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USE OF NEUTRON-CAPTURE PLASTIC FIBERS FOR NONDESTRUCTIVE ASSAY

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Abstract

Neutron-capture plastic fibers can be used as a nondestructive assay tool. The detectors consist of an active region assembled from ribbons of boron-(^{10}B) loaded optical fibers. The mixture of the moderator and thermal neutron absorber in the fiber yields a detector with high efficiency (ε) and a short die-away time (τ). The deposited energy of the resultant charged particles is converted to light that is collected by photomultiplier tubes mounted at both ends of the fiber. Thermal neutron coincidence counters (TNCC) made of these fibers can serve to verify fissile materials generated from the nuclear fuel cycle. This type of detector may extend the range of materials now accessible to assay by ^{3}He detectors.

Experiments with single fibers of diameters 0.25, 0.50, and 1.00 mm test their ability to distinguish between the signals generated from neutron interactions and those from gamma rays. These results are compared with those obtained from simulation analyses for the same purpose. Light output and attenuation, neutron detection efficiency, and the signal-to-noise ratios of these fibers have also been investigated. The experimental results for light attenuation and neutron detection efficiency are consistent with the values obtained from simulation studies. A comparison of the performance of various configurations of the plastic scintillating fibers with that of other neutron-capture devices such as ^{3}He detectors is also discussed.

INTRODUCTION

Boron-loaded plastic scintillating fibers (PSF) are a mixture of neutron moderator and thermal neutron absorber. The combined effects of these two properties yield a neutron detector with high efficiency and a short die-away time, because of the close proximity of neutron moderator and the absorber. Within a fiber, an 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 3840b for thermal neutrons [1]. The Q-value of the $^{10}B(n,\alpha)^{7}Li$ reaction is 2.792 MeV (94%), with 2.31 MeV going to the charged particles along with a 478-keV gamma ray. Energy deposited by the 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. For fast electrons, Birks [2] 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 is the deposited energy of the charged particle, r is the path length 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.* [3] use another semi-empirical formula based on the work by Birks [2] and Chou [4]:

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

where C is a measured parameter expressed in g^2-cm^{-4}-MeV^{-2}. These functions have been empirically determined for many types of scintillators [5-7]. For α particles, Birks [2] 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 due to the strong nonlinearity of the light response [3]. Although the fiber includes a layer of cladding, its contribution to the interaction is also ignored. Miller [8] 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 "keV electron-equivalent" expresses the light output relative to that obtained for electrons of the specified energy. The elastic scattering of neutrons with hydrogen yields protons with 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 [3]. Therefore, the recoil protons can contribute to the noise, but thermalizing 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.

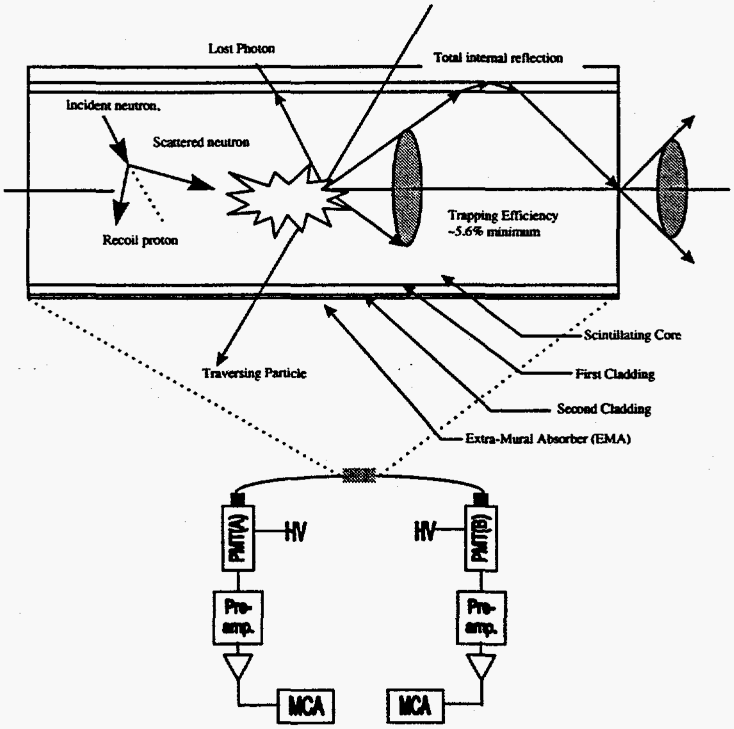

Fig. 1. Thermalization and Subsequent capture of fission neutrons in a boron-loaded PSF. The scintillation light from the resultant charged particles is guided through the PSF to the photomultiplier.

