Neutron Detector Based on Particles of ^6Li Glass Scintillator Dispersed in Organic Lightguide Matrix

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Abstract

Most ^3He replacement neutron detector technologies today have overlapping neutron-gamma pulse-height distributions, which limits their usefulness and performance. Different techniques are used to mitigate this shortcoming, including Pulse Shape Discrimination (PSD) or threshold settings that suppress all gammas as well as much of the neutrons. As a result, count rates are limited and dead times are high when PSD is used, and the detection efficiency for neutron events is reduced due to the high threshold. This is a problem in most applications where the neutron-gamma separation of ^3He detectors had been essential. This challenge is especially severe for neutron coincidence and multiplicity measurements that have numerous conflicting requirements such as high detection efficiency, short die-away time, short dead time, and high stability. ^6Li -glass scintillators have excellent light output and a single peak distribution, but they are difficult to implement because of their gamma sensitivity. The idea of reducing the gamma sensitivity of ^6Li -glass scintillators by embedding small glass particles in an organic light-guide medium was first presented by L.M. Bollinger in the early 60s but, to the best of our knowledge, has never been reduced to practice. We present a proof of principle detector design and experimental data that develop this concept to a large-area neutron detector. This is achieved by using a multi-component optical medium (^6Li glass particles attached to a glass supporting structure and a mineral oil light guide) with matches the indices of refraction and minimizes the absorption of the 395 nm scintillator light. The detector design comprises a 10" long tube with dual end readout with about 3% volume density of ^6Li glass particles installed. The presented experimental data with various neutron and gamma sources show the desired wide gap between the neutron and gamma pulse height distributions, resulting in a true plateau in the counting characteristics similar to that of ^3He detectors.

Keywords: neutron detectors, ^6Li glass, neutron/gamma separation, ^3He replacement, nuclear safeguards, neutron coincidence counters, active interrogation.

I. Introduction.

The ^3He (and to some extent $^{10}\text{BF}_3$) proportional counters have been established as a gold standard for thermal neutron detectors because of their unique combination of properties: very high efficiency for thermal neutrons, separated neutron and gamma distributions resulting in inherent insensitivity to gain (HV) instability and simple (shaper/discriminator) signal readout. The detection concept of ^3He thermal neutron detectors surrounded by a High Density Polyethylene (HDPE) moderator is the backbone of most fielded neutron measurement systems. Most ^3He replacement neutron detector technologies today have neutron and gamma pulse-height distributions which overlap each other [1]. This complicates their design and limits their performance. Different techniques are used to mitigate this shortcoming, including Pulse Shape Discrimination (PSD) [2] or threshold settings that suppress all gammas as well as many of the neutrons [3,4,5]. As a result, count rates are limited and dead times are high when PSD is used, and the detection efficiency for neutron events is reduced due to the high threshold. This is a problem in most applications where the neutron-gamma separation of ^3He detectors had been essential. This challenge is especially severe for neutron coincidence and Differential Die-Away (DDA) [6] measurement techniques that have numerous conflicting requirements such as high detection efficiency, short die-away time, short dead time, and high stability.

The motivation of this work is to develop a neutron detector, primarily for fission spectrum neutrons, with a combination of properties similar to ^3He tubes embedded in a HDPE moderator but using ^6Li glass scintillator [7] as a thermal neutron detector because of its attractive properties: single peak neutron response (~ 7000 photons/neutron), very good thermal neutron macroscopic cross section (15 cm^{-1}), fast ($\sim 70\text{ ns}$) light decay time [8], and ability to be fabricated in different shapes. The major showstopper of the bulk glass material is its high gamma sensitivity, which leads to overlapping neutron and gamma pulse-height distributions. The classical approach of using ^6Li scintillation-clad fibers did not provide good gamma/neutron separation [9]. The idea of reducing the gamma sensitivity of ^6Li glass scintillators by embedding small glass particles in an organic light-guide medium was first proposed by L.M. Bollinger in the early 60s but, to the best of our knowledge, has never been reduced to practice [10,11]. A similar approach using ^6Li glass scintillator particles embedded in PVTPlastic scintillator was presented at the SORMA XV conference [12].

We present a proof of principle detector design and experimental data that develop this concept into a large-area neutron detector. This is achieved by using multi-component optical medium (^6Li glass particles attached to a glass supporting structure and a mineral oil light guide) with matches the indices of refraction and minimizes the absorption of the 395 nm scintillator light.

The detector design comprises of a 10" long tube with dual end readout and $\sim 3\%$ volume density of ^6Li glass particles installed. The presented experimental data with various neutron and gamma ray sources show the desired wide gap between neutron and gamma pulse-height distributions, resulting in a true plateau in the counting characteristics similar to that of ^3He detectors.

