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Changing the World's Energy Future

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Advanced Inorganic Fluors in the Scintillation Hydro-Gel for Isotopic Neutron Emitters (SHINE) Neutron Detection Material for Nuclear Reactor Monitoring Systems

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NARRATIVE

Currently, the market for neutron detectors is an estimated $583.80 million for 2017, with expected compound annual growth at a rate of 11.25% during the next forecast period, i.e., 2018-2023[1]. With an ever-growing need for neutron detectors, there is an outstanding market potential for new technologies. Scintillation Hydrogel for Isotopic Neutron Emitters (SHINE) is an alternative neutron detection material that addresses the current He-3 shortage for neutron detection[2]. SHINE is an environmentally friendly, first-of-its-kind scintillation gel capable of detecting thermal neutrons that can be poured into any shape at room temperature. SHINE converts neutrons to charged particles, which then excite quantum dots (QD) to release photons. Lithium-6 (6Li), in the form of lithium chloride (LiCl), has a high neutron capture cross-section and is highly soluble in water which increases Li loading. The indium phosphate (InP) QD component used in the SHINE material are wavelength-tunable and have a high light yield due to greater surface area. The QD core/shell structures have a semiconductor core and are passivated by a shell of a wider bandgap semiconductor such as zinc sulfide (ZnS) which prevents self-absorption of light thereby improving light yield over current neutron detection materials.

SIGNIFICANCE

The SHINE scintillation material for neutron detection is a significant improvement over current state-of-the-art technologies, and it will be the frontrunner for the next generation of scintillation detection. These scintillation gels incorporate the best properties of plastic and liquid scintillators without the disadvantages. For solid scintillators, such as plastics, the disadvantages include reduction of detector efficiency due to lithium-6 (^{6}Li)“dead areas” in solids and fewer neutrons generated for capture. The disadvantages of liquids include the use of toxic chemicals and the potential for spills. Liquids are also high maintenance because it is difficult to keep liquid scintillator clean. For crystals, disadvantages include lengthy fabrication times, high costs, and the fragility of the crystals. By using a novel scintillation gel, the disadvantages of other materials can be mitigated while retaining equivalent detection of special nuclear material (SNM) using a low-cost, stable, non-regulated, and environmentally-friendly material. Neutrons have mass but no electrical charge. Therefore, neutrons cannot directly produce ionization in a detector, meaning they cannot be directly detected. Neutron detectors must rely upon a conversion process where an incident neutron interacts with a nucleus to produce a secondary-charged particle. These charged particles are then directly detected, and, from them, the presence of neutrons is deduced. Based on previous proof-of-concept and current work, SHINE technology was developed using a unique combination of polyacrylamide, water-soluble lithium-chloride (^{6}LiCl), and new fluorescent compounds including nanoparticles. The nanoparticles consist of water-soluble zinc sulfide (ZnS) core-shells, as well as indium-phosphate/zinc-sulfide (InP/ZnS) nanoparticles incorporated into the gel, which can fluoresce in the presence of energy signatures such as gamma rays.

The SHINE technology has been tested for neutron detection using a gamma source, cesium-137 (^{137}Cs) and a californium-252 (^{252}Cf) source (neutron).

