Transparent lithium loaded plastic scintillators for thermal neutron detection

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ABSTRACT

The fabrication of a series of novel, optically transparent, bulk plastic scintillators loaded with lithium methacrylate, and incorporating 2,5-diphenyloxazole and 5-phenyl-2-[4-(5-phenyl-1,3-oxazol-2-yl)phenyl]-1,3-oxazole fluorescent centres, is described. The attenuation length, photoluminescence, and both gamma ray and thermal neutron scintillation responses were compared over a range of lithium methacrylate concentrations. The maximum concentration corresponded to a weight percentage of lithium-6 of 0.63%. The photoluminescence shows a composite 2,5-diphenyloxazole and 5-phenyl-2-[4-(5-phenyl-1,3-oxazol-2-yl)phenyl]-1,3-oxazole broad band with vibronic features in the range 350–500 nm, and lifetimes in the range 0.9–2.7 ns. An increasing luminescence in a thermal neutron beam with increasing lithium-6 content is demonstrated.

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1. Introduction

Due to their comparatively low cost, ability to be machined into complex shapes and fast response times, plastic scintillators have found application in a wide variety of photon and particle detection applications [1]. Therefore, methods to fabricate these scintillators, especially loading with other elements that enhance their performance are of considerable interest. For example, low resolution plastic scintillators loaded with bismuth or lead complement higher cost medium resolution lanthanum halide and sodium iodide detectors [2]. Commercially available scintillators loaded with boron-10, gadolinium, lead and lithium-6 to increase the thermal neutron detection efficiency are available [3]. However, many of these commercially available scintillators are either liquids [4] or glasses [5] which may not offer all of the advantages of plastic scintillators. Plastic scintillators have very fast decay times (e.g. 5 ns) and thus exhibit high count-rate capabilities (e.g. 10^6 per second) [6]. The process of loading plastic scintillators with specific elements is limited by a reduced light output that often limits the elemental concentrations that can be achieved.

Although lithium-6 has a high thermal neutron absorption cross-section, it has proven difficult to load into plastic scintillators. It is an objective of this paper to describe the performance of transparent polystyrene-based plastic scintillators loaded with

lithium-6. Currently lithium-6 containing scintillators are commonly glasses [5] or inorganic crystals and lithium compounds dispersed in phosphor powders [3]. The topic of transparent plastic bulk scintillators containing lithium is relatively unexplored; previous reports of the incorporation of lithium into plastic organic scintillators mainly focus on the mixing of inorganic salts and nanoparticles such as lithium fluoride [7,8], lithium chloride [9] or lithium phosphate [10] into a polymer which results in a heterogeneous material with reduced transparency. A paper relating to a transparent scintillator containing lithium by Negina et al. [11] gives no details of the scintillator's preparation and composition and is therefore difficult to compare to this current work.

Thus there have been only a few reports of promising lithium-loaded plastic scintillators for thermal neutron detection. Yet achieving loading with lithium-6 may offer advantages over boron-10 loaded scintillators, as discussed in a recent report on lithium loaded liquid scintillators by Bass et al. [12]. For example, a feature of the lithium-6 reaction is its comparatively high Q-value (4.78 MeV compared with 2.79 MeV for the boron-10 reaction). Unlike the boron-10 reaction, there is an absence of gamma emissions associated with the charged particle reaction products. The products of both reactions are otherwise short range energetic charged particles, enabling use of small detectors without significant edge effects [2]. In the case of thermal neutron absorption by lithium-6 the product triton and alpha particle are emitted with energies of 2.73 MeV and 2.05 MeV respectively, for a combined deposited energy of 4.78 MeV. In the case of the boron-10 reaction, the product lithium-7 nucleus is produced 94% of the time in an excited state emitting a 478 keV gamma ray

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which often escapes the detector and is thus not detected but may contaminate the detection environment. Without detection of the gamma ray, the combined particle detected energy from the alpha particle (1.47 MeV) and the lithium ion (0.84 MeV) is 2.31 MeV and is thus considerably lower than 4.78 MeV obtained from the lithium-6 reaction. However, in practice the performance of the scintillator depends on the cross section for the neutron absorbing reaction, the percentage elemental loading that can be achieved, and the comparative light outputs of lithium loaded and boron loaded plastic scintillators. Some light absorption in lithium loading of organic scintillators at 0.15 wt% level and a relatively low neutron detection efficiency compared to boron loaded organic scintillators have been noted [2]. Historically usage has strongly favoured the boron loaded scintillators which are commercially available from Saint Gobain and Eljen Technology [3]. Improvement of the lithium loaded plastic scintillators may lead to a commercially competitive product whereby the higher reaction Q-value and the absence of gamma emission may be utilised.

