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The Feasibility of Using Boron-Loaded Plastic Fibers for Neutron Detection

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Abstract

The results from simulations and laboratory experiments with boron-loaded plastic scintillating fibers as a nondestructive assay tool are presented. Single and multiclad fibers in three diameters of 0.25, 0.5, and 1 mm were examined for their application in neutron coincident counting. For this application, the simulation results show that various configurations of boron-loaded plastic scintillating fibers have a die-away time (τ) of 12 μs with an efficiency (ε) of 50%. For a comparable efficiency, ^{3}He proportional tubes have a typical die-away time of 50 μs. The shortened die-away time can reduce the relative error for measurement of similar samples by up to 50%. Plastic scintillating fibers also offer flexible configurations with the potential to discriminate between signals from gamma-ray and neutron events.

To date, the emphasis of the investigation has been the detection capability of plastic scintillating fibers for neutrons and gamma rays and evaluation of their ability to discriminate between the two events. Quantitative calculations and experiments have also been conducted to determine the light output, evaluate the noise, quantify light attenuation, and determine neutron detection efficiency. Current experimental data support the analytical results that boron-loaded plastic fibers can detect thermal neutrons with performance metrics that are comparable or better than those of ^{3}He proportional tubes.

Introduction

Since their introduction, scintillating fibers have had numerous applications for radiation detection. Leutz [1], White [2], and Kirkby [3] provide an excellent overview of scintillation fibers and their potential application as radiation detectors. Axmann [4], Bliss, *et al.* [5], and Ottonello, *et al.* [6] have investigated the use of lithium- and boron-loaded plastic and glass fibers for thermal neutron detection. Angelini, *et al.* [7] compare the characteristics of glass and plastic scintillating fibers (PSF) for high-resolution tracking of charged particle beams. Takada, *et al.* [8], Sailor, *et al.* [9], Wurden, *et al.* [10], and Singkarat, *et al.* [11] have investigated the use of PSF for fast neutron detection and spectrometry. Other specific applications of PSF include the work by Yariv, *et al.* [12], Imai, *et al.* [13], Binns, *et al.* [14], and Finocchiaro, *et al.* [15] for radiation detection, track imaging, and profiling low-intensity ion beams.

One application area that could benefit from the development and application of PSF is nondestructive assay (NDA) of special nuclear materials. For detection and characterization of fissionable materials, such as plutonium and uranium present in the nuclear fuel cycle [16], boron-loaded PSF can also be used as neutron coincidence counters. At present, the most commonly used and effective neutron counter is the polyethylene-moderated ^{3}He thermal neutron coincidence counter (TNCC). This counter has a relatively high efficiency, $\approx 50\%$, but a long die-away time of ≈ 50 μs. Because of this long die-away time, a wide coincidence timing gate is needed for counting correlated neutrons. This sometimes results in a high accidental coincidence rate from random neutrons such as those produced in (α, n) reactions. The high accidental coincidence rate results in a longer assay time in order to achieve the desired relative error. When used as a neutron coincidence counter, boron-loaded plastic detectors can have a much shorter die-away time and high neutron-detection efficiency. Miller [17] reports a 50% efficiency with 3–4 μs die-away time for BC454[1]/BGO-phoswich detectors. With a shorter die-away time, a more sensitive and accurate determination of the actual sample mass may be possible. Cylindrical boron-loaded optical fibers of various diameters and lengths as well as ribbons of 200 fibers have been investigated. The fibers examined consist of a boron-loaded (1% by weight ^{10}B) polystyrene (PS) core surrounded with polymethylmethacrylate (PMMA) cladding. Detectors built from fibers of this type have a flexible structure and, if configured properly, can have a die-away time and efficiency comparable to those of BC454. The fiber detectors are also less sensitive to gamma rays because of the small diameter of the fiber. Used in multifiber ribbons, the signals from the PSF can also be logically sorted for gamma-ray discrimination.