The detection efficiency for a single PSF depends 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, transmission efficiency is nominally about 2.4% which translates to 8,000 photons per MeV from minimum ionizing radiation.* 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 [3]. For a cylindrical fiber with a diameter of 1 mm or less, the minimum trapping efficiency is 3.4% with a single layer of cladding material and 5.6% with multiple layers. An upper value of 12% has been reported for multiclad PSF. The optical attenuation factor is a function of both the material and geometry of the fiber.

Since their introduction scintillating fibers have had numerous applications for radiation detection, but their use in neutron detection via thermalization has not been fully explored. Leutz [9], White [10], and Kirkby [11] provide an excellent overview of scintillation fibers and their potential application as radiation detectors. Axmann [12], Bliss, *et al.* [13], and Ottonello, *et al.* [14] have investigated the use of lithium- and boron-loaded plastic and glass fibers for thermal neutron detection. Angelini, *et al.* [15] compare the characteristics of glass and plastic-scintillating fibers for high-resolution tracking of charged particle beams. Takada, *et al.* [16], Sailor, *et al.* [17], Wurden, *et al.* [18], and Singkarat, *et al.* [3] have investigated the use of PSF for fast neutron detection and spectrometry. Other specific applications of PSF include the work by Yariv, *et al.* [19], Imai, *et al.*[20], Binns, *et al.* [21], Finocchiaro, *et al.* [22], and Agoritsas, *et al.* [23] for radiation detection, track imaging, and profiling low-intensity ion beams.

The most commonly used and effective thermal neutron coincidence counter TNCC in nuclear materials safeguards is the polyethylene-moderated ^{3}He proportional counter [1]. This counter has a relatively high efficiency, ≈ 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 results in a high accidental coincidence rate from random neutrons such as those produced in (α,n) reactions, which results in a longer assay time in order to achieve the desired relative error. One alternative method designed to alleviate this limitation is the use of high-pressure ^{3}He detectors [12, 24, 25]. In this approach, ^{3}He tubes are filled with high-pressure gas (10 atm. as compared to 4 atm. for a standard design) to increase their efficiency and reduce their die-away time. It is, however, unclear how these detectors operate in high gamma-ray application areas, because their sensitivity to the gamma rays is unknown. Another alternative detector design is boron-loaded PSF which is the subject of the remainder of this paper.

When used as a neutron coincidence counter, boron-loaded plastic detectors can achieve a much shorter die-away time and a high neutron detection efficiency. Using computer simulations, Miller [8] reports that a 50% efficiency with 3-4 μs die-away time can be expected from BC454† BGO-phoswich detectors. Experimental validation of similar computer simulations, with 10% efficiency and 7 μs die-away time, has been recently performed. With a shorter die-away time, more sensitive and accurate determination of the actual sample mass may be possible. Various configurations of 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 (Fig. 1). 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 monolithic BC454/BGO. Furthermore, the fiber detectors are less sensitive to gamma rays because of the small diameter of the fiber. Used in multifiber ribbons, the signals from the scintillating fibers can also be logically sorted for additional gamma-ray discrimination.

SIMULATION RESULTS

The efficiency and die-away time of several configurations of the proposed detector system have been examined using simulated data obtained from MCNP4B with the ENDF/B-V cross section library [26]. A basic arrangement of the system appears in Fig. 2. The geometry consisted of a can, 18 cm in diameter and 25 cm tall. A polyethylene liner wrapped with alternating layers of polyethylene sheets and fiber ribbons surrounded the sample. A ribbon consisted of 0.25-mm fibers in a single layer. To simplify the geometry, the ribbons and polyethylene sheets were reduced to alternating concentric cylinders around the central can. To minimize neutron leakage, a nickel reflector was placed around the ribbons, and graphite plugs on the top and bottom of the ribbons and sample cavity.

