Characterization of Fissile Material Using Low Energy Neutron Interrogation
by
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Characterization of Fissile Material Using Low Energy Neutron Interrogation

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Abstract

The glaring need to develop methods for detecting and interdicting illicit nuclear trafficking has resulted in the exploration of various methods for active neutron interrogation, specifically for the presence of special nuclear material (SNM) in cargo containers. The proposed system aims to defeat the ability of terrorists to import SNM into the United States via maritime shipping, thus greatly reducing the possibility of a successful nuclear terrorist attack.

The proposed system uses 60-100 keV neutrons, produced by the $^7\text{Li}(p,n)^7\text{Be}$ reaction in a linear accelerator and kinematically beamed into various targets. In the event that fissile material is present, highly energetic neutrons will be emitted from the fissioning of a nucleus and some of these neutrons will eventually radiate from the container. Inevitably, high energy photons will also radiate from the target due to the interactions of neutrons and host materials. Utilizing a neutron detection system that is able to discriminate low energy neutrons, high energy gamma rays and the high energy neutrons from fission enables the detection of fissile material in various containers. An increase in discriminated high energy neutron events during active neutron interrogation selectively indicates the presence of SNM, since neutron energies on the order of 1 MeV are required for the SNM-equivalent fissioning potential when incident upon $^{238}$U and other high-Z nuclei. Furthermore, neutrons with less than approximately 100 keV do not undergo nuclear processes such as (n,2n) and (n,n'), but rather lose their energy through kinematic collisions.

Results obtained validate this proof-of-concept, in that observed high energy neutron events increase significantly in the presence of gram-quantities of SNM. Further, attempts made to shield the SNM from active interrogation do not defeat the proposed system's ability to identify the presence of SNM.

With a fully-functional proof-of-concept, further work towards developing a complete and deployable prototype active neutron interrogation system will serve to augment the ability of the United States to detect, deter and interdict illicit nuclear trafficking.

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## Table of Contents

Abstract............................................................................................................................... 3
Acknowledgments............................................................................................................. 4
1.0 Introduction................................................................................................................... 9
  1.1 Motivation............................................................................................................. 9
  1.2 Background............................................................................................................ 10
  1.3 Thesis Objectives.................................................................................................... 11
Task Descriptions...................................................................................................... 11
2.0 Low Energy Neutron Interrogation Theory............................................................... 13
  2.1 Criteria for Selection of Neutron Energy.............................................................. 13
  2.2 Theoretical Neutron Interrogation Effectiveness.................................................... 19
    2.2.1 Neutron Transport in Various Media with Varied Energy.............................. 20
    2.2.2 Neutron Interrogation Effectiveness in Simulated Cargo Environments......... 29
3.0 Lithium Target Design.............................................................................................. 37
  3.1 Lithium Target Description..................................................................................... 37
  3.2 Thick Target Neutron Yield.................................................................................. 39
4.0 Neutron Detection System......................................................................................... 43
  4.1 Description of Hardware........................................................................................ 43
    Liquid Organic Scintillator Detectors....................................................................... 44
    Tandem Particle Accelerator..................................................................................... 46
    Digital Oscilloscope.................................................................................................. 47
  4.2 Digital Pulse Shape Discrimination........................................................................ 48
5.0 Fissile Material Characterization............................................................................... 63
  5.1 Bare Fissile Material Induced Signal...................................................................... 65
  5.2 Effect of Shielding on Induced Signal................................................................... 67
6.0 Future Work and Conclusions.................................................................................... 71
  6.1 Conclusions............................................................................................................. 71
  6.2 Future Work............................................................................................................ 74
References......................................................................................................................... 76
List of Figures

Figure 1 - Total Fission Cross Sections for Various Isotopes [Lanza, 2006].................. 15
Figure 2 - Total Fission Cross Sections for U235 and Pu239 [ENDF/B-VI].................. 17
Figure 3 - Neutron Flux-Dose Conversion (ANSI-91)........................................... 18
Figure 4 - Neutron Absorption Cross Sections for H, Al and Fe.......................... 19
Figure 5 - Penetration MCNP Problem Geometry [Johnson, 2006]......................... 21
Figure 6 - Total Neutron Flux Through HDPE (60 keV neutrons) [Johnson, 2006]...... 22
Figure 7 - Total Neutron Flux Through HDPE (14 MeV neutrons) [Johnson, 2006]..... 23
Figure 8 - Total Neutron Flux Through Concrete (60 keV neutrons) [Johnson, 2006]... 24
Figure 9 - Total Neutron Flux Through Aluminum (60 keV neutrons) [Johnson, 2006] 25
Figure 10 - Total Neutron Flux Through Aluminum (100 keV neutrons) [Johnson, 2006] .......................................................... 26
Figure 11 - Total Neutron Flux Through Tungsten (100 keV neutrons) [Johnson, 2006] 27
Figure 12 - Total Neutron Flux Through Tungsten (14 MeV neutrons) [Johnson, 2006] 27
Figure 13 - MCNP Problem Cargo Geometry [Johnson, 2006]............................... 30
Figure 14 - Fast Neutron Flux With HEU [Johnson, 2006]..................................... 31
Figure 15 - Fast Neutron Flux With Lead [Johnson, 2006]...................................... 31
Figure 16 - Fast Neutron Flux With Pu [Johnson, 2006]........................................ 31
Figure 17 - Fast Neutron Flux With Concrete Shielded (5 cm) HEU [Johnson, 2006]... 33
Figure 18 - Fast Neutron Flux With Concrete Shielded (25 cm) HEU [Johnson, 2006] 33
Figure 19 - Lithium Target Assembly....................................................................... 37
Figure 20 - Proton Energy Vs. Depth in Lithium [Blackburn, 2006].............................. 40
Figure 21 - Cross Section Vs. Depth in Lithium [Blackburn, 2006].............................. 41
Figure 22 - Scionix Liquid Organic Scintillator Detector........................................ 44
Figure 23 - NSI Tandem Linear Accelerator.............................................................. 47
Figure 24 - Neutron and Gamma-ray Pulse Shapes in Liquid Organic Scintillators ...... 49
Figure 25 - Neutron/Gamma-ray Discrimination from DT Neutron Generator............ 50
Figure 26 - Histogram of Counts from DT Neutron Generator.................................. 52
Figure 27 - Total Charge Deposition Spectra from DT Source.................................. 54
Figure 28 - Neutron/Gamma-ray Discrimination from AmBe Neutron Source .......... 55
Figure 29 - Histogram of Counts from AmBe Neutron Source.................................... 56
Figure 30 - Total Charge Deposition Spectra from AmBe Source............................... 57
Figure 31 - Neutron/Gamma-ray Discrimination from DD Neutron Generator.......... 58
Figure 32 - Histogram of Counts from DD Neutron Generator.................................. 59
Figure 33 - Total Charge Deposition Spectra from DD Source.................................... 60
Figure 34 - Experimental Setup for Active Neutron Interrogation of SNM............... 64
Figure 35 - Neutron/Gamma-ray Discrimination with No SNM................................. 66
Figure 36 - Neutron/Gamma-ray Discrimination with Unshielded SNM................. 67
Figure 37 - Neutron/Gamma-ray Discrimination with 1" HDPE Shielded SNM............ 68
Figure 38 - Neutron/Gamma-ray Discrimination with 2" Ricorad Shielded SNM........... 69
Figure 39 - Low Energy Neutron Interrogation Results Summary.............................. 71
Figure 40 - Complete Waveform Integration Values for Interrogation Sessions........... 72
Figure 41 - Neutron Event Differences from Background for Interrogation Sessions .... 73
List of Tables

Table 1 – Penetration Depths for Neutrons of Varied Energy in Select Materials
   [Johnson, 2006]........................................................................................................ 28

Table 2 – Fission Probability per Source Neutron [Kerr, 2005]................................. 35

Table 3 – Near Threshold Lithium Compound Target Yields [Lee, 1998]...................... 38

Table 4 – Thick Target Neutron Yields Versus Incident Proton Energy [Lee, 1998].... 40
1.0 Introduction

Since 1993, there have been over 500 documented cases of illicit trafficking of nuclear and radiological materials [GAO Report, 2005]. Each year, approximately 7 million shipping containers transit seaports in the United States [GAO Report, 2005]. With less than 3% of these containers undergoing any type of security screening, the magnitude of vulnerability becomes alarming. Should a terrorist organization succeed in smuggling a nuclear weapon into the United States followed by detonation, the results would be catastrophic – both in terms of loss of life and economic ramifications.

1.1 Motivation

To address the vulnerability inherent in the shipping and transportation industry, the United States Government has invested heavily in new technologies for container security. The largest efforts to date involve collaboration between multiple national laboratories trying to assess and improve port security at many foreign seaports, including the installation of radiation detectors. The goal of this program is to deter and interdict illicit nuclear proliferation activity at foreign ports, before the nuclear material can reach American soil.

