Spectroscopic radiography with monoenergetic neutrons for homeland security applications

by

Jill Marie Rahon, Captain, U.S. Army
B.S., United States Military Academy (2006)

Submitted to the Department of Nuclear Science and Engineering
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Abstract

In this thesis, I discuss the hypothesis, testing, and applications of a radiographic system which uses the spectral analysis of fast neutrons generated in the $^{11}$B(d,n$\gamma$)$^{12}$C reaction. Neutron radiography in current use systems employs a total count analysis to reconstruct hydrogenous content in scanned cargoes. This research presents a technique which leverages the large differences in the energy dependence of neutron interaction cross sections between hydrogenous materials and those of higher atomic number. These dependencies result in characteristic spectral details of hydrogenous materials which may be analyzed to classify type and areal density of cargo contents. The studies presented here demonstrate that this technique is feasible and suited to the application of national security port monitoring at industry-requisite speeds and cargo dimensions.

Thesis Supervisor: Areg Danagoulian
Title: Assistant Professor of Nuclear Science and Engineering

Thesis Reader: Richard Lanza
Title: Senior Research Scientist, Nuclear Science and Engineering
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A.1 Optimized integration bounds and corresponding $R$ values for various cargoes and areal densities ........................................... 66
Chapter 1

Introduction

Neutron radiography is an imaging technique well suited for the discernment of low atomic number (Z) elements, based on their high propensity to interact with neutrons. Leveraging the uniquely high variability of the neutron interaction cross section in hydrogen and hydrogenous materials, it is possible to generate radiographs via novel analysis of transmitted neutron energy spectra.

Chapter one discusses radiographic applications to national security and presents a perspective on the enormous task of securing U.S. borders. It covers relevant background information and an overview of nuclear reaction based gamma and neutron radiography.

Chapter two begins with a short literature review of the research investigating the $^{11}\text{B}(d,n\gamma)^{12}\text{C}$ reaction to establish a scientific basis for the results expected from the experiment. It discusses the neutron spectrum generated by the reaction and presents the underlying physics of fast neutron radiography.

Chapter three provides an overview of the experimental scope and describes the experimental set-up of the radiography system.

Chapter four describes the experiments and motivations for the design decisions made during the experimental process, as well as data analysis.
Chapter five presents results and discussions of the technique’s effectiveness and applicability in radiography supporting national security. It also discusses conclusions and possible expansions to the research and application of the spectroscopic technique.

1.1 The role of radiography in national security

The 50,000 containers that enter U.S. ports daily provide myriad opportunities for the illicit transportation of nuclear weapons, as well as conventional contraband into the country [1]. From 1993 to 2015, there have been 18 reported cases of attempted smuggling of special nuclear material (SNM) — defined as various forms and isotopes of uranium, plutonium, and thorium, including one in 2015 conceived with the objective of transporting uranium to members of the Islamic State of Iraq and Syria [2]. In addition to the nuclear and radiological threat, land and sea cargo must be effectively and rapidly screened for low-density contraband, including narcotics and high explosives.

Radiography provides a powerful tool for security purposes in the non-destructive imaging and testing of sealed cargoes. Its ability to detect dangerous and illicit materials from a standoff distance increases safety for inspectors while preserving the availability of forensic evidence required to prosecute smugglers. The Department of Homeland Security (DHS) and U.S. Customs and Border Protection (CBP) rely upon risk-based models to determine supply chain vulnerabilities that necessitate additional screening for a container once it reaches U.S. territory [3]. However, due to changing host country and political and legal constraints, it is difficult to assess the threat degree of a particular container, a problem readily mitigated by rapid scan inspection systems in numbers high enough to contend with the throughput of domestic ports.

The Domestic Nuclear Detection Office (DNDO) has awarded an Academic Re-
search Initiative (ARI) collaborative research grant to MIT, Georgia Institute of Technology, and Pennsylvania State University to develop a novel approach to the active detection of shielded nuclear materials hidden in container cargo. The universities are investigating the use of the $^{11}$B(d,n$\gamma$)$^{12}$C reaction to generate high-energy $\gamma$ rays for high contrast transmission imaging and fissile material confirmation via photofission. This multi particle reaction also generates neutrons in quasi-monoenergetic groups which may be used to identify fissile material through neutron induced fissions, as well as generate radiographs sensitive to materials of low atomic numbers. This thesis investigates the radiographic use of neutrons generated in the $^{11}$B(d,n$\gamma$)$^{12}$C reaction, as well as the application of a new concept of spectroscopic analysis.

1.2 Fundamentals of radiography

Radiographic systems provide a non-destructive method to image and evaluate sealed cargo by detecting transiting particles such as photons (X-rays and $\gamma$ particles), neutrons, and muons, then inferring cargo information based on signature provided by the particle counts and energy.

1.2.1 X-ray and gamma radiography

In the most basic form of X-ray radiography, the relative strength of the mass attenuation coefficient of the scanned material, $\mu$, is measured as photons from the incident beam attenuate on atomic particles at rates based on the material’s atomic number ($Z$). At energies used in medical imaging and baggage scanning, X-rays interact via photoelectric effect or Compton scattering. Dual energy X-ray imaging is an established technique for these applications, in which the dominance of one photon interaction or the other is measured via attenuated fluxes at two known interrogation beam energies. $\mu$ is strongly dependant on $Z$ and is used to reconstruct an image of
internal structure [4].

In the case of dense objects like shipping and air cargo containers, the amount of material the interrogating beam must traverse adds greater complexity to the determination of $\mu$. An effective solution is the use of dual high energy X-ray or $\gamma$ radiography employing widely separated beam energies, such as the system being developed under ARI. Here, the interrogation beam is sufficiently energetic to penetrate the steel container walls as well as induce pair production interactions in the scanned material. Similar to dual low energy radiography, the $\mu$ of high Z materials experiences a greater contribution from the effects of pair production than from Compton scattering, while this effect is reversed in the $\mu$ of lower Z materials. The ratio of attenuated flux at a photon beam energy favoring pair production and one at an energy favoring Compton scattering is evaluated alongside the cargo’s areal density to determine the atomic number of medium and high Z materials (aluminum, $Z = 13$, and greater). Gammas used for this process may be generated by nuclear interactions or reactions, while X-rays are produced via bremsstrahlung radiation from the acceleration of electrons in a high Z conversion target.

1.2.2 Neutron radiography

In neutron radiography there exist several analogs to $\gamma$ radiography, though the underlying physics largely differs. Like x-ray imaging, a material’s composition may be inferred from the number of transmitted neutrons, measured with a photographic plate or neutron sensitive detector. Here, neutron attenuation is the measured value and is related to the material’s neutron cross section, $\sigma$. This is a material-specific measurement of the likelihood for a neutron of a given energy to interact via a specific mechanism (scattering, absorption, fission, etc.), measured in units of barns, an areal quantity. The macroscopic cross section, $\Sigma$, is the product of nuclei $N$ per unit volume and $\sigma$, yielding the probability per unit path length for a specific interaction.
in units of inverse distance. The probability that a neutron will undergo any type of interaction while transiting a material is given by [5]:

$$\Sigma_{tot} = \Sigma_{scatter} + \Sigma_{fission} + \Sigma_{rad.capture} + ...$$  \hspace{1cm} (1.1)

which is in analogy to the linear absorption coefficient for \( \gamma \)-rays. The known flux of the incident beam is compared to the transmitted flux to determine a \( \Sigma_{eff} \) of the scanned material.

The propensity of neutrons to attenuate via scattering or absorption interactions in light elements such as hydrogen, boron, and lithium while penetrating through heavier elements makes this form of radiography suited to detecting variations of hydrogenous content, even when shielded by heavy metals. Basic neutron transmission radiography is now complemented by more advanced techniques including digital imaging and tomography, used commonly to non-destructively examine electronic components, fuel cells, engine turbine blades, and items of cultural significance [6]. Fast neutron resonance radiography (FNRR) is another form of transmission imaging in which an object is exposed to a broad spectrum neutron beam \( (E_n = 1\text{-}10 \text{ MeV}) \). Elements possessing sharp cross section resonances, such as carbon, nitrogen, and oxygen, alter the transmitted spectrum, which is collected via a detection method with high enough energy resolution to discern the changes. This resonance mapping combined with total count derived density mapping can determine the stoichiometric elemental ratio in the subject, permitting discernment between common explosives and benign cargo [7–9].

Intense neutron sources are required to provide meaningful measurement statistics; currently nuclear reactors, deuterium-tritium (DT) neutron generators, and accelerator-based spallation or nuclear reaction sources provide beams of sufficient intensity and energy.  

---

\(^1\)For a comprehensive list of neutron radiographic techniques and applications, see [6]
1.3 Motivations for comprehensive methods of detection

Compounding upon the concerns of speed and throughput discussed in Section 1.1, the sheer volume and scope of materials that present harmful potential to the people, property, or economy of the U.S. and its allies is daunting. On a typical day, the CBP conducts 1,000 apprehensions and 54 arrests, discovers 4,437 materials for quarantine (including plant, meat, animal by-product and soil), and seizes 11,660 pounds of drugs and $274,065 in undeclared and illicit currency [10]. Lawrence Livermore National Laboratory conducted a study of the composition of containerized cargo passing through the Port of Long Beach, select results of which are presented in Table 1.1. Of note is the wide variety of materials and densities encountered on a typical day, requiring multiple scan methods sensitive to very high Z weapons threats and low Z contraband such as alcohol, drugs, and explosives. A distribution of average cargo effective densities recorded in this study is available in Appendix A, Fig. A-1.

