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# A VARIETY OF NEUTRON SENSORS BASED ON SCINTILLATING GLASS WAVEGUIDES

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# A VARIETY OF NEUTRON SENSORS BASED ON SCINTILLATING GLASS WAVEGUIDES

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## ABSTRACT

Pacific Northwest Laboratory (PNL) has fabricated cerium-activated, lithium-silicate glass scintillating fiber neutron sensors via a hot-downdraw process. These fibers typically have a transmission length ($e^{-1}$ length) of greater than 2 meters. The underlying physics of, the properties of, and selected devices incorporating these fibers are described. These fibers constitute an enabling technology for a wide variety of neutron sensors.

## INTRODUCTION

The development of neutron-sensitive scintillating fibers was initiated to produce large-area, solid-state, thermal-neutron detector packages which are insensitive to gamma rays. Based on experience and literature data,[1] it was determined that cerium-activated, lithium-silicate glass-fiber waveguides would be the best candidate for the active part of the detector packages. Because such fibers are not available commercially, a fiber fabrication facility was developed.

The glasses are lithium aluminosilicates activated with cerium. It is well-known that this composition family scintillates in the presence of a thermal neutron flux.[2,3,4] These glasses are drawn into scintillating waveguides using a hot-downdraw fiberizing tower. The glass fiber is clad with a thermal-curing silicone that serves as both an optical cladding and a physical buffer. The use of a polymer for the cladding gives the waveguides a high numerical aperture, which is vital for sensing neutron interaction.

As recently as 1993, cerium-activated lithium-silicate fibers were described as having "...useful lengths of fiber [limited] to <10 cm."[5] By developing glass compositions specifically for fiber drawing, using strict oxidation-state controls, and quenching rapidly, fiber with useful lengths in excess of 200 cm can be produced. The use of a hot-downdraw system to manufacture the waveguide avoids crystallization of the lithium-rich glass.

The active portion of the sensor is based on detecting scintillation induced in the glass by a $^6Li(n,\alpha)^3H$ reaction. This reaction is exothermic, releasing approximately 4.7 MeV of which the majority is carried away by the triton (Fig. 1). The triton and alpha both interact with the glass matrix to produce a trail of electronic excitations (ionization). These excitations can transfer energy by exciting $Ce^{3+}$ ions that will emit optical photons as they relax to the ground state. An optical waveguide is formed when the fiber cladding has a smaller refractive index than that of the core. In this case, a fraction of the scintillation photons will be trapped in the fiber and guided towards each end. The fraction of the scintillation light that is captured is determined by the ratio of the cladding-to-core refractive index. For the silicone-clad, lithium-aluminosilicate fiber, the captured fraction is approximately 0.033 towards each end.

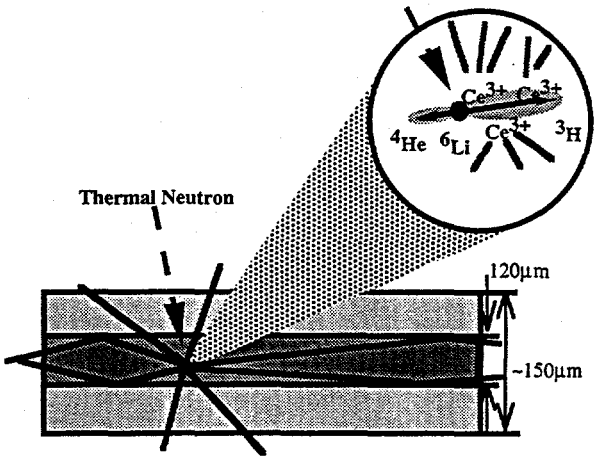


Fig. 1. Schematic showing the major nuclear, electronic, and optical processes in the detection of a neutron with a scintillating-fiber waveguide.

Sensors for a wide variety of applications have been produced. Sensors may consist of a single fiber or tens of thousands of fibers, depending on the neutron flux to be detected.

