An Analysis of the Scintillation Properties of Several Materials for Radiation Detection

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AN ANALYSIS OF THE SCINTILLATION PROPERTIES OF
SEVERAL MATERIALS FOR RADIATION DETECTION

By
LUKE JOHN LAFFEY

A thesis submitted in partial fulfillment of
the requirements for the degree of
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in Materials Science and Engineering

ROCHESTER INSTITUTE OF TECHNOLOGY
College of Science
School of Chemistry and Materials Science

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To the Faculty of Rochester Institute of Technology:

The members of the Committee appointed to examine the thesis of LUKE JOHN LAF-FEY find it satisfactory and recommend that it be accepted.

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AN ANALYSIS OF THE SCINTILLATION PROPERTIES OF SEVERAL MATERIALS FOR RADIATION DETECTION

by Luke John Laffey,
Rochester Institute of Technology
August 2020

Advisor: Sheth Nyibule, Ph.D.

Abstract

The accurate detection and identification of ionizing radiation and radioisotopes is of significant interest to governments, industry and the scientific community, particularly for use in detecting illicit radioactive weapons, among other uses. Of all the methods to detect radiation, scintillation has been a mainstay among handheld or portable detectors owing primarily to its simple equipment requirements. In particular, inorganic crystals and plastic scintillators are two scintillating materials with promising characteristics for use in particle identification. In this work, two promising materials are compared to more common detectors of the same type. The inorganic scintillator SrI$_2$:Eu$^{3\%}$ is compared to 2 other inorganic crystals, NaI:Tl and LaBr$_3$:Ce on the basis of its energy resolution. The energy resolution for several $\gamma$-ray energies were measured and SrI$_2$:Eu$^{3\%}$ was found to have an energy resolution of $(6.7\pm0.1)\%$ for the 662 keV photopeak. SrI$_2$:Eu$^{3\%}$ shows significant promise for use in handheld radiation detectors due to its better energy resolution than NaI:Tl and simpler background than LaBr$_3$:Ce. Also investigated was plastic scintillator EJ-299-33A on the basis of its n/$\gamma$ discrimination capabilities. Two sizes of EJ-299-33A, 2” $\times$ 1” and 2” $\times$ 2” were compared to NE-213 through several different pulse shape discrimination methods.
Both sizes of EJ-299-33A were found to have very similar discrimination capability, and were comparable to NE-213. The best discrimination for all detectors was by comparing the PID vs. the pulse height. EJ-299-33A was found to have a figure of merit of 1.03 at energies of about 250 keVee for both sizes. EJ-299-33A shows promise for use in n/γ discrimination where liquid scintillators are not practical.
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Dedication

This thesis is dedicated to my mother and father who provided everything I needed to be successful in my education and in life.
Chapter One

Introduction

1.1 Overview of Strontium Iodide

Measurement of radiation interactions by scintillation mechanism is one of the oldest and most employed methods of radiation detection. Due to high Z-composition, inorganic scintillators play a vital role in the detection of gamma-rays (γ-rays) and charged particle interactions. Neutron detection is achieved using organic scintillation due to their high $^1\text{H}$ content. Thallium-doped sodium iodide crystals (NaI:Tl) and cerium-doped lanthanum bromide crystals (LaBr$_3$:Ce) are the two most common inorganic scintillators in use today. NaI:Tl crystals are relatively easy to grow, but have relatively poor light output, slow response times, and energy resolutions of about 7-10%. LaBr$_3$:Ce has better energy resolution of about 3-4%, and better light output, but is harder to grow and has internal radiation (Milbrath et al., 2006). Situations requiring high resolution and simple background spectrum had no solution in current inorganic scintillators. Efforts to solve this problem led to the rediscovery of europium-doped strontium iodide (SrI$_2$:Eu). SrI$_2$:Eu has the advantage of increased light output over NaI:Tl, comparable energy resolution to LaBr$_3$:Ce, and does not have internal radiation. The high light yield and lack of internal radiation makes SrI$_2$:Eu desirable for use in handheld radiation monitors.

Many efforts have been made to optimize the performance of SrI$_2$:Eu crystals. The ease
and stability of growth and crystal size have been thoroughly investigated. Doping with Europium to optimize light output and energy resolution has also been investigated, with doping rates in the range of 1% - 7% being of primary interest. The goal of our experiment is to characterize a 1” by 1”, 3% Eu-doped SrI$_2$ crystal. The effect of tapering the end of the crystal facing the radioisotope is also under investigation by other researchers in the field but was not considered in the present work. Preliminary results suggest tapering leads to better energy resolution. The SrI$_2$:Eu crystal used in the present experiment was untapered. SrI$_2$:Eu is currently finding applications in experimental handheld detectors for field identification of radioisotopes (Milbrath et al., 2006) (N. J. Cherepy et al., 2017).

1.2 Gamma Radiation Detectors

The detection of faint, radioisotope signals using gamma spectroscopy has found application in a variety of fields. These include astronomy in mapping the radioisotope composition of extraterrestrial bodies and in radioisotope identification (RIID), particularly for security applications. The detection of faint signals requires detectors with high light output, short relaxation time, and good energy resolution. Energy resolution is a measure of the spread of the photopeak relative to its energy. High-Purity Germanium (HPGe) detectors are the gold standard by which other detectors with full-energy photoelectric absorption are judged. Their energy resolution is orders of magnitude better than Sodium Iodide (Prettyman et al., 2015)(Knoll, 2000).

One difficulty with HPGe detectors is that they require complicated supporting equipment, increasing cost and decreasing ease of use, which limits their use in space and security applications. Inorganic scintillators are the detector of choice in these cases due to their high light output and linearity. The 2 major inorganic scintillators in use today are thallium doped sodium iodide (NaI:Tl) and cerium doped lanthanum bromide (LaBr$_3$:Ce). NaI:Tl is relatively cheap and easy to manufacture, but has energy resolutions that are too poor for
highly sensitive applications. LaBr$_3$:Ce has higher light output and better energy resolutions than NaI:Tl, but suffers from difficult crystal growth and a complex internal spectrum. LaBr$_3$:Ce has a broad peak due to internal radiation from the $\beta$-decay of lanthanum at 1436 keV, contributions from x-rays around 35 keV, and a very complex spectrum above 1436 keV that is theorized to be contributions from $\alpha$-particles. These obfuscating factors make LaBr$_3$:Ce’s use in RIID difficult. Both detectors have relatively slow response times, an issue with inorganic scintillators as a whole. (Milbrath et al., 2006)

1.3 Europium Doped Strontium Iodide

Several europium-doped alkaline earth halogens have recently been investigated as inorganic scintillators for use in place of NaI:Tl or LaBr$_3$:Ce. SrI is a promising crystal for us in inorganic scintillation due to several factors: excellent linearity in its response to radiation, a light output higher than LaBr$_3$:Ce, a relatively fast response time, a lack of internal radiation, and excellent energy resolutions. Recent experiments have found energy resolution for SrI$_2$:Eu to be less than 3% for the 662 keV photopeak. Its potential for use in particle identification has spurred significant research into characterizing its use in various applications. The work done here seeks to add to this body of research.

1.3.1 Crystal Growth

Much research into SrI$_2$:Eu has gone into studying the growth of SrI$_2$ crystals, which have an orthorhombic crystal structure. Early experiments showed that normal growth methods led to frequent cracking in SrI$_2$ crystals despite the low anisotropy in the coefficient of thermal expansion that allows for fast cooling without issue. The development of the vertical Bridgman technique has successfully overcome these challenges in crystal growth for SrI$_2$ and helped to secure its place as a detector for use in RIID applications. (Wilson et al., 2008)(N. Cherepy et al., 2009)
1.3.2 Europium Doping

The amount of europium doping and how it affects the performance of SrI has been of great interest in recent experiments. Doping from 0.5% to 8% europium or more has been explored. The emission spectrum and decay time for SrI$_2$:Eu were found to be independent of the amount of Eu doping. The peak emission wavelength is approximately 430 nm. Linearity is also very consistent across doping concentrations. The light yield and energy resolution for a 1.5” × 1.5” cylindrical crystal were measured to be best for doping concentrations of 3% - 6%. Light yields for these concentrations were found to be >80,000 Photons per MeV, in some cases as high as 120,000 Photons/MeV. Energy resolution were found to be <4% at 662 keV (Wilson et al., 2008)(N. Cherepy et al., 2009)(N. J. Cherepy et al., 2017).

