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FERMILAB-Conf-98/123

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August 1998

Published Proceedings of the *Scintillating Fibers '97*,
University of Notre Dame, Indiana 46556, November 3-6, 1997

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Abstract. A decade of research into plastic scintillation materials at Fermilab is reviewed. Early work with plastic optical fiber fabrication is revisited and recent experiments with large-scale commercial methods for production of bulk scintillator are discussed. Costs for various forms of scintillator are examined and new development goals including cost reduction methods and quality improvement techniques are suggested.

INTRODUCTION

Over the past 15 years, a great deal of new work on plastic scintillators and plastic scintillating fiber (PSF) has been performed. Much of the renewed interest in scintillator detectors stems from the tremendous progress that has been made in plastic optical fibers during this period¹ and techniques involving PSF and wavelength-shifting (WLS) fiber have produced a new generation of scintillation detectors in high energy physics.^{2,3} In 1988 the Scintillator Fabrication Facility (SFF) was formed at Fermilab with the purpose of extending the technology of plastic scintillator and plastic optical fiber. The original fiber R&D program at Fermilab is now continued under the auspices of the Scintillator Detector Development Laboratory (SDDL), and new work on different forms of plastic scintillator has emerged. We have developed three methods of scintillator manufacture, with the most recent method centering on a manufacturing process designed to produce large volumes of scintillator from commercially available polystyrene (PS).

The earliest method we have used, and notably the most likely to produce very high quality scintillator, produces cast shapes directly from monomer/dopant solutions which are carefully prepared and then polymerized under closely controlled cleanroom conditions. The second and somewhat more recent method is also a casting process, but begins with commercially available pre-polymerized pellets rather than monomer. By adding dopants to polystyrene pellets, we are able to quickly and inexpensively produce small scintillator samples suitable for optical and other laboratory tests. Finally, in an attempt to reduce cost and improve quality and consistency of very large ($>5 \times 10^5$ Kg) quantities of plastic scintillator, we have recently begun working on a third method involving commercial plastic processing techniques including compounding, pelletizing, and profile extrusion. There is hope this third method may reduce scintillator fabrication costs substantially. We believe the third method holds the most promise for producing very large quantities of detector grade scintillator.

PLASTIC OPTICAL FIBER-THE FACILITY AND PROCESS

In 1987 a small R&D program began at Fermilab within the Particle Detector Group. The original program intention was to investigate physical, chemical, and

optical properties of various fluorescent compounds and polymers that might be used in plastic scintillator. Early studies determined that commercially available monomers, polymers, and fluorescent compounds were often lacking in purity, and unsuitable for high quality scintillator use. A chemistry lab and cleanroom were subsequently constructed and utilized in a program to further understand and foster improvements in this area. Although originally envisioned as a research facility, the SFF also became a small-scale fiber preform (the scintillator rod from which fiber is drawn) production facility, initially producing over a million meters of scintillating fiber. The facility now continues as a research and development area.

The process that produces scintillating fiber preforms begins with preparation of the various chemicals used and requires substantial attention to detail if high purity preforms are to be obtained. Preform purity impacts directly on the quality of the eventual fiber and undesirable chemical impurities as well as excessive inert particulate contamination must be avoided. The facility itself must be carefully inspected and cleaned prior to each polymerization cycle, and then the styrene monomer and dopants to be used are purified. All equipment preparation, especially cleaning and assembly of the parts that come in contact with monomer and solvent, is carried out in the cleanroom. For this purpose, the cleanroom is outfitted with three large fume hoods; one serving as a distillation area, another for general cleaning and fluid transfer, and a third which houses the actual polymerization baths (Figure 1).

