Lithium-Glass Neutron Detection

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A senior thesis submitted to the faculty of
Brigham Young University
in partial fulfillment of the requirements for the degree of

Bachelor of Science

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April 2011

ABSTRACT

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We have designed a neutron detector using ^{6}Li glass scintillator. ^{6}Li has a large neutron capture cross section, which gives high neutron detection rates. ^{6}Li captures the neutrons and the resulting ^{7}Li rapidly decays into charged particles which are subsequently detected. We have measured neutron detection efficiency of 30 percent with gamma contamination of only 1 part in 10000.

Keywords: Neutron, Detection, Radiation

ACKNOWLEDGMENTS

Thanks to Dr. Lawrence Rees and Dr. J. Bart Czirr.

Parts of this work were funded by NNSA Grant no. DE-FG52-10NA29655 and DHS Award no. 2010-DN-077-ARI039-02.

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Chapter 1

Introduction

1.1 Motivation

Every day thousands of cargo containers are scanned by nuclear particle detectors. The U.S. Department of Homeland Security (DHS) is seeking to prevent proliferation of nuclear materials. For years DHS has been using ^{3}He neutron detectors at ports of entry. However, ^{3}He is becoming increasingly difficult to obtain [1]. We have received funding from DHS and the National Science Foundation to create a replacement detector for ^{3}He. As part of this research, we have designed a lithium glass neutron detector that could replace ^{3}He detectors.

1.2 Background

1.2.1 Neutron Detection

Because neutrons are one of the fundamental building blocks of matter, their detection is important to several branches of physics. Every nucleus has a neutron interaction cross-section, which is related to its probability of interacting with a neutron. Nuclei with large cross-sections have high probabilities of neutron interaction. Neutron interaction cross-section is a function of neutron energy [2]. The most important neutron interactions that occur in our detector include neutron capture, elastic collisions, and inelastic collisions.

When a neutron collides with a nucleus, the neutron loses energy. If the neutron energy is above the excitation energy of the nucleus, then the neutron has a probability of colliding inelastically; the neutron excites the nucleus. If the neutron energy is below this excitation energy, the neutron can only capture or collide elastically. For elastic collisions we only need to understand the basics of neutron-hydrogen and neutron-carbon collisions. When a neutron collides with a hydrogen nucleus, the neutron's direction can change up to a 90 degree angle and the neutron can transfer up to 100 percent of its energy to the proton. On average a neutron will lose about half its energy in an elastic collision with hydrogen. Carbon is 12 times heavier than hydrogen. When a neutron collides with carbon, its direction can change up to 180 degrees; additionally, the neutron can only transfer up to 28 percent of its initial energy [2]. Thus hydrogen is much more effective at reducing (moderating) neutron energy.

Because they lack charge, neutrons are difficult to detect. Neutrons are detected as they interact with other particles, producing charged particles which are subsequently detected. Gamma rays are common sources of background radiation because they can Compton scatter, which produces electrons that can create a signal similar to that from the products of neutron interactions.

1.2.2 Lithium-6 Detector

Our neutron detector is composed of ^{6}Li-doped glass scintillator, plastic scintillator, and a photomultiplier tube. ^{6}Li has a high probability of capturing neutrons, creating charged particles that can be easily detected. Neutrons are absorbed by ^{6}Li, exciting the nucleus. The newly formed, highly energetic ^{7}Li undergoes fission, splitting into an alpha particle and a triton:

$$\mathrm{n} + {}^6\mathrm{Li} \rightarrow {}^7\mathrm{Li}^* \rightarrow \alpha(2.056\mathrm{MeV}) + {}^3\mathrm{H}(2.729\mathrm{MeV}). \tag{1.1}$$

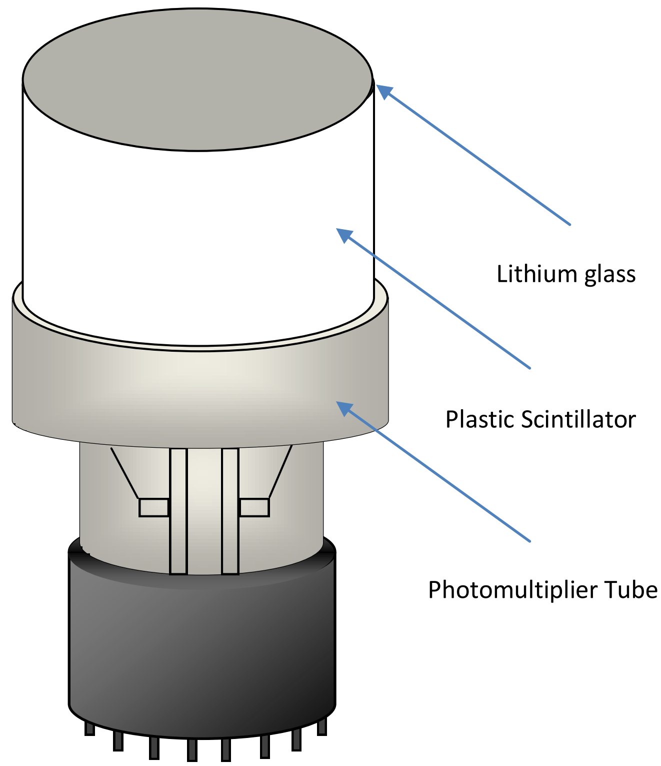

Figure 1.1 Lithium Glass Neutron Detector

The alpha particle has a range of 7 μm in the lithium glass while the triton has a range of 40 μm. The glass we use is 1 mm thick, thus the alpha and triton usually deposit all of their energy in the glass. The cross section of this interaction increases with decreasing neutron energy.

