MODELS AND CHARACTERIZATIONS OF NEUTRON SCINTILLATION DETECTORS FOR FEASIBILITY OF SPENT FUEL CASK MONITORING

By

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A DISSERTATION PRESENTED TO THE GRADUATE SCHOOL OF THE UNIVERSITY OF FLORIDA IN PARTIAL FULFILLMENT OF THE REQUIREMENTS FOR THE DEGREE OF DOCTOR OF PHILOSOPHY

UNIVERSITY OF FLORIDA

2017
I dedicate this work to my family.
ACKNOWLEDGMENTS

I would like to express my deepest gratitude to my advisor, the chair of my thesis committee, Prof. Andreas Enqvist for being very supportive over the years. Thanks for all the valuable advice and consistently reminding me to think critically and improve myself. Working under Prof. Enqvist, I had many opportunities to explore different fields and to present in professional conferences. It would be impossible to accomplish all the work without his trust and patience. I would also like to thank Prof. Kelly Jordan for everything he has done for me. I am very grateful for his lectures on reactor physics, encouragement on my career and certainly for providing the D-D neutron generator. The days spent with him and his research group inside the underground tunnel at Ohio University were memorable. I would also like to thank Prof. Edward Dugan who was the course advisor when I first joined the Nuclear Engineering Program. I am especially grateful for his valuable lessons, standards, work attitude, encouragement and stories about the history of the Nuclear Engineering Program at the University of Florida.

Additional thanks are also due to my committee members: Prof. James Baciak for accepting me into Nuclear Engineering Program and showing me his passion for work, kindness, incredible consideration and care towards students and the department. He has been a wonderful role model for many of us. At the same time, I would also like to thank Prof. Adam Veige and Prof. Assel Aitkaliyeva for serving on my thesis committee and reading my updates. Thanks also to Prof. James Tulenko and Prof. Ghatu Subhash for providing the research opportunity in novel nuclear fuel developments in my first year of graduate study at the University of Florida. The work on carbon nanotubes and the application of spark plasma sintering was very interesting and enabled me to explore the micro world.
Thanks are extended to the nuclear physicist, Prof. Thomas Massey and the electronic engineer, Don Carter and other personnel at the Ohio University Edwards Accelerator Facility for their assistances in using their facility and valuable discussions during my measurement campaigns in 2014 and 2016. Their cooperation made the accurate time-of-flight measurements possible. I would also like to say thanks my colleagues and friends: Andrew, Jonathan, Xianfei, Ting, Kelsey, Erica, Robert, Noah, Ira, John and many others who I have worked with. Without them, it would be much less fun moving bricks around in the lab. Additionally, special thanks to Bob who read through my thesis and provided valuable feedback on my writing.

Also, I would like to take this opportunity to express my deepest gratitude to my family for their unconditional love and encouragement. I wouldn’t have pursued education in the U.S. without their understanding and trust. In the end, I sincerely wish the UF Nuclear Engineering Program continues to grow and flourish.
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Abstract of Dissertation Presented to the Graduate School
of the University of Florida in Partial Fulfillment of the
Requirements for the Degree of Doctor of Philosophy

MODELS AND CHARACTERIZATIONS OF NEUTRON SCINTILLATION DETECTORS
FOR FEASIBILITY OF SPENT FUEL CASK MONITORING

By

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May 2017

Chair: Andreas Enqvist
Major: Nuclear Engineering Sciences

The goal of this research is to investigate different scintillation-based neutron
detectors to understand their applicability and limitations for monitoring the condition of
spent fuel casks. It starts with an analytical model for finite deuterated scintillation
detectors in cylindrical shape. The anisotropic model would be beneficial to understand
responses of scintillation materials heavier than hydrogen.

In fast neutron related experiments, neutron scatter signals and neutron beam
triggers were recorded in coincidence mode, and the incident neutrons were
categorized by energy as obtained by time-of-flights. The recognition of fast neutron
detection pattern was mainly based on pulse shape discrimination algorithms.
Scintillators were characterized by a neutron light-response matrix and its
corresponding light output function. The nonlinear neutron response was modelled and
fitted with advanced functional shapes to capture the pulse height response of the
detector. To capture thermal neutrons released from a loaded TN-32 fuel cask, $^{6}\text{Li}$-
enriched Cs$_2$LiYCl$_6$ was also investigated to study its performance in identifying fast and
thermal neutrons. A smoothing filtering method was applied to modify the existing
waveform to separate its slow components. Neutrons were additionally analyzed to identify thermal and fast neutrons.

Additionally, the spatial resolution is severely affected because of the relatively large size of an organic scintillator when compared to the gamma detector active volumes. To compensate for this drawback, a spatial dependent model of photon generation and tracking in the scintillation medium considering the amount of deposited energy of incident neutrons was created.
CHAPTER 1
INTRODUCTION

The objectives of this research were to investigate neutron interaction mechanisms with scintillation materials, algorithms and characterization of scintillation detectors to determine how different detectors can be best used in spent fuel cask structure evaluation. The main focuses were on a deuterated organic scintillator, EJ-315, and a $^{6}\text{Li}$-enriched Cs$_2$LiYCl$_6$: Ce (CLYC) crystal. Through the study of EJ-315, an analytical model was created to mimic the neutron collision series and the energy transport histories and to simulate the signals of light outputs. The performance of several pulse shape discrimination algorithms were investigated and compared. Slow and fast decay constants were calculated by pulse shape fittings with exponentially modified Gaussian distributions. Through the study of CLYC, smoothing filtering functions were implemented to simulate a circuit module to enhance the separation of the tail integral which improved the performance of pulse shape discrimination algorithm. Additionally, a Monte-Carlo model of photon transportation was created to evaluate the location dependence of photon transportation in a cylindrical detector.

1.1 Thesis Layout

In this thesis, Chapter 1 covers the motivations of this research and the fundamentals of the scintillation mechanisms. In order to better understand how a detector responds to neutrons and how materials can contribute differently to the light outputs, Chapters 2 and 3 describe the neutron collision probabilities and light generation models to predict the neutron pulse height distribution of an EJ-315 detector, respectively. Additionally, a complete pulse height model uses the light output function as a part of the inputs of the model, which requires a thorough study of pulse shape
discrimination algorithms and their applications to extract the response matrix. Thus, we will first go through different pulse shape discrimination algorithms and study their consistency in performance in Chapter 4, and then show the application of pulse shape discrimination algorithms in EJ-315 to study its properties including the response matrix, light output function and resolutions in Chapter 5. For Chapter 6, we calculate the decay constants based on neutron and gamma ray pulses and to confirm the scintillation mechanisms. Chapters 1 to 6 mainly focus on the organic scintillators for fast neutron detection, and we will explore the study on thermal neutron detection using a CLYC detector while making use of the algorithms we have studied or created. Finally, we intend to put the photon tracking model at the end of this thesis, which is used to investigate the dependence between the locations of where photons are generated and the final recorded results.

1.2 Motivation

Due to recent developments of digital equipment in fast analysis and high recording speed, the door has opened to digital signal processing for direct fast neutron detection which has been hard to do previously. The popular applications of neutrons due to electrical neutrality and small wavelength are mainly restricted to thermal neutrons, where their penetrating and probing nature can be used to characterize materials such as crystals. However, the wavelength of fast neutrons is even smaller when compared to that of thermal neutrons. The major obstacles that limit the fast neutron applications are: 1) the availability of fast neutron beams, and 2) the effective detection of fast neutrons in a limited space.

The University of Florida has a 100 kW light water Training Reactor which has resumed its operation since April 2015. It provides a promising facility for fast neutron
beams. Meanwhile, our nuclear program has a D-D generator emitting isotropic 2.45 MeV neutrons at a maximum output of $4 \times 10^9$ per second. Strong Plutonium-Beryllium sources are also available at UF for detector characterizations. To make full use of those fast neutron facilities and explore the applications of fast neutrons in fundamental science and in the monitoring of spent fuel cask, more study in neutron detection is essential.

If fast neutrons can be effectively detected using scintillation materials with advanced algorithms, fast neutrons can be used in scattering research like small angle scattering. One of the direct benefits of effective detection is that the detection of fast neutrons does not require any moderation, which could potentially simplify the detector setup and significantly improve fast neutron diffraction applications. The main work of my PhD thesis is on the neutron scattering, pulse height distribution and the optical model on photon tracking in the EJ-315 detector. Other part of my research relates to scintillation material itself, algorithms and nuclear data visualization. As mentioned previously, all the work aims to provide fundamental science advances in fast neutron detection and promote applications of fast neutron sources at the University of Florida in nuclear technology.

1.3 Spent Fuel Cask Evaluation

Currently, the demand for dry cask storage facilities for nuclear spent fuel is on rise due to the recent increase in nuclear power in some countries and continued operation of currently existing nuclear plants. The dry cask storage is the primary solution to temporarily store the spent fuel that has already been cooled in a spent fuel pool. However, the structure of cask is continuously exposed to the remaining heat emitted by the spent fuel, which will cause material degradation and other radiation
damages. Since the cask is permanently sealed after the load of spent nuclear fuel, as a result it has been a challenge since the early 1980s to monitor the condition of spent fuel inside a cask and the structure materials without physically penetrating the spent fuel container. It has been verified in our group that despite of the heavy shielding layers on the cask, some radiation penetrate the shielding materials and reach the surrounding air. The emitted primary and scattered or induced secondary radiation can be utilized to provide a correlation to the material inside the fuel canister. The interior image reconstruction procedure relies on the inversion of the gamma-ray and neutron transform. Thus, it would be essential to have a comprehensive system consisting of multiple well characterized scintillators for radiation detection.

Figure 1-1. The TN-32 spent fuel cask view in auto-CAD.

The inventory of spent fuel in a cask, like TN-32, as shown in Fig. 1-1, is large and heavy, and the imaging of the cask requires sophisticated detectors and imaging system. Direct measurements in spent fuel storage field could be time-consuming and
lack of details. Thus, a full understanding of the radiation characteristics and the detector themselves is the key to the project.

1.4 Neutron Detection

The detection of thermal neutrons and fast neutrons is very different. The nuclear cross-sections of thermal neutrons are much larger than those of fast neutrons. Thermal neutrons cause nuclear reactions and create products such as protons, alpha particles, gamma rays and fission fragments that can be detected efficiently. As a result, thermal neutron detection is dominating.

![Figure 1-2. Cross section versus neutron energy for some reactions of interest in neutron detection [1].](image)

Fast neutrons can be detected based on a number of commonly used slow neutron-induced reactions during which directly detectable charged particles are produced. For instance, the $^{10}\text{B}(n,\alpha)$ reaction causes the release of amount of energy (more than 2 MeV) depending on the status of $^7\text{Li}$. Unfortunately, the probability that a neutron will interact by this reaction decreases rapidly with increasing neutron energy,
as shown in Fig. 1-2 [1]. This phenomenon also applies to other typical thermal neutron detectors as seen in Fig. 1-2. Nevertheless, in principle, such reaction could be applied to detect fast neutrons, and the main approach is to slow down neutrons with moderators before being captured by $^{10}$B or other materials. However, such a detection method loses the energy information of detected neutrons. Thus it is energy insensitive and the approach would be disadvantageous when the energy of neutrons are needed to investigate the origin source information for purposes like nuclear nonproliferation.

Instead of relying on the fast neutron moderation and nuclear reactions induced by thermal neutrons, the detection of fast neutrons mainly uses liquid organic scintillators relying on elastic scattering and the application of pulse shape discrimination methods.

1.4.1 Energy Transformation in Neutron Elastic Scattering

Currently, the most common method of fast neutron detection is based on elastic scattering of neutrons by light nuclei, rather than nuclear reactions. The scattering interaction transfers a portion of the neutron kinetic energy to the target nucleus, resulting in a recoil nucleus. In such a reaction, the Q-value of elastic scattering is zero due to the conservation of the total kinetic energy before and after the collision. This continues until the scattered neutron energy is too low to generate a considerable amount of signals, or it simply escapes from the detector since the active volume of a detector is limited. For single scattering, the fraction of the incoming neutron energy that is transferred to the recoil nucleus can range anywhere from zero to the full energy, depending on the target nucleus and the scattering angle. For hydrogen, the maximum energy deposited in a single collision can be the full incident neutron energy, and its energy distribution can be treated homogeneously [2].
However, for an isotope of hydrogen, like deuterium, the energy distribution of a scattered neutron has to be described as a function of its scatter angle. This is also true to other elements like carbon [3]. Nevertheless, conservation of momentum and energy in the center-of-mass coordinate system gives the following relation for the energy of the scattered neutron kinetic energy ($E'_n$):

$$E'_n = \frac{A^2 + 1 + 2A \cos(\theta)}{(1 + A)^2} E_n,$$

(1-1)

where $A$ is the atomic mass number of the target nuclei, $\theta$ is the scattering angle of the neutron in the center-of-mass coordinate system, and $E_n$ is the incoming neutron kinetic energy. The energy difference between the incident neutron and the scattered neutron is the deposited energy. The scattered neutron can undergo further collisions with other atoms and deposit more energy inside the scintillation material. As Eq. 1-1 shows, the most efficient moderator is hydrogen because a neutron can lose up to all its energy in a single collision with a hydrogen nucleus. For heavier nuclei, only a partial energy transfer is possible. Thus, in our study, only light materials are used. However, regarding materials that are heavier than hydrogen, double differential cross-sections will have to be considered since the energy of the recoil nuclei is angle-dependent.

1.4.2 Scintillation Mechanism in Organic Scintillators

Before discussing the physical process, it is important to introduce the concepts of fluorescence and phosphorescence. Fluorescence is the prompt emission of visible radiation from a substance following its excitation by some means, and phosphorescence corresponds to the emission of longer wavelength light than fluorescence, and with a characteristic time that is generally much slower.
During a random collision, if enough energy is transferred to a recoil nucleus which is a charged particle, the recoiling nucleus excites or ionize molecules by both direct collisions and long range transfers when it passes through scintillation medium. The recoil nucleus can be a single charged proton or just a regular nucleus composed of protons and neutrons.

In aromatic hydrocarbons, such as those typified by the ring structure of benzene, three of the four valence electrons of carbons are in the hybridized orbitals ($\sigma$). Those are strongly localized between each carbon, its two carbon neighbors, and a single hydrogen. The fourth electron in the so-called $\pi$ orbital, is not as well localized and does not participate in the bond as strongly as the $\sigma$ electrons. Thus, it is believed that the $\pi$ electron is most responsible for the scintillation process. The $\pi$-electronic energy levels are illustrated in Fig. 1-3 [4]. When one of the two electrons of opposite spins is promoted to a molecular orbital of higher energy, its spin is in principle unchanged so the total spin quantum number remains zero. The multiplicities of both the ground and excited states is equal to 1, and both are called singlet state. A series of singlet states (spin 0) are labeled as $S_0$, $S_1$, $S_2$, ... A similar set of triple (spin1) electronic levels are shown as $T_1$, $T_2$, $T_3$, ... whose total spin quantum number is 1 and the multiplicity is 3. According to Hund’s Rule, the triplet state has a lower energy than that of the single state of the same configuration.

For molecules of interest in the organic scintillators like EJ-309, the energy space between $S_0$ and $S_1$ is in the order of eV, which is usually higher than the spacing between higher-lying states. Additionally, each of these electronic configurations is further subdivided into a series of levels with much finer spacing that correspond to...
various vibrational states of the organic molecule. The typical spacing of this category is of the order of 0.15 eV. A secondary subscript is added to distinguish these vibrational states. For instance, $S_{00}$ represents the lowest vibrational state of the ground state. However, the spacing between vibrational states is much larger compared with average thermal energies (0.025 eV). Thus, according to the Boltzmann population distribution $e^{-E/kT}$, at room temperature nearly all molecules are in the $S_{00}$ states.

Figure 1-3. The energy levels of an organic molecule with $\pi$-electron structure. The ionization level $I_{\pi}$ is shown at the top. The excited states as well as vibrational sublevels are shown as dashed horizontal lines. Internal degradation is a nonradioactive process [4].

In Fig. 1-3, the absorption of energy by the molecule and the scintillation light emission are represented by the upward and the downward arrows, respectively. In the case of an organic scintillator, the absorption of energy from a charged particle passing through an aromatic system causes the excitation of electrons which are quickly de-excited to $S_1$ electron state through non-radiative internal conversion. Any state with excess vibrational energy is not in thermal equilibrium and quickly loses the vibrational energy. Therefore, the outcome of the excitation process in a simple organic scintillator
produces a population of excited molecules in the $S_{10}$ states within a negligibly short period of time.