This paper reports on the use of boron-loaded plastic fibers for detection of neutrons in high intensity gamma-ray fields. A short discussion on the principle

[1] 1% by weight ^{10}B in polyvinyltoluene-based material.

of the scintillation process in PSF and the results from analytical and simulation analyses are presented.

Principle

The mixture of the moderator and thermal neutron absorber in the fiber yields a detector with high efficiency and a short die-away time. Energy deposited by charged particles is converted to light that is collected via total internal reflection in the fibers coupled to photomultiplier tubes. As shown in Fig. 1, the incident neutron with energy E_n is thermalized through a series of scattering collisions and is subsequently captured by ^{10}B, which has a cross-section of 3838 b. The Q-value of the $^{10}B(n,\alpha)^7Li$ reaction is 2.792 MeV (94% of the time), with 2.31 MeV going to the charged particles along with a 478-keV gamma ray.

The amount of deposited energy of the resultant particles is converted to scintillation light. For fast electrons, Birks [18] suggests a semi-empirical relation for production of light if quenching of primary excitation can be assumed unimolecular:

$$\frac{dL}{dr}=\frac{S\,dE/dr}{1+kB\,dE/dr},$$

where E refers to the deposited energy of the charged particle, r is the range of electrons in the scintillator measured in cm air equivalent, S is the scintillator efficiency, and kB is a material dependent factor expressed in g-cm^{-2}-MeV^{-1}. For recoil protons, Singkarat, *et al.* [11] use another semi-empirical formula:

$$L(E)=S\int_0^E dE\left(1+kB\left(\frac{dE}{\rho dr}\right)+C\left(\frac{dE}{\rho dr}\right)^2\right)^{-1},$$

where C is a measured parameter expressed in g^2-cm^{-4}-MeV^{-2}. For α particles, Birks [18] suggests the following linear relationship for the scintillation response per unit path length:

$$\frac{dL}{dr}=\frac{S}{kB}=\text{constant}.$$

The light output from the $n+^{12}C$ interactions (elastic and inelastic) is assumed to contribute little because the strong nonlinearity of the light response [11]. Although the fiber includes a layer of cladding, its contribution to the interaction is also ignored. These functions have been empirically determined for many types of scintillators [19], [20],[21]. Miller [17] reports a value of 93 keV$_{ee}$ for the reaction production of $^{10}B(n,\alpha)$ in plastic scintillators. The unit of keV$_{ee}$ or expresses the light output relative to that obtained for electrons of the specified energy.

Recoil protons produced from the elastic scattering of neutrons with hydrogen yield an energy, $E_p = E_n \cos^2\varphi$. Depending on the recoil angle, φ, the range of the proton can be shorter or longer than its maximum path length available in the fiber [11]. Therefore, the recoil protons can contribute to the noise, but themalizing neutrons prior to their entry into the fiber can both increase the probability of the (n,α) reaction and reduce the number of recoil protons from elastic scattering collisions.

For a single PSF, the detection efficiency depends mainly on three factors: the transmission efficiency of the absorbed energy to scintillation photons, the photon trapping efficiency, and the optical attenuation effect. For BCF-10, -12, and -20, the first factor is nominally about 2.4% which translates to 8,000 photons per MeV from a minimum ionizing radiation.[2] The trapping efficiency (the percentage of photons collected by the fiber) depends on the shape and refractive indices of the core and the surrounding medium [11]. For a cylindrical fiber with a diameter of 1 mm or less with an extra-mural absorber (EMA) coating, the trapping efficiency is approximately 4%. The optical attenuation factor depends both on the material and cross-sectional dimension of the fiber.