## FABRICATION

Lithium carbonate obtained from Oak Ridge National Laboratory is purified via a recrystallization process. The lithium is 97% enriched in $^6Li$. All other raw materials are fiber-optic grade. Melting is done in a platinum crucible under reducing conditions in an induction furnace. Melts are usually batched to produce 100 g of glass, and yields are typically over 99%. Melts are quenched in air by pouring onto water-cooled plates. The glass remaining in the crucible is quenched by placing the crucible into a water bath. The water is not allowed to contact the glass. Premelting the batch allows for preliminary characterization of the glass and prevents off-gases from contaminating the fiber-drawing system.

To draw the fiber, the glass cullet is placed into a platinum bushing. The bushing has a single orifice. The bushing is heated in a controlled-atmosphere induction furnace. A cold finger seals the tip of the bushing until the glass is ready to draw. The fiber draw is started by removing the cold finger and then pulling a small gather of glass from the bushing tip. The fiber is then threaded through the drawing tower to the take-up drum. When the fiber draw is established and stabilized, the cladding cup is placed around the fiber and a flow of silicone cladding material is started (Fig. 2). The take-up drum translates as it rotates to allow the manufacture of mono-and bi-layer ribbons up to 2 m long.

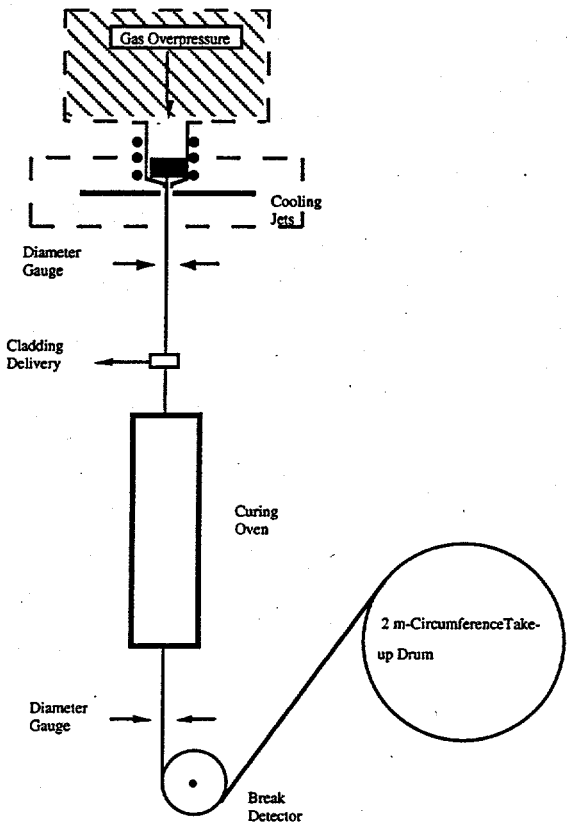


Fig. 2. Schematic of the PNL hot down-draw tower. The tower is approximately 2 m high.

To make ribbons, the fibers are bound together while still on the drum, using a room-temperature, air-curing silicone; an allowance of 10 to 20 cm must be left free of adhesive at each end of the ribbon. The fibers are then cut off the drum, usually in 1- or 2-meter lengths, and processed into detector panels. The loose fiber ends are gathered into ferrules, glued with another heat-curing silicone, which is moderately stiff, and cut even with a slow-speed diamond saw. The fiber ribbon is then sandwiched between two sheets of Gella™ to protect it from water vapor and provide some neutron moderation.

## FIBER TRANSMISSION

The transmission length ($e^{-1}$ distance) of the fiber is routinely measured using ultraviolet (UV) fluorescence excitation. A sample fiber, typically 2 meters long, is cleaved and is placed at the focus of a fiber-optic spectrometer. The fiber is laid on a flat 2-meter-long reflective surface. A mercury-vapor lamp (mounted on a rail system) with a solar-blind filter illuminates approximately 1 cm of fiber (Fig. 3). The mercury-vapor lamp excites the $Ce^{3+}$ in the fiber, which fluoresces. The behavior of the light is identical with that for the nuclear excitation; however, the intensity of the fluorescence is many orders of magnitude greater than that under neutron stimulation. Periodically, transmission values obtained using UV stimulation are compared to values obtained from fiber ribbons using

nuclear excitation. These values are within the uncertainty normally obtained for single-fiber measurements (± 8 cm transmission length).