Although these radiation monitors installed at foreign ports of interest are effective in detecting sources of radioactivity that could possibly be used in a Radiological Dispersal Device (RDD), they are not as effective in detecting Special Nuclear Material (SNM). Depending on the type of SNM (various forms of Pu, U, Th, etc.), the radiation signal emitted may not be detectable by installed radiation monitors or
can be easily shielded. Such deficiencies can be addressed by using active neutron interrogation techniques [Slaughter, 2003].

1.2 Background

Among the new technologies being developed for the detection of SNM in cargo containers is the use of low energy neutron interrogation. Specifically, 60-100 keV neutrons are produced by the $^7\text{Li}(p,n)^7\text{Be}$ reaction in a linear accelerator and are kinematically beamed into various targets. In the event that fissile material is present, highly energetic neutrons will be emitted from the fissioning of a nucleus and some of these neutrons will eventually radiate from the container. Inevitably, high energy gammas will also radiate from the target due to the interactions of neutrons and host materials.

Utilizing a neutron detector that is able to discriminate low energy neutrons, high energy gamma rays and the high energy neutrons from fission will enable the detection of fissile material in various containers. An increase in high energy neutron flux during active neutron interrogation will indicate the presence of SNM specifically, since neutron energies on the order of 1 MeV are required for efficient fissioning of U238 and other high-Z elements [Johnson, 2006]. Furthermore, neutrons with less than approximately 100 keV do not undergo nuclear processes such as $(n,2n)$ and $(n,n')$, but rather lose their energy through kinematic collisions [Dietrich, 2004].

Lawrence Livermore National Laboratory has designed and tested an RFQ-based 60 keV neutron interrogation system with limited configurations [Dietrich, 2004]. It has been suggested that this method of neutron interrogation is able to detect and discriminate the induced neutron and/or gamma radiation in both fissile and non-fissile materials.
Specifically, small amounts of highly enriched uranium have been detected using a low-dose, low energy neutron interrogation system.

The success of preliminary testing has validated the basic principles of this concept, yet many variables have not been investigated. Such variables include: the implementation of a digitizer to increase the performance of neutron discrimination independent of gamma and neutron fluxes, the characterization of bare Plutonium and highly enriched Uranium signatures, the effects of various methods of shielding, detector position, source neutron energy as well as many other variables.

1.3 Thesis Objectives

The goal of this thesis is to (1) implement a neutron detection system on the Van de Graff accelerator (used in this case as a low energy neutron generator) capable of discriminating low energy neutrons, high energy gammas and high energy neutrons from fission. Furthermore, this detection system will be used (2) to characterize SNM sources in various configurations and to (3) perform blind tests for the identification of fissile material.

Task Descriptions
1. The cornerstone of this project will be the integration of a neutron detection system on the linear accelerator. Research will have to be done to decide what size and configuration of neutron detector to use and more importantly, this neutron detector will have to be coupled to a digitizer and data acquisition system. The efficiency of this neutron detection system is crucial to the project’s ability to detect and discriminate different types of radiation, such as low energy neutrons, high energy gammas and high energy neutrons from fission.
2.1 – Once the neutron detection system is assembled and functioning, the first step will be to characterize the neutron flux and spectra from the linear accelerator and target setup.

2.2 – Next, natural and induced radiation signals of various forms of bare SNM can be characterized. This data will serve as reference for further neutron interrogation studies in various configurations.

2.3 – Further, the effects of shielding material and configuration will be investigated. This will allow for the efficiency and overall feasibility of the neutron interrogation system to be gauged. Based on these results, slight changes to the system may be made.

3 – Lastly, the proof of principle concept will be tested with a series of blind tests seeking to identify the presence/absence of SNM. Once this ability has been demonstrated, further research in neutron interrogation techniques and progress towards Phase II and III of the low energy neutron interrogation project [Lanza, 2005] can commence.
2.0 Low Energy Neutron Interrogation Theory

The most effective neutron interrogation system will have a combination of deep penetration ability and high fissioning rate (in the presence of SNM) per source neutron. Unfortunately, these two characteristics are contradictory, in that increased penetration capability calls for higher source neutron energies, while lower source neutron energies increase the probability of an interaction between a source neutron and fissile atom resulting in a fission event. Simply increasing the source neutron energy would improve penetration capabilities, but would increase the risk of fissioning non-fissile isotopes such as uranium-238 and other high-Z elements. Additionally, higher energy neutrons will appear similar to neutrons from fission events, complicating the discrimination system. Conversely, lowering neutron energies to an arbitrarily low level might severely limit the system’s ability to scan deep within containers.

Due to this conflict, careful design considerations must be implemented to ensure the end product’s effectiveness. Probabilistic analysis using Monte Carlo simulation models can be used to determine the optimal neutron source characteristics [Johnson, 2006]. The following sections present the methodology used to determine optimal source neutron energies as well as their simulated effectiveness.

2.1 Criteria for Selection of Neutron Energy

The keystone of this project is the selection of source neutron energy, as this will ultimately dictate the overall system efficiency. The source neutron energy directly affects the penetration ability, fissioning rate and SNM selectivity of the interrogation
system. For the neutron interrogation system presented here, it has been determined that the optimal source neutron energy range is from 60 keV to 100 keV.

In terms of radiation transport and penetration, higher energy neutrons travel farther in various media than neutrons of lower energies when considering only elastic scattering. The two most significant factors in determining a neutron’s penetration is the rate of energy loss with respect to distance and the mean free path of interactions (density dependent). The maximum energy that a neutron of mass \( M \) and kinetic energy \( E \) can transfer to a nucleus of mass \( m \) in a single elastic collision is given by equation 2.1 below [Turner, 1995]:

\[
Q_{\text{max}} = \frac{4mME}{(M + m)^2}
\]  

2.1

Simply put, higher energy neutrons have more kinetic energy to lose in elastic collisions and therefore must undergo more scattering events through larger distances before their final interaction. For an SNM detection system, this ability to scan deeper into containers or through layers of shielding is extremely valuable. Unfortunately, the presence of higher energy neutrons can negatively affect the accuracy of the system in detecting fissile material. Non-fissile isotopes such as uranium-238 and thorium-232 have relatively negligible fission cross sections for neutrons less than approximately 500 keV, with rapidly increasing cross sections thereafter as shown in Figure 1 [ENDF/B-VI] below.
The figure above clearly shows that while the total fission cross sections for uranium-235 and plutonium-239 are relatively high throughout the depicted energy range, non-fissile isotopes such as uranium-238 and thorium-232 typically need a much higher neutron energy to undergo a fission event with comparable probability, i.e. greater than 600 keV. Note that neutron energies in excess of 1 MeV are needed before the total fission cross section for uranium-238 is comparable to uranium-235. With a high-energy neutron interrogation system, one risks mistaking benign fissile material for SNM. In the maritime shipping industry, detectors that have high false positive alarm rates would result in very high costs of inspection and time lost – two factors that most shipping agents are not willing to sacrifice. For this proposed application, liquid organic scintillators have many beneficial properties which make them uniquely efficient.
A benefit to using low energy source neutrons with a liquid organic scintillator neutron detection system (explained further in Chapter 4.0) is that the relative discrimination efficiency for neutrons below several hundred keV is extremely low [Knoll, 2000]. This is simply an effect of the light emission from elastic scattering of neutrons with recoil nuclei; neutrons below several hundred keV simply don't transfer enough energy to the recoil proton for significant light output. Therefore, liquid organic scintillators are effectively blind to the source neutron population while remaining highly effective at discriminating fission energy neutrons (discussed further in Chapter 4.2). Utilizing a high-energy neutron source would diminish this SNM selectivity and force a reliance on accelerator pulse-trigger timing for determining the source of high energy neutrons.

It is well known that fission cross sections for fissile material (e.g. uranium-235 and plutonium-239) are much larger at lower incident neutron energies. In fact, the total fission cross section for uranium-235 at thermal neutron energies (0.0235 eV) is more than a factor of 280 times the same cross section for 14 MeV neutrons [Johnson, 2006], as indicated in Figure 2.

Roughly the same sensitivity to lower energy neutrons is exhibited by plutonium-239 as illustrated in Figure 2 below. Note that from approximately 10 keV to 14 MeV, there is little increase in the cross sections for both U235 and Pu239; however, dramatic increases are seen as incident neutron energies are moderated and lose energy. Although some resonance fission cross sections are exhibited by U238 at lower neutron energies, the overwhelming effect of using low energy source neutrons is improved fissile material selectivity.
Figure 2 – Total Fission Cross Sections for U235 and Pu239 [ENDF/B-VI]

An additional motivation to utilize low energy source neutrons is the omnipresent need to minimize the total possible radiation dose to the public and personnel. As depicted in Figure 3 below, direct absorbed dose in tissue increases exponentially with increasing neutron energy. 14 MeV neutrons deliver 40 times the direct absorbed dose than 60 keV neutrons, thus necessitating the need for much more shielding/personnel control. Also, increased neutron activation is noted at higher source neutron energies due to reactions such as (n, 2n) [Kerr, 2005].
Further, 14 MeV deuterium-tritium neutron generators are unable to produce naturally forward directed beams due to the isotropic nature of the reaction kinetics. However, the $^7$Li($p,n$)$^7$Be reaction is kinematically beamed forward, with an average neutron emission angle of 51° for proton energies of 2.1 MeV (75 keV neutrons) [Lee, 1998].

