Extruded Plastic Scintillation Detectors

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As a way to lower the cost of plastic scintillation detectors, commercially available polystyrene pellets have been used in the production of scintillating materials that can be extruded into different profiles. The selection of the raw materials is discussed. Two techniques to add wavelength shifting dopants to polystyrene pellets and to extrude plastic scintillating strips are described. Data on light yield and transmittance measurements are presented.

I. INTRODUCTION

Plastic scintillation detectors have been used in nuclear and high energy physics for many decades [1]. Their advantages and disadvantages are recognized. Among their benefits are fast response, ease of manufacture and versatility. Their main drawbacks are radiation resistance and cost. Many research projects have concentrated on improving the fundamental properties of plastic scintillators [2,3], but little attention has focussed on their cost. Currently available plastic scintillating materials are high quality products whose cost is relatively expensive, and because of that, their use in very large detectors has not been a feasible option. For instance, MINOS (Main Injector Neutrino Oscillation Search) will require 400,000 Kg of plastic scintillator for its detector [4]. With the price of cast scintillator at approximately \$40 per Kg, such a detector would not be affordable. However, recent studies using commercial polystyrene pellets as the base material for extrudable plastic scintillators have allowed the MINOS collaboration to consider a less expensive alternative in building a plastic scintillator to build and upgrade their Forward and Central Preshower Detectors. In this case, the driving force was not the cost of the material, since only 2,000 Kg of plastic scintillator were needed, but the opportunity to use a particular shape (triangular bar) that would have been expensive to machine out of cast plastic scintillator sheets [5,6].

II. EXTRUDED PLASTIC SCINTILLATORS

Several factors contribute to the high cost of plastic scintillating sheets and wavelength shifting fibers. The main reason is the labor-intensive nature of the manufacturing process. The raw materials, namely styrene, vinyltoluene, and the dopants, need to be highly pure. These purification steps often take place just prior to the material utilization. Cleaning and assembly of the molds for the polymerization process is a detail-oriented operation that adds to the overall timeline. The polymerization cycle lasts several days. It consists of a high temperature treatment to induce full conversion from monomer to polymer, followed by a controlled ramp-down to room temperature to achieve a stress-free material. Finally, there are machining charges for sheets and tiles, and drawing charges for fibers that cannot be overlooked.

In order to significantly lower the cost of plastic scintillators, extruded plastic scintillation materials need to be considered. In an extrusion process, polymer pellets or powder must be used. Commercial polystyrene pellets are readily available, thus eliminating monomer purification and polymerization charges. In addition, extrusion can manufacture nearly any shape, increasing detector geometry options. There are, however, some important disadvantages. The extruded plastic scintillator is known to have poorer optical quality than the cast material. The main cause is high particulate matter content within the polystyrene pellets. General purpose polystyrene pellets are utilized in numerous products but few of them have strict optical requirements. A way to bypass the short attenuation length problem is to extrude a scintillator shape and use a wavelength shifting (WLS) fiber as readout. Our first approach was a two-step process that involved adding dopants to commercial polystyrene pellets to produce scintillating polystyrene pellets, which were then used to extrude a scintillator profile with a hole in the middle for a WLS fiber (Figure 1). The goal in the first step was to prepare scintillating pellets of acceptable optical quality in a factory environment. In addition to careful selection of the raw materials, the manufacturing concerns dealt with possible discoloration of the scintillating pellets because of either residues present in the equipment or degradation of the polymer pellets and the dopants in the processing device. The latter could be induced by the presence of oxygen at the high temperatures and pressures which constitute the typical operating conditions.



FIG. 1. Rectangular scintillator profile with a hole in the middle for a WLS fiber.

After producing the first batch of scintillating pellets, samples were cast to perform light yield and radiation degradation studies. Samples of standard cast scintillators such as BC404 and BC408, and samples prepared through bulk polymerization at Fermilab were also included in the studies (Table I). All the samples had similar dopant composition and were cut as 2-cm cubes. The light yield measurements were performed using a ²⁰⁷Bi source (1 MeV electrons). The light yield results (Table I) showed no significant difference among them. The Bicron samples are made of poly(vinyltoluene) instead of polystyrene which accounts for the 20% increase in light output [1]. The samples for radiation damage studies were placed in stainless steel cans and connected to a vacuum pump for two weeks to remove dissolved air and moisture. The cans were then back-filled with nitrogen and irradiated with a ⁶⁰Co source at the Phoenix Memorial Laboratory of the University of Michigan. The irradiations took place at a rate of approximately 15 KGy/h to a total dose of 1 KGy. After irradiation and annealing, the extruded scintillator cubes showed a 5% decrease in light yield which is similar to the losses observed in regular scintillator of this composition. Based on these tests, there was no sign of degradation in the scintillating pellets. The material was then used to produce extruded scintillator of different profiles with a hole in the middle for a WLS fiber.