<table>
<thead>
<tr>
<th>Category</th>
<th>% TEU</th>
<th>Category</th>
<th>% TEU</th>
</tr>
</thead>
<tbody>
<tr>
<td>Furniture, toys, misc articles</td>
<td>20.18</td>
<td>General merchandise</td>
<td>3.09</td>
</tr>
<tr>
<td>Machinery &amp; appliances</td>
<td>15.48</td>
<td>Vegetable products</td>
<td>2.66</td>
</tr>
<tr>
<td>Textiles &amp; textile articles</td>
<td>10.75</td>
<td>Wood pulp products</td>
<td>2.47</td>
</tr>
<tr>
<td>Base metals</td>
<td>7.11</td>
<td>Hides &amp; skins</td>
<td>2.24</td>
</tr>
<tr>
<td>Plastic and rubber</td>
<td>7.00</td>
<td>Animals &amp; animal products</td>
<td>1.60</td>
</tr>
<tr>
<td>Prepared foodstuffs</td>
<td>5.29</td>
<td>Instruments measuring-musical</td>
<td>1.41</td>
</tr>
<tr>
<td>Transportation equipment</td>
<td>5.04</td>
<td>Mineral products</td>
<td>0.63</td>
</tr>
<tr>
<td>Articles of stone, plaster, cement</td>
<td>4.00</td>
<td>Animal &amp; vegetable fats</td>
<td>0.15</td>
</tr>
<tr>
<td>Footware, headgear</td>
<td>3.98</td>
<td>Works of art</td>
<td>0.07</td>
</tr>
<tr>
<td>Wood &amp; Wood products</td>
<td>3.64</td>
<td>Pearls, precious stones &amp; metals</td>
<td>0.05</td>
</tr>
<tr>
<td>Chemical products</td>
<td>3.12</td>
<td>Arms &amp; ammunition</td>
<td>0.03</td>
</tr>
</tbody>
</table>

Note: Data gathered on the 14 highest throughput days between June 2004 and June 2005 [11]

Fixed and mobile radiation portal monitors (RPM) and other passive forms of
detection abound at ports, though their deployment is suited towards detecting materials emitting a radioactive signal well in excess of background levels, such as an unshielded radiation source \[5,12,13\]. In the instance of even a lightly shielded significant quantity of SNM (i.e. 12 kg of weapons grade uranium in a 3 cm tungsten tamper) total neutron and $\gamma$ counts at the surface of a typical shipping container are reduced to $1.7 \times 10^{-4} \text{ particles cm}^{-2} \text{sec}^{-1}$; an insufficient signal in practical backgrounds \[14\].

Simple x-ray attenuation measurements permit non-unique combinations of cargo thickness and density — a small amount of a very high atomic number (Z) material such as SNM can look the same as a large thickness of low Z material. Dual energy x-ray scanners provide excellent contrast between materials of low and high Z, but are unable to distinguish between low Z materials.

It becomes obvious that there is no universal scanning solution that can determine the presence of threats, both those of a high and low Z nature. Critical to defeating smuggling is a comprehensive system, capable of detecting elements of very high atomic number (Z) as well as rapidly image the cargo for other contraband. The widely separated 4 and 15 MeV $\gamma$ lines used in the ARI radiography system described in Section 1.1 provided a marked advantage over 5 and 9 MeV dual energy bremsstrahlung interrogation systems, presenting an image contrast sensitivity for high Z a factor of five times than that achievable with the latter \[8\]. Inherent in the reaction central to this and other nuclear reaction based radiography systems is the generation of quasi-monoenergetic neutrons, a source of low Z quantification that can be used to complete the radiographic assessment of cargo. Brandis, et al. are among those combining dual energy $\gamma$ radiography with nuclear reaction generated neutrons to conduct FNRR with the broad neutron beam generated in the $^{11}\text{B}(\text{d},\text{n}\gamma)^{12}\text{C}$ reaction \[8\].

\[2For an analytical description of the challenges and benefits of active interrogation over passive detection in terms of background and signal flux density and detector parameters, refer to \[12\]
1.4 A new analysis method for neutron radiography

FNRR has the ability to provide elemental composition-specific cargo reconstructions with high spatial resolution, however it necessitates precise resolution of neutron energy to exploit resonances. This requires the use of pulsed beams, time of flight spectroscopy, and in some cases, complex apparatus for selecting neutron production angles, which could complicate or preclude its application to mobile and rapidly deployable field-use systems. In situations calling for less precise material analysis and expedient set-up, standard radiography presents a viable option.

In standard transmission radiography, total counts of transmitted neutrons are compared with the known flux of the incident beam to generate a ratio representative of neutron interaction within the scanned material. This research proposes and investigates a new method for the reconstruction of specimen content based on spectral analysis of neutron beams. It leverages the information within the energy spectrum by comparing the energy-dependant attenuation properties of different materials. The experiments in this thesis have been conducted as a component of the $^{11}\text{B}(d,n^{\gamma})^{12}\text{C}$ reaction radiography system, exploiting the wide spectral range of the quasi-monoenergetic neutron energies. However, the described spectral analysis technique can be applied in any apparatus using dual or multi energy neutron groups.
Chapter 2

Fast neutron radiography with the $^{11}$B(d,n\(\gamma\))$^{12}$C reaction

Chapter two presents the underlying physics of fast neutron radiography and discusses the neutron spectrum generated in the $^{11}$B(d,n\(\gamma\))$^{12}$C reaction. It includes a literature review of the reaction to establish a scientific basis for the results expected from the experiment.

2.1 The $^{11}$B(d,n\(\gamma\))$^{12}$C reaction

2.1.1 Reaction review

A short literature review of the research history of the $^{11}$B(d,n\(\gamma\))$^{12}$C reaction serves to describe its characteristics which make it suitable for a radiographic interrogation source. Several investigations of this reaction have been undertaken, the goals of which can be classified into three general groups. Throughout the 1950s and early 1960s, efforts focused on \(\gamma\) and neutron angular yields resulting from the increasing deuteron beam energies made available by innovations such as the tandem Van de Graaff accelerator [15–17]. In the 2000s, the reaction’s utility as a \(\gamma\) interrogation
source necessitated modern measurements of its excitation function\footnote{These reassessments found good agreement between previously reported values and present day measurements after correcting for the detector response function from the earlier measurements [18].} \cite{18,19}.

The third group of studies is comprised of those determining the mechanics of the $^{11}\text{B}(d,n\gamma)^{12}\text{C}$ reaction and the roles of the compound nuclear theory and direct interaction (stripping) theory in generating the resulting products. In the mid-1960s, Class, Price, and others sought to determine the contributions of each type of interaction on the resulting neutron spectrum by determining the angular distributions and excitation functions of observed neutron groups \cite{20–22}. This search is of relevance to the reaction’s applications to fast neutron radiography because it introduces the mechanism by which the spectrum of mono-energetic neutron groups arises.

### 2.1.2 Reaction characteristics

In the compound nucleus model, nuclear reactions proceed in two distinct parts: first, the formation of the compound nucleus, $C$, from the reactants $a$ and $b$, and second, the disintegration of this intermediary nucleus to the reaction products, $d$ and $e$:

\[
a + b \rightarrow C \rightarrow d + e.
\]  

(2.1)

By this model, the reaction of interest in this research proceeds as:

\[
^{11}\text{B} + ^3\text{D} \rightarrow ^{13}\text{C}^* \rightarrow ^{12}\text{C}^* + n + \gamma,
\]  

(2.2)

where the kinematics of the reaction govern the excitation energy of $^{12}\text{C}^*$. The resulting neutrons are produced in discrete energy groups corresponding to the disintegration path of $^{13}\text{C}^*$ to the ground or an excited state of $^{12}\text{C}^*$, as illustrated in the energy level scheme in Fig. 2-1.

It is convenient to refer to each energy group by the resulting state of $^{12}\text{C}^*$ with
which it corresponds—therefore, the $n_0$ group corresponds with ground state, $n_1$ with the first excited state, and so forth. $n_x$ is used in certain referenced publications as an inclusive higher-excitation energy group (the $^{12}$C 12.71 MeV level and above), as they are difficult to individually resolve due to convolution with each other and neutrons from other source reactions [19].