Monte Carlo Simulation

The efficiency and die-away time of several configurations of our proposed detector system were simulated with MCNP4B using the ENDF/B-V cross section libraries [22]. A basic arrangement of the system appears in Fig. 2. The geometry consisted of a can, 18 cm in diameter and 25-cm tall, surrounded by a polyethylene liner wrapped with alternating layers of polyethylene sheets and fiber ribbons. A ribbon consisted of 0.25mm fibers in a single layer. As a simplification of the geometry, the ribbons and polyethylene sheets were simulated as alternating concentric cylinders around the central can. The fiber ribbons were adjusted for density and material composition when simulated as concentric cylinders. A nickel reflector was positioned around the ribbons, and carbon plugs were put on top of the ribbons to reflect back any lost neutrons.

The simulations used ^{252}Cf and ^{240}Pu point sources assuming a Watt's fission spectrum distribution, expressed by

$$f(E)=C\exp(-E/a)\sinh(bE)^{1/2},$$

where a = 1.025 MeV and b = 2.926 MeV^{-1} for ^{252}Cf, and a = 0.799 and b = 4.903 for ^{240}Pu. The goal of the simulations was to optimize the efficiency and die-away time with a constant number of ribbons by varying the parameters of the materials. The efficiency

[2] Bicron Corporation, Newbury Ohio.

of the detector system is defined as the number of ^{10}B neutron captures per source neutron. It was assumed that all of the ^{10}B captures would result in a signal as the range of the alpha is on the order of a few micrometers. The average efficiency obtained was ≈ 50%. The actual efficiency would, however, be lower once factors, such as the quantum efficiency of the PMT and the attenuation length of the fiber are incorporated in the analysis.

The die-away time was calculated by taking the time distributions of ^{10}B events. The die-away time was strongly influenced by the thickness of the polyethylene sheets as shown in Fig. 3. The die-away time was reduced to ≈ 10–12 µs with the addition of a cadmium liner placed between the source and the ribbons.

In another series of simulations, neutron and gamma-ray energy depositions in a single fiber were simulated to determine the best fiber diameter for neutron/gamma discrimination. As shown in Fig. 4, it is impossible to distinguish between the energy deposited from a 1-MeV photon and a ^{10}B neutron capture event with a 1-mm fiber. This finding is consistent with the observation by Abel, *et al.* [23]. The same observation is true for the 0.5-mm diameter. But the 0.25-mm diameter fiber yields only a slight overlap of the photon and neutron peaks. For this reason, the 0.25-mm diameter fibers were chosen for the simulations and the final design. The light output of a ^{10}B neutron capture event in the fiber is equivalent to a 93-keV electron [17], which corresponds to the peak in the neutron capture spectrum.

Application to Neutron Detection

Single fibers have been tested with neutron and gamma-ray sources for neutron detection and pulse-height discrimination of gamma-ray events. The spectra were obtained using single 0.25-, 0.5-, and 1-mm-diameter fibers. The length of the fibers varied from 2 m to 3 m. To enhance light collection, the fiber was coupled directly to two Burle S83049F photomultiplier tubes. To cancel the effects of light loss caused by the 2.3-m attenuation length along the fiber, the geometric mean of the values from the two ends of the fiber should be used [24].

First, energy calibration measurements were taken using a BC454 log (diameter is 7.5 cm, length = 30 cm) and ^{137}Cs, ^{22}Na, and ^{109}Cd with activities of 10 µCi, 10 µCi, and 100.7 µCi, respectively, were performed. These sources provide gamma rays at 662, 1274, and 88 keV, respectively. The Compton energies for 180° scattering are 478 keV for ^{137}Cs and 341 keV for the 511 keV annihilation gamma rays from ^{22}Na. The energy of the neutron peak from the ^{10}B nuclear reaction is 93 keV, which is consistent with that reported by Miller [17]. The energy calibration measurements were also used to properly configure the electronics for data collection with a single 1-mm-diameter fiber.

