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Development and characterization of a neutron detector based on a lithium glass–polymer composite

M. Mayer [a], J. Nattress [a], V. Kukharev [a], A. Foster [a], A. Meddeb [a], C. Trivelpiece [b], Z. Ounaies [a], I. Jovanovic [a,*]

[a] Department of Mechanical and Nuclear Engineering, The Pennsylvania State University, University Park, PA 16802, United States
[b] Materials Research Institute, The Pennsylvania State University, University Park, PA 16802, United States

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ABSTRACT

We report on the fabrication and characterization of a neutron scintillation detector based on a Li-glass–polymer composite that utilizes a combination of pulse height and pulse shape discrimination (PSD) to achieve high gamma rejection. In contrast to fast neutron detection in a PSD medium, we combine two scintillating materials that do not possess inherent neutron/gamma PSD properties to achieve effective PSD/pulse height discrimination in a composite material. Unlike recoil-based fast neutron detection, neutron/gamma discrimination can be robust even at low neutron energies due to the high *Q*-value neutron capture on ^{6}Li. A cylindrical detector with a 5.05 cm diameter and 5.08 cm height was fabricated from scintillating 1 mm diameter Li-glass rods and scintillating polyvinyltoluene. The intrinsic efficiency for incident fission neutrons from ^{252}Cf and gamma rejection of the detector were measured to be 0.33% and less than 10^{-8}, respectively. These results demonstrate the high selectivity of the detector for neutrons and provide motivation for prototyping larger detectors optimized for specific applications, such as detection and event-by-event spectrometry of neutrons produced by fission.

1. Introduction

Fast neutron detection can be accomplished by two classes of methods. In the first approach, neutron elastic scattering produces nuclear recoil, resulting in ionization or a combination of ionization and scintillation. In the second approach, neutrons are first thermalized and then captured, whereby energetic charged particles produced from the neutron capture induce ionization or a combination of ionization and scintillation. The neutron is typically thermalized in an organic or water-based moderator, providing a high density of protons that have favorable moderating properties, including low mass, high scattering cross-section, and low absorption cross-section. The use of nuclear recoil also allows measurements of the neutron spectrum, which is accomplished by a process of unfolding using the known, separately measured detector response [1]. Since the unfolding method relies on the entire spectrum, neutron energy cannot be measured on an event-by-event basis. A hybrid approach based on measurement of the energies produced by both thermalization and capture does not exhibit this limitation, and is referred to as capture gating [2]. To date, capture-gated scintillation detectors have relied on the use of a thermalization agent that exhibits pulse discrimination (PSD) properties in order to reject the gammas that usually accompany neutrons. The Saint-Gobain BC-523A, a boron-loaded liquid scintillator, is an example of such capture-gated scintillators [3,4]. This material produces three PSD bands, which allow distinguishing among gammas, neutron scatters, and neutron capture events.

It has been proposed and demonstrated first by Berlman and Marinelli [5] and later by Knoll [6] that anisotropic, composite scintillators can be used to achieve favorable neutron detection properties that are available only in a limited range of isotropic materials. The operational principle of a composite scintillator is based on the combined use of scintillation materials with varying fluorescence decay time constants, large differences of cross-sections for different particle types, and the differences in the typical range of charged particles produced in the neutron and gamma interactions. Individually, each scintillating material does not possess PSD capabilities. In the suitably designed composite, a gamma interacts primarily through Compton scattering, producing recoil electrons, whereas a neutron interacts through a combination of elastic scattering (at high energies) and capture (at thermal energies), producing heavy charged particles with energies determined by the *Q*-value of the neutron capture reaction. The amplitude and the shape of the resulting scintillation pulse are dependent on the location of interaction and the path traversed by the produced charged particle. By constricting geometrical

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* Corresponding author.
E-mail address: ijovanovic@psu.edu (I. Jovanovic).

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dimensions and careful material selection, discrimination between neutrons and gammas can be achieved through both pulse height and shape analysis. Recently, there has been a renewed interest in composite neutron detectors, with several new types of detectors demonstrated using lithium–gadolinium-borate [7] shards, lithium glass [8] cubes, and interlaced polymer scintillator and ^{6}Li doped glass plates [9].

In this work, we make use of lithium-doped glass in a form of thin, long rods as a thermal neutron detector component and scintillating polyvinyltoluene (PVT) as a moderator component of the detector. Lithium-doped glass in the form of rods is commercially available, while the use of PVT permits relatively easy fabrication into a variety of shapes. The thermal neutron is captured in the lithium glass in the reaction

$$^6\mathrm{Li}+\mathrm{n}\rightarrow{}^3\mathrm{H}(2.05\ \mathrm{MeV})+{}^4\mathrm{He}(2.73\ \mathrm{MeV}). \qquad (1)$$

^{6}Li has a high thermal neutron capture cross-section of approximately 940b, with a high Q-value as compared to the competing energy depositions of recoil electrons produced in typical gamma interactions, making it an attractive choice for use in a composite detector. The choice of scintillating PVT allows the thermalization energy of the neutron to be measured, which could facilitate capture-gated fast neutron spectrometry. The relevant results of the detector simulation are provided, followed by the description of detector fabrication and characterization in fast neutron and gamma fields.