The plastic in the detector is composed of hydrogen and carbon, which moderate neutron energy. As neutrons enter the plastic, they collide with the hydrogen and carbon atoms; each collision changes the neutron's energy and direction. Figure 1.1 shows a diagram of a simple lithium glass detector. Neutrons may initially pass though the lithium glass but can still scatter back from the plastic and be captured in the glass. Because of this backscattering, adding additional plastic increases the number of neutrons that capture in the glass.

Both the plastic and the glass are scintillators. Scintillators are materials that become excited as charged particles pass through them. The excited atoms then release the excitation energy by isotropically emitting light. The amount of light a scintillator emits increases as the energy the particle deposits increases. This amount differs depending on the type of particle depositing the energy. To quantify this difference, the energy deposited by any particle is given in units equal to the amount of energy deposited by an electron of equal energy. For example, the alpha particle and triton from a ^{6}Li-neutron interaction together deposit 1.6 MeV electron equivalent (MeVee) [3]. That is equivalent to the amount of energy that a 1.6 MeV electron would deposit. The light emitted by the scintillators reflects inside the detector until it enters the photomultiplier tube (PMT).

The incident light ejects electrons from a thin cesium coating inside the PMT. These electrons are accelerated into a dynode where additional electrons are emitted. This acceleration process continues through several stages. A PMT multiplies the current produced by photoelectrons by roughly a factor of a million. This output pulse is sent to a waveform digitizer where the input voltage is converted to a digital output.

The digitizer records the output voltage every four nanoseconds. If the voltage increases above a threshold, the digitizer triggers and begins writing an event to an internal buffer. Each event consists of a small amount of initial background followed by a larger voltage pulse (see Fig1.2). The output voltage is proportional to the amount of light that enters the PMT. Thus, a particle that deposits a large amount of energy in a scintillator, causing it to emit a large amount of light, produces a large pulse.

1.2.3 Detection Difficulties

For the detector to be useful, it must be able to differentiate between gamma rays and neutrons, have a large solid angle, and detect neutrons in a wide spectrum of energies.

Gamma rays Compton scatter in the detector, transfering energy to electrons. These electrons excite the scintillator exactly the same way as an alpha particle and triton from a ^{6}Li-neutron interaction. A scintillator emits light regardless of whether the charged particle is positive or

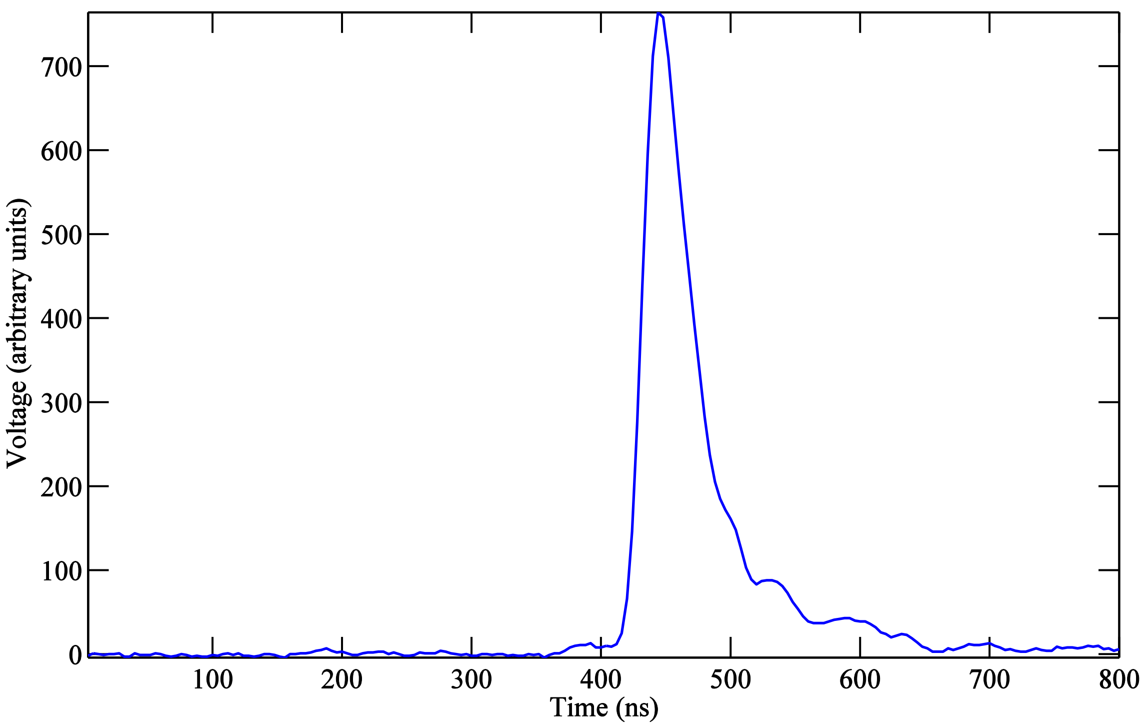

Figure 1.2 A Digitized Light Pulse

negative. Interactions from both gamma rays and neutrons produce light pulses in a scintillator. However, the pulses produced by neutrons and gamma rays differ in shape, so analysis of the digitized waveform can determine what type of particle produced the pulse. This process is called pulse-shape discrimination.

The detector's efficiency is defined as the ratio of the number of neutrons detected to the number entering the detector. The number of neutrons entering the detector is calculated by first determining the number of neutrons emitted by the source, based on the known source activity. This number is then multiplied by the solid angle of the detector divided by 4π. The solid angle is a measurement of how large the object appears to a source. This measurement takes into account the size of the object and the distance to the source. The solid angle is the surface integral over the angle of the object as seen by the source:

$$\Omega = 2\pi \int \sin\theta d\theta. \tag{1.2}$$

If a source were placed in a sphere then the solid angle would be 4π.

