Development and Operation of a $^6\text{LiF}:\text{ZnS}(\text{Ag})$ - Scintillating Plastic Capture-Gated Detector

K. Wilhelm^{a,*}, J. Nattress^b, I. Jovanovic^b

^a*Department of Mechanical and Nuclear Engineering, The Pennsylvania State University,
University Park, PA 16802, USA*

^b*Department of Nuclear Engineering and Radiological Sciences, University of Michigan, Ann Arbor, MI 48109, USA*

Abstract

We report on the design, construction, and operation of a capture-gated neutron detector based on a heterogeneous scintillating structure comprising two scintillator types. A flat, 500 μm thick sheet composed of a mixture of lithium-6-fluoride capture agent, ^6LiF , and zinc sulfide phosphor, $\text{ZnS}(\text{Ag})$, is wrapped around scintillating polyvinyl toluene (PVT) in a form of cylinder. The $^6\text{LiF}:\text{ZnS}(\text{Ag})$ sheet uses an aluminum foil backing as a support for the scintillating material and as an optical reflector, and its optical properties have been characterized independently. The composite scintillator was tested using ^{252}Cf , DD fusion, ^{137}Cs , and ^{60}Co sources. The intrinsic detection efficiency for neutrons from an unmoderated ^{252}Cf source and rejection of gammas from ^{137}Cs were measured to be 3.6% and 10^{-6} , respectively. A figure of merit for pulse shape discrimination of 4.6 was achieved, and capture-gated spectroscopic analysis is demonstrated.

Keywords: Neutron scintillation, composite detector, capture-gated spectroscopy, fast neutron detection, pulse shape discrimination, $^6\text{LiF}:\text{ZnS}(\text{Ag})$ scintillator

1. Introduction

Efforts to detect special nuclear materials, including those that are heavily shielded and in transit, have resulted in a need to develop improved radiation detectors, including fast neutron detectors that can be readily scaled to achieve large area coverage. Among the various neutron detection technologies, ^3He detectors combined with moderating materials still serve as a benchmark. As a consequence of the large demand for ^3He and its scarce supply, the specific cost of ^3He is high, making large coverage prohibitively expensive. One approach to efficiently detect fast neutrons is by use of proton recoil in pulse shape discriminating (PSD) materials, including liquids [1, 2, 3] and plastics [1, 4, 5]. Detectors based on ^4He recoil in a high pressure gas have received significant attention in the recent period [6]; however, the need for high pressure presents technological challenges. An alternative to recoil based detection is harnessing the benefits of neutron capture reactions, such as $^6\text{Li}(\text{n},\text{t})^4\text{He}$ or $^{10}\text{B}(\text{n},\alpha)^7\text{Li}$. Unlike detectors based on heavy charged particle (HCP) recoil, detectors based on neutron capture offer high neutron/gamma discrimination even at low neutron energies. This makes these types of detectors also suitable for detection of fast neutrons that have been well moderated (for example, by intervening shielding) or thermal neutrons. Recent advances

*Corresponding author.

Email address: kvw5368@psu.edu (K. Wilhelm)

in the development of homogeneously doped scintillators with PSD capabilities based upon this principle have been reported, using ^{10}B [1, 7, 8, 9] and ^6Li [1, 9, 10, 11, 12] dopants. Detectors that rely on neutron capture can also be used for neutron spectroscopy via the capture-gating technique [10].

The other approach to realize neutron capture-based detectors with neutron/gamma discriminating properties is by combining scintillators of varying scintillation characteristics and neutron sensitivity into heterogeneous structures. Composites of lithium-doped glass embedded within a scintillating polymer [13, 14, 15, 16] and lithium gadolinium borate (LGB) composites [17, 18, 19], have been demonstrated to be scalable to high efficiency while maintaining moderate cost, and have been used to perform capture-gated spectroscopy [18, 19, 20, 21]. Composite designs that use $^6\text{LiF:ZnS(Ag)}$ scintillators have recently accomplished high levels of gamma discrimination [22]. A heterogeneous design using $^6\text{LiF:ZnS(Ag)}$ scintillator sheets has been recently demonstrated in a coincidence design and used to resolve the energy spectra of incident fast neutrons [23]. That design requires the use of two photodetectors, and only incorporates the $^6\text{LiF:ZnS(Ag)}$ on one edge of the detector, thereby sacrificing neutron detection efficiency in order to achieve fast neutron spectroscopy. We demonstrate a non-segmented $^6\text{LiF:ZnS(Ag)}$ -PVT detector design that is readily assembled and scaled to large volumes, has high neutron efficiencies, and maintains high neutron/gamma discrimination. The detector uses a single photomultiplier, and shows the potential for use of the $^6\text{LiF:ZnS(Ag)}$ scintillator in a simple, fast neutron spectrometer through the capture-gating technique, while maintaining high neutron detection efficiency. These characteristics make it highly suitable for a number of applications in nuclear security, but also for applications in basic research such as antineutrino detection [24, 25, 26] and medical applications [27, 28]. Additionally, we report on the independently characterized optical properties of the $^6\text{LiF:ZnS(Ag)}$ scintillator and its performance within the heterogeneous fast neutron detector. We quantify the standard performance characteristics such as PSD figure of merit, efficiency, and gamma rejection, and also demonstrate capture-gated spectroscopic analysis using monoenergetic and spontaneous fission neutrons.

