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Author(s): **D. R. Mayo , N. Ensslin, R. Grazioso, A. S. Heger, D. Mercer, M. Miller, P. Russo, and M. Sweet**

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Applications of Boron-Loaded Scintillating Fibers as NDA Tools for Nuclear Safeguards

Douglas R. Mayo*, Norbert Ensslin*, Ronald F. Grazioso†, A. Sharif Heger†, David J. Mercer*, Michael C. Miller*, Phyllis A. Russo*, and Martin R. Sweet*

*Los Alamos National Laboratory[1]
Los Alamos, New Mexico 87545
† *University of New Mexico*
Albuquerque, New Mexico 87131

Abstract. Nuclear safeguards and nonproliferation rely on nondestructive analytical tools for prompt and noninvasive detection, verification, and quantitative analysis of nuclear materials in demanding environments. A new tool based on the detection of correlated neutrons in narrow time windows is being investigated to fill the niche created by the current limitations of the existing methods based on polyethylene moderated ^{3}He gas proportional tubes. Commercially produced Boron-loaded (^{10}B) plastic scintillating fibers are one such technology under consideration. The fibers can be configured in a system to have high efficiency, short neutron die-away, pulse height sensitivity, and mechanical flexibility. Various configurations of the fibers with high density polyethylene have been considered which calculationally result in high efficiency detectors with short die-away times. A discussion of the design considerations and calculations of the detector efficiency, die-away time, and simulated pulse height spectra along with preliminary test results are presented.

INTRODUCTION

Neutron coincidence/multiplicity counting (NCC/NMC) is a nondestructive assay (NDA) approach applicable to the bulk assay of plutonium [1]. The samples in question are residues, aged compounds, and other impure forms. The NCC method addresses safeguards needs by quantifying plutonium stored in thousands of cans and drums. NCC systems have the advantages of economy and safety, and the benefits of ruggedness and reliability.

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The detection of multiple neutrons that are correlated in time from their point of origin, i.e., the fission of an actinide nucleus, is a powerful tool in nuclear safeguards and nonproliferation. Neutrons, unlike gamma rays, penetrate dense shielding or large quantities of nuclear materials. The requirement of neutron coincidences is effective in eliminating background events. Passive NCC/NMC of plutonium uses the count rate of correlated neutrons from spontaneous fission to determine the mass of even-A isotopes, with ^{240}Pu usually the major even isotope present. Active NCC/NMC of uranium by interrogation with uncorrelated (i.e., not fission) neutrons at energies below threshold for the fission of ^{238}U induces fission in ^{235}U to produce a count rate of correlated neutrons that is a measure of the mass of ^{235}U.

The current detector technology typically thermalizes neutrons in a polyethylene moderator and captures the neutrons in a ^{3}He gas tube detector. The detector and moderator are macroscopically distinct and separate, resulting in a wide (in time) coincidence gate for counting correlated neutrons because of the long (50-100 μs die-away) time between thermalization and capture. Although these detector systems provide discrimination against gamma rays, high neutron detection efficiency, and reliable, stable, long-term performance in variable environments, numerous field applications, including many *in-situ* measurements, are precluded by the long die-away time.

Because of the relatively long neutron die-away times in polyethylene-moderated thermal (^{3}He) neutron counters, NCC assay sensitivity is limited by high accidental coincidence rates from α,n (uncorrelated) neutron yields. The uncorrelated neutron yields can greatly exceed the yields of (correlated) fission neutrons produced by these impure materials. Sensitivity is governed by the neutron detection efficiency and the total neutron count rate, which determines the accidental coincidence rate. The sensitivity of the NMC assay decreases as the fraction of uncorrelated neutrons in the total neutron signal rises. For materials with neutron yields dominated by uncorrelated neutrons, the NMC assay precision and its sensitivity (in g of ^{240}Pu or ^{235}U) are lessened by the greater excess of uncorrelated neutrons.