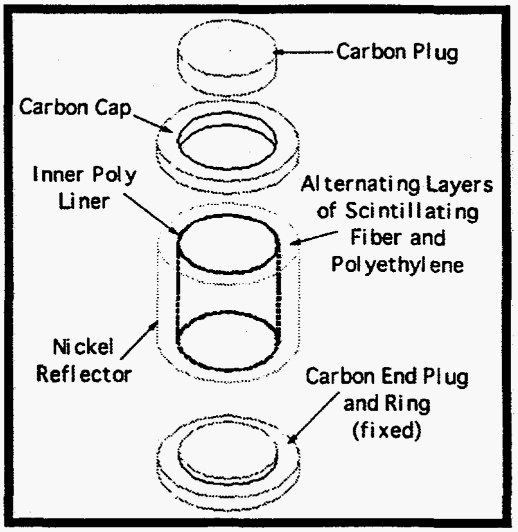

Fig. 2. Basic geometric arrangement of the proposed detector system for the simulation analyses.

The simulations investigated both ^{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 $a=0.799$ MeV and $b=4.903$ MeV^{-1} for ^{240}Pu. The goal of the simulations was to optimize the efficiency and die-away time of the detector by varying the thickness of the moderator while maintaining a constant number of ribbons of 0.25-mm fibers. The efficiency of the detector system was 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 because the range of the alpha particle is on the order of a few micrometers. The die-away time was calculated by taking the time distributions of ^{10}B events. With a layer of 0.41 mm of cadmium between the source and the first layer of polyethylene, the die-way time varied from 6.7 to 10.1 μs with corresponding values of efficiency ranging from 39.6% to 51.3%. Without the cadmium absorber, the detector die-away time varied from 5.2 to 14.1 μs. The detector efficiency varied from 6.0%to 61.9%. The influence of the moderator thickness on the detector die-away time and efficiency is shown in Fig. 3.

Another series of simulations compared the deposition of neutron and gamma-ray energy in a single fiber to determine the best fiber diameter for neutron/gamma-ray discrimination. As shown in Fig. 4, it is impossible to distinguish between the energy deposited from a 1-MeV photon and that of a ^{10}B neutron capture event with a 1-mm fiber. This finding is consistent with the observation by Abel, *et al.* [27]. 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, 0.25-mm-diameter fibers were selected for additional simulations and the final design. The light output of a ^{10}B neutron capture event in the fiber is equivalent to a 93±1-keV electron [8, 28], which corresponds to the peak in the neutron capture spectrum.

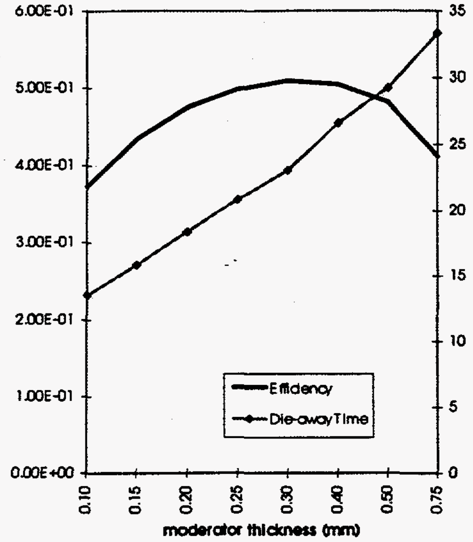

Fig. 3. Efficiency and die-away time as a function of polyethylene moderator sheet thickness.