TABLE I. Relative light yield of samples with similar compositions but from different manufacturing p	rocesses.
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$\operatorname{Scintillator}$	Bicron^a	$\operatorname{extruded}$	bulk polymerized
404	1.0	0.80	0.78
408	1.0	0.85	0.77
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^aBicron scintillator has a poly(vinyltoluene) matrix which yields 20% more light than a polystyrene one [1].

A. Selection of Raw Materials

There are many manufacturers and grades of polystyrene pellets. Most of them fall under the category of general purpose polystyrene. Only a few offer optical quality polystyrene pellets. Needless to say, there is a substantial difference in price. Nonetheless, the first plastic scintillating pellets were prepared using an optical grade polystyrene from Dow, labeled XU70251, which was later superseded by XU70262 (Dow 262). The price for Dow 262 is about \$4.5 per Kg. After confirmation by the initial tests that high quality extruded plastic scintillators were feasible, the quest began to replace the costly optical grade pellets with general purpose material. Various samples of different polystyrene grades were received from Dow, Fina, Nova, BASF, Huntsman, etc. These samples had been selected based on price, availability and melt flow rate for ease of extrusion. These materials were cast into cylinders up to 3 inches long. Transmittance measurements were performed using a Hewlett-Packard 8452 spectrophotometer. The materials tested were compared to cast samples of Dow 262 pellets. Often polystyrene contained additives that absorbed at long wavelengths such as the Fina pellets illustrated in Figure 2. This absorption would diminish the

amount of light produced by the dopants that need to be added to make a particular scintillator. Other features observed were long absorption tails and haziness caused by additives and debris in the pellets. There were a couple of materials that were repeatedly tested and showed high clarity and lack of absorptions at long wavelengths. Dow Styron 663 (Dow 663) was chosen as the general purpose polystyrene grade to conduct our extrusion studies. Its price ranges from \$1.3/Kg to \$1.7/Kg depending, among other things, on the quantity ordered.



FIG. 2. Transmittance data of commercial polystyrene pellets.

A variety of organic fluorescent compounds can be used as primary and secondary dopants in plastic scintillator applications. The primary dopant is commonly used at a 1-1.5% (by weight) concentration. The secondary dopant or wavelength shifter in scintillator is utilized at a concentration of 0.01-0.03% (by weight). The goal was to prepare a blue-emitting scintillator that could be readout with a green WLS fiber. Most green fibers are doped with K27, and thus the emission of the scintillator would have to match as best as possible the absorption of K27 in the fiber. The selection of dopants was based on these spectroscopic requirements as well as price and ease of manufacture. *para*-Terphenyl (200-225/Kg) and PPO (100-160/Kg) were considered as primary dopants. POPOP and bis-MSB (both at 0.5-1/g) were tested as secondary dopants. The final choice for the extruded plastic scintillator was PPO and POPOP in Dow 663. Figure 3 plots the transmittance spectrum of an extruded scintillator sample of this type.

B. Manufacturing Techniques

The majority of the extruded scintillator prepared has used Method 1, a two-step process conducted at two separate facilities. Figure 4 depicts the flow chart for this method. The first step was carried out at a company whose function was to add the dopants to the polystyrene pellets. (In the plastics industry, this trade is typically referred to as a color or compounding business.) Prior to the coloring run, polystyrene pellets were purged for several days with an inert gas, generally argon, to remove dissolved oxygen and moisture. The coloring step was a batch process where polystyrene pellets and dopants were tumble-mixed for 15 min. and then added to the hopper of an extruder. Each batch prepared 45 Kg of mixture. A silicone oil was used as a coating aid to achieve better distribution of the dopants on the pellet surface. An argon flow was also added to the hopper to minimize the presence of oxygen in the extruder. The die at the extruder head generated several strings of material which were cut yielding the scintillating pellets. At

the end, the scintillating pellets collected in many containers during the run were blended to homogenize the material. These pellets could now be used to produce plastic scintillators through several procedures — namely extrusion, casting and injection molding. In this case, the scintillating pellets were taken to an extrusion company to extrude the desired scintillator profile.



FIG. 3. Transmittance data of extruded plastic scintillator.



FIG. 4. Two-step process: batch coloring and extrusion (Method 1).

Using this batch process, there is also the possibility of directly extruding the scintillator profile and thus by-passing the pelletizing step. This is the route that the MINOS collaboration has chosen to investigate. This variation of Method 1 can be less expensive since all the work is done in one facility. It also reduces the heat history of the product by removing its exposure to another high temperature cycle and minimizes the chance of optical degradation. The drawback is in the batch work since the polymer and the dopants still need to be weighed for each mixture, and in the tumble-mixing step which is susceptible to contamination and prone to errors.

