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Procedia**www.elsevier.com/locate/procedia**Gadolinium loaded plastic scintillators for high efficiency neutron detection**Lena Ovechkina¹, Kent Riley¹, Stuart Miller¹, Zane Bell² and Vivek Nagarkar¹

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Keywords: plastic scintillators, neutron detectors, gadolinium**Abstract**

Gadolinium has the highest thermal neutron absorption cross section of any naturally occurring element, and emits conversion electrons as well as atomic X-rays in over 50% of its neutron captures, which makes it a useful dopant in scintillators for detecting thermal neutrons. Gadolinium isopropoxide was studied as a possible dopant for styrene-based plastic scintillators as a convenient and inexpensive method to produce high-efficiency thermal neutron detectors. Plastic scintillators with gadolinium weight concentrations of up to 3% were transparent, uniform and defect-free and were characterized with spectral measurements performed under x-ray and neutron irradiation. The new material has the same characteristic emission of styrene with a maximum at approximately 425 nm, and a light output of 76% relative to the undoped plastic. A 13 mm thick sample containing 0.5% gadolinium by weight detected 46% of incident thermal neutrons, which makes this an attractive material for a variety of applications.

Introduction

Gadolinium is a naturally occurring element which has two isotopes with very high thermal neutron absorption cross-sections: Gd¹⁵⁵ (14.7% natural abundance) and Gd¹⁵⁷ (15.7% natural

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abundance) with 6.1×10^4 b and 2.6×10^5 b, respectively. These cross sections are significantly higher than nearly every other isotope, which makes gadolinium an attractive dopant for a variety of neutron detectors. Thermal neutron absorption in gadolinium produces low energy internal conversion electrons as well as a cascade of associated Auger electrons, x-rays and gamma rays ranging in energy from a few eV to several MeV. These radiations are readily absorbed in plastic and will produce significant light output in a suitable scintillating material. Gadolinium doped plastic scintillators are therefore of great interest as inexpensive, efficient and versatile neutron detectors that could be fashioned in small arrays with high spatial resolution.

The concept of using a digital readout such as a CCD in combination with a neutron imaging scintillator is established, but few scintillators suitable for this task are available and further research is needed to develop practically useful devices [1]. The most widely used scintillator types in neutron radiography/diffraction studies are ${}^6\text{LiF}+\text{ZnS}(\text{Ag})$ and $\text{Gd}_2\text{O}_3\text{S}$ (GOS) [2, 3] but these often do not have the desired neutron detection efficiency and are sensitive to photons. Making the scintillator thicker, for higher efficiency, is not practical, because these scintillators are used as powder screens and therefore scatter scintillation light, resulting in significantly reduced spatial resolution. Also, these materials have limited optical transparency for their own light, and when combined with the optical scattering it causes a significant reduction in the light transmission to the underlying detector. Thicker scintillators are also problematic because of their enhanced gamma sensitivity.

Scintillating crystalline materials, such as $\text{GdI}_3:\text{Ce}$ [4] and $\text{Cs}_2 \text{LiYCl}_6:\text{Ce}$ (CLYC) [5] have excellent sensitivity to neutrons but are also sensitive to gamma rays. Materials such as CLYC can discriminate between neutrons and gamma rays based on differences in pulse shape. Unfortunately, due to the complex nature of CLYC, at present it cannot be synthesized in a convenient fashion that would make it suitable for a wide variety of applications.

Plastic scintillators are not very sensitive to low energy neutrons, because they do not produce enough light by the proton recoil mechanism responsible for neutron detection. Thin plastic scintillators are also relatively insensitive to photons as they are low density and contain low Z elements for which the photon interaction probabilities are smaller. Loading the plastic with additives having a high thermal neutron absorption cross-sections may therefore be an effective method to produce inexpensive and efficient thermal neutron detectors with low gamma-ray sensitivity that can be produced in large areas or complex arrays.. The feasibility of this approach was investigated using common polymers and luminescent additives together with gadolinium isopropoxide, which is commercially available and has not previously been used in this application.

Materials and Methods

Gadolinium-loaded polystyrene-based plastic scintillators were produced using a thermal polymerization. Liquid styrene (99.5%-Alfa Aesar) was mixed with concentrations of 1% 2,5-diphenyloxazole (PPO) and 0.1% 4-bis(5-phenyl-2-oxazole) (POPOP) additives (99%-Sigma Aldrich) that were optimized to maximize light yield under x-ray irradiation. Gadolinium isopropoxide $\text{Gd}[\text{OCH}(\text{CH}_3)_2]_3$ was added as a neutron-sensitive dopant in concentrations ranging from 0 to 6.4% which yielded corresponding gadolinium weight concentrations of 0 to 3%. Polymerization of the doped styrene solution was performed at 125-140 °C in an Argon atmosphere over several days without a catalyst. Samples were polymerized in glass vials having an inner diameter of 14 mm, and were prepared for measurements by cutting and polishing into cylinders with thickness ranging from 3-13 mm.