Of note in Fig. 2-1 is the reaction’s high positive Q-value of 13.73 MeV and the large separation between $^{12}$C levels and their corresponding neutron groups. These features enhance the suitability of the $^{11}$B(d,n$\gamma$)$^{12}$C reaction to radiographic purposes as the high Q value contributes to prolific generation of widely separated $\gamma$ lines useful for dual energy radiography. Likewise, the quasi-monoenergetic neutron groups are optimally spaced to exploit the greatest variability of hydrogen’s $\sigma_n(E)$ and are easily resolved by low cost spectrometers. Increasing the energy of the bombarding deuteron accesses higher excited states of $^{12}$C [25]. To generate the higher energy 15.1 MeV photon for use in the project’s radiographic system, the $^{11}$B(d,n)$^{12}$C($\gamma_{15.1}$) reaction has a deuteron threshold energy of $E_d = 1.627$ MeV, resulting in a $\sigma_{reaction} = 1.1$ barns [19]. The energy of the resulting neutrons, $E_n$, is dictated by the energy
of the reaction not emitted as a $\gamma$, in addition to the kinetic energy of the deuteron [26], given by

$$\sqrt{E_n} = \sqrt{\frac{m_d m_n E_d}{(m_n + m_{12C})^2} \cos \theta \pm \sqrt{\frac{m_d m_n E_d}{(m_n + m_{12C})^2} \cos^2 \theta + \frac{m_{12C} - m_d}{(m_n + m_{12C}) E_d} + \frac{Q m_{12C}}{(m_n + m_{12C})}}}.$$

(2.3)

<table>
<thead>
<tr>
<th>Group</th>
<th>Carbon-12 Level [MeV]</th>
<th>Neutron Energies [MeV]</th>
<th>$J^n$</th>
<th>$\frac{d\sigma}{d\Omega} \ [mb/sr]$</th>
<th>$\dagger$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$n_0$</td>
<td>Ground</td>
<td>16.51</td>
<td>0+</td>
<td>1.9</td>
<td></td>
</tr>
<tr>
<td>$n_1$</td>
<td>4.43</td>
<td>12.20</td>
<td>2+</td>
<td>4.5</td>
<td></td>
</tr>
<tr>
<td>$n_2$</td>
<td>7.65</td>
<td>8.86</td>
<td>0+</td>
<td>0.9</td>
<td></td>
</tr>
<tr>
<td>$n_3$</td>
<td>12.71</td>
<td>4.01</td>
<td>1+</td>
<td>100$\dagger$</td>
<td></td>
</tr>
</tbody>
</table>

Note: All neutron energies include the energetic contribution from a 3.0 MeV deuteron.

$\dagger$ The cross section data is from plots of neutron angular distributions for the listed energy groups (Fig. 17 in Ref [19]). Data from a deuteron beam energy of 3.25 MeV and a 3.17 mg/cm$^2$ $^{11}$B target.

$\dagger$ The cross sections for neutron energies corresponding to $^{12}$C excited states greater than 8.0 MeV are summed due to indistinguishable individual neutron energy peaks when measured by time of flight spectroscopy.

Table 2.1 displays the most prominent neutron energies by yield in the $^{11}$B(d,n)$^{12}$C($\gamma_{15,1}$) reaction, as generated by the 3.0 MeV deuteron used in this research. The neutron energy groups broaden as a function of deuteron’s stochastic energy straggling within the thickness of the source target. The mean range of a 4.0 MeV deuteron (the energy used in the data comprising Fig. 2-2) in a boron target is on the order of hundreds of $\mu$m [5]. Those deuterons that straggle but are able to interact above the threshold energy contribute to a peak broadening of approximately 2 MeV for the $n_0$ and $n_1$ energy groups, as measured by a time of flight spectrometer [19]. This effect is visible in Fig. 2-2 and prevents individual resolution of the $n_1$ and $n_0$ groups in the thick target data, as well as in the data collected in this research.
2.1.3 Other sources of neutrons

Other physical processes arising from or occurring during the $^{11}$B(d,n)$^{12}$C($\gamma$) reaction generate neutrons which affect the collected spectrum. $^{12}$C, present in the target backing foil, and $^{16}$O from target oxidization experience (d,n) reactions, contributing neutron peaks at 1.05 and 3.59 MeV for $^{12}$C and 1.71 and 2.24 MeV for $^{16}$O [19]. As target thickness increases however, the effects of these reactions become obscured by low energy neutron production and scatter from (d,n) reactions on $^{11}$B. As a diagnostic tool, Taddeucci, et al. have proposed the neutron signature from these reactions can be used to identify target contaminants. Likewise, true target thickness can be measured by neutron yields compared to those of an stopping thickness target.

The asymmetric charge distribution and low binding energy of the bombarding deuteron make it prone to stripping reactions, in which one constituent particle is absorbed and the other escapes as the reaction product [16, 27]. Given 3.0 MeV deuterons on a thick target of $^{11}$B, stripping reactions have a $\sigma$ of 0.05 barns; with stripped neutrons contributing to collected spectra in a continuum based on the kinematics of the reaction [21]. An incident deuteron with energy in excess of the target
nucleus' Coulomb barrier that experiences a stripping reaction emits the escaping particle in a distinct angular distribution. Price and Almond found the distribution maxima of ground state neutrons \( n_0 \) at \( \theta_{\text{LAB}} = 20^\circ \), with distributions becoming increasingly anisotropic as \( E_d \) increases above 5 MeV, an effect which could distort neutron spectral analysis in set-ups involving higher energy beams \([15,20]\). Similarly, Ward and Grant noted the anisotropy of the \( n_1 \) group and theorized it a result of captured p-wave deuterons accessing spin states in the compound nucleus which result in \( j_n = \frac{5}{2} \) or \( \frac{3}{2} \). These \( l = 2 \) neutrons are emitted non-isotropically, yielding a distribution nearly symmetrical about \( \theta_{\text{LAB}} = 90^\circ \) with peaks at \( 20^\circ \) and \( 150^\circ \) \([16]\)–a second process which could result in differences between measurements taken at different angles from the source.

In situations where fissile material is present in the combined \( \gamma \) and neutron beam, neutron induced fissions, as well as photofissions by the 15.1 MeV \( \gamma \) will result in the addition of fission spectrum neutrons \([28]\). \( ^{238}\text{U}, \, ^{235}\text{U}, \, \text{and } ^{232}\text{Th} \) each have a maximum cross section for photofission of approximately 480 mb for an incident photon energy of 14 MeV, with an average neutron fission energy of 2.0 MeV for \( ^{238}\text{U} \) and \( ^{235}\text{U} \) and 1.5 MeV for \( ^{232}\text{Th} \) \([29]\). Total fission and photoneutron yield and its impact on the radiographic spectra is dependant on fissile material mass and shielding. Awareness of angular and spectral peculiarities arising from the aforementioned processes is critical to the maintenance of experimental parameters that permit consistent radiographic analysis. Section 3.1 presents experimental design decisions to mitigate these factors.

### 2.2 Fast neutron radiography

Neutrons of a fixed energy have a constant probability per unit path length for each type of interaction mechanism, expressed as a cross section, \( \sigma \). The relative probabil-
Figure 2-3: Energy-dependent total neutron interaction cross sections
Data for high-density polyethylene (HDPE), aluminium and lead (natural isotopic abundance). The HDPE total $\sigma$ is a result of a concentration-weighted sum of carbon and hydrogen cross sections per the reduced polyethylene molecule, CH$_2$ ($\sigma_{\text{HDPE}} = \sigma_C + 2\sigma_H$)

...
Figure 2-4: Energy dependant neutron attenuation
Plot for HDPE, aluminium, and lead of ratios of transmitted to incident flux as a function of incident neutron energy. The areal density for each scenario is $30 \frac{g}{cm^2}$ and the vertical dashed lines correspond to the neutron energies listed in Table 2.1.

by

$$I = I_o \exp \left\{ -\sigma_n(E) \rho x N_A / A \right\}, \tag{2.4}$$

or more generally by

$$\frac{I}{I_o} = e^{-\Sigma_{tot} x} \tag{2.5}$$

where $I_o$ is the incident beam energy and $\Sigma_{tot}$ is the total macroscopic cross section.

Using Eq. (2.4), Fig. 2-4 plots the ratio of transmitted to incident flux as a function of incident neutron energy. The plot indicates that for equal areal densities of lead, aluminum, and high density polyethylene (HDPE), there exists a significant difference in low to high energy neutron attenuation in HDPE as compared to other materials. As the behavior of carbon’s cross section mirrors closely that of other higher $Z$ materials, it is hydrogen’s uniquely varying total neutron interaction cross section that drives the effect in HDPE. This research explores the technique of assessing the variation in energy dependant attenuation present in a spectroscopic measurement and its correlation to hydrogenous materials in the scanned cargo.

Using a beam of quasi-monoenergetic neutrons incident on a target for a certain
length of measurement time, the transmitted fluence of a given neutron energy group, \( \phi \), can be obtained by integrating counts over the desired energy region. By selecting two energy regions optimally spaced to exploit the greatest variation in \( \sigma_n(E) \) for hydrogen, the ratio of the fluences yields a measurement of the attenuation in the sample due to hydrogenous material. It is useful to define this ratio, \( S \), as:

\[
S = \frac{\phi_{[E_2, E_3]}}{\phi_{[E_0, E_1]}},
\]

where \([E_2, E_3]\) and \([E_0, E_1]\) are the high and low energy regions, determined by the search procedure described in Section 4.2. The hypothesis of this research is that the \( S \) value generated by this technique provides a determination of the amount of HDPE within the scanned voxel.

It is valid to neglect the effects of material-specific resonances visible in Fig. 2-3 and Fig. 2-4 provided the selected transmitted energy ranges are sufficiently broad to encompass resonance effects within statistical error. The resolution of the detector used in this research was low enough that individual resonances were not visible in transmission spectra, however higher resolution set-ups must be aware of the potential for these effects to skew data interpretation in overly narrow energy regions. Herein lies the benefit of a quasi-monoenergetic source over a continuous spectrum; providing neutron generation at distinct energy groups provides better counting statistics within each group. By allowing the downscatter from the sole high energy source to comprise counts in the low energy region, the spectrum becomes more susceptible to the effects of cargo moderation and resonances. A spectrum of discrete neutron production energies, while still affected by downscatter, produces a greater number of low energy counts of neutrons that were able to transit the entire sample thickness. A random sample of any two neutrons in a quasi-monoenergetic spectrum is more probable to be further spaced apart in energy than a sample of two neutrons from a ‘white’ or sole neutron energy spectrum. This allows the quasi-monoenergetic beam to be more
efficient at signal detection rooted in the energy regions of which it spans.
Chapter 3

Experimental setup

Chapter three discusses the design and technical specifications of experiment, including beam source, collimation, cargo, and data acquisition.