II. Design considerations.

The implementation of Bollinger's idea faces both design (selection of scintillator particle size, particle spacing, organic light-guide materials, detector geometry, and signal readout) as well as fabrication challenges (preparation of scintillator beads or cubes, and creation of a uniform scintillator distribution in the lightguide matrix). The prior work based on two components (^6Li particles and a solid lightguide matrix) did not show promising results because of excessive absorption of the scintillation light by the lightguide matrix. Therefore, we chose a multi-component system: ^6Li glass particles, particle-supporting structure (glass and/or acrylic) and mineral oil with closely matched refractive indices (1.56 for ^6Li glass, 1.505 for PMMA, 1.55 for ^6Li glass, and 1.47 for mineral oil) and low attenuation for scintillation light (395 nm wavelength). The design considerations and selection criteria for the detector components are described below:

A. Size of ^6Li scintillator particles.

The size of each scintillator particle should be small enough to limit the energy deposited by gammas, photo and Compton electrons (Fig. 1, based on data from [13]) to below the energy distribution of ^6Li reaction products (typically a peak at 1.6 MeV_{ee} with a FWHM of 10-14% [7,8]) and therefore provide a gap between the gamma and neutron capture distributions.

{HERE FIG.1}

On the other hand, the lower end of the particle size is limited by the following factors:

- increased light scattering due to imperfect matching of refractive indices increases the path length of the scintillation light and reduces the gap between the neutron and gamma pulse-height distributions
- increased complexity and manufacturing cost of the detector
- increased fraction of incomplete charge deposition for neutron reactions near the surface layer of the glass particles leads to a neutron distribution below the peak of full energy transfer.

The minimal spacing between adjacent glass particles should be large enough to prevent interaction of high energy Compton electrons with more than one glass particle (the rest of the energy is then absorbed by surrounded non-scintillating material).

B. Light transport and selection of non-scintillating (light guide and support) materials

Prior experience with neutron sandwich scintillators, such as a ^6Li foil, indicates non-uniformity of detection efficiency and additional mixing of the already overlapping neutron and gamma ray distribution due to light losses along the scintillator slab [14]. The light losses in a 3D heterogeneous ^6Li glass particles scintillator are proportional to the average light path and the optical attenuation in the materials. The average light path is much longer than the detector geometrical size because of multiple scattering due to the mismatch of the refraction indices and thus the light attenuation due to multiple passes through the material with initially good transparency. Therefore reducing the light scattering and associated light losses during photon transport in the detector is critical for the separation of the neutron and gamma distributions in large scale detectors.

C. Overall detector design considerations

The overall performance of a neutron detector based on ^6Li glass particle scintillator (detection efficiency, gamma sensitivity, dead-time, count rate capability etc.) will depend on selection of size, and spacing (ratio of scintillator versus light guide material) of ^6Li glass particles as well as the detector volume and shape. The possibility for the selection of

scintillator components offers a great level of flexibility to tailor the detector performance to the specific application. As an example we will consider a neutron well coincidence counter that could be a replacement for the ^3He based HLNCC-II detector [15]. The MCNPX modeling of a ^6Li foil sandwich scintillator based coincidence counter (same geometry as the HLNCC-II) containing 133 grams of $50\text{ }\mu\text{m}$ ^6Li foil sandwiched in 15 layers of 6.25 mm light guide strips of PMMA [5] shows about 34% reaction rate efficiency and 16 μs die away time for ^{252}Cf source in the middle of the cavity. A design based on ^6Li glass particles would contain about 850 cm^3 (2100 g) of ^6Li glass (GS20) scintillator uniformly dispersed in the 33 liter detection volume (about 2.8% of detection volume is filled with glass scintillator) and should have similar detection efficiency (the neutrons reaction rate is equal to the count rate for detectors with separate neutron and gamma distributions like ^3He) and faster die-away time. The scintillator to detector volumetric ratio will be used as a guideline for design of our detector prototypes.

III. Proof-of-principle prototype design and experimental results.

We have built and explored two prototypes with different light guide matrices: one based on solid PMMA and the other based on mineral oil.

A. Prototype design based on PMMA as a supporting structure and light guide

The prototype comprises unpolished ^6Li glass cubes (GS20) with 1-mm side (Fig. 2a) inserted in blind holes (at a 0.2" pitch) in $\frac{1}{4}$ " thick $2'' \times 2''$ plates of PMMA. Eight plates were stacked in a square box of PMMA mounted on the top of a 3" diameter PMT (Fig. 2b) forming a $2'' \times 2'' \times 2''$ detector with single end light readout. The internal surface of PMMA box and lid were laminated with light reflective tape. The remaining voids in the detector volume were filled with mineral oil that serves mainly as a light interface between components rather than light-guide/moderator.