In nuclear non-proliferation applications the amount of shielding used is not under our control. Therefore, neutron detectors must be able to detect moderated or unmoderated neutrons. Water can be used as a moderator to decrease neutron energy. Additionally, hydrogen in the water can capture neutrons to form deuterium. Thus many neutrons may be captured by the water and never reach the detector. We need to be able to detect the presence of neutrons despite this decrease in the number and energy of neutrons that reach the detector.

1.3 Monte Carlo for Neutral Particles

Monte Carlo is a method of simulating a system based on probabilities [4]. The probabilities of particular interactions occurring are well know for most nuclear interactions. A program called Monte Carlo for Neutral Particles (MCNP) was developed by scientists at Los Alamos National

Laboratory to calculate how neutrons and photons interact with matter. Using MCNP we can model a neutron detector by defining its chemical makeup and spacial layout. A radioactive source is defined by its emission rate as a function of neutron energy. Each individual neutron is traced as it leaves the source. Every time the neutron comes near a particle, its probability of interacting with the particle and the outcome of the interaction is determined. Because the interactions are all probabilistic, this gives a good simulation of what would actually occur. By testing millions of neutrons, we can find the number of neutrons that would interact in particular parts of the detector within a reasonable uncertainty.

MCNP provides efficiency optimization without the effort of building many detectors. By modeling different configurations of the detector we can determine how to best build our detector and design our experiments.

Chapter 2

Experimental Method

In this chapter I describe the configuration and efficiency optimization of the neutron detector. Then I describe the different methods used to test the detector and analyze the output data.

2.1 Experimental Set Up

2.1.1 Configuration

The spatial configuration of plastic and glass determines neutron detection efficiency. Our original concept for ^{6}Li glass neutron detectors employed multiple layers of glass. Each glass layer was placed downstream from a 2.4 cm layer of plastic scintillator. We believed that each layer of plastic would moderate the neutron energy and the neutrons would then be captured by subsequent layers of glass. We began MCNP simulations of a detector with five layers of glass and six of plastic.

We produced a source of moderated neutrons by placing containers of water between a californium-252 source and the detector. We found that the multi-layer detector had a much lower detection rate for these moderated neutrons. This was counterintuitive because ^{6}Li has a higher cross-section for capturing low-energy neutrons. As the neutrons lost energy in collision with the water, the neutrons should have been more likely to capture in the glass. However, the plastic on the front surface of the detector was continuing to moderate the already low energy neutrons. As these low-energy neutrons thermalized in the plastic, they did not move deeper into the detector. Thus fewer neutrons would penetrate the plastic and be captured in the glass. The amount of moderation in a five-layer detector proved to be too much.

Removing the first layer of plastic increased neutron capture efficiency for moderated neutrons. Without a front layer of plastic, neutrons would first interact in lithium glass. Moderated neutrons were readily captured in the glass because of ^{6}Li's high capture cross-section for low-energy neutrons. If a source is not moderated, many of the neutrons pass through the front layer of glass. However, the neutrons then enter the plastic and many scatter back into the glass. Placing glass on the detector's front surface gave increased capture efficiency for moderated neutrons without sacrificing efficiency for unmoderated neutrons.

The back four layers of glass in a five-layer detector captured fewer neutrons. In fact, MCNP suggested that the first layer of glass accounted for over 90 % of all neutrons captured when the source was placed in the center of a sphere of water 60 cm in diameter. A single layer lithium-glass detector with plastic scintillator is nearly as effective at detecting neutrons as a five-layer detector and costs significantly less.

We used a single layer detector with 1 mm of glass and 4 cm of plastic for most of our experimental tests. According to MCNP, simulations this small amount of plastic does increase the lithium capture efficiency. Although MCNP indicated that the capture efficiency is somewhat larger with slightly thicker plastic, we had 4 cm scintillator disks available. This simple detector allowed us to experimentally test the efficiency of the glass. This model was also sufficient for determining the best method to differentiate between neutrons and gamma rays.

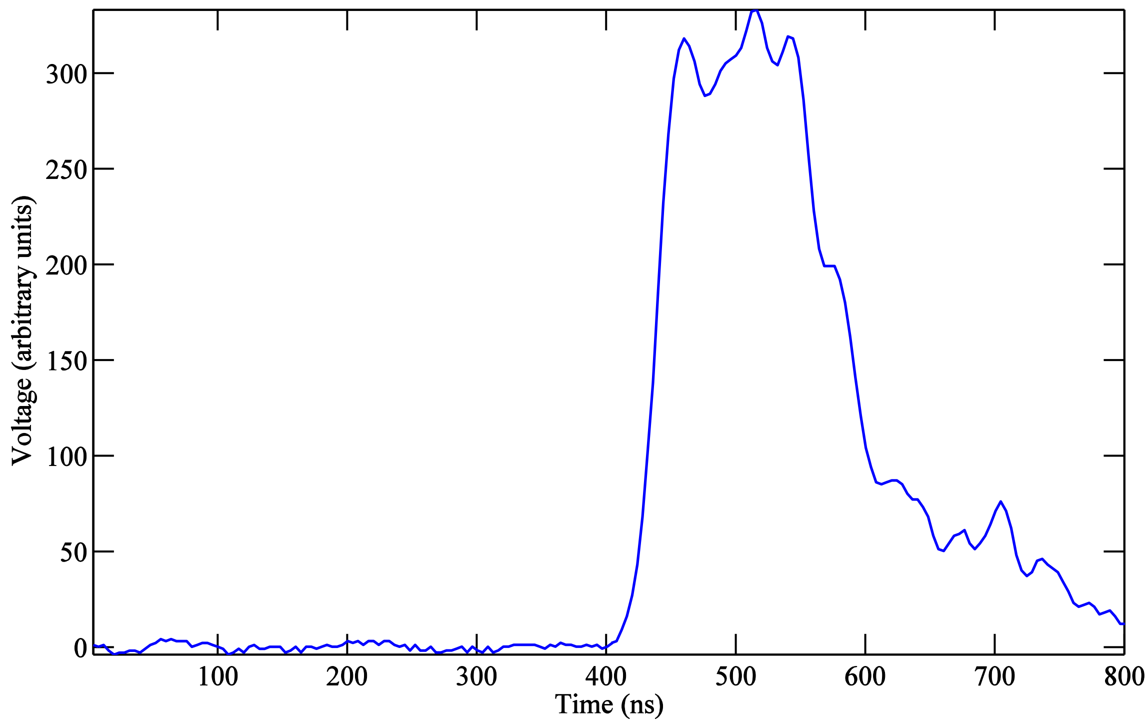

Figure 2.1 A Typical Glass Pulse.