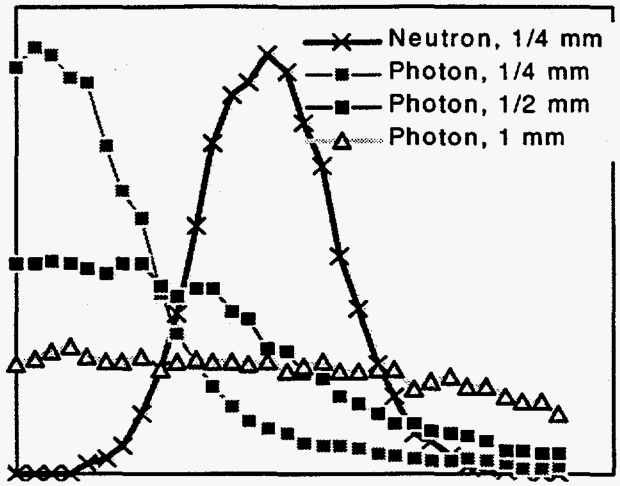

Fig. 4. Simulated pulse-height spectra of 0.25-, 0.5-, and 1-mm-diameter fibers for fiber diameter selection to discriminate against gamma rays.

BENCHMARKING

To validate the results from analytical and simulation analysis, a series of experiments were conducted. First, the energy calibration was measured using a BC454 log (with a diameter of 7.5 cm and a length of 30 cm) and ^{137}Cs, ^{22}Na, and ^{109}Cd with activities of 10 μCi, 10 μCi, and 100 μCi, respectively. These sources emit 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 peak from the ^{10}B neutron reaction was found to be 93 keV, which is consistent with that reported by Miller [8].

Following these calibration measurements, 1-mm-diameter fibers were used to determine the sensitivity of boron-loaded PSD to neutrons and gamma rays and the maximum fiber length that could resolve a peak. Initially, 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 coupled directly to a Burle S83049F photomultiplier tube.

Comparing the spectra with and without the polyethylene cylinders, an increase in the counts between channels 16–64 with the former arrangement was observed. This increased count was due to thermal neutron capture because it dropped with the addition of a cadmium absorber. The sum of channels 16–64 yielded 14842 counts in that peak area. This is a factor of 2 higher than MNCP prediction of 7960±1042 for ^{10}B-capture events. This discrepancy in counts is due to the fact that the simulated model counts only the (n,α) reactions. The counts from the experiments, however, can stem from several factors such as the recoil protons and the gamma rays interacting in the fiber in addition to the (n,α) reactions. A similar relationship was found between the measured and simulated spectra with and without the cadmium liner, counting about twice as many events as those predicted by MCNP. The results, however, clearly demonstrated the sensitivity of the boron-loaded PSF to gamma rays and thermal neutrons.

In another set of experiments, a ^{237}Np source was placed at the end of 34-, 67-, and 134-cm long, 1-mm-diameter fibers. The goal in this experiment was to determine the maximum fiber length that can resolve an alpha peak. Neptunium-237 emits alpha particles in the range 4.6–4.9 MeV. The alpha peak was clearly seen with the 34- and 67-cm long fibers, but could not be resolved with the 134-cm fiber. Furthermore, scintillation light from a ^{90}Sr beta source resulted in attenuation lengths ranging from 1 to 1.5 m with a 1-mm fiber. Simulation analyses and experiments with a green monochromatic pulser resulted in an attenuation length of 2.3 m, which is consistent with that reported by the PSF manufacturer [29].

FUTURE PLANS

Current experiments include a 200-fiber ribbon tested under the same conditions as those for the single fibers. The fiber diameter is 0.25 mm. While it appears impractical to test single fibers of this diameter, the ribbon should improve the signal-to-noise ratio. The signals from this ribbon could help to determine if logical discrimination in conjunction with pulse-height discrimination could eliminate gamma-ray events. In addition, a configuration of ribbons of the PSF interlaced with thin layers of polyethylene could further validate the expected performance of boron-loaded plastic fibers as a neutron detector. The plan for the test ribbon includes a coincidence counter setup to count known ^{240}Pu and ^{252}Cf samples. Further tests with thermal neutron beams from a reactor and a high gamma-ray flux environment are being considered. The results of these measurements will be used to optimize the final system.

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[*] Scintillating materials manufactured by Bicron Corporation, Newbury Ohio.

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