1.3.3 Crystal Size

Crystal size has significant impact on the energy resolution and light yield. SrI$_2$:Eu has substantial overlap in its emission and absorption spectra, which can lead to re-absorption of the emitted light. Larger crystals can experience many absorption and emission events, substantially increasing their response time, while decreasing resolution and light yield. Smaller crystal size decreases the amount of absorption that can occur. One way to mitigate the effects of absorption in larger crystals is to taper the crystal slightly. Tapering the crystal shows energy resolution improvement for crystals of size 1.5” x 1.5” in both digital and analog analysis. (Wilson et al., 2008)(N. Cherepy et al., 2009)(N. J. Cherepy et al., 2017)

Several radiation detection companies have developed SrI$_2$:Eu scintillating crystals for use in laboratory research. These scintillators produce varying results, but measurements are comparable to what scientific research has concluded. Some of these companies have developed SrI$_2$:Eu integrated detectors that include integrated photomultiplier tubes and basic electronic outputs. These commercialized scintillators show a promising outlook for SrI$_2$ in future radiation detection applications. (N. J. Cherepy et al., 2017)
1.4 EJ-299-33A - A Plastic Scintillator

EJ-299-33A is a plastic scintillator developed by Lawrence Livermore National Laboratory and produced by Eljen Technologies that holds particular promise in neutron/γ discrimination. Unlike many other plastics capable of this discrimination, EJ-299-33A is significantly more stable. Prior plastics suffered significant degradation over time that limited their use outside of the laboratory. EJ-299-33A is found to have a good long term stability, and is safe from degradation due to water, weak acids and bases, and low alcohols. EJ-299-33A’s stability and n/γ discrimination capabilities make it an enticing choice for several applications, but more research is needed.

EJ-299-33A has already been shown to be capable of discriminating alpha particles and protons from neutrons and γ-rays, and its light output is well studied. EJ-299-33A’s discrimination capabilities have been compared to liquid scintillators in several other studies, but primarily for detectors of larger size or at higher energies. This thesis seeks to add to this body of research by studying the discrimination capabilities of EJ-299-33A for smaller detector size and lower energy. EJ-299-33A's of size 2'' × 2'' and 2'' × 1'' are compared. The best method of comparison is also investigated. (Liao and Yang, 2015) (Nyibule et al., 2013) (Pozzi, Bourne, and Clarke, 2013) (Pozzi, Bourne, Dolan, et al., 2014) (E. Pagano et al., 2018) (Woolf et al., 2015)
Chapter Two

Background

The detection of gamma-rays by scintillation mechanism is one of the most commonly employed techniques in radiation detection. The principle operation of a scintillator is the conversion of high-energy x-rays and γ-rays into lower energy photons that are easier to detect, most often in the visible spectrum. A high-energy photon impingement upon a scintillating material deposits its energy by ionizing charged particles in the detector, often electrons. As these ions move through the detector, they create excited states. For organics, these excited states are within the electronic structure of the molecule. For inorganic scintillators, the excited states are due to electrons exciting into higher energy bands in the crystal lattice. These excited states decay primarily through the emission of a photon, though they can decay through other means. Both quickly decaying states (singlet states, normally excited states) and slowly decaying states (triplet states, states with forbidden transitions) can become populated and do so over a specific rise time. Both types of states decay with their own decay time, referred to as the characteristic time, contributing to the overall light emission by the scintillator. Response times of scintillating materials are dependent on the characteristic times of these excited states. The energy difference between excited states in the scintillator determines the emission spectrum. (Knoll, 2000)
2.1 Gamma-ray Interactions

There are several ways that γ-rays can impart their energy to a detector material. One way is through photoelectric absorption, where a γ-ray deposits all of its energy in a bound electron. The electron becomes ionized and its kinetic energy is equal to the γ-ray energy minus the electron binding energy. In practice, this binding energy is negligibly small relative to the total γ-ray energy, and the kinetic energy of the electron is taken to be equal to the γ-ray energy. The mechanisms of γ-ray interaction are described below in brief.

2.1.1 Photoelectric Absorption

The probability of photoelectric absorption is a function of atomic number, $Z$, and photon energy, $E$. High probabilities of photoelectric absorption are found in high-$Z$ materials. The cross section, $\sigma$ is given by eq. 2.1, where $n$ is a number that varies between 4 and 5. This relationship is used to the advantage of inorganic scintillators, whose high $Z$-value causes photoelectric absorption to be the dominant interaction mechanism. (Knoll, 2000)

$$\sigma \propto \frac{Z^n}{E^3}$$  \hspace{1cm} (2.1)

2.1.2 Compton Scattering

Another mechanism through which γ-rays can interact with matter is Compton Scattering. This occurs when the γ-ray ionizes an electron but imparts only a portion of its energy and scatters within the scintillator. The γ-ray may scatter several more times within the scintillator before leaving the detector. The several scattering events happen much faster than the population of the excited states, and as such the light from several Compton scattering events due to a single γ-ray reaches the photodetector at approximately the same time. Therefore all Compton scattering events from a single γ-ray can be considered a single event, and the total energy imparted is determined by the number of scatters and the angle during each scatter. (Knoll, 2000)
The energy imparted to an electron in a single Compton scattering event, $E'$, is a function of the scattering angle, $\theta$, and is given by eq. 2.2, where $m_e c^2$ is the rest energy of an electron and $E_\gamma$ is the pre-scatter energy of the incident photon. If the scattering angle is very small, such that the $\gamma$-ray is only slightly deflected from its pre-scatter path, the $\gamma$-ray will impart only a small amount of energy to the electron. The $\gamma$-ray will impart the most energy when the scattering angle is 180° ($\pi$ rad) from the initial trajectory, however this energy is less than the full energy of the $\gamma$-ray. The maximum energy able to be imparted by a photon during a single Compton scattering is the Compton Energy, $E_C$, given in eq. 2.3. For small detectors ($< 1$-$2$ cm), a $\gamma$-ray will only scatter once inside the detector, and the resultant scattered photon will leave the detector. In this case, resulting possible energies distribute as a plateau of energies up to the Compton energy. (Knoll, 2000)

$$E' = \frac{(E_\gamma) \frac{E_\gamma}{m_e c^2} (1 - \cos \theta)}{1 + \frac{E_\gamma}{m_e c^2} (1 - \cos \theta)}$$   \hfill (2.2)

$$E'|_{\theta=\pi} = E_\gamma \frac{1 + \frac{2E_\gamma}{m_e c^2} - 1}{1 + \frac{2E_\gamma}{m_e c^2}}$$

$$E_C = E_\gamma (1 - \frac{1}{1 + \frac{2E_\gamma}{m_e c^2}})$$ \hfill (2.3)

For medium sized detectors such as used in the experiments described herein, one $\gamma$-ray can scatter multiple times. In such cases, the energy can exceed the Compton energy as the imparted energy from multiple scattering events add together. The distribution of energies due to Compton scattering greater than the cutoff energy decreases with increasing imparted energy. (Knoll, 2000)

### 2.1.3 Pair Production and Annihilation

A third mechanism by which $\gamma$-rays can interact with a detector is through pair production. This process is only likely for high-energy $\gamma$-rays, and increases for high-Z detectors. For $\gamma$-rays with energy greater than twice the rest mass of an electron, 1.02 MeV, the $\gamma$-ray can
Figure 2.1 Schematic spectrum from a detector undergoing photo-absorption and pair production. The single escape peak is found at an energy 511 keV less than the photopeak and the double escape peak is 1020 keV less. Also shown is the annihilation peak, which can occur from $\beta^+$ sources. The $\beta^+$ annihilates with an electron, generating 2 photons of energy 511 keV. Not to scale.

Decay into an electron and a positron through interaction with the nucleus. Energy beyond the required 1.02 MeV is converted into kinetic energy in the electron and positron, which create excited states within the detector. Once the positron has deposited all of its kinetic energy, it will recombine and annihilate with another electron creating 2 $\gamma$-rays each with energy 511 keV. The positron deposits its energy very quickly, such that the annihilation happens a very short time after the pair production. (Knoll, 2000)

In detectors of medium size and high Z number, both, neither, or only one of the two 511 keV $\gamma$-rays can leave the detector without interacting. The $\gamma$-rays interact almost exclusively via photoelectric absorption, the entire process of which will occur over a short enough time scale to be nearly coincident with the original pair production. If both $\gamma$-rays are absorbed,
the full-energy of the original incident $\gamma$-ray will have been absorbed and it will contribute to the full-energy photopeak. In the event that one $\gamma$-ray escapes, the resulting energy will appear as a peak about an energy 511 keV less than the the photopeak. If both $\gamma$-rays escape, then the deposited energy is only equal to the kinetic energy deposited by the electron and positron, resulting in an energy peak that is 1.02 MeV less than the photopeak. These are labeled the single-escape peak and the double-escape peak, respectively. an example is shown in fig. 2.1. (Knoll, 2000)

### 2.1.4 Backscatter

Some $\gamma$-rays may scatter off of surrounding materials before being detected. It can be shown that for a given energy and scattering at angles greater than 120°, the scattered photons have a nearly identical energy. This energy ranges from 0.2-0.25 MeV, 0.25 MeV as the limit for high incident energy. A monoenergetic source of $\gamma$-rays will result in many backscattered gamma-rays of similar energy. As detectors are often surrounded by shielding which $\gamma$-rays can scatter off of, backscattered $\gamma$-rays can result in a peak in the energy spectrum around 0.25 MeV. (Knoll, 2000)

### 2.1.5 Spectrum Complications

There are several additional complications to the spectrum that can be encountered; 2 major ones are escape electrons and Bremsstrahlung radiation. Escape electrons are primarily electrons produced near the surface of the detector. These electrons escape the detector before they are able to impart all of their energy, distorting the spectrum. Events will be shifted towards lower energies, in particular increasing the relative intensity of the Compton plateau to the photopeak. Escape electrons are more likely to occur when the incident photon has a high energy causing the resultant electron to have a high kinetic energy. (Knoll, 2000)