2.1.2 Components

A good neutron detector not only has high efficiency, but also can effectively discriminate between neutrons and gamma rays. In our detectron this is accomplished by comparing the shape of pulses emitted by lithium glass and by plastic scintillator.

We chose two types of plastic for pulse-shape discrimination [5]. Eljen 240 (EJ 240) is a slow-emission plastic scintillator. While energy is deposited into EJ 240 in less than a nanosecond, the plastic emits the light over about 230 ns. This slow emission time gives pulses that have a long tail. These wide pulses are compared with the pulses from the glass, which has an emission time of 70 ns (see Fig.2.1).

Eljen 212 (EJ 212) is a fast-emission plastic. The light from EJ 212 is emitted over 2.4 ns. Fast emission time means the light is quickly collected by the PMT and gives a pulse that is narrow in time (see Fig. 2.2). This can be compared to the light pulse from glass, which is much wider (see Fig. 2.1).

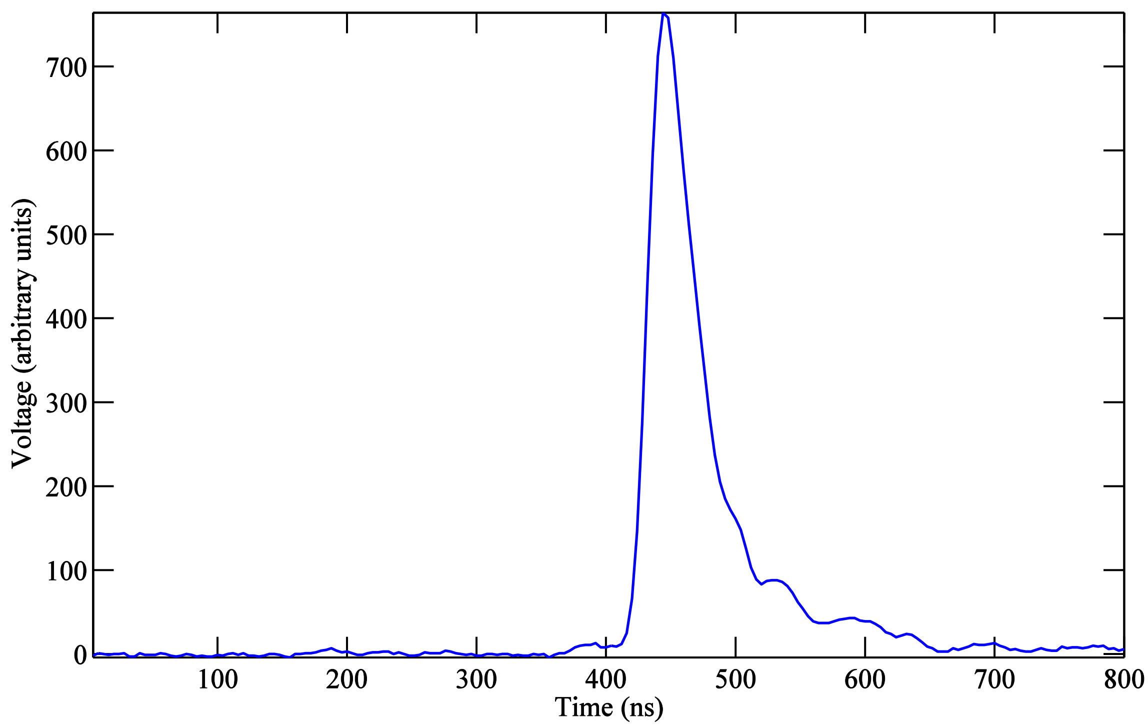

Figure 2.2 A Typical EJ 212 Pulse

Once light is emitted from the scintillators, it reflects in the detector until it reaches the PMT. We placed a thin reflective layer of aluminized mylar on the outside surface of the detector to help increase light collection. We placed a thin layer of mineral oil between the glass and plastic to reduce reflection at the interface. Additionally we coupled the plastic to the PMT with a layer of optically transparent silicon grease.

2.1.3 Sources

We used a radioactive ^{252}Cf source to test the neutron capture efficiency. ^{252}Cf spontaneously fissions, emitting gamma rays and neutrons. Fission gamma rays and background gamma radiation are similar in effect. Therefore the increase in gamma radiation from the fission source does not introduce new or unexpected elements into the experiment. ^{252}Cf emits a wide energy spectrum of neutrons (see Fig.2.3), which allows us to test the response of our detector over a wide energy spectrum.

