



^{10}B enriched plastic scintillators for application in thermal neutron detection

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ABSTRACT

We report here on the synthesis and characterization of a novel ^{10}B enriched aromatic molecule that can be incorporated into common poly(vinyltoluene) (PVT) based plastic scintillators to achieve enhanced thermal neutron detection. Starting from relatively inexpensive ^{10}B enriched boric acid, we have prepared 4,4,5,5-tetramethyl-2-phenyl-1,3,2-dioxaborolane (MBB) in three high yield steps. MBB is soluble and compatible with PVT based formulations and results in stable plastic scintillators. Chemical synthesis, solubility limit in PVT, and the physical properties of the dopant were explored. The relevant response properties of the resulting scintillators when exposed to neutron and gamma radiation, including light yield and pulse shape discrimination properties were measured and analyzed.

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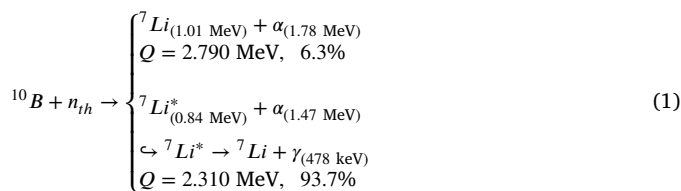
1. Introduction

Over the past decade, the rapid increase in deployment of radiation portal monitors to address domestic and international security concerns has produced a significant demand for ^3He gas. ^3He is a stable isotope of helium that is used in the detection of neutron radiation as a counting gas in proportional counters. These detectors are not only used for security purposes but also found in large scale neutron science experiments as well as in nuclear scattering and imaging research fields. With the demand for ^3He gas far outweighing the production, the US federal government began rationing the store of ^3He , only allocating it for certain groups or purposes [1]. Research aimed toward finding a viable alternative to be used for detection of neutrons is currently ongoing and motivated the research presented here.

Plastic scintillators are traditionally utilized as a first line detection method in radiation detection systems. Their large volume, relatively low cost, and fast response times allow for their efficient use as gross counters, but they provide very limited spectroscopic information about the incident radiation. Due to their large hydrogen content, plastic scintillators are sensitive to fast neutron radiation via proton collisions that in turn have a thermalizing effect on the incident neutrons, allowing them to be detected by the currently employed ^3He based proportional

counters. By incorporating material into the plastic that has a high thermal neutron interaction cross section, these detectors can directly measure the neutron radiation, that may reduce the need for ^3He based technology [2].

Although there are several possible neutron sensitive isotopes to explore, each with their own pros and cons, we are focusing our efforts on incorporating the ^{10}B isotope into plastic scintillators via organo-boron based molecules. The relatively high natural abundance of ^{10}B (~20%) in boron, established organic chemistry methods, and the nuclear properties of the capture reaction (Eq. (1)) are attractive for the development of new detector materials [3–5].



Eq. (1): Reaction scheme for thermal neutron capture on ^{10}B ; at thermal neutron energies (25 meV), ^{10}B has a neutron reaction cross section of 3837 barns (compared to ^3He : 5333 barns) [6].

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Carboranes have traditionally been used as additives to increase boron content in plastic scintillators however, they are very expensive. Searching for more cost effective alternatives, we first utilized the commercially available bis(pinacolato)diboron (B_2Pin_2) as the boron additive in plastic scintillators and also as a chemical precursor for borylated benzenes explored for the same purpose [7,8]. Although we achieved good neutron detection capabilities, we reached a solubility limit that corresponded to a ^{10}B content of ~ 0.2 wt%. To increase our ^{10}B content, and therefore enhance neutron detection, we explored the use of ^{10}B enriched precursors. This would lead to lower material loading into the plastic matrix while maintaining or increasing neutron sensitivity, and enhancing the mechanical integrity of the resultant plastic scintillators. We report here the synthesis of ^{10}B enriched 4,4,5,5-tetramethyl-2-phenyl-1,3,2-dioxaborolane, termed monoborylated benzene (MBB), in three high yield steps (Scheme 1) and its incorporation into poly(vinyltoluene) (PVT) based scintillators for use in thermal neutron detection. MBB was admixed into PVT at loadings up to 20.0 wt% (1.06 wt% ^{10}B) [9].

2. Preparation of monoborylated benzene (MBB)

Initially, the synthesis of ^{10}B enriched B_2Pin_2 was explored however, the toxicity and high price of the enriched starting material ($^{10}BF_3$) led us to look for safer synthetic routes and affordable alternatives [10]. From a synthetic standpoint the most readily available source of enriched ^{10}B is boric acid, so we developed our chemistry starting from this material. Scheme 1 shows the chemistry we designed and performed in order to prepare a PVT compatible and stable MBB from 1-bromobenzene. Initial attempts to prepare di- and triborylated ^{10}B enriched aromatics starting from 1,3-dibromobenzene and 1,2,4-tribromobenzene and t-butyl lithium were unsuccessful.