3.1 Concept of the experiment

The neutron transmission radiography experiment follows as a natural extension of the larger gamma radiography experiment. When passing through a hydrogenous sample, the ratio of transmitted high to low energy neutrons changes due to the energy-specific $\sigma_n(E)$ of the material. Thus the $\Sigma_{eff}$ for hydrogen-containing materials is significantly higher for low energy neutrons than their high energy counterparts, resulting in preferential low energy beam attenuation. The research concept was to use the quasi-monoenergetic neutron groups arising from a nuclear reaction to interrogate various materials and determine the extent to which the above relation holds in experimental conditions. Neutrons emitted isotropically from the source reaction pass through a set of vertical collimators, through the cargo, and into a collimated neutron sensitive detector, as presented in Fig. 3-1. Analysis of the recorded count ratios in select energy regions provides information on cargo composition.
3.1.1 The goal

The goal of the experimental portion of this research was to collect neutron transmission spectra that established a dependence between energy specific attenuation and the quantity of hydrogenous material in the cargo. The necessity for 'clean' spectra obtained in a complex radiation environment was the driving force behind several adjustments to the set-up and acquisition procedures.

3.2 Experimental set-up

3.2.1 Radiofrequency quadrupole linear accelerator

This experiment was conducted using 3.0 MeV deuterons accelerated by a AccSys Model DL-3 Linac System radiofrequency quadrupole (RFQ) linear accelerator at MIT’s Bates Accelerator Center in Middleton, MA. A RFQ simultaneously focuses and accelerates ions via exact matching of the RF period and the longitudinal modulation of the copper quadrupole vanes. The velocity-independent acceleration, focusing, and bunching effects permit the RFQ to accept low velocity ions at much higher currents than a drift tube linear accelerator [30,31]. The high currents produced in the machine translate to improved counting statistics (therefore faster scans and lower uncertainty) in a transmission radiography set-up, though a smaller accelerator
or other neutron source can be used with this technique to the same effect. The runs conducted in this experiment were limited to a 15 μA current due to available shielding and the existing exclusion area at the RFQ facility.

The deuteron beam was incident on a 1.0 mm thick natural boron target (isotopic composition: 19.9% $^{10}$B and 81.1% $^{11}$B by mole percent) which is mounted to a tantalum backing substrate. Tantalum is used due to its high strength properties, good adhesion through its readiness to form boride tantalum, and good hydrogen solubility and diffusivity [12,32]. The RFQ was operated at a current of 2 to 20 μA, a beam repetition rate of 300 Hz, and a pulse length of 30 μs.

3.2.2 Shielding, collimation and cargo mock-up

The primary purpose of shielding in this set-up was to moderate and capture the fast neutrons generated in the isotropic reaction that were not collimated into the beam. This mitigates the background by thermalizing neutrons not in the interrogation beam prior to reaching the detection assembly, as well as maintains neutron induced $\gamma$ reactions to within radiation safety standards. 5% borated HDPE in 2.5 cm sheets was stacked around the target, moderating all neutrons but those produced in the range of $\theta_{LAB}=0-10^\circ$, defining the interrogation beam as the unmoderated neutrons. This shielding configuration also minimized the contributions from the 20$^\circ$ angular production maxima of the deuteron stripping reaction. $\gamma$-ray shielding was also placed around the boron target and beam line to reduce the neutron and $\gamma$-ray dose. As the detector used in the experiment was sensitive to neutrons and $\gamma$, 5 cm of lead shielding was placed in the beam line prior to the cargo in order to remove a significant quantity of produced $\gamma$, thereby decreasing the likelihood of $\gamma$-neutron pile-up events. This lead shielding remained in place for the entirety of the data collection. Collimators were constructed of dual density concrete and resulted in a neutron beam width of 2 cm incident on the cargo. In later measurements, the post cargo collimation visible
in Fig. 3-1 was removed to limit the effects of secondary neutrons ricocheting off
collimator walls and into the detector. This adjustment resulted in a slight qualitative
improvement in noise at neutron energies below 2 MeV, the effects of which are visible
in the example in Appendix A, Fig. A-3.

The cargo mock-up consisted of a modular steel shelving system mounted to a
stepper motor driven linear actuator, designed to emulate the movement of an LD-3
aircraft shipping container through a scanner. The forward edge of the cargo was
located 2.2 m from the neutron production target and the detector 5.4 m from the
forward edge of the cargo. Phantoms of varying composition and areal densities were
loaded on to the motion system or directly adjacent to the post-source collimation.
Phantom materials consisted of 5% borated HDPE (0.94 $\frac{g}{cm^3}$), reactor-grade graphite
(2.25 $\frac{g}{cm^3}$), aluminum (2.7 $\frac{g}{cm^3}$), and lead (11.3 $\frac{g}{cm^3}$), each in a variety of sizes and areal
densities.\(^1\) Graphite and lead were chosen as mock cargo for their availability and
representation of the extremes of the atomic number range. In select runs, sheets of
depleted uranium (19.1 $\frac{g}{cm^3}$) were scanned and compared to matching areal densities
of lead to determine the effects of induced \(^{238}\)U fission neutrons on the spectrum.
Analysis of this data did not show any differences.

### 3.2.3 Detector

An EJ-309 organic liquid scintillation detector was chosen for its ability to discrimi-
nate between neutrons and $\gamma$-rays through pulse shape discrimination (PSD) analysis.
The scintillating components in EJ-309 liquid have a short decay time of 3.5 ns, useful
in PSD. This liquid also has as a high flashpoint as compared to industry standard
solvent xylene, improving handling safety in the lab and during field use. The 5.08 x
5.08 cm cylindrical detector used in this experiment was mounted to a Hamamatsu
R7724 photomultiplier tube, the technical drawing and specifications for which are

\(1\) Areal density is the measure of a material's thickness and density, $\alpha = \rho \cdot \Delta x$, in units of $\frac{g}{cm^2}$.
available in Appendix A, Fig. A-4. The detector was calibrated frequently using the pulse shape discrimination selected $\gamma$ spectra from $^{137}$Cs, $^{60}$Co, and PuBe sources.

3.2.4 Data acquisition

The Accelerator-based In-situ Materials Surveillance (AIMS) Data Acquisition (ADAQ) software package was used for data collection. The ADAQ libraries are an in-house developed system providing acquisition and analysis of experimental and simulated particle detector data through integration of C++, Python, and the ROOT data analysis framework. ADAQAcquisition is a user application which interfaces the CAEN acquisition hardware and the user’s computer in order to control high voltage and digitizer settings. It layers in additional functionality by enabling the user to view individual waveforms, apply live-time pulse shape discrimination (PSD) cuts and other utilities to visualize and analyze data as it is collected. ADAQAnalysis is a separate user application utilizing the ADAQ framework and ROOT to further enhance and analyze data, the usage of which will be discussed in Section 4.2 [33].

Analog waveforms from the detector were digitized using a CAEN V1720 12-bit 250MHz digitizer and the photomultiplier tube voltage provided by a SY1527 CAEN high voltage mainframe unit.

3.3 Data collection

3.3.1 Characterizing the neutron environment

Prior to the radiography experiments, an investigation was conducted to determine the nature and effects of the neutron environment in the ARI system set-up. The neutron background during the operation of the RFQ is significantly great to impact both $\gamma$-ray and neutron spectroscopy. For each $^{12}$C that is created a neutron is emitted; a $\gamma$ only occurs when $^{12}$C is left in an excited state. Neutron-induced $\gamma$ from
(n,\(\gamma\)) and (n,n') reactions form a significant continuum over top of the spectrum used in \(\gamma\) radiography, visible in Appendix A, Fig. A-2. This background obscures the energy peaks of interest; to reduce its impact, 50 cm of in-beam neutron shielding were used during \(\gamma\) collection.

In order to maximize the signal to background ratio in the region of the detector, a survey was conducted to determine the extent of beam spreading and strength of the neutron background. The neutron background in this experiment is defined as any neutron which is not emitted from the source, transits the cargo, and enters the detector. The background therefore includes neutrons which scatter within the cargo and subsequently enter the detector, as well as cosmic and other naturally occurring neutrons. For the energy range of concern in this experiment, the cosmic neutron differential flux is approximately 0.7 s\(^{-1}\)cm\(^{-2}\) [5,34]. At typical experimental conditions, the in-beam flux was 1\(\times\)10\(^2\) s\(^{-1}\)cm\(^{-2}\), therefore no specific attempt was made to shield nor coincidence-remove cosmic neutrons. Fig. 3-2 displays total neutron counts made over a five minute period for a shielded and unshielded detector, both in the beam line and at 25 and 50 cm lateral displacement.