The emission spectra of 3 mm thick plastic scintillators containing different concentrations of gadolinium were measured using 8 keV Cu K_α x-ray excitation and a calibrated monochromator. The relative light output of these samples was determined from the position of the 59.7 keV (^{241}Am) photopeak in the pulse height spectrum generated using pulse-processing electronics

modules with the detector coupled to a Photonis XP2203 photomultiplier tube. The light output in terms of photons/MeV was determined from similar measurements using a comparably sized bismuth germanate oxide (BGO) crystal for which the absolute light output is known.

Neutron irradiations were performed using the same pulse processing electronics and a ^{252}Cf spontaneous fission source encapsulated in a 20 cm diameter polyethylene cylinder that results in a thermal spectrum of low energy neutrons. The source may be shielded with 5mm of boron-loaded flexible polymer.

Results

The mixing and curing process produced solid, colorless cylindrical plastic scintillators pictured in Figure 1 that are free of voids and obvious visual defects. Two out of nine samples containing gadolinium were cloudy or only semitransparent, but this effect did not correlate with increased gadolinium concentration and may have instead been related to different batches of gadolinium isopropoxide that were used.

Emission intensity is plotted as a function of photon wavelength in Figure 2 for an undoped plastic scintillator as well as three different gadolinium weight concentrations. The gadolinium dopant has no observable effect on the spectral emission characteristics of the plastic, which has a maximum in the blue region at approximately 430 nm. Trace concentrations of gadolinium up to approximately 0.5% seem to exhibit stronger luminescence, but this may result from increased absorption in the high-Z gadolinium metal. This effect is illustrated in the pulse height spectra plotted in Figure 3 where the total area under the curve increases with increasing gadolinium concentration due to greater absorption and the 59.7 keV photopeak moves gradually to the left, indicating lower light output. Increasing the gadolinium concentration beyond 0.5% appears to reduce light output as quenching processes begin to overcome the effects of increased absorption. The light output for plastics steadily decreases with increasing gadolinium concentration as illustrated in Table 1 where light output determined from the

spectra in Figure 3 is given relative to the undoped plastic as well as a BGO specimen with known light output. The measured light output for the undoped plastic scintillator is consistent with published values for similar organic scintillators (6) and the reduced brightness in the most highly doped sample is still comparable to BGO which emits 8,200 photons/MeV.

Pulse height spectra are plotted in Figure 4 that were measured using a 6 mm thick scintillator containing 2.5% gadolinium and a thermalized ^{252}Cf source with and without neutron-absorbing shielding. A broad peak corresponding to neutron capture in gadolinium is evident during neutron irradiation that is reduced when the source is shielded and cannot be discerned amongst the photon background. Lower channels in the spectrum also exhibit increased activity during neutron irradiation that may be associated with reaction products from ^{155}Gd which has a lower neutron absorption cross section than ^{157}Gd or recoil protons created by fast neutrons that elastically scatter with hydrogen contained in the polymer. The intrinsic detection efficiency of a 13 mm thick scintillator containing 0.5% gadolinium was 46% based upon an independent measurement of the neutron source intensity.

Conclusions

The metallo-organic compound gadolinium isopropoxide can be used as an additive to synthesize polystyrene-based plastic scintillators. Gadolinium-isopropoxide is soluble in polystyrene up to at least 6.4% which results in 3% gadolinium by weight. The relative light output for these plastic scintillators is not seriously degraded by gadolinium, measuring 76% for 3% gadolinium by weight. A 13 mm thick scintillator loaded with 0.5% Gd detects approximately 46% of incident thermal neutrons.

The achieved concentrations of gadolinium are high enough to attain desirable neutron detection efficiencies in plastic scintillators that are only a few millimeters thick. Parameters such as detector thickness and gadolinium concentration may be adjusted to minimize the photon sensitivity of these scintillators relative to the desired neutron signal. Perhaps most

importantly, these plastics are inexpensive, readily available and may be configured in a variety of complex shapes, including micro-capillary arrays which are useful for attaining high spatial resolution.

Acknowledgements

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| Gadolinium content (wt. %) | relative to undoped plastic | Light output | |
|-------------------------------|--------------------------------|-----------------|-------------|
| | | relative to BGO | Photons/MeV |
| 0.0 | 1 | 1.43 | 11,800 |
| 0.5 | 0.93 | 1.34 | 10,980 |
| 1.0 | 0.86 | 1.23 | 10,150 |
| 2.5 | 0.82 | 1.18 | 9,680 |
| 3.0 | 0.75 | 1.08 | 8,850 |

Table 1. Summary of light output for gadolinium-containing scintillating plastic measured relative to undoped plastic as well as BGO with a known light output.