{Here Fig.2}

Pulse height spectra using ^{60}Co and ^{232}Th gamma ray sources, and ^{252}Cf neutron source sources were measured with only three PMMA plates to avoid deterioration of the neutron/gamma distribution due to light losses encountered with all eight plates. The pulse height spectra in Fig.

3 show the neutron and gamma separation and the regions of energy deposited by electrons and products of the neutron capture reaction. The neutron distribution is a peak at 1600 keV_{ee} with ~15% FWHM. The gamma distribution appears independent of source energy and was limited below 1000 keV_{ee} which, according to Fig.1, corresponds to about 2 mm electron range in glass and the 1.7 mm main diagonal of cubes with 1 mm size.

{Here Fig.3}

The initial results obtained with this prototype demonstrated good and predictable separation between the neutron and gamma distributions but also revealed substantial attenuation of scintillation light and distortion of the neutron capture peak when all eight plates were used.

B. Prototype design based on mineral oil as light guide

Based on the results of the first prototype, a second prototype was built and tested in which the solid light guide of PMMA was replaced by mineral oil. The size of the ⁶Li glass cubes was increased to 1.5 mm, and a large amount of cubes were diced and packaged in tape and reel for future automated assembly. The cubes were mounted on a support structure consisting of 2.5 mm × 2.5 mm × 250 mm hollow square glass rods to minimize light scattering and attenuation (see Fig. 4). A cylindrical geometry with 7.6 cm diameter by 25 cm length with dual side readout using Hamamatsu R 6263 3" PMTs was selected. The body of the detector was fabricated from Teflon tubing lined inside with a light reflective sheet (see Fig. 5). An additional 25 cm long tube with 2.5 cm ID was fabricated to investigate the light attenuation for a detector with lower light collection solid angle. Based on estimates in Section II.C, five rods with ⁶Li glass particles inserted in a 1" diameter tube or ~45 rods in a 3" diameter tube (about 1 rod per cm²) will have the same ⁶Li volume density as the one modeled in the coincidence counter [5]. The few rods necessary for the measurements with a 1" diameter geometry were assembled manually in-house, whereas an automated assembly using a pick and place machine has been arranged for the 45 rods necessary for the 3" detection geometry. In the present experiments, the 3" configuration was only used to obtain the baseline peak position (channel 706) and the FWHM (11%) from a 6 cm² ⁶Li glass plate placed directly the bottom PMT and covered with

reflective tape for optimum light collection.

{Here Fig.4}

{Here Fig.5}

Most of the measurements were carried out in the 1" tube configuration. A standard spectroscopy setup of charge sensitive preamplifier (Ortec 672 amplifier at 0.5 μ s bipolar shaper and Ortec MCB mod 921) was used. The ^{252}Cf pulse height spectra for one, two, etc. rods installed in the 1" tube were measured for a fixed source position, and the obtained neutron capture peak position and intensity (peak area) as function of number of rods are shown in Fig. 6.

{Here Fig.6}

The data of Fig. 6 show only 20% light attenuation for a five times increases of ^6Li glass content up to the projected 2.7% necessary for a 30% efficient fission neutron detector. The setup with all five rods installed was used to characterize the 1" by 10" long prototype geometry (Fig. 5a) with various sources. The plot in Fig. 8 show the pulse height spectra obtained with a ^{252}Cf source. The 8 cm Pb shielding was used in this measurement to estimate the impact of direct gamma radiation. The right wing of the neutron capture peak is due to non-uniformity of light collection along the detector length (a similar but more severe effect was observed in the 2" $\times$ 2" $\times$ 2" prototype with the PMMA light guide and single ended light readout). The plot in Fig. 8 shows the pulse height spectra of sources with mixed neutron and gamma distribution: a PuO_2 sample (containing mainly ^{239}Pu) and a combination of ^{252}Cf and ^{137}Cs sources.

{Here Fig.7}

{Here Fig.8}

The quality of ^3He -based neutron detection systems is generally judged by the shape of the plateau curve (i.e. the detector counting rate at a fixed discriminator threshold versus applied HV) [15]. Plateau measurements were taken for all four pulse height measurements shown in Fig. 7-8 using a bipolar shaper with fixed discriminator threshold and a Canberra JSR-15 neutron counting system [16]. The normalized plateau characteristics are shown in Fig. 9. The count rate data from 1200 V to 1300 V, which have the same slope, were used for normalization. Unlike most ^3He replacement technologies, this detector shows a high quality plateau with only a small slope after the initial rise. This shows that the vast majority of neutron events are recorded. (The slope on the plateau is caused by neutron events with pulse height below the main peak). There is no gamma contribution until the gain is increased considerably, therefore there is a wide interval over which the neutron count rate is not very sensitive to HV (gain), indicating that the detector should have good stability.