Unfortunately, lower energy neutrons are also more readily absorbed by common materials such as hydrogen, aluminum and iron. Figure 4 illustrates the general trend of neutron absorption cross sections from thermal energies to 14 MeV. Lower energy neutrons are more likely to be absorbed by these materials than high energy neutrons; however, the cross sections for (n,g) reactions are still relatively low (on the order of
barns) compared to the fission cross sections for SNM at the same energy (hundreds of barns).

![Graph showing neutron absorption cross sections for H, Al, and Fe (ENDF/B-VI)](image)

Given the complicated nature of the innumerable, energy-dependent cross sections, a great deal of Monte Carlo simulation is needed. Without tangible evidence of overall neutron interrogation system effectiveness using a variety of materials, the selection of optimal source neutron energy is not possible. MCNP code can be used to attain invaluable information regarding neutron flux through materials as well as simulated induced fission signal output.

### 2.2 Theoretical Neutron Interrogation Effectiveness

Extensive modeling using MCNP simulations can help determine the optimal source neutron energy for use in an effective neutron interrogation system [Johnson,
2006; Kerr, 2005; Dietrich, 2004]. The first order of business is to assemble a visual data set representing the benefits and ramifications of utilizing different source neutron energies in various media. This visual data set consists of mesh tallies overlaid over the problem geometry giving two-dimensional representations of neutron flux penetration. This allows the project management to assess the possible benefits of using lower energy neutrons versus the above stated consequences associated with this option. Once optimal source neutron energy is selected, the overall neutron interrogation effectiveness in simulated cargo environments can be assessed. This ultimately determines the feasibility of the proposed system and helps to set operational parameters for improved performance.

2.2.1 Neutron Transport in Various Media with Varied Energy

The primary Monte Carlo simulation study used for this project [Johnson, 2006] was performed utilizing three incident neutron energies: 60 keV, 100 keV and 14 MeV. The neutron sources are all modeled as ideal monoenergetic beams directed in a single direction towards the center of one face of a cube (500 cm sides) of varied atomic composition. The MCNP problem geometry is illustrated by Figure 5 below. A sphere of air with radius of 500 cm surrounds the cube to account for possible scattering events outside of the material where neutrons may be reflected back into the cube of material.
A cylindrical mesh with the origin at the point nearest to the neutron source is selected for the tallying method. This method allows for the flux to be symmetrically projected across the radial and vertical cross section of the cube. Contour plots with the scaled flux plotted versus radial and vertical distance in the cube allow for the visual characterization of penetration ability for source neutrons of various energies.

With regards to neutron transport, several key categories of matter are most representative of the different modes of neutron propagation. These include elements ranging from high atomic number (high-Z) to low-Z elements and hydrogenous material. Due to the nature of elastic collisions, neutrons do not lose energy in collisions at the same rate in high-Z material as low-Z material, with hydrogenous material being most effective at stopping neutrons in the least number of interactions. However, certain materials exhibit resonance absorption cross sections complicating a blanket approach to determine penetration depths for neutrons based on energy.
Two hydrogenous materials that are fairly common and thus likely to be used in an attempt to shield SNM from an active neutron interrogation system are high-density polyethylene (HDPE) and concrete. Both of these materials contain significant amounts of hydrogen nuclei and also carbon. Obviously, carbon is not a hydrogenous material, but as a low-Z moderator it contributes significantly to scattering and thermalizing neutrons. Figures 6 and 7 below illustrate the penetration characteristics of 60 keV and 14 MeV neutrons through HDPE, respectively. The scaled flux is shown by the colored contour plot overlaid over the problem geometry described above. The axes scale here represents the first quadrant on the XZ plane of Figure 5 above.

Figure 6 – Total Neutron Flux Through HDPE (60 keV neutrons) [Johnson, 2006]
As expected, the penetration ability of 60 keV neutrons through hydrogenous material is greatly attenuated, while 14 MeV neutrons appear to be somewhat less affected. Although 14 MeV neutrons have much greater penetration ability in hydrogenous material, the penetration depth is no larger than twice that for any given flux [Johnson, 2006]. Due to the high rate of energy transfer between the neutrons and hydrogen nuclei, as well as the increased tendency for neutron absorption at lower neutron energies (Figure 4), the disparity in neutron penetration ability between 60 keV and 14 MeV is greatest for hydrogenous material. Concrete exhibits similar penetration characteristics (Figure 8 below), although the penetration ability is somewhat larger due to the reduced hydrogen nuclei density when compared to HDPE. Although it may appear that low energy source neutrons might be defeated by shielding with hydrogenous
material, these data simply indicate a need for longer interrogation duration when hydrogenous shielding is detected (possibly via x-ray).

![Figure 8](image)

**Figure 8 – Total Neutron Flux Through Concrete (60 keV neutrons) [Johnson, 2006]**

Common low-Z (non-hydrogenous) materials that may likely be found in cargo environments are steel (primarily iron-56) and aluminum. The total neutron flux characteristics for steel are strikingly similar to those found for concrete (Figure 8); however, inspection of the energy spectrum reveals that the thermal neutron flux at any given penetration depth in steel is approximately 3 orders of magnitude less than that found in concrete [Johnson, 2006]. This can be attributed to the resonance absorption cross sections present in both steel and aluminum (Figure 4).

These resonances lead to a surprising result: When modeling the penetration capabilities of low energy neutrons and 14 MeV neutrons in aluminum, it was found that
60 keV neutrons actually have a greater penetration depth than neutrons of higher energy [Johnson, 2006]. Figures 9 and 10 below illustrate the penetration characteristics of 60 keV neutrons and 100 keV neutrons in aluminum.

Figure 9 – Total Neutron Flux Through Aluminum (60 keV neutrons) [Johnson, 2006]

Also significant is the distance at which notable thermalization of the impinging neutron beam occurs: 115 cm for the 60 keV beam and 125 cm for the 100 keV neutron beam. Therefore, one can conclude that the presence of aluminum in shipping containers does not readily interfere with the neutron interrogation system at lower source neutron energies.
Depending on the nuclear trafficker’s level of sophistication, attempts may be made to shield the SNM with high-Z elements such as lead or tungsten. As explained by Equation 2.1, neutrons transfer much less energy per collision with high-Z nuclei than with lower-Z nuclei (1/Z dependence). Thus, a large mechanism of flux attenuation for neutrons propagating through high-Z media is due to absorption. Given the resonant nature of absorption cross sections in many materials, this may have little effect on source neutrons of lower energy compared to those with energies above a resonance. This behavior is shown in figures 11 and 12 below, which provides total neutron fluxes through tungsten for 100 keV neutrons and 14 MeV neutrons.
Figure 11 – Total Neutron Flux Through Tungsten (100 keV neutrons) [Johnson, 2006]

Figure 12 – Total Neutron Flux Through Tungsten (14 MeV neutrons) [Johnson, 2006]
Table 1 summarizes the penetration characteristics of neutrons of various energies in different media [Johnson, 2006]. Penetration here is defined as the point where the flux is reduced to $10^{-4}$ neutrons/cm²/source neutron. Penetration $\Delta$ is defined as the distance required for the flux to be reduced from $10^{-3}$ to $10^{-4}$ neutrons/cm²/source neutron. Note that penetration depths vary most dramatically for hydrogenous shielding material, while those for aluminum and iron are less dramatic and even somewhat diminished for higher energy neutrons.