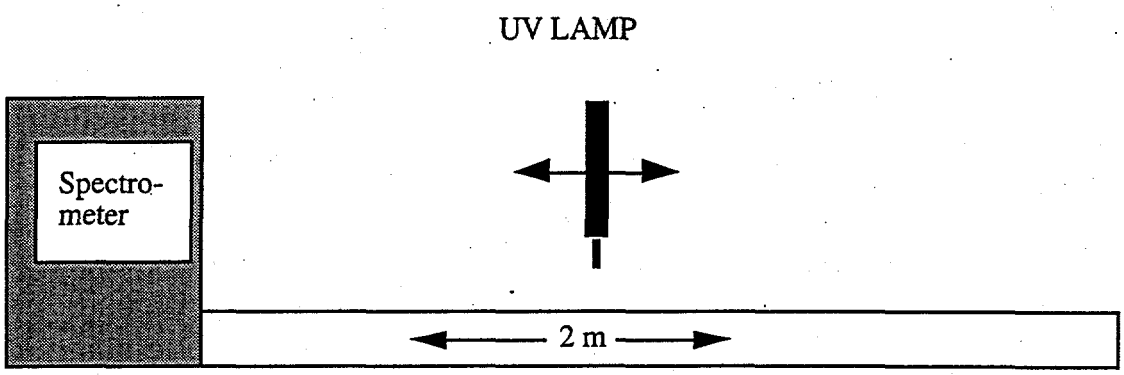


Fig. 3. Set-up used to measure fiber transmission. The UV lamp is used to excite fluorescence in approximately 1 cm of the fiber.

Figure 4 shows the data that were measured on a single fiber to 10 m away from the detector. To compare various fibers to each other, the transmission length is used as a quality factor. The transmission length is taken from the best linear fit to the slope (on a logrithmatic plot) of the integrated intensity versus distance between points 20 cm and 80 cm away from the detector. It is necessary to define a distance range because the transmission varies non-exponentially as the excitation point is moved away from the detector. The 20-to-80-cm range was chosen because it is physically easy to measure and is relevant to the use of the fibers in actual detectors.

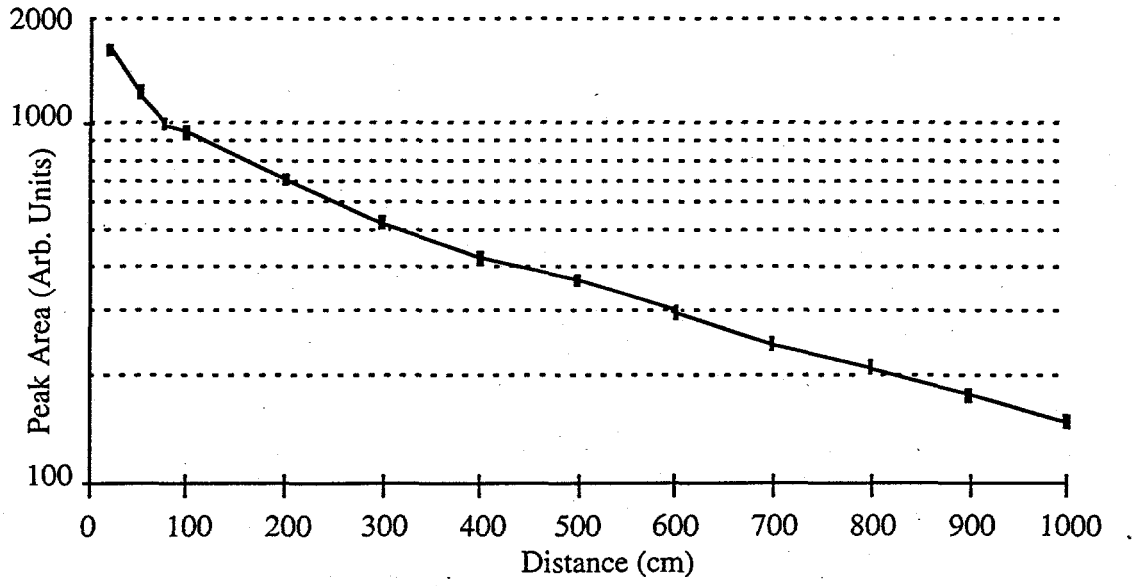


Fig. 4. Integrated fluorescence peak area as a function of distance from the detector. The short-distance region has a 1/e value of approximately 1 m. The long-distance region has a 1/e value of approximately 5 m.

