Fabrication and characterization of a lithium-glass-based composite neutron detector

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ABSTRACT

A novel composite, scintillating material intended for neutron detection and composed of small (1.5 mm) cubes of KG2-type lithium glass embedded in a matrix of scintillating plastic has been developed in the form of a 2.2 in.-diameter, 3.1 in.-tall cylindrical prototype loaded with $(5.82 \pm 0.02)\%$ lithium glass by mass. The response of the material when exposed to ^{252}Cf fission neutrons and various γ-ray sources has been studied; using the charge-integration method for pulse shape discrimination, good separation between neutron and γ-ray events is observed and intrinsic efficiencies of $(1.15 \pm 0.16) \times 10^{-2}$ and $(2.28 \pm 0.21) \times 10^{-4}$ for ^{252}Cf fission neutrons and ^{60}Co γ rays are obtained; an upper limit for the sensitivity to ^{137}Cs γ rays is determined to be $< 3.70 \times 10^{-8}$. The neutron/γ discrimination capabilities are improved in circumstances when a neutron capture signal in the lithium glass can be detected in coincidence with a preceding elastic scattering event in the plastic scintillator; with this coincidence requirement, the intrinsic efficiency of the prototype detector for ^{60}Co γ rays is $(2.42 \pm 0.61) \times 10^{-6}$ while its intrinsic efficiency for unmoderated ^{252}Cf fission neutrons is $(4.31 \pm 0.59) \times 10^{-3}$. Through use of subregion-integration ratios in addition to the coincidence requirement, the efficiency for γ rays from ^{60}Co is reduced to $(7.15 \pm 4.10) \times 10^{-7}$ while the ^{252}Cf fission neutron efficiency becomes $(2.78 \pm 0.38) \times 10^{-3}$.

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1. The ^{3}He supply problem

The well-documented shortage of ^{3}He [1] has motivated numerous investigations into novel neutron detector technologies which can suitably replace ^{3}He detectors in their many applications. Replacement of ^{3}He-based detection systems is not trivial, however, as they are robust with a very broad application space.

Any competitive replacement must boast several particularly important characteristics: reasonable neutron detection efficiency across a broad range of neutron energies; limited sensitivity to, or the ability to discriminate against, γ rays; and a stable efficiency for neutron detection in a mixed radiation field, where both neutrons and γ rays are present. While boron-lined proportional counting tubes have shown promise [2,3], several groups have been working on novel neutron detection materials with promising recent results, notably Cs_2LiYCl_6 (or simply CLYC) [4] and PSD-

enabled plastic scintillators [5,6]. Other groups have investigated composite scintillators – heterogenous materials composed of neutron-sensitive grains embedded in a supporting plastic matrix – for their potential application in both basic neutron detection [7–9] and capture-gated neutron spectrometry [10,11].

2. Operating principles of composite scintillators

Neutrons incident on a composite detector are intended to interact predominantly through two mechanisms: scattering on nuclei in the supporting, plastic matrix and capture on nuclei in the embedded grains. Though many other design features may vary, the plastic matrix serves as an effective moderator for fast neutrons incident on a composite detector, making such detectors sensitive to a broad range of incident neutron energies without the need for additional moderation [7]. Early composite detector work by Knoll et al. embedded thin-walled glass spheres containing high-pressure ^{3}He in scintillating plastic; neutrons would enter the volume and be captured on ^{3}He nuclei, with the escaping reaction products depositing energy in the scintillating matrix and

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producing detectable signals [8,9]. Numerous recent efforts have focused on composites loaded with the inorganic, neutron-sensitive scintillator lithium gadolinium borate (LGB), which have shown promise as both a neutron detector [7,10–12] and an antineutrino detector [13]; in these composites, the scintillation light produced by neutron capture on any of the constituent nuclei of LGB originates within the embedded grains themselves.

LGB-based composites have been fabricated with both scintillating and non-scintillating matrices, but the use of scintillating plastic can provide a mechanism by which neutron- and γ-ray-generated signals could be distinguished. In the case of these composites with scintillating matrices, neither the embedded LGB nor the plastic matrix has inherent pulse-shape discrimination capabilities; while PSD-capable plastic has been developed [5,6], to our knowledge it has not yet been used as a matrix for composite scintillators. Despite the lack of inherent PSD in either utilized material, the characteristic decay times of the pulses originating in LGB are distinct from those of pulses originating in the plastic scintillator; by virtue of the fact that composite scintillators are largely composed of plastic and that the cross-section for capture of low-energy neutrons on the nuclei in the embedded scintillator is very high, it can be argued that γ-ray pulses have timing characteristics similar to the plastic matrix while neutron-capture pulses have timing characteristics similar to the embedded scintillator [7].