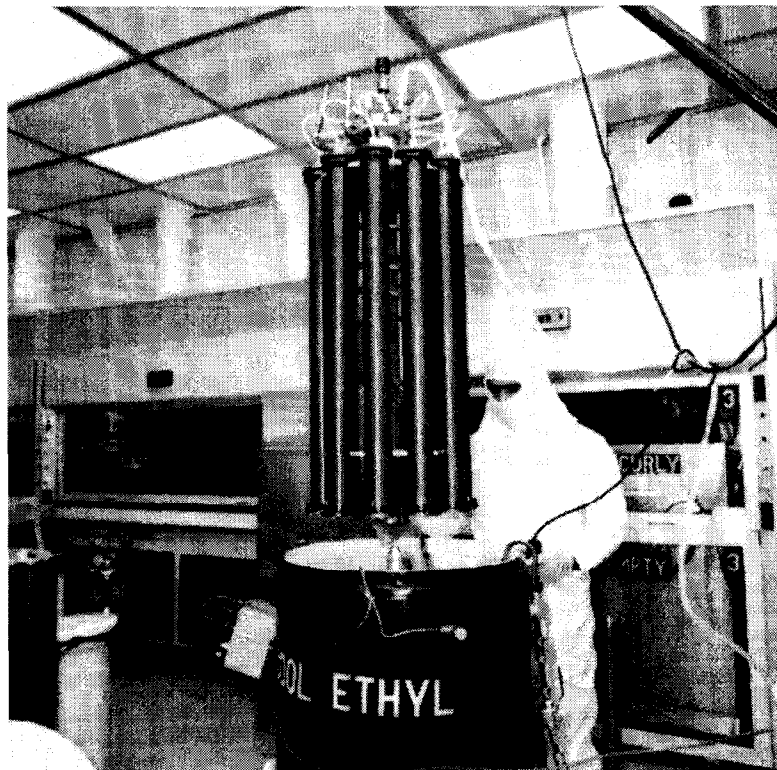


FIGURE 1. View of Scintillator Fabrication Facility cleanroom. This picture shows a mold set being lowered into the alcohol bath at the start of a freeze-pump-thaw cycle.

Styrene monomer preparation is the first step in the production of fiber preforms. The monomer is first de-inhibited using an alumina packed column and is then vacuum distilled. Samples of de-inhibited or distilled product may be taken for spectrophotometric or chromatographic analysis to determine purity. This purification step is critical to the success of the eventual scintillator. Dopants are prepared by recrystallization or other purification techniques, carefully weighed out, and are then dissolved into the monomer on the day of the polymerization run. A pressure vessel is used to accomplish the mixing step and also delivers the dopant/monomer solution through a series of filters to a set of fluoropolymer (FEP) lined cylindrical aluminum molds. Once filled the molds are placed in a low temperature bath of ethyl alcohol and evacuated in order to remove dissolved gasses from the monomer/dopant solution. The bath temperature is maintained at -90 C , just above the freezing point of the alcohol. The assembly is allowed to cool until the monomer/dopant solution solidifies and is then removed from the bath and warmed back to room temperature while pumping on the head space of the mold. This “freeze-pump-thaw” cycle is repeated several times. Next, the mold set is lowered into a heated oil bath and maintained at elevated temperature ($110\text{-}140\text{ C}$) for a period typically lasting several days. During this phase the monomer/dopant solution polymerizes and the molecular weight and polydispersity of the resultant polymer are fixed according to the heat profile and duration of the cycle. This polymerization cycle must be accurately controlled. Depending on initial bath temperature, the polymerizing monomer can produce significant exothermic heat about one hour into the polymerization cycle, and this heat must be removed in order to keep the mold and its contents at optimum temperature. Accomplishing this under some conditions is not trivial. A test was conducted using a mold instrumented with nine thermocouples to determine how serious a problem this might be. The experimental data indicate that the center of the preform can run 40 C hotter than the surrounding bath temperature, depending on the initial temperature of the bath and thus the initial rate of polymerization in the monomer. This large temperature difference as measured within the inner most part of the forming polymer core and the outer mold surface can yield large density variations in the finished preform which subsequently can affect fiber performance. In order to avoid this problem, the initial bath temperature is lowered thereby reducing the polymerization rate during the early part of the cycle. Temperature rampdown, especially the portion involving the transition through the glass transition ($\approx 110\text{ C}$) of the polymer is also critical. This step determines to a lesser but still significant extent, the eventual physical properties of the preform. Finally at the end of the heating cycle, the finished preforms are removed from their molds and samples are cut from the preform for documentation and later analysis.

When the preforms are removed, they are inspected with a 1 mW green (543nm) HeNe laser. The preform ends are diamond cut and the laser light is passed through the preform, along the axis, while looking by eye for light scattered by particles entrapped in the material. Slightly changing the entrance angle of the laser beam and scanning across the preform face gives a quick measure of the level of particulate contamination. We also use a setup that incorporates the laser, an optical chopper, reference and signal photodiodes, and a lock-in amplifier in order to measure

precisely the Rayleigh scattering in the material. Preforms are next checked for roundness and then are ready to be drawn into fiber.