## SELECTED DEVICES

A number of devices have been fabricated or are under development based on the PNL neutron-sensitive scintillating-fiber technology. Several prototype detectors for high- and low-flux

applications are described to illustrate the utility of this technology.

Figure 5 shows pulse-height spectra of radiation-induced events detected by a single fiber for neutron and for beta excitation. Beta-induced excitation is representative of that produced by gamma rays because the gamma ray interacts with a scintillator via a Compton electron or photoelectron. Electrons produce much smaller pulses than neutrons because the range of the electron is much greater than the diameter of the fiber. It is seen that neutron events produce a broad isolated peak centered at channel 55, which electron events do not.

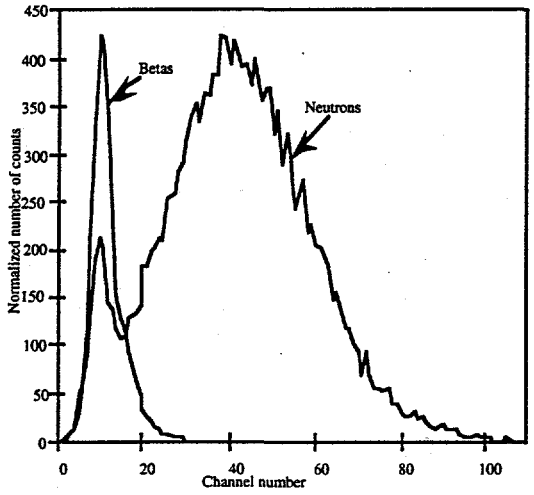


Fig. 5. Pulse-height spectra of a single scintillating glass fiber in neutron and beta-ray fields using two photomultipliers at opposite ends of the fiber in coincidence.

Fig. 6 illustrates a concept for a real-time dosimeter to be used in boron-capture neutron therapy.[6] Boron-neutron capture therapy (BNCT) is an experimental treatment for a class of malignant brain tumors. Dose rates in BNCT are, however, particularly difficult to calculate. Calculations must deal with beams that are a mix of neutrons of various energies. The cross-sections for the desired therapeutic reaction and reactions with natural nuclei in the patient are functions of the energy of the neutron and vary by several orders of magnitude for an epithermal beam mix. Thus, it would be highly desirable to have a real-time measure of dose rate at the delivery site that is also proportional to the other reaction rates of interest. Current practice for BNCT dose measurement is to implant a gold wire at, or near, the tumor-treatment site. Part way through the treatment, the wire is removed and neutron-activated radionuclides are counted to determine the time required to complete the treatment. The sensitivity of PNL fibers to neutron energy varies in the same way as that of the boron; therefore, their use provides the possibility for measuring the dose rate in real time. These devices are currently being tested.

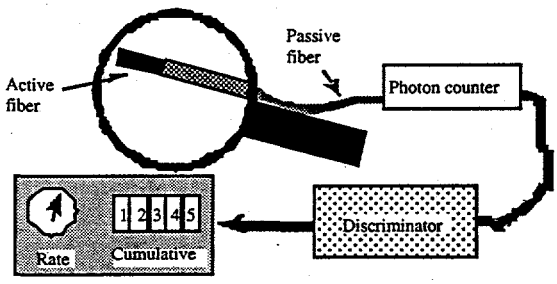


Fig. 6. Conceptual design of real-time neutron dosimeter for use in BNCT relying on neutron-sensitive scintillating-glass waveguides for the active element.