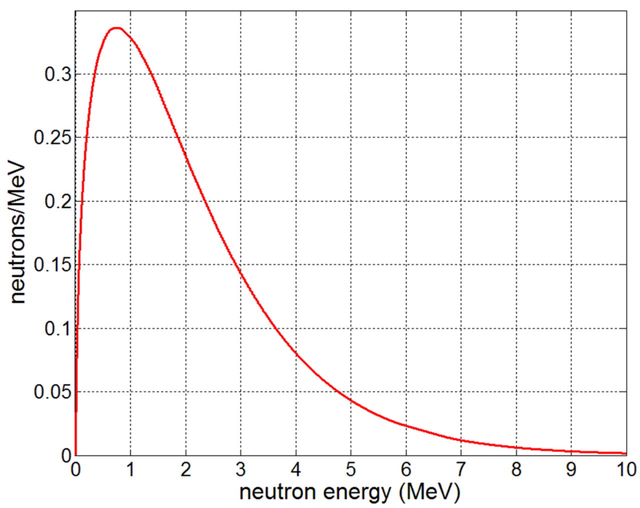

Figure 2.3 Neutron Emission Spectrum for ^{252}Cf Spontaneous Fission

We used a pure gamma source, ^{60}Co, to determine the response of the detector to gamma rays. ^{60}Co spontaneously emits two gamma rays in uncorrelated directions. Both of these gamma rays are released within a picosecond of each other. Because of the limited response time of a PMT (on the order of nanoseconds), the ^{60}Co gamma rays can be considered to be emitted simultaneously. The two gamma rays have energies of 1.17 MeV and 1.33 MeV. If both gamma rays were to Compton scatter in the glass, they could deposit as much energy as a triton and alpha from a neutron-^{6}Li interaction.

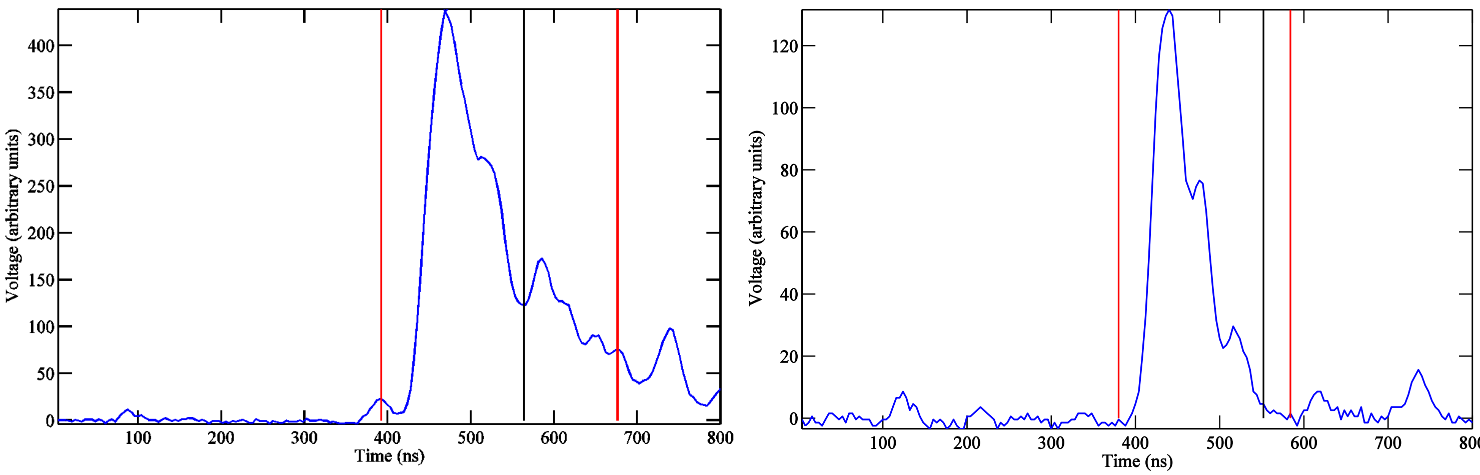

Figure 2.4 Glass and plastic pulses divided into early and late area

2.2 Data Analysis

The difference in pulse shapes of lithium glass and the plastics provides a method of discriminating between neutron and gamma radiation. The digitized PMT signal is imported into MatLab and placed in an array. Each pulse is characterized by area, peak height and location, width, early area, late area, and tail area. A line is drawn dividing the pulse(See Fig.2.4). The dividing line is place a fixed time after the start of the pulse. All of the area between the first red line and the black line is defined as early area. Late area is the area after the black line to the second red line. The scintillator emits some small pulses of light after the main pulse. Tail area consists of any area after the main pulse. By breaking up area into components, differences in the shape of glass and plastic pulses can be characterized.

We compared the different components of glass and EJ 212 plastic pulses to determine which pulse characteristics would yield the best separation. For glass pulses only around 70 percent of the total area is early area (See Fig.2.4). For plastic pulses early area is a large fraction of total area(See Fig.2.4). This allowed us to define a range of early area for glass pulses that was separate from the range for plastic pulses.