2. Simulation

The detection properties of the composite detector were simulated using the Monte Carlo simulation framework Geant4.10.0.p1 [10]. Preliminary simulations focused on geometrical optimization of the detector, specifically by determining the optimal size of the PVT matrix and analyzing the geometrical shape and positioning of the lithium glass [11]. The results indicated that a 127 mm diameter detector thermalizes (below 0.08 eV) approximately 12% of the neutrons with a neutron spectrum characteristic for spontaneous fission of ^{252}Cf that are incident on the detector, and has an intrinsic efficiency of 6%. In conclusion, it was determined that the exact placement of the lithium glass rods when the average Li glass-to-PVT ratio is fixed does not significantly affect the intrinsic efficiency.

For prototyping of the composite detector that employs this general design, a smaller size detector than in Ref. [11] was fabricated and simulated. Specifically, we first simulated a 50 mm diameter by 50 mm high cylindrical PVT detector with 1 mm square rods made of GS20 lithium glass. The rods were aligned parallel to the wall of the detector and placed in a square pattern. Due to physical fabrication restrictions, the rods were not ideally homogeneously distributed. The placement pattern and alignment of the rods can be seen in Fig. 1.

The simulations only took into consideration the deposited energy in the lithium glass, but the production and propagation of optical photons needed to fully simulate the optical pulse shapes incident onto the photomultiplier were not simulated at this time. Future simulations that include optical photons allow more accurate prediction of intrinsic efficiency and neutron/gamma discrimination properties. The simulations used ^{252}Cf fission neutrons and ^{208}Tl gamma rays (2.614 MeV) that were emitted by an isotropic point source located 1.8 m from the detector. Geant4 built-in particle definitions were used, and the energy spectrum of the neutrons was generated using the Watt parametrization within the range of energies of 1 meV–15 MeV [12].

Three different sets of simulations were performed. The first simulation used 10^6 ^{252}Cf fission neutrons and 10^7 ^{208}Tl gamma rays (2.614 MeV) emitted in a collimated beam with the size and shape matching the footprint of the detector, with the detector oriented such that the particles were directly incident upon the entire side wall of the detector from one direction. This allowed for an estimate of the expected intrinsic efficiency. For the subsequent simulations, 3.2×10^8 ^{252}Cf fission neutrons were emitted isotropically. The top of a 15 cm thick concrete slab with surface dimensions of $10\times10\ \mathrm{m}^2$ was placed 130 cm below the bottom of detector and source. The center of the concrete slab was directly below the center of the detector. Air was also introduced into this simulation, as the first simulation was in a vacuum. Since the exact composition of the concrete floor at the laboratory is unknown, the concrete floor was simulated once using the predefined concrete formula in Geant4's material database and the second time using a slightly higher hydrogen concentration to account for uncured concrete and moisture in the ground. Lastly, the simulation was performed again without the concrete floor to allow comparison with the first simulation in which the detector was placed in vacuum.

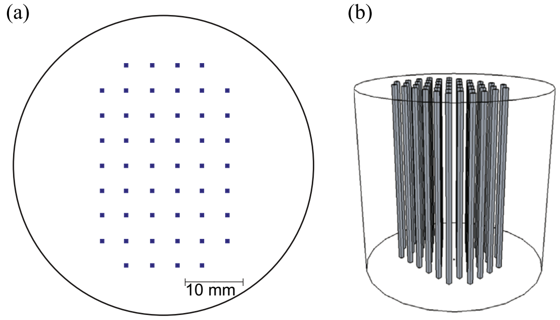

Fig. 1. Simulated detector shape and rod pattern: (a) top view; (b) perspective view.

The deposited energy in the lithium glass was calculated in each simulation. The energy following neutron capture on ^{6}Li is quenched by a factor of 0.35 in Li-glass, calculated from the ratio of electron-equivalent scintillation for neutron capture on ^{6}Li in glass (1.5–1.8 MeV) and the Q-value of neutron capture (4.78 MeV) [13]. For this analysis, the electron-equivalent scintillation yield produced from neutron capture on ^{6}Li-loaded glass is 1.65 MeV. A finite resolution of 10% with a Gaussian shape is fitted to the neutron capture peak to approximately simulate the finite detector resolution. The number of counts within 3σ of the capture peak is counted and divided by the number of particles incident on the detector to provide an intrinsic capture efficiency. For the first simulation, the number of particles incident upon the detector from the source was known, while the number of particles incident upon the detector from the source was calculated from the solid angle for the last simulations. Although a neutron capture may produce higher and lower deposited energies in Li-glass, we conservatively consider only the counts in the Li capture peak for the purpose of estimating the efficiency. Similarly, the number of events resulting from energy depositions of gammas into the same 3σ capture peak applied in the neutron simulation is used to calculate the expected gamma rejection if only the pulse height analysis is used.

The simulated intrinsic efficiency for this detector was calculated to be 0.49% with approximately 0.0033% gammas misidentified as neutrons using only the pulse height rejection criterion. The detector simulation performed with the addition of the concrete floors had a measured efficiency of 1.6% and 5% for a 1% hydrogen concrete floor and 9% hydrogen concrete floor, respectively. The concrete floor was removed for a final simulation using an isotropic source and using the number of particles incident upon the detector from the solid angle. The intrinsic efficiency was again calculated to be 0.49%. As can be concluded from those