Thermal neutron scintillators using unenriched boron nitride and zinc sulfide

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Abstract. Thermal neutron detectors based on powdered zinc sulfide intimately mixed with a neutron capture compound have a history as long as scintillation technique itself. We show that using unenriched boron nitride powder, rather than the more commonly used enriched lithium fluoride, results in detection screens which produce less light but which are very considerably cheaper. Methods of fabricating large areas of this material are presented. The screens are intended for the production of large area low cost neutron detectors as a replacement for helium-3 proportional tubes.

1. Introduction
\textsuperscript{3}He proportional tubes were the industry standard neutron detectors over the period \textasciitilde 1970-2008. Recently, however, the international shortage of \textsuperscript{3}He has led to the abandonment of these and an urgent search for alternative technologies [1].

Large numbers of neutron detectors are needed for Radiation Portal Monitors (RPMs) which are used at international borders and other locations to detect illegally shipped nuclear materials [2]. Identical monitors are employed in the metal recycling industry to detect orphan sources or other radioactive materials in scrap feeds before melting [3]. A further application is ensuring the security of sites where nuclear materials are used or stored.

As the expected fluxes are low, the detectors used must be as large as possible, square metres being typical. The detection efficiencies must be high, but also unambiguous, so good signal-to-noise ratios are desirable. Real-time signal discrimination is essential, as opposed to the computed post-processing encountered in many experimental physics environments. The detectors must be deployable, that is, reasonably robust and stable over many years operation in harsh environments. They must be manufacturable, transportable and installable with minimal health and safety implications and operable by unskilled staff. Lastly, because of the large areas required, there are constraints on price, which translate into a requirement to use readily available materials.

2. \textsuperscript{6}LiF-ZnS scintillation detectors
Thermal neutron detectors based on powdered zinc sulfide intimately mixed with a neutron capture compound have a history as long as scintillation technique itself.

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Large volume thermal neutron detectors based on a mixture of isotopically enriched lithium fluoride and zinc sulfide scintillator powders held in a binder were demonstrated by Barton et al. [4, 5]. These used the scintillator mixture in layers, interleaved with acrylic wavelength shifting lightguides and polypropylene moderators. The lightguides were end-viewed by photomultipliers and neutron discrimination was provided by pulse counting discriminators similar to those described by Caines [6] and Davidson [7]. These detectors were originally built to study rare fission events and were operated in harsh environments in underground low-background sites.

The individual detectors had an active volume $90 \times 14.4 \times 14.4$ cm and a stack of eight detectors completely surrounding a $^{252}$Cf source had a neutron detection efficiency of 37%. The thin sensitive layers and the pulse counting discrimination technique ensured that these detectors were totally insensitive to gammas or muons. They have provided robust stable operation over the intervening years, having been deployed in three different underground sites.

3. Neutron scintillation detectors for portal applications

A program to re-investigate this type of detector for use as the neutron sensitive component in RPMs has been undertaken.

Initially consideration was given to replacing the zinc sulfide. The first complaint about zinc sulfide is that it absorbs its own light and can only be used in thin layers. However, it is just this that gives the Barton design its immunity to gamma rays, as no gamma with typical nuclear decay energy will ever deposit sufficient energy in the thin layer to compare with the products of neutron capture on $^{6}$Li. Zinc sulfide also has problems with long decay time and poor pulse height discrimination but these were largely dealt with by the pulse counting discriminators. Problems with long afterglow remain, but as high event rates are not expected, this is unlikely to be a problem. The fact remains that, despite half a century of research into inorganic scintillators, zinc sulfide remains the brightest low cost scintillator. The choice was made to continue with it.

$^{6}$LiF is a controlled material and is also increasingly expensive, so it was necessary to consider the alternatives. The only three capture reactions of interest are:

\[ ^{3}\text{He} + n \rightarrow ^{3}\text{H} + ^{1}\text{H} + 0.764\text{MeV} \]
\[ ^{6}\text{Li} + n \rightarrow ^{4}\text{He} + ^{3}\text{H} + 4.8\text{MeV} \]
\[ ^{10}\text{B} + n \rightarrow ^{7}\text{Li} + ^{4}\text{He} + 2.3\text{MeV} + 0.48\text{MeV}(\gamma) \]

Gadolinium, cadmium and europium are excluded, since their capture reactions involve emission and subsequent detection of gamma rays and, consequently, it is difficult to get good neutron/gamma discrimination with these elements. There are also issues of cost and toxicity.