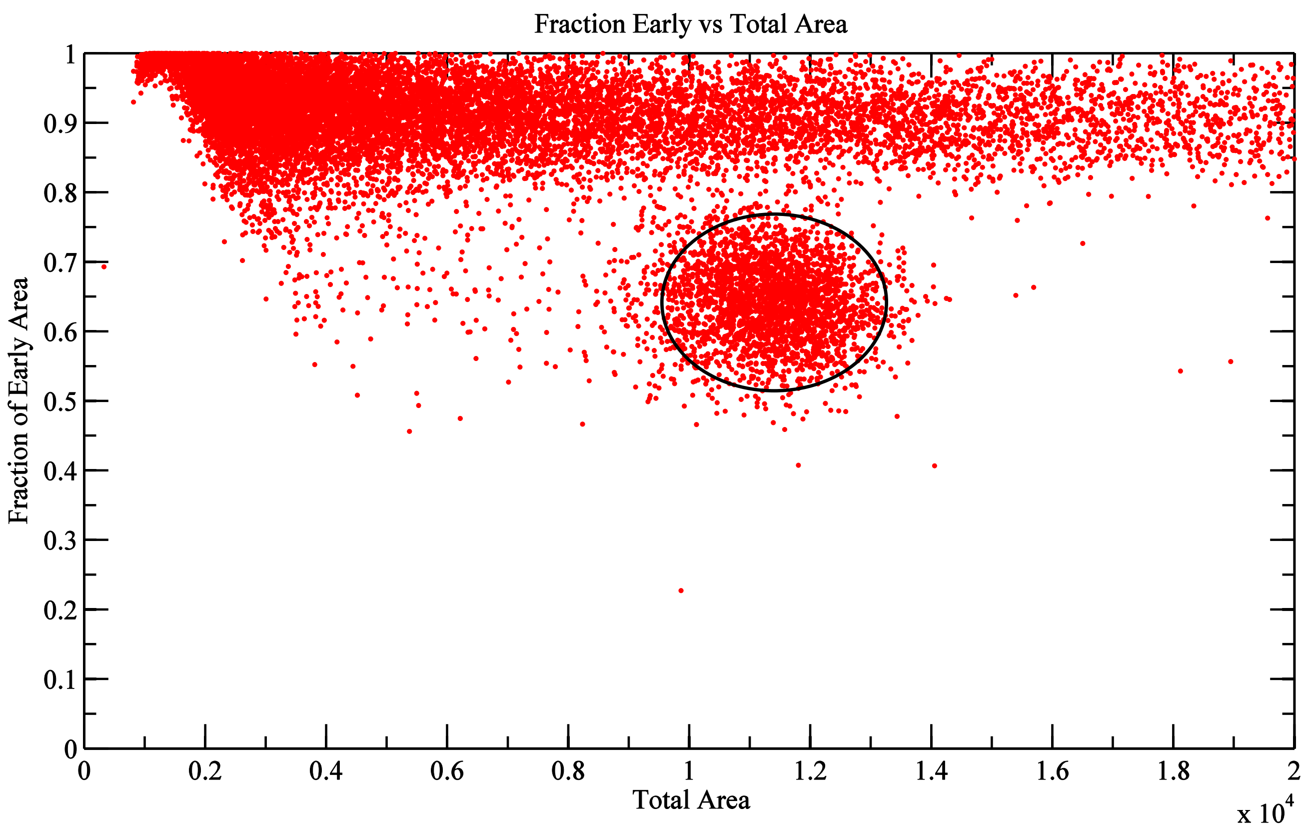

Figure 2.5 Data taken by detector under a ^{252}Cf source. Pulses inside the window are attributed as neutrons

We defined a window of early area vs total area as a neutron window. The triton and alpha particle from the neutron-lithium interaction deposit 1.6 MeVee in the glass. Because this capture interaction is mono-energetic, all neutron pulses have similar total area. However, the amount light collected by the PMT is not always the same because of the detector's optical properties. Additionally, the amplification from the PMT may differ from pulse to pulse. This gives a small spread in area that encompasses most neutrons. The total area and early area from every pulse generated in the glass was very similar. Using this small spread we defined a window in which 90 percent of the neutrons were found (see Fig.2.5).

When gamma rays Compton scatter in the glass, the resulting electrons may remain in the glass. If they do, the electrons produce pulses that have same shape as those produced by neutrons. Thus we can only differentiate between glass pulses and plastic pulses. Electrons have a range of roughly 1 mm in lithium glass, but because the glass is only 1 mm thick, many of the energetic electrons that are produced in the glass deposit a portion of their energy in the plastic. The emission time of plastic gives the resulting pulses a very different early area than that of a captured neutron. Gamma rays only produce pulses that appear to be neutrons if they scatter electrons that remain in the glass and have roughly the same energy as triton and alpha particles from a neutron-lithium interaction.

For gamma rays from ^{60}Co to give a signal in the neutron window, both gamma rays must scatter in the glass. Additionally both electrons must deposit most of their energy in the glass. We tested this effect by placing the cobalt source 2 inches away from the detector. With such a large solid angle, there was a larger probability that both gamma rays would interact in the glass. However, because the solid angle for a cylinder facing the source drops off roughly as $1/r^2$, the probability of two gamma rays simultaneously interacting in the glass decreases by $1/r^4$. This means a more radioactive ^{60}Co source further away from the detector would be less of a problem than a weaker source close to the detector.

Chapter 3

Results

3.1 Computational Results

The MCNP code simulates the detector and shows the effects of each component of a ^{6}Li detector. In this section I discuss simulations that show how moderation, plastic thickness, glass thickness, and multiple layers of glass and plastic affect neutron detection efficiency (See Fig. 1.1).

Monte Carlo simulations show that neutron capture efficiency of ^{6}Li glass is affected by moderation of neutron energy. If a source is placed in a sphere of water with a 16 cm radius, the water captures 59 percent of the neutrons. With a radius of 32 cm, 95.4 percent of the neutrons never escape the water (See Fig. 3.1). Because of the decrease in the number of neutrons that can interact in a detector, most methods of neutron detection are negatively affected by moderation. However, because of the increase in neutron-capture cross section with decreasing energy, moderation improves the efficiency of ^{6}Li neutron detectors. Figure 3.2 shows how the intrinsic efficiency of a single sheet of glass is affected by an increasing amount of moderator being placed around the source. Most importantly, this graph shows that the detector is more effective with shielding than it is without.