An approach to overcome the uncorrelated neutron limitation in thermal counters is to homogenize the moderator and detector materials to shorten die-away time, thus reducing the accidentals fraction. For this approach to be successful, it must be done without sacrificing the high neutron detection efficiency and essential gamma-ray discrimination capability provided by (^{3}He) counters. A boron-loaded plastic scintillator that combines detector (^{10}B) and moderator (plastic) at a molecular level for order-of-magnitude reductions in neutron die-away time has been developed and investigated at Los Alamos National Laboratory (LANL) [2]. When used in monolithic forms such as rods or blocks, high neutron detection efficiency is readily achieved, neutron die-away time is reduced by more than a factor of ten, and benefits of neutron spectroscopy are also realized. To eliminate signals from gamma rays, capture events are tagged by simultaneous detection of capture gamma rays with an inorganic scintillator bonded to the plastic. A prototype counter that uses this approach is being tested at LANL [2].

Boron-loaded plastic can be extruded in the form of fibers. Reducing the diameter of a cylinder of plastic from rod to fiber dimensions results in a reduction in the amplitudes of pulses that arise from gamma-ray interactions, possibly enabling threshold gamma-ray discrimination. The reduction in diameter of a fiber limits the track length of electrons passing through a single fiber, and consequently the signal measured is less. From a practical standpoint, the fiber's characteristics lend themselves to a unique combination of capabilities: short coincidence resolving times, pulse-height discrimination, logical sorting, three dimensions of position sensitivity, and flexibility of configuration. Additional capabilities could include detection of neutrons with high efficiency and discrimination against gamma rays, both of which are essential criteria that are presently achieved with existing neutron-capture detectors.

Gamma-ray discrimination is further enhanced by the logical sorting of fiber readouts to eliminate multi-fiber gamma-ray events within a layer. The enhancements provided by the new detector will extend the measurement sensitivity, characterize the neutron energy spectrum, enable the imaging of the actinide materials, and support multiple applications by mechanical reconfiguration.

DETECTOR SIMULATION AND OPTIMIZATION

A well counter for a can that is 18-cm diameter x 25-cm tall was simulated to check the feasibility of the proposed detector. A crude design is shown in Figure 1. The alternating layers of scintillating fiber and polyethylene are achieved by wrapping a stack of 3-m long boron-loaded fiber ribbons interleaved with thin flexible polyethylene sheets. The photon energy deposition in fibers and neutron capture probabilities in the ^{10}B were calculated using the MCNP code [3].

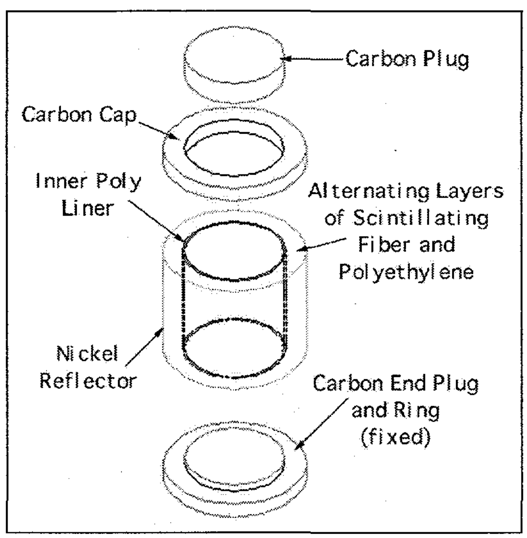

FIGURE 1.: Conceptual diagram of detector for 18-cm diam. x 25- cm tall cans.

Additional constraints were placed on the design with an attempt to maintain a cost of the overall system, comparable to that of a ^{3}He NCC system. A detector made solely of ribbon layers would be costly. As such, thin sheets of polyethylene between ribbon layers were considered as a means to increase efficiency by moderating neutrons and decrease cost by lessening the amount of fiber needed. Configurations were tested in simulations to maximize the efficiency (ε) while minimizing the die-away time (τ), as shown in Figure 2, to