Measurements were taken with a 1-mm-diameter fiber to determine their sensitivity to neutrons and gamma rays. A ^{252}Cf source was placed inside a tungsten shield to eliminate gamma rays. The tungsten shield was inside multiple, concentric cylinders of polyethylene to thermalize the neutrons. A 2-m length of 1-mm-diameter fiber was wrapped around a Lucite cylinder that fit around the polyethylene cylinders. The fiber was attached to the PMT by a threaded coupling.

Comparing the spectra with and without the polyethylene cylinders shows an increase in counts between channels 16–64 with the polyethylene cylinders. A sum of channels 16–64 yielded 14842 counts in that peak area. This is a factor of 2 higher than the MCNP prediction of 7960 ± 1042 for ^{10}B capture events. This discrepancy in counts is because of recoil protons interacting in the fiber. The same relationship was found between the measured and simulated spectra with and without the cadmium liner. MCNP predicted half as many counts as were obtained. The spectra obtained from these experiments are shown in Fig. 5a. When the lead liner was placed between the polyethylene and the fiber, there was virtually no effect on the spectrum, as shown in Fig. 5b.

A ribbon of 200 fibers will be tested under the same conditions as those for the single fibers to determine if logical discrimination in cooperation with pulse-height discrimination is useful in eliminating gamma-ray events. The test ribbon will also be used in a coincidence counter setup to quantify the mass of known ^{240}Pu and ^{252}Cf samples. Further tests will be conducted in a high gamma-ray flux environment, the results of which will be used to help optimize the final system.

A proposed fiber coincidence counter consists of thousands of fibers in 30-cm-wide single-layer ribbons optically coupled to an array of PMTs. The geometric arrangement of the fibers and their logical coupling to the PMTs will be configured to reduce noise, discriminate neutron and gamma-ray events, and identify coincidence neutron events.

Conclusions

Monte Carlo results of the response of boron-loaded plastic fibers of various diameters have been discussed. These results yield a die-away time of 12 µs with 50% efficiency, which point to the potential application of this type of detection system for the NDA of special nuclear materials. Laboratory experiments to date have shown detection of thermal neutrons with these fibers. The investigation into various configurations of a detector system based on this method of detection is in progress.

References

[1] H. Leutz, "Scintillating Fibres," *Nucl. Instr. and Meth.* A364 (1995) 422-448.

[2] T.O. White, "Scintillating Fibres," *Nucl. Instr. and Meth.* A273 (1988) 820-825.

[3] J. Kirkby, "Today and tomorrow for scintillating fibre (SCIFI) detectors," European Organization for Nuclear Research, CERN-EP-87060, March 1987.

[4] A. Axmann, "Hardware aspects of position-sensitive detectors, in Position-sensitive detection of thermal neutrons," P. Convert and J.B. Forsyth, Eds. Academic Press 1983.

[5] M. Bliss, *et al.*, "Glass-fiber-based neutron detectors for high- and low-flux environments," SPIE 2551 (1995) 108-117.

[6] P. Ottonello, *et al.*, "Slow Neutron Imaging using Scintillating Glass Optical Fibers," *Nucl. Instr. and Meth.* A 349 (1994) 526.

[7] C. Angelini, *et al.*, "Comparison of glass and plastic scintillating microfibers for high-resolution tracking," *Nucl. Instr. and Meth.* A295 (1990) 299-314.

[8] E. Takada, *et al.*, "Neutron distribution sensor using flexible plastic scintillating fiber combined with the time-of-flight technique," IEEE Transactions on Nuclear Science, 42 (4) (1995) 570-573.

[9] W.C. Sailor, *et al.*, "Conceptual design for a scintillation-fiber neutron detector for fusion reactor plasma diagnostics," *Rev. Sci. Instrum.* 66(1) (1995) 896-903.

[10] G.A. Wurden, *et al.*, "Scintillating-fiber 14 MeV Neutron Detector on TFTR during DT Operation," *Rev. Sci. Instrum.* 66 (1) 901 (1995).