Table 1 shows the natural abundances of the useful isotopes together with the thermal capture cross-sections for these reactions. The use of natural lithium is unattractive due to the low concentration of $^{6}$Li. Natural boron on the other hand is attractive, with the 19.8% abundance of the $^{10}$B being offset by the very much higher capture cross-section. It is clear that the use

| nuclide | % natural abundance | thermal cross-section |
|---------|---------------------|-----------------------|
| $^{3}$He | 0.00014 | 5333 |
| $^{6}$Li | 7.50 | 940 |
| $^{10}$B | 19.80 | 3842 |

Table 1. Natural abundances and cross-sections in barns for useful capture reactions.
of material which is not isotopically enriched is likely to result in considerable savings and that, even if the resulting detectors are slightly worse in performance, they may still be very attractive. If a detector can be demonstrated using unenriched boron, then it is clear that a considerable improvement can subsequently be obtained by switching to enriched material, but that this would also dramatically increase the cost.

The obvious disadvantage of the $^{10}$B reaction is that the reaction products have much less energy than is the case in the $^6$Li reaction. This will result in less light produced in the zinc sulfide and may require better designed optics. The other problem is that the products of the capture reactions have reduced range. Simulations using SRIM [8] indicated that the mean range of the $^7$Li nucleus was 2.5$\mu$m while the alpha particle had a range of 4.3$\mu$m. This indicated that very fine grain size powdered boron compounds were needed.

Boron compounds mixed with zinc sulfide scintillator were demonstrated [9, 10] as neutron detectors at an early stage in the development of scintillation technique and much subsequent work was done, but this was largely abandoned when $^3$He tubes became dominant. No attempt had been made to read it out using the Barton volume configuration [4], so all the reports of detectors using boron describe rather small detectors.

The ideal boron compound for this application is required to be completely chemically stable and transparent to the light emitted by the zinc sulfide. It should also be commercially available at low cost in controlled grain sizes. A boron compound which meets these requirements is boron nitride, BN, which is available in two allotropes; a hexagonal form and a cubic form. Hexagonal BN is a white slippery solid with a layered structure, physically similar to graphite. It is manufactured for lubricant and cosmetics applications.

Hexagonal BN has a density of 2100 kg m$^{-3}$ and is birefringent with refractive indexes of 1.65 and 2.13. The pure material is highly transparent in the visible and near UV having essentially no optical absorption at wavelengths longer than 230nm [11]. The macroscopic neutron cross section of $^*BN$ is 38.93 cm$^{-1}$, compared to 57.53 cm$^{-1}$ for $^6$LiF, so the resulting detectors will be of comparable efficiency.

4. Optimization

The Barton detectors [4] were optimized for maximum efficiency and lowest background in a volume configuration. Security applications need to optimize effective area per unit cost:

$$\frac{\text{area} \times \text{efficiency}}{\text{cost}}$$

MCNPX [12] simulations indicated that in the original eight layer design, the four best layers contributed 76% of the efficiency of the detector. This suggested that the four worst layers can be redeployed at the side to double the area giving an effective area 1.52 times the original.

5. Scintillating layers

The layer production technology has been revised for ease of use and to reduce waste. Originally a thermoset resin was used as a binder, but this was problematic in that it set rapidly and could not be reworked or easily cleaned from equipment. Experiments on replacing this with solvent soluble polymers were successful, a thick paint being made from ZnS and BN powders, polymer and solvent.

This is applied in a controlled thickness to a substrate of aluminized mylar. When the solvent evaporates, the scintillator layer remains firmly attached to the mylar and the result is sufficiently robust and flexible to withstand handling and even curving to quite a tight radius. Small pieces can be made by a doctor-blade spreading technique. To produce larger areas, a coating machine has been developed, which uses a peristaltic pump to apply the scintillator paint mixture to the mylar through a small nozzle. This can be seen in figure 1.
6. Wavelength shifting lightguides
With less light available from the boron reaction, than the lithium reaction, light collection becomes more important. The original BBQ loaded acrylic (Plexiglas GS2025) is no longer available. A number of alternative materials have been investigated. Some of the commercial wavelength shifting materials are based on polyvinyltoluene and are consequently intrinsic scintillators. Their use degrades the overall neutron/gamma discrimination of the current design. Success has been achieved with a commercial green fluorescent acrylic rod as can be seen in figure 2, however, supplies of this material may prove unreliable. Experiments have been made with dyed acrylic [13] and these are ongoing.

7. Current status
At present two experimental detectors using boron nitride are working. One is based on sheet wavelength shifter, while the other is based on cylindrical rods. They have the same gamma ray rejection as the original designs and similar detection efficiency to $^3$He proportional tubes. Considerably more details can be found in Marsden’s thesis [14]. More accurate efficiency measurements are in preparation and will be published shortly.

8. Acknowledgements
This work was supported by the Home Office Scientific Development Branch and subsequently by STFC CLASP award ST/K000179/1.
Figure 2. The detector based on cylindrical lightguides

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