2. Detector Design

2.1. Composition and Principle of Operation

In this work, a cylindrical block of scintillating PVT has been encased within a thin layer of an inorganic thermal neutron scintillator. No coupling agent was used to optically pair the PVT to the neutron scintillator; as a result, a small air gap is present on their interface. The use of two different scintillators with different scintillation decay time constants provides excellent neutron/gamma discrimination. The scintillating PVT is EJ-230 manufactured by Eljen Technology with a prompt fluorescence decay time of 1.5 ns [29] and has no reported PSD characteristics. The thermal neutron scintillating matrix, $^6\text{LiF:ZnS(Ag)}$, is EJ-426 with a prompt fluorescence decay time of 200 ns [30], and is also manufactured by Eljen Technology. The shorter pulse from the PVT scintillator can be associated with interactions of gamma rays producing energetic electrons (for example, by Compton scattering) or proton recoils. The longer pulses from the $^6\text{LiF:ZnS(Ag)}$ scintillator originate from the neutron capture reaction on ^6Li , with a Q-value of 4.78 MeV. Light from both components of the scintillator propagates with little loss through PVT; however, the air-coupling on the PVT- $^6\text{LiF:ZnS(Ag)}$ interface reduces the fraction of the emitted light that is transported to the PMT and detected. This loss results from guiding the light by reflection within

the interface, and also by light first entering and then exiting the PVT volume on the opposite side and being absorbed by the ${}^6\text{LiF:ZnS(Ag)}$ scintillator. [26]. PSD makes the neutron capture reaction in the ${}^6\text{LiF:ZnS(Ag)}$ scintillator distinguishable from interactions in the PVT. A time correlation between the scintillation from the PVT and the scintillation attributed to neutron capture in ${}^6\text{LiF:ZnS(Ag)}$ is used to identify the scintillation from PVT as a neutron thermalization event. In this case, the total scintillation light output from PVT is correlated to the incident neutron energy, and can be used for capture-gated spectroscopy [1].

2.2. Detector Simulation

All simulations were performed using Geant4.10.1 [31]. The size and shape of the detector have been chosen based on the general guidance from prior analyses [13, 32]. The volume of the scintillating PVT can be chosen for a given neutron spectrum to achieve efficient thermalization, thereby maximizing the capture-gated efficiency, (*i.e.* the fraction of incident fast neutrons that are thermalized and subsequently captured). The detector assembled in this work is a cylinder of scintillating PVT with a height of 12.7 cm and diameter of 12.7 cm. The entire side-wall and one base of the cylinder are covered with a 0.5 mm thick layer of ${}^6\text{LiF:ZnS(Ag)}$. The opposing base of the PVT cylinder is left uncovered for coupling to a 12.7 cm photomultiplier tube (PMT). In all simulations the neutrons are produced by a point source and are incident upon the side-wall and filling the entire aperture of the detector. Though not a requisite for capture-gating, the significant majority of incident fast neutrons are captured by ${}^6\text{Li}$ only after being nearly fully thermalized (98.6% of captured neutrons are thermalized to <100 keV), as illustrated by Monte Carlo simulation in Figure 1. This phenomenon is a result of the relatively large thermal neutron cross section of ${}^6\text{Li}$ (~ 940 b) and its $1/v$ dependence on neutron speed (v).

Figure 1: Simulated neutron kinetic energy at time of ${}^6\text{Li}$ capture. 10^7 neutrons from an unmoderated ${}^{252}\text{Cf}$ spontaneous fission source were simulated.

The impact of detector size on neutron capture efficiency was investigated by Monte Carlo simulation. The PVT cylinder diameter was varied while keeping the aspect ratio of the cylinder at unity. The ${}^6\text{LiF:ZnS(Ag)}$ scintillator dimensions were varied accordingly, so that the EJ-230 polymer is always enclosed by the EJ-426 thermal neutron scintillator. An important consideration for this detector design is the competing thermal neutron capture reaction ${}^1\text{H}(n,\gamma){}^2\text{H}$. The thermal neutron capture cross section for ${}^1\text{H}$ (~ 0.33 b) [33] is much smaller than that of ${}^6\text{Li}$, but increasing the PVT volume significantly increases the probability for a neutron to be lost in this process. The ${}^1\text{H}(n,\gamma){}^2\text{H}$ reaction yields a 2.2 MeV gamma that has a high probability ($\sim 72\%$ according

Figure 2: Simulated dependence of the neutron capture efficiency on detector diameter. The capture by ${}^6\text{Li}$ without the presence of the competing ${}^1\text{H}$ capture reaction (a), the capture by ${}^6\text{Li}$ with the presence of the competing ${}^1\text{H}$ capture reaction (b), and the capture by only the ${}^1\text{H}$ (c) are depicted. The aspect ratio of the detector is kept at unity. Chosen detector diameter is circled. Neutrons are incident from an unmoderated ${}^{252}\text{Cf}$ spontaneous fission source.