[11] S. Singkarat, *et al.*, "The Feasibility of Using Plastic Scintillation Fibers for Fast Neutron Spectrometry," *Nucl. Instr. and Meth.* A335 (1993) 248-254.

[12] Y. Yariv, "Simulations of neutron response and background rejection for a scintillating-fiber detector," *Nucl. Instr. and Meth.* A292 (1990) 351-358.

[13] S. Imai, *et al.*, "New radiation detector of plastic scintillating fiber," *Rev. Sci. Instrum.* 62 (4) (1991) 1093-1097.

[14] W.R. Binns, *et al.*, "Scintillator-fiber charged-particle track-imaging detector," *Nucl. Instr. and Meth.* 216 (1983) 475-480.

[15] P. Finocchiaro, *et al.*, "A scintillating fiber-based profiler for low intensity ion beams," *Nucl. Instr. and Meth.* A385 (1988) 31-36.

[16] D. Reilly, *et al.*, "Passive Nondestructive Assay of Nuclear Materials," Nuclear Regulatory Commission Report NUREG/CR-5550, LA-UR-90-732 (March 1991).

[17] M.C. Miller, "Neutron detection and multiplicity counting using a boron-loaded plastic scintillator/bismuth germanate phoswich detector array," Los Alamos National Laboratory Thesis, LA-13315-T (1997).

[18] J.B. Birks, "The theory and practice of scintillation counting," Pergamon Press (1964).

[19] R.A. Cecil, *et al.*, "Improved predictions of neutron detection efficiency for hydrocarbon scintillators from 1 MeV to about 300 MeV," *Nucl. Instr. and Meth.* 161 (1979) 439-447.

[20] F.D. Brooks, "Development of organic scintillators," *Nucl. Instr. and Meth.* 162 (1992) 477-505.

[21] R. Madley, *et al.*, "The response of NE-228A, NE-228, NE-224, and NE-102 scintillators to protons from 2.43 to 19.55 MeV," *Nucl. Instr. and Meth.* 151 (1992) 445-450.

[22] J.F. Briesmeister, Ed., "MCNP4B Monte Carlo N-Particle Transport Code System," Los Alamos National Laboratory report LA-12625-M (November 1993).

[23] K.H. Abel, *et al.*, "Performance and applications of scintillating-glass-fiber neutron sensors, Proceedings of the SCIFI93 conference workshop on scintillating fiber detectors," October 1993.

[24] R.C. Byrd, *et al.*, "Measurements and calculations of the gamma response," *Nucl. Instr. and Meth.* A313 (1992) 457-463.

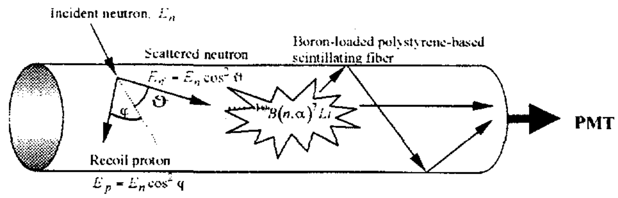

Figure 1. Thermalization and subsequent capture of fission neutrons in a boron-loaded PSF. The scintillation light from the resultant charged particles is guided through the PSF to the photomultiplier tubes at the ends of the fiber.

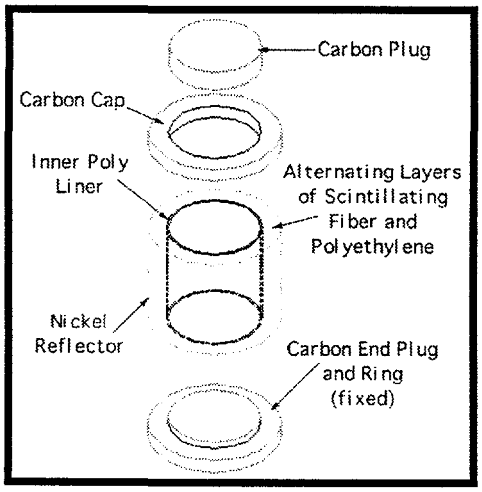

Figure 2. Basic geometric arrangemement of the proposed detector system for the MCNP calculations.