EXPERIMENTAL

Experimental procedures began with 5 g enriched lithium chloride (LiCl) dissolved in 6 ml water and left overnight to dissolve completely. Once transparency (79% transmission at 450 nm wavelength) was achieved after 24 hours, 6 ml of bis-acrylamide monomer was poured into the solution, along with 0.5mL of 10% ammonium persulfate. Water-soluble cadmium selenide capped with zinc sulfide (CdSe/ZnS) quantum dots (QD) were poured into the monomer solution, and Tetramethylethylenediamine (TEMED) was added to polymerize the solution into gel form. The final form of the gel is shown in Figure 1 (Appendix 1). The detection system for the new scintillation hydrogel material uses lithium-6 (^{6}Li) which has a large cross section for thermal neutron capture. High loadings of ^{6}Li can be achieved by using enriched LiCl, which is highly soluble in water (84.25 g of LiCl can dissolve in 100 mL of water at 25 °C). Since the gel is water-based, ^{6}LiCl is the perfect candidate for maximizing ^{6}Li content. The amount of ^{6}LiCl and water was determined by optimizing the following three factors: maximizing ^{6}Li weight %, minimizing water content, and maximizing optical transmission at 450 nm (nanometers). The ^{6}Li(n,α) reaction has a Q-value of 4.78 MeV and produces a 2.05 MeV alpha particle and a 2.73 MeV triton upon interacting with a thermal neutron. The light yield of the triton is about 5 times greater than that of the alpha particle, but both can excite water-soluble CdSe/ZnS quantum dots with carboxylic acid ligands to produce scintillation light peaking at 450nm. If this scintillation light is collected by a photodetector such as a photomultiplier tube (PMT), information regarding the radiation event can be obtained[3]. The water content is minimized, because the water itself can perform as a separate detector—producing Ĉerenkov scintillation light. Before loading the gel with LiCl or QDs, an "empty" gel was fabricated that was composed of mostly water and the polymerized monomer, and the additional catalysts. Shown in Figure 2 (Appendix 1), a 48-hour (black) and 10-min (red) measurement of the empty gel spectra without any sources present was performed. A noticeable "hump" is observed at approximately channel 400nm, but for only the 48-hour measurement. The hump was theorized to have originated from muons interacting with the water in the gel, and since mons are rare events, they were only noticeable on the longer 48-hour measurement.

The ^{6}LiCl + CdSe/ZnS gel was wrapped in Teflon and coupled to the PMT window using optical glue. A measurement was run without any source, and a noticeable bump was once again observed. Then, a cesium-137 (^{137}Cs) source (whose activity matched the gamma-ray flux of the californium-252 (^{252}Cf) source) was used to irradiate the gel. This was performed to determine the gamma-ray event contribution to the ^{252}Cf spectra, and to set a lower limit detection (LLD) to reduce background noise and gamma radiation. Before irradiating the gel with ^{252}Cf, the PMT (without gel) was irradiated with ^{252}Cf to determine the Ĉerenkov contribution that originated from gamma-rays and neutrons interacting with the PMT window. This contribution was quite significant. Finally, upon irradiation of the gel with the ^{252}Cf, a higher count rate than the ^{252}Cf + PMT measurement was observed above LLD 400nm (determined by the ^{137}Cs + gel measurement). The count rates for each of the measurements are given Figure 3 (Appendix 1). Interestingly, between channels 0 and 100, the count rate for the ^{252}Cf + PMT is higher than that of the ^{252}Cf + gel. This is due to the gel acting as a shield for the neutrons and gamma rays, slowing down their speed and reducing their energies. Above LLD 400nm, the count rate for the ^{252}Cf + gel is higher than that of the ^{252}Cf + PMT, indicating additional scintillation events over Ĉerenkov scintillation events. To verify the two separate events, an oscilloscope was used to record the waveforms exiting the PMT directly before undergoing amplification and pulse shaping. Waveforms upon irradiating the gel with ^{252}Cf had three exponential decay times, the shortest being 23.5 μs (microseconds), and the longest being 512 μs (Figure 3). Waveforms upon irradiating the PMT with ^{252}Cf had only one exponential decay time of 20 ns (Figure 3). The waveforms with the gel irradiation, Figure 4, had much longer decay times than the PMT irradiation, validating the separate events in both the cases. The results indicate a need for performing pulse shape discrimination for distinguishing between the two events, as an LLD might not be sufficient to separate the two events and might also lead to a lowered count rate.