to simulation for the chosen detector dimensions, if the gamma is produced in the center of the detector) of escaping the detector volume without interaction. Any electron recoils resulting from these gammas would result in a short pulse as recorded by the digitizer. Results of simulation of neutron capture are shown in Figure 2 and include the neutron captures on both ${}^6\text{Li}$ and ${}^1\text{H}$. Another important consideration for this detector is the probability that a thermalized neutron is captured by ${}^6\text{Li}$ as it begins to diffuse to the outer edge of the detector. Monte Carlo simulations show that $\sim 99.6\%$ of thermal neutrons (< 0.05 eV) incident on the internal edge of the 0.5 mm thick ${}^6\text{LiF}:\text{ZnS}(\text{Ag})$ scintillator are captured prior to leaving the detector volume. While the light collection efficiency has not been simulated, it can still be concluded that the neutron capture efficiency has a strong dependence on detector size, as expected given the increasing probability for thermalized neutrons to be captured. For this reason, a practical choice of 12.7 cm diameter was made for the experiment, which corresponds to a simulated neutron capture efficiency of 7.8% for unmoderated neutrons from spontaneous fission of ${}^{252}\text{Cf}$. Several steps of the detector assembly process are shown in Figure 3.

2.3. Optical Properties of EJ-426 Scintillator

It is desirable to understand the detailed optical characteristics of the ${}^6\text{LiF}:\text{ZnS}(\text{Ag})$ scintillator with aluminum foil backing (EJ-426) as this directly contributes to detection efficiency. It is known that the light yield from the ${}^6\text{LiF}:\text{ZnS}(\text{Ag})$ scintillator resulting from HCP energy deposition is approximately three times higher than that of the anthracene crystal [34]; however, much of the light generated in ${}^6\text{LiF}:\text{ZnS}(\text{Ag})$ is attenuated due to its poor transparency. The exact type of the scintillator used in this study is EJ-426-0, which has a ${}^6\text{LiF}:\text{ZnS}(\text{Ag})$ mass ratio of 1:3, and is enriched to 95% ${}^6\text{Li}$ [30]. The mass ratio of 1:3 was chosen for its performance when detecting thermal neutrons [34]. Modeling the complete neutron detection process, from capture to electronic signal generation within the PMT, is challenging due to the heterogeneous nature of the powdery matrix [22]. For this reason, and because this design only incorporates the ${}^6\text{LiF}:\text{ZnS}(\text{Ag})$ on the outer edge of the detector, a study was conducted to better understand the characteristic light

Figure 3: Detector assembly: (a) EJ-426 sheets and EJ-230 PVT cylinder, (b) EJ-426 wrapped around EJ-230 PVT, and (c) detector fully assembled and connected to a H6527 Hamamatsu PMT.

production, transport, and reflection within this medium. Of particular interest is the effect of light attenuation, which could compromise the identification of some capture events produced deeper within the volume of the ${}^6\text{LiF}:\text{ZnS}(\text{Ag})$ scintillator and contribute to a difference between capture and detection efficiency.

Samples of EJ-426 were placed within a Perkin Elmer Lambda 950-UV/VIS spectrometer with an integrating sphere for light reflection and transmission measurements. These samples comprised the matrix of ${}^6\text{LiF}:\text{ZnS}(\text{Ag})$ on a clear polymer backing to better allow for unimpeded light transmission. The samples were then exposed to light over a broad spectral range (250 nm–600 nm). In all measurements the light was incident on the ${}^6\text{LiF}:\text{ZnS}(\text{Ag})$ material, and not the clear polymer backing. The light was collected on both sides of the sample. Figures 4 and 5 show the resulting reflective and transmission characteristics, respectively.

Figure 4: Fraction of incident light reflected from a 0.5 mm thick sample of EJ-426-0 with a clear polymer backing. The emission spectrum of EJ-230 PVT is shown, ranging over 350 nm–490 nm and peaking at 391 nm [29]. The emission spectrum of EJ-426-0 scintillator is also shown, ranging over 400 nm–560 nm and peaking at 450 nm [30]. The emission spectra have been normalized to the light reflectance scale for comparison purposes.

The diffuse nature of ${}^6\text{LiF}:\text{ZnS}(\text{Ag})$ reduces the fraction of its scintillation light that escapes and is transported to the PMT, as shown in Figure 5. Scintillation light generated from ${}^4\text{He}$

Figure 5: Fraction of light transmitted through a 0.5 mm thick sample of EJ-426-0 with a clear polymer backing. The emission spectrum of the EJ-426-0 scintillator is also shown [30]. The emission amplitude has been normalized to the light transmitted scale for comparison purposes.

and ^3H HCPs, which deposit their energy near the surface of the EJ-426 scintillator, escapes to the PVT and is transported to the PMT with higher efficiency than the scintillation from energy deposited deeper in the EJ-426 volume. The detection probability of neutron capture within EJ-426 is, therefore, affected by the distance the scintillation light must travel through the $^6\text{LiF:ZnS(Ag)}$ matrix.