Table 1-1. Main mechanisms describing interchanges of molecule states [6]

<table>
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<tr>
<th>Interaction</th>
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<tr>
<td>$T_n \rightarrow T_1 (n \geq 2)$</td>
<td>internal conversion</td>
</tr>
<tr>
<td>$S_n \rightarrow T_n \rightarrow T_1$</td>
<td>intersystem crossing followed by internal conversion</td>
</tr>
<tr>
<td>$S_1 \rightarrow T_1$</td>
<td>intersystem crossing</td>
</tr>
<tr>
<td>$S_1 \rightarrow S_0 + hv$</td>
<td>fluorescence</td>
</tr>
<tr>
<td>$I^+ + e^- \rightarrow \begin{cases} T_n \text{($\sim 75%$)} \ S_n \text{($\sim 25%$)} \end{cases}$</td>
<td>ion recombination</td>
</tr>
<tr>
<td>$T_1 \rightarrow S_0 + en$</td>
<td>internal conversion (non-radiative)</td>
</tr>
<tr>
<td>$T_1 \rightarrow S_0 + h\nu_p$</td>
<td>phosphorescence</td>
</tr>
<tr>
<td>$T_1 + T_1 \rightarrow \begin{cases} T_1 + S_0 + en \ 2S_0 + en \ S_1 + S_0 \end{cases}$</td>
<td>partial self-quenching</td>
</tr>
<tr>
<td>$T_1 \rightarrow S_1$</td>
<td>production of $S_1$ which leads to p-type delayed fluorescence</td>
</tr>
<tr>
<td>$T_1^{thermal} \rightarrow S_1$</td>
<td>production of $S_1$ which leads to e-type delayed fluorescence</td>
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Through the inter-system crossing, some excited singlet states may be converted into triplet states. However, this or the reverse transition involves a change in electronic state, and is less likely to happen when compared to a singlet-to-singlet transition. As a result, the lifetime for the first triplet state $T_1$ is characteristically much larger than that of the singlet state $S_1$. From the aspect of observation, the radiation emitted in a de-excitation from $T_1$ to $S_0$ is therefore a delayed light emission, characterized as phosphorescence. As shown in Fig. 1-3, the energy level of $T_1$ lies below $S_1$, the wavelength of the phosphorescence spectrum will be slightly longer than that of the
fluorescence spectrum. Additionally, some molecules may be thermally excited back to the $S_1$ state and subsequently decay through regular fluorescence, which is the delayed fluorescence.

For a more detailed theory which explains how the excited molecules are formed, we have adopted G. Laustriat et al.’s early work [5]. The basic idea is that when a recoil nucleus travels through scintillation medium, it produces excited molecules and ionizations by both direct collisions and long range transfers. Most of the excitations and ionization occur along the path of the particle within a given radius $r_0$. During the ionization, electrons can be given various amounts of kinetic energy. For the very slow secondary electrons, they are quickly slowed down and trapped by the ions within the interacting distance $r_0$; the more energetic ones will escape the track region and behave as primary particles. They further produce ionizations and excitations along their track. These phenomena happen frequently especially by the end of their tracks. As a result, there are ions, free electrons and excited molecules produced. Meanwhile, the ions and the free electrons recombine rapidly to form excited molecules which can be at different electronic levels as shown in Fig. 1-3. The main processes are shown in Table 1 [6].

1.4.3 Scintillation Process Induced by Gamma-ray

We also want to briefly mention the gamma-ray interactions with scintillation materials to grasp the differences among mechanisms involved in the scintillation process starting with recoil nuclei. They interact primarily through photoelectric absorption, Compton scattering and pair production. In the photoelectric effect, a photon is absorbed by an atom, which emits one atomic electron. Additionally, the exact probability for photoelectric absorption is difficult to calculate. However, it is most significant for low energy photons and the probability increases quickly with the atomic
number of Z or the number of atomic electrons. Compton scattering is the process by which a photon scatters with a nearly free atomic electron, resulting in a less energetic photon and a scattered electron carrying the energy lost by the photon. The last process is pair production, in which the photon is absorbed and an electron-positron pair is produced. In this process, there is clearly an energy threshold of $2m_ec^2$. Thus, the pair production in general is only important to photons of high energy and only Compton scattering dominates in our research.

![Graph](image)

Figure 1-4. Comparison of electron and proton stopping power distributions. Data colored in red are for electrons, while in blue are for protons. The original cross section data were cited from the National Institute of Standards and Technology (NIST) Physical Measurement Laboratory.

### 1.4.4 Scintillation Differentiation Summary

Gamma rays in the background of fast neutron fields are primarily in the energy range within which they are predominantly interacting with electrons through Compton scattering. For heavy charged particles, like recoil nuclei, Rutherford scattering is an important process in nuclear physics. However, it has very limited influence on the loss...
in energy as it travels through the scintillation materials, because the nuclei are only about $10^{-15}$ of the volume of their atoms [7] [8]. Instead, the dominant mechanism for the energy transfer is Coulomb scattering by the atomic electrons in the scintillation materials. It's important to notice that because of the infinite range of the Coulomb force, the recoil nucleus interacts simultaneously with many electrons and loses energy gradually and continuously along its travel path. As a result, a recoil nucleus travels much less distance than a scattered electron because the recoil nucleus interacts intensively with surrounding scintillation material and reduces its energy very quickly to zero. Such a phenomenon can be confirmed according to the stopping powers of proton and electron over various energies in the medium of stilbene, as shown in Fig.1-4 and a summation of the interaction mechanisms is shown in Fig. 1-5.

Figure 1-5. Mechanisms of gamma rays, fast and slow neutrons interacting with scintillation matters inside the detector cell and the corresponding photomultiplier tube.
CHAPTER 2
NEUTRON COLLISION MODEL

This chapter presents the theoretical model for collision sequences and the corresponding collision probability distributions. Specifically, based on the Peierls-formula, a complete and self-contained analytic model of pulse height distribution for EJ-315 is created. In the model, neutrons with an energy range from 1 to 10 MeV are investigated. The pulse height distribution and elaborate measurements at Ohio University using the deuterated organic scintillation detector will be explained in Chapter 3. Because of the length, we decided to split the neutron pulse height model into the neutron collision model (in Chapter 2) and the light signal generation model (in Chapter 3). The neutron collision model allows us to investigate collision probabilities and build a matrix of deposited energy and probability. The matrix will be used as inputs of the light signal generation model. In measurements, a threshold in the order of 100 keV is applied to minimize noises and leads of the pulse deficiency at the low pulse height region. The complete model compensates for the pulse deficiency.

2.1 Why Deuterated Liquid Scintillators

Liquid organic scintillation detectors are frequently used in fast neutron detection in nuclear fields such as nuclear non-proliferation and nuclear material accountability. \(^3\)He is normally used in gas proportional counters. However, due to significant demand in homeland security, and decrease in supply from the decay of tritium, it is therefore desirable to find alternatives to \(^3\)He [9]. Organic scintillators as promising candidates are used as neutron and gamma-ray detectors because of their fast response and large-volume availability at relatively low cost. Unlike conventionally used \(^3\)He gas tubes, liquid organic scintillators do not require neutron moderation [10, 11], and can be
applied to separate gamma rays and neutrons by applying the charge-integration based pulse shape discrimination algorithms [12, 13]. Additionally, higher mass isotopes such as deuterium have been suggested for use to obtain improved spectroscopic information in nuclear safeguards [14] or nuclear science [15].

Some studies have investigated light pulse distribution of organic liquid scintillation detectors. For instance, light pulse amplitude distribution of EJ-309 detectors mainly composed of carbon and hydrogen have been reported [2] [16]. Their calculations were carried out based on an assumption that incident neutrons are uniformly scattered after collisions. Only in the case of scattering on hydrogen nuclei, $^1$H, is the isotropic assumption a highly accurate one. It is not suitable for incident neutrons with energy of several MeV considering that the scattering angular distribution is quite anisotropic and dependent on incoming energies and nuclei with which neutrons collide. Quantitative simulation or analytical modeling work on the elastic scattering with and without angular dependence has not been extensively investigated for deuterated scintillators.

In experiments, V. Bildstein et al. reported that the performance of deuterated scintillation detectors for fast-neutron detection is comparable to or surpasses that of non-deuterated scintillation ones with additional benefits on discriminating multiple-scattering events [17] [18]. For instance, pulse-height responses to mono-energetic 2.5 MeV neutrons for deuterated EJ-315 and hydrogen-based EJ-309 are shown in Fig. 2-1 [14]. However, the light intensity in measurement induced by neutrons is a result of stochastic collisions, and cannot provide any details on the energy transfer history of an
individual collision. Thus, the motivation of this research further arises from the need for enhanced information extraction for unfolded spectra.

![Figure 2-1. Illustration of pulse-height responses to mono-energetic 2.5 MeV neutrons for deuterated EJ-315 and hydrogen-based EJ-309 [14].](image)

### 2.2 Analytic Collision Model

The slowing down of fast neutrons is caused by multiple scatters on deuterium (D) and carbon (C) nuclei. A scattered neutron deposits a fraction of its kinetic energy inside the detector until its energy is too low to initiate a considerable amount of signal, or it simply escapes from the detector since the active volume of detector itself is limited. Then, the fluorescence light is converted into an electric signal by the photocathode and then collected by a photomultiplier tube which is mounted to the organic liquid cell’s surface and amplifies the electron output. However, most neutron detectors can only provide the total light output generated in measurements; while in the model calculation, each individual stochastic neutron collision with different nuclei can be investigated allowing us to quantify the probability and spectrum of a light signal produced due to a specific collision sequence.
The collision probability theory is based upon an implicit assumption that, once scattered, the neutrons are isotropically distributed inside the detector. As reported in Ref. [19], the collision probability in a sample volume V for an isotropic neutron source is an integral over space for a transcendental function. The function is energy, geometry, and scintillation material composition dependent and is defined as the Peierls-formula. The function can be expanded into an amenable form, \( P_c \), such that numerical calculation is possible in the case of cylindrical geometry to suitably fit the detector.

\[
P_c(E) = 1 - \frac{1}{h} [E_3(0) - E_3(h)] + \frac{4}{\pi d^2 h} \int_0^d dt \left\{ E_3(t) - E_3 \left[ \frac{(t^2 + h^2)^{1/2}}{2} \right] \right\} (d^2 + t^2)^{1/2} - \]

\[
\frac{4}{\pi d^2 h} \int_0^h du (h - u) \int_0^d t^2 dt e^{\frac{-((t^2 + u^2)^{1/2})}{2}} (d^2 - t^2)^{1/2}
\]

with \( E_n(z) = \int_1^\infty \frac{e^{-zu}}{u^n} du \), where the height \( h \) and the diameter \( d \) are expressed in mean free paths. In the study of EJ-315, \( h(E) = d(E) \).

Figure 2-2. Scattering cross section of carbon and deuterium for neutron with kinetic energy up to 15 MeV.
Figure 2-3. Recoil nucleus kinetic energy distribution of scattered neutrons due to (a) deuterium, and (b) carbon. $E_R$ stands for energy of scattered neutron, and $E_n$ kinetic energy of incident neutron.

The scattering cross sections of both deuterium and carbon are plotted (Fig. 2-2).

Obviously, the carbon cross section changes significantly over energy and gives rise to
the variation in collision possibility. Table 2-1 tabulates the cylinder detector dimensions d and h in mean free path (MFP) for neutrons with energies of 1 MeV. As MFPs of neutrons increase when they have higher kinetic energies, dimensions of d and h expressed in MFPs decrease.

Table 2-1. Detector dimensions of cylinder geometry expressed in mean free paths for neutrons with energy of 1 and 10 MeV

<table>
<thead>
<tr>
<th>Dimensions (cm)</th>
<th>MFPs (1MeV)</th>
<th>MFPs (10 MeV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.2223</td>
<td>0.0885</td>
</tr>
<tr>
<td>5</td>
<td>1.1116</td>
<td>0.4423</td>
</tr>
<tr>
<td>10</td>
<td>2.2232</td>
<td>0.8847</td>
</tr>
<tr>
<td>15</td>
<td>3.3348</td>
<td>1.3270</td>
</tr>
</tbody>
</table>

Since neutrons in the detector volume can collide either with carbon or deuterium, or escape away from the detector, probability of escape, \( W(E) \), from the volume is given as:

\[
W(E) = 1 - C_D(E) - C_C(E),
\]  

where

\[
C_C(E) = P_C(E) \frac{\Sigma_C(E)}{\Sigma_D(E) + \Sigma_C(E)},
\]  

and

\[
C_D(E) = P_C(E) \frac{\Sigma_D(E)}{\Sigma_D(E) + \Sigma_C(E)}.
\]

Furthermore, the initial escape probability \( W(E) \) can be calculated from an exponential distribution in some cases, such as a known mono-directional incident flux.

For fast neutrons the absorption probability is relatively low unless neutrons are thermalized. In our calculation model, the absorption is assumed not to contribute to the light generation. Rather than being isotropically scattered after collision, as calculated in Ref. [2], neutrons are scattered at various angles with a probability that is based on the
conservation of momentum and energy in the center-of-mass coordinate system. As a result the recoil neutron kinetic energy \( E_R \) in scintillation materials is given by:

\[
E_R = \frac{A^2+1+2A \cos\phi}{(1+A)^2} E_n,
\]

(2-5)

where \( A \) is the atomic mass number of the target nucleus, \( \phi \) the scattering angle of the neutron in the center-of-mass coordinate system, and \( E_n \) the incoming neutron kinetic energy.

According to Eq. 2-5, the energy ratio of \( E_R \) to \( E_n \) is a function of \( \phi \). The probability distribution of energy is determined by the double differential cross-section \( \sigma(E, \phi) \). For convenience, we rearrange recoil neutrons with different kinetic energy based on the energy ratio embedded with \( \sigma(E, \phi) \). The incident neutron elastic scattering differential cross-sections of both deuterium and carbon, cited from ENDFB-VII, after interpolation are plotted in Fig. 2-3.

The carbon cross-section changes significantly with energy and gives rise to large variation in collision probability with respect to both energy and scattering angle. As shown in Fig. 2-3, after the collision with a carbon nucleus, the minimum energy ratios between a recoil neutron and the corresponding incident neutron are calculated as 0.111 for deuterium and 0.716 for carbon, respectively. The corresponding scattering probability is written as \( P_s(E_n/E_R, E_R) \), whose integral over the whole possible ratio is unity. Further, we have

\[
P_{x^n}(E_n, E_R) = \frac{P_s(E_n/E_R, E_R)}{E_R},
\]

(2-6)

where \( X \) stands for collision nuclei. Then, the energy probability distribution of neutrons collided once with deuterium and once with carbon, respectively, is expressed as
\[ f_D(E_1, E_0) = H(E_1 - \alpha_D E_0) C_D(E_0) P_{Dn}(E_1, E_0), \quad 0 < E_1 < E_0, \quad (2-7) \]

and

\[ f_C(E_1, E_0) = H(E_1 - \alpha_C E_0) C_C(E_0) P_{Cn}(E_1, E_0), \quad 0 < E_1 < E_0. \]

\(H(x)\) is the unit step function used to control the energy transfer induced by collision and the collision parameter \(\alpha = (A - 1)^2/(A + 1)^2\). However, for the probability of only one collision, the leakage function \(W(E)\), which is adopted to terminate the calculation from further interactions, must be considered to eliminate the chance of further energy deposition.

Table 2-2. Probabilities of various collision histories in the case of incoming neutrons with 1 MeV kinetic energy

<table>
<thead>
<tr>
<th>Collision History</th>
<th>Detector Dimensions ((h=d))</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>1 cm</td>
</tr>
<tr>
<td>(P_0 = W(E_0))</td>
<td>0.7833</td>
</tr>
<tr>
<td>(P_D)</td>
<td>0.0883</td>
</tr>
<tr>
<td>(P_C)</td>
<td>0.0804</td>
</tr>
<tr>
<td>(P_{CC})</td>
<td>0.0085</td>
</tr>
<tr>
<td>(P_{CD})</td>
<td>0.0086</td>
</tr>
<tr>
<td>(P_{DC})</td>
<td>0.0107</td>
</tr>
<tr>
<td>(P_{DD})</td>
<td>0.0089</td>
</tr>
<tr>
<td>(P_{CCC})</td>
<td>0.0009</td>
</tr>
<tr>
<td>(P_{DDD})</td>
<td>0.0009</td>
</tr>
<tr>
<td>(P_{CDD})</td>
<td>0.0009</td>
</tr>
<tr>
<td>(P_{DDD})</td>
<td>0.0013</td>
</tr>
<tr>
<td>(\sum P_{xxx})</td>
<td>0.0082</td>
</tr>
<tr>
<td>(\sum P_{xxxx})</td>
<td>0.0019</td>
</tr>
<tr>
<td>Probability of first four collisions</td>
<td>0.9988</td>
</tr>
</tbody>
</table>

In the case of neutrons that have collided exactly twice the probability distribution is given by Eq. 2-8. The collision consequence \(f_{DC}\) indicates that the neutron first collided with a deuterium nucleus followed by a collision with a carbon nucleus. The probability of two collisions is given in the following:
\[ f_2(E_2, E_0) = f_{DD} + f_{DC} + f_{CD} + f_{CC} = \int_{E_2}^{\text{Min}[E_0, E_2]} f_D(E_1, E_0) f_D(E_2, E_1) W(E_2) dE_1 + \]
\[ \int_{E_2}^{\text{Min}[E_0, E_2]} f_D(E_1, E_0) f_C(E_2, E_1) W(E_2) dE_1 + \ldots. \] (2-9)

For higher order collisions, the energy distribution of a specific history can always refer back to the scattered neutrons from a previous collision in the form of a recursive function. For instance,

\[ f_{DDCD}(E_4, E_0) = \int_{E_4}^{\text{Min}[E_0, E_4]} f_{DDC}(E_3, E_0) f_D(E_4, E_3) W(E_4) dE_3. \] (2-10)

### 2.3 Deposited Neutron Energy

The deposited energy inside the scintillation materials is given as the complement of the scattered neutron energy. \( E_d \) is written as:

\[ E_d = E_0 - E_R. \] (2-11)

Thus, the probability distribution of deposited energy after collisions, such as DDCD is given as

\[ f_{d_{DDCD}}(E_d, E_0) = f_{DDCD}(E_0 - E_4, E_0). \] (2-12)

### 2.4 Result and Discussion

Table 2-2 lists various individual collision probabilities for incident neutrons with 1 MeV kinetic energy, indicating that detectors with small dimensions have less ability to constrain incoming neutrons and are relatively transparent. As expected for the larger dimensions, incident neutrons are transported within a relatively larger volume and tend to collide multiple times before terminating the collision history either due to leakage or absorption.
For the numerical inputs of the analytical model only angular dependent scattering cross-sections of deuterium and carbon, the detector size, and incoming neutron energy are required. This method not only allows us to evaluate the individual contribution of the collision probability distribution (Fig. 2-3), but also helps to quantify the effects of the geometry of detectors and their scintillation materials on the neutron energy probability. It is important to remember that this analytic model is also suitable for nuclei other than carbon and deuterium. As a curious example, the second derivative, or the change of slope of the distribution for twice-collided neutrons with carbon is reversed when anisotropic scattering is implemented. The distribution thus results in an overestimation of the transferred energy, and the consequence of the pace of neutron moderation per scatter, if not taking anisotropic scattering into account at these energies [20]. Probability distributions of multiple collisions are shown in Fig. 2-4. More thorough discussion is available in Chapter 3 along with the light signal generation model.
Figure 2-4. Energy probability distributions of recoil neutrons which have collided with nuclei inside a detector with dimensions of 10 x 10 cm after the second collision (a), the third collision (b), and sum of probabilities with different collision numbers (c, d). The total collision probability of the first collision is labelled as “col.1”, the first two “col. 1+2”, etc. The initial incident neutron energy is fixed at 1 MeV, except in c-2 whose corresponding neutron incident energy is 3 MeV.