Figure Captions

Figure 1. Photograph of plastic scintillator samples doped with 0, 0.5 and 1.0% gadolinium by weight.

Figure 2: Emission spectra of styrene-based scintillators containing weight concentrations of gadolinium ranging from 0 – 3%.

Figure 3: Pulse height spectra of ^{241}Am using styrene-based scintillators with weight concentrations of gadolinium ranging from 0 – 3%.

Figure 4: Pulse height spectra of a shielded and unshielded thermalized ^{252}Cf source measured using a 6 mm thick scintillator containing 2.5% gadolinium by weight.

Figures

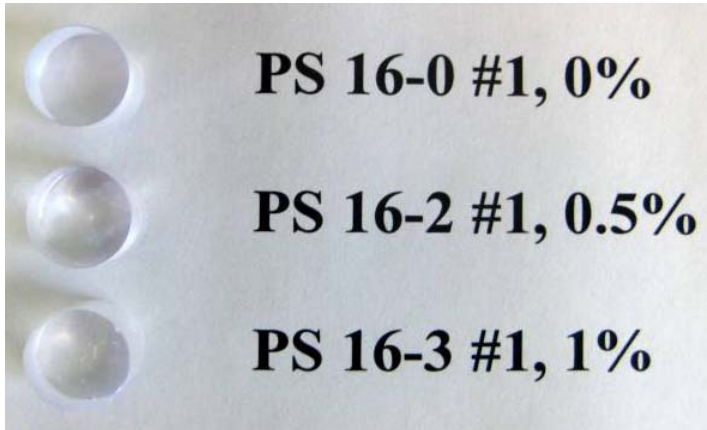


Figure 1

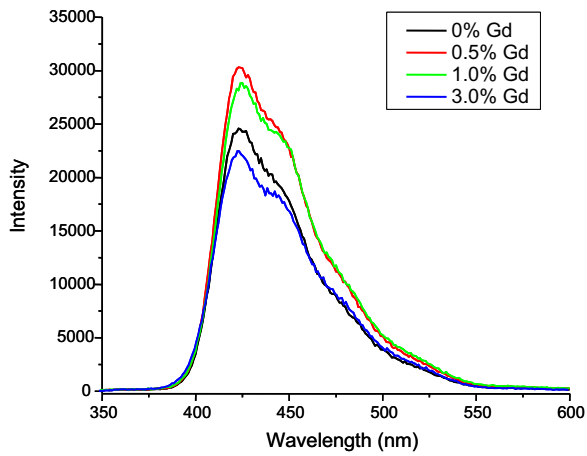


Figure 2

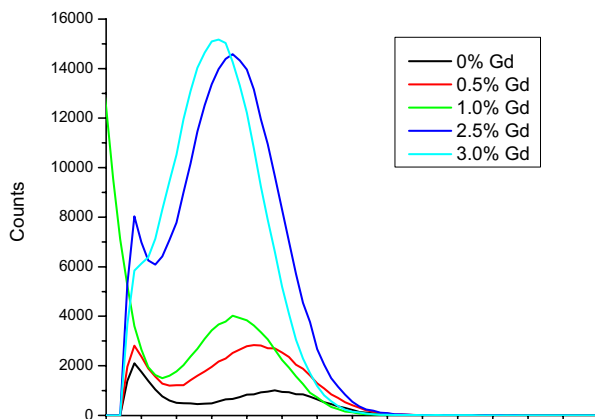


Figure 3

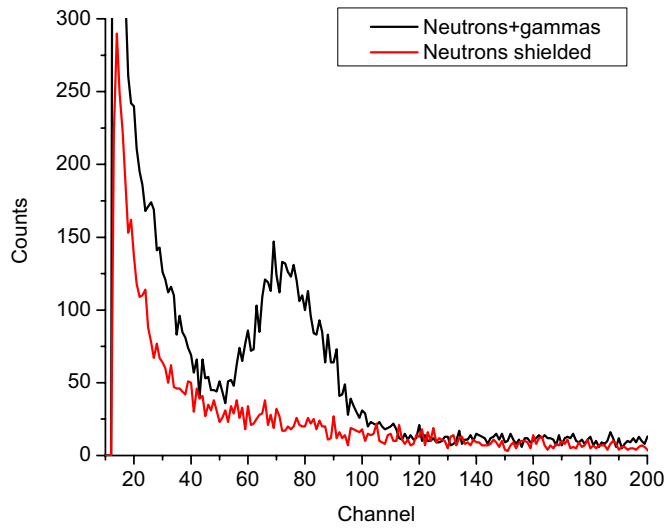


Figure 4