Detector shielding consisted of a lead block castle, surrounded by 5 cm of HDPE. Its primary purpose was to minimize \(\gamma\) counts which could interfere with neutron spectroscopy and to limit secondary neutron scatters into the detector. Though off-beam counts comprise 2% of those recorded in-beam, they are entirely comprised of a continuum of 1-6 MeV neutrons. This background is likely neutron scatters on the cargo or collimation which then enter the detector. The presence of a background of secondary scatters has the potential to obscure the spectrum’s low energy signal, discussed further in Chapter 4.
Figure 3-2: Neutron counts in a lead and borated-HDPE shielded vs. unshielded detector at various positions. Counts recorded by an EJ-309 organic liquid scintillation detector. Offset measurements were displaced laterally from the beam line, while remaining at the same height as the beam. Shielding consisted of 5 cm of lead and 5 cm of HDPE surrounding the detector, leaving a 2 x 2 cm opening.
3.3.2 Transmission experiments

After the background characterization, several iterations of transmission measurements were conducted at varying detector positions, beam currents, and cargo areal densities. Throughout this process, many updates to data acquisition procedures and software were completed to contend with high data throughput. A total of 161 transmission runs of homogeneous and heterogeneous cargoes were recorded for analysis and radiographic imaging between September 2015 and April 2016.
Chapter 4

Data analysis

Chapter four presents the organization and analysis of the data collected in the previous chapter. Section 4.1.1 and Section 4.1.2 discusses elements of fast neutron detection and how their effects impact assessment of collected spectra.

4.1 Fast neutron spectroscopy

4.1.1 Proton recoil detectors

The most common technique for fast neutron detection is based on the high elastic scattering cross section for neutrons by light elements. In this method, a scattering interaction on hydrogen nuclei (or deuterium, helium, etc.) in the detector volume results in some portion of the neutron’s kinetic energy transferred to the target. In the instance of a hydrogen-containing detection medium, as is present in EJ-309 scintillation liquid, the recoil nucleus is a single proton. A neutron may transfer all of its energy in a single collision with a hydrogen nucleus, with the maximum fractional energy transfer per single collision decreasing as the target increases in nucleon number. The energy per collision transferred to a hydrogen recoil nucleus, $E_R$, by a neutron of energy $E_n$ is a function of the angle of recoil (in the lab frame)
given by \( E_R = (\cos^2 \theta E_n) \) [5].

Because all scattering angles are permitted, the distribution of recoil energies mirrors a plot of the the differential scattering cross section, \( \sigma(\Theta) \), as a function of the neutron scattering angle (in the center of mass frame). However, a convenient approximation may be made in the case of s-wave neutron inelastic scattering, which is isotropic in the center of mass frame. Therefore \( d\sigma(\theta)/d\Omega \) does not vary with angle and is a constant \( \sigma_{tot}/4\pi \), leading to a continuous uniform distribution of recoil proton energy, beginning at zero and terminating at \( E_n \) [5, 35]. A beam of monoenergetic neutrons transfers energy according to this distribution and the recoil protons deposit all of their transferred energy in the medium, giving rise to a roughly rectangular energy spectrum. A beam of quasi-monoenergetic neutrons therefore manifests as a series of stacked rectangular distributions for each energy group.

### 4.1.2 Pulse shape discrimination with liquid organics

Liquid organic scintillators operate on the same principles as other scintillation detection media— the conversion of ionizing radiation into detectable luminescence, which is transmitted to a PMT or other light sensor. Organics fluoresce due to transitions in the energy level structure of individual molecules, as opposed to the crystalline lattice dependencies of sodium iodide and other inorganics, therefore they can operate as scintillators independent of physical state. Uncharged ionizing radiation interacts in the detection volume and transfers energy to charged particles of the medium, which then transfer kinetic energy to organic molecules, leaving them in excited states. \( \gamma \)-rays interact primarily through Compton scattering in low-Z organic scintillators, producing a negatively charged electron which travels some distance in the detection volume, generating singlet and triplet electronic excitation states in the molecules along its track. The singlet state of EJ-309 scintillation liquid decays via prompt fluorescence in approximately 3.5 ns, manifesting as the fast component in a digitized
A neutron-induced recoil proton experiences a significantly higher specific energy loss ($-dE/dx$), creating a high density of excited state molecules. Though a direct decay of a triplet state is forbidden, triplet-triplet annihilation among adjacent molecules results in a nonradiative transition to a singlet state [5,37]. Occurring with a rate proportional to triplet concentration squared, this singlet transition and subsequent prompt decay results in a delayed component in the waveform, the intensity of which may be quantified to determine the nature of the original incident particle. The pulse shape discrimination (PSD) analysis in this research was implemented with the Digital Dual Gate Charge Integration method in CAEN firmware, in which the waveform charge is integrated between programmable gates set to encompass the total waveform and the ‘tail’ or slow component, as shown in Fig. 4-1.

The tail integral is divided by the total integral to generate a PSD parameter, which is plotted against pulse area, permitting energy-dependant discrimination of pulses exhibiting greater occurrence of the slow component light emission. When
plotted as such, the neutron counts group into an upper ‘lobe’ while the photons comprise the lower one, visible in Fig. 4-2. Neutron or \( \gamma \)-ray specific events may be selected by a 2D cut and further processed in ROOT. Recoil protons with a high ionization density lead to a greater number of triplet-triplet annihilations and therefore a higher delayed component. A more energetic proton results in lower ionization densities, a circumstance which results in a non-linear light output function for neutron events. PSD cuts were applied to select the \( \gamma \)-ray spectrum from the three calibration sources, \(^{137}\)Cs, \(^{60}\)Co, and PuBe. The Compton edges from their characteristic decay energies were identified, \( \text{e.g.} \) a 476 keV Compton edge corresponds to the 661 keV \(^{137}\)Cs decay, and fitted with a Gaussian edge finder in ADAQAnalysis to generate a calibration curve. As the detector was calibrated using \( \gamma \) sources, neutron energy deposited is reported in units of MeV-electron equivalent [MeVee]. Neutron energy in proton equivalents may be achieved using known dependencies in which the light output, \( L(E_p) \) is approximated by a polynomial rational: \( L(E_p) = L_0(E_p^2/(E_p + L_1)) \) where \( E_p \) is proton energy and \( L_0 \) and \( L_1 \) are fitted parameters from a characterization of detector material and geometry [5, 39, 40]. The plots presented in this research remain in units of MeVee as relative counts, not precise neutron energies, are of greater relevance towards the experimental goal.

### 4.1.3 Spectrum features

The particularities of proton recoil detectors and PSD analysis, as well as other experimental details generate certain features in the collected spectra worth noting. Fig. 4-3 is a typical homogeneous cargo transmission spectrum recorded during this research. The neutron energy groups generated in the \(^{11}\)B(d,n\( \gamma \))\(^{12}\)C reaction are visible as stacked, roughly rectangular energy groups with each leading edge corresponding to a neutron group energy. The scintillator non-linearity is evidenced by the distortion of each rectangle towards lower energies, compounded by the prevalence of
Figure 4-2: PSD parameter versus pulse area
Typical PSD plot from experimental data, this plot is of a 20-minute transmission run through 30 cm of HDPE. The neutron and γ-ray lobes have a high degree of separation for all data collected.

downscattered neutrons in energies below 2 MeVee.\(^1\) The finite resolution of the detector due to nonuniform light collection and other sources of noise results in neutron energy edges dispersed in a Gaussian distribution, rather than in a sharp cutoff at \(E_n\). Further convolving the spectrum are the effects of deuteron energy straggling in the thick target as discussed in Section 2.1.2, leading to increased broadening of the neutron energy groups.

It becomes readily apparent that the method of spectroscopy used in this experiment is ill-suited for individual neutron energy group determination. However, this resolution is more than adequate to exploit the greatest variability in hydrogen’s \(\sigma_n(E)\), occurring between 2 and 10 MeV incident neutron energy. The detector’s low resolution has the effect of obscuring neutron cross section resonances from individual materials in the set-up or cargo, providing a ‘smoothing’ function.

\(^1\)Knoll, Fig. 15.18 presents examples of the distortion of the proton-recoil energy distribution due to scintillation non-linearity.
Figure 4-3: Typical neutron spectrum from the $^{11}$B(d,$n\gamma$)$^{12}$C reaction

Transmission spectrum of 30 $\frac{g}{cm^2}$ of graphite, recorded with an EJ-309 detector. The graphite transmission spectrum was chosen to illustrate neutron energy groups due to the discernibility of the $4.0\text{ MeV}$ $n_3$ neutron group, which becomes obscured due to low energy secondary scatters in cargoes of higher Z. The horizontal lines mark neutron groups produced in the $^{11}$B(d,$n\gamma$)$^{12}$C reaction, which were converted to MeVee using data from Verbinski, et al. [41] Group $n_2$ does not correspond with a visible energy edge due to its low production yield.

4.2 Spectrum Analysis

A significant portion of the analysis work in this research went towards identifying suitable integration regions which would optimize a spectrum’s $S$ value. $S$, the value defined in Section 2.2 as the ratio of the neutron fluences in two selected energy regions, can yield a measurement of the attenuation in the sample due to the presence of hydrogenous material. To determine the optimal $S$ value, we must determine the two energy regions optimally spaced to exploit the greatest variation in $\sigma_n(E)$ for hydrogen. The first analysis method approached, discussed in Section 4.2.1 and Section 4.2.2, used an iterative optimization search to determine the regions in two transmission spectra which yielded the greatest variation in attenuation. The second method was a more simple, intuitive selection of energy regions that sought to improve upon the first method by mitigating nuisance downscattered neutrons. The develop-
ment of both methods is discussed in this section and results from their application to heterogeneous cargoes discussed in Section 5.1.