2.4. Characteristics of the Interface between Scintillators

The $^6\text{LiF:ZnS(Ag)}$ scintillator is located solely along the surface of PVT and only a mechanical contact is applied. This geometric configuration, in which there is no non-scintillating interface layer, has consequences on the distribution of energy deposition from HCPs. The HCPs deposit their energy in EJ-426 with a mean travel distance of $29.5\text{ }\mu\text{m}$ and $9.0\text{ }\mu\text{m}$ from the point of capture for ^3H and ^4He , respectively. As shown in Figure 6, the majority of neutron captures occur on the inner edge of the EJ-426, which is an indication of the effect of moderation of the PVT. Because the neutron captures extend throughout the EJ-426, the scintillation produced deeper within EJ-426 is substantially attenuated. This attenuation will result in a broad range of detected light output from neutron capture.

For neutron captures that occur close to the interface between EJ-426 and EJ-230 scintillators, a fraction of the HCP energy can be deposited in EJ-230. As a result, it is expected that these events can be recognized by pulse shapes that resemble a superposition of characteristic pulse shapes from the two scintillators. Similarly, fast proton recoils in EJ-230, which occur close to the interface between EJ-426 and EJ-230 scintillators, may also deposit a fraction of their energy in EJ-426. Such events would be indistinguishable from the neutron captures taking place near the interface using only the pulse shape, but in some cases it may be possible to identify them by also considering the total light output. In future work it is desirable to further quantify the effect the air-coupling-interface has on optical photon transmission.

3. Experimental Setup

The detector was mounted to a Hamamatsu H6527 PMT using Eljen Technology's EJ-550 optical grease. The detector was then fully wrapped in black electrical tape in order to prevent

Figure 6: Simulated location of neutron captures in ${}^6\text{LiF:ZnS(Ag)}$ (black) and locations at which emitted HCPs stop (gray). Negative distances correspond to locations within the PVT, while the distances between 0 μm and 500 μm represent the locations within the ${}^6\text{LiF:ZnS(Ag)}$ scintillator. Distances greater than 500 μm are outside of the detector volume. For normalization purposes, only the release of ${}^3\text{H}$ was included to maintain a unity ratio of captures to HCPs. An unmoderated ${}^{252}\text{Cf}$ spontaneous fission source is used for the simulation. [Color version available online]

light leaks. A CAEN DT1470ET high voltage source was used to power the PMT at a voltage of -1500 V. Waveforms were digitized using a CAEN DT5730 14-bit 500 MS/s digitizer. A record length of 400 ns was used. The digitizer uses standard CAEN firmware and the data was collected and stored using the CAEN Wavedump software. Post-processing of all data for integration and data analysis was performed using the ROOT framework [35].

Gamma sources were used for both energy calibration and gamma rejection measurements. Calibration was performed using a 1 μCi ${}^{60}\text{Co}$ and a 5 μCi ${}^{137}\text{Cs}$ source according to the method established in Ref. [36]. Gamma rejection measurements were performed using only the ${}^{137}\text{Cs}$ source and were conducted over a time interval of 7 hours. This measurement time was used both for the source measurement and the background measurement. The source was placed approximately 30 cm away from the side-wall of the detector in the center plane of the detector. Gamma rejection measurements were performed far from any artificial neutron source.

Neutron measurements were performed using a 5.4 mCi ${}^{252}\text{Cf}$ source and DD fusion-generated, 2.45 MeV monoenergetic neutrons. Measurements were conducted by varying the time collection windows in the range of 0.5–4 hours and with distances varying in the range of 30–300 cm in order to ensure consistency. Intrinsic efficiency measurements were performed by using a shadow cone and an unmoderated ${}^{252}\text{Cf}$ spontaneous fission source, and followed the method described in Ref. [37]. For efficiency measurements, the source to cone distance was ~ 19 cm, and the cone to detector distance was ~ 194 cm, making the overall source-to-detector distance ~ 263 cm. For these measurements, the source was placed so as to ensure center alignment with the side-wall of the detector. The detector was supported by a thin aluminum rack at a height of 135 cm, with a source height of ~ 141.5 cm for alignment to the detector center plane. Capture-gated measurements were performed by using the ${}^{252}\text{Cf}$ and DD fusion-generated neutron sources. For DD-fusion measurements, a portable neutron generator (Thermo Scientific MP 320) was used to produce monoenergetic, 2.45 MeV neutrons. The detector was placed approximately 60 cm from the DD generator such that the neutrons are incident on one side-wall of the detector volume. Relocating either the ${}^{252}\text{Cf}$ or DD neutron sources to a reduced neutron scatter environment was not possible due to facility restrictions governing radiation safety.

4. Analysis

4.1. Pulse Shape Discrimination

The method used for PSD analysis is described in Ref. [32] and uses charge integration. Baseline for each waveform was established by averaging the first 60 ns of the digitized record. The primary means of PSD used in this analysis involves comparison of the tail and total integrals of the waveforms. The integration windows were set relative to the waveform peak as follows: t_0 (start of the pulse integration) is 12 ns before the peak, t_1 (beginning of the tail of the pulse integration) is 30 ns after the peak, and t_2 (end of the pulse integration) is 280 ns after the peak. Figure 7 shows representative waveforms originating in EJ-426 and EJ-230 with the corresponding integration time windows used in PSD analysis. Neutron capture waveforms extend beyond the established record time window and integration limits, but the chosen integration windows are nevertheless suitable for identifying neutron capture events, as shown in Figure 8.