Bremsstrahlung radiation occurs when a charged particle is accelerated, such as when an
electron is slowed or changes direction due to interactions with a solid material, and releases X-rays. Most of the X-rays produced by secondary electrons in the detector are promptly reabsorbed, and their energy is still picked up by the detector. However some of these X-rays can escape, decreasing the detected energy from the event and shifting features in the spectrum in a similar manner to escape electrons. This effect is increased if the electron has high energy, or for high-Z materials. (Knoll, 2000)

If a radioisotope decays through beta-minus decay, the electrons may interact with shielding around the detector. As this shielding is intended to protect against wayward $\gamma$-rays it will often be of high-Z materials with appreciable cross-sections to stop $\gamma$-rays (such as lead). Beta particle interaction with the shielding can produce Bremsstrahlung radiation that can complicate the spectrum at lower energies. (Knoll, 2000)

If beta-plus decay occurs, the emitted positron is likely to interact with any casing around the detector or the radioisotope, producing two 511 keV gamma-rays through pair production. The created $\gamma$-rays will travel in opposite directions, so for most detector geometries, only one of the $\gamma$-rays will travel towards the detector. This $\gamma$-ray may be picked up by the detector, producing a single peak at 511 keV, shown in fig. 2.1. (Knoll, 2000)

### 2.2 Neutron Interactions

Neutrons, due to their uncharged nature, are more difficult to detect than charged particles and will often move through several centimeters of material with no effect before an interaction occurs. These interactions are with the nucleus of the absorbing material and are primarily either a scattering of the neutron off of a nucleus or a neutron-induced nuclear reaction. The cross-section, or probability per unit length, of different types of interactions is strongly a function of the incident neutron’s energy. Subsequently neutrons are divided into "slow neutrons" and "fast neutrons" based on their kinetic energy.
2.2.1 Slow Neutrons

Slow neutrons are those with an energy less than 0.5 eV. Their most probable way to react is via elastic scattering events with the absorbing material’s nucleus. Because their kinetic energy is so small, these scattering events serve to "thermalize" the neutron, or bring it to thermal equilibrium with the absorber. They also generally fail to produce enough energy in the recoil nucleus to be detectable. Despite the prevalence, low-energy elastic scattering is not an effective way to measure slow neutrons.

Slow neutrons do have a reasonable cross-section for neutron-induced nuclear reaction. These reactions create heavy charged particles in the detector, which are generally detectable. There are several different types of reactions that can occur, such as (n,p), (n,α), (n,fission), and (n,γ), the last of which is not desirable due to γ-rays not being directly detectable. Slow neutrons have not been investigated in this work.

2.2.2 Fast Neutrons

Fast neutrons are those with an energy greater than 0.5 eV. The cross-section of neutron-induced nuclear reactions goes down significantly with increasing neutron energy. Also at higher energies, the amount of energy imparted to a nucleus during elastic scattering becomes significant. Most detectors of fast neutrons rely on recoil nuclei from elastic scattering.

During each scattering event, the neutron imparts some of its energy, "moderating" the neutron. The most effective moderator is hydrogen, as a neutron can impart nearly all of its energy in a single scattering event. The moderating effect of an absorber decreases with increasing size of the absorber nucleus. The best detectors of fast neutrons have high densities of hydrogen. (Knoll, 2000)
2.2.3 Scintillation with Fast Neutrons

While several methods exist to detect fast neutrons, the detection of recoil nuclei via scintillation is one of the most commonly employed. Recoil nuclei, or recoil protons if the nuclei are \( ^1 \text{H} \) nuclei, create excited states in much the way other charged particles like secondary electrons do. The details of these excited states is discussed further in section 2.4.

Polymers derived from hydrocarbons primarily interact with neutrons through their hydrogen atoms. The recoil protons can potentially absorb the entire energy of the incident neutron if the collision is head-on. For non-relativistic neutrons (neutrons with energy \( E_n << 939 \text{ MeV} \) undergoing elastic collisions, conservation of energy and momentum can be applied to the collision to get the recoil energy, \( E_R \), as shown in eq. 2.4

\[
E_R = \frac{4A}{(1 + A)^2} \cos^2 \theta E_n
\]  

where \( A \) is the atomic mass of the absorbing material, \( \theta \) is the scattering angle of the recoil nucleus in the lab frame, and \( E_n \) is the energy of the incident neutron. The maximum energy is imparted to the recoil nuclei when the scattering angle is 180°, during head on collisions, giving:

\[
E_R = \frac{4A}{(1 + A)^2} E_n
\]  

We can see from eq. 2.5 that the \( E_R \) decreases with increasing atomic mass of the absorber, shown in table 2.1, and that for \( A = 1 \) we get the simple result \( E_R = E_n \). It is for this reason that light nuclei and in particular hydrogen are of the most interest in recoil scintillators.

Further investigations into the probability of scattering in a particular direction and the distribution of recoil energy shows that the distribution of the energy is the same shape as the cross-section of the recoil nuclei, which is itself a function of \( \Theta \), the scattering angle in the centre of mass reference frame. This result tells us that for most nuclei, the energy distribution of the recoil nuclei will be distributed somewhat like a plateau from 0 to \( E_R \) but will peak slightly at either end, corresponding to a higher probability of head-on collisions
Table 2.1 Maximum fractional energy that can be imparted by a neutron to a nucleus of a given A value.

<table>
<thead>
<tr>
<th>Absorber Nucleus</th>
<th>A</th>
<th>$\frac{E_R}{E_0}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^1$H</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>$^2$H</td>
<td>2</td>
<td>$\frac{8}{9} = 0.889$</td>
</tr>
<tr>
<td>$^3$He</td>
<td>3</td>
<td>$\frac{3}{4} = 0.750$</td>
</tr>
<tr>
<td>$^4$He</td>
<td>4</td>
<td>$\frac{16}{25} = 0.640$</td>
</tr>
<tr>
<td>$^{12}$C</td>
<td>12</td>
<td>$\frac{48}{169} = 0.284$</td>
</tr>
<tr>
<td>$^{16}$O</td>
<td>12</td>
<td>$\frac{64}{289} = 0.221$</td>
</tr>
</tbody>
</table>

and glancing blows. The exception to this is hydrogen, for which the cross-section is isotropic in the center-of-mas reference frame. For Hydrogen, the distribution of recoil energy will be a rectangle from 0 to $E_R$ if the incident neutrons are of a singular energy, as it is not uncommon for them to be (Knoll, 2000)

2.2.4 Spectrum Complications

In most scintillators, and in particular plastics, contributions from other light elements can be a major source of deviation in the spectrum distribution. In particular, the energy distribution of recoil $^{12}$C nuclei, an isotope abundant in hydrocarbon-based polymers, is complicated. The presence of nitrogen, oxygen, or deuterium can further complicate the energy spectrum, making it hard to predict what the energy spectrum should look like.

Another factor complicating the energy spectrum from fast neutrons is size of the detector. Large scintillators will interact with more neutrons, increasing detector efficiency, but will suffer from less uniform light collection, decreasing resolution. Additionally it is possible that neutrons that scatter off of hydrogen and do not deposit all of their energy may scatter again, sometimes several more times. Because the several scattering events will happen over time scales much shorter than the time is takes for light collection to occur,
all of this energy will be gathered in a single pulse and will weight the response function towards higher energies. For small detectors, the recoil nuclei may escape from the detector before it has deposited all of its energy, leading to an increased weighting at lower energies.

Many organic scintillators also exhibit noticeable degrees of non-linearity in light output with energy. Even when a detector shows acceptable linearity for electrons, there is often still significant non-linearity for larger charged particles. (Knoll, 2000)

If the effects of complications are small, the energy of the incoming neutrons can be determined by taking a derivative of the energy distribution. The derivative of the flat portion of the distribution will be zero, but the function will spike at the end of the distribution as it reaches the cut-off. This peak occurs at $E_n$ and can be easily distinguished. If the effects of complications are not small, more advanced deconvolution techniques are required. (Knoll, 2000)

### 2.3 Inorganic Scintillators

The scintillation mechanism in inorganic crystals is dependent on the energy band structure determined by the crystal lattice. Electrons in inorganic crystals have discrete energy bands which they can occupy. In insulators and semiconductors, these electrons normally lie in the *valence band* of the material, but upon the absorption of energy can excite into the higher energy *conduction band*, leaving behind a positively charged hole. Between them lies the *forbidden band*, which is inaccessible to the electron in the pure crystal. Impurities within the crystal lattice can create excited states within the forbidden band that an excited electron can now be found in. The intentional introduction of impurities, called *activators* is known as *doping*.