4.2.1 Optimization of integration bounds for a fixed areal density

The first method of optimizing the $S$ value compared integrated counts from two regions in a homogeneous non-hydrogenous spectrum with those from a homogeneous HDPE spectrum of the same areal density. The regions indicating the greatest divergence between the spectra while preserving low statistical error were hypothesized to yield the highest $S$ values for any cargo of that areal density. Defining $S_H$ (calculated in Section 2.2) as the $S$ value for the HDPE spectrum and dividing it by $S_X$, the $S$ value for some non-hydrogenous material, $X$, we can create a ratio of strengths of attenuation. The absolute difference of this ratio from unity is divided by the propagated error, $\sigma_{S_X/S_H}$, to yield units of standard deviation. The $R$ value is defined as:

$$R = \left| 1 - \frac{S_X}{S_H} \right| \frac{\sigma_{S_X/S_H}}{S_H}. \quad (4.1)$$

$R$ is a figure of merit that describes the analyzing power of a particular set of integration windows, $\{E_0, E_1, E_2, E_3\}$, where $E$ corresponds to an energy bin for like-binned histograms. $R$ provides a measure of effectiveness for any configuration of energy windows in determining the relative strengths of attenuation between two sets of homogeneous-Z cargoes. For example, $R = 2$ represents a 95% discrimination capability between HDPE and material $X$.

It should be noted that the distribution of the ratio of two normally distributed, independent random variables with non-zero means has no defined mean, variance, nor higher moments. For normal random variables $X$ and $Y$ with strictly positive means
and variances \((\mu_x, \sigma^2_x)\) and \((\mu_x, \sigma^2_x)\) and coefficients of variation, \(\delta_{x,y} = \sigma_{x,y}/\mu_{x,y}\), less than 1, the ratio \(Z = X/Y\) is approximately normally distributed. Both ratios occurring in this work (that of integrated counts in the \(S\) value calculation and that of the \(S\) values themselves in the \(R\) value) fit these criteria, permitting good approximation of their ratio distribution to a Gaussian and their errors propagated as such [42].

To determine the energy bins conveying the strongest signal of cross section based attenuation, an iterative search procedure was performed using the ROOT data analysis framework. The search algorithm selected the largest \(R\) value for any two spectra by iterating over every possible bin combination, providing inherent noise reduction through penalization of low count integration regions. Table 4.1 shows the highest \(R\) value for each comparison between HDPE and carbon, aluminum, lead, and uranium (all in areal densities of 30 \(g/cm^2\)).

<table>
<thead>
<tr>
<th>Cargo</th>
<th>(S) value</th>
<th>(R) value (this configuration)</th>
<th>(R) value (at 50 cm/s scan)†</th>
</tr>
</thead>
<tbody>
<tr>
<td>HDPE</td>
<td>0.939 ± 0.011</td>
<td>48.19</td>
<td>4.4</td>
</tr>
<tr>
<td>Carbon</td>
<td>0.362 ± 0.001</td>
<td>47.35</td>
<td>4.4</td>
</tr>
<tr>
<td>Aluminum</td>
<td>0.375 ± 0.001</td>
<td>49.80</td>
<td>5.5</td>
</tr>
<tr>
<td>Lead</td>
<td>0.288 ± 0.001</td>
<td>49.81</td>
<td>5.4</td>
</tr>
<tr>
<td>Uranium</td>
<td>0.289 ± 0.001</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

†: \(R\) value extrapolated to a 1 mA beam current, permitting an industry-requisite scan speed of 50 cm/s. Integration step size, \(E_0\), is 0.06 MeV for all runs; \(S\) values calculated using 0.6 - 1.9 MeV and 2.2 - 8.8 MeV regions.

The integration regions used to generate the \(S\) values in Table 4.1 were the averaged integration windows resulting in the highest \(R\) value for each material’s comparison with HDPE. The spectra of the 30 \(g/cm^2\) cargoes were segmented into 200 bins of 0.06 MeV each and the optimal bin widths calculated to be \(\{E_0, E_1\} = \{10, 32\}\) and \(\{E_2, E_3\} = \{36, 146\}\). These integration regions are overlaid on the consolidated 30 \(g/cm^2\) transmission spectra in Fig. 4-4. Visible in the figure is the search procedure’s

2Due to the non-availability of uranium in variable areal densities, only carbon, aluminum, and lead were used for subsequent runs
Figure 4-4: Normalized 30 $\frac{g}{cm^2}$ runs with optimized integration regions overlaid. Spectra have been normalized to total integrated counts with overlaid optimized integration regions, \( \{E_0, E_1\} = \{10, 32\} \) and \( \{E_2, E_3\} = \{36, 146\} \).

Successful identification of spectral regions in which the unique variability of the \( \sigma_n(E) \) of hydrogen has the greatest attenuation effect on HDPE’s spectrum relative to the attenuation in the higher-Z spectra.

### 4.2.2 Optimization of integration bounds for cargoes of various areal densities

In realistic scenarios, scanned cargo units are packed non-uniformly, resulting in varying effective areal density and atomic number throughout the container. It is therefore essential that the hydrogenous content assessment be either agnostic of areal density or able to apply density-specific integration bounds given a pixel density. Table 4.2 catalogues the \( R \) value resolving power of our spectral assessment technique in a more realistic scenario where the effective areal density of a cargo is known \textit{a priori} and areal density-specific integration regions are applied to determine hydrogenous content.\(^3\)

\(^3\)Areal density may be determined through absolute neutron or 4.4 MeV \( \gamma \) counts given a precise determination of integrated beam current [14].
<table>
<thead>
<tr>
<th>Areal density [g/cm²]</th>
<th>Integration regions</th>
<th>Carbon</th>
<th>Aluminum</th>
<th>Lead</th>
</tr>
</thead>
<tbody>
<tr>
<td>10</td>
<td>10-24 35-180</td>
<td>71.78</td>
<td>74.65</td>
<td>78.16</td>
</tr>
<tr>
<td>20</td>
<td>10-27 34-148</td>
<td>59.43</td>
<td>60.59</td>
<td>65.26</td>
</tr>
<tr>
<td>30</td>
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<td>9.7</td>
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<td>16-48 77-144</td>
<td>6.89</td>
<td>2.31</td>
<td>4.30</td>
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Note: Integration regions, corresponding to 0.06 MeV bin sizes, are the averaged carbon, aluminum, and lead integration regions for each areal density. For example, the integration regions used for the 10 g/cm² group were averaged from the optimal integration windows for 10 g/cm² of carbon, 10 g/cm² of aluminum, and 10 g/cm² of lead. Data from 20 minute runs at 8-10 μA.

As the areal densities of cargoes in Table 4.2 increase, the counts decrease, limiting the technique’s ability to discern a hydrogenous spectrum from a non-hydrogenous one. Despite tailored integration regions, the R value decreases nearly linearly as areal density increases, due primarily to the high attenuation of counts in the HDPE runs. For a 20 minute run at 10 μA, there were $2.6 \times 10^5$ total neutron counts transmitted through 10 g/cm² of HDPE, but only $8.8 \times 10^3$ counts through 40 g/cm². The limiting of R values due to worsening statistics is also apparent in the full table of R values and specific integration regions for areal densities 10 through 60 g/cm², which may be found in Appendix A, Table A.1. 60 g/cm² was selected as the upper limit due to low neutron count rates in higher HDPE densities, as well as space constraints in the experimental set-up. Despite the wide range of atomic number between carbon and lead, the R values for each material trend strongly with areal density, with no observed material-specific trends.

Extending the scanning scenario a step further to one in which an areal density determination is unavailable, there is a benefit in determining the effectiveness of universal integration regions. The averaged integration regions for all runs in Table 4.2 are $\{E_0, E_1\} = \{10, 31\}$ and $\{E_2, E_3\} = \{42, 149\}$. The results of these universal
Figure 4-5: Normalized 10 and 40 $g/cm^2$ runs with universal integration regions overlaid.

Fig. 4-5a. is the total count-normalized spectra for 10 $g/cm^2$ and Fig. 4-5b. the spectra for 40 $g/cm^2$ for comparison. Both sets of histograms are overlaid with the universal integration regions, $\{E_0, E_1\} = \{10, 31\}$ and $\{E_2, E_3\} = \{42, 149\}$.

Integration bounds applied to each spectrum are listed in Table 4.3 and overlaid on both the consolidated 10 and 40 $g/cm^2$ spectra in Fig. 4-5.

<table>
<thead>
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<th>Areal density [g/cm²]</th>
<th>R value (% change)</th>
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<tr>
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<td>69.37 (3.4)</td>
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<td>20</td>
<td>58.82 (1.3)</td>
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<td>30</td>
<td>47.69 (0.9)</td>
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<td>12.66 (0.8)</td>
</tr>
<tr>
<td>60</td>
<td>4.65 (33.6)</td>
</tr>
</tbody>
</table>

**Note:** The percent change in each R value from its best value in Table 4.2 is listed in parentheses. Values are derived using the universal integration bounds, $\{E_0, E_1\} = \{10, 31\}$ and $\{E_2, E_3\} = \{42, 149\}$. Data from 20 minute runs at 8-10 $\mu$A.

A visual comparison of the density-optimized bounds in Table 4.2 and the universal bounds in Fig. 4-5 shows that they are quite similar until 50 $g/cm^2$. Noting the percent change next to each R value in Table 4.3, the ‘one size fits all’ integration regions result in an average R value of 2.7% lower than the optimized bounds for each areal density.
for cargoes 40 $\frac{g}{cm^2}$ and less. For 50 and 60 $\frac{g}{cm^2}$, the low statistics and incidence of low energy scatter precludes an accurate hydrogenous content assessment at neutron fluxes available in this experiment. The universal integration bounds remain a viable option for systems in which an effective assessment of areal density is infeasible or impractical. For a slight overall reduction in $R$ value, these regions provide the ability to determine hydrogenous content agnostic of areal density, given adequate statistics.