Table 1 – Penetration Depths for Neutrons of Varied Energy in Select Materials [Johnson, 2006]

<table>
<thead>
<tr>
<th>Material</th>
<th>Energy (keV)</th>
<th>Average Mean Free Path (cm)</th>
<th>Penetration (cm)</th>
<th>Penetration $\Delta$ (cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>HDPE</td>
<td>60</td>
<td>0.45</td>
<td>20</td>
<td>15</td>
</tr>
<tr>
<td></td>
<td>100</td>
<td>0.46</td>
<td>20</td>
<td>10</td>
</tr>
<tr>
<td></td>
<td>14000</td>
<td>3.02</td>
<td>55</td>
<td>25</td>
</tr>
<tr>
<td>Concrete</td>
<td>60</td>
<td>1.05</td>
<td>30</td>
<td>15</td>
</tr>
<tr>
<td></td>
<td>100</td>
<td>1.05</td>
<td>30</td>
<td>15</td>
</tr>
<tr>
<td></td>
<td>14000</td>
<td>3.03</td>
<td>45</td>
<td>25</td>
</tr>
<tr>
<td>Steel</td>
<td>60</td>
<td>1.80</td>
<td>35</td>
<td>15</td>
</tr>
<tr>
<td></td>
<td>100</td>
<td>2.02</td>
<td>35</td>
<td>20</td>
</tr>
<tr>
<td></td>
<td>14000</td>
<td>3.07</td>
<td>60</td>
<td>30</td>
</tr>
<tr>
<td>Aluminum</td>
<td>60</td>
<td>13.80</td>
<td>70</td>
<td>45</td>
</tr>
<tr>
<td></td>
<td>100</td>
<td>13.10</td>
<td>50</td>
<td>30</td>
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<tr>
<td></td>
<td>14000</td>
<td>16.20</td>
<td>75</td>
<td>45</td>
</tr>
<tr>
<td>Tungsten</td>
<td>60</td>
<td>1.38</td>
<td>25</td>
<td>15</td>
</tr>
<tr>
<td></td>
<td>100</td>
<td>1.55</td>
<td>25</td>
<td>10</td>
</tr>
<tr>
<td></td>
<td>14000</td>
<td>1.92</td>
<td>45</td>
<td>15</td>
</tr>
<tr>
<td>Depleted U</td>
<td>60</td>
<td>1.82</td>
<td>25</td>
<td>10</td>
</tr>
<tr>
<td></td>
<td>100</td>
<td>1.82</td>
<td>25</td>
<td>10</td>
</tr>
<tr>
<td></td>
<td>14000</td>
<td>2.21</td>
<td>55</td>
<td>20</td>
</tr>
</tbody>
</table>

The decision to pursue the design of a low energy neutron interrogation system is based on the penetration characteristics derived through these MCNP simulations and the issues discussed in Chapter 2.1. Through the use of a lithium-7 neutron generator, varied neutron energies in the range of 60 – 100 keV are easily achieved without modification of
hardware. This system’s overall neutron interrogation effectiveness in simulated cargo environments as modeled in MCNPX is presented in the next section.

2.2.2 Neutron Interrogation Effectiveness in Simulated Cargo Environments

A thorough study of the feasibility of a low energy neutron interrogation system in simulated cargo environments was performed [Johnson, 2006] to evaluate the ability and effectiveness of the proposed system to detect SNM. This study analyzes the induced fission neutron signal in SNM in various configurations, including unshielded and shielded with hydrogenous material. The ability to predict whether the proposed system would be able to detect SNM while shielded with hydrogenous material is paramount, in that if this system was deemed unable to do so there would be little use in developing the technology.

A basic cargo container is modeled as a 3 meter tall cylindrical shell with a radius of 1.5 meters and filled with air at standard temperature and pressure. Several layers exist within the shell which may comprise shielding. The target material is either highly enriched uranium (HEU) enriched to 93.5% (atom %) uranium-235, pure lead, plutonium (91.95 atom % plutonium-239) or depleted uranium (DU) (0.25 atom % uranium-235). This target material is situated in the center of the cylindrical cargo shell as depicted in Figure 13 below. The neutron source consists of an MCNP modeled source [Kerr, 2005] that is described further in Chapter 3. The average source neutron energy is 60 keV.
The first step in this stage of simulation is to analyze the induced neutron signal in the cargo geometry with fissile material present and no shielding. For a benchmark, 3 different scenarios were simulated: 1 – 7 kg of HEU; 2 – 4.5 kg of Pb; 3 – 1 kg of Pu. Plotting the total neutron flux against the cargo geometry for all three cases yielded virtually identical graphs; however, this is simply due to the fact that both cases with SNM were subcritical assemblies and thus no dramatic neutron flux multiplication occurred. Although the total neutron flux in each case is similar, the difference in neutron energy spectrum for fissile and non-fissile scenarios can be exploited. When the mesh tally was binned according to energy and only the fast neutron (kinetic energy > 1 MeV) flux was plotted against the cargo geometry (Figures 14 – 16), the appearance of SNM was unmistakable, as 60 keV neutrons are incapable of inducing fission in lead.
Figure 14 – Fast Neutron Flux With HEU [Johnson, 2006]

Figure 15 – Fast Neutron Flux With Lead [Johnson, 2006]

Figure 16 – Fast Neutron Flux With Pu [Johnson, 2006]
It is interesting to note that neutrons emitted naturally through spontaneous fission events can be neglected in HEU, as a one kilogram will emit 0.98 fission neutrons/sec [Johnson, 2006]. Averaging this over $4\pi$ and at a distance of even half a meter reduces this signal to well below background. However, dependent on the atomic fraction of plutonium-240 in the plutonium target material, spontaneous fission events can be as high as $5.21 \times 10^4$ fission neutrons/sec/kg [Johnson, 2006]. Depending on the distance of the detector and level of shielding, this natural signal may be detectable without active neutron interrogation. This signal, however, should not be viewed as interfering with the effectiveness of an active neutron interrogation system because any high energy neutrons detected would be indicative of SNM, regardless of whether they were induced by source neutrons or spontaneous events.

With proven neutron interrogation effectiveness with bare SNM sources, the focus shifts to the effect of shielding on induced neutron signals. Here, it is only necessary to model cargo simulations using hydrogenous material as shielding due to the fact that this type of shielding is the most effective at attenuating any neutron fluxes. Use of any other type of shielding materials for this energy range would only diminish its effectiveness and result in improved induced neutron signals. However, the amount of hydrogenous shielding used is extremely influential in two very contrasting manners: Minimal amounts of shielding will augment the induced neutron signal, while larger amounts of shielding will suppress the induced neutron signal. If only a small amount of hydrogenous shielding is used, the overall effect will simply be to moderate the source neutrons down towards thermal energies and disproportionally increase the effective fission cross sections. On the other hand, if excess moderating material is used neutrons
will be reflected or slowed down and captured, thus greatly reducing the induced neutron flux.

Figures 17 and 18 below illustrate the effect of adding various levels of hydrogenous shielding. In the simulations, a uniform layer of shielding material was added surrounding the target material and the resulting fast neutron fluxes can be compared to the previous bare source simulations.

Figure 17 – Fast Neutron Flux With Concrete Shielded (5 cm) HEU [Johnson, 2006]

Figure 18 – Fast Neutron Flux With Concrete Shielded (25 cm) HEU [Johnson, 2006]
Comparing Figure 14 with Figure 17 shows that a small amount of hydrogenous material is not only ineffective at shielding source neutrons, it actually increases the induced fission neutron flux. Higher flux levels are present at further distances from the target material in the shielded case than with the bare SNM scenario. However, the opposite is true when 25 cm of concrete shielding are used. Figure 18 shows that a drastically reduced fast neutron flux is present throughout the cargo geometry, indicating that source neutrons are being effectively shielded and that induced fission neutrons are also moderated below threshold for detection and/or absorbed. In the latter case, approximately 3 induced fission neutrons would be observed per neutron source pulse (assuming 10% detector efficiency and $10^9$ source neutrons/pulse) [Johnson, 2006]. An event rate this low would require extremely precise neutron discrimination capability to identify SNM – something that is not implausible but had yet to be tested.

Further work at modeling active neutron interrogation systems has been performed [Kerr, 2005] which only serves to reinforce the decision to use low energy source neutrons. Table 2 below summarizes the results of performing similar cargo environment simulations utilizing multiple neutron sources, such as the 2.5 MeV deuterium-deuterium reaction as well as the 14.1 MeV deuterium-tritium reaction. The cargo container modeled in this simulation is filled uniformly with a density of 0.4g/cm³ for the various materials listed. Note that in every shielding case studied, with the exception of hydrogenous material (water), the 60 keV neutron source has a higher fission probability rate per source neutron.
Table 2 – Fission Probability per Source Neutron [Kerr, 2005]

<table>
<thead>
<tr>
<th>Source</th>
<th>Cargo</th>
<th>Water</th>
<th>Aluminum (Al)</th>
<th>Iron (Fe)</th>
<th>Lead (Pb)</th>
</tr>
</thead>
<tbody>
<tr>
<td>60 keV</td>
<td>Water</td>
<td>3.92e-8 ± 0.15</td>
<td>4.69e-4 ± 0.008</td>
<td>5.05e-4 ± 0.007</td>
<td>5.75e-4 ± 0.007</td>
</tr>
<tr>
<td>(directional)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2.5 MeV</td>
<td>Water</td>
<td>1.03e-6 ± 0.043</td>
<td>1.26e-4 ± 0.012</td>
<td>9.09e-5 ± 0.15</td>
<td>9.31e-5 ± 0.019</td>
</tr>
<tr>
<td>(isotropic)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>14.1 MeV</td>
<td>Water</td>
<td>3.37e-5 ± 0.007</td>
<td>1.18e-4 ± 0.013</td>
<td>1.22e-4 ± 0.15</td>
<td>1.48e-4 ± 0.018</td>
</tr>
<tr>
<td>(isotropic)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Nevertheless, it can be stated with confidence that the proposed low energy neutron interrogation system should, in theory, be very effective at identifying SNM in cargo environments. Clearly, this demonstrates justification for further research and development into this type of SNM detection system.
3.0 Lithium Target Design

Given the neutron interrogation system criteria explained in the previous Chapter, a proton-lithium neutron generator was commissioned for this project. This consists of a particle accelerator and a lithium target which together utilize the $^7\text{Li}(p,n)^7\text{Be}$ reaction to produce sub-MeV energy neutrons. The goal in designing the lithium target is to maximize neutron production rates while maintaining desired neutron energy levels and neutron emission geometry. The following sections provide the general design characteristics and estimated neutron yields for the lithium target.