The relaxation of the electron from the conduction band back into the valence band through the emission of a photon is an inefficient process, and for most materials results in too high of an energy to be detected by a standard photomultiplier tube. Relaxation from
the excited states created by impurities is a more efficient process that gives rise to photons in the visible spectrum, which can more closely match the absorption spectrum of common PMTs. The excited states created by impurities are called recombination centers. (Knoll, 2000) (Birks, 1964b)

2.3.1 Excitation from Ionizing Radiation

When a charged particle, (such as a free electron created by the absorption of a $\gamma$-ray) passes through the detector, it will generate many electron-hole pairs. A hole will quickly ionize an activation site as it will be at a lower energy for the hole. Excited electrons will be free to move throughout the detector until they find an ionized activator, where they will fall into a lower energy state. The activator site has its own structure of excited states that the electron will fall into. The relaxation from these excited states to the ground state of the activator is what gives rise to the scintillation mechanism, as this process has a high probability for the emission of a photon. These transitions often have have lifetimes of 50 - 500 ns (Knoll, 2000) that are responsible for the time characteristics of the scintillator, as the movement of the electron to the activator happens over much shorter times. (Birks, 1964b)

2.3.2 Phosphorescence and Quenching

Two other processes can occur other than the quick process described above. Some transitions from the excited state to the ground state of the detector are forbidden. These states require the introduction of a small amount of additional energy to excite to a state without a forbidden transition to the ground state. This additional energy often comes from thermal excitation, and can be a very slow process. Called phosphorescence, this can be a source of 'afterglow' in the scintillator.

Some excited states can transition to the ground state through radiationless processes. These processes are highly undesirable and are called quenching. Good scintillating detectors
have minimal quenching so as to maximize light output, defined as the energy emitted as light per incident radiation. (Knoll, 2000)

2.4 Organic Scintillation

2.4.1 Excitation and Emission

The scintillation mechanism in plastic scintillators differs from inorganic scintillators in several important ways. It begins in the same way, with an ionized electron being created by an incident radiation (such as a $\gamma$-ray through Compton Scattering). As this free electron travels through the scintillating material and interacts with the surrounding electrons, it excites them out of the $S_0$ state (the ground state) and into a higher energy singlet state. The excited state can be the first state, $S_1$, but is generally a higher energy state. The $S_2$ and higher states decay into the $S_1$ state with very short decay times, primarily through radiationless internal conversion. Once in the $S_1$ state, the electron can either decay back to the ground state through the prompt emission of a photon or instead transition into a triplet state, $T_1$.

Transitions from $S_1$ to $S_0$ are slower than transitions from the $S_2$ and higher states to $S_1$, but still relatively quick when compared to other decay times and are on the order of nanoseconds. Emission of a photon from the decay of the $S_1$ state is known as **prompt fluorescence**. (Knoll, 2000)

Alternatively, the $S_1$ state can transition into a triplet state, $T_1$, through a process called intersystem crossing. There is no $T_0$ for the state to decay to and transitions from the $T_1$ state to the $S_0$ state are ‘forbidden’ transitions. The $T_1$ state can decay to the $S_0$ state anyway, albeit with a much larger decay time. The photon emitted by this process, known as **phosphorescence**, is of slightly less energy than that emitted by fluorescence. Decay times range from 10s to 100s of nanoseconds. (Knoll, 2000) (Birks, 1964a)
The T$_1$ state will sometimes excite back to the S$_1$ state, where it will decay with the emission of a photon. Transitions back to the S$_1$ state require a small increase in energy, so these transitions have a relatively large characteristic time, similar to those of phosphorescence. This process is called delayed fluorescence. (Knoll, 2000)

![Schematic Diagram showing the excitation of the singlet and triplet states and the relaxation through fluorescence and phosphorescence. (Birks, 1964a)](image)

**Figure 2.2** Schematic Diagram showing the excitation of the singlet and triplet states and the relaxation through fluorescence and phosphorescence. (Birks, 1964a)

Each of the electronic excited states are further subdivided into smaller vibrational states. These vibrational states are often denoted by a second subscript. At room temperature, these vibrational states quickly decay into their ground vibrational states via thermal processes. Decays from the S$_{10}$ and T$_{10}$ state rarely decay into the the S$_{00}$ ground state, instead decaying into one of the excited vibrational modes of the ground electronic state. These states, S$_{01}$, S$_{02}$, etc. quickly de-excite to the S$_{00}$ state. Transitions to the vibrational modes emit light that is of a smaller energy than required for the S$_{00} \rightarrow$ S$_{10}$, so reabsorption is minimal. This is the source of the high light-output of organic scintillators. A diagram of the relaxation of the singlet and triplet states is shown in fig. 2.2.

The overall characteristic decay times of organic scintillators is usually on the order of a
few nanoseconds, though for some materials it can be as low as tenths of nanoseconds and as
great as tens of nanoseconds. Some organic scintillators for specific applications have decay
times on the order of microseconds. (Knoll, 2000) (Birks, 1964a)

2.4.2 Liquid Scintillators

Pure organic crystals for scintillation are not often used, the only 2 with common use being
Anthracene and Stilbene. Organic crystals are fragile and exhibit anisotropy in their scintilla-
tion efficiency. Other organic scintillators, such as liquid scintillators, find much more
widespread use.

Liquid scintillators consist of an organic scintillator that has been dissolved in an ap-
propriate solvent. These can be the only two compounds present, but sometimes a third is
added that acts as a wavelength shifter, to better match the emitted light with the response
spectrum of common photomultiplier tubes. In some uses, a radioactive material may also
be dissolved in the solution. This provides for a nearly 100% count rate efficiency for the
dissolved radioisotope and can be a very useful for detecting low-activity materials. Another
benefit of liquid scintillators is their lack of a crystalline structure that can be damaged by
intense radiation. This makes them highly resistant to radiation damage and its effects (such
as decreased light emission). (Knoll, 2000)

Liquid scintillators often come in a sealed glass container that has had most of the
oxygen removed. Dissolved oxygen can act as a strong quenching agent, where quenching
is the absorption of light through non-radiative processes. Care must be take to ensure
optical coupling between the glass of the scintillator and that of the photomultiplier tube.
Additionally even very small leaks that allow the transfer of gas can ruin the scintillator.
Many liquid scintillators or their solvents are toxic or environmental hazards. The risk of
spills can make them a hazard in the lab and limits their application outside of the lab.
(Knoll, 2000)
2.4.3 Plastic Scintillators

Plastic scintillators are made when an organic scintillator is dissolved in a solvent that can be polymerized. The polymerization of the solvent leads to a solid solution being formed with the polymer and the scintillating material. Common plastics for scintillation are polystyrene (PS), polyvinyltoluene (PVT), and polymethylmethacrylate (PMMA).

Plastic scintillators have all of the benefits of organic scintillators, but are cheaper and easier to produce. They can be manufactured in a wide range of shapes and sizes allowing for application in many fields. They are easy to handle, and in applications that require large detectors or those with complex geometries, such as in heavy-ion reactions involving $4\pi$ detectors like the CHIMERA detector array (A. Pagano et al., 2004), plastic scintillators are the only cost effective solution. Similar to liquid scintillators, they have little to no anisotropy in the scintillation efficiency. Plastic scintillators vary in their resistance to radiation damage, with significant degradation occurring for exposures as little as $10^2\text{ Gy}$ in some plastics, but no degradation being observed for exposure as large as $10^5\text{ Gy}$ is others. (Knoll, 2000)
Chapter Three

Scintillation Measurement

The general process of detecting an ionizing particle begins with the interaction of the particle with a detector material. In the scintillation process, this interaction produces light that is turned into an electrical signal via a photomultiplier tube. A histogram is often generated from a collection of signals. There are several features of the waveform that they can be binned by, but the most common is the pulse height. Newer technology has allowed for easy analysis of individual waveforms as well.

3.1 Photomultiplier Tubes

When light is emitted by a scintillator, it is first detected by a photomultiplier tube. The light is impinges on a photocathode, a thin metal with a work function below the energy of the incident photons of interest. Each photon with energy above the work function imparts the electron with its energy in excess of the work function as kinetic energy. The electrons escape the surface of the photocathode and interact with a chain of dynodes, gaining energy and losing some of that energy to the production of additional electrons. Those electrons are eventually absorbed by the anode of the photomultiplier tube and generate an electrical signal that can be detected via conventional means.

In practice, the conversion of light energy to electrical energy is not 100% efficient. The...
electrons lose some of their kinetic energy to electron-electron interactions while moving to
the surface of the photocathode. Additionally not every photon may have enough energy
to overcome the work function of the material, or to impart enough energy to escape the
photocathode, leading to no production of an electron. The thinness of the photocathode
leads some photons to pass through unabsorbed, further reducing efficiency. The measure of
how many electrons are produced per incident photon is known as the quantum efficiency of
the photomultiplier tube. The quantum efficiency is dependent on the incident wavelength,
but average quantum efficiencies are often quoted as 25%. For maximum light collection
efficiency, the choice of a PMT for a given application is one whose absorption spectrum
closely matches the emission spectrum of the scintillating material.

3.2 Energy Spectrum

3.2.1 Detector Calibration

The exact details of every interaction between a photon and a doped, scintillating crystal is
dependent on many factors, such as the number and location of lattice defects and dopants,
differences in shape, and temperature. These factors can change the response function of the
scintillator. Additionally, each PMT (photomultiplier tube) a scintillator is coupled to may
have a different absorption spectrum. These have the net effect of causing every scintillating
detector to require its own calibration. The calibration characterizes the relationship between
the bins pulses can be sorted into (channels) and the energy corresponding to each channel.