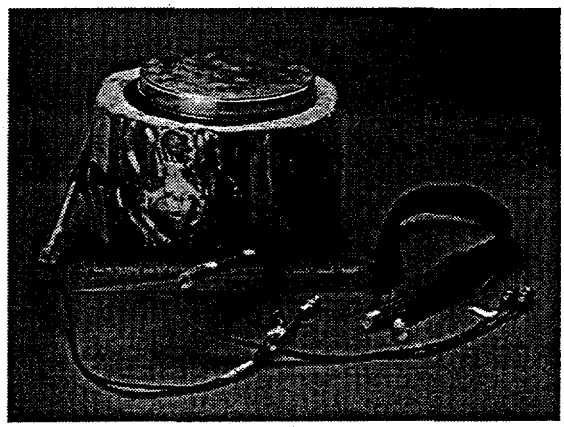

Fig. 7. Prototype of the neutron-sensing fiber-optic detector. The detector was designed to be the equivalent of a 7.5-cm-diameter $^{3}He$ tube wrapped around the sample. The samples were contained in a 20-cm-high by 20-cm-diameter can.

Figure 7 shows a detector designed to provide continuous monitoring of a storage can.[7] This detector was designed to encircle a small storage container ~20 cm tall and ~20 cm in diameter. Such containers are used to store solid scraps of plutonium and plutonium compounds that are by-products of weapons production. By monitoring the neutron flux emanating from a storage container, the integrity of its contents can be ensured. This detector exhibited standard deviations from the mean in neutron counting rate of ~3% for samples emitting on the order of $10^5$ neutrons/sec. This translates to a sensitivity, at, 3 standard deviations, of approximately $10^3$ neutrons/sec emitted. One gram of weapons-grade plutonium oxide emits approximately $10^2$ neutrons per second.[8]

For large-area detectors made from layers of fiber ribbons, the fibers are gathered into bundles to attach to photomultiplier tubes (PMTs). Figure 8 shows a typical 2-m fiber panel bilayer. Typically, all the fibers from a single bilayer of a panel are gathered into a single bundle. (Top end of panel in Fig. 8.)

Fig. 8. Typical 2-m scintillating-fiber bilayer. See text for description.

Fig. 9. Neutron-sensing panel to be installed in roadbed.

Figure 9 shows a detector fabricated from a bilayer similar to that shown in Fig. 8. All detector electronics, including power supply, logic circuitry, and analysis functions, are included in the package. This detector was developed to demonstrate the capability of detecting nuclear weapons in vehicles. The detector was constructed with a minimal quantity of moderator; the roadway material was intended to function as moderator. The device demonstrated the capability of detecting and distinguishing simulated nuclear weapons passing at traffic speeds.

To enhance the discrimination against gamma rays, PNL flat detector panels are fabricated with PMTs on both ends (Figs. 7 and 8). Most of the sensitivity to gamma rays comes from the region near the PMT at which the fibers are bundled. The added thickness of the bundle region increases the probability that a gamma will result in a valid count. Depending on the application, improved gamma-ray rejection is desirable. In such cases, fibers from several stacked bilayers are collected and additional coincidence techniques applied. (Bottom end of panel in Fig. 8.) Sum and coincidence requirements in such configurations give a counting efficiency ratio for neutrons, relative to gammas, of greater than 8000 when a coincidence-plus-a-sum requirement of four or more photoelectrons is adopted.

## OTHER APPLICATIONS

The most persistent of the radionuclides contaminating defense production sites include neutron emitters. Because neutron-sensing scintillating optical fibers are flexible, these can be used as large-area detectors in application-specific geometries. They can be wrapped around a drum to assay its sealed contents. They can also be used in a flat-plate geometry for measuring soil concentrations of radionuclides. Thus, this technology can be used in environmental cleanup operations.

Also, in an application that has yet to be tested, neutron-sensing scintillating optical fibers can provide ultra-high spatial-resolution detection for thermal neutron scattering in research applications. Neutron scattering is used in materials-science research to determine the structure of materials. In this kind of research, a beam of cold neutrons is incident upon the material to be studied. The scattered neutrons are measured at a distance; the angular distribution of scattered neutrons provides the information about the structure of the material under study. Presently, an array of $^3He$ tubes (with a diameter of ca. 6 mm), arranged like a picket fence, defines the spatial resolution of the scattered neutrons. Using PNL fibers instead of $^3He$ tubes could improve this resolution by approximately a factor of 40. This, in turn, would provide improved information about the material structure and the mechanisms that operate in condensed matter.

## ACKNOWLEDGEMENTS

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