With the successes of composites utilizing LGB, there is reason to explore potential areas for improvement. Though each of the eponymous atomic constituents of LGB has isotopes with large thermal-neutron capture cross-sections, only captures on ^{6}Li result exclusively in charged-particle emission; captures on other isotopes in the LGB can result in γ-ray emission which may be largely indistinguishable from an external γ-ray background. There also exist other scintillators whose index of refraction is better matched to that of plastic scintillator ($n = 1.58$) [14] than LGB ($n = 1.66$) [12]; this mismatch can adversely affect the scintillation-light collection efficiency for larger volumes or for composites with higher concentrations of embedded grains.

3. Design and fabrication of a lithium-glass-based prototype composite

The materials used in the prototype, as well as the geometry of the embedded scintillator grains, were chosen in an effort to maximize neutron sensitivity and minimize sensitivity to γ rays. This was informed and motivated by recent experimental and simulation work by Kazkaz et al., who explored the use of numerous materials as an alternative to LGB as the embedded scintillator [7]. The composite of Ref. [7] used as its matrix EJ-290, a polyvinyl toluene (PVT)-based plastic scintillator from Eljen Technology [14], and this same material was selected as the matrix for the present composite. Numerous materials were considered as candidates to serve as the embedded, neutron-sensitive scintillator. Consideration was also given to the number density of the element on which most neutron captures would occur and the ability to produce the material enriched in the capture isotope. To improve upon the neutron efficiency and PSD capabilities realized with LGB-based composites, it was desirable to select a material with an isotope on which neutron capture results exclusively, or predominantly, in the emission of charged particles; a related concern is that the material possess a minimal number of isotopes on which neutrons are likely to capture and produce signals which are difficult to distinguish from γ-ray backgrounds due to the release of γ rays after capture, effectively competing for neutrons with the isotopes which produce charged particles after capture, thereby potentially reducing both the neutron detection efficiency

and the neutron/γ discrimination capabilities of the composite. Other important factors included: the index of refraction, which should be closely matched to that of plastic scintillator; the light output; the decay time, which is ideally distinct from that of plastic scintillator; and the quenching factor for recoiling nuclei, which determines the electron-equivalent energy of neutron capture signals in the material. An extensive discussion on alternative materials can be found in Ref. [7]. Ultimately, KG2-type lithium glass was selected for the prototype composite for its high atomic fraction of lithium, its enrichment in ^{6}Li, its index of refraction, and its desirable scintillation decay time [15,16].

Successful fabrication of a composite featuring a continuous, even distribution of the embedded scintillator pieces is difficult due to settling of the pieces and formation of bubbles during curing of the plastic matrix. To address the issue of settling, a stratified geometry, where cubes of the scintillator are located only at discrete heights along the axis of the detector, was considered; such a geometry can be fabricated by addition of successive layers of scintillator cubes and uncured EJ-290 on top of previous, semi-cured and nearly-solid layers. Monte Carlo simulations were carried out using the LUXSim [17] front end for GEANT4 [18,19] to compare the intrinsic neutron detection efficiencies of different potential geometries for both thermal and ^{252}Cf-fission-spectrum neutrons: within statistical uncertainties, there was no distinction between the stratified and continuous distributions of embedded scintillator cubes. Simulations comparing the neutron detection efficiencies as a function of the number of layers present in the stratified geometry suggested there was little dependence on this parameter. After a qualitative evaluation balancing the areal density of Li-glass cubes on each layer and the distance between adjacent layers, it was decided that the prototype would be divided into 11 layers. We ignored the effect of optical photon propagation and absorption when determining the optimal number of layers.

In preparation, a large boule of KG2-type lithium glass purchased from Applied Scintillation Technologies [20] was diced into 1.5-mm cubes; these cubes were not polished, though specimens with obvious damage from the machining process were rejected. The total mass of lithium glass added to the sample was 12.14 $\pm$ 0.03 g, divided equally among the 11 layers. The prototype detector was fabricated at Lawrence Livermore National Laboratory (LLNL) over the course of 10 days and was carried out in a glass vessel using an incremental, additive approach. EJ-290 resin, with catalyzing agent added in proportions prescribed by Eljen [21], was added to the vessel and partially polymerized by heating through submersion in an oil bath of temperatures ranging between 54 and 60 °C for between 6 and 12 h. Cubes of lithium glass were then added on top of this partially-cured layer, distributed as evenly as possible across the surface. The polymerization of the underlying layer was sufficient to increase the viscosity to a point where the added cubes remained largely on top of, or very near, the surface; following addition of the glass cubes, another layer of EJ-290 resin was added to the vessel. With the resin added, the fabrication vessel was placed in a desiccator which was subsequently evacuated using a small diaphragm pump. The rough vacuum in the desiccator removed much of the air trapped in the scintillator resin and effectively prevented the permanent formation of bubbles in the prototype as curing took place. Following this evacuation procedure, the top-most scintillator layer was partially polymerized and the steps described here were repeated until the desired number of layers was reached. Throughout the fabrication procedure, dry-nitrogen gas was flowed over the prototype vessel when possible to minimize oxidation of the plastic scintillator.

After the final layer had been added and evacuated, the vessel was moved to a convection oven for final curing at ~70 °C for