This relationship is highly linear for inorganic scintillators but is less so for other types. To
calibrate a detector, radiation of known energy is collected by the detector and a histogram
is generated. The channel number corresponding to the peak energy can be extracted (see
below) and plotted against the energies. A straight line fit through these points has a
slope that gives a multiplicative conversion factor between channel number and energy for
the detector. Detectors that display significant non-linearity may require more complicated curve-fitting for calibration.

3.2.2 Curve-fitting to Peaks

Radioisotopes whose decays products include γ-rays often emit γ-rays of well defined energies (e.g. Na-22 emits of 2 γ-rays of energy 0.511 MeV and 1.274 MeV). During the scintillation process, there are many random mechanisms in which the amount of light emitted from a monoenergetic source deviates from a single value. For example, in inorganic crystal scintillators this can be due to lattice defects, thermal processes, and which shell an electron is ionized from. Additional uncertainties arise from the fact that each photon interaction with the detector can only give rise to a discrete number of excited states. Similarly any light emitted from the decay of these excited states can only create a discrete number of free electrons in the connected photomultiplier tube.

The energy spectrum of the SrI₂ detector for the 662 KeV γ-ray showing the photopeak is shown in fig. 3.1. The distribution of the photopeak is assumed to be a Gaussian. This Gaussian distribution will have its centroid, \( x_0 \), located at the energy of the incident γ-ray with a standard deviation \( \sigma \). The amplitude, \( A \), gives the number of counts above the background and the offset, \( y_0 \), gives the background number of counts. An example fit is given in fig. 3.1.

\[
f(x) = Ae^{-\frac{(x-x_0)^2}{2\sigma^2}} + y_0
\]

3.2.3 Energy Resolution

The energy resolution, \( R \), can be calculated using eq. 3.2,

\[
R = \frac{FWHM}{E} \times 100\%
\]
Figure 3.1 A sample fit for the 662 keV photopeak from $^{137}$Cs, collected with SrI$_2$:Eu during our experiments. This fit is used to extract the centroid and standard deviation of the photopeak. The standard deviation is taken to be the uncertainty in the centroid location. It is also used to calculate the full width at half maximum, FWHM, of the peak. For the sample fit, $FWHM = (44.8 \pm 0.4)$ keV.

$FWHM$ can be found from the standard deviation by

$$FWHM = 2\sqrt{2\ln 2} \sigma$$

The energy resolution characterizes the ability of the detector to resolve fine features, and is important in determining whether a detector is suitable for a given application. Because the energy resolution is a unitless ratio, it can also be calculated using the channel number of the peak and the $FWHM$ in channel number for a linear detector.

The uncertainty in the energy resolution was determined using the quadrature method, given generally for a function $f(x, y)$ by eq. 3.3. The uncertainty in $f$, $\delta f$, is dependent on
the uncertainty in $x$ and $y$, $\delta x$ and $\delta y$.

$$\delta f = \sqrt{(\frac{\partial f}{\partial x})^2(\delta x)^2 + (\frac{\partial f}{\partial y})^2(\delta y)^2} \quad (3.3)$$

The uncertainty in the energy resolution, $\delta R$, is given by eq. 3.4:

$$\delta R = \sqrt{\left(-\frac{\text{FWHM}}{E_0}\right)^2(\delta E_0)^2 + \left(\frac{1}{E_0}\right)^2(\delta F)^2 \times 100\%} \quad (3.4)$$

where $E_0$ is the centroid, $\delta E_0$ is the uncertainty in the centroid, and $\delta F$ is the uncertainty in the FWHM. The uncertainty in the variables was taken to be the standard deviation of the variables given by the fit of the Gaussian distribution to the photopeaks.

### 3.3 Neutron-Gamma Discrimination

When radiation interacts with the detector (an event), the signal produced is often referred to as a waveform. For scintillating materials, this waveform is characterized by a sharp peak followed by a much longer decay. The initial sharp peak occurs because the radiation creates many excited states over a very short time period ($\sim 0.1$ ns) and many of these states immediately begin to decay. The longer decay of the waveform occurs because the number of excited states decaying at any given moment decreases as the number of excited states decreases.

Two features of the waveform are directly proportional to the energy of the incident radiation for linear detectors. The first is the total integral of the waveform. The second is the peak signal from the waveform, called the pulse height. While both of these features are proportional to the energy, they will have independent constants of proportionality. The overall shape of the waveform is constant for a given radiation and detector setup. Finding the constants of proportionality for these methods is the purpose of calibrating the detector, as described in section 3.2.1.

One thing many scintillating detectors are unable to do is discriminate between different types of radiation. While the constant of proportionality may vary for different particles
that have the same initial energies, the shape of their waveform may be the same. When it is different, Pulse Shape Discrimination techniques (PSD) may be used to analyze individual waveforms and determine the identity of incident radiation.

A particularly useful capability of organic scintillators is their use in neutron-gamma discrimination. This is possible because the 2 different states that can be populated by radiation - the singlet and the triplet states - have different characteristic decay times. For a given amount of energy deposited by an event, the number of populated states will be approximately the same. Because the triplet states are much longer lived, events that populate a greater fraction of triplet states over singlet states will have lower pulse heights and longer tails. Similarly, a state that has a greater fraction of singlet states will have a larger pulse height and shorter tail. An example is shown in fig. 3.2 where the neutron has a larger fraction of its energy stored in the tail due to populating a greater fraction of triplet states.

Comparisons of different aspects of the waveform, such as the ratio of the light contained in the tail region to the total light output of the waveform will yield differences in the results for a given type of radiation due to the different ratios of singlet and triplet states excited by the radiation. These comparisons are often referred to as Particle Identifications. PID can be done when different types of radiation populate different ratios of singlet and triplet states, leading to different values for particle identification (PIDs).

The division between the prompt and tail regions of the waveform is not well defined. The decay of some excited states begins while the population of the states is ongoing, and as such the tail region starts were the influence of the newly populating states becomes sufficiently small relative to the number decaying states, where sufficiently is not well defined. The determination of where the tail region begins could constitute its own investigation.

Here we chose to find an approximate location for the tail region instead of doing an extended review. We determined the tail region by examining several waveforms of varying amplitude on a semi-logarithmic scale and marking where the waveform becomes straight on
Figure 3.2 An example of the differences in signal from NE213 due to γ-rays and neutrons. Note how the waveform produced by the neutron has a longer decay time. This is due to a higher proportion of triplet states populated by neutrons.
the semi-log graph. This denotes the region where the exponential decay of the tail region dominates. The prompt-tail boundary was found to be approximately 100 channels after the rise time. An example of the described method is given in fig. 3.3.

![Example of Prompt-Tail Approximation for NE213]

**Figure 3.3** An example of the method used to determine an approximate boundary between the prompt and tail regions of the waveform. The boundary shown marks approximately where the tail begins, and makes sure not to include any portion of the prompt region. Small portions of the tail region may be marked as prompt, however in these sections the differences between waveforms are small.

### 3.3.1 Particle ID and Figure of Merit

The process of capturing a waveform adds many types of error into the determination of a PID. Sources of random variation during light capture, from electrical signal noise and during the digitization process, cause the PID to have a distribution about some mean value. This distribution is approximately a normal distribution whose average value is the actual PID of the radiation. This PID is dependent upon the detector and is not suitable for direct
The spread of the distribution is often a function of energy, and may be wide at low energies, but narrow significantly at higher energies. When more than one type of radiation is present, the distributions may overlap for certain energies. At very low energies, this overlap may make the individual distributions indistinguishable. One must be careful when assigning a particle as one type or the other, as different particles may have the same PID at these low energies.

It is useful to discuss the degree to which two PIDs are separated. It is only when the particles are separated to an appreciable degree that discrimination can occur with an acceptably low number of misidentifications. The quality of a separation is given by the figure of merit, or FOM. The figure of merit is calculated using eqn. 3.5

\[
FOM = \frac{|x_0 - x'_0|}{FWHM + FWHM'}
\]  

(3.5)

where \(x_0\) and \(x'_0\) are the centroids of the two distributions (the average PIDs) and \(FWHM\) and \(FWHM'\) are the respective full widths at half-maximum. Large differences in the average PID value of the peaks and small FWHMs correspond to large values of the figure of merit. Thus larger figures of merit correspond to better separation of the distributions and better discrimination of the detected radiation. For NE213, a minimum figure of merit is about 0.75, while for EJ-299-33A a minimum of about 0.80 is required for separation.

This definition of the figure of merit has advantages in that it is mostly independent from the relative intensities of the two peaks. It is mostly independent because the intensities need to be close enough that one peak does not obscure the other. In the event that one peak obscures the other, fitting a Gaussian distribution will not be possible and the figure of merit will not be calculable until there is enough of a separation for both peaks to be visible. As long as no peaks are obscured, the figure of merit will be calculable.
Chapter Four

Experimental Details - Strontium Iodide

The goal of the experiments described here was to characterize the response function of a recently rediscovered, inorganic scintillator SrI$_2$:Eu. In particular, its energy resolution for several different radioisotopes was measured and compared to two other inorganic scintillators, NaI:Tl and LaBr$_3$:Ce. For all three detectors, a gamma-ray energy spectrum was acquired for the following sources: $^{137}$Cs, $^{22}$Na, $^{60}$Co, and $^{54}$Mn. These sources were chosen as they span a range of common $\gamma$-ray energies. Additionally they are byproducts of the creation of nuclear weapons from uranium and of nuclear power, making them of particular interest in radiation safety and security. The experiment was performed at the Nuclear Chemistry Laboratory, University of Rochester.