3.1 Lithium Target Description

The lithium target consists of electrochemical grade lithium foil (99.9% Li) secured to a basic KF-40 flange with conductive carbon tape. The lithium foil is 107 μm thick and covers the entire exposed inner surface of the flange (18mm radius) as pictured in Figure 19 below. The isotopic abundance of lithium-7 is greater than 93% in the foils, with less than 7% lithium-6 and negligible impurities.

![Figure 19 – Lithium Target Assembly](image)
Due to the highly corrosive nature of pure lithium metal, all assembly had to be
done in an argon atmosphere to prevent the rapid oxidation of lithium in the presence of
oxygen. A layer of oxidation covering the lithium target would degrade performance of
the neutron generator system by causing protons to interact with non-lithium nuclei,
resulting in a net loss of neutron production. Other lithium compounds such as LiF and
Li$_2$O are commercially available and are essentially non-reactive in normal atmospheric
conditions. However, the presence of non-lithium nuclei in the target material severely
affects total neutron production rates (neutrons/milliCoulomb) as shown in Table 3 [Lee,
1998].

<table>
<thead>
<tr>
<th>Incident Proton Energy (MeV)</th>
<th>Neutron Yields (neutrons/mC)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Li</td>
</tr>
<tr>
<td>1.89</td>
<td>6.34E+09</td>
</tr>
<tr>
<td>1.90</td>
<td>1.49E+10</td>
</tr>
<tr>
<td>1.91</td>
<td>2.41E+10</td>
</tr>
<tr>
<td>1.92</td>
<td>3.35E+10</td>
</tr>
<tr>
<td>1.93</td>
<td>4.30E+10</td>
</tr>
<tr>
<td>1.94</td>
<td>5.25E+10</td>
</tr>
<tr>
<td>1.95</td>
<td>6.21E+10</td>
</tr>
<tr>
<td>1.96</td>
<td>7.16E+10</td>
</tr>
<tr>
<td>1.97</td>
<td>8.12E+10</td>
</tr>
<tr>
<td>1.98</td>
<td>9.08E+10</td>
</tr>
<tr>
<td>1.99</td>
<td>1.00E+11</td>
</tr>
<tr>
<td>2.00</td>
<td>1.10E+11</td>
</tr>
</tbody>
</table>

Throughout the range of proton energies, the total neutron yield for pure lithium is
approximately 3 times that for LiF and double the production rate for Li$_2$O. In addition
to the enhanced neutron production rate, lithium metal has a much higher thermal
conductivity than either of the two compounds. This will also increase target longevity.

On the non-vacuum face of the flange a heat exchanger made from 5 mm copper
tubing was soldered to the base. Circulating water through this heat exchanger enables
cooler operation at high beam current while maintaining proper vacuum conditions on the accelerator. Heat generation rates as high as 40W can easily be achieved, and without a means to remove this heat the metal vacuum fittings may expand, resulting in loss of vacuum.

3.2 Thick Target Neutron Yield

The interdependency of proton energy with desired neutron energy, production rate and angle of emission makes optimization of a low energy neutron generator system non-trivial. The optimal configuration is unique to each interrogation scenario; optimal neutron energy varies depending on the geometry and material composition of the interrogation object. A thorough summary of thick lithium target neutron yields versus incident proton energy is given in Table 4 [Lee, 1998].

As determined by the criteria presented in Chapter 2 of this thesis, the optimal source neutron energy lies in the range of 60 – 100 keV. Therefore, the potential range of incident proton energies goes from just above threshold, 1.89 MeV to approximately 2.10 MeV. At lower incident proton energies, total neutron yield is decreased due to the fact that the total integrated (p,n) cross section is lower. Figure 20 illustrates the one-dimensional energy profile of protons passing through lithium metal foil [Blackburn, 2006].
Table 4 – Thick Target Neutron Yields Versus Incident Proton Energy [Lee, 1998]

<table>
<thead>
<tr>
<th>Incident Proton Energy (MeV)</th>
<th>Total Neutron Yield (n/sec/mA)</th>
<th>Maximum Neutron Energy (keV)</th>
<th>Mean Neutron Energy (keV)</th>
<th>Maximum Neutron Angle (degrees)</th>
<th>Mean Neutron Angle (degrees)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.89</td>
<td>6.34 x 10^9</td>
<td>67.1</td>
<td>34.0</td>
<td>30.0</td>
<td>16.5</td>
</tr>
<tr>
<td>1.90</td>
<td>1.49 x 10^10</td>
<td>87.6</td>
<td>38.3</td>
<td>45.2</td>
<td>23.0</td>
</tr>
<tr>
<td>1.91</td>
<td>2.41 x 10^10</td>
<td>105.3</td>
<td>42.4</td>
<td>60.3</td>
<td>27.8</td>
</tr>
<tr>
<td>1.92</td>
<td>3.35 x 10^10</td>
<td>121.4</td>
<td>46.5</td>
<td>180</td>
<td>31.9</td>
</tr>
<tr>
<td>1.93</td>
<td>4.30 x 10^10</td>
<td>136.6</td>
<td>50.6</td>
<td>180</td>
<td>35.3</td>
</tr>
<tr>
<td>1.94</td>
<td>5.25 x 10^10</td>
<td>151.1</td>
<td>54.4</td>
<td>180</td>
<td>38.3</td>
</tr>
<tr>
<td>1.95</td>
<td>6.21 x 10^10</td>
<td>165.1</td>
<td>58.1</td>
<td>180</td>
<td>41.0</td>
</tr>
<tr>
<td>1.96</td>
<td>7.16 x 10^10</td>
<td>178.8</td>
<td>61.6</td>
<td>180</td>
<td>43.5</td>
</tr>
<tr>
<td>1.97</td>
<td>8.12 x 10^10</td>
<td>192.1</td>
<td>65.0</td>
<td>180</td>
<td>45.6</td>
</tr>
<tr>
<td>1.98</td>
<td>9.08 x 10^10</td>
<td>205.1</td>
<td>68.4</td>
<td>180</td>
<td>47.6</td>
</tr>
<tr>
<td>1.99</td>
<td>1.00 x 10^11</td>
<td>218.0</td>
<td>71.7</td>
<td>180</td>
<td>49.4</td>
</tr>
<tr>
<td>2.00</td>
<td>1.10 x 10^11</td>
<td>230.6</td>
<td>75.1</td>
<td>180</td>
<td>51.1</td>
</tr>
<tr>
<td>2.10</td>
<td>2.13 x 10^11</td>
<td>350.4</td>
<td>108.4</td>
<td>180</td>
<td>63.0</td>
</tr>
<tr>
<td>2.20</td>
<td>3.62 x 10^11</td>
<td>463.4</td>
<td>158.9</td>
<td>180</td>
<td>68.7</td>
</tr>
<tr>
<td>2.30</td>
<td>5.78 x 10^11</td>
<td>573.1</td>
<td>233.1</td>
<td>180</td>
<td>66.3</td>
</tr>
<tr>
<td>2.40</td>
<td>7.48 x 10^11</td>
<td>680.6</td>
<td>286.5</td>
<td>180</td>
<td>63.8</td>
</tr>
<tr>
<td>2.50</td>
<td>8.83 x 10^11</td>
<td>786.7</td>
<td>326.4</td>
<td>180</td>
<td>62.9</td>
</tr>
</tbody>
</table>

Proton Energy Vs. Depth in Lithium

Figure 20 – Proton Energy Vs. Depth in Lithium [Blackburn, 2006]
Utilizing tabulated stopping power data for protons in lithium, the energy profile versus penetration depth can be constructed. Note that 2.0 MeV protons traverse roughly 15 microns before falling below the \((p,n)\) threshold of 1.89 MeV, while 1.9 MeV protons travel a mere 1 micron before doing the same. Knowing the energy profile as a function of distance, the total integrated cross section can be obtained utilizing tabulated cross section data based on energy. Figure 21 shows the \((p,n)\) cross section versus distance into the lithium target [Blackburn, 2006].

![Cross Section vs. Depth in Lithium](image)

**Figure 21 – Cross Section Vs. Depth in Lithium [Blackburn, 2006]**

As expected, the higher energy protons have a much higher integrated cross section (4058 mb·μm vs. 419 mb·μm), nearly 10 times that of protons only 100 keV less energetic. Therefore, for any given proton beam current, higher energy protons will produce higher source neutron fluxes. However, increased proton beam current may be
able to compensate for the decreased production rate if lower energy neutrons are desired. An additional note of concern may be the average emission angle of neutrons. As shown in Table 4, the average emission angle for neutrons increases with higher proton energies. If a more focused beam is desired, then lower energy protons must be used for increased kinematic collimation; however, optimal interrogation beam geometry will vary.