{Here Fig.9}

IV. Discussion

The pulse height spectra obtained with the 1" diameter 10" long geometry make us believe that a detector module with 3" diameter and 20" length would have at least the same, if not better, neutron to gamma separation, allowing the device to be scaled up for practical applications. Reducing the concentration of ^6Li glass cubes would reduce the specific neutron absorption efficiency but will allow for a larger geometrical size. The dependence of light intensity at both ends of the detector can be used both for crude position information of neutron interaction as well as to correct amplitude signal dependence due to light losses along the slab.

The effective light decay time constant of ^6Li glass is ~ 70 ns, therefore a symmetrical bipolar shaper with a time constant of a similar value would allow the collection of a substantial fraction of ~ 2000 photo-electrons corresponding to a single neutron capture in the ^6Li scintillator. This short time constant and a symmetrical bipolar shaper will allow the system to

perform a t h i g h c o u n t i n g r a t e s w h i l s t m i n i m i z i n g b o t h d e a d t i m e a n d g a m m a p u l s e p i l e - u p (gamma pulses exceeding the neutron threshold). In case of very high gamma background there will be multiple Compton scattering events that will be treated as single pulse. In this case, a reduction of scintillator particle size can be used to increase the gamma tolerance.

The fabrication, using automated assembly of rods with ^6Li glass particles (see Fig. 4), of a 3" by 10" long prototype is in progress. We intend to carry out comprehensive light transport and neutron transport modeling on this device. In addition to tests targeted towards neutron multiplicity counting, we will also be testing a device with a pulsed neutron source in order to confirm its suitability for active interrogation applications.

V. Conclusions

We have built and tested a heterogeneous detector prototype based on ^6Li glass particles dispersed in an organic lightguide matrix. The experimental results show very good separation between the neutron and gamma distributions, leading to a plateau characteristic similar to that of a ^3He detector. The demonstrated properties of the device indicate that it could be not only a practical replacement for ^3He detectors in many common applications but may also have equivalent or even superior performance. For example, the detection module could be scaled up to make a neutron coincidence well-counter with a performance better than HLNCC-II. The efficiency and die-away time would be improved, and the dead time would be much lower. In addition, the device has potential for optimization for particular applications such as the ability to tolerate very high gamma ray count rates by reducing the size of the particles.

Acknowledgments

This work was supported by the Office of Defense Nuclear Nonproliferation Research & Development (DNN R&D), National Nuclear Security Administration, US Department of Energy. The authors are indebted to Dr. C. Hurlbut (Eljen Technology) for enlightening discussions.

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FIGURE CAPTIONS

Fig. 1: Electron range versus deposited energy. For PMMA data of polystyrene

Fig. 2: (a) Unpolished ^6Li glass particles inserted in a 2"by2" plate of PMMA; (b) top view of 2"× 2"×2" detector assembly filled with mineral oil (assembly lid removed).

Fig. 3: Neutron and gamma pulse height spectra obtained from ^6Li glass in PMMA with different gamma energies and intensities.

Fig. 4: Hollow glass rods containing about 200 GS20 glass cubes installed 6 mm apart.

Fig. 5: (a) Detector assembly on vertical stand; (b) top view of 1" diameter tube with 5 rods inserted in 3" diameter tube (PMT removed).

Fig. 6: Neutron capture peak position and intensity as function of number of rods.

Fig. 7: ^{252}Cf pulse height spectra: Fig.7(a) with lead gamma shield, and Fig.7(b) without lead shield. The spectra are normalized to the neutron peak to show the difference in the gamma pulse-height distribution.

Fig. 8: Pulse height spectra of a PuO_2 sample (Fig.8(a)) and of a combination of ^{252}Cf & ^{137}Cs sources (Fig.8(b)). The spectra are normalized to the neutron peak to show the difference in the gamma pulse-height distribution.

Fig. 9: Normalized plateau characteristics with different neutron and gamma sources.

Fig.1

Fig.2

(a)

(b)

Fig.3

Fig.4

Fig.5

(a)

(b)

Fig.6

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Fig.7(a)

Fig.7(b)

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461 Fig.8

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Fig.8(a)

Fig.8(b)

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Fig.9