2.5 Conclusion

An analytical model of the collision model for the finite deuterated scintillation detector is created using the impulse approximation. The energy distribution of a scattered neutron is calculated based on an existing collision probability scheme for general cylindrical shaped detectors taking into account double differential cross-sections. A non-uniform angular scattering distribution has been mapped to the collision model, and the neutron energy up to 10 MeV has been investigated. The model reveals the detail of collision history in sequences to capture deposited neutron energies in deuterium based scintillation materials. Regardless of significant observed differences
in light outputs, the probabilities of neutron collisions with carbon and deuterium nuclei are comparable.
CHAPTER 3
LIGHT SIGNAL GENERATION MODEL

This chapter is the extension of the previous work which describes the energy deposition within organic medium. The effects on light output of multiple factors, including detector geometry, neutron leakage, and anisotropic angular distribution, have been taken into consideration. The completed calculation exhibits the quantitative probability of fast neutron slowing down history because of stochastic collisions with carbon or deuterium nuclei; the neutron light yield function of a deuterated liquid scintillator is measured and calibrated vs. electron response. The quantitative collision probability convoluted with the light output function for continuous neutron flux gives the light output for various collisions. Furthermore, at the end of this article, individual shape of the light pulse spectrum induced by 5 MeV neutrons is analyzed and compared to the corresponding pulse height distribution from measurements. For future application, this model can be used to evaluate the property of a targeted detector which is not commercially available, and to predict its qualitative response matrix as modified only by the light generating function of the new compound.

3.1 Analytical Model of Light Generation

As previously described, this model calculates the probability distribution of remaining and deposited energy for neutrons having undergone scatters with various nuclei. Using functions describing the relationship between the conversion of the deposited energy and light, we can then calculate the detector pulse height distributions as a function of the collision histories associated with both D and C. As reported [16], the existence of a non-linear relationship between the deposited neutron energy and the corresponding converted light pulse gives rise to pulse height variations. The fitting of
the light function for EJ-315 is based on measurements which were completed in the tunnel of the Edwards Accelerator Laboratory at Ohio University. References [2] and [16] applied an empirical quadratic polynomial function to describe the light generation:

\[ L(T) = c_1 T^2 + c_2 T, \]  

(3-1)

where \( c_1 \) and \( c_2 \) are constants, and \( T \) is the deposited energy. However, for a small amounts of transferred energy, the polynomial fitting does not properly capture the light generation function. As suggested in Ref. [20], a semi-empirical exponential fit is used in our work which agrees better with the measured light function. The light output empirical formula for deuterium is expressed as:

\[ L_D(T) = aT - b[1 - \exp(1 - cT)]. \]  

(3-2)

Then the inverse function \( T \) is written as

\[ T_D(L) = \frac{bc+cl+a\cdot F\left(-\frac{bc}{a}\cdot\exp\left[-\frac{c(b+L)}{a}\right]\right)}{ac}, \]  

(3-3)

where \( a = 1.1216, b = 15.0451, c = 0.0708, \) and \( F(z) \) is defined as the principal solution for \( w(z) \) in \( z = w \cdot \exp(w) \). For carbon, the light generation function is approximated as a linear function of the transferred energy with \( d = 0.02 \text{ MeVee} \) [16]:

\[ L_c = dT. \]  

(3-4)

Even though Eqs. 3-2 and 3-4 are simplified and empirical, they are believed to be coarse approximations to the physical mechanisms relevant to light generation and absorption inside the detector. This includes the light spectrum emitted by the scintillator with associated scintillation efficiency, light absorption of scintillation materials and the detector walls, and the response of the photomultiplier to light.
According to the principle used to describe the light amplitude distribution [21], the light output from the first collision with deuterium is written as

\[ h_D(l_1, E_0) = H[L_D(E_0(1 - \alpha_D)) - l_1]C_D(E_0) \times P_{Dn}[E_0 - T_D(l_1), E_0] \left| T_D'(l_1) \right|, \]  

where \( T_D(l) \) is the inverse function of \( L_D(E) \). Likewise, the first collision with carbon is given by:

\[ h_C(l_1, E_0) = H[L_C(E_0(1 - \alpha_C)) - l_1]C_C(E_0) \times P_{Cn}[E_0 - T_C(l_1), E_0] \left| T_C'(l_1) \right|. \]  

For higher-order collisions, the light pulse distribution cannot be calculated in a manner similar to the case of the probability distribution for collisions which requires only the neutron energy spectrum from the previous collision, as shown in Eq. 2-10. In order to obtain the total light output induced by an incoming neutron, each collision with either a carbon or a deuterium nucleus must be taken into consideration in an independent fashion. The pulse width of our organic scintillator is on the order of 10+ nanoseconds. Thus, the detector has enough time to aggregate signals induced by a series of collisions of an incident neutron into a single accumulated pulse. In computer simulations, the light amplitude due to an incoming neutron is normally taken as a summation of the light output from each individual collision. This mimics the characteristics of a detector with limited physical size in which all the notable energy transfers occur during a short time generating only a single voltage pulse from a photomultiplier tube. There might be some deviation from this behavior if using peak amplitude for pulse height rather than pulse integral if the time between fast neutron scatters in the detector approaches that of the pulse width. This is especially true when the light generation function is predominantly obtained by single scatter data.
As an example, the light pulse distribution coming from the collision history of DCD, beyond which no further interaction occurs, is given by

\[
h_{DCD}(L, E_0) = \int_0^L \int_0^{L-l_1} h_D(l_1, E_0) h_c(l_2, E_0 - T_D(l_1)) \times h_D(l_3, E_0 - T_D(l_1) - T_C(l_2)) \times W[E_0 - T_D(l_1) - T_C(l_2) - T_D(L - l_1 - l_2)] dl_1 dl_2
\]

(3-7)

As shown in Eq. 3-7 the energy spectrum is given as a convoluted integral, where \( l_1 \) and \( l_2 \) indicate the quantity of light generated in the first and second collision, respectively, and \( L - l_1 - l_2 \) is the light output due to the third collision. It is important to note that due to the conversion relationships of the transferred energy, the pulse distribution for a scattering history with \( n \) collisions is derived from \( (n - 1) \) integrals rather than as a recursive function. As for the total light distribution due to an incident neutron, it is given as the summation of all possible separate collision histories:

\[
h(L, E_0) = h_D + h_C + h_{DC} + h_{CD} + h_{CC} + h_{DD} + h_{CCC} + \cdots
\]

(3-8)

3.2 Light Yield Function

In the measurement of the light output of the Eljen Technology EJ-315 deuterated liquid scintillator, an underground tunnel in the Edwards Accelerator Facility was utilized. The detector was placed at 10 m away from a neutron generating target. The \(^{27}\)Al target was exposed to 7.44 MeV deuterons accelerated by a tandem van de Graaff accelerator to generate neutrons with energies of several MeV [22]. The incident neutron beam was well collimated after passing through a thick collimator wall. This maximized the uncollided flux hitting the detector and reduced secondary interactions and background.
A remote controlled and monitored VME-based high-voltage supply system, manufactured by CAEN, was used to power the EJ-315 detector at 1470 V. A 9821B PMT was used with blue-green sensitive bialkali photocathode on a Plano-concave window. The gain at the supplied voltage is about $10^5$. The data acquisition was performed using a 14-bit, 250 MHz, 16-channel SIS3316 Struck digitizer. In measurements, a pulsed beam with a frequency of 625 kHz and the detector output were connected to the first and the second channels, respectively, and a coincidence mode between them was enabled. The digitizer started saving neutron and beam pulses within a 1600-ns time window only when a signal was observed by the detector and the beam channel in coincidence. The recorded data was then continuously transmitted to a disk through 1000 MBit/s Ethernet communication.

Neutrons were separated from gamma rays using the standard charge-integration based discrimination method [23] [24]. The energy of each individual neutron was characterized by its time-of-flight, and 100 keV wide energy bins were used to categorize neutron energy groups. As reported in Ref [25], the light yield response to recoil protons and recoil nuclei produced from neutrons is a non-linear function of the incident neutron energy. In our measurement, the light yield from neutrons approaches a linear function of neutron energy at high energies. Similarly, the light output from gamma rays is a weakly varying non-linear function for the equipment used in our study. To convert pulse amplitude to units of MeV-electron-equivalent (MeVee), several gamma sources including $^{22}\text{Na}$, $^{54}\text{Mn}$, $^{60}\text{Co}$, and $^{137}\text{Cs}$ have been measured. Additional detail of the measurement is described in Ref. [26]. The light output function of the EJ-315 from measurements with exponential fit is given in Fig. 3-1.
The neutron scintillation light emission function for a 7.6-by-7.6 cm EJ-315 detector. The exponential fit captures the examined data region well.

### 3.3 Result and Discussion

This part of discussion covers models described in Chapters 2 and 3. Deuterated hydrocarbon based scintillation detectors with dimensions 1-by-1, 5-by-5, 10-by-10, and 15-by-15 cm have been investigated using the model. The concentration ratio of D to C is 0.99. The anisotropic elastic scattering distribution and the kinematic limitations in transferred energy leads to interesting energy transfer and light pulse distributions. According to Table 2-2, especially for smaller detector sizes, the probability of the escape dominates all possible collision consequences. For detectors of larger dimensions, high-order collision sequences are more likely to happen than the same order collision histories in detectors of smaller dimensions.
Figure 3-2. Scintillation light pulse distribution for neutrons scattered exactly twice within a detector before escape. Specifically, (a) displays scintillation pulse distributions of different collision histories in a 5 cm diameter detector; (b) shows the scintillation pulse distributions of twice collided neutrons for various detectors sizes. The incident neutron energy is 5 MeV.
Taking a 5-cm diameter detector as an example, collisions (CC, DD, CD and DC) in the scintillation materials help to expand the scattered neutron energy range from the initial stage to a corresponding minimal value which is determined by collision constraints due to conservation of momentum and energy (shown in Fig. 2-3a). As expected, neutrons that collide with deuterium nuclei twice give the widest possible energy range, or equivalently, incident neutrons tend to deposit a larger amount of energy after collisions with deuterium nuclei. However, for collisions only with carbon nuclei (CC), the energy distribution is quasi-symmetrical, but quite narrow due to only a small amount of energy being transferred. As for collision probabilities of both sequences of DC and CD, they show distinct feature differences but end with the same range of scattered energy. In addition, the energy spectrum of the third collision (Fig. 2-3b) reveals that scattered neutrons are primarily composed of low energy ones, especially those neutrons scattered by deuterium nuclei. Thus, from our analytical model, it can be quantified to what extent collisions with deuterium are more beneficial. Those scatters enable a larger energy transferring efficiency that is obtained by depositing more energy of an incoming neutron in the scintillation materials and further initializing considerable signals through the photomultiplier tube. This is a known relationship and the reason for trying to maximize the light nuclei content in many recoil detectors. However the model can easily quantify the potential gain as a function of nuclei mass and concentration ratio. Additionally, it can be used to calculate the effect in more complex compounds containing multiple isotopes and elements of lesser atomic mass separation, or strongly varying double differential scattering cross-sections.
As Figs. 2-4c & d show, after a second collision, a minute extension of the transferred energy range is hard to observe due to the nature of Eq. 2-5. The sharp drop at lower energy is due to the first collision with carbon nuclei, while the sharp drop at higher energy is due to the first collision with deuterium nuclei. By including more collisions, the probability of having more energy transferred increases. However, it does not necessarily improve the light output amplitude. When showing the distribution of all neutrons colliding four times or more, the energy distribution does not change as much as when considering a second or third collision. Since the fourth collision does not contribute a considerable amount to the accumulated collision probabilities, it is often of negligible impact for the scintillation pulse distribution depending on detector sizes.

The analytical model utilizes the light output function in terms of light amplitude which could be changed to light integral without needing any change in the model itself. Fig. 3-2a shows the light pulse distribution generated by various collision sequences. Light pulse spectra from collision histories ending with carbon, like DC, have more characteristics than that from CD stemming from features in the energy-dependent carbon cross-section (Fig. 2-2). According to the neutron-induced pulse distribution (Fig. 3-2a), the pulse spectra reveal that the upper tail of the pulse distribution in calculations following the spectrum peak is caused by the second and following collisions. The light output from CC is included in Fig. 3-2b. It's evident that the signals generated by carbon nuclei are mainly confined to the very lowest part of light pulse distribution, but its probability is significant when compared to the probability of collisions with deuterium nuclei. Such a feature of the pulse amplitude distribution ensures that the light output function measurement of deuterated scintillation materials indeed is the light output from
deuterium nuclei rather than carbon nuclei. It is important to note that the signals generated by different collision-sequence orders of the detector nuclei contribute to different regions of pulse amplitude. The analytical model shows that the shape of the light pulse distribution depends heavily on neutron collisions with both deuterium and carbon nuclei, even though the lighter nuclei dominate the light generation.

In a realistic measurement setup or a post-measurement analysis, a reasonable detection threshold would be applied which inevitably truncates the contribution from collisions with only carbon nuclei, leading to part of the pulse distribution being lost at the low pulse amplitude region. The empirical light output curve for carbon is highly simplified and uncertain due to the lack of measured data, and this could somewhat affect the low light output region. However, it is not the light generated by carbon nuclei, but rather the change in the neutron energy distribution from those anisotropic collision distributions that matters for the overall light pulse distribution.

In analysis, a 100 keV wide energy bin was used to categorize each neutron energy group. For example, the experimental curve of 5 MeV neutrons is comprised of pulses induced by neutrons with energy ranging from 4.95 to 5.05 MeV. This widens the pulse distribution and introduces additional uncertainty to the detector energy resolution. The obtainable resolution of the detector in the measurement is thus limited by statistical and physical processes converting the light to an electronic signal. The analytic model can be reshaped to a realistic measured pulse height distribution by applying a Gaussian smoothing function to match the distribution edge to that of the pulse peak from measurement, as shown in Fig. 3-3, similar to the Gaussian energy broadening that can be applied in MCNP simulations.
Figure 3-3. Scintillation light pulse height distribution for neutrons: (a) shows the total light output from various size detectors; (b) exhibits the pulse height distribution comparison between simulation and measurement for 7.6-by-7.6 cm detector. As for the embedded plot, it has been re-normalized for the amplitude comparison purpose. The incident neutron energy is 5 MeV.
In order to display the measurement distribution and the analytical model result in a comparable scale, the curves have been normalized. The analytical model calculation covers the low pulse height region which has been removed in measurements due to applied pulse height thresholds. The pulse distribution from the model calculation exhibits the characteristics shown in the measurements, however an increased agreement could be achieved by implementing a cutoff mimicking a detection threshold more accurately. In the embedded plot (Fig. 3-3b), it has been re-normalized for amplitude comparison within the range of interest. The proportion of pulse distribution in the lower region is highly affected by the value of coefficient k associated with carbon in Eq. 3-4. A minor change could lead to overestimation or underestimation of the impact of carbon nuclei. The lower light region of deuteron scatters use extrapolation from the measured data, which could also explain part of the discrepancy at low pulse heights.