For this proof of concept, it was decided to generate 75 keV neutrons (mean energy) using 2.0 MeV protons on a thick lithium metal target. Due to gamma-ray interference (explained further in Chapter 5), the tandem accelerator was run at a very low 0.04 μA average current. This results in an expected neutron yield of approximately 4.4x10^6 neutrons/second. To put this into perspective, we may simplify the neutron emission geometry and assume neutrons are emitted isotropically into 4π, and a 25 cm^2 target at a distance of 40 cm from the lithium target can expect to see approximately 5500 neutrons/second, or 219 neutrons/second/cm^2.
4.0 Neutron Detection System

The keystone of this project is undoubtedly the neutron detection system. Without an effective way of discriminating low energy source neutrons, high energy neutrons from fission and gamma rays, the concept of active neutron interrogation would be extremely difficult to implement. Each component of the neutron detection system was carefully selected to maximize the effectiveness of the entire neutron detection system. The main components that comprise the neutron detection system are as follows: Liquid organic scintillator detectors, digital storage oscilloscope, Van de Graaff particle accelerator and most importantly, the digital pulse shape discrimination (PSD) software.

The following sections describe each component in detail and explain the methods of implementation.

4.1 Description of Hardware

In regards to active neutron interrogation systems, a major reason for utilizing low energy source neutrons as opposed to high energy source neutrons (MeV range or above) is that it is possible to design a neutron detection system which is non-responsive to the low energy source neutrons, but highly sensitive to higher energy neutrons from fission events. Furthermore, it is necessary to discriminate high energy neutron events from gamma-ray events due to the inherently high level of gamma radiation present in high neutron radiation fields [Kerr, 2005].

A common approach to this problem is to utilize liquid organic scintillators which allow for excellent neutron/gamma-ray pulse shape discrimination. The light output caused by neutrons elastically scattering off hydrogen nuclei in the organic scintillator
bulk material differs slightly from the light output caused by Compton electrons from gamma-ray interactions in the scintillator fluid. Through advanced digital pulse shape discrimination techniques, this difference in light output can be exploited for purposes required in active neutron interrogation.

**Liquid Organic Scintillator Detectors**

Two liquid organic scintillators from Scionix (model # V250A82/3M-LS-X2-NEG) have been tested for use in this project. The detectors contain LS 309 scintillation fluid and are coupled to Photonis XP4312B photomultiplier readouts. The optimal operating voltage for both detectors was determined to be -1500V, based on count-rate optimization utilizing a calibrated 10mCi AmBe source. Figure 22 below shows one of the two identical neutron detectors provided by Scionix.

![Figure 22 – Scionix Liquid Organic Scintillator Detector](image)

The selection of detector size in this project involves a compromise between neutron detection efficiency, energy resolution and gamma-ray event pileup. Obviously,
a detector having a larger cross section perpendicular to incoming radiation will comprise a larger solid angle and thus have a higher chance of counting a radiation event. Also, the effect of detector thickness on overall efficiency is greatly pronounced with regards to neutron detection. As neutrons propagate through the scintillation fluid, the chance of interaction and detection is directly proportional to the pathlength (d), as indicated by the equation for intrinsic efficiency [Knoll, 2000] below,

\[ \varepsilon = 1 - \exp(-N\sigma_s d), \]  

where \( N \) is the number density of target nuclei and \( \sigma_s \) is the scattering cross section for these nuclei. Due to the energy dependent nature of scattering cross sections, the efficiency is indirectly proportional to the square root of the neutron energy \( (E_n) \) [Knoll, 2000].

\[ \sigma_s(E_n) = \frac{4.83}{\sqrt{E_n}} - 0.578 \text{ barns} \]  

For 1 MeV neutrons, the intrinsic detection efficiency for liquid organic scintillators is 78% [Kerr, 2005]. However, the intrinsic efficiency assumes only neutron-proton interactions. Neutrons interacting with carbon, oxygen or any other nuclei present in the scintillation material will inevitably detract from overall detection efficiency. Therefore, the only accurate way to determine the detector's neutron detection efficiency is to use a calibrated neutron source. Utilizing a 10mCi Am(α,n)Be source, our neutron detection efficiency was determined to be approximately 1.42%. It should be noted that the average neutron energy emitted from an AmBe source is 4.5 MeV. Considering the average fission neutron energy is approximately 2 MeV, Equation 4.2 above predicts that
the neutron detection efficiency for neutrons from fission events to be approximately 2.13%.

In order to maximize neutron count rates, it was decided to use fairly large liquid scintillator detectors (12” x 12”). Although gamma-ray count rates will exceed fission neutron count rates, it is assumed that through the use of PSD software these events can be excluded. Although gamma-ray events can indeed be isolated and ultimately rejected, gamma-ray event pileup in the detector was discovered to be a major limiting factor in the overall system efficiency. This is discussed further in Chapter 6.

**Tandem Particle Accelerator**

For the proof-of-concept phase of this project, a tandem particle accelerator manufactured by Newton Scientific Instruments serves as the source of high energy protons. This accelerator is normally run with continuous extraction current and has been shown capable of achieving 100 µA beam currents for sustained periods of time. For the purposes outlined in Chapter 3, the terminal voltage was set at 1.0 MV, resulting in proton energies of 2.0 MeV at the lithium target. The ion source was supplied by high purity hydrogen gas at approximately 4 sccm. A picture of the accelerator is shown in Figure 23.
Completing the neutron/gamma-ray discrimination system is the digital oscilloscope provided by Ztec Incorporated. After testing the 8-bit, 2.5 GSamples/second digital storage oscilloscope, it was decided to purchase a 16-bit, 400 MSamples/second digital storage oscilloscope (model #ZT410PCI-51) for enhanced waveform resolution. This digitizer is capable of running at 400 MSamples/second with one channel of input or simultaneously running two channels at 200 MSamples/second.

For digital waveform acquisition, each waveform is digitally reconstructed using a fixed number of points. Higher resolution digitizers more accurately place the point at the correct voltage, while digitizers with higher sampling rates simply reproduce the waveform using more sample points with less accuracy. At 400 MSamples/second, each point represents 2.5 nanoseconds in the waveform. Typical waveforms for this application are on the order of 300 nanoseconds, meaning each waveform is digitally constructed from approximately 120 points. Obviously, larger amounts of data points
reconstructing the waveform is desirable; however, neutron/gamma-ray discrimination has been achieved with sampling rates as low as 150 MSamples/second at 12-bit resolution [Kaschuck, 2004]. As long as approximately 120-200 MSamples/second sampling rates can be achieved, the most important factor in selecting a digital storage oscilloscope is the resolution – the higher, the better [Lanza, 2006]. As the current state of technology for digital storage oscilloscopes progresses, this will no longer be an issue as 16-bit, 400 MSamples/second digitizers are not considered state-of-the-art at this point.

This unit was installed on a desktop PC, running Windows XP at 3.2 GHz and 1 GB of RAM. Using the drivers provided by Ztec Inc., the digital oscilloscope is configured and controlled by a LabVIEW 7.1 program written for the purpose of neutron/gamma-ray discrimination.

4.2 Digital Pulse Shape Discrimination

The slight difference in neutron and gamma-ray pulse shapes acquired from a digitizer coupled to a liquid organic scintillator detector can be seen in Figure 24 below. As depicted, the leading edge of the neutron and gamma pulses are virtually indistinguishable and pulse separation becomes apparent at approximately 50 nanoseconds after the leading edge of the waveform. The key point to this “charge comparison method” of pulse shape discrimination is to integrate the waveform over certain key regions of the pulse. These key regions have been designated $\Delta T_f$ and $\Delta T_s$, pertaining to the fast and slow components of scintillation light decay (trailing edge of pulse after peak) as depicted.
Figure 24 - Neutron and Gamma-ray Pulse Shapes in Liquid Organic Scintillators