4.2.3 Improvements to $R$ value selected integration bounds

Prior to discussing the second method of integration bound determination, it is important to briefly discuss additional spectral features which limit hydrogenous content assessment using the $S$ ratio analysis method. By first approximation, the $R$ value, which measures the effectiveness of the $S$ ratio in identifying a hydrogenous spectrum, should increase with increasing cargo thickness and therefore increased interactions. To investigate this hypothesis, the $R$ values from runs of increasing areal densities were normalized by the square root of the corresponding density’s HDPE counts, removing the effects of worsening statistics in high areal densities. Following this adjustment, the $R$ value, or strength of the analysis method, increases as expected until 40 $\frac{g}{cm^2}$, when it begins to decline again. This is presumably due to a competing process either mimicking the effects of hydrogen’s $\sigma_n(E)$ in higher-Z materials or masking the effects of $\sigma_n(E)$ in hydrogenous scans.

An investigation of HDPE spectra in increasing areal density runs shows a gradual increase in sub-1 MeVee energy neutrons relative to total low energy (> 2 MeVee) counts. The fit of an exponential function between 0.5 MeVee and 1.0 MeVee for each spectrum in Fig. 4-6 shows the slope becoming increasingly negative with higher density, indicating the dominance of low energy scatter with increasing cargo thickness.\footnote{The 10 $\frac{g}{cm^2}$ spectrum does not follow the slope trend, due to very low neutron attenuation in 10 cm of HDPE.}

The process of neutron attenuation significant impacts the analyzing power of the
Figure 4-6: Consolidated HDPE spectra in varying areal densities

HDPE transmission spectra of 10 - 70 \( \frac{g}{cm^2} \) phantoms, each recorded during a 20 minute, 10 \( \mu \)A run. The spectrum slope \((m)\) for an exponential fit between 0.5 and 1.0 MeV is detailed by each areal density in the legend. The red line on the 10 \( \frac{g}{cm^2} \) displays the fit boundaries.

technique around 40 \( \frac{g}{cm^2} \), removing the high energy neutrons upon which the \( S \) value relies and masking the effects of \( \sigma_n(E) \) by allowing scattered neutrons to dominate the low energy spectrum. A neutron which undergoes a low angle scatter looses a fraction of its energy and continues on a similar path, entering the detector and confounding lower energy assessments, especially when statistics of non-interacting neutrons are low. Accurate assessments of cargoes with an excess of 40 \( \frac{g}{cm^2} \) hydrogenous materials may be possible with higher neutron fluxes or longer count times, but were not pursued in the scope of this research.

As discussed in Section 3.3.1, the neutron background off beam axis comprises roughly 2% of total counts. A background subtraction procedure matched to count time and beam current was applied to collected spectra. However, this procedure proved insufficient for accurate removal of the low energy scatter, as the type and thickness of scanned cargo plays a significant role in the magnitude and spectrum of room background as well as low-angle scatters entering the detector.
A new set of integration bounds was devised in attempt to mitigate the effects of low energy scatter impacting the $S$ value determination. The universal integration bounds calculated in Section 4.2.2 perform adequately well for all materials at or below $40 \frac{g}{cm^2}$. The second set of bounds, called the adjusted bounds for taxonomic purposes, are based on rejecting the effects of the low energy scatter to test the viability of assessment at greater areal densities. Like the universal integration bounds, the adjusted bounds, \( \{E_0, E_1\} = \{16, 33\} \) and \( \{E_2, E_3\} = \{36, 160\} \) are areal density-independent. They place the low energy integration window at approximately 1 MeVee, where the slopes of the spectra in Fig. 4-6 are more constant across areal densities, indicating less density-specific scatter build up. The high energy integration region is also expanded to correspond with the best statistics in combination with the new low energy region, as determined by the $R$ value search. The ability of both integration regions to determine hydrogenous content was tested by application to heterogeneous cargoes in Section 5.1.
Chapter 5

Results and conclusions

Chapter five presents results and discussions of the technique’s effectiveness and applicability in radiography supporting national security. It also discusses conclusions and possible expansions to the research and application of the spectroscopic technique.

5.1 Application of spectroscopic analysis techniques to heterogeneous cargoes

Several scans of mixed cargoes were performed and grouped by total areal density, e.g., 33 $\frac{g}{cm^2}$ of HDPE and 7 $\frac{g}{cm^2}$ of lead grouped into the 40 $\frac{g}{cm^2}$ section. The integration bounds discussed in Chapter 4 were applied to the PSD-selected neutron transmission spectra of each mixed cargo and resulting $S$ values plotted against the areal density of HDPE present in each scan. Fig. 5-1, Fig. 5-2, and Fig. 5-3 present these plots for total areal densities of 30, 40, and 50 $\frac{g}{cm^2}$.

The spectrum-derived $S$ value increases nearly linearly with increasing HDPE for all material combinations and at all total areal densities until approximately 35 $\frac{g}{cm^2}$ of HDPE. Fig. 5-1 indicates a strong correlation between $S$ value and composition HDPE, with the ability to discriminate between cargoes differing by 2 to 3 cm of
Figure 5-1: $S$ value versus HDPE content in cargoes with 30 $\frac{g}{cm^2}$ total areal density

Figure 5-2: $S$ value versus HDPE content in cargoes with 40 $\frac{g}{cm^2}$ total areal density
Figure 5-3: $S$ value versus HDPE content in cargoes with 50 $\frac{g}{cm^2}$ total areal density. Mixed cargo scans with total areal density held constant and amount of HDPE varied. All scans were 20 minutes and 9-11 $\mu$A. Adjusted integration bounds (see Section 4.2.3) abbreviated to ‘adj’ and universal bounds to ‘uni’.

HDPE. Poor statistics limited the two spectrum-determined $R$ value assessment of resolving power to 40 $\frac{g}{cm^2}$. As $S$ value relies on a single spectra to generate a hydrogenous content estimation, it is more susceptible to the noise occurring at higher HDPE thicknesses. 30 - 35 $\frac{g}{cm^2}$ of HDPE remains the maximum amount the technique is able to resolve, after which incidence of low energy scatter and high energy attenuation drive the $S$ value to lower, non-unique values. Improvements to bolster the resolving limits of this technique and prevent misidentification due to non-unique $S$ values are discussed in Section 5.2.1.

The attempt of the adjusted integration bounds to extract additional information experiences limited success over that of the universal bounds, visible in Fig. 5-2. The $S$ value for 33 $\frac{g}{cm^2}$ of HDPE, 7 $\frac{g}{cm^2}$ of aluminum continues on the linear trend of increasing $S$ value, while the same data point analyzed by the universal bounds shows an $S$ value lower than the point preceding it. The adjusted integration bounds provide some mitigation of the effects of low energy scatter, however they remain insufficient to provide an independent hydrogenous content determination at areal
densities above \(35 \frac{g}{cm^2}\). By design, the universal integration bounds have the lowest fractional error of all possible sets. The merits of both integration region methods may be weighed and applied based on the scanning system requirements. For subsequent assessments applied in this research, unless otherwise stated, the universal bounds are applied due to their lower relative error.

5.1.1 Effect of total cargo areal density on hydrogenous cargo assessment

To determine the impact of various concentrations of non-hydrogenous materials on the assessment of a fixed quantity of HDPE, transmission derived \(S\) value plots were compiled by material type. Fig. 5-4 through Fig. 5-6 present a varying amount of HDPE ‘shielded’ behind carbon, aluminum, and lead, respectively.

The presence of graphite in increasing quantities has a significant impact on the accurate determination of hydrogenous content. Fig. 5-4 shows good agreement be-
Figure 5-5: $S$ value determination of aluminum-shielded HDPE in various areal densities

Figure 5-6: $S$ value determination of lead-shielded HDPE in various areal densities

Mixed cargo scans in Fig. 5-4 through Fig. 5-6 have total areal density held constant and amount of HDPE varied. All scans were 20 minutes and 9-11 $\mu$A. Adjusted integration bounds abbreviated to ‘adj’ and universal bounds to ‘uni’.
tween derived $S$ value and quantity HDPE for total cargo densities of 30 and 40 $g/cm^2$. However, the 50 $g/cm^2$ scans indicate a sharp divergence from the trend of the $S$ value’s direct correlation to HDPE thickness. The datum from the 30 $g/cm^2$ HDPE / 20 $g/cm^2$ graphite scan experiences a $21.52 \pm 0.03\% S$ value change from data at 30 $g/cm^2$ HDPE / 10 $g/cm^2$ graphite and 30 $g/cm^2$ HDPE / 0 $g/cm^2$ graphite scans. This effect is due to a broad, non-Breit-Wigner distributed unresolved resonance region in the $\sigma_n(E)$ of carbon from 1 to 2.3 b, occurring between 7.1 and 8.5 MeV incident neutron energy. The effect of this cross section increase is to preferentially attenuate and therefore remove neutrons from the high energy regions, rendering the $S$ value for a fixed quantity of HDPE lower than the same quantity shielded by an alternate material such as lead. The combined spectra of the data points referenced in this example are plotted in Appendix A, Fig. A-5 and the carbon-induced high energy attenuation visible.