The impact of detector size is shown to be nonlinear with respect to the collision probability (Fig. 3-2b). A detector with larger dimensions like 15-by-15 cm is more likely to provide higher light output amplitudes. However, the increase at higher light output amplitude is diminishing with increasing size, largely due to the nonlinearity of the light generation function. In the energy ranges where the mean free path is large with respect to the detector material constituents, further increase in detector size continues the increase of interaction probability. The physical phenomena affecting light output from the measurement, such as light self-absorption of the scintillation materials, internal refraction, and the response functions of the photomultiplier and photo cathode, are believed to be the active factors affecting the pulse height distribution by modifying the constants in the exponential semi-empirical function (Eq. 3-2).
For an extremely large-sized detector, the self-absorption phenomena within scintillation materials will begin to play a more significant role, but for the sizes considered here and this type of scintillation material it is rather small. For application purposes, the size of a scintillation detector, the scintillation light spectrum and the cost of material supplies should all be taken into consideration, as well as the cost of the photomultiplier tube, which should ideally match the size of scintillation materials. This can practically be of larger importance even beyond the isotropic collisions and light generation effects examined by this model.

The normal method of choice for computational detector response and studies would be a much more general use Monte Carlo based code. As noted such codes (for example MCNP) only requires the same inputs in form of light response function and differential cross-sections. This also points to the possible use of the model: it does not have the statistical limitations of MC and would thus not require massive computational times for looking at specific collision sequences; the ability to study the neutron kinematics for a specific case is easily done with the model, especially when that event can be outnumbered by many orders of magnitude depending on detector size and compositions. Beyond these special cases for detailed parametric studies of collision kinematics or cross-section manipulations, MC simulation is naturally a better option in general due to its versatility as mentioned. The dependence on light output functions and the ability to apply detector resolutions to the modeled pulse distributions are analogous to that of MCNP, offering a suitable alternative tool for specific studies.
3.4 Summary and Conclusion

Given the fact that elastic scatterings dominate inelastic scatterings within the investigated neutron energy, an analytical model of pulse height distribution and the measurement of the light output function of the EJ-315 detector have been described. In the model, the energy distribution of an incident neutron after scattering was determined according to the impulse approximation and utilizing the neutron double differential cross-sections of carbon and deuterium nuclei. The probability of each collision sequence based on the Peierls formula involving one or multiple collisions has been calculated recursively. The light output function was fitted with a modified exponential formula and its inverse function was used in the analytical model. The model combines the measured light output function with the collision histories to analytically and numerically calculate the estimated signal output distribution of the deuterated scintillation detectors induced by an incident neutron.

This completed model provides a good description of scattered neutrons’ energy and the transferred energy distributions inside various cylindrical dimensional detectors. It shows that the dimension plays an important role in the neutron detecting behavior, and clearly reveals the contribution of the probability of each neutron’s collision sequence. Additionally, the result shows the chance of collisions with carbon nuclei is comparable to that with deuterium nuclei. However, due to the fact that carbon has much lower efficiency of energy conversion to light output than deuterium, the light pulses generated by collisions with carbon nuclei are primarily small. As a result, the light pulse height distribution can be coarsely separated into two characteristic zones. The first part is mainly due to neutron collisions with carbon, and the second part is
mainly due to collisions involving deuterium nuclei. This feature verifies that the light output function from measurements is the light output generated from deuterium nuclei. A comparison study on the 7.6-by-7.6 cm deuterated scintillation detector between the light pulse height distributions of the analytical model and measurements at Ohio University was conducted. A very high probability of an incident neutron generating a low amplitude pulse due to carbon scatters has been observed. Therefore, the contribution of carbon nuclei to the pulse height distribution should be subject to additional investigations to remove the uncertain impact of carbon scatters both in this model and in MCNP simulations. This emphasizes the usefulness of understanding the response of the scintillation materials and extract nuclei information from recorded pulses from a model such as this one. This model is applicable to scintillation materials other than carbon and deuterium, with known angular dependent elastic cross-sections and an estimated light output function for the corresponding scintillation materials.
The pulse shape discrimination (PSD) algorithms based on charge-integration are essential in fast neutron detection. The application of the algorithms is not limited to liquid organic scintillators. When multiple algorithms are being applied to the same amount of data, it leads to a concern of the performance consistency between different algorithms. In this chapter, we investigate two fundamental methods: Digital Charge Integration (DCI) and Pulse Tail Analysis (PTA). Particles are produced by a Tandem Van de Graaff generator with a continuous neutron energy spectrum up to or even higher than 10 MeV. About 30 million pulse events have been recorded with a deuterated organic scintillator detector, EJ-315, operated in a coincidence mode with a 625 kHz trigger beam. Neutron events identified according to DCI and PTA, respectively, are shown for multiple pulse height thresholds, ranging from 13 mV to 0.2 V (15 to 270 keVee). The bending trend of the discrimination curve for PTA at the low pulse height region becomes less important for scenarios with high pulse height thresholds. With constant and predetermined discrimination curves, results show a better agreement between both methods at higher pulse height thresholds. Additionally, neutron and gamma-ray population distributions are obtained based on the relative distance between each event to the corresponding discrimination curve and the coordinate origin, and are fitted with a double-Gaussian function to achieve an optimal $R^2$ using the simplex method. Figure-of-merits (FOMs) are calculated to show the peak separation of neutron and gamma-ray plumes for both algorithms. Results consistently indicate that, considering the relative distances between data to the corresponding
polynomial differential curve and the coordinate origin, DCI gives the best event separation in terms of FOM for the deuterated organic scintillator, EJ-315.

4.1 Application of Neutrons

Since their discovery in 1932, neutrons are a unique and increasingly indispensable tool in broad areas of the physical, chemical, and biological sciences, as well as in materials technology and nuclear medicine [27] [28] [29]. For instance, neutrons have been extensively used to characterize the dynamical properties like flexibility and resilience of bio-systems by referring to mean square displacements of atomic motions and time scales [30] [31] [32]; and they also have been used to study molecular assemblies [32] [33], dispersion, alignment and mixing of nanoscale materials [34] [35], porous structures and inclusions [36] [37], and battery energy states [38].

Additionally, neutron imaging is an important technique for nondestructive testing that is complementary to x-rays, and suitable for visualization of light elements in the interior of heavy objects. Take the exploration of Mars as an example. Mars lacks a magnetic field and has a thin atmosphere. As a result, cosmic rays propagate to the surface and cause the release of secondary neutrons within its subsurface. Thus a significant proportion of high-energy neutrons is moderated in those regions with an abundance of water or ice, inevitably leading to deficits of fast neutrons [39] [40].

However, the nuclear capture cross-sections of thermal neutrons are much larger than fast neutrons, and thermal neutrons cause nuclear reactions, and create products such as protons, alpha particles, gamma rays, and fission fragments that can be detected efficiently [4]. As a result, among those applications, only thermal neutrons with a wavelength in a scale of Å have been widely explored and used. Nevertheless,
the detection of fast neutrons based on moderated ones is not a practical task, since neutrons collide with moderator atoms, in each collision losing only a fraction of their energy: about 50% with H in H$_2$O and 40% with D in D$_2$O. Thus, the volume of moderation material would be considerable.

So far, the detection of fast neutrons is mainly based on liquid organic scintillators due to elastic scatters and the application of pulse shape discriminations, namely the charge integral based pulse shape discrimination algorithm [39] [41].

4.2 Fundamentals of PSD Algorithms

Through elastic scattering with scintillation materials, fast neutrons transfer part of kinetic energy to nuclei and generate fluorescence and phosphorescence lights [3] [12]. A typical pulse consists of a prompt component and a slow, or delayed, component. It has been shown that the relative intensity of the slow component compared to the total intensity depends on the specific ionization of the ionizing particle with which molecules interact. The fundamental principle of PSD algorithms is to integrate the delayed component, due to triple-triple interactions, and to compare it to the total integral of the pulse [42] [43] [44]. The ratio is dependent on the energy loss per unit distance of the particle in the scintillator, and such particle dependent difference is seen in the amount of radioluminescence. So far, the variation of intensity ratio of different incident particles has been observed not only in conventional and deuterated liquid organic scintillation materials [45] [46], but also pressurized $^4$He gases [47] [48] and some plastics [49].

Previously, Gamage et al. investigated the efficiency of pulse gradient analysis, charge comparison method, neutron-gamma model analysis, and simplified digital charge collection based on the measurements using a BC501 (equivalent to EJ-301)
organic liquid scintillator detector [50]. They showed that the simplified digital charge collection (SDCC) has the best discrimination performance in terms of FOM. The SDCC is based on the peak amplitude and a discrimination parameter that is associated with the logarithm calculation of the short integral. However, BC501 has a low flash point of 26 °C, and SDCC doesn't perform well for the EJ-315 detector. Compared to EJ-301, EJ-315 has similar scintillation efficiency, but with a fourfold higher flash point.

This work implements particle identifications based on two PSD methods: DCI compares the tail integral with the integral of the total pulse; while PTA mainly analyzes the delayed decays in the tail, minimizing signal noises. Both methods are based on the basic idea of the charge comparison. The performance of PSD methods is not only evaluated based on the neutron ratios, but also FOMs at different energy thresholds. With predetermined polynomial discrimination curves, incident events are identified with these two methods in Boolean matrices. The same pulse that has contrary indexes with different methods is flagged and the neutron to gamma-ray ratios of the number of differences are carefully calculated. Those ratios are being investigated for different pulse height thresholds.

4.2.1 Data Preparation

The white neutron source provided by the Edwards Accelerator Facility at Ohio University is ideal for this work due to the diversity of pulse beams and the availability of a wide energy range of fast neutrons. For our study, a deuterium beam is accelerated by a Van de Graaff generator and guided by the Beam Swinger Magnet to bombard a $^{27}$Al target.

Neutrons with continuous dynamic energy from a few hundred keV up to more than 10 MeV [20] were emitted. The bore on the concrete collimator wall is about 0.1 m
in diameter, and the distance between the swinger axis to the poured concrete wall is 2.45 m. To obtain proper timing and energy resolutions, a 3-inch by 3-inch liquid organic scintillator (EJ-315) is set 10 meters away from the Al target. The detector assembly contains an inert nitrogen expansion volume to allow for expansion of the liquid because of changing temperature. As reported, the orientation of the detector, leading to the different bubble position, changes the light outputs and shifts the Compton edge [51]. During the whole measurement period, the detector has been stabilized horizontally, lining up with the center of the collimator bore, and powered at 1450 V, by a high voltage system (NDT1470), manufactured by CAEN.

The data acquisition system consists of a SIS3316 digitizer unit (manufactured by Struck), a compact system which has a sampling frequency of 250 MHz. The trigger algorithm implemented in the digitizer uses a moving average. The moving average window can be defined by users. During measurements, the trigger threshold has been set at 12 mV (14 keVee). The number of photons produced during the full absorption of a 1 MeV electron by the scintillation materials is about 9,200. To minimize the interference from background gamma rays due the collisions between fast neutrons and concrete walls and to minimize the data storage, a coincidence mode with a window length of 1600 ns between the detector and the trigger beam with an operation frequency of 625 kHz was enabled. The typical individual pulses of fast neutron and gamma-ray are demonstrated in Fig. 4-1. The record of signal waveform is of great advantage in analysis of the digital features: each pulse consists of 400 samples taken at 4 ns intervals. More information about the digitizer system and system parameters can be referred to our previous work [3] [46].
4.2 Pulse Filter

Considering that the maximum dynamic range of our digitizer is 2 V, we have filtered pulses during post analysis which were clipped. The maximum threshold is set
at 1.95 V. A pulse whose amplitude is greater than the predefined value is eliminated from the total event population.

Figure 4-2. Pulse shape discriminations: (a) DCI, (b) PTA. No software based pulse height threshold has been applied. The black lines are the discrimination curves. The cyan and red regions indicate controversial events.
A good pulse has to have at least 5 data points before its maximum pulse height and 60 data points following the peak, and both the total and tail integrals must be positive. The ratio of the tail integral to the total integral should be reasonable. Meanwhile, a simple pileup rejection algorithm is used to exclude a pulse with abnormal secondary peak. For instance, a case of double pulse is found when a peak in the tail exceeds 10% of the pulse maximum amplitude. With the effect of the trigger algorithm, even though the threshold is 12 mV, the minimum pulse height is 13 mV and the pulse height stabilizes around 18 mV, as shown in Fig. 4-1. The total population of recorded events, including both gamma rays and neutrons by the deuterated scintillator is 29.66 million.

4.2.3 Standard Digital Charge Integration

For DCI, we have calculated the tail and the total integrals of a pulse. Assume $P_{\text{arr}}(i)$ is a valid pulse in the array. Let us define DCI as the ratio of the tail integral to the total pulse integral. Then it can be simply written as

$$\text{DCI}(i) = \frac{\int_a^b p_{\text{arr}}(i)(j) \, dj}{\int_c^b p_{\text{arr}}(i)(j) \, dj}, \quad (4-1)$$

where $a$ denotes the beginning of the tail in a pulse, $b$ the end of the tail in the pulse, which is equivalent to the end point of the pulse, and $c$ is the beginning of the pulse. Defining $\text{Ind}$ as the index of the pulse peak location in the array, and $\text{Lim}$ as the end point of the tail, then we have $a = \text{Lim} + \text{Ind}$; naming $\text{BeF}$ the distance between the pulse initial point, then we have $b = \text{Ind} - \text{BeF}$. Further, by defining $\text{Pas}$ as the distance between the beginning of a tail and the peak, then $c = \text{Ind} + \text{Pas}$. In analysis, the peak can be located, and those defined parameter values are fixed. The ratio of the tail to the total pulse is calculated and sorted to plot a density distribution.
Without any software based pulse height threshold being applied to data, Fig. 4-2a is the output of event distribution based on DCI, within which the bright region (colored in yellow) has a larger population than that of the dark region (colored in blue), and the black curve is a polynomial curve used to differentiate neutron and gamma-ray plumes. The ratio of the tail to the total pulse is calculated and sorted to plot a density distribution.

### 4.2.4 Pulse Tail Analysis

For PTA, we have emphasized the individual decay which causes the emission of photons, leading to an increase of signal because of the production of photoelectrons. Rather than integrating all data points in the tail, we sum the data point which is greater than its previous consecutive data point, and which is larger than a noise value, 1 mV. Here, we are interested in the slope of two consecutive sample points, and the valid tail segment in the pulse. We can express the PTA in the following equation:

\[
PTA(i) = \int_a^b \left| P_{\text{arr}}(i)(j) - P_{\text{arr}}(i)(j-1) \right| \, dj \times \frac{\int_a^b \frac{P_{\text{arr}}(i)(j) - P_{\text{arr}}(i)(j-1)}{P_{\text{arr}}(i)(j)} \, dj}{\int_c^b P_{\text{arr}}(i)(j) \, dj}. \tag{4-2}
\]

Where \( P_{\text{arr}}(i)(j) > P_{\text{arr}}(i)(j-1) \), and \( P_{\text{arr}}(i)(j) > \text{noise} \) in the second part of the equation, otherwise the integrand is forced to be zero at the corresponding location. The slope is calculated in both the first and the second part of the equation. As defined in the conditions, a valid data point requires a positive slope, and the digital value has to be larger than the noise level. The values of parameters in Eq. 4-2 are the same as used in Eq. 4-1. The event distribution based on PTA is shown in Fig. 4-2b without any
software-based pulse height threshold being applied to data. The associated black
curve is the fixed event discrimination criterion.

4.3 Result and Discussion

For convenience, the conversion between the pulse heights in Volt and in MeVee can be treated in a linear manner. However, a nonlinear function can be achieved through calibrations and helps to increase the accuracy of measurements. Previously, we have measured the slightly non-linear response function of our detector system to various gamma rays, emitted by $^{22}$Na, $^{54}$Mn, $^{60}$Co, $^{122}$Ba, and $^{137}$Cs [46]. Units conversion from V to MeVee is the numerical solutions of

$$f = \log(1.0678x + 1) - 0.1968x,$$  \hspace{1cm} (4-3)

where $x$ is the Compton energy, and $f$ is the corresponding pulse height. A clear separation between the neutron and gamma-ray plumes is achieved even with a low trigger threshold (12 mV) for the EJ-315 detector.

The separation is gradually improved by increasing the pulse height threshold, as shown in Figs. 4-2 and 4-3. Noticeably, while the plots show that both neutron and gamma-ray regions with small tail integrals based on DCI are shrinking along with the increase on the pulse height threshold; results based on TPA suggest both the neutron and gamma-ray regions become smaller not only for those with the small tail-to-pulse ratios, but also for those with large ratios. In order to quantitatively evaluate the performance of both methods for the same inputs, we have calculated the index matrices based on the ratios of the tail integral to the total integral. Conflicting events given by two methods are identified. The cyan and red dots flag the conflict events identified with DCI and PTA.
Figure 4-3. Pulse shape discriminations for different thresholds: (a & b) 0.05 V (60 keVee), (c & d) 0.10 V (124 keVee), and (e & f) 0.2 V (270 keVee).
The cyan dots denote events identified as neutrons according to DCI, while as gamma rays according to PTA; on the contrary, the red dots denote events identified as gamma rays according to DCI, while as neutrons based on PTA. Without a software based pulse height threshold being applied, the total valid event population is given as 29,659,246, within which 19.05% and 13.98% are neutrons according to DCI and PTA, respectively. Interestingly, the disagreement is mostly lining up with the differential curve, especially for plots of DCI. Surprisingly, even though the cyan colored region seems to be very narrow, it includes 1.5 million controversial events. However, only about 150 controversial events are counted in the red region. We name the particles that are identified as neutrons according to both PSD methods as regular neutrons, and that with controversial events as misidentified neutrons. We further calculate the ratios of the regular neutron and the misidentified neutron to the total population, and the ratios are sorted by the total pulse integral to achieve a histogram distribution, as shown in Fig. 4-4. The controversial ratio decreases dramatically along with the increase of the value of total pulse integral, which reassures that the performance of PSDs is better for pulses with larger pulse height. According to Fig. 4-5, the neutron number, or the ratio given by DCI, drops significantly, and both methods almost give identical results at a pulse height threshold of 270 keVee. The zoomed view of Fig. 4-4b verifies that a good agreement on the regular neutrons in the high total integral region between two methods has been achieved. Thus, even though the overall event differential agreement in the low value region of the ratio of the tail can be significantly improved by bending down, rather than up, the beginning part of the
discrimination curve associated with PTA, the shape of the discrimination curve is less important in event identification in a case with a higher pulse height threshold.