Figure 7: Sample waveforms originating in EJ-230 (black) and in EJ-426 (gray). PSD integration time limits t_0 , t_1 , and t_2 are shown. A ^{252}Cf source was used. [Color version available online]

Figure 8: A 4-hour measurement with ^{252}Cf produces a neutron band resulting from capture reactions occurring in the EJ-426 scintillator. The capture region extends in the range of $0.64 \lesssim \text{PSP} \lesssim 0.88$ and $\text{Light Output} \lesssim 5000 \text{ keVee}$. A pulse-height cut has been implemented at the trigger threshold and is evident in the lower energy of the neutron band beginning at $\sim 100 \text{ keVee}$. [Color version available online]

Due to the self-attenuation occurring in the EJ-426 scintillator, the neutron region is not an isolated island resulting from the discrete release of 4.78 MeV of energy from the ${}^6\text{Li}(n,t){}^4\text{He}$ reaction. Instead, the neutron region comprises a continuous band of light output that extends from the upper limits of the capture reaction energy to 0 keVee. A small fraction of events producing high light output in the ${}^6\text{LiF:ZnS}(\text{Ag})$ scintillator result from captures at higher neutron energies. A curved region connects the neutron band to the lower scatter/recoil band. This bridge-like structure is partially a result of a superposition of long and short pulses formed by the energy deposition of the HCPs in both the ${}^6\text{LiF:ZnS}(\text{Ag})$ and PVT (see Figure 6). This is made possible by neutron capture events occurring on the inner edge of the ${}^6\text{LiF:ZnS}(\text{Ag})$, whereby one of the HCP deposits energy within the ${}^6\text{LiF:ZnS}(\text{Ag})$ and the other within the PVT. Based on simulation, this superposition of pulses accounts for approximately 2.8% of neutron captures. Inclusion of these superimposed waveforms increases the intrinsic efficiency by 0.07%. Superimposed waveforms were identified by $PSP < 0.64$ and where the waveform never returns to the established baseline as a result of prolonged fluorescence. Figure 9 shows the superposition region and an average waveform found within this bridge-like structure compared to an average waveform found within the neutron capture and scatter/recoil bands.

Figure 9: (a) PSD of bridge-like region with all superposition waveform cuts implemented. (b) Average waveform found in neutron capture region (dotted) in the range of $0.64 \lesssim PSP \lesssim 0.88$. Average waveform found in the bridge-like structure (solid) in the range of $0.50 \lesssim PSP \lesssim 0.55$. Average waveform found in scatter/recoil region (dashed) in the range of $0.0 \lesssim PSP \lesssim 0.2$. All waveforms are an average of 50 waveforms from each region and have a light output of 850–900 keVee. Waveforms were normalized to their maximum amplitude and aligned to their peak position for comparison. [Color version available online]

The neutron capture band is further isolated and defined by applying several cuts. First, a Gaussian was fit to the Pulse Shape Parameter (PSP) of the neutron band (upper band of PSD shown in Figure 8), and a 3σ cut was applied such that a neutron capture is in the range $0.64 \lesssim PSP \lesssim 0.88$, as shown in Figure 10. PSP is defined as the ratio of the tail to total waveform integral, with bounds shown in Figure 7. Second, to eliminate low energy noise within this PSP range, an additional cut is applied to this region such that the full integral must be greater than 70 keVee. This region, shown in Figure 11, was used to evaluate the neutron intrinsic efficiency and gamma rejection performance.

The intrinsic efficiency of the detector for neutron capture was determined using a ${}^{252}\text{Cf}$ source by taking the ratio of the shadow cone-emplaced, background-subtracted detected neutron rate and the calculated incident neutron rate, including the subtended solid-angle and the neutron rate

Figure 10: *PSP* of the neutron region. A Gaussian fit is shown in red between $0.67 \lesssim PSP \lesssim 0.85$ and the standard deviation ($\sigma \approx 0.03$) was used for further defining the neutron capture region. [Color version available online]

Figure 11: Neutron capture region with all cuts applied, rejecting the bridge-like superposition region. [Color version available online]

222 produced by the source. Gamma rejection measurements were conducted in the same manner,
 223 counting the number of gammas that would be misidentified as neutrons by falling within the
 224 established neutron region. A figure of merit (FOM) was then determined using a light output of
 225 70 keVee as

$$FOM = d / (FWHM_g + FWHM_n). \quad (1)$$

226 Here, d is the distance between the two distribution centroids, $FWHM_g$ is the full-width-half-
 227 maximum of the scatter/recoil distribution, and $FWHM_n$ is the full-width-half-maximum of the
 228 neutron capture distribution (see Figure 12). This method follows the approach used in Refer-
 229 ence [38] because of the non-Gaussian nature of the gamma distribution. The gamma distribution
 230 of the PSP is skewed by small, but finite differences between nuclear and electron recoil pulse
 231 shapes in the EJ-230 scintillator.

Figure 12: PSP distribution at a light output of 70 keVee obtained from a 4-hour measurement using a ^{252}Cf source. Quantities used in FOM calculations are annotated. Non-Gaussian nature of the gamma distribution is evident and has a centroid of $PSP \approx 0.1$.