Figure 24 represents actual waveforms acquired from a deuterium-tritium (DT) neutron generator whose pulse-shapes have been averaged. With the exception of a slightly higher voltage offset at the beginning of the waveform, no obvious differences between neutron and gamma-ray pulses is apparent from the leading edge of the pulse through $\Delta T_f$. However, the difference in pulse light decay times from recoil protons from neutron events and Compton electrons from gamma rays is extremely evident through $\Delta T_s$. Integrating each waveform over the two regions and plotting the integrated charge of $\Delta T_s$ versus the integrated charge of $\Delta T_f$ will further enhance the difference in radiation events, as shown in Figure 25.
In this figure, the red colored events represent neutron interactions and the black colored events are from gamma-ray interactions. A very clear separation of neutron and gamma-ray events is produce by this digital pulse shape discrimination method. The acquisition and storage of entire waveforms also allows for enhanced post-processing of data for use in determining figure of merit and radiation field characterization. While real-time pulse shape discrimination is easily achieved, further attempts to characterize incoming waveforms may contribute to computer dead time and result in the unintended rejection of data.
In order to systematically count the apparent number of neutron/gamma-ray interactions, it is necessary to transform the above two-dimensional plot into a one-dimensional histogram by projecting the discriminated events onto one axis. First, the entire plot is rotated clockwise by an angle $\theta$, so that the "line of discrimination" which separates neutron from gamma-ray events is parallel to an arbitrary axis (we can use the x-axis for reference). Then, the distance of each point from the reference axis (y-axis value in this reference case) is binned and plotted in a histogram displaying the relative number of events for both neutrons and gammas. In this case, x-values are represented as the integration value for $\Delta T_I$, or short integration value, and y-values are represented by the integration value for $\Delta T_s$, or long integration value. Therefore, the reference axis can be represented by the Equation 4.3:

$$\cos \theta \int_{\text{long}} V_{dt} - \sin \theta \int_{\text{short}} V_{dt}$$  \hspace{1cm} 4.3

Here, $V_{dt}$ is simply the section of acquired waveform that is to be integrated. Figure 26 represents the values obtained from Figure 25.
Figure 26 – Histogram of Counts from DT Neutron Generator

As shown in the figure, the neutron to gamma ratio is approximately 0.84, with 7,664 neutrons and 9,136 gammas. Although the neutron/gamma ratio is useful in determining induced neutron radiation fields, it alone can not give an accurate measure of the quality of discrimination achieved. For this purpose, the Figure of Merit (FOM) is defined by Equation 4.3, where $S$ is the separation between the neutron and gamma-ray peaks, and $\sigma_N$ and $\sigma_\gamma$ are directly related to the neutron and gamma-ray peak full width at half-maximum (FWHM), respectively. $\sigma_N$ and $\sigma_\gamma$ are the respective square-roots of the variance ($\sigma^2$) for either peak, and are related to the FWHM as defined in Equation 4.4.
Figure of merit values for successful PSD are generally greater than 1 [Sellin, 2003], with typical values ranging from 1.3 to 2.6. FOM values are highly dependent on signal shape, resolution and integrity. As discussed in Chapter 4.1, very high gamma radiation fields lead to pileup rejection and waveform distortion. This effect distorts the time and voltage characteristics of the neutron and gamma-ray waveforms acquired, leading to degraded discrimination performance. For the DT neutron generator data pictured above, the FOM was found to be 2.17.

Additionally, it is possible to simply integrate over the entire waveform to establish a measure of the total energy deposited by the incoming radiation event and then plot the values in a histogram. This is similar to obtaining a pulse-height spectrum in gamma-ray spectroscopy. With gamma rays, the total charge deposition spectra will exhibit peaked features corresponding to the energy distribution of the gamma field as seen with gamma-ray pulse height distribution graphs. However, the spectra for neutrons will be rectangular, ranging from a fixed minimal value to a maximum value relative to the maximum energy of the neutrons. This is due to the fact that neutrons, transferring their kinetic energy to protons in the scintillation fluid through elastic scattering, depend on the scattering angle (0 to $\pi$) to determine the amount of energy transferred (0 to total KE) [Knoll, 2000]. Therefore, it is not possible to determine the kinetic energy of a single neutron interaction; however, it is possible to correlate the maximum charge deposition value with a maximum possible neutron kinetic energy level to help
characterize the neutron radiation field. Figure 27 illustrates the total charge deposition spectra for the DT neutron generator source.

![Total Charge Deposited](chart)

*Figure 27 – Total Charge Deposition Spectra from DT Source*

This method of analysis was performed on multiple neutron sources in order to verify the validity and accuracy of the experimental setup and analytical procedure. Figures 28 - 30 represent data obtained using a 10 mCi AmBe neutron source. The average neutron energy emitted from this source is 4.5 MeV. Figures 31 - 33 represent data obtained from a deuterium-deuterium (DD) neutron generator source, with average neutron energies of approximately 2 MeV.
As shown in Figure 28, the AmBe neutron source used resulted in a very clean neutron/gamma-ray discrimination plot. A FOM of 2.55 was obtained for this data, largely due to the relatively low gamma-ray emissions rate of the AmBe source. Although the neutron/gamma ratio was low (0.25), the rate at which gamma rays interacted with the detector was much lower than those encountered with accelerator-based neutron generators; the absence of false neutron events caused by gamma pileup in the area above the neutron event region of interest is a large indication of the purity and accuracy of results.
The two distinct peaks are clearly visible in this figure, with minimal overlapping of the peak distributions around approximately $0.8 \times 10^{-9}$. A perfectly discriminated plot would show both the trailing edge of the gamma peak and leading edge of the neutron peak reaching zero counts. The lesser the degree of overlap present in this figure, the higher the confidence in neutron/gamma count rates.
Figure 30 shows the energy deposition spectra for the 4.5 MeV AmBe neutron source. When compared to the 14 MeV DT neutron generator source, the average energy deposited has shifted to the left of the graph, indicated a clear response to the energy of incident neutrons. Although the distribution now seems slightly peaked, this is most likely just a result of the reduced 4.5 MeV charge deposition range coupled with limited statistics. Increased waveform acquisition counts are needed to determine a truer representation of the charge deposition spectrum.
Figure 31 - Neutron/Gamma-ray Discrimination from DD Neutron Generator

Figure 31 depicts the neutron/gamma-ray discrimination obtained from a 2 MeV DD neutron generator source. Due to a relatively large gamma field, the separation is nowhere near as clean as that obtained with the AmBe neutron source. However, a FOM of 2.20 was achieved, with neutron/gamma ratio equaling 0.38. Inspection of Figure 32 helps to unravel the discrepancies between a higher FOM and seemingly lower quality discrimination.
Figure 32 – Histogram of Counts from DD Neutron Generator

Compared with Figure 26 (DT neutron generator source), the histogram of counts from the DD neutron generator source reveals that, although the peak separation is diminished ($4.96 \times 10^{-10}$ Vs. $6.74 \times 10^{-10}$), the respective $\sigma_N$ and $\sigma_\gamma$ values (sum) are also smaller ($2.25 \times 10^{-10}$ Vs. $3.11 \times 10^{-10}$). The overall effect increases the FOM to an artificially high level. This can be explained by the relatively low FWHM for the neutron peak in Figure 32. Comparison of Figure 31 with Figure 25 reveals a much higher neutron/gamma ratio for the DT neutron generator source and also wider neutron peak distribution, resulting in a smaller FOM.
Figure 33 - Total Charge Deposition Spectra from DD Source

Figure 33 clearly indicates a much lower range of charge deposition from the 2 MeV neutron events when compared with the 14 MeV neutron spectra shown in Figure 27 and the 4.5 MeV neutron spectra shown in Figure 30. Moreover, this particular charge deposition spectrum is extremely valuable, in that the 2 MeV DD neutrons are roughly equivalent to fission energy neutrons which will appear in active neutron interrogation and may thus serve as verification of induced fission events. The long trailing edge of the neutron events distribution can be attributed to the inevitable distorted waveforms acquired due to gamma pileup. These waveforms are usually double-peaked or largely
saturated signals which have very high integrated values as seen by the multitude of “stray neutron events” present above the neutron event region of interest in Figure 31.

Although both the DT and DD neutron generator sources are accelerator based, the DT neutron generator source is an RFQ accelerator and the DD neutron generator source utilized the tandem linear accelerator. As evidenced in the above figures, the tandem linear accelerator based neutron generator source simply emits a much higher gamma radiation field per source neutron (neutron/gamma ratios of 0.84 for DT versus 0.38 for DD), which in turn leads to somewhat diminished neutron/gamma-ray discrimination quality.

Nevertheless, the data analysis performed herein clearly demonstrates a successfully designed neutron/gamma-ray discrimination system capable of resolving fission energy neutrons.
5.0 Fissile Material Characterization

With a fully functioning neutron/gamma-ray discrimination system, the low-energy neutron interrogation system proof-of-concept was tested with 42 grams of highly enriched (93w/o U235) uranium. Clearly, 39 grams of uranium-235 is not a significant amount in terms of the amount needed to construct a nuclear weapon (kg quantities); however, it is felt that this small amount would represent a lower bound in the detection abilities of the proposed system. Additionally, multiple shipments of smaller amounts of SNM may be smuggled into the United States separately and the weapon constructed domestically, thus validating a need to explore the limits of sensitivity for such active neutron interrogation systems.