Figure 4-4. Quantitative analysis on regular neutrons and controversial neutrons based on DCI and PTA. (a) No additional pulse height threshold applied other than the measurement trigger threshold, 12 mV, and (b) 0.2 V (270 keVee), respectively. The histogram distributions have been normalized.
To evaluate the separation of neutron and gamma-ray plumes, the distributions of tail-to-total pulse ratio with or without considering the relative distance to the curve and the origin are given in Fig. 4-6. Namely, Fig. 4-6a shows the event distributions of the tail-to-total pulse ratio, calculated based on DCI, and the ratio only considers the separation based on a linear discrimination curve; Figs. 4-6b & c are the event distributions considering the point distance to a polynomial function and the distance to the coordinate origin. For each histogram distribution, the bin number is 2000, a number large enough so that even without data smoothing, the variation on the shape of the event distribution is invisible. The corresponding FOM value for each threshold is calculated in the following:

\[
FOM = \frac{\text{Peak separation}}{\text{FWHM}_r + \text{FWHM}_n}. \tag{4-3}
\]

Figure 4-5. The performance comparison between DCI and PTA methods. The neutron numbers and ratios are shown as a function to the threshold.
Event distributions are fitted with a double Gaussian function with an optimization function of \(1-R^2\), and we have:

\[
R^2 = 1 - \frac{\sum (y_i - f_i)^2}{\sum (y_i - \bar{y})^2},
\]  

(4-4)

where \(y_i\) is the known data set with an average value \(\bar{y}\) and \(f_i\) the predicted value. With six parameters, we have achieved the best fitting using the simplex algorithm [52] [53].

---

Figure 4-6. Event population distributions with various pulse height thresholds: (a) Single ratio, (b) DIC, (c) PTA. (d) The event distribution based on DCI without applying additional software based pulse height threshold.

The better the double Gaussian fits the data, the closer the value of \(R^2\) is to 1. The fitted curves are plotted in red, as shown in Fig. 4-6, and the results show that the majority of
fittings capture the distribution peaks quite well, except for the distribution with pulse height threshold of 15 keVee (the measurement trigger value) (Fig. 4-6d).

Figure 4-7. Fitting correction and FOM comparison. (a) New fitting for the event distribution with a threshold of 15 keVee based on DCI, and (b) FOM comparison among three methods.
The main reason for this issue is that while achieving an optimal $R^2$ the fitting sacrifices the precision on the neutron peak by capturing the edge of the secondary peak. By definition, $R^2$ is the fraction of the total squared error that is explained by the distribution, and can be a lousy measure of goodness-of-fit, as in the case mentioned previously. To achieve a better fitting, we have forced the fitting to capture the secondary peak, while, at some degree, we have compromised the value of $R^2$. The new corrected result is given in Fig. 4-7a. FOMs considering the separation based on a linear or a polynomial discrimination curve have been summarized in Fig. 4-7b. The error bar has been estimated with one standard deviation of uncertainty. It consistently shows that the separation between the neutron and gamma-ray regions have been improved when the input threshold increases. Among them, for the deuterated organic scintillator, the DCI method gives the best event discrimination performance in terms of FOM at various pulse height thresholds. Meanwhile, it reveals that the improvement on FOM by increasing the pulse height threshold is more effective at low value thresholds (<124 keVee) than at high pulse height thresholds (>124 keVee). When the threshold reaches 352 keVee, the change on FOM is insignificant based on DCI, while that based on PTA is larger.

4.4 Summary and Conclusion

In this chapter, two fundamental charge-integration based algorithms (DCI and PTA) used in fast neutron detection were investigated. DCI compares the tail integral with the integral of the total pulse, while PTA mainly focuses on the delayed decays in a tail rather than the full tail integral. The input pulse data were obtained from measurements using the deuterated organic scintillation material. To obtain the neutron energy source with energy up to 10 meV, the Van de Graaff generator at Ohio
University was used. Based on measured data, the consistency between DCI and PTA was investigated. Results show that the number of controversial events decrease dramatically along with the increase of the pulse height threshold from 15 to 270 keVee, and that eventually DCI and PTA give the almost identical event discrimination ratio when the threshold reaches 270 keVee. Meanwhile, event population distributions have been calculated and fitted with a double Gaussian function with an optimal R². FOMs suggest that when taking the relative distance from a data point to the polynomial differential curve and the coordinate origin into consideration, DCI provides the best event discrimination result for the deuterated organic scintillation detector.

In addition to the standard digital charge integral method, we recently have expanded the two-lifetime description of pulse shape formation in organic scintillators to a high-order statistics in the form that is similar to the definition of the Fisher-Person standardized moment coefficient. The method is much more sensitive to photons possibly generated directly by the interaction of photocathode while still separating fast neutrons and gamma rays. During the interaction, one or more electrons are multiplied by secondary emission to a level whose voltage surpasses the threshold in the digital acquisition system and consequently trigger the fast photons. The work will be further studied in the near future.
CHAPTER 5
PULSE HEIGHT MEASUREMENTS AND RESOLUTIONS OF EJ-315

In this chapter we investigate the light response to fast neutrons and estimate the pulse height resolution of a deuterated liquid organic scintillator, EJ-315, considering the detector's non-linear light response to gamma rays. Initially, collision data and a neutron beam trigger are recorded in coincidence mode, and incident neutron energy is calculated with a time-of-flight technique. Fast neutrons are further discriminated from gamma-ray background based on the scintillation material decay patterns using a pulse shape discrimination algorithm. A light response matrix composed of multiple neutron energy and their corresponding light outputs is derived. The pulse height resolution property of the EJ-315 is characterized utilizing the derivatives of the pulse height distributions with corrections of the measurements’ setup uncertainties. Additionally, the EJ-315's pulse height resolution is also characterized by comparing the smoothed derivatives of quasi-monoenergetic neutron pulse height distributions, given by the Peierls-formula-based analytic model, to match the measurement data. Results show rather consistent 10 - 13% pulse resolution for mono-energetic neutrons with kinetic energy above 2 MeV. The resolution decreases slightly with an increase in neutron energy indicating the improved resolution performance of EJ-315 in the higher energy events.

5.1 Background

Due to intrinsic properties of liquid scintillators, pulse shape discrimination (PSD) and time-of-flight (TOF) are presently used in fast-neutron detection [54] [55]. Compared to deuterated benzene-based detectors, regular aromatic hydrocarbon based detectors like EJ-301, EJ-309 and NE-213 have relatively higher light outputs
higher collision probabilities and better capabilities at depositing a larger proportion of neutron energy in elastic scatters [2]. These features are mainly due to the existence of hydrogen which is frequently used as one of the basic components of neutron shielding materials [57]. The efficient moderation of fast neutrons by measuring neutron spectra makes the exploration of hydrogen abundance at subsurface of Mars a feasible option [40]. However, according to the study [14], deuterated liquid scintillators have excellent n-gamma discrimination properties and better performance in spectra unfolding because of n-d scatters, which are anisotropic and favor backwards and forwards scattering, generating more features in measured pulse height distribution as a function of neutron energy. This compensates for its slightly inferior performance in view of light output and indicates the usage for de-convoluting pulse height spectra without needing to have a long flight path and a time-of-flight setup. These aspects potentially bring new possibilities for fast neutron detection within a small area and make applications of fast neutrons in neutron imaging and nondestructive testing more feasible [58] [59].

Continuous neutron spectra can be categorized as multiple quasi-mono-energetic groups if their time-of-flight information is available. Gamma rays interacting with scintillators may give up all or part of their energy in single events through the photoelectric effect, Compton scattering and pair production. The detection of fast neutrons on the other hand is based on elastic scatters during which only a limited fraction of the neutron kinetic energy is transferred to the scattered heavy nuclei. Fast neutrons can only deposit partial energy in an individual collision with materials encapsulated in a deuterated scintillation detector. In the case of deuteron nuclei, the
maximum fraction transferred in a single collision is 8/9 of the incident neutron energy. Even if multiple collisions with an incident fast neutron are involved, the maximum light output is not guaranteed due to possible collisions with carbon rather than hydrogen isotopes. Single collisions normally dominate all the collision sequences depending on the detector size [3]. As a result, the fast neutron pulse height distribution (PHD) lacks full energy peak, and the conventional resolution function associated with the full width at half maximum (FWHM) and the corresponding peak energy [60] is not appropriate to be adopted to characterize the resolution of the deuterated organic liquid scintillator, EJ-315.

As pointed out [61], the last minimum in the derivatives of a neutron pulse height distribution denotes the proton recoil events corresponding to the maximum incident neutron energy. The detector energy resolution can be represented and determined by the FWHM of a fitted Gaussian for the minimum of the derivatives. This approach has been well accepted for conventional EJ-309, non-deuterated organic scintillators [20] [62], which is different from the typical method used to characterize the energy resolution of an inorganic scintillator based on the full energy peak measured.

The light response matrix provides a measure of how well the detector transfers incident particle energy to detectable signals. The light response matrix affected by factors like quantum efficiency is also essential to evaluating other key detector performance metrics such as detector resolution. This work applies a standard charge-integration-based algorithm to discriminate neutron events from gamma rays, and obtains the neutron energy based on its TOF spectrum. The light output function in terms of the leading edge of the pulse height distribution of each quasi-monoenergetic
neutron group is measured. Both the pulse peak and the pulse integral are evaluated, respectively, to obtain the optimal result for the conversions from V to MeV electron equivalent (MeVee) considering that the deployed detector has a nonlinear light response to gamma rays. The light response matrix is derived. Additionally, we investigate the EJ315's energy resolution by achieving optimal fittings of the minimum derivatives of the theoretical neutron pulse distributions with Gaussian spread functions. The pulse distribution model used takes the neutron elastic double differential cross-section into consideration, and analytically and numerically simulates scintillation pulse height distributions for quasi-monoenergetic neutrons based on the Peierls-formula convoluted with the material light output function from measurements [3].

5.2 Measurement Setup

Detector calibration and measurement were performed with a 10 μCi $^{137}$Cs source and a continuous neutron flux, respectively at Ohio University. The Edwards Accelerator Facility contains a Tandem Van de Graaff Generator along with a well-shielded 30-meter long underground tunnel which allows us to set the deuterated organic scintillation detector at a distance such that neutrons are well-categorized in energy by time-of-flights with digital high-precision timing. In this configuration, the front central surface of the alumina cell of the applied detector was placed at a 10-meter distance from the neutron generator which emits fast neutrons with a broad energy range by bombarding a thick $^{27}$Al target with 7.44 MeV deuterons [22]. Counting uncertainties arise from the fact that radiative decay is a random process, and thus any measurement based on observing emitted nuclear decay is subject to statistical fluctuations. In order to reduce background caused by cosmic rays and other undesirable radioactive sources, the experiment was completed in the underground
tunnel, as shown in Fig. 5-1. The setup figure is not to scale considering the fact that the detector is very small when compared to the 10-meter distance between the detector and the collimator wall. The bore on the concrete collimator wall is about 0.30 meters in diameter. The approximate distance from the swinger axis of rotation to the poured concrete wall of the tunnel is about 2.45 meters.

Figure 5-1. Simplified sketches: a) of the facility in Edwards Accelerator Laboratory, showing the measurement tunnel that was utilized in this research in the right upper corner, and b) EJ-315 setup toward the collimator and the neutron source (not to scale).

To avoid gain variations that could inevitably give rise to fluctuation and bias in pulse height distribution, the photo-multiplier was consistently powered at 1470 V by a remotely controlled high-voltage power supply (NDT1470), manufactured by CAEN with a maximum voltage ripple less than 30 mVpp. Generated neutrons are emitted in all directions with an angular anisotropy that is a function of the incoming deuteron beam direction. The incident neutron beam was selected by guiding it through a thick collimator wall toward the detector’s front surface (Fig. 5-1b). Thus, the majority of neutrons emitted in other directions were eliminated, which otherwise could have added a significant amount of signal uncertainty due to neutron scattering against the tunnel's
concrete walls and resulting in a significant change to the incident neutron energy spectrum.

The detector consisted of a highly purified deuterated benzene-based material, with a D to H ratio of 141:1. It has 60% of the light output of anthracene, a wavelength of maximum emission of 425 nm and a time constant of fast decay in the order of nanoseconds. The detector used was cylindrical, and of identical dimensions in diameter and height (7.36-by-7.36 cm). The cell was encapsulated in a nitrogen environment to minimize the light quenching effect due to oxygen. Photons are emitted while ionizing particles pass through the scintillation medium. The scintillation light is first absorbed by a blue-green sensitive bialkali photocathode, and then converted into an amplified electronic signal through an ET Enterprises 9821B PMT.

The data acquisition system is a SIS3316 digitizer unit manufactured by Struck with a sampling rate of 4 ns. It contains a high resolution (14 bits) analog-to-digital converter (ADC) and a field-programmable gate array (FPGA). Each ADC FPGA group (4 channels) has two DDR3-memory-chips with 256 Mbyte each, and 128 Mbyte for an individual channel. During measurements, the internal gate generation for coincidence between channels 1 and 2 was enabled. The two channels were connected to a 625 kHz beam pulse and the EJ-315 digital output, respectively. The coincidence gate length was set to 1600 ns to match the repetition frequency of the neutron generator. The coincidence setup ensured relevant data and minimized disk memory needed during irregular generator operations. Accidental coincidences were mostly generated by gamma rays from the activated aluminum target and contributed only negligibly to the neutron data due to TOF and PSD cleaning. The detector signals were digitized by
the ADC, and compared to the threshold value predefined in a trigger logic. The trigger algorithm implemented in the digitizer utilizes a zero-crossing mechanism with comparison of two moving average windows. The length and the separation of the averaging windows can be user defined to fit the shape of the detector pulses. Pulses were stored with additional relevant information including the specific channel number and recorded time. The data were then transferred from memory to a PC through Gbit Ethernet communications.

5.3 Response Matrix Measurement

5.3.1 Detector Response to Gamma Rays

The response of an organic liquid scintillator is dependent on the energy transfer of the detected charged particle. For instance, gamma rays and neutrons can be detected by creating recoil electrons and recoil nuclei, respectively. The response of the organic scintillator is notably nonlinear to deposited particle energy for charged particles of mass higher than electrons [63] [64]. Non-linear PMT gain or digitizer response can contribute to an observed non-linear response also to gamma rays (electrons) for the detector system. Thus the response calibration is necessary not only for interpretation of the measured neutron pulse height spectra, but also for gamma-ray response. In order to acquire the energy dependent conversion ratio of the pulse amplitude in V to MeVee, several gamma-ray sources including $^{22}\text{Na}$, $^{54}\text{Mn}$, $^{60}\text{Co}$, and $^{133}\text{Ba}$, and $^{137}\text{Cs}$ have been measured multiple times under the same laboratory conditions. Each calibration measurement was taken such that the Compton edge of a pulse height distribution was well-defined. The detector's responses to gamma-ray sources are shown as pulse height distributions (Fig. 5-2). The electron equivalent detector output
describes the relationship between energy deposited by incident gamma rays and the observed responses of the scintillator and neutrons.

Figure 5-2. Gamma-ray pulses from the EJ-315 neutron detector consistently powered at a voltage of 1470 V.

The light response is normally described as a function of the pulse amplitude at a specific Compton edge such as that of $^{137}$Cs. The resolution, detector size and gamma-ray energy all have impacts on the perceived position of the Compton edge vs. the real position, regardless of taking the Compton max or half-height [64]. In our analysis, the pulse amplitude value at the half height of the pulse peak of the upper part was used as the light output of the corresponding energy.

However a gamma-ray source whose energy is greater than 2 MeV is rarely available in normal laboratory conditions. The calibration of electron equivalent response into the high-energy region beyond the energy of the used gamma-ray sources is based on the extrapolation of data. As a result, this method possibly overestimates the response of the organic liquid scintillator leading to inadequate
neutron light output at high light pulse outputs. The neutron pulse height data used is in or near the calibrated electron response range. During offline analysis, we compare the detector response from the digital pulse-peak amplitude to that of the corresponding pulse integral response. To ensure response curves anchored at origin\((0,0)\), the data is fitted to a function with a nonlinear component in the form of \(f(x) = \log(a \cdot x + 1) + c \cdot x\), here the input \(x\) is Compton energy of an incident gamma-ray.