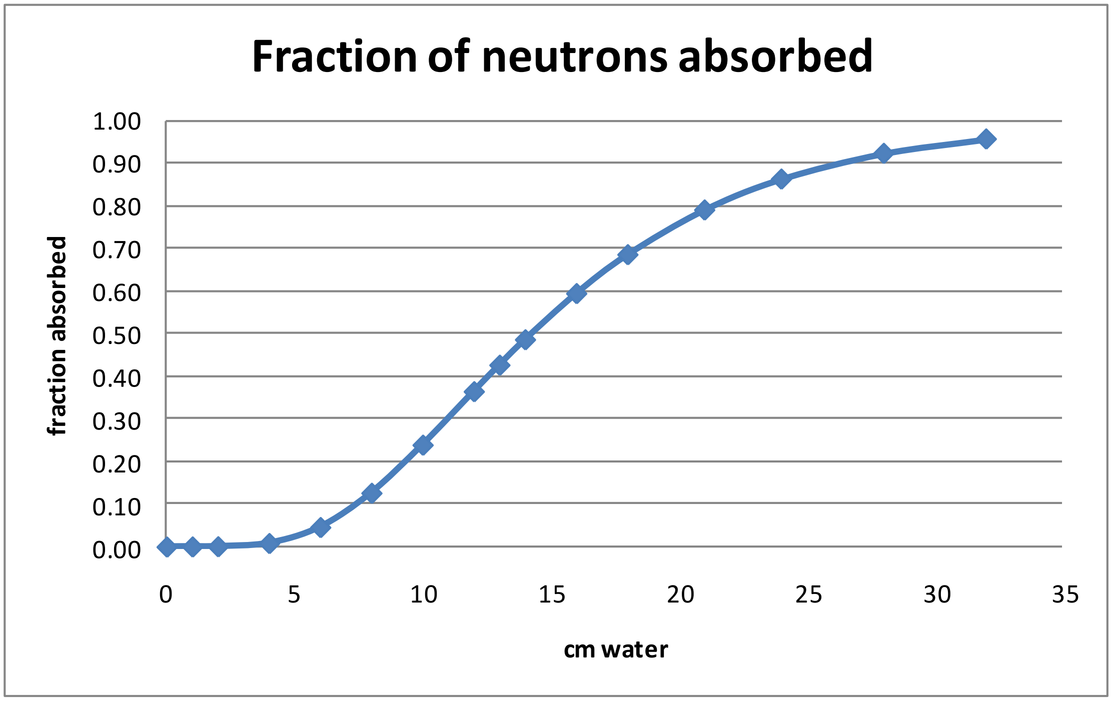

Figure 3.1 Fraction of neutrons that get absorbed in water

For non-proliferation applications, the amount of moderation used is unknown and uncontrollable. In the absence of an external neutron moderator, plastic thickness increases detection efficiency. Without external moderation, 16 cm of plastic between the glass and PMT increases efficiency by more than a factor of 95. However, if the source is placed at the center of a 16 cm radius sphere of water, the difference between no plastic and 15 cm is only a factor of 1.33. The plastic acts as a moderator and is necessary in applications when the amount of moderator is unknown. The detector is nearly at optimum efficiency for all thicknesses of shielding with 4 cm of plastic (See Fig. 3.2). Regardless of shielding more than 8 cm of plastic is not effective.

Increasing glass thickness improves efficiency at the cost of increasing the number of gamma rays that are misattributed as neutrons. With 30 cm of water the increasing the glass thickness from 1 mm to 1.5 mm increases efficiency by 1.18 percent. However, because MCNP does not

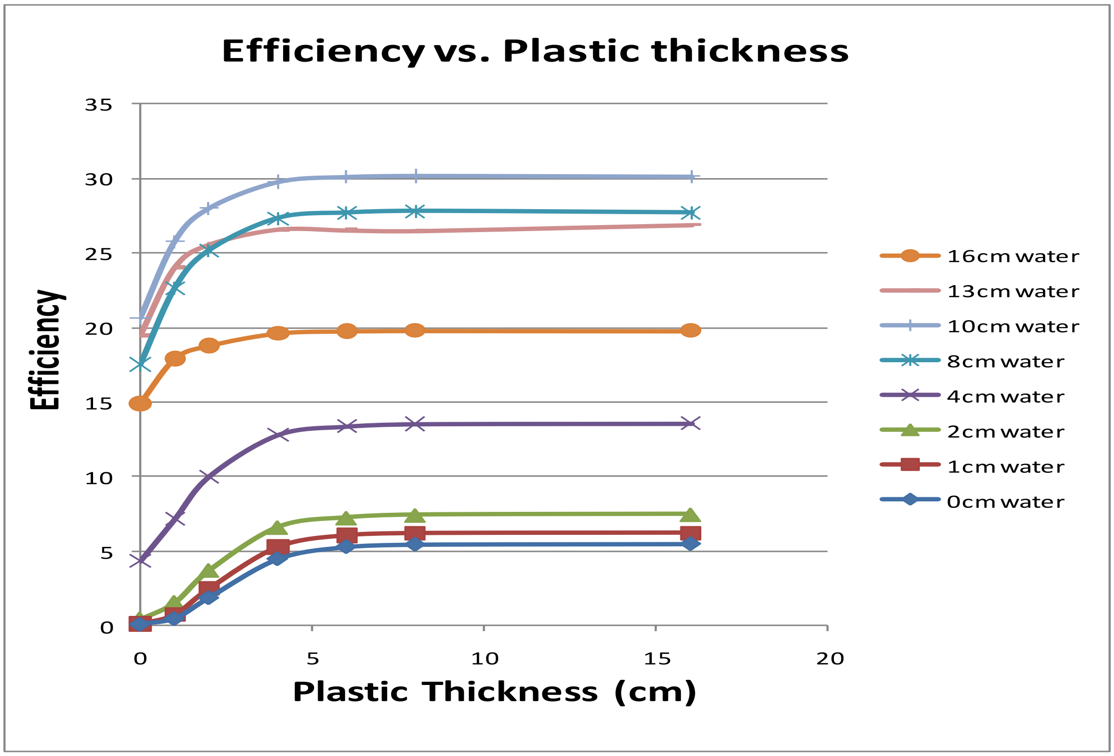

Figure 3.2 Effectiveness of plastic with various amounts of shielding

take gamma radiation into account, these data do not tell the full story. Thicker glass allows more gamma rays to Compton scatter and deposit their energy in the glass. That means more gamma rays falsely appear to be neutrons. Therefore, the small increase to neutron detection efficiency due to thicker glass may not be advantageous unless good gamma-neutron discrimination is not required.