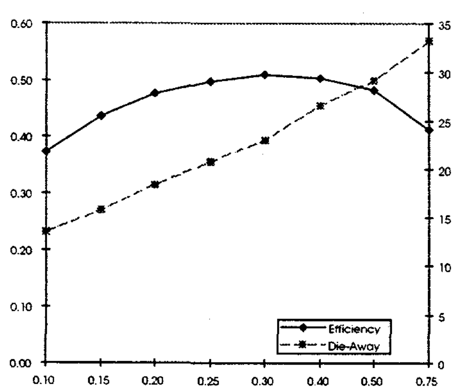

Figure 3. Efficiency and die-away time as a function of polyethylene moderator sheet thickness.

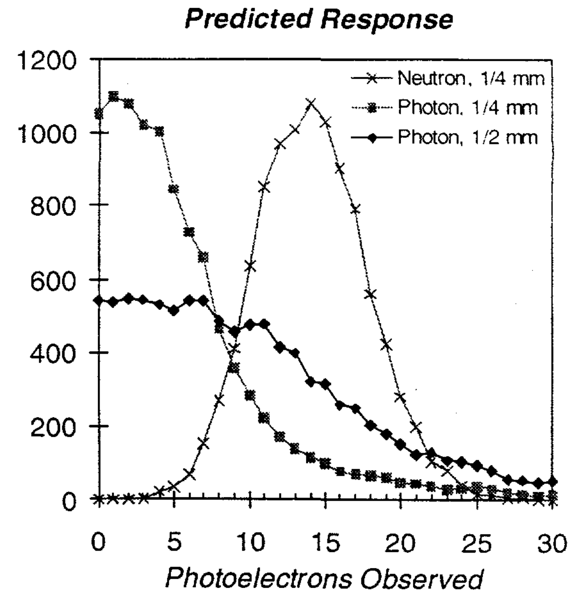

Figure 4. Simulated pulse-height spectra of .25-mm, 0.5-mm, and 1-mm-diameter fibers for fiber diameter selection in order to discriminate against gamma rays..

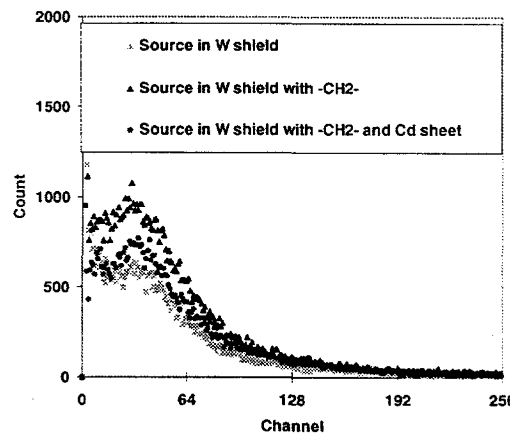

Figure 5a. Spectra of ^{252}Cf neutron source with and without polyethylene cylinders and cadmium liner.

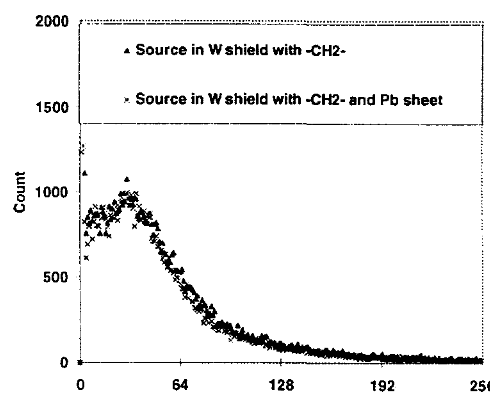

Figure 5b. Spectra of ^{252}Cf neutron source with and without lead liner.