Indium-phosphate/zinc-sulfide (InP/ZnS) QDs were tested since they are environmentally friendly and with a large stokes shift which prevents self-absorption of light. Light yield in the InP/ZnS QDs is also improved due to the semiconductor core of InP that is passivated by a shell of the wider band-gap found in the ZnS. The InP/ZnS showed good water solubility and along with ^{6}LiCl which allowed for high loading and a good gamma ray detection response, Figure 5. Further studies are needed to evaluate pulse shape discrimination for gamma ray rejections and polymer gel radiolytic stability over time.

References

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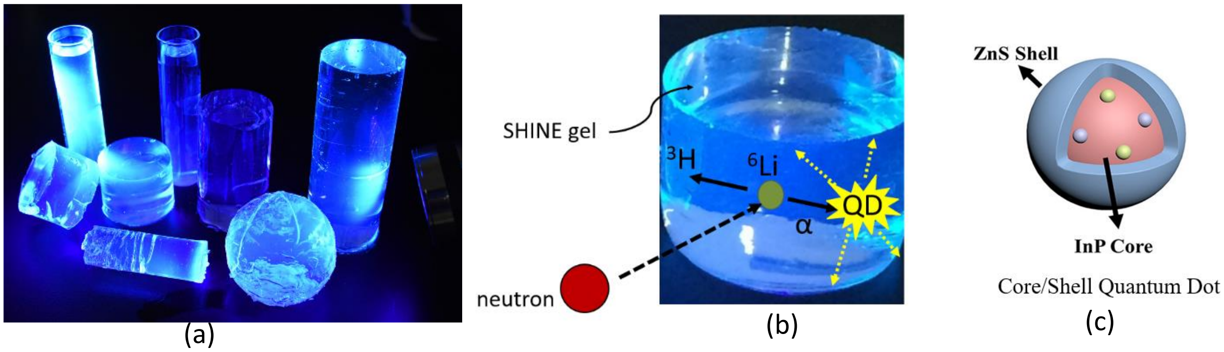
2) Zeitelhack, K., Search for alternative techniques to helium-3 based detectors for neutron scattering applications, Neutron News, 23:4, 10-13, DOI 10.1080/10448632.2012.725325, (2012).
3) Ghosh, P., et al, A high-efficiency, low-Cerenkov Micro-Layered Fast-Neutron Detector for ˆ the TREAT hodoscope, Nuclear Inst. and Methods in Physics Research, A (2018).

APPENDIX 1.



(a) (b) (c)

Figure 1. a) The SHINE scintillation gel can be poured into any shape and solidifies in minutes at room temperature. b) Schematic of the Li-6/QD (quantum dot) nanoparticle function within the hydrogel. c) the InP/ZnS nanoparticle core.

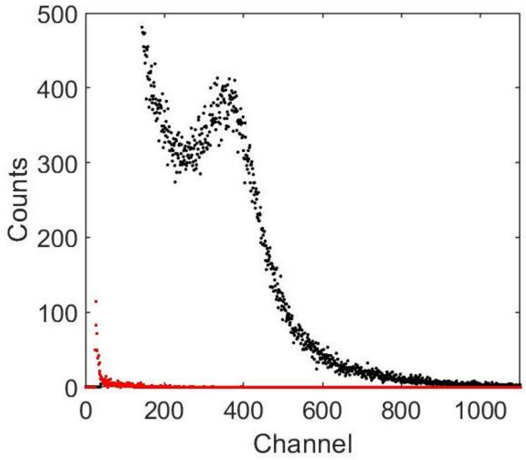
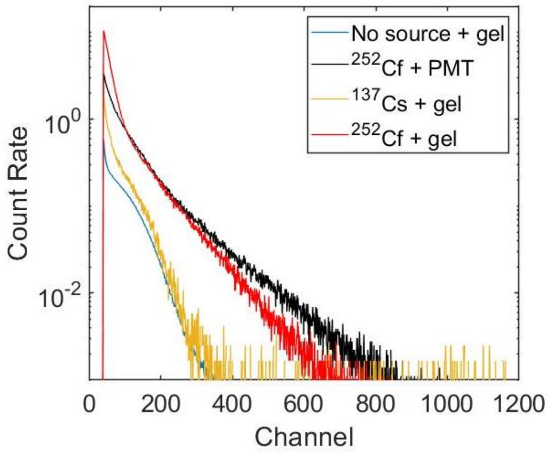


Figure 2. A 48-hour (black) and 10-min (red) measurement of the polymer gel (no nanoparticles present) spectra without any sources present.