Figure 5-3. Measured response functions of the EJ315 organic liquid scintillator to electron energy in terms of the pulse amplitude and the pulse integral. Close circles at the same Compton energy were due to multiple measurements.

As Fig. 5-3 reveals, the measured response in view either of the pulse amplitude or the pulse integral is a non-linear function to Compton energy for the detector setup used here. Thus when transforming the deuterated scintillator's response matrix in units of V to MeVee a pulse-height dependent formula needs to be used rather than a single conversion value as commonly used in post-analysis. The uncertainty of the measurements based on the position of pulse peak was found to be less than 0.7%.
The pulse integral uncertainty is lower mostly due to the random nature of the signal noise of around 1 mV$_{pp}$. It is more susceptible to baseline bias compared to the pulse amplitude, and baseline evaluation was performed to mitigate that. Fig. 5-3 shows a consistent performance of our detection system for both cases.

5.3.2 Detector Response to Fast Neutrons

Neutrons and gamma rays are categorized using the pulse shape discrimination technique, whose principle has been thoroughly studied and reported [42]. Unlike charged particles which produce signals in proportion to its kinetic energy, neutrons only deposit part of their energy in scattering. Thus it is impossible to determine neutron energy simply based on the amplitude of the detected light pulse. To obtain an accurate estimate for neutron energy, time-of-flight data was used.

Figure 5-4. Neutron pulse height distributions in Volts for various neutron energies before MeVee conversion. The bin width is 100 keV. The data for neutrons with kinetic energy of 0.55 to 0.65 MeV is labelled as 0.6 MeV. Arrow $a$ denotes the trend of how pulse edge propagates from low to high energy neutron pulses, and arrow $b$ denotes the contribution from scattering between incident neutrons and hydrogen-nuclei lighter than deuterons.
The energy range of 0.55 to 10.05 MeV was measured, and the energy binning width was 100 keV referring to neutron energy TOF windows. The number of neutrons with higher energy is less than that of neutrons with lower energy due to the neutron energy spectrum [14]. Therefore the pulse height distribution of each neutron energy was binned with varying numbers of amplitude bins. At higher energies the features are wider and adequate identification of the PHD features can be achieved with fewer bins. For the visualized PHDs 300 pulse height bins were used for consistency.

Figure 5-3 shows that the light output response is non-linear to the gamma-ray energy. Similarly, according to the asymptotic line, notated as the arrow \( a \) (Fig. 5-4), the light yield is slightly non-linear with respect to neutron energy. In addition, the light output increase due to multiple collisions is small compared to the primary pulse amplitude [3]. The original deuterium collision is able to transfer up to 8/9 of the neutron energy, and the additional light from multiple scatters that specifically involve deuterium rather than carbon nuclei only generates a minor extension of the PHD because of low probabilities. Each PHD extends further in amplitude as indicated by the arrow \( b \). These extensions are consistently about two orders of magnitude lower than the counts of the corresponding maximum pulse amplitude. Regular hydrogen has higher light yield than deuterium, and the amount of hydrogen in the active cell is about 140 times less than that of deuterium. Thus this extension seen is not related to the deuterium scattering but rather to the small hydrogen content. The effect of hydrogen nuclei on the light output of the deuterium in EJ-315 content is negligible for analysis purposes, but it is still clearly noticeable in the logarithmic scale in Fig. 5-4.
Figure 5.5. Response matrix: (a) neutron pulse height distribution based on pulse peak amplitude value after conversion to MeVee from Volt, and (b) is equivalent to (a) but in a 3-dimensional view. The maximum neutron energy shown is 8 MeV.
Figure 5-6. Light output curve comparison before and after electron-response conversion correction: (a) shows the light output curves from pulse integral and maximal pulse; and (b) is the counterpart of (a) after conversion. The deuterium energy is the transferred energy of a neutron that underwent elastic scattering on deuterium nuclei.
The Compton edge response of the detector to various gamma-ray energies was used to calibrate the electron-equivalent response for the detector system. After the pulse amplitude conversion from V to MeVee, the neutron pulse height distribution is shown in Fig. 5-5. The changes of PHDs are demonstrated in Fig. 5-5b showing the full response matrix as a function of incident energy and pulse height.

Figure 5-7. The light emission function for the 7.6-by-7.6 cm EJ-315 detector. The exponential function captures all the pulse region very well, and data colored in red is EJ-315 light output with a detector of 11.43 cm diameter and 2.54 cm thickness [18].

The MeVee amplitude has been stretched out because the ratio, given by the reciprocal of the gradient of $\log(1 + 1.0678 x) - 0.1694 x$ (Fig. 5-3), is greater than unity within the investigated energy range. To evaluate the accuracy of the conversion, the deuterated organic scintillator’s light output based on the pulse peak amplitude as well as the pulse integral are given in Fig. 5-6. Figure 5-6a shows a close agreement between the light outputs in terms of V and V-ns before the light conversion. However, a
disagreement is observed after the conversion for neutrons with initial energy greater than 6 MeV (Fig. 5-6b). This disagreement is partially in the extrapolation range of the gamma-ray calibration source data. It is further partially caused by not having a significant delayed light portion for the gamma rays, which the neutrons do exhibit. This discrepancy is of smaller effect when using the peak pulse amplitudes rather than pulse integrals, when comparing neutrons and gamma rays.

Figure 5-8. Pulse height distributions after conversion to MeVee and their corresponding derivatives. Gaussian functions were applied to fit the minimum of the derivatives. The data is from 3.40 to 4.90 MeV (from left to right) with an energy width of 0.2 MeV.

Despite the disparity in the higher neutron energy section, the conversion from V to MeVee for neutrons with incoming energies less than 6 MeV is not a concern. In measurements the light induced is comparable to that induced by 6 MeV neutrons. This reduced energy upper limit is beneficial for limiting the effect of deuterium breakups in the high energy region. The light output function with the exponential format is given in Fig. 5-7. The result shows a similar trend to the measurement completed by
[18], but is consistently slightly lower than their results. The main differences are expected to come from the size and shape of the deployed detectors [20], which was thinner and more optimized for light collection with a larger diameter PMT than the size of the detector cell.

![Graph showing pulse shape distributions of 5 MeV neutrons from measurements and the analytic model calculation.](image)

**Figure 5-9.** Pulse shape distributions of 5 MeV neutrons from measurements and the analytic model calculation. 10.0% resolution has been applied to smear the sharp edge of the spectrum. The actual neutron energy covers 4.95 to 5.05 MeV. All abbreviations of legends (from top to bottom) are explained in the following: analytical PHD, measured PHD, smoothed analytical PHD derivative, smoothed analytical PHD, analytical PHD derivative fitting, measured PHD derivative, and measured PHD derivative fitting.

The energy resolution of a scintillation spectrometer is a measure of the ability to distinguish a specific category of particles of various energies. Rather than looking at the maximum pulse edge, the first order derivatives of PHDs for quasi-monoenergetic neutrons with an energy range of 0.6 to 6 MeV are investigated. Among them several quasi-monoenergetic groups are plotted in **Fig. 5-8**. It shows that the PHDs do not exhibit sharp features that can only be seen in ideal (perfect resolution) pulse height
spectra like those (colored in red) derived from the analytical model as exhibited in Fig. 5-9 and Fig. 5-10. The main difference is the detector resolution observed in measurements.

Gaussian fittings have been applied to the minimum of the derivatives, which capture the PHD features reasonably well. This approach gives not only a peak position of the pulse amplitude, but also the variance and the full width at half maximum [61]. Corrections based on the time of the flight uncertainty and the detector's responses are applied with the following equation:

\[
\frac{dE}{E} = \sqrt{\left(\frac{2 dt}{l}\right)^2 + \left(\frac{2 dl}{l}\right)^2}.
\]  

(5-1)

Figure 5-10. Pulse shape distributions of 2 MeV neutrons from measurements and the analytic model calculation. 13.0% resolution has been applied to smooth the analytical spectrum. The actual neutron energy covers 1.95 to 2.05 MeV. The abbreviations of legends are the same as used in Fig. 5-9.

where \(dt\) is the time resolution of the deuterated scintillator, \(l = 10\) m is the flight path length, and \(dl = 7.6\) cm is the flight path spread due to the detector size. The recording
time interval between two consecutive data points is 4 ns. To analyze the time resolution of our detector, two detectors were used to measure a cobalt-60 source which emits two gamma rays simultaneously in coincidence mode. The detector's timing resolution can be characterized as the timing differences between the two detectors' responses to two simultaneously emitted gamma rays. The detector's time resolution was about 1.5 ns, giving a more detailed timing than the sampling rate. In addition, we applied a correction to the energy due to the neutron energy-bin width, which worsens the resolution of the measured data and affects the Gaussian resolution applied to the analytic PHD. The resolution curves both before and after correction are plotted in Fig. 5-11. The error bars are derived from fitting uncertainties. For the low energy region the insufficient data and large bin widths generate large uncertainties.

Figure 5-11. Deuterated scintillator EJ315's resolutions before and after corrections. The triangles denote the obtained resolutions for the optimal results using the analytical PHD comparison for monoenergetic neutrons of 2, 3, 4, and 5 MeV with energy width of 0.1 MeV.
The calculated light output resolutions are fitted with the following function [61] [65] to evaluate the effects on the light output of several components including the scintillation material medium and the PMT used in measurements:

\[
\frac{dL}{L} = \sqrt{\alpha^2 + \frac{\beta^2}{L} + \frac{\gamma^2}{L^2}}
\]

(5-2)

where \( \alpha \) describes the locus-dependent light transmission from the scintillation materials to the bialkali photocathode, limiting the resolution of the detector at high pulse heights; the stochastic term \( \beta \) is the statistical variations of the light relating to photon production, attenuation, photon to electron conversion and electronic signal amplification; and \( \gamma \) represents the noises from both the PMT and the data acquisition system. In this study, \( \alpha = 0.161 \), \( \beta = 0.087 \), and \( \gamma = 0.033 \) are the fitted parameters and comparable to the parameters calculated by Enqvist et al. [20] for a conventional 7.6-by-7.6 cm EJ-309 detector in terms of detector resolution.

It is important to notice that even after applying the convolution of the smoothing function to the PHD from our analytical model, the pulse at the lower light output region (Fig. 5-9) is much higher than that of the measured PSD. It is mainly due to the small light output accumulation because of neutron collisions with carbon nuclei, and the uncertainty in the carbon light value used in the model (seemingly overestimated vs. that observed here in EJ-315). These collisions have comparable probabilities to those involving deuterium nuclei [3]. However, the light output from hydrogen isotopes is about 10 - 50 times larger than one from carbon depending on the range. As expected, when the incident neutron energy decreases from 5 to 2 MeV, a corresponding increase in resolution is observed, as shown in Fig. 5-10. The deuterium nuclei dominate photon production when neutrons interact with the organic scintillation medium. Therefore, the
light output shown in Fig. 5-7 from our measurements can indeed be considered as the light output generated by deuterium.

![Graph showing error vs resolution](image)

Figure 5-12. Calculated error between fitted curves with different resolutions and measured pulse shape distribution at neutron energy of 5 MeV.

The derivative resolution described above indicates the full measured detector system and is sensitive to a diffuse upper range of energy transfer caused by neutrons not being able to deposit their full energy in a single collision. In order to investigate the intrinsic light output resolution of EJ-315, an algorithm applying a Gaussian smoothing function is proposed to find optimal fittings. Mathematically, a Gaussian spreading function is convoluted with the pulse height distribution from the Peierls-formula-based analytical model [3] and the fitting error is defined as the difference of FWHMs of the minimum derivatives from our analytical model and from measurements. Specifically, the optimization function is written as the following formula:

\[
\min \left[ \sqrt{(l_{it} - l'_{it})^2 + (l_{ir} - l'_{ir})^2}, \ i = 1,2, \ldots, n \right],
\]  

(5-3)
where $l_{il}$ and $l'_{il}$ are the lower light amplitude changes at the FWHMs of derivatives of measured PHD and of analytic PHD, respectively; while $l_{ir}$ and $l'_{ir}$ are the upper light amplitude changes at FWHMs. The resolution step length is 0.5%. For neutrons with kinetic energy of 5 MeV, the error of different applied resolutions, as defined in Eq. 5-3 is shown in Fig. 5-12.

The reason why this algorithm refers to FWHMs of derivatives rather than the directly measured PHDs as reported by [65], is that the low light output regions of measured PHDs are lost due to the applied detection threshold. The resolution value is only relevant near the maximum transferred energy since it is highly dependent on the deposited energy. For PHDs containing many distinct features one could envision fitting a full energy-dependent resolution-smoothing to the analytical model. In this case it is applied only to the upper edge due to the PHD features there. The low pulse height region is also best avoided due to large background influence in the measurements and significant carbon-light output uncertainty which affects the analytical distributions significantly.

As indicated by green triangles in Fig. 5-11, the convolution obtained resolutions around 10-13% are rather consistent for neutrons with energy of 2, 3, 4 and 5 MeV. With increasing neutron energy the resolution decreases slightly. If the same mechanism used for the derivative resolution correction were applied, the final resolutions would change about 1-2%, depending on the incident energy. The trend is similar to the resolutions obtained purely based on minimum derivative fittings of the measured PHDs. Finally, it is important to note that a more thorough study of the analytical model that considers inelastic collisions between neutrons with scintillation
materials would also help to limit the error in resolution evaluation, especially when applied to high energy neutrons.

5.5 Summary and Conclusion

In this chapter we have investigated the light responses of a 7.6-by-7.6 cm deuterated EJ-315 detector to several gamma-ray sources and a white neutron source. The detector response was analyzed using both pulse peak amplitude and pulse integral values. The light output induced by fast neutrons has been converted from V to MeVee based on both the pulse amplitude and the pulse integral. An equivalence was found with regards to using either pulse peak amplitude or pulse integral for the PHDs from the digitized pulse data in the relative low energy region. Due to the disparity of light outputs after conversions, the light output conversion is upper limited to 6 MeV neutrons. This was also done to limit the impact of deuterium breakup induced by high-energy neutrons.

The energy-dependent resolution based on the PHD-derivative minima of PHDs was calculated for our digital acquisition system. The detector’s resolution parameters including $\alpha$, $\beta$, and $\gamma$ have been calculated for the EJ-315 detector with values of 0.144, 0.094 and 0.032, respectively. Results indicate that EJ-315 is comparable to EJ-309 in view of detector resolution. Additionally, a new method for the intrinsic light output resolution has been proposed by matching the FWHMs of the minimum derivatives of convoluted analytic PHDs to those of the measured PHDs. A rather consistent and expected resolution result has been observed for the EJ-315. The detector resolution is among 10-13% for neutrons with an energy range of 2-5 MeV.
Previously, we have discussed the analytical models of neutron collisions, pulse height distributions and the implementations of pulse shape discrimination algorithms and pulse height measurements. Molecular states transitions generated by fast neutron and gamma-ray are different. The difference is caused by the decay processes. The mathematical model used in decay time calculation is adapted from the analysis of the pulse shape in plastic scintillator implemented by N.P. Hawkes [67].

6.1 Decay Time Measurement

This chapter continues the work described in the previous sections and data from early measurements are used. To quantitatively investigate the scintillation properties of a deuterated scintillation detector, EJ-315, average pulses of several groups of neutrons with different energy are analyzed. A mathematical model consisting of exponentially modified Gaussian distributions has been applied to fit scintillation pulses. Reasonable fittings with convergence during optimal parameter searching were achieved by a simplex algorithm method. The short and the long decay time constants were measured as 7.75 ns and 63.25 ns, respectively. The reasons for measurement discrepancies with declared values are discussed in the subsection of measurement setup within Chapter 5.