232 4.2. Capture-Gated Spectroscopy

233 The thin sheet of $^6\text{LiF:ZnS(Ag)}$ scintillator is initially transparent to the incident fast neutrons,
 234 which first enter the detector volume and interact within the PVT scintillator. A fraction of
 235 energetic neutrons lose energy by elastic scattering, with a smaller fraction being moderated to low
 236 energies, which increases the probability of their capture on ^6Li . Figure 13 shows the location within
 237 the detector volume where proton recoil via neutron interactions occur. Because nearly all of the
 238 kinetic energy is transferred through proton recoil within ~ 50 ns [1], the many individual photons
 239 released from multiple interactions are observed as a single thermalization pulse. The pulse is
 240 strongly correlated to the total energy deposited in thermalization, and the incident neutron energy
 241 spectrum can therefore be examined through capture-gated analysis. Much of the capture-gated
 242 technique used in this analysis follows the procedure outlined in Ref. [20].

Figure 13: Simulated locations of interactions within the detector volume. A slice of the detector volume is shown. Blue color indicates the location of neutron capture on ${}^6\text{Li}$, red color indicates the location at which a HCP stops, and green indicates the location of neutron scatter. An enlarged sub-region depicts the nature in which HCPs are emitted following neutron capture. A ${}^{252}\text{Cf}$ point source was used in this simulation. [Color version available online]

Neutrons are then captured by ${}^6\text{Li}$ in the ${}^6\text{LiF}:\text{ZnS}(\text{Ag})$ scintillator. Monte Carlo simulations indicate that on average, 99% of all neutrons that are captured are first thermalized to energies at or below 0.025 eV, within 220 μs prior to capture, for both ${}^{252}\text{Cf}$ and DD neutrons. Using this time correlation, a cut is applied in order to only accept the events in PVT that occur within the time window of 0–220 μs prior to neutron capture. The time window is affected both by the ${}^6\text{Li}$ loading of the scintillator and the mean distance a thermal neutron travels prior to capture. The uncorrelated event rate $I(t)$ is governed by the distribution function

$$I(t)dt = r \exp(-rt)dt, \quad (2)$$

where t is the time interval and r is the average event rate [1]. Using the experimental data taken with the ${}^{252}\text{Cf}$ source and DD generator, the time to capture distributions were calculated. An exponential fit corresponding to Eq. (2) was calculated using the events in the range $220 \mu\text{s} < t < 500 \mu\text{s}$. This fit was then extended into the capture window (Figure 14) as a means of subtracting uncorrelated events.

The corrected inter-event time distribution, following the subtraction of uncorrelated coincident events, was compared to Monte Carlo simulations. Both the experimental and simulated data sets were normalized to the integral of the distribution between 0 μs and 250 μs . The results are shown in Figure 15 and are in good agreement with the simulation, indicating that the relevant aspects of detector architecture and composition are appropriately accounted for by the simulation.

5. Results and Discussion

The detector has an intrinsic neutron detection efficiency of 3.6% for unmoderated ${}^{252}\text{Cf}$ neutrons, and a gamma rejection rate of 10^{-6} . The FOM was calculated to be 4.6 at a light output of 70 keVee, showing that excellent neutron/gamma separation can be achieved using this design. The decrease in detection efficiency from that of the simulated 7.8% capture efficiency can be attributed,

Figure 14: An exponential fit is applied to the event distribution in the range 220–500 μs (black line). The fit was then extended into the capture time window 0–220 μs (gray line). Data from measurements made with the ^{252}Cf source are shown. [Color version available online]

Figure 15: Time to capture from first scatter event. The gray line indicates the experimental results, while the black line indicates the simulated results. Time distributions have been normalized over an integral calculated from 0 μs to 250 μs . Data from measurements made with the ^{252}Cf source are shown. [Color version available online]

in large part, to the light attenuation resulting from the opaque nature of the $^6\text{LiF:ZnS(Ag)}$ scintillator. The reason for this is because the detection efficiency of $^6\text{LiF:ZnS(Ag)}$ scintillators primarily depend on two factors: (1) the abundance of ^6Li for neutron capture, and (2) the thickness of the scintillator that optical photons must traverse [34].

Figure 16 shows the *PSD* and light output corresponding to the thermalization of fast neutrons from ^{252}Cf and DD sources, including uncorrelated events, and extracted from capture-gated analysis. Despite the presence of uncorrelated events, fast neutron spectroscopy with this detector design is achievable, even though neutron events cannot be identified from the PVT pulses alone. The light output spectrum for the ^{252}Cf source is indicative of a broad distribution neutron spectrum that is characteristic of ^{252}Cf spontaneous fission neutrons [39]. A feature corresponding to the monoenergetic 2.45 MeV neutrons from the DD reaction is clearly identifiable as well. Also of interest, Figure 16 suggests that for both sources, the EJ-230 PVT provides sufficient pulse shape differential between Compton scatter and proton recoil events to be visually observed, although

Figure 16: Capture-gated PSD depicting neutron thermalization events within EJ-230 PVT from exposure to (a) ^{252}Cf source and (c) DD fusion neutron source. Capture-gated light output spectra resulting from neutron thermalization within EJ-230 polymer from exposure to (b) ^{252}Cf source and (d) DD fusion neutron source. [Color version available online]

it is not sufficient for reliably distinguishing the two events. This phenomenon is also evident in Figure 12 and contributes to the asymmetry of the scatter/recoil PSP peak.