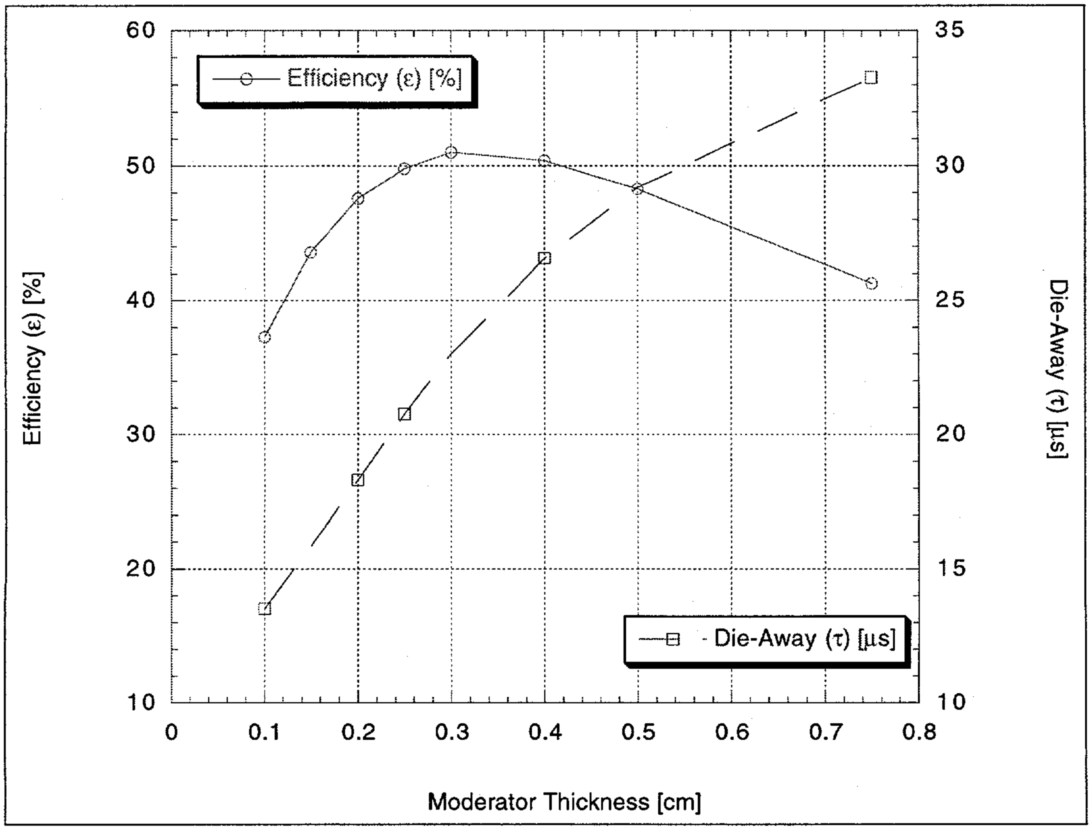

FIGURE 2. Efficiency (ε) and die-away (τ) for a 32 ribbon detector (2 stacks of 16) as a function of polyethylene sheet thickness between ribbon layers. The simulations were performed for 0.5-mm fibers.

produce an optimal detector that meets the design criteria.

Photon and Neutron Discrimination

Early calculations with 0.5-mm diameter fibers pointed to the economic feasibility of layering polyethylene between each of the ribbon layers while achieving an $\varepsilon \approx 50\%$ with a $\tau \approx 10\mu s$. As determined experimentally in Abel, *et al.* [4], the 1.0-mm boron-loaded plastic fibers were incapable of separating electron and neutron events on the basis of a threshold cut. This observation led to a study of energy deposition as a function of fiber diameter as shown in Figure 3. The capture of a neutron by ^{10}B and subsequent emission of an α-particle produces a 93-keV electron-equivalent light output [2]. The simulated data shown in Figure 3 is in terms of the number of photoelectrons produced by the photocathode of the photomultiplier tube (PMT). The figure points out that for a 1-MeV photon incident upon a bare fiber, the photon events in 1.0-mm and 0.50-mm fibers can not be separated from the neutron induced events. The 0.25-mm fiber shows promise. By reducing the diameter, however, the amount of energy the Compton scattered electrons can deposit decreases, thus allowing for the possible discrimination of