4.1 Description

The experimental setup was designed for accurate detection of $\gamma$-rays from the radioisotope while minimizing the effects of cosmic and background radiation and deterring pileup (the detection of multiple events in overlapping time periods). As shown in fig. 4.1, the $\gamma$-ray source was placed approximately 34 cm from the 2.54 cm (1") diameter strontium iodide detector. Due to the relatively small size of the SrI$_2$:Eu detector, care was taken to ensure the source was properly aligned with the detector. SrI$_2$:Eu was optically coupled to an
XP2041 photomultiplier tube (PMT) operated at -1750 volts.

**Figure 4.1** Schematic layout of the experiment. The detector and PMT were encased in lead housing with only a single cylindrical opening to allow gamma rays to enter. The source was carefully aligned with the center of the detector. The entire setup was draped with a dark fabric and experiments were done in a darkened room.

The LaBr$_3$:Ce detector used was a 5.08 cm (2") diameter, commercial, integrated detector produced by Saint-Gobain Crystals. The in-built PMT was of design R6231. It was placed approximately 34 cm from the source and was operated at 600 volts. Both of these two detectors were set up in a 5 cm (2") thick lead housing to minimize background radiation during measurement. An aperture larger than the source and the detector face was open to the source.

The NaI:Tl detector used was a commercial 7.72 cm (3") diameter detector, also of integrated design. Due to the size and fixed location of this detector, the NaI:Tl detector was not encased in lead housing. The source was placed approximately 30 cm away from the detector and it was operated at 900 volts.
4.1.1 Analog-to-digital Conversion

For all detectors, the output was connected to a FemtoDAQ Digital Analyzer by SkuTech Instruments, shown in fig. 4.2. The FemtoDAQ is fashioned with 2 analog inputs each of which is capable of 14 bit analog to digital conversion operating at 100 Ms/s. Data was collected until enough events were captured for ease of analysis, on the order of $10^3$ counts for the highest-count peaks. The amount of time that collection occurred for (the collection time) varied between detectors but was held constant across isotope sources for a given detector.

![FemtoDAQ Analog-to-Digital Converter](image)

**Figure 4.2** FemtoDAQ Analog-to-Digital Converter. All data was converted to digital and waveform analysis was done off line. Shown are the front face of the FemtoDAQ (top) and the back face (bottom).
Chapter Five

Results for Strontium Iodide

In this chapter the combined energy spectra for each inorganic scintillating detector is presented, along with a table of the energy resolutions for different energies and detectors. The measured energy resolutions are compared and discussed.

5.1 Energy Spectra

Experimental results are illustrated in figs. 5.1 - 5.3. In fig. 5.1, energy spectra for various radioisotopes measured with SrI$_2$ are presented, along with the background measurement. The full energy peaks corresponding to the $\gamma$-ray energies are marked. As clear from the figure, SrI$_2$ does a good job identifying $\gamma$ rays.

In fig. 5.2, energy spectra for the same radioisotopes as measured by NaI are presented. As expected, NaI also does a good job identifying the characteristic $\gamma$-ray energies for the various radioisotopes.

Fig. 5.3 presents the energy spectra for the same radioisotopes as measured by LaBr$_3$·Ce. The characteristic $\gamma$-rays are clearly identifiable. Note the complex internal structure, which could obscure peaks from weaker sources. Of particular note in this background is the broad peak that coincides with the $^{54}$Mn photopeak.

The measured energy resolution is presented in table 5.1 and calculated using eq. 3.2.
Figure 5.1 The combined spectrum of a SrI$_2$:Eu detector for several different radioisotopes. Selected full energy peaks are labeled. Compton Edges associated with these photopeaks are also more noticeably present compared to NaI:Tl or LaBr$_3$:Ce. Note the lack of any internal radiation and low background.
Figure 5.2 The combined spectrum of a NaI:Tl detector for several different gamma ray sources. Selected full energy peaks are labeled. The response from $^{54}$Mn (835 keV) was difficult to distinguish from the background and was not included in the energy resolution comparison. The small peak at high energies is background radiation – likely potassium that is present due to the lack of lead shielding around the NaI:Tl detector.
Figure 5.3 The combined spectrum of a LaBr$_3$:Ce detector for several different gamma ray sources. Full energy peaks (labeled) are easily distinguishable from the background. Note the 1436 keV peak due to internal radiation present in all spectra.

Table 5.1 Measured resolution for several $\gamma$-ray energies are compared across 3 different inorganic crystal scintillators. Energy resolutions for Strontium Iodide (SrI$_2$:Eu) are comparable to more common detectors. The bottom row includes the energy resolution of the internal radiation in LaBr$_3$:Ce from $^{138}$La.
5.2 Conclusions

We measured the $\gamma$-ray energy spectrum for SrI$_2$:Eu$^{3\%}$ using Cs$^{137}$, Na$^{22}$, Co$^{60}$, and Mn$^{54}$. The quality of energy spectra for SrI$_2$:Eu$^{3\%}$ is compared to that of LaBr$_3$:Ce (Brilliance) and NaI:Tl. The energy resolution shows that SrI$_2$:Eu has a slightly superior energy resolution compared to the NaI detector. SrI shows a slightly lower resolution compared to LaBr$_3$:Ce, but measures a simpler background spectra. With tapering, SrI$_2$:Eu is expected to produce even more remarkable energy resolution. Further improvements to the energy resolution are expected with slightly higher doping levels. The excellent energy resolution with a lack of internal radiation found for SrI$_2$:Eu makes it a detector of choice for hand-held radiation detection monitors.
Chapter Six

Experimental Details - Organic Scintillators

The experiments described here were performed to characterize the quality of n/γ discrimination by a recently developed plastic scintillator EJ-299-33A of two different lengths. In particular, a figure of merit was calculated for the separation of neutrons and γ-rays from an americium-beryllium (AmBe) n/γ source. This was done for two EJ-299-33A scintillators, cylindrical in shape, and of sizes 5.08 × 2.54 cm and 5.08 × 5.08 cm (2" × 1" and 2" × 2" respectively) in size, where the first parameter indicates the diameter of the cylinder and the second parameter indicates the length of the cylinder. The analysis was also done for a 7.62 × 5.08 cm (3" × 2") NE213 liquid scintillator. The figure of merit was compared for all 3 detectors.

For all three detectors, a γ-ray energy spectrum was acquired for the following sources: $^{137}$Cs, $^{22}$Na, and $^{54}$Mn. All experiments were performed at the same University of Rochester.

6.1 Description

The setup for the experiments with organic scintillators was very similar to that for the experiments with inorganic scintillators. A schematic layout is shown in fig. 4.1. The
source was placed approximately 34 cm from the scintillator. Care was taken to ensure the source was properly aligned with the detector. NE213 was optically coupled to an XP2041 photomultiplier tube (PMT) operated at -1750 volts. Both EJ-299-33A scintillators were attached to a Hamamatsu R7724 PMT operated at -1600 Volts. There are two major differences between the setup for the inorganic scintillator experiments and those done here:

1. Both $\gamma$ and n/$\gamma$ sources were used

2. When the n/$\gamma$ source was used, a 5.04 cm thick lead brick was used to shield some of the $\gamma$-rays

Energy spectra were collected through histogramming for both $\gamma$-ray only sources and for n/$\gamma$ sources. Additionally over 20,000 individual waveforms were recorded for both source types.

The output of the detector was connected to a FemotDAQ Digital Analyzer made by SkuTech Instruments (http://skutek.com/). The FemtoDAQ is fashioned with 2 analog inputs each of which is capable of 14 bit analog to digital conversion operating at 100 Ms/s. For the energy spectra, data was collected until enough events were captured for ease of analysis, on the order of $10^3$ counts for the highest-count peaks. The data collection time varied between detectors but was held constant across isotopes for a given detector. Waveform collection duration was based on the number events recorded. The FemtoDAQ is show in fig. 4.2.

6.2 Numerical Analysis

The data from the energy spectra was stored in text files and analyzed in Python. The spectra were analyzed to calibrate the detector using the methods described in section 3.2.

Calibration of the organic scintillators was done by fitting half-Gaussian to the Compton edge produced in a 1D histogram from the $\gamma$-rays from 3 sources: $^{137}$Cs, $^{54}$Mn, and $^{22}$Na.
allowing for 4 data points to do the fit. The channel number corresponding to the Compton energy was extracted from the Gaussian fits and plotted against the theoretical Compton energy values. A linear fit was used to produce a calibration factor.