Here we have studied the incorporation of lithium-6 into bulk plastic polystyrene scintillators using new lithium containing compounds to give transparent scintillators. We report a method of preparing scintillators containing lithium which utilises a solution of a lithium complex which fully dissolves in the precursor monomer solution. This mixture can then be poly-merised giving a transparent scintillator. Our investigation clearly shows an increase in light output with increases in the lithium-6 concentration when the scintillator is placed in a thermal neutron beam.

2. Preparation of scintillators

The main goal of our research was to produce a transparent plastic scintillator loaded with lithium to act as a detector of thermal neutrons. Upon designing this scintillator we needed to incorporate the components of a conventional plastic scintillator into our material so that the products of the lithium neutron capture reaction could be converted to light, which is able to be detected by a photomultiplier tube. Styrene was chosen as the aromatic base material for our scintillator as it is cheap and has been extensively studied, as have the primary and secondary shifters 2,5-diphenyloxazole (PPO) and 5-phenyl-2-[4-(5-phenyl-1,3-oxazol-2-yl)phenyl]-1,3-oxazole (POPOP) [13]. To produce our plastic scintillators we polymerised the styrene solution using the free radical initiator azobisisobutyronitrile (AIBN).

A range of lithium containing compounds was trialled for solubility in the polymerisation solution. Lithium methacrylate (LiME) was investigated as the methacrylate ligand has a vinyl group which is complementary to the vinyl groups in styrene and allows the compound to be incorporated into the polymer. LiME is sparingly soluble in the styrene monomer and copolymers have been previously reported [14]. However the application of these copolymers to scintillator applications has not been investigated. To increase the lithium loading in the polymerisation solution a mixture of methacrylic acid and styrene was used.

A range of scintillators with LiME concentrations varying from 1.25 to 10 wt% were fabricated using both natural lithium and enriched lithium-6. The percentage of lithium-6 by weight in 95% enriched ^{6}Li-ME is 6.3% thus in a 4 g scintillator containing 0.4 g LiME the lithium-6 content is 0.63 wt%. The calculated lithium-6 weight percentages for both the natural lithium (7.5% lithium-6 by weight) scintillator series and the enriched scintillator series are shown in Table 1.

The following is an example of the method used to fabricate the scintillators. LiME (0.4 g) was dissolved in methacrylic acid

Table 1
LiME content, lithium-6 percentage by weight and attenuation length of the bulk scintillator samples.

	LiME wt %	Li-6wt %	Attenuation length (mm)
Natural (7.5 wt% Li-6)	1.25	0.007	41
	2.5	0.014	25
	5	0.028	32
	7.5	0.042	30
	10	0.057	27
Enriched (95 wt% Li-6)	1.25	0.08	22
	2.5	0.16	31
	5	0.31	37
	7.5	0.47	41
	10	0.63	34

Fig. 1. Photo of scintillator samples, 1.25 wt% enriched LiME (left) and 10 wt% enriched LiME (right).

(1.6 g) and this solution was then mixed with a solution of PPO (20 mg) and POPOP (2 mg) in styrene (2 g). AIBN (2 mg) was then added to this solution and the mixture sealed in a glass ampoule under argon then incubated at 50 °C to facilitate polymerisation. The scintillators fabricated by this method were 20 mm diameter, 10 mm long, transparent solid plastic bullets; however a range of sizes and shapes could be produced. One end face of each scintillator was machined flat to match the photocathode of a photomultiplier tube.

The attenuation length of the flattened scintillator samples (Table 1) was measured using an Ocean Optics ST 2000 fibre-optic spectrometer. The samples were all transparent as can be seen in Fig. 1, with an average attenuation length of 32 mm for light at 429 nm the emission wavelength of POPOP (Fig. 2).

3. Luminescence measurements

3.1. Photoluminescence

Photoluminescence (PL) spectra were recorded on a Fluorolog 3 fluorometer at room temperature equipped with a 450W Xe arc lamp source. Typical spectra for fluorescence and excitation are shown in Fig. 2. The fluorescence spectra show clear vibronic features which assist in attributing the spectra to two principal bands, assigned to PPO and PPO-pumped POPOP as shown. However a third weak band in the red region of the spectrum remains unattributed, and possibly arises from the polymerising agent. The excitation spectra at wavelengths characteristic of PPO and POPOP are very similar, supporting the idea of the PPO acting to pump emission from the POPOP dye in a wavelength shifting