Scaling this design to even larger volumes would increase the efficiency (see Figure 2, noting the competition from the $^1\text{H}(n,\gamma)^2\text{H}$ reaction), but would hamper the detector’s ability to perform fast neutron spectroscopy due to a corresponding increase in the mean time to neutron capture. Lengthening this time increases the uncorrelated coincident event rate.

Loading the detector volume with additional ^6Li would further improve capture-gated performance metrics and further increase detection efficiency. An increase in the $^6\text{LiF:ZnS(Ag)}$ thickness does increase the neutron capture rate; however, the limitations posed by the opaque nature of the $^6\text{LiF:ZnS(Ag)}$ scintillator would become more important, since much of the scintillation light produced would be attenuated to low levels. The additional capture agent would thus be better situated within the PVT moderator, thereby decreasing the distance between neutron moderation and capture. One way to accomplish this is to load the detector volume with a homogeneously dispersed capture agent.

6. Conclusion

In this work, a composite detector design that uses a combination of PVT scintillating polymer and $^6\text{LiF:ZnS(Ag)}$ thermal neutron scintillator was designed, assembled, and characterized. This design, readily manufactured and easily scaled to larger volumes, was shown to be capable of performing thermal neutron detection with high efficiency while maintaining good neutron/gamma discrimination. It has also been shown that this design can be used for fast neutron spectroscopy through capture-gated analysis. Increasing the detector volume would increase neutron detection efficiency, but would have a degrading effect on the performance of fast neutron spectroscopy. At the current specifications, this detector offers efficient neutron detection and is capable of determining incident neutron energies through the capture-gating technique. The complexities associated with the use of the $^6\text{LiF:ZnS(Ag)}$ scintillator are difficult to fully model. Much work has already been, and is currently being performed to better understand and harness the capabilities of this material in both neutron and antineutrino detection. Future work would seek to further expound upon the performance metrics associated with this detector’s ability to perform fast neutron spectroscopy, and to develop a more complete understanding of the scintillation light transport in the heterogeneous detector structure.

Acknowledgments

The authors are grateful to Michael Mayer, Marc Wonders, Albert Foster, Zoubeida Ounaies, and Amira Meddeb for their comments, suggestions, and assistance with measurements. Additionally, we want to thank Eljen Technologies for providing the EJ-426 sample material used in light reflection/transmission measurements. The work was supported by the U.S. Department of Homeland Security under Grant Award Number 2014-DN-077-ARI078-02 and 2015-DN-077-ARI096. The research of J.N. was performed under appointment to the Nuclear Nonproliferation International Safeguards Graduate Fellowship Program sponsored by the National Nuclear Security Administration’s Next Generation Safeguards Initiative. K.W. is an active, Nuclear and Counter-Proliferation Officer in the U.S. Army, and is under the appointment of the DOD, U.S. Army, and United States Military Academy.

References

- [1] G. F. Knoll, Radiation Detection and Measurement, John Wiley & Sons, 2010.
- [2] A. Kaplan, M. Flaska, A. Enqvist, J. Dolan, S. Pozzi, Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment 729 (2013) 463–468.
- [3] M. Flaska, S. Pozzi, Pulse-shape discrimination for identification of neutron sources using the BC-501A liquid scintillator, in: Proceedings of the 8th Joint International Topical Meeting Mathematics & Computation and Supercomputing in Nuclear Applications (M&C+ SNA 2007), Monterey, 2007.
- [4] S. Pozzi, M. Bourne, S. Clarke, Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment 723 (2013) 19–23.
- [5] D. Cester, G. Nebbia, L. Stevanato, F. Pino, G. Viesti, Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment 735 (2014) 202–206.
- [6] R. Chandra, G. Davatz, H. Friederich, U. Gendotti, D. Murer, Journal of Instrumentation 7 (2012) C03035.
- [7] S. Normand, B. Mouanda, S. Haan, M. Louvel, Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment 484 (2002) 342–350.
- [8] L. M. Bollinger, G. E. Thomas, Review of Scientific Instruments 28 (1957) 489–496.
- [9] N. J. Cherepy, R. D. Sanner, P. R. Beck, E. L. Swanberg, T. M. Tillotson, S. A. Payne, C. R. Hurlbut, Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment 778 (2015) 126–132.
- [10] B. Fisher, J. Abdurashitov, K. Coakley, V. Gavrin, D. Gilliam, J. Nico, A. Shikhin, A. Thompson, D. Vecchia, V. Yants, Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment 646 (2011) 126–134.
- [11] C. Bass, E. Beise, H. Breuer, C. Heimbach, T. Langford, J. Nico, Applied Radiation and Isotopes 77 (2013) 130–138.
- [12] S. Ait-Boubker, M. Avenier, G. Bagieu, J. Cavaignac, J. Collot, J. Favier, E. Kajfasz, D. Koang, A. Stutz, B. Vignon, Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment 277 (1989) 461–466.
- [13] M. Mayer, J. Nattress, C. Trivelpiece, I. Jovanovic, Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment 784 (2015) 168–171.
- [14] G. Rich, K. Kazkaz, H. P. Martinez, Evaluation of a lithium-glass based composite neutron detector for He-3 replacement, in: APS Division of Nuclear Physics Meeting Abstracts, volume 1, 2012.