Drawing is an operation that must be done in a clean environment and with excellent tension and temperature control. The core/clad interface greatly influences the amount of light that can be efficiently piped in the fiber and surface contamination can be extremely detrimental to the cladding process. Lack of core uniformity over the length of the preform, including variations in molecular weight or residual stress, can be problematic in maintaining a consistent draw thereby yielding poor diameter control and possible fiber tensile strength problems.

Many manufacturers now produce high quality scintillating fiber. The present attenuation benchmark for this type fiber is 9-10 meters @ 525 nm wavelength as measured for undoped Kuraray multicladd fiber. Increasing use of plastic scintillating fiber has stimulated further research, and progress is being made in availability, quality, and cost.

LOW COST SCINTILLATOR PROGRAM-SCINTILLATING PELLETS

The advances in plastic optical fiber technology as applied to WLS fiber readout has launched a new generation of scintillator detectors, particularly in the area of calorimetry. Although cast plastic scintillator, usually polyvinyltoluene (PVT) or polymethylmethacrylate (PMMA), has been a mainstay in detector construction for decades, the cost for very large detectors using these materials becomes prohibitively expensive. WLS fiber readout has extended the applications in which plastic scintillator can be applied and in many instances has reduced assembly costs. The cost of the bulk scintillator, however, has remained high. In response to this cost problem, we began pursuing the idea that less expensive plastic scintillator could be produced using conventional plastic extrusion techniques and machinery. Discussions with vendors began in June of 1993 and the first successful attempt at producing scintillator by compounding scintillation and waveshifting dyes into commercially available polystyrene pellets using conventional extrusion equipment was accomplished in March 1994. Several different compositions have been produced using this method. The technology appears to be reasonable in cost, allows additional secondary operations, and could be widely adopted and used once standardized methods are established. The concept is promising and several groups are conducting experiments using pellets as possible detector scintillation material. The resultant material, however, is best suited for WLS fiber readout applications due to the relatively poor optical attenuation properties of the bulk polystyrene pellets as compared to conventional "cast-from-monomer" PVT, or PS scintillator. Table 1 indicates relative light yields for polystyrene scintillator samples with like geometry (2 cm cubes) and dopant systems but produced by different methods. All are referenced to Bicon BC 404, a commercially available PVT-based scintillator. The ratio between the light yield in otherwise identical PVT and PS scintillators has been reported⁴ in the

literature to be 1:0.8 and has been attributed to differences in the π -orbital structure of the two polymers.

TABLE 1. Relative light yield of scintillators

Sample/Method	Relative Yield	Light
Bicron BC 404 (PVT)	1.0	
Fermilab PS-404 (cast from monomer)	0.8	
Fermilab 404-C (cast from previously compounded/extruded scintillator pellets DOW 70262)	0.8	

We have tested many commercially available polystyrene pellets. Test samples are formed by casting the material of interest in test tubes and then heating and melting the contents inside a vacuum oven. Discs are cut from the resultant castings and these samples are subjected to optical, scintillation, and mechanical tests. Samples can be clear polymer or dopants can be added to produce scintillator. These test samples are used to select an initial set of materials to be used in extrusion studies. Two polymers, DOW XU 70262 and DOW Styron 663W have been found suitable for scintillator applications. Styron 663W is low-cost, readily available in commercial markets, and produces a good scintillator.

The concept of producing scintillator pellets using extrusion or compounding machinery has proven successful. This versatile material can be manufactured in small to very large quantities and yields quality high-light-output scintillator. Profile extrusion of previously compounded scintillating pellets has been accomplished, with shapes generally optimized for a particular detector geometry. Usually a hole or groove is included along one axis of the profile to contain a WLS fiber for collection of scintillation light produced in the profile. We have now used this process to successfully produce finished scintillator shapes over a wide range of production rates; from 30 to 200 pounds per hour (see Figure 2).