The synthesis of MBB began with enriched boric acid ($>96\%$ ^{10}B) as the precursor which was purchased from Ceradyne Inc. (\$1.74/gram). Precursors 1 and 2 (Scheme 1) were synthesized following literature procedures [11,12]. For MBB preparation, a dry two-neck 250 mL round-bottom flask equipped with a stir bar and a dropping funnel was evacuated and refilled with argon four times. Tert-butyl lithium (tBuLi) (50mL of a 1.7M solution in pentane, 0.085 mol) was added to the flask using a cannula followed by 45.0 mL of anhydrous pentane. The solution was cooled to -78 °C and stirred for 30 min. To a dry 50.0 mL round bottom flask was added bromobenzene (8.45 mL, 0.079 mol) followed by vacuum/argon refill four times then by 10–15 min of bubbling argon through the solution followed by an additional vacuum/argon refills four times. Anhydrous THF (10 mL) was added to the bromobenzene and the resultant solution transferred to a dropping funnel that was then slowly added dropwise into the -78 °C tBuLi solution over a period of 30 min. The solution went from pale yellow to off-white after approximately 2 h of stirring. Then dry isopropoxy $^{10}Bpin$ (2) (15.9 g, 0.085 mol) was added to the dropping funnel and slowly dripped into the reaction over 30 min. The reaction was stirred at -78 °C for a day. The reaction vessel was removed from the cold bath and allowed to warm to room temperature with stirring for 5 h. Isopropyl alcohol was slowly added to the completed reaction in order to quench any excess tBuLi . The reaction was then extracted with diethylether, dried with magnesium sulfate, and concentrated to remove the majority of the solvents. The product was vacuum distilled to remove the remaining volatile materials and then collected as a white solid and stored in the freezer until needed. (Yield 46%, 7.54 g) The final product was characterized by proton and carbon nuclear magnetic resonance (1H and ^{13}C NMR). 1H NMR (500.0 MHz, $CDCl_3$) δ 1.34 (s, 12 H), 7.34–7.38 (t, 2 H), 7.44–7.47 (t, 1 H), 7.80–7.81 (d, 2 H). ^{13}C 1 NMR (125.8 MHz, $CDCl_3$): δ 135.13 (CH), δ 131.65 (CH), δ 128.10 (CH), δ 84.15 (CCH₃), δ 25.24 (CH₃), n.o. (C-B).

3. Preparation and testing of plastic scintillator samples

Details on the preparation and testing of plastic scintillators can be found in our previous publications [7,8]. Briefly, purified vinyltoluene monomer (VT) (inhibitor removed by flowing through an alumina gel plug) and recrystallized azobisisobutyronitrile (AIBN) radical initiator were mixed with a specific wt% of dopants. After being sparged with argon to de-oxygenate the mixture, the samples were polymerized in glass jars (the surfaces are silanized to prevent sticking) in an inert atmosphere oven at 80 °C for four days then at 90 °C for one day. After reaching ambient temperature, the glass jars were broken to provide the 20 g plastic scintillators (4.7 cm diameter by 1.1–1.3 cm) that were then machined and polished on a single side. Each finished sample was wrapped in a reflective tape, coupled to a PMT (on the finished side), and exposed to fields of ionizing radiation. The samples response was measured with a DAQ system based on a 250 Msample/s waveform digitizer built in-house, controlled by a MIDAS software interface [13]. Pulse integration occurs over a 120 ns interval. In this study we used 2,5-diphenyloxazole (PPO) as the primary dopant and 1,4-bis(5-phenyloxazol-2-yl) benzene (POPOP) as a wavelength shifter.

Light yield (LY) of a sample was determined by comparing the positions of the Compton Edge (C.E.) feature resulting from exposure to a ^{137}Cs source, to a Saint Gobain Crystals BC-408 commercial sample. The known energy of the C.E. allowed us to calibrate each sample individually and establish an individual sample energy scale (in keV_{ec}). Next, the samples were exposed to a $^{244}Cm/^{13}C$ source in various shielding configurations, which produce different mixed neutron and gamma fields. The sample responses to fast and thermalized neutron fluences were recorded to identify and analyze the ^{10}B neutron capture reaction as well as any n/ γ PSD capabilities. Gamma background spectra were taken with the use of a cadmium shield (~ 1 mm) around the coupled sample. The ROOT data analysis framework was utilized to analyze the collected data, and develop relevant output spectra [14].

4. Results

We first explored the solubility limits of MBB in vinyltoluene to determine how much ^{10}B could be added (as indicated earlier, the detection of thermal neutrons is achieved via the ^{10}B), as well as testing the compatibility of MBB with a plastic scintillator formulation (PVT overdoped with 20.0 wt% PPO) that allowed for PSD for further particle identification [15]. Table 1 details the various samples that were produced and tested for radiation response. MBB was still soluble in PVT at 20.0 wt%, however, polymerization of the final plastic was already significantly hindered at this level and an upper limit for the solubility was not determined. For comparison, we prepared m-carborane based samples in order to compare radiation response and physical properties such as dopant solubility and ^{10}B content, total light yield, energy of boron capture peak.

The measured ^{137}Cs response of samples containing MBB and m-carborane are shown in Table 1 (a 5% error is assumed in the LY measurements) and it is seen that the LY trends lower both with increasing MBB and m-carborane concentrations. The two samples containing MBB with the lowest LY (sample 4 and 6) were too soft to fully machine or polish, due to the amount of dopants (combined MBB and PPO) in the sample, leading to a practical limit of <15 wt% MBB for usable plastics. Likely, incomplete polymerization lowered the LY as well as poor optical coupling to the photodetector. The LY of the samples vary from $>50\%$ to $\sim 90\%$ of a commercial control sample (BC-408; app. 11,000 photons/1 MeV; 64% anthracene). The m-carborane containing samples all polymerized to clear, colorless samples, which were hard enough to machine and polish. However, the drop off in LY is more pronounced for m-carborane samples containing equivalent amounts of ^{10}B to MBB samples, likely due to the absence of aromatic groups in m-carborane compared to MBB.

Fig. 1 shows the thermal neutron capture peaks resulting from exposing the samples to thermalized neutrons. Data was collected

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