- [15] G. Rich, K. Kazkaz, H. Martinez, T. Gushue, Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment 794 (2015) 15–24.
- [16] J. B. Czirr, G. L. Jensen, Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment 349 (1994) 532–539.
- [17] P. Nelson, N. Bowden, Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment 660 (2011) 77–82.
- [18] J. B. Czirr, D. B. Merrill, D. Buehler, T. K. McKnight, J. L. Carroll, T. Abbott, E. Wilcox, Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment 476 (2002) 309–312.
- [19] M. Flaska, S. A. Pozzi, J. B. Czirr, Use of an lgb detector in nuclear nonproliferation applications, in: 2008 IEEE Nuclear Science Symposium Conference Record, IEEE, 2008, pp. 3376–3380.
- [20] J. Nattress, M. Mayer, A. Foster, A. B. Meddeb, C. Trivelpiece, Z. Ounaies, I. Jovanovic, IEEE Transactions on Nuclear Science 63 (2016) 1227–1235.
- [21] N. Mena, M. Villani, S. Croft, R. B. McElroy, S. A. Philips, J. B. Czirr, IEEE Transactions on Nuclear Science 56 (2009) 911–914.
- [22] J. H. Ely, M. Bliss, R. T. Kouzes, A. T. Lintereur, S. M. Robinson, E. R. Siciliano, M. T. Swinhoe, M. L. Woodring, Pacific Northwest National Laboratory, Richland, WA (2013).
- [23] V. Potapov, S. Ignatov, O. Ivanov, K. Balygin, V. Zaitsev, E. Meleshko, A. Klimov, M. Karetnikov, S. Korotkov, V. Ryzhkov, Instruments and Experimental Techniques 58 (2015) 329–336.
- [24] B. R. Kim, S. C. Kim, K. K. Joo, C. D. Shin, S. H. So, I. S. Yeo, Journal of the Korean Physical Society 66 (2015) 768–773.
- [25] S. D. Kiff, N. Bowden, J. Lund, D. Reyna, Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment 652 (2011) 412–416.
- [26] M. Sweany, J. Brennan, B. Cabrera-Palmer, S. Kiff, D. Reyna, D. Throckmorton, Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment 769 (2015) 37–43.
- [27] C. L. Tessa, T. Berger, R. Kaderka, D. Schardt, S. Burmeister, J. Labrenz, G. Reitz, M. Durante, Physics in Medicine and Biology 59 (2014) 2111.
- [28] D. W. Nigg, Physical dosimetry and spectral characterization of neutron sources for neutron capture therapy, in: Neutron Capture Therapy: Principles and Applications, Springer Berlin Heidelberg, Berlin, Heidelberg, 2012, pp. 227–257.
- [29] EJ-230 Data Sheet, Eljen Technology, August 2016. Available at http://www.eljentechnology.com/images/products/data_sheets/EJ-228_EJ-230.pdf.

- 392 [30] EJ-426 Data Sheet, Eljen Technology, August 2016. *Available at [http://www.](http://www.eljentechnology.com/images/products/data_sheets/EJ-426.pdf)*
393 *[eljentechnology.com/images/products/data_sheets/EJ-426.pdf](http://www.eljentechnology.com/images/products/data_sheets/EJ-426.pdf).*
- 394 [31] S. Agostinelli, J. Allison, K. a. Amako, J. Apostolakis, H. Araujo, P. Arce, M. Asai, D. Axen,
395 S. Banerjee, G. Barrand, et al., Nuclear instruments and methods in physics research section
396 A: Accelerators, Spectrometers, Detectors and Associated Equipment 506 (2003) 250–303.
- 397 [32] M. Mayer, J. Nattress, V. Kukharev, A. Foster, A. Meddeb, C. Trivelpiece, Z. Ounaies, I. Jo-
398 vanovic, Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spec-
399 trometers, Detectors and Associated Equipment 785 (2015) 117–122.
- 400 [33] B. Hamermesh, G. Ringo, S. Wexler, Physical Review 90 (1953) 603.
- 401 [34] C. Wu, B. Tang, Z. Sun, Q. Zhang, Z. Yang, J. Zhang, Y. Yang, J. Liang, J. Wu, Radiation
402 Measurements 58 (2013) 128–132.
- 403 [35] R. Brun, F. Rademakers, Nuclear Instruments and Methods in Physics Research Section A:
404 Accelerators, Spectrometers, Detectors and Associated Equipment 389 (1997) 81–86.
- 405 [36] E. R. Siciliano, J. H. Ely, R. T. Kouzes, J. E. Schweppe, D. M. Strachan, S. T. Yokuda,
406 Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers,
407 Detectors and Associated Equipment 594 (2008) 232–243.
- 408 [37] C. Eisenhauer, J. Hunt, R. Schwartz, Radiation Protection Dosimetry 10 (1985) 43–57.
- 409 [38] R. S. Woolf, A. L. Hutcheson, C. Gwon, B. F. Philips, E. A. Wulf, Nuclear Instruments and
410 Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated
411 Equipment 784 (2015) 80–87.
- 412 [39] C.-K. C. Wang, Progress in californium-252 neutron brachytherapy, INTECH Open Access
413 Publisher, 2012.