The $S$ value-determined amount of HDPE when shielded by aluminum, presented in Fig. 5-5, shows better agreement, though still suffers some divergent effects at higher total effective areal density. Fig. 5-6 displays excellent agreement between $S$ value and quantity HDPE, which is largely unaffected by the magnitude of higher-Z contribution to the total areal density. Combinations of lead, aluminum, and HDPE permit a quantitative determination of hydrogenous content up to 30 $g/cm^2$, independent of effective areal density.

### 5.2 Radiographs using $S$ value reconstruction

Two HDPE step wedges were assembled in the cargo mock-up and one shielded by 2 cm (5.4 $g/cm^2$) of aluminum. The cargo was scanned at a beam current of 12 $\mu$A for 10 minutes per pixel, using 10 cm x 30 cm pixels. Fig. 5-7 displays a cargo mock-up photograph and a $S$ value reconstructed radiograph side by side. The vertical color scale shows the $S$ value as well as the areal density of HDPE per pixel, calibrated by
Figure 5-7: Experimental set-up and radiograph of shielded and un-shielded HDPE step wedges

a least-squares fitted polynomial.

The cargo mock-up in Fig. 5-8 was scanned at a speed of .03 cm/s with a beam current of 12 μA. The leftmost phantom is 12.7 cm of HDPE shielded by 20.6 cm of aluminum; to its immediate right is 12.7 cm of HDPE shielded by 5 cm of lead. The rightmost two phantoms are 12.7 cm of unshielded HDPE alongside 13.3 cm of aluminum, areal densities of each configuration are listed in the graphic. Using the earlier referenced calibration data, Fig. 5-9 presents the radiograph generated from $S$ values. The image demonstrates the effectiveness of the technique in reconstructing the areal density of hydrogenous materials, even when combined with higher Z materials. Though the aluminum-pure phantom is nearly three times the areal density of the HDPE-pure phantom, the HDPE registers an $S$ value of 0.58 ± 0.007, while the aluminum yields 0.358 ± 0.003. This suggests a strong dependence of the $S$ value on the variations in the $\sigma_n(E)$ of hydrogen as hypothesized.

5.2.1 Discussion and recommendations for improvements

A more accurate correlation of $S$ value to hydrogenous content density is possible given a measure of total areal density with which to compare. This knowledge fur-
Figure 5-8: Cargo schematic and phantom areal densities
From left to right, total phantom areal densities are: 33.3, 70.1, 12.7, and 36.0 $\text{cm}^2/\text{s}$.
Inset shows sidelong view of the mock-up, with 12.7 $\text{cm}^2/\text{s}$ of HDPE behind the aluminum.

Figure 5-9: Transmission radiograph of mock-up in Fig. 5-8
Scan performed at a speed of .03 $\text{cm}/\text{s}$ with a beam current of 12 $\mu\text{A}$. Each pixel contains a relative error of 0.3 to 1.2%. Phantom outlines are overlaid on the radiograph for placement.
ther enables the use of total areal density tailored integration bounds, returning the resolving power of the technique to the values reported in Table 4.2. It is possible to utilize additional information in the spectrum, such as the slope determination in Fig. 4-6 in combination with the effective areal density assessments and $S$ value to generate a set of unique spectrum-derived values for all possible hydrogenous/non-hydrogenous combinations.

Recording spectra which display as few effects as possible resulting from room background is critical for analysis which compares relative counts at different energies. The background subtraction that was attempted as a method of spectrum deconvolution in this research was an oversimplification as the room background changes greatly with cargo type. A concurrently running detector, out of the beam line and shielded identically to the imaging detectors will provide a more accurate determination of the neutron background during each scan. Simulations modelling the background as well as the radiography system as a whole are in progress and will inform future developments and data processing.

Spatial resolution can be drastically improved by the construction of a neutron detection array which spans the height of scanned cargo. At this point, resolution is limited by counting statistics, which may be improved by longer scan times or increased beam current. The scan presented in Fig. 5-9 contained an average relative error of 0.5% per $S$ value assessment at a cargo speed amounting to 3 minutes per 5 x 7.5 cm pixel. To reach a relative error of 0.1% per $S$ value assessment, a similar resolution at a speed of 0.5 $\text{m/s}$ requires a beam current of 1 mA. The $R$ values, or standard deviations of hydrogenous cargo discrimination capability, for this scenario are reported in Table 4.1. The mean density of air cargo commodities classified primarily as 'plastic products' is $212.54 \frac{kg}{m^3}$ [43]. Adjusting for uneven and non-homogeneous loading profiles, the average air cargo ULD container declared as plastic commodities presents a $32 \frac{g}{cm^2}$ areal density of hydrogenous material along its scanned 1.5 m
axis. All remaining classifications of cargo may include some plastic or hydrogenous components, but likely not in areal densities exceeding those of the plastic products designated containers.

Increased beam current translates to increased dose to cargo and increased shielding requirements. In this system, dose is proportional to the $R$ value squared, necessitating a balance between accurate reconstruction and risk to cargo and operators. In addition to compliance with ANSI N42.46 standards, neutron radiographic systems are subject to ANSI/HPS N43.14-2011: Radiation Safety for Active Interrogation Systems: Safe Operating Practices for Active Interrogation Systems for Security Screening Using Fast Neutrons [44]. A shielding and radiological nonproliferation benefit lies in the on/off nature of a nuclear reaction induced by a particle accelerator, which requires operational shielding only when in use as opposed to radioactive sources.

5.3 Conclusions and further research

The development of more sophisticated algorithms utilizing principle component analysis and other predictive modelling strategies can potentially provide more accurate cargo type reconstruction with the same or fewer statistics, thereby reducing cargo dose. Future work should also focus on obtaining a clearer signal from neutrons that have truly transited the cargo limiting or removing background.

Three dimensional radiographs are possible using this system, one such method is the use of the filtered back projection (FBP) algorithm, which is the computed tomography industry standard for three dimensional sample reconstructions. In a cargo scanning system, the container can be rotated on a stage through a fan beam as radiographs are generated at various angles. The FBP algorithm sums the back projected radiographs through a filtering deconvolution function that contains a posi-
tive core with negative side lobes to cancel image blur, thereby generating a top down view from a series of side-on images [45,46].

This technique may be applied to any neutron scanning system employing energy sensitive detectors and a quasi-monoenergetic neutron beam sufficiently spaced to exploit the $\sigma_n(E)$ of hydrogen. Mono-energetic sources, such as a 14.1 MeV D-T generator, may be selectively moderated to replicate the multiple neutron energy groups produced in a nuclear reaction. The application of this technique is well suited to ruggedized and rapid-deployable field uses in which the complex apparatus required for FNRR or other radiographic assessments are infeasible. In more permissible environments it can also be applied as the density reconstruction function necessary for FNRR.

This research has demonstrated the feasibility of radiography based on a spectral analysis technique applied to nuclear reaction produced neutron groups. Extrapolation of the results to scenarios with high container throughput speeds suggests the suitability of the technique in field-use environments. The salient aspect of this research is the ability of complementary neutron and $\gamma$-ray systems to provide a comprehensive radiographic assessment of sealed cargo using the $^{11}$B(d,n$^\gamma$)$^{12}$C reaction.
Appendix A

Supplemental information
Figure A-1: Distribution of average cargo density
Histogram data taken on the 14 highest throughput days between June 2004 and June 2005 at the Port of Long Beach, CA. [11].

Figure A-2: Simulated $\gamma$ spectrum as an effect of neutron interactions
GEANT4 simulation of a NaI(Tl) response to 4.4 and 15.1 $\gamma$ peaks with and without neutron background. The simulation was programmed for the $\gamma$ radiography system to determine neutron effects in resolving the peaks of interest, but serves to illustrate $(n,\gamma)$ and $(n,n')$ contributions to the radiation environment of the experiment. Plot courtesy of Z. S. Hartwig, MIT PSFC.
Figure A-3: Spectral effects of removing post-cargo collimation
Plots are indicative of the effects of removing the post-cargo collimators as described in Section 3.2.2. The example shows the normalized, combined spectra from various cargoes of 40 $\frac{g}{cm^2}$; the top spectrum is with both collimators in place and the bottom is with the post-cargo collimators removed. Bin-to-bin variation in counts for the HDPE spectrum is slightly lessened without the scattering effects of the second collimator set. This effect is present to some degree in all measurements of cargoes from 10 to 50 $\frac{g}{cm^2}$ areal density.
Figure A-4: Technical drawing and specifications for EJ-309 detector
<table>
<thead>
<tr>
<th>Areal density [g cm⁻²]</th>
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<th>Integration regions</th>
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<td>Pb</td>
<td>4.94</td>
<td>16-26 84-136</td>
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Note: Integration step size, $E_0$, is 0.06 MeV; all runs 20 minutes and 8-10 μA.
Figure A-5: Combined transmission spectra for heterogeneous graphite and HDPE cargo scans
Spectra plotted to illustrate an overall increase in attenuation with the incremental addition of graphite, as well as the preferential attenuation of high energy neutrons due to a broad resonance region. All scans were 20 minutes and 10 $\mu$A.
Bibliography


[21] C.M. Class, J.E. Price, and J.R. Risser. Angular distributions from the $^{11}$B(d,n$\gamma$)$^{12}$C and $^{11}$B(d,n$\gamma$)$^{12}$*C reactions for deuteron energies from 1.5 to 4.7 MeV. Nuclear Physics, 71:433–440, 1965.