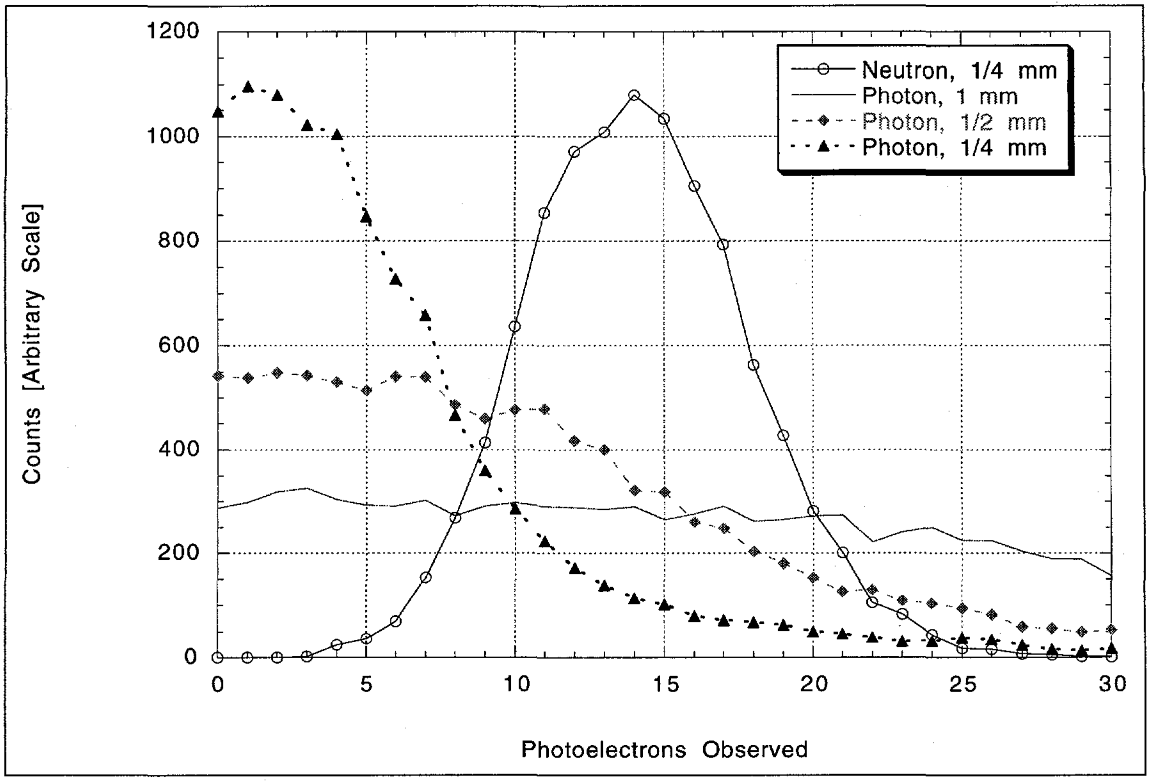

FIGURE 3. Simulated number of photoelectrons produced in the photomultiplier photocathode due to neutron and photon events produced in 1.0-mm, 0.50-mm, and 0.25-mm fibers.This model assumes that the light collected from neutron-induced events are independent of the fiber diameter as the range of the α-particle is a few μm.

photon and neutron events.

The model used to generate the number of photoelectrons assumes geometric (ray-like) light propagation. The events are randomly distributed along the 2.5-m length of the fiber. The attenuation length was taken to be 2.2-m, which has since been confirmed in a benchtop experiment for a 1.0-mm fiber to be $\approx$ 2.3-m. At this level of simulations, bending and coupling losses were assumed to be negligible compared to the light lost due to attenuation and reflection/transmission at the core/cladding interface. The interactions assumed single wavelength emission with a 20% quantum efficient tube. The model represents an idealistic result, not accounting for the PMT resolution and other factors which tend to smear the response and/or reduce the number of photoelectrons produced.

Efficiency and Die-Away Time

The efficiency and die-away time of a given detector geometry were simulated using MCNP. The neutrons were generated assuming a ^{252}Cf point source with a Watt's fission spectrum

TABLE 1. Summary of 0.25-mm fiber Simulations.