Measurement	Count Rate
No source + gel	0.48 ± 0.003
^{137}Cs + gel	0.52 ± 0.021
^{252}Cf + PMT	1.74 ± 0.022
^{252}Cf + gel	3.66 ± 0.031

Figure 3. Count rates for ^{6}LiCl + CdSe/ZnS polymer gels. In all cases of using the ^{252}Cf, both a bismuth shield and polyethylene moderator was used, while for the ^{137}Cs, the same bismuth shield was used.

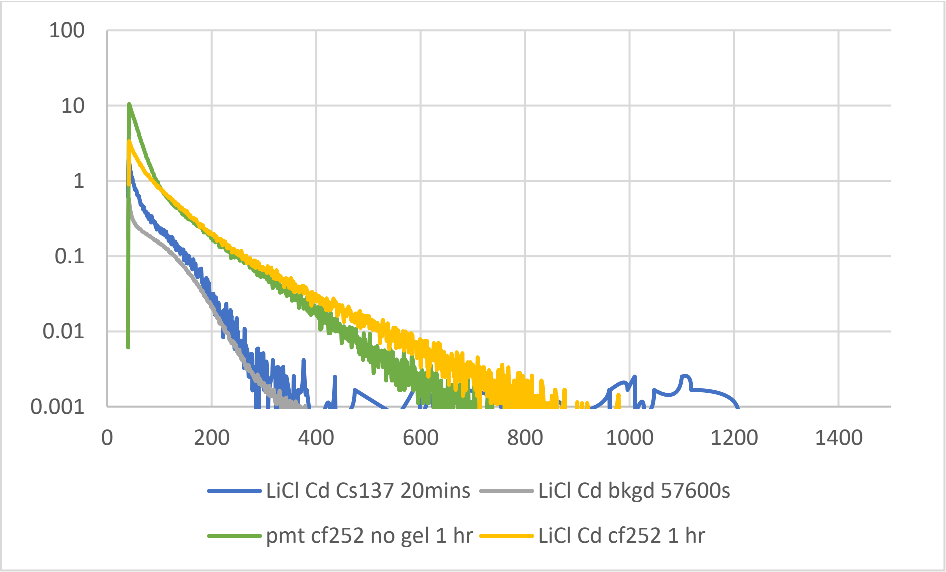


Figure 4. Spectra counts for LiCl/Cd using a gamma source (^{137}Cs) and neutron source (^{252}Cf) for 1hr and 20min count times, respectively.

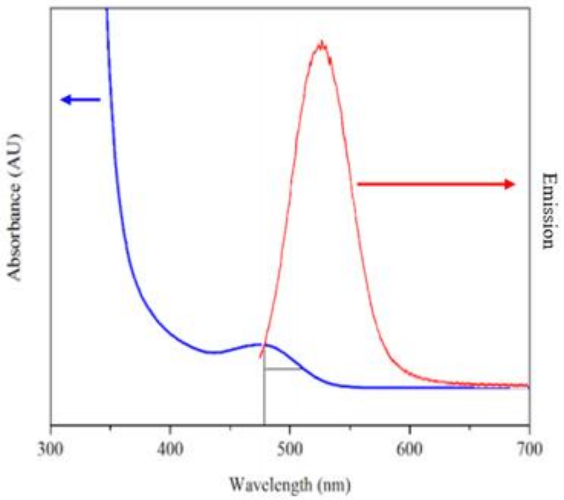


Figure 5. InP/ZnS emission, absorption spectra.