The five alternating layers of glass and plastic is not the most efficient use of materials. Even with only an 8 cm radius sphere of water for moderation the front layer of glass is 2.25 times more effective than the other four layers combined. Any attempt to bring radioactive materials into the US can be expected to be heavily moderated to block neutrons. By splitting a five-layer detector into five single layer detectors we can increase the solid angle and therefore absolute efficiency by a factor of five. Five single layer detectors gives the most efficient use of glass.

3.2 Detector Efficiency

Intrinsic efficiency is the ratio of the number of incident neutrons to the number detected. With an optimum amount of shielding (10 cm), we measured an intrinsic efficiency of 30 percent.

Using pulse shape discrimination, we reduced gamma contamination to 1 in 10 000. Plotting early light vs pulse area on a graph gives a small window where pulses are defined as neutrons. When a neutron source is absent there are still a few counts that get classified as neutrons. Figure 3.3 shows data from the detector when placed under a ^{60}Co source. The neutron window is circled. The events in this window are thought to be due to high energy gamma rays that produce electrons, which deposit most of their energy in the glass. Based on the number of gamma rays emitted from the source and the solid angle of the detector there is only one gamma ray pulse in the neutron

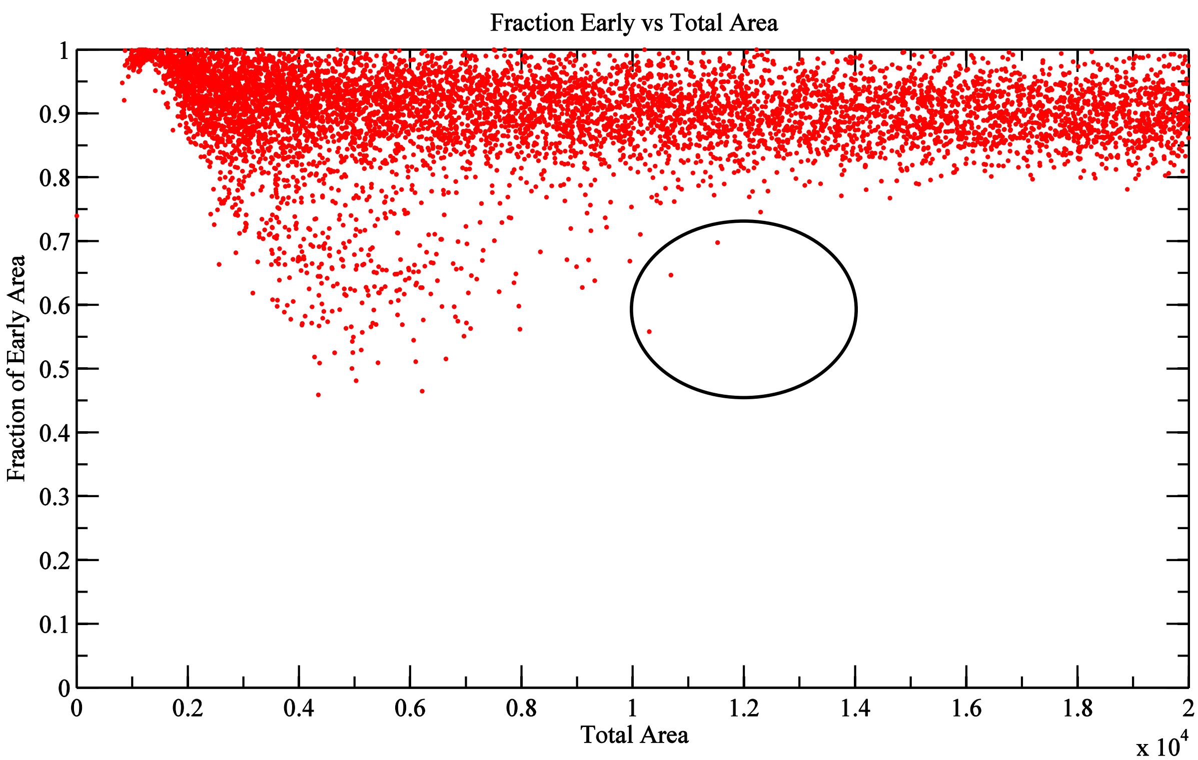

Figure 3.3 Data taken by detector under a ^{6}0Co source. The pulses inside the neutron window are misattributed neutrons.

window for every 10 000 incident gamma rays.

3.3 Conclusion

^{6}Li provides an efficient, inexpensive means of detecting neutrons. Optimally built ^{6}Li detectors include a PMT, an 8 cm thick disk of EJ 212 plastic and a 1 mm sheet of glass. The plastic moderates the neutron energy; in the absence of an external moderator, 8 cm of plastic increases neutron detection efficiency by a factor of 94. Additionally, EJ 212 provides a method of pulse-shape discrimination that allows us to eliminate all but one out of ten thousand gamma rays.

The number of gamma rays misattributed as neutrons in our detector is sufficiently low for most applications. However, the Department of Homeland Security wants a ^{3}He replacement to have only one out of a million gamma rays detected by the detector to give a false neutron count [6]. Having extensively examined the data from gamma rays in ^{6}Li glass detectors, there is little chance that improved post processing will be able to achieve the requirement. Work is currently being done to try to reduce the amount of energy a Compton-scattered electron will be able to deposit in the glass. Any methods used must still allow triton and alpha particles to deposit all of their energy. This work will hopefully create a detector that is very efficient at detecting neutrons, with almost no misattributed gamma rays.

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