Case #	Inner Moderator Thickness [cm]	CH_2 Moderator Thickness [cm]	Efficiency ε [%]	Die-Away Time τ [μs]	Ribbon Stacks
1	1.0	0.2	47.6	18.3	2
2	1.0	0.1	37.3	13.5	2
3	1.0	0.1	54.9	12.4	4
4	0.5	0.1	55.3	11.9	4
5	0.5 0.041 Cd	0.1	51.3	10.1	4

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

where $a = 1.025$ and $b = 2.926$. This differs slightly from the neutron spectra of plutonium, but the codes will initially be benchmarked with ^{252}Cf sources. The efficiency was calculated from the total number of neutrons captured by the ^{10}B. It was assumed that all of the captures resulted in a detectable signal. The fraction of capture events that are recorded by the PMT is a quantity that can be determined through benchtop experiments.

The die-away time can be calculated by taking the time history from the simulations. A single exponential was assumed for the function, and the die-away parameter was calculated from the fit. Table 1 lists some of the results from different configurations for 0.25-mm fibers. The detector optimization involved varying the thickness of polyethylene sheet between the ribbon layers as well as the thickness of the polyethylene inner shell, adding the layers of Pb and Cd in the well for shielding, adding graphite top and bottom plugs and introducing neutron reflecting material on the exterior. The graphite end plugs and external nickel reflector increased the overall efficiency without a large increase in neutron die-away time. The effect on die-away time and efficiency as a function of moderator thickness is shown in Figure 2.

TESTING AND BENCHMARK PLANS

Prior to investing the time and resources in building a prototype detector system, a number of tests have been considered to prove the feasibility of a boron-loaded scintillating fiber NCC system. Currently a set of 1.0-mm, 0.50-mm, and 0.25-mm singly clad boron-loaded fibers are being tested. The first goal of the project is to determine the idealized characteristics of neutron detection with independent fibers and to compare the characteristics for different fiber sizes and configurations. A validation of the calculations, including a determination of what, if any, bias exists, prompted the benchmark experiments which are being performed. The set of test fibers will allow for a comparison similar to that shown in Figure 3.

A set of 0.25-mm multiclad fibers will be compared to the singly clad fibers used to provide information on the benefits of the additional coatings. A 3.0-m ribbon of 0.25-mm fibers (200 fibers wide) will be used in the construction of a miniature well detector which will provide benchmark data and allow for experimenting with different layers of polyethylene and metal foils.

Preliminary Data

Initially the electronic equipment were setup with a 2.54-cm diam. x 1.27-cm thick bismuth germanate (BGO) crystal mounted on a Hamamatsu 5-cm R329-02 PMT. The preamplifier pulse passed through an ORTEC 855 dual spectroscopy amplifier to a Canberra 1510 ADC, which was fed into a Canberra S100 card and read into MCA software. The BGO provided easily resolved photoelectric peaks to test the electronics.

The BGO cylinder was replaced by a 2.54-cm diam. x 2.54-cm thick BC454 plastic cylinder to determine whether neutron events could be observed. Figure 4 shows the clear neutron peak from a ^{252}Cf source along with the scattering continuum underneath.

A cylinder was made from a hollow polyethylene annulus with $\approx$ 100 of the 1.0-mm fibers, 2.54-cm long, epoxied in the center and polished on both ends. The fiber cylinder was considered a first step in determining whether neutron events would stand out without the effects of light propagating through a wrapped 2.5 - 3.0-m long fiber. Going from a 2.54-cm aperture in the case of the BC454 crystal to a 1.0-mm aperture in the fiber should reduce the light collected by a factor $\approx 4-10$ (numerical aperture). The data indicate that the tubes currently used may not be optimal for this purpose, and that a lower noise level and better energy resolution is required. Several tubes are being considered, including the hybrid tubes.