6.2 Mathematical Model of Pulse Shape Distribution

To quantitatively analyze the characteristics of the deuterated scintillator materials, a mathematical model to fit the regular neutron pulse shape is necessary. The response of a PSD-capable scintillator has been described as two types of excited states, one significantly shorter lived than the other. PSD is possible because of gamma
and neutron interactions producing the two excitation states in different proportions. In an ideal case, the decay rate of short-lived singlet states (short-lived states) is treated as radioactive isotopes, and the distribution model of the production of scintillation light is thus proportional to its activity [66]:

\[ L(t) = \frac{A}{t_1} \exp \left( -\frac{t}{t_1} \right) + \frac{B}{t_2-t_1} \left( \exp \left( -\frac{t}{t_2} \right) - \exp \left( -\frac{t}{t_1} \right) \right), \]  \hspace{1cm} (6-1)

where \( t_1 \) is the time constant for the short-lived state, \( t_2 \) for the long-lived state, and \( A \) and \( B \) stand for the weight of the light due to the short-lived and the long-lived types at \( t = 0 \), respectively. The first term in Eq. 6-1 represents the decay of the initial population of the short-lived types to the ground states, while the second term convoluting an exponential component represents light production due to the de-excitation from the long-lived types to the short-lived type states. However, in a realistic scenario, a detector response takes accumulated time consumed in multiple steps, including transformations of atomic states due to collisions, recoil neutron and photon movements inside a scintillation material volume, and the light absorption and electron emission processes in a PMT. Signal production and transmission with associated distortion and delay can be represented by an exponentially modified Gaussian distribution (ex-Gaussian) [67] [68]:

\[ f(x|x_0, \mu, \sigma) = \frac{1}{x_0} \exp \left( \frac{\sigma^2}{2x_0^2} - \frac{x-\mu}{x_0} \right) \phi \left( \frac{x-\mu}{\sigma} - \frac{\sigma}{x_0} \right). \]  \hspace{1cm} (6-2)

Here \( \phi(x) \) is the normal cumulative distribution function, and \( \mu \) is the difference between the mean and the skewness of the distribution. The final expression for the observed pulse shape is therefore written as

\[ L(t) = (1-r)Nf(t|t_1, \mu, \sigma) + rNf(t|t_2, \mu, \sigma), \]  \hspace{1cm} (6-3)

with

\[ r = \frac{t_2}{(A+B)(t_2-t_1)}. \]  \hspace{1cm} (6-4)
The scintillation model with six parameters was used to fit the neutron pulse distribution by optimizing the target function of $R^2$ to be 1, or converging $(1-R^2)$ to be zero. Because of multiple variables the simplex algorithm method with operations of reflection, contraction and expansions was applied [52]. The target function defines a fit space in a multi-parameter space. Starting with predetermined parameter values, the simplex method uses the steepest gradient on the fitted surface to determine how the parameter values should be adjusted. However, the target function often converges to a local minimum, which is not necessarily the global minimum and is dependent on the starting points; the initial points were determined by considering an estimated value of the short decay constant and trial and error runs. Taking a neutron group with kinetic energy of 2 to 2.5 MeV as an example, the pulse shape distribution is fitted and plotted as the blue curve in Fig. 6-1. The parameter values of several regular neutron energy groups are given in Table 6-1.

Figure 6-1. Neutron pulse shape function $L(t)$ fitted to the neutron group with kinetic energy of 2 to 2.5 MeV.
Table 6-1. Parameter values determined by fitting the pulse shape function to four selected neutron energy groups and gamma rays

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Symbol</th>
<th>Neutron energy (MeV)</th>
<th>Gamma Mean</th>
</tr>
</thead>
<tbody>
<tr>
<td>Short time constant (ns)</td>
<td>$t_1$</td>
<td>7.530 8.184 7.560</td>
<td>8.132 7.782</td>
</tr>
<tr>
<td>Long time constant (ns)</td>
<td>$t_2$</td>
<td>58.973 70.267 64.788</td>
<td>71.407 64.882</td>
</tr>
<tr>
<td>Mean transmission time (ns)</td>
<td>$\mu$</td>
<td>12.761 12.869 12.717</td>
<td>12.914 12.805</td>
</tr>
<tr>
<td>St. dev. of transmission time (ns)</td>
<td>$\sigma$</td>
<td>3.311 3.344 3.429</td>
<td>3.311 3.374 3.354</td>
</tr>
<tr>
<td>$R^2$</td>
<td></td>
<td>0.9982 0.9987 0.9986</td>
<td>0.9984 0.9984</td>
</tr>
</tbody>
</table>

As compared to the declared fast decay time in the data sheet for EJ-315, the observed fast decay time is much longer. This is a result of not measuring the light output directly but instead converting it to electronic signals with associated delays and pulse distortions. Forcing the short decay constant to assume the reported value of 3.5 ns results in severe degradation of the pulse fitting. It also results in a severely underestimated long decay constant as the simplex algorithm will optimize it in order to compensate for the too small short-time constant. The long time constant however is so long as to not be significantly affected by this pulse generating process. As can be observed there is a significant difference of an order of magnitude or more between the decay state time constants.

6.3 Summary and Conclusion

The fitting function considers the process of signal production and transportation in the data acquisition system. To optimize the goodness of fitting, a target function of $(1-R^2)$ and the simplex method have been applied to search six parameter values. The theoretical shape function is seen to be capable in producing good fits to capture the scintillator light photons induced by various energy neutrons. It gives an average short decay time constant of 7.75 ns and a long decay time constant of 63.25 ns. In addition,
the mean transmission time and the standard deviation of the transmission time is provided as 12.76 ns and 3.31 ns, respectively. For comparison, the mathematic model of the light scintillation will be applied to fit the pulse shape of gamma rays.
CHAPTER 7
NEUTRON DETECTION WITH $^6$Li-ENRICHED CLYC

Previous work mostly focused on fast neutron detection. While in application, thermal neutrons could be important to us in the mapping of radiation for a loaded spent fuel cask. The elpasolite crystal Cs$_2$LiYCl$_6$: Ce (CLYC) is used in this work for neutron and gamma-ray detection because of its capability of identifying gamma rays, thermal and fast neutrons. Instead of natural Li, 95% enrichment of $^6$Li was used in the detector, which makes it much more appealing in the work of thermal neutron detection. However, the detection in a continuous environment and thermal neutron overestimation of CLYC have not been well studied. In this work, neutrons with continuous energy up to 10 MeV were generated by bombarding a beryllium target with deuterium ion beams, and each detected pulse dimension is reduced to two based on the well-accepted pulse shape discrimination algorithm.

To improve its performance, each pulse has been shaped with a simulated smooth function to amplify its tail component. Thermal neutrons which are shown as a condensed group with electron equivalent energy centered at 3.5 MeV and fast neutrons as a relatively wide spread group are described with Gaussian mixture models optimized according to the mean and covariance matrix. Time-of-flights show there are mainly two energy groups in the overcounting region and fast neutrons interacting with $^{35}$Cl and $^6$Li are the main reasons leading to the thermal neutron overestimation issue. Additionally, the energy spectrum of fast neutrons is calculated according to time-of-flight spectra with deficit counts in the high energy region.
7.1 CLYC Interacting with Neutrons

Radioluminescence spectra of CLYC contain two main emission components coming from core-to-valence luminescence (CVL) between 250 and 350 nm and Ce\(^{3+}\) d-f transitions [69] [70]. The features of CVL only appear under the exposure of γ rays, which is the primary reason of pulse shape discrimination for neutron and gamma differentiation. In the early development of CLYC crystal, C. Combes et al. pointed out that Ce\(^{3+}\)-doped CLYC exhibits high photon yields under both X-ray and γ-ray excitation, however, the low content of \(^{6}\text{Li}\) per cubic volume makes neutron/gamma discrimination less efficient [69]. Radiation Monitoring Device (RMC) and Bubble Technology Industries have changed the crystal growth from natural LiCl to \(^{6}\text{Li}\)Cl with a high capturing cross section (~940 barns), which significantly improved its thermal neutron detection efficiency [71] [72].

The \(^{6}\text{Li}\)-enriched CLYC has been demonstrated to have excellent performance in terms of gamma-ray and neutron identification and measurement [71] [72] [73] [74]. The estimated light yield is about 20,000 photons per MeV. As mentioned previously, CVL is critical to the PSD performance. However, the spectrum of this luminescence overlaps with the cerium absorption bands [75]. As a result, the existence of Ce\(^{3+}\) leads absorption of CVL and shifts on the emission spectrum slightly to a longer wavelength [76] [77]. As shown by E. van Loef et al., in a larger CLYC crystal, CVL can be fully absorbed by Ce\(^{3+}\) [69] [75]. However, self-absorption is not the dominating factor limiting the dopant concentration because this effect is compensated by photon remission. The rise time in the scintillation pulse and the presence of more than one decay time component have been observed when involved with the Ce\(^{3+}\) [72] [73]. The energy resolution at 662 keV has been reported reaching 5% or even better, affected by
the experimental setup, crystal sizes, surface processes, operating temperatures and signal processing algorithms.

In our work, the CLYC crystal is 1” x 1” and was grown by the vertical Bridgman method in a vacuum chamber at RMD with 95% enrichment of $^6$Li and 0.5% of Ce. The shape and transparency is demonstrated in Fig. 7-1. The response of CLYC has been tested based on the pulse height, the pulse tail integral, the total pulse integral and the fetch filtered total pulse integral. Unlike most research where monoenergetic sources were heavily studied, a continuous neutron source was used in our work. The diversity of the incident neutron energy complicates the response measurement of the CLYC detector because some interactions between fast neutrons and $^{35}$Cl and $^6$Li have a similar light output due to neutron thermal absorption. The energy dependent clusters are identified using Gaussian Mixture Models. The separation outcome is also shown in the form of time-of-flight (TOF).

![Figure 7-1. A CLYC crystal grown by RMD is shown to be transparent. The image is cited from the original work of Radiation Monitoring Devices, Inc. [71].](image)

### 7.2 Experiments

The CLYC crystal was set at the center of a well-shielded 30-m long underground tunnel and its front surface was 10.37 meters away from the neutron generator. Fast
neutrons were generated by bombarding a beryllium target with deuterium ion beams at the Van de Graaff accelerator of University of Ohio. The emitted signals were collected by a bialkali photocathode based Hamamatsu R6231-100 PMT. Then the digital signal was then processed by a linear amplifier.
Figure 7-2. CLYC characterization: (a) CLYC’s responses to $^{137}$Cs in terms of pulse height, tail integral, pulse integral, tail integral with smooth function and pulse integral with smooth function; (b) CLYC’s responses to shielded/unshielded Cs and/or Cf sources in terms of pulse height; and (c) CLYC’s responses to shielded/unshielded Cs and/or Cf sources in terms of pulse integral. The maximum value of each pulse height distribution is fixed for comparison purposes.

The response of a detector to different particles are particularly important. The typical $^{137}$Cs gamma source was used to calibrate the location of 662 keV energy peak. Events with an extremely small pulse height, double peaks, or clipped peaks were removed. The responses of CLYC to $^{137}$Cs have been evaluated based on pulse height, tail integral, pulse integral, tail integral with filters which will be discussed in the next section, and pulse integral with filters, as shown in Fig. 7-2. According to Fig. 7-2a, it is obvious the pulse height is not a good indicator of the CLYC’s response to gamma rays, and both tail integral and pulse integral help to improve the resolution. However, that was not the case, at least not an obvious one, when characterizing neutron pulses with deuterated EJ-315 scintillators [3] [46].
The calibration has also been done using the $^{252}\text{Cf}$ neutron source, and the mixture sources of $^{252}\text{Cf}$ and $^{137}\text{Cs}$ with or without polyethylene layers. The purpose of adding polyethylene layers is to moderate neutron and to test CLYC’s responses to fast and thermal neutrons. The responses to gamma rays and neutrons are shown in terms of pulse height and pulse integral in Figs.7-2b & c, respectively. The thermal peak of the pulse integral spectrum has been shifted to the end side, while there is a long tail following the thermal peak in the pulse height spectrum. During measurements, fluorescent light is collected as the total amount of the accumulated signal within a sampling period. Calibration based on pulse height may only capture the CVT signal while missing the big chunk of useful information which truly represents the CLYC’s responses to thermal neutrons.

Besides, the thermal neutron capture reaction emits charged alpha and triton particles with a $Q$ value of 4.8 MeV. Additionally, it’s important to mention that the pulse generated by CLYC may last a few microseconds. In a measurement, if one continue to record a pulse for 1 $\mu$s, the following tail may trigger another pulse which will give a large number of unnecessary events. In general, those events are generally identified as gamma rays.

### 7.3 Simulated Smooth Function

The idea of the smooth function was inspired by C. Bioano et al. [73] [78]. The original module was designed to discriminate the fast component of the signal using a sophisticated analog stretcher circuit [78] [79]. However, the signal recording process has been taken care of by the SIS3316 unit and the pulse peak and time information are relatively easy to be extracted with our Fortan/Matlab programs, and an extra unit is not necessary. Because of the existence of operational transconductance amplifier, the
stretcher circuit can be simplified as a \( CR - (RC)^n \) shaping module. The differentiation and the integration time constants are set to 1 \( \mu s \). The outcome of the stretcher is equivalent to the following circuit equation:

\[
E_{out} = \frac{E_{in}}{n} \left(\frac{t}{\tau}\right)^n e^{-t/\tau}.
\]  

(7-1)

Figure 7-3. Demonstrations of different smooth functions: (1) a default Gaussian smoothing function built in Matlab; (2) CR-RC smooth over 100 ns; and (3) CR-RC smooth over 200 ns. Each smooth function has been normalized.

In this particular case, only one stage of RC integration is used, and the time interval of consecutive sampling points is 4 ns. The results after smoothing by a standard Gaussian smooth function and the CR-RC filter with different window lengths are given in Fig. 7-3. With multiple trails, stretching over 200 ns is used in the following work for pulse shape discrimination processes.
7.4 PSD and Energy Spectrum

7.4.1 PSD Comparison

Three continuous runs were implemented. The neutron gamma discrimination was performed by integrating signals within two different windows: the first one ($W_1$) starting from 120 ns away from the pulse peak to 800 ns, and the second one ($W_2$) from the onset of the signal to 800 ns. The PSD ratio is defined as: $R = W_1/W_2$. A two dimensional matrix was calculated with the PSD ratio and deposited energy in terms of pulse tail integral or pulse integral. Results from one of benchmarks were given in Fig. 7-4. However, they do not tend to show clear fast peaks due to $^{35}\text{Cl}(n,\alpha)^{32}\text{P}$ and $^{35}\text{Cl}(n,p)^{35}\text{S}$, since the energy of incident neutrons have a wide spread range and those nuclear reactions do not relate to a specific energy.

![Graph showing PSD ratio vs. Energy (keVee)]
7.4.2 Energy Spectrum

With CR-RC filters on, the neutron and gamma regions are gotten narrower. The energy spectrum of thermal neutrons are clearly separated from gamma rays as shown in Fig. 7-4d and Fig. 7-5. Even though the energy spectra in Fig. 7-5a in view of either pulse integral or pulse integral with CR-RC filter do not show a clear characteristic feature of thermal neutrons, the PSD plot (Fig. 7-4c) does display a clean separation between the plumes of neutron and gamma pulses according to their tail ratios.

7.5 Energy Clusters and TOFs

The $^6$Li isotope absorbs thermal neutrons with a large cross-section (940 barns), which gives Q-value of about 4.8 MeV. However, it does not require to have a neutron to reach to a thermal level, before it can be captured by $^6$Li, as Fig. 7-5a shows the
probability is becoming greater when its energy decreases. The emitted tritium and the alpha particles deposit approximately about 3.45 MeVee, and the kinetic energy of the neutron itself adds spreads to deposited energy.

Figure 7-5. Spectra distribution: (a) energy spectra for one benchmark, and (b) energy spectra for 3 measurements with/without filters in view of tail integral.
Figure 7-6. Cross-sections: (a) of Cl isotopes interacting with neutrons, and (b) of $^{35}\text{Cl}$ and $^6\text{Li}$ producing charged particles from 1 to 20 MeV. The total cross-section in (b) is the sum of three reactions the raw data was cited from National Nuclear Data Center.
The fast neutron cross-section is of the order of 0.3 barns at energy from 3 to 5 MeV interacting with $^{35}\text{Cl}$ and $^6\text{Li}$. Additionally, the Q-value of the reaction $^{35}\text{Cl}(n, p)^{35}\text{S}$ is 0.62 MeV, and of the reaction $^{35}\text{Cl}(n, \alpha)^{32}\text{P}$ is 0.94 MeV [73]. Thus a fast neutron could also potentially deposit an amount of energy that is similar to the value of Q-value of thermal neutron capture.

As a result, the number of thermal neutron can be over counted, especially in a scenario where no moderation materials are around the CLYC crystal other than the tunnel walls. Nevertheless, the particle detected and shown in the thermal neutron region should have characteristic deposit energy and should be able to be identified based on their distances.

The neutrons are identified based the PSD ratio values and energy in the Fig. 7-4d. The thermal neutron group is further extracted by the clustering algorithm: Gaussian Mixture Model [80] [81]. The cluster is determined by the expectation maximization technique. The process is to calculate the probability of each event belongs to each cluster, and the probability density function of a multivariate Gaussian for cluster \( j \) is defined in the following:

\[
g_j(x) = \frac{1}{\sqrt{(2 \pi)^n |\Sigma_j|}} e^{-\frac{1}{2} (x - \mu_j)^T \Sigma_j^{-1} (x - \mu_j)}.
\]  

(7-1)

Here, \( x \) an input vector, \( n \) the vector length, and \( \Sigma_j \) the \( n \times n \) covariance matrix for cluster \( j \). The probability of a certain sample point \( i \) belongs to the cluster \( j \) is calculated in the following:

\[
w_{ji} = \frac{g_i(x) \Phi_j}{\sum_{k=1}^{K} g_i(x) \Phi_k}
\]

(7-2)

where \( \Phi_j \) is the fraction of the dataset belonging to cluster \( j \). During iterations, we re-calculate the mean and covariance till the changes are negligible. The particular way
how the mean of cluster j and the covariance matrix are calculated can be found in J. Bilmes [82] and Matlab tutorial. The group value is set at 3, including the fast neutron group, thermal neutron group and unwanted noises. The result is given in Fig. 7-7a: the group colored in blue (cluster 1) is the unwanted noises or small pulses, the group colored in green (cluster 2) is the thermal neutron region with a part of counts from fast neutron reaction, and the wide-spread region in red (cluster 3) is the major fast neutron group. In measurements, the proportions of events in cluster1 and cluster 2 are 43%, respectively.

