PROCEEDINGS OF SCIENCE

Neutron Detectors for Security Applications

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Due to the global shortage of ^{3}He, a replacement for the ^{3}He proportional thermal neutron detector is urgently sought. A previously successful neutron scintillation detector design has been reconstructed to use cheaper and more readily available materials, with the intention that it can be used in radiation portal monitors. This report outlines the design considerations.

19th International Workshop on Vertex Detectors - VERTEX 2010
June 06 - 11, 2010
Loch Lomond, Scotland, UK

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PoS(VERTEX 2010)031

1. Introduction

There is a growing requirement for particle detectors in Radiation Portal Monitors (RPMs) which are used at international borders and other locations to detect illegally shipped nuclear materials [8]. Identical monitors are also employed in the metal recycling industry to detect orphan sources or other radioactive materials in scrap feeds before melting occurs [10]. A further application can be found in ensuring the security of sites where nuclear materials are used or stored. Both gamma and neutron detectors are usually deployed together for all these applications.

The detectors used are radically different in design to the majority of vertex detectors described elsewhere in this conference. They are rarely position sensitive, having a single pixel, but this must be as large as possible, square metres being typical. They must detect low particle fluxes giving low event rates in an untriggered environment. The particle energies are low, typically <10MeV for both gammas and neutrons. In the case of gamma rays, RPMs have no or very limited ability to measure energy spectra, while in the case of neutrons, no energy measurement is possible. The detection efficiency must be high, but also unambiguous, so good signal-to-noise ratios are desirable. Real-time signal discrimination is essential, as opposed to the computer-intensive 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 front line officers (Border Agency staff, etc). Lastly, because of the large areas required, there are extreme constraints on price, which translate into a requirement to use readily available materials.

Current typical RPMs use NaI or PVT plastic scintillators for gamma detection combined with ^{3}He proportional tubes for neutrons. ^{3}He proportional tubes are thermal neutron detectors and are normally surrounded by hydrogenous moderator to provide a broad energy response. The construction, operation and performance characteristics of ^{3}He proportional tubes are given by Crane and Baker [4].

2. The ^{3}He supply problem

The global shortage of ^{3}He is an international scientific crisis and, as of November 2009, the US Department of Homeland Security has suspended all installations of neutron detectors in RPMs at ports and borders. The (US) annual demand for ^{3}He is estimated at 65000 litres and there is essentially no source that can meet this demand. Supply is dwindling due to reduced production and use of tritium. The price has risen from \$100 to \$2000/litre in recent years [9] and is still rising. A replacement detector design for the ^{3}He proportional tube is urgently sought.

3. ^{6}LiF-ZnS scintillation detectors

Large volume thermal neutron detectors based on an intimate mixture of isotopically enriched lithium fluoride and zinc sulfide scintillator powders held in a binder were demonstrated by Barton et al. [2]. These used the mixture spread in $\sim$100μm layers and interleaved with acrylic wavelength shifting panels and polypropylene moderators. The panels were end-viewed by two photomultipliers and neutron discrimination was provided by pulse counting discriminators similar to those described by Caines [3] and Davidson [5]. 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.

4. 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 and it may be that gated charge integration is even better. 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 to detector designers are:

$$
\begin{aligned}
{}^3\mathrm{He} + \mathrm{n} &\rightarrow {}^3\mathrm{H} + {}^1\mathrm{H} + 0.764\mathrm{MeV} \\
{}^6\mathrm{Li} + \mathrm{n} &\rightarrow {}^4\mathrm{He} + {}^3\mathrm{H} + 4.8\mathrm{MeV} \\
{}^{10}\mathrm{B} + \mathrm{n} &\rightarrow {}^7\mathrm{Li} + {}^4\mathrm{He} + 2.3\mathrm{MeV} + 0.48\mathrm{MeV}(\gamma)
\end{aligned}
$$

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

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.

^{10}B being offset by the very much higher capture cross-section. It is clear that the use 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 less energy and consequently reduced range. Simulations using SRIM [13] for indicated that the mean range of the ^{7}Li nucleus was 2.5μm while the alpha particle had a range of 4.3μm. This indicated that very fine grain size powdered boron compounds were needed.