In evaluating each Compton edge, one needs to determine the position of the actual maximum electron recoil energy from the distribution, however a consensus is lacking on the exact position for a given scintillator and in the best method to determine it. E. Pagano et al. (2018) suggests that the feature of the Gaussian that corresponds to the maximum electron recoil energy, the Compton energy, depends on the detector and the electronics used. They propose a method based on other research of finding the locations which minimized the variance in the subsequent linear fit. This approach was used here, but found that for NE213 and for the 2″ × 2″ EJ-299, the Compton Energy corresponded to the peak of the Gaussian fit. For the 2″ × 2″ EJ-299, the variance was not able to be minimized. We believe these results to be unrealistic. Due to several authors showing similar results for these two scintillators despite the differences in electronics, the locations used by them were used here as well. For NE213, the Compton Energy was taken to be at 89% of the peak. For both EJ-299-33As, the Compton Energy was taken to be at 50% of the peak. These numbers align with those found by Pozzi, Bourne, and Clarke (2013), Zaitseva et al. (2012), and E. Pagano et al. (2018).

For calibrating the 2D histograms, the scale factor of the FemtoDAQ had to be taken into account as well. When measuring individual waveforms, the FemtoDAQ digitizes the incoming voltage into 8192 channels (spanning a range of 0 to 1 V). When histogramming, the FemtoDAQ compresses this data by a factor of either 2 or 4 into 4096 and 2098 channels respectively. The calibration discussed above only calibrates from the scaled channel number into energy. To calibrate the raw channel number into energy, the scale factor must be accounted for. Both NE213 and the 2″ × 2″ EJ-299 were scaled by a factor of 2. The 2″ × 1″ EJ-299 was scaled by a factor of 4.

This data was fit using a linear fit and scipy.optimize.curve_fit, which utilizes
**Figure 6.1** Calibration plots for all 3 Organic Scintillators. While not as ideal as NE213, the linearity of EJ-299-33A is acceptable for the work done here.
a least squares method to fit a given function to a 1D set of data. An sufficient degree of linearity is observed, consistent with the findings of others that NE213 exhibits good linearity between 0.1 MeV and 1MeV and that EJ-299-33A shows a fair degree of linearity over the range of interest. (Scherzinger et al., 2016) (Flynn et al., 1964) (Nyibule et al., 2013) (E. Pagano et al., 2018)

### 6.2.1 Waveform Analysis

To analyze the waveform data, each wave was integrated using the trapezoidal method via `scipy.integrate.trapz`. Several regions of the wave were integrated, with integrals denoted as "Q":

- The entirety of the wave form, \( Q_{\text{total}} \)
- The initial "fast" region of the waveform, \( Q_{\text{fast}} \)
- The decaying "tail" region, \( Q_{\text{tail}} \)

Additionally the Particle Identification (PID) was calculated using eq. 6.1. The pulse height was also found, and was taken to be the maximum voltage of the waveform.

\[
\text{PID} = \frac{Q_{\text{tail}}}{Q_{\text{total}}} \tag{6.1}
\]

The "trigger point," where the FemtoDAQ recognizes an event is occurring and begins recording data and is set a specific number of data points in, was used as the start of the waveform. An average "tail location", which denoted the boundary between the fast and the slow regions, was chosen by plotting the wave on a logarithmic scale and visually deducing an approximate location where the waveform appeared to turn over for several waves. This resulted in a tail location a few samples after the peak.
6.2.2 Identifying Particles

While a histogram may show that there are two particles with different PID, it is necessary to determine which PID is associated with which particle. One way to do this is to attenuate different types of particles and see how the histogram changes. A low-Z material high in hydrogen content, such as paraffin wax, absorbs many neutrons but few $\gamma$-rays. Comparison between histograms with and without a neutron absorber show that one peak will have far fewer counts relative to the other when shielding is in place. This would imply that the weaker peak would correspond to neutrons. The same can be done with a high-Z material, such as lead, which will attenuate the $\gamma$-ray signal but not the neutrons. This allows the assignment of particular PID to a type of particle. In agreement with theory, the larger PID corresponds to neutrons and smaller PID corresponds to $\gamma$-rays for all experiments done here.

6.2.3 2D Histogram of Waveform Integrals

The calculated information that was extracted from the aggregate waveform data was plotted as a 2D histogram with several different metrics on the axes. The number of bins was 160 along the y-axis and 234 along the x-axis. This provided a good resolution for the histogram while keeping the number of counts in a given bin large enough that reasonable 1D statistics could be applied in the analysis described below. $160 \times 234$ also closely matches the pixel ratio of the graph produced by pyplot, giving a clean, square appearance to the bins. An example is given in figure 7.7 on p. 52, where PID is plotted on the y-axis and Pulse Height is plotted on the x-axis. The color on each bin corresponds to the number of waveforms of a given PID and total energy. The 2d histogram was made using matplotlib.pyplot.2Dhist.

To find the figure of merit, the counts for each bin as a function of the y-axis value for a given x-axis value was extracted and plotted as a 1D histogram using matplotlib.pyplot.hist. The sum of two Gaussian, or normal, distributions was fitted to these 1D histograms using
scipy.optimize.curve_fit. The centroids and standard deviations were extracted and used to calculate the figure of merit at various x-axis values. The equation for the figure of merit is given in eq. 3.5 on p. 29.

The AmBe source produces far more $\gamma$-rays than neutrons. In order to prevent the detector from measuring too few neutrons over the course of 20,000 events, the $\gamma$-rays were attenuated with a 2 in. block of lead. This has the effect of decreasing the amplitude of the $\gamma$-ray peak. While this does not affect the figure of merit, which is only dependent on the centroid separation and the full-width at half-max of the 2 peaks, large differences in the intensities can cause the smaller peak to be obscured by the larger peak, and appear indistinguishable from the larger peak. Discrimination is only possible in places where the centroid separation, width, and amplitude are such that both peaks are visible. Large differences in amplitude may conceal discrimination that would be possible if the amplitudes were more similar. As long as both peaks are visible, the determination of a figure of merit is possible.
Chapter Seven

Results - EJ-299-33A

For the three detectors, EJ-299-33A in two different sizes and an NE213 Liquid Scintillator, three 2D histograms were generated:

1. $Q_{\text{total}}$ vs the Pulse Height

2. The PID calculated using eqn. 6.1 vs the Pulse Height

3. The PID vs $Q_{\text{total}}$

Histograms of different types are discussed and a comparison of different detectors is found in the section for each histogram.

7.1 Energy Spectra

The energy spectra for $^{137}$Cs, $^{22}$Na, and $^{54}$Mn were captured with each detector. For all detectors, the spectra show a distinct Compton edge easily distinguishable from the background (see figures 7.1 - 7.3). The resolution of the edges are similar for both EJ-299-33A and NE213. All spectra show simple backgrounds, however they exhibit a significant signal at low energies ($< 100$ keV). We believe these may be x-rays, but they could also be attributed to false triggers, or both. Proximity to the trigger threshold further obscures the source of this peak.
**Figure 7.1** Energy spectra captured by NE213. Compton edges for the $\gamma$-ray energies are labeled.

**Figure 7.2** Energy spectra captured by the $2'' \times 2''$ EJ-299-33A. Compton edges for the $\gamma$-ray energies are labeled.
Figure 7.3 Energy spectra captured by the 2" × 1" EJ-299-33A. Compton edges for the γ-ray energies are labeled.

7.2 Total Light Output vs. Pulse Height

The measured total light output ($Q_{\text{total}}$) is plotted as a function of pulse height. The plots are shown in figures 7.4, 7.5, and 7.6 for NE-213, EJ 2"×1", and EJ 2"×2" respectively. For peak height lower than 400 keVee for NE213 and 600 keVee for both EJ-299-33As, PID between neutrons and γ-rays is not possible as the neutron ridge and the γ-ray ridge are merged. However for the the pulse heights greater than these values, neutrons and γ-rays do become distinguishable. While the discrimination becomes possible for NE213 at lower energies than for EJ-299-33A, at the lowest energies measured NE213 has a wider spread of values for the total light output. The 2"×1" and 2"×2" exhibit very similar discrimination using this method. As shown further on, other comparisons offer better separations at much lower energies. This is of particular interest because the use of $Q_{\text{Total}}$ vs. Pulse Height is common in pulse shape discrimination.
Figure 7.4 A 2D-Histogram of the total integral vs. pulse height for NE213. Red represents the greatest number of counts and blue represents the least, with white indicating no count. The top ridge is made of neutrons and the lower made of $\gamma$-rays.
Figure 7.5 A 2D-Histogram of the total integral vs. pulse height for EJ-299-33 2×1. Red represents the greatest number of counts and blue represents the least, with white indicating no count. The top ridge is made of neutrons and the lower made of γ-rays.
Figure 7.6 A 2D-Histogram of the total integral vs. pulse height for EJ-299-33 2×2. Red represents the greatest number of counts and blue represents the least, with white indicating no count. The top ridge is made of neutrons and the lower made of γ-rays.
7.3 PID vs. Pulse Height

Pulse Height vs. PID is shown in 7.7 - 7.12. As is clearly visible EJ-299-33A and NE213 achieve similar PID. All three scintillators show good PID resolution as low as 200 keVee. The 2"×2" EJ-299 detector shows the best resolution at low energies, achieving a figure of merit of 1.1 at 135 keVee. The 2"×1" EJ-299 achieves a figure of merit of only 0.894 at the same energy, with FOM = 0.97 at 165 keVee. NE213 has the worst response, with a figure of merit equaling 0.71 at 135 keVee, not achieving a figure of merit greater than 0.90 until 195 keVee. This suggests that small EJ-299-33A detectors discriminate slightly better than NE213 at low energies. EJ-299-33A has promise for use in situations where organic crystal or liquid scintillators are not suitable without sacrificing discrimination capabilities.