Although not fully disclosed, the dimensions of the thin sheet of SNM were approximately 3 cm wide by 12 cm length, with a depth of approximately 0.25 cm. This SNM foil was housed inside a cylindrical plastic container approximately 8 cm in diameter and 23 cm in length. The SNM was placed approximately 40 cm from the lithium neutron generator source as illustrated in Figure 34 below (NOT TO SCALE). With the extremely low beam current producing only 219 neutrons/cm²/sec at the SNM target (this beam current was determined to be the maximum allowable before complete failure of discrimination ability), the maximum fission rate that can be expected assuming ideal geometry and 2 neutrons per fission is 320 fission neutrons per second, emitted into 4π. The fission neutron emission rate (R) for 60-100 keV source neutrons was calculated using Equation 5.1, where Σ is equal to 0.81cm⁻¹, φ is 219 n/cm²·sec and the volume (V) is equal to 3 cm x 12 cm x 0.25 cm.

\[ R = 2\phi \Sigma V \]  

5.1
As shown in Figure 34, the detector was placed approximately 87 cm from the SNM, partially shielded from the lithium target by a double-density concrete wall, but unshielded from fission neutrons emitted from the SNM target. With the fission rate calculated above, the maximum fission neutron interaction rate at the detector is approximately 3.13 neutrons/sec. With roughly 2% detection efficiency for 2 MeV neutrons, this system in its current state is not suitable for rapid cargo inspection applications. The total expected discriminated neutron events for a 25-minute interrogation system is approximately 94. For this reason, additional HDPE was added surrounding the rear and right side of the SNM container for the purpose of
reflecting/moderating source neutrons and effectively increasing the neutron source flux without equally increasing the gamma radiation field.

5.1 Bare Fissile Material Induced Signal

Four 25-minute interrogation sessions were performed to evaluate the proposed system’s effectiveness in characterizing the induced signal on unshielded SNM. Two sessions included SNM as depicted in Figure 34, and two sessions were conducted without SNM for the purpose of determining the basal radiation signal from the lithium neutron generator system. Figure 35 presents the data collected with no SNM in the interrogation beam.

As shown in Figure 35, very few (101 in total) neutron events were recorded during both 25-minute intervals run without SNM (both sessions produced virtually identical data). These neutron events can be attributed to the low energy source neutrons being scattered into the detector and recorded; however as explained previously, the neutron detection efficiency for neutrons with less than several hundred keV is very small compared to the already minimal 2% efficiency for 2 MeV neutrons. Most likely, the events are a result of gamma pileup caused by neutron absorption in equipment and shielding, as well as background from gamma radiation produced intrinsically by the accelerator itself (proton interactions with hardware as well as bremsstrahlung from the ion source). Figure 36 below presents the data obtained from the interrogation of unshielded SNM.
A very slight, yet noticeable increase is apparent when comparing Figure 36 with Figure 35. Total neutron events recorded increased from the background of 101 (both background sessions produced 101 neutron events) to 177 and 185 neutron events for both unshielded SNM runs. This average increase in fission neutron events of 80 events is slightly short of the predicted 94 events, but still within range when considering absolute counting uncertainty; however, this may simply be an artifact of SNM geometry assumptions.
Unfortunately, the severely limited number of neutron events prevented the actual determination of a FOM due to the fact that not enough points were available for the formation of a Gaussian distribution of neutron events. The data are completely dominated by gammas and the software used to analyze datasets was unable to resolve any neutron peaks.

![Figure 36 - Neutron/Gamma-ray Discrimination with Unshielded SNM](image)

**5.2 Effect of Shielding on Induced Signal**

The same procedure described above was repeated for two different shielding configurations: High density polyethylene (HDPE) and Borated polyethylene (Ricorad).
In both cases, very large sheets (15 inches x 24 inches) of shielding were placed between the lithium neutron generator and the SNM target. For the HDPE sessions, one 1 inch thick sheet was used, and two 1 inch thick sheets were used for the borated polyethylene sessions. Figures 37 and 38 below present the neutron/gamma-ray discrimination results obtained for the HDPE and Ricorad shielded cases, respectively.

Figure 37 – Neutron/Gamma-ray Discrimination with 1” HDPE Shielded SNM

As predicted in Chapter 2, a minimal amount of hydrogenous shielding would serve to moderate the source neutrons, effectively increasing the fission cross sections and result in increased SNM selectivity. The data presented in the above figure supports
that conclusion, as the neutron events increased from a background of 94 counts without SNM to a total of 267 neutron events with SNM. This represents an increase of 184% over background.

A common, neutron-absorbing material used for shielding is borated polyethylene. The boron content acts as a black absorber for low energy neutrons, while the high hydrogen content thermalizes higher energy neutrons. Although registered neutron events increased from a background of 93 to 139 events with SNM, the incremental increase of 49% was the smallest of all scenarios.
6.0 Future Work and Conclusions

6.1 Conclusions

A summary of the low energy neutron interrogation experiment is provided in Figure 39 below. As counts, the associated uncertainties are expressed as simply the square root of the value, with the error bars indicated at the top of the respective column.

![Figure 39 - Low Energy Neutron Interrogation Results Summary](image)

Even with extremely limited neutron source conditions, Figure 39 clearly indicates that the proof-of-concept design presented herein is fully functional and responds accordingly to various changes in shielding configurations and presence of SNM. Considering that only 39 grams of uranium-235 was interrogated at extremely low beam currents, these results are a testament to the accuracy and robustness of the neutron/gamma-ray discrimination system designed.
As for the blind SNM tests to be conducted, this was inadvertently performed as the last interrogation session performed was not documented properly in a lab notebook. Upon review of the data, there was no question that this session was indeed the second No Shielding with SNM run. This was confirmed upon inspection of the sessions originally planned, yet not assigned a dataset. Unfortunately, due to a number of administrative reasons, it was not possible to interrogate different samples of fissile material such as depleted uranium or plutonium.

Unfortunately, the source neutron generation limitations described previously severely limit the ability to accurately resolve the induced neutron energy spectrum. With event counts in the range of a couple hundred, the charge deposition spectrums are not resolvable. Figure 40 presents a histogram of the waveforms acquired for the respective interrogation sessions.

![Figure 40 - Complete Waveform Integration Values for Interrogation Sessions](image)
The far right region of the above graph shows that binned values for sessions including SNM begin to diverge from averaged background values. Plotting the normalized difference of the SNM session from background, a general neutron peak can be unfolded. This data is plotted in Figure 41. Most importantly, it is shown that the histogram values obtained for the neutron region circled in red correspond to values expected for fission energy neutrons as given by Figure 32, supporting the claim that neutrons from fission events are being resolved only in the active interrogation sessions with SNM.

Figure 41 – Neutron Event Differences from Background for Interrogation Sessions
6.2 Future Work

Although this proof-of-concept has demonstrated successful functionality, there lies much room for improvement and refining of the experimental apparatus. Obviously, the major limiting factor to this system’s overall efficiency is gamma pileup caused by excessive gamma radiation produced by both the accelerator and lithium neutron generator.

As far as the accelerator is concerned, not much can be changed in the way of hardware configuration due to the prototypical design of the linac unit. Originally, as the second phase of this project, it was planned to design and construct a prototype RFQ-based accelerator much like the DT neutron generator used which produced fantastic results. Construction of this RFQ based system using materials such as tungsten may help to reduce the induced gamma field.

The same can be recommended for a future lithium neutron generator design. The steel flange used in this experiment can be substituted with a design constructed from tungsten or aluminum – materials which have lower (n,g) cross sections.

Additionally, the size of detector chosen for this project may be too large and therefore more susceptible to gamma pileup [Knoll, 2000]. Using multiple smaller liquid organic scintillators could possibly regain any loss in overall neutron detection efficiencies experienced by reducing the available total solid angle for neutron interaction.

Furthermore, given that 100 keV neutrons move at a speed of $4.4 \times 10^6$ m/second whereas gamma radiation have a speed of $3 \times 10^8$ m/second, it would be extremely beneficial to use the timing of the accelerator pulse and placement of the detector(s) to
effectively pre-discriminate the acquired waveforms. For instance, if the detector is placed 1 meter from the SNM, which itself is placed 1 meter from the lithium target, approximately 280 nanoseconds would elapse before one could expect induced fission neutrons to arrive at the detector. Furthermore, this assumes that all induced fission neutrons are produced instantaneously and a source neutron energy of 100 keV. In reality, thermalization time, delayed neutron events and lower source neutron energies would augment the time delay between the arrival of gammas and induced fission neutrons at the detector. Each of the above recommendations would serve to limit the effects caused by gamma pileup and seen manifested in this proof-of-concept. The combined effect of all these changes should not only increase the accuracy and quality of discrimination, but it would also allow for higher beam current to be run which would produce more source neutrons, which in turn induce more fissions and substantially reduce the required interrogation times.

Additional suggestions for the continuation of this project include varying the proton energies/neutron source energies. As explained in Chapters 2 and 3, the neutron energy is not only directly related to the penetration ability and probability of inducing fission, but also the reaction kinematic geometry. Obviously, there are an infinite amount of shielding scenarios to simulate; however, if this type of system is ever to be implemented, careful consideration should be taken into what data is published given the nature of this research. Lastly, a sensitivity analysis to determine the effects of varied amounts of fissile material should be performed with a highly refined low energy neutron interrogation system.
References