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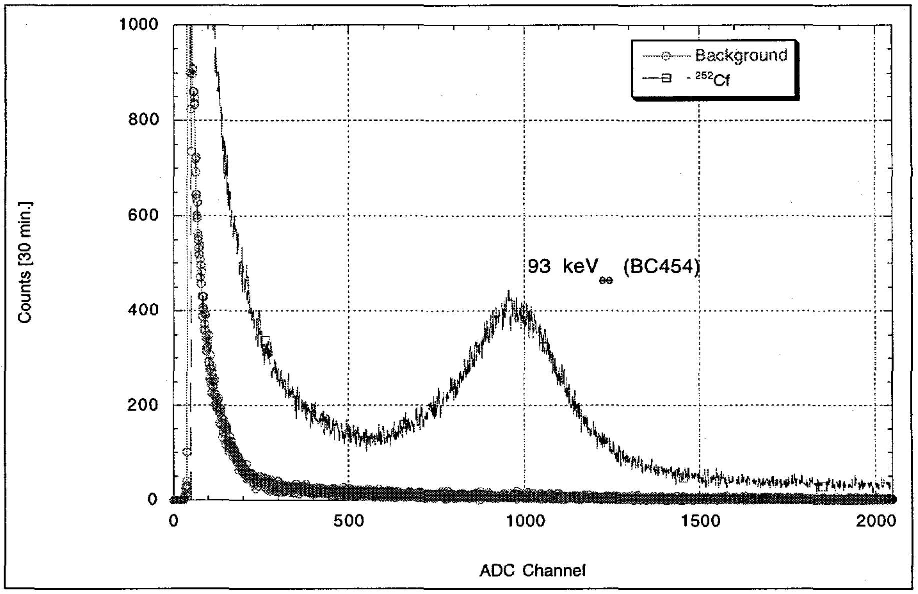

FIGURE 4. Background and moderated neutron spectra from 2.54-cm diam. x 2.54 cm BC454 cylinder. The neutron capture peak is clearly seen.

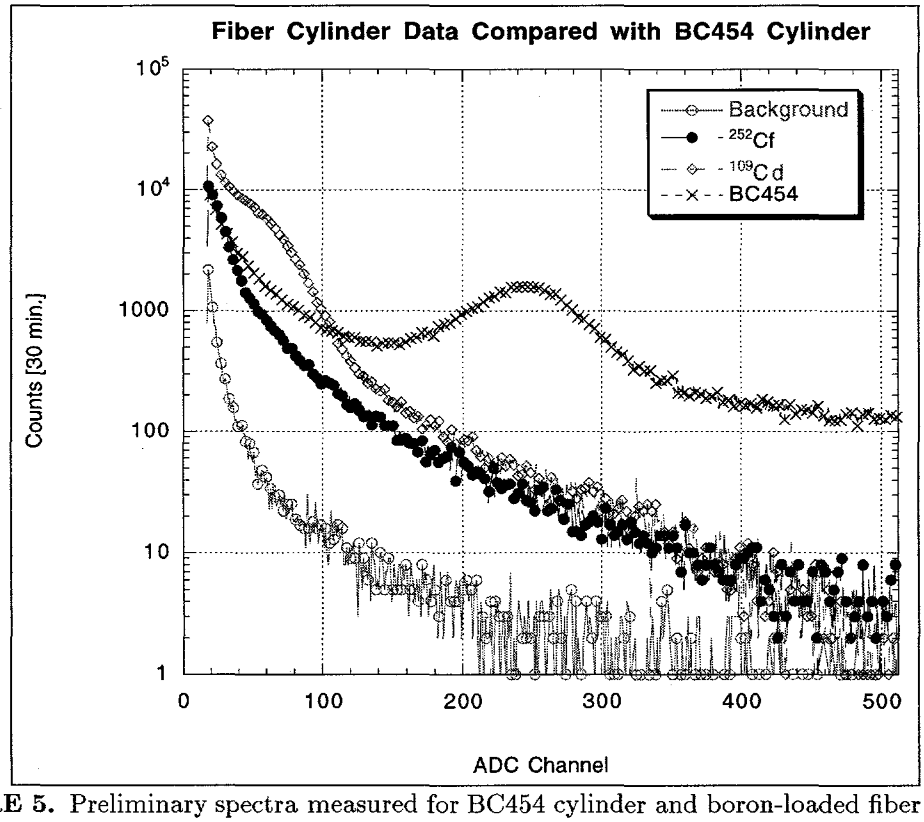

FIGURE 5. Preliminary spectra measured for BC454 cylinder and boron-loaded fiber cylinder. Although the ^{252}Cf spectra measured by the fibers does not exhibit an distinct peak, it does show a response to neutrons.