7.4 PID vs. Total Light Output

The figure of merit calculated from the 2D histogram of the Total Light Output, $Q_{\text{total}}$, vs PID is compared. $Q_{\text{total}}$ is not converted into keVee because the FemtoDAQ bins by pulse height. Without conversion, comparisons of different detectors are difficult to compare, so more attention here is given to comparing two different sizes of EJ-299-33A.

As shown in figures 7.14 - 7.19, the comparison of $Q_{\text{total}}$ vs PID overall provides a clear separation of the neutrons and gamma rays, and provides reliable separation at reasonable energies for use in n/γ discrimination. NE213 provides figures of merit of 1.10 at values for $Q_{\text{total}}$ as low as $2.84 \times 10^5$ (arb. units). EJ-299-33A shows nearly identical separation based on PID and $Q_{\text{total}}$ for the $2 \times 2$ and $2 \times 1$ sizes. The same figure of merit, FOM = 1.08, is found for the energy range of $1.197 \times 10^6 - 1.496 \times 10^6$. Fig. 7.20 shows that for low energies, the figure of merit increases with increasing energy.
Figure 7.7 A 2D-Histogram of the PID vs. Pulse Height for NE213. Red represents the greatest number of counts and blue represents the least, with white indicating no count. The top ridge is made up of neutrons, $PID = 0.815 \pm 0.008$. The lower ridge is made up of $\gamma$-rays, $PID = 0.786 \pm 0.006$. Notice the narrowing spread with increasing pulse height, suggesting that our separation continues to improve with increasing pulse height. The high number of data points at high energy and their spread is due to waveforms whose pulse height exceeded the maximum voltage of the FemtoDAQ.
Figure 7.8 NE213: The set of 4 graphs shows the progression through successive bins of the separation between \(\gamma\)-rays and neutrons for the pulse height vs PID discrimination method. The index of the bin from 0 pulse height is given by \(i\). The bottom right graph, for energies in the range 180 - 210 keVee, it becomes visibly clear that a separation is present. The bottom left graph has a Figure of Merit equal to 0.80, while the bottom right has a figure of merit of 0.93.
Figure 7.9 A 2D-Histogram of the PID vs. Pulse Height for EJ299-33 2×2. Red represents the greatest number of counts and blue represents the least, with white indicating no count. The top ridge is made up of neutrons, \( PID = 0.917 \pm 0.005 \). The lower ridge is made up of \( \gamma \)-rays, \( PID = 0.898 \pm 0.003 \). Notice the narrowing spread with increasing pulse height, suggesting that our separation continues to improve with increasing pulse height.
Figure 7.10 EJ-299-33A, 2×2: The bottom 4 graphs shows the progression through successive bins of the separation between γ-rays and neutrons for the pulse height vs PID discrimination method. The bottom left graph, for energies in the range 120 - 150 keVee, it becomes visibly clear that a separation is present. The middle left graph has a figure of merit equal to 0.73, while bottom left graph has a figure of merit equal to 1.10, and the bottom right has an FOM = 1.09. The top left graph exhibits 2 peaks for pulse heights between 0 and 30 keVee that vanishes in the next graph, giving the illusion of good separation at low energies. However the top left graphs has FOM = 0.64, implying that this apparent separation is rather poor.
Figure 7.11 A 2D-Histogram of the PID vs. Pulse Height for EJ299-33 2×1. Red represents the greatest number of counts and blue represents the least, with white indicating no count. The top ridge is made up of neutrons, $PID = 0.912 \pm 0.003$. The lower ridge is made up of $\gamma$-rays, $PID = 0.898 \pm 0.002$. Notice the narrowing spread with increasing pulse height, suggesting that our separation continues to improve with increasing pulse height.
Figure 7.12 EJ-299-33A, 2×1: The set of 4 graphs shows the progression through successive bins of the separation between γ-rays and neutrons for the pulse height vs PID discrimination method. In the bottom right graph, for energies in the range 150 - 180 keVee, a clear separation is present. This energy range has a figure of merit equal to 0.97. The figure of merit for the bottom left graph is equal to 0.89.
Figure 7.13 At low energies the figure of merit rises rapidly with increasing energy. At higher energies, the figure of merit continues to increase but at a slower pace, matching our expectation that the discrimination capabilities continue to improve with increasing energy.
Figure 7.14 A 2D-Histogram of the PID vs. total integral for NE213. Red represents the greatest number of counts and blue represents the least, with white indicating no count. The top ridge is made up of neutrons, $PID = 0.81 \pm 0.01$. The lower ridge is made up of $\gamma$-rays, $PID = 0.762 \pm 0.007$. Notice the narrowing spread with increasing $Q_{total}$, suggesting that the separation may continue to improve with increasing $Q_{total}$. The drift at high energies is due to waveforms whose pulse height exceeded the limits of the FemtoDAQ.
Figure 7.15 NE213: The set of 4 graphs shows the progression through successive bins of the separation between $\gamma$-rays and neutrons for the $Q_{total}$ vs PID discrimination method. The bottom left graph, $1.81 \times 10^5 - 2.11 \times 10^5$, has a figure of merit equal to 0.93. The bottom right graph, for energies in the range $2.69 \times 10^5 - 2.99 \times 10^5$, it becomes visibly clear that a separation is present. This corresponds to a figure of merit equal to 1.10.
Figure 7.16 A 2D-Histogram of the PID vs. total integral for EJ-299-33 2×2. Red represents the greatest number of counts and blue represents the least, with white indicating no count. The top ridge is made up of neutrons, $PID = 0.916 \pm 0.005$. The lower ridge is made up of $\gamma$-rays, $PID = 0.897 \pm 0.003$. Notice the narrowing spread with increasing $Q_{total}$, suggesting that the separation may continue to improve with increasing $Q_{total}$. 
Figure 7.17 EJ-299-33A, 2×2: The set of 4 graphs shows the progression through successive bins of the separation between γ-rays and for the $Q_{total}$ vs PID discrimination method. The top right graph, $5.18 \times 10^5 - 7.77 \times 10^5$, has a figure of merit equal to 0.84. The bottom left graph, for energies in the range $7.77 \times 10^5 - 10.4 \times 10^5$, it becomes visibly clear that a separation is present. This corresponds to a figure of merit equal to 0.93. Further separation is seen in the bottom right graph, which has $FOM = 1.07$. 
Figure 7.18 A 2D-Histogram of the PID vs. total integral for EJ-299-33 2×1. Red represents the greatest number of counts and blue represents the least, with white indicating no count. The top ridge is made up of neutrons, $PID = 0.912 \pm 0.003$. The lower ridge is made up of $\gamma$-rays, $PID = 0.898 \pm 0.002$. Notice the narrowing spread with increasing $Q_{total}$, suggesting that the separation may continue to improve with increasing $Q_{total}$. 
Figure 7.19 EJ-299-33A, 2×1: The set of 4 graphs shows the progression through successive bins of the separation between $\gamma$-rays and neutrons for the $Q_{\text{total}}$ vs PID discrimination method. The top right graph, $6.05 \times 10^5 - 9.06 \times 10^5$, has a figure of merit equal to 0.92. The bottom left graph, for energies in the range $9.06 \times 10^5 - 12.1 \times 10^5$, it becomes visibly clear that a separation is present. This corresponds to a figure of merit equal to 0.95. Further separation is seen in the bottom right graph, which has FOM = 1.08.
Figure 7.20 At low energies the figure of merit rises rapidly with increasing energy. At higher energies, the figure of merit continues to increase but at a slower pace, matching our expectation that the discrimination capabilities continue to improve at high energies.
7.5 Conclusions

We have compared the capability of NE213, a 2" × 2" EJ-299-33A, and a 2" × 1" EJ-299-33A to separate neutrons from γ-rays by pulse shape analysis. The pulse shape analysis methods investigated here are total light output, $Q_{total}$, vs. pulse height, particle identification (PID, using eq. 6.1) vs. pulse height, and PID vs. $Q_{total}$. At lower energies (pulse height < 400 keVee for NE213 or < 600 keVee for both EJ-299-33As), the neutron and γ-ray ridges are merged and no separation is possible for $Q_{total}$ vs. pulse height for the distribution considered. Even at energies as low as 200 keVee, the ridges are distinguishable for the PID vs. pulse height method. Comparisons find that the size of EJ-299-33A in the axial direction have little effect on the n/γ discrimination capabilities at low energy for lengths on the order of 1-2 inches. When comparing using PID and pulse height, EJ-299-33A offers equivalent or slightly better discrimination capabilities than NE213. A more thorough investigation may better quantify how much better EJ-299-33A provides n/γ separation at low energies.
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