Boron compounds mixed with zinc sulfide scintillator were demonstrated [1, 7] 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 [2], so all the reports of detectors using boron describe small detectors. Initial tests have now been made by us using fine grain hexagonal boron nitride produced for cosmetic applications. Cubic boron nitride is also available in controlled sizes as an abrasive. Engels et al. [6] identified sintered boron nitride as a potential neutron detector, however no light emission could be detected from any of the fine powder boron nitride samples obtained by us.

The layer production technology has also been revised, largely to reduce wastage. 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. The original detectors used a spreading technique and, while various other techniques, such as spray painting, powder coating, serigraphy and ink-jet systems, were examined, coating using doctor-blade spreaders remained the best choice for pilot scale production.

5. Optimization

The Barton detectors [2] 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 indicate that in the original eight layer design, the four best layers contribute 76% of the efficiency of the detector. This suggests that the four worst layers can be redeployed at the side to double the area giving an effective area 1.52 times the original.

Such a planar detector will require a new optical configuration. The original detectors used wavelength shifting panels fabricated from a commercial acrylic loaded with the wavelength shifting dye BBQ (Plexiglas GS2025). Experiments have been undertaken using a low cost technique in which BBQ dye loaded lacquer was sprayed onto the surface of clear acrylic sheet, following the ideas of Viehmann and Frost [11]. Tests with small samples suggest that this is 1.2 times better than the original material. An alternative approach using dip-coated cylindrical acrylic rods has also been examined and this is also giving good results.

The original pulse counting system used hard-wired TTL and a new computer based monitoring system controlling hardware neutron discrimination is being constructed to replace this.

6. Current status

At present two experimental detectors using boron nitride and based on these new ideas are working. They appear to have the same gamma ray rejection as the original designs and similar detection efficiency to ^{3}He proportional tubes, though as yet, no accurate calibrations have been performed. The exact discrimination criteria is being optimized with respect to the characteristics of the zinc sulfide used. A more detailed report will be published when this has been completed.
This work was supported by the Home Office Scientific Development Branch.

References

[1] D.E. Alburger. *A slow neutron counter with high efficiency. Rev. Sci. Instrum.*, 23:769, 1952.

[2] J.C. Barton, C.J. Hatton, and J.E. McMillan. *A novel neutron multiplicity detector using lithium fluoride and zinc sulphide scintillator. J Phys G: Nucl Part Phys*, 17:1885–1899, 1991.

[3] P.J. Caines. *The development of an efficient neutron detector for cosmic ray studies.* Master's thesis, University of London, 1972.

[4] T.W. Crane and M.P. Baker. Neutron detectors. Chap. 13, *Passive Nondestructive Assay of Nuclear Materials*, ed. Reilly, D. et al., Technical Report NUREG/CR-5550; LA-UR-90-732, Los Alamos National Laboratory, NM, USA., 1991.

[5] P.L. Davidson. *A new discriminator principle for slow neutron counting.* Technical Report RL-77-106/A, Rutherford Laboratory, Chilton, Oxon, UK, 1977.

[6] R. Engels et al. *Boron nitride, a neutron scintillator with deficiencies. IEEE Nucl. Sci. Symp. Conf. Rec.*, 3:1318– 1322, 2005.

[7] E. Gatti et al. *Boron layer scintillation neutron detectors. Nuovo Cimento*, 9:1012–1021, 1952.

[8] IAEA. *Illicit nuclear trafficking: collective experience and the way forward.* Technical Report STI/PUB/1316, International Atomic Energy Agency, Vienna, Austria, 2008.

[9] R.L. Kouzes. *The ^{3}He supply problem.* Technical Report PNNL-18388, Pacific Northwest National Laboratory, Richland, Washington, USA, 2009.

[10] United Nations. *Report on the Improvement of the Management of Radiation Protection Aspects in the Recycling of Metal Scrap*. United Nations Economic Commission for Europe, United Nations, New York, USA, 2001.

[11] W. Viehmann and R.L. Frost. *Thin film waveshifter coatings for fluorescent radiation converters. NIM*, 167:405–415, 1979.

[12] L.S. Waters. *MCNPX user's manual: Version 2.3.0.* Technical Report LA-UR-02-2607, Los Alamos National Laboratory, NM, USA, 2002.

[13] J.F. Ziegler, J.P. Biersack, and M.D. Ziegler. *SRIM – The Stopping and Range of Ions in Matter*. Lulu Press, Morrisville, NC, USA, 2008.