The TOF spectra of fast neutron and thermal neutron are plotted in Fig. 7-7b. The thermal neutron TOF is mostly relatively flat and shows a stable absorption of thermal neutrons, except that the cyan-colored region between 200 and 500 ns shows double peaks. Those peaks are seem due to $^{35}\text{Cl}(n, p)^{35}\text{S}$, $^{35}\text{Cl}(n, \alpha)^{32}\text{P}$ and $^6\text{Li}(n, \alpha)^4\text{T}$. Even the reaction of $^6\text{Li}(n, \alpha)^4\text{T}$ whose Q-value is about 1 MeV larger than that of the thermal neutron absorption is possible to contribute the thermal neutron overcounting in the region of interest because of the relatively wide range of the thermal neutron region. The peak with larger value of TOF in the region of interest associates with neutrons whose energy is less than 3 MeV. It is mainly due to $^{35}\text{Cl}(n, p)^{35}\text{S}$ and $^6\text{Li}(n, \alpha)^4\text{T}$ according to Fig. 7-5b. The wider peak with smaller TOF value associates with neutrons whose energy is from 3 to 10 MeV is mainly due to $^{35}\text{Cl}(n, p)^{35}\text{S}$ and $^{35}\text{Cl}(n, \alpha)^{32}\text{P}$. This reaction shall mostly due to all three nuclear reactions. However, the particular contributions on the thermal neutron overestimation due to those reactions or other unidentified nuclear reactions are not yet quantified. Nevertheless, the accuracy of the thermal neutron detection can be improved using numerical Monte-Carlo method.
within which a PDF describing distribution of neutrons counting variances in the
deposited energy and the pulse ratio, and the misidentified fast neutron events can be
deduced referring to the thermal neutron background fitting curve.

Figure 7-7. Energy clusters: (a) application of Gaussian mixture model with 3 energy
groups, and (b) TOF distributions of the clusters 1 and 2. The fitting curve
was implemented based on the data points outside the region of interest.
Figure 7-8. The neutron spectrum provided by the Edwards Accelerator Laboratory, and TOF was generated based on the energy spectrum. (b) The measured fast neutron spectrum based on the countings from the $^6$Li enriched CLYC detector.

The fast neutron spectrum was generated by redistributing the timing information of events in the fast neutron region. For comparison, the Edwards Accelerator
Laboratory provided their neutron source spectrum, as shown in Fig. 7-8. The spectrum from CLYC shows a deficit region at the relatively high energy region whose reaction cross-section is decreasing.

The exact value of the trend change on the TOF spectrum from CLYC measurement can be affected significantly by the timing resolution, depending on the energy region. Here, the narrow peak next to origin is the photon peak and can be totally cut out by rising the energy threshold. This small amount of photons is remained for the purpose of TOF adjusting and achieving a better timing resolution.

7.6 Summary and Conclusion

In this chapter, we presented the PSD performances of 1”x 1” 6Li enriched CLYC detector in view of the pulse peak, pulse integral and tail integral. Its performance has been significantly improved by the CR-RC fetch function which amplifies the tail of the pulse and increases the ratio difference between neutron and gamma rays. Continuous neutrons events are separated into 3 clusters, including the unwanted small pulse region, the quasi-elliptic thermal neutron region and the wide spread fast neutron region according to Gaussian mixture models. The optimization was implemented according to convergences on the mean and the covariance matrix. The energy-related thermal neutron overcounting has been identified according to the thermal neutron TOFs: the left and wider peak in the spectrum is mainly due to reactions of $^{35}\text{Cl}(n, p)^{35}\text{S}$ and $^{35}\text{Cl}(n, \alpha)^{32}\text{P}$, while the right and narrower peak in the spectrum is mainly due to $^{35}\text{Cl}(n, p)^{35}\text{S}$ and $^{6}\text{Li}(n, \alpha)^{4}\text{T}$. Additionally, the fast neutron spectrum from the CLYC measurements was compared to the original neutron energy spectrum. The deficit region agrees well with the decreasing on cross-sections over 6.5 MeV.
The concept of light is familiar to everyone, especially visual light which is naturally captured by our eyes and which makes our world to be colorful and wonderfully weird. However, in our research the light generated by the scintillation material is critical to the type of photocathode used in PMT because the energy of photons needs to best fit the energy gaps of materials in order to emit a maximum amount of electrons. Those electrons are normally further accelerated along with secondary electrons to enhance the electron charges.

When coming to measurements, photons emitted by the scintillation materials will travel through the medium and be bounded around in the cell. Eventually, photons hit the photocathode if not escaped or absorbed. This part of work simulates the light transmission process in view of statistics and investigate the signal spatial dependence between photon origins and signal outputs.

8.1 Existing Particle Transportation Simulation Review

Monte Carlo N-Particle simulation is popular in the study in particle transmission, especially particles like neutrons and gamma rays. It gives good results when we consider the average outcome of a large number of histories and it’s also well suited for the biased calculations. However, it fails when considering a single history [84]. In standard MCNP/MCNPX, secondary photons are generated independently from the type of neutron collision.

However, MCNP-PoliMi code samples the neutron collision type first, and then try to generate photons accordingly. Their system is adapted from the standard MCNP/MCNPX and relies on the data libraries to find information on neutron collisions,
cross sections, reaction Q-values, outgoing neutron energies and angular distributions. Unfortunately, there is no simple correspondence between neutron reactions and photon-production reactions. In MCNP-PoliMi, it determines the number of photons based on the total energy released by gamma rays, $E_g$ [85]. However, as discussed in Chapter 1, there is nonradioactive energy transmission and those will lead to that the sum of the energy of photons is much smaller than the total released energy.

Additionally, SCINFUL code was designed to provide the full response anticipated for neutron interactions in either liquid or solid cylindrical organic scintillators by means of Monte Carlo method. It covers energy from 0.1 to 80 MeV and the primary reactions like $H(n,n)$, $C(n,n)$, and etc. Their simulation results show a better agreement was achieved with measurements in the higher energy [86]. However, the code was made for NE-213 and NE-110 only. While, in our system, we focus on the simulation on the statistic results and track the transition of photons generated by the deposited neutron energy.

8.2 Light Emission Spectrum of EJ-315

The electromagnetic spectrum is a vast continuum stretching from the very short wave cosmic, gamma, x-rays, ultraviolet, visible, and infrared to the long wavelength radio waves, as seen in Fig. 8-1. Particularly, those gamma rays are also photons but with much higher energy when compared to visible light and with a wavelength in the order of nanometers. Gamma rays are also sensitive to fast neutron detectors. As we have discussed in Chapter 1, after energy deposition from gamma or fast neutrons, photons are eventually emitted by detector cells. For EJ-315, the energy spectrum was measured by Eljen Technology, as shown in Fig. 8-2.
When coming to simulation, the number of photons generated should be strictly controlled by the amount of energy deposited inside the scintillation materials. The original physical processes of photon interaction in quantum level is complex and impossible to precisely track in current situation. However, if viewing the light generation process as a distribution of probability, then when produced, a photon can be given a certain amount of energy with a random direction. Additionally, in order to increase the calculation speed, the light generation process was written in C++ language. The probability function is viewed as discrete data points. In our simulation, the energy was generated randomly through a mapping of the non-uniform light spectrum to a uniform binary distribution. The non-uniform distribution is calculated from the fitted multiple Gaussian functions and is its cumulative distribution function before normalization (CDF), as shown in Fig. 8-3. As a result, the photon energy or the wavelength is a random process and can be easily determined by a value between 0 and 1.
8.3 Light Reflection and Diffraction

A ray tracing program is normally used to compute their pixel at a time, and for each pixel the basic task is to find the object that is seen at that pixel’s position in the
image. Each pixel looks in a different direction. For our purposes, the ray is tracked to record where it eventually ends with when absorbed by a photocathode if it not escapes already. Our liquid scintillators are encapsulated within aluminum cells with a clear window for coupling to a photomultiplier tube. As known, reflected light obeys the law of reflection. There are two types of reflection: specular and diffuse.

Figure 8-4. EJ-522 reflective paint reflectance spectrum.

For smooth surfaces, specular reflection occurs; while diffuse reflection occurs on rough surfaces. The rougher the surface is, the more diffuse the reflection experiences. For smooth surfaces like aluminum cells, the light reflection is reasonably assumed to be specular. However, for the white paint coated on the interior wall, the light reflection is diffuse. According to the law of reflection, the angle of incidence is equal to the angle of reflection.

Additionally, when the light moves from one medium to another, it is refracted. If it moves from the first medium with refractive index $n_1$ to the second medium with
refractive index $n_2$, with the incident angle and the refraction angle to the surface normal $\theta_1$ and $\theta_2$, respectively, according to Snell’s law it has the following relation:

$$n_1 \sin \theta_1 = n_2 \sin \theta_2. \quad (8-1)$$

The refractive index of deuterated benzene is 1.498, however, the refractive index of aluminum is much larger than that of the deuterated benzene [83]. The probability of light reflection is evaluated according to the Fresnel equation:

$$R_0 = \left| \frac{n_1 - n_2}{n_1 + n_2} \right|^2. \quad (8-2)$$

The reflectivity index is 473 for aluminum for photons with a wavelength of 248 nm\(^1\). However, photons emitted by deuterated benzene have a wavelength over 400 nm as shown in Fig. 8-2. With the increasing tendency, the refractive index of aluminum is higher than 470 for photons emitted by our detector volume. Thus, more than 98.5% of light is reflected by the aluminum cell. Despite the light reflection strictly follows the mirror reflection, the surface normal of the walls is extremely important in order to simplify the calculation of the reflected angle. However, the EJ-522 reflective paint was coated on the interior surfaces of the aluminum cell. The bright white paint is consisted of titanium dioxide pigment and of a paint base selected for its inertness to the solvent reaction of nearly all liquid scintillators. The combining results of the reflective paint and the aluminum wall is plotted in Fig. 8-4.

8.4 Ray-plane Intersection

Ray is bounded back and forth by the aluminum cell. It’s important to keep track the intersection point before specular reflection. In a 3-dimensional space, assume an

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\(^1\) This information was found at the Refractive Index Database, created by M. N. Polyanskiy. The link has been inserted.
incident ray with direction vector \( d = (l, m, n) \) that passes a point \( p_0 = (x_0, y_0, z_0) \), the equation of a line is written as

\[
p_1 = p_0 + t \cdot d. \tag{8-3}
\]

Considering the detector is 3-by-3 inches with cylindrical shape, the wall can be described in the following:

\[
x^2 + y^2 = R^2, \quad \left( -\frac{H}{2} \leq z \leq \frac{H}{2} \right), \tag{8-4}
\]

where \( R = \frac{H}{2} = 1.5 \) inches. Substitute Eq. 8-4 into Eq. 8-3, the intersection can be easily calculated. However, it is important to notice the boundary condition that the \( z \) value of the intersection point should be in the range of \( z \), as shown in Eq. 8-4. For computing, it’s important to avoid infinite loops because of the incident angle and the intersection position. As a result, a proper spatial error should be considered to eliminate those undesirable scenarios. In our case, the error has been set to be \( 10^{-6} \) cm. Additionally since only one side of EJ-315 is mounted to a PMT, the light absorption mainly happens in the side with photocathode, which has been set to be the bottom surface.

### 8.5 Polarization

Light is an electromagnetic wave, and the electric field of this wave oscillates perpendicularly to the direction of propagation. Light is un-polarized if the direction of the electric field fluctuates randomly in time. On the contrary, if the direction of the electric field of light is well defined, it is polarized light. In our case, photons are generated in a random direction, and the transmission of single photon is very similar to the generation of sun rays. Thus, the light induced by the scattering of a fast neutron is un-polarized.
8.6 Light Attenuation

In many cases, the absorbance of a sample follows the Beer-Lambert Law [84]:

\[ A(\lambda) = \log \frac{I_0}{I_{\lambda}} = \varepsilon(\lambda)lc, \]  

(8-5)

where \( I_0 \) and \( I_{\lambda} \) are the light intensities of the beams entering and leaving the absorbing medium, respectively. \( \varepsilon(\lambda) \) is the molar absorption coefficient expressed in L mol\(^{-1}\) cm\(^{-1}\), and \( c \) is the concentration of absorbing species in mol L\(^{-1}\) and \( l \) is the absorption path length in cm. Sometimes, physicists use the Napierian absorption coefficient \( \alpha(\lambda) \) to express the relationship between incident intensity and leaving intensity in the following

\[ I_{\lambda} = I_0 e^{-\alpha(\lambda)l}. \]  

(8-6)

\( l \) is the absorber thickness g/cm\(^2\). Thus Eqs. 8-5 and 8-6 can be used to calculate the probability of light attenuation. The raw data of mean free path of photons is measured by Eljen Technology and plotted in Fig. 8-5.

![Figure 8-5. The mean free path \( \lambda \) (cm) of photons in deuterated benzene.](image-url)
As discussed in Section 8.1, the light spectrum released by the deuterated benzene is 400 nm. However, the absorption spectrum indeed only covers the light with much shorter wavelength. It’s even more obvious for H-based benzene whose absorption mainly happens for photons with wavelength less than 270 nm. Additionally, there may have additional compound that affects the light absorption and re-emission properties. Currently, the absorption in our simulation is considered based on the mean free path from 320 to 440 nm. For photons with wavelength that is higher than 440 a fixed value is considered. The light absorption indeed in significant considering the size of the detector we have.

8.7 Result and Discussion

![Starting position (0,0,-3.7)](image)
Figure 8-6. Photon absorption position: (a) for 1 million events with initial fast neutron energy deposition position at (0,0,-3.7), (b) for 10 million events with start position at (1,1,-3.7), and (c) for 10 million events with the start position at (1,2.5, -3.7).
This numerical simulation was implemented in C++ platform. With initial conditions like the interaction position and the event numbers, individual photon is generated traveling in a random direction. Its travelling path is tracked until the photon hits the photocathode layer. The result of each event is not determined by if the event is lost or not. Instead, the event is evaluated based on the weight of how much it contributes to the signal collected.

A reasonable weight is between 0 and 1 for each individual photon. The result visualization is implemented in Matlab. The size of memory required for computation is minimized by outputting each individual result, instead of accumulating events in a lengthy queue. At the moment, generating one million events requires about 3 s of CPU time. The final position of different event is plotted in Fig. 8-6, within which three different locations are simulated. In Fig. 8-6a, the original of position is $(0,0, -3.7)$ which is close to the center of the photocathode surface with 1 million events; in Fig. 8-6b, the original position is $(1,1, -3.7)$ with 10 million events; and in Fig. 8-6c, the original position is $(1, 2.5, -3.7)$ with 10 million events. The results showing that the signal distribution patterns are very different regarding their initial positions.

8.8 Summary and Conclusion

A primeval frame work of the photon generation and emission model has been developed in our lab. It allows us to track the travel path of each photon and to evaluate the contribution of individual photon to the final signal collected by our digital data acquisition system. With the light output function from EJ-315 as measured previously, we could actually further calculate the energy deposited by each incident neutron and calculate the signal distribution of photons. Reversely, the model would be able to
predict the interaction position based on the output signal or each individual waveform and potentially to improve the signal spatial resolution.
CHAPTER 9
SUMMARY AND FUTURE RESEARCH

This work has investigated the characteristic responses of the deuterated organic scintillator and the thermal neutron detection crystal (CLYC) to neutrons. Particularly, for fast neutron detection, deuterated organic scintillators are most frequently used in my research. The collision model describes the details of neutron collision history in a certain interacting medium and energy depositions. The pulse height generation model picks up the deposited energy from the collision model and refers to light output function from measurements to provide the pulse height spectrum. Despite the model is created for EJ-315, it’s relatively easy to transfer this work to other cylindrical scintillation detectors with other scintillation material. In near future, the analytical neutron collision model will be combined with the optical photon transportation model to predict the behavior of EJ-315 detector.

For measurements and post signal analysis, the algorithms used in fast neutron/gamma-ray discrimination have been discussed. The energy of fast neutrons was measured based on time-of-flights at Ohio University. Data extraction, dimension reduction, denoising, fitting and mathematical models were mainly implemented in Matlab/Fortran. The whole process will be further modified and built as a standard process as a fast neutron detection protocol.

However, most organic scintillators are only sensitive to fast neutrons, thermal neutrons released from a loaded spent fuel cask requires both the radiation information from not only fast neutrons but also from thermal neutrons. A $^6\text{Li}$-enriched CLYC crystal has been investigated. Unsupervised training technique: Gaussian mixture model was implemented to separate neutrons into 3 categories: noise, thermal neutron and fast
neutrons. The energy-related over estimation has been identified according to thermal neutron time-of-flights.

Eventually, we would like to move forward to use the characterized detectors to build a transportable detection system that supports thermal and fast neutron detection at the same time, and further to apply the detection system to monitor the nuclear spent fuel cask.
LIST OF REFERENCES


BIOGRAPHICAL SKETCH

Growing up in Wenzhou, China, Haitang Wang graduated with honors of Outstanding Undergraduate Student of Shandong Province and Best Senior Design from China University of Petroleum (East China) in 2012 with a Bachelor of Science degree in applied physics. Afterward, he began pursuing his master’s and Ph.D. in nuclear engineering sciences at the University of Florida.