An alternative to these operations is given by Method 2 which is summarized in Figure 5. Method 2 is a continuous in-line coloring and extrusion process. It emphasizes the most direct pathway from polystyrene pellets to the scintillator profile with the least handling of raw materials. In this situation, the purged polystyrene pellets and dopants are metered into the extruder at the correct rate for the required composition of the scintillator. An argon flow is still used at the hopper. Coating agents are no longer needed. The appropriate die profile gives rise to the extruded scintillator form of choice. If the die can produce strands, these can also be pelletized and the scintillating pellets used in other processes. Method 2 has been tested and produces plastic scintillator of high quality and homogeneity. Although it is a simple concept, the equipment needed to accurately meter small quantities of powders such as the dopants and to achieve a good distribution of the powders in the molten polymer is not widely available. The difficulty in testing this process was finding a facility with the adequate instrumentation.



FIG. 5. Continuous in-line coloring and extrusion process (Method 2).

III. LIGHT YIELD OF EXTRUDED PLASTIC SCINTILLATORS

Light yield studies have been performed on many samples of extruded plastic scintillators. Although a variety of shapes and sizes is available, the measurements have mostly been carried out on 11.5-cm long rectangular extrusions (1cm x 2cm) with a hole in the middle for a green WLS fiber. Each extrusion is tightly wrapped in Tyvek for this test. The WLS fiber utilized is BC91A (0.835 mm diameter, 1.5 m long) with one mirrored end. The light yield test setup uses an electron spectrometer with ¹⁰⁶Ru source whose 3-MeV beam is momentum selected. There is a small trigger counter in front of the extruded sample. The photomultiplier tube used is a Hamamatsu R2165 which has excellent single photo-electron resolution. The fiber is held at a fixed position from the PMT surface to minimize fluctuations among measurements. The light yield is determined from the following calculation:

$$LightYield = \frac{Mean - Pedestal}{Gain}$$

where the mean and the gain are defined as:

$$Mean = \frac{\sum_{i}^{n} v_{i} x_{i}}{\sum_{i}^{n} v_{i}}$$

Gain = FirstPeak - Pedestal

where v_i is the number of entries for each ADC value, x_i . The data are fitted to locate the position of the first and second peaks, and the pedestal. Figure 6 presents the light yield distribution of an extruded sample and the fit for the first and second electron peaks.

The results from a series of light yield measurements are listed in Table II. RDN 262 extrusions were prepared by the two-step batch process (Method 1) using Dow 262 optical grade pellets. Leistritz 262 and 262P samples were produced by the continuous procedure (Method 2) using Dow 262 polymer. Leistritz 663 samples were also prepared by Method 2 but used general purpose polystyrene pellets (Dow 663). Although the samples are from different runs, their light output is similar. The Leistritz 262 samples show a slightly lower light yield but their profile is smaller than that of the remaining samples. These samples were collected early in the extrusion run when the profile was not completely to specification. These results indicate that there is no major difference in light yield between Method 1 and Method 2. This test proves that the continuous in-line coloring and extrusion process (Method 2) yields a homogeneous part with the right concentration of dopants. This aspect is less of a concern in Method 1 since the first step includes batch tumble-mixing and post-blending of the scintillating pellets before the scintillating profile is extruded. In addition, these numbers confirm that Dow 663 (general purpose polystyrene pellets) can replace the optical grade pellets initially utilized. More measurements are underway to compare these extrusions to samples of commercial plastic scintillator sheets which have been cut to the same profile.



FIG. 6. Light yield distribution of extruded plastic scintillator (RDN 262) with WLS fiber. Solid curve is 2-gaussian fit to first and second peaks.

TABLE II. Light yield of extruded plastic scintillator samples.					
Scintillator	No. of samples	Light Yield	St. Dev.	Characteristics	
RDN 262	30	2.05	0.09	Dow 262, Method 1	
Leistritz 262	10	1.81	0.14	Dow 262, Method 2	
Leistritz 262P	10	2.02	0.10	Dow 262, Method 2	
Leistritz 663	15	2.22	0.07	Dow 663, Method 2	

IV. CONCLUSIONS

Research on extruded plastic scintillator was driven by the high cost of cast plastic scintillator. The goal was to use commercially available polystyrene pellets, in particular from a general purpose grade, and standard extrusion equipment to lower the price of plastic scintillators. Extruded plastic scintillator strips have been manufactured and tested. The estimated price for extruded scintillator ranges from \$3.5/Kg to \$6/Kg. About 50% of the cost is due to raw materials with the remaining 50% due to processing. The results indicate that the extruded scintillator profile with a WLS fiber as readout is a valid system for scintillation detectors. The MINOS experiment will build a very large detector using this technology. The D0 experiment is assembling the Central and Forward Preshower detectors using extruded scintillating triangular strips.

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