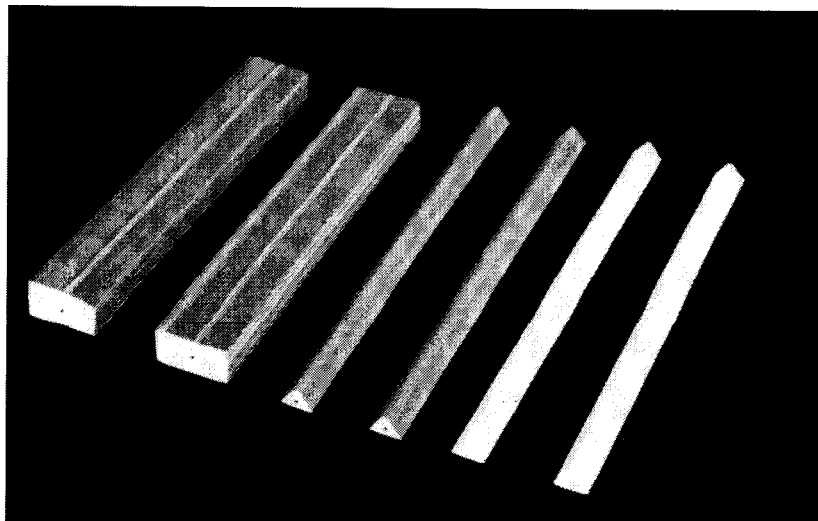


FIGURE 2. Rectangular (1 X 2 cm) and triangular profile extrusions with a hole down the axis for WLS fiber.

Production rates in the hundreds of pounds per hour range and above should markedly reduce costs, as well as provide higher quality material due to reduced residence time in the heated section of the line. Injection molding, a similar process, also uses scintillator pellets and is being used by others to produce “tiles” and various other shapes formerly machined from cast scintillator plate.⁵

This new method of scintillator manufacture uses existing equipment and technology with subtle variations from the industrial norm to make the difference between good scintillating material and just brightly colored plastic. Good quality scintillator manufacture using these methods will require some changes on the part of the processors. Companies that manufacture “clean” plastic parts, such as ophthalmic lenses, catheters, and other medical or optical devices have an advantage in that they understand contamination problems. These companies often produce parts in cleanrooms or controlled environments, providing a potential reduction in particulate loading with correspondingly less light scattering.

A recent concept that we have prototyped is the combining of the compounding step with the profile extrusion step, thereby creating a single-step process. Aside from a large cost savings brought about by the reduced handling, another potentially significant advantage of this idea would be the elimination of two heat histories and the possible loss of light yield in the scintillator. This may be critical for some polymer/dopant combinations, but would certainly be a good general practice. The process would take commercial polymer pellets and dopants and meter them through one extrusion line yielding a finished high quality scintillator shape. We have experimented with this concept and are continuing work in this area. Direct inline-doping at a concentration of approximately one percent (typical of the primary dopant in plastic scintillator) pushes the limits of ordinary extrusion practice, but

products are made using similar techniques and therefore we have hope this idea may yet be achieved.

SUGGESTIONS FOR IMPROVEMENT AND FUTURE R&D

Plastic scintillator has improved in a variety of ways over the last decade. New manufacturing processes have been identified and material quality, availability, and new detector readout geometries have all significantly progressed. Lowering the cost while still producing a high-quality scintillator remains a goal. Table 2 indicates known costs for commercial PVT scintillator sheet vs. estimated and projected costs for finished profiles of PS scintillator using a similar dopant system, but produced by new methods.

TABLE 2. Cost comparison of conventional cast scintillator to extruded scintillator.

Manufacturer/Method	Cost
Bicron Cast Sheet: BC-404	~\$40/Kg
Extruded Profiles from scintillator pellets	\$5-6/Kg
Extruded Profiles using direct in-line doping	\$3.5-4.0/Kg

Part of the challenge in continuously producing low cost quality extruded scintillator will be to understand the intuitive aspect of this technology and convert it to science. Assuming very high production rates become possible, online quality control for profile size and scintillator optical characteristics will become essential. Although very high rates have the potential to reduce scintillator costs substantially, we must be certain the parts are usable.

Finally, cultivating vendor resources, especially large polymer and chemical (dopant) manufacturers has potential for delivering advances in scintillator. Polystyrene pellets, for example, sometimes exhibit quality or consistency differences from batch to batch. Cooperation from suppliers will help us understand whether this can be resolved or needs to be factored in. Hopefully, as this new scintillator technology grows and vendor channels are established, material supply conditions will improve.

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