ELSEVIER

Nuclear Instruments and Methods in Physics Research A 466 (2001) 482–491

NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH Section A

www.elsevier.com/locate/nima

Low-cost extruded plastic scintillator

Anna Pla-Dalmau, Alan D. Bross*, Kerry L. Mellott

Fermi National Accelerator Laboratory, Mail Station 121 (WH 9E), P.O. Box 5000, Batavia, IL 60510, USA

Received 1 August 2000; received in revised form 2 October 2000; accepted 2 October 2000

Abstract

Motivated by a need for lower cost plastic scintillation detectors, we have tested commercially available polystyrene pellets in order to produce scintillating materials that can be extruded into various shapes. Selection of the raw materials is discussed. Two techniques are described that add wavelength shifting dopants to polystyrene pellets and extrude plastic scintillating bars using these materials. Data on light yield and transmittance are presented.

PACS: 29.40.Mc

Keywords: Scintillation; Neutrino detector; Calorimeter

1. Introduction

Plastic scintillation detectors have been used in nuclear and high energy physics for many decades [1]. Although there are some disadvantages in using plastic scintillation detectors, their strengths are numerous. Among their benefits are fast response time, ease of manufacture and versatility. Their principal drawbacks are relatively low radiation resistance and high cost. Currently available plastic scintillating materials are high quality products whose cost (above $ 40 per kg) is relatively expensive, and because of this, their use in very large detectors has not been a feasible option. Recently, many research projects have concentrated on improvement of the fundamental properties of plastic scintillators [2,3], but only limited attention has been focussed on their cost. Most of the development work on producing lower cost plastic scintillator was performed in the 1970s and 1980s. In 1975, the acrylic scintillator Plexipop was developed.[1] Plexipop lowered the cost, but the scintillation light output was only one-fourth that of conventional plastic scintillator. Work on other acrylic-based plastic scintillators [4] improved on the light yield problem. However, the resultant scintillator suffered from slow response time and relatively poor mechanical properties. The extrusion technique was first applied to plastic scintillator detectors in 1980 [5] and produced a polystyrene-based scintillator with good light yield, but demonstrated a relatively poor attenuation length. These developments never resulted in high quality scintillator that was commercially available at low cost. During the late 1980s and early 1990s, wavelength shifting (WLS) fiber became commercially available and was utilized in numerous scintillation detector applications. The use

*Corresponding author.
E-mail addresses: bross@fnal.gov (A.D. Bross).

[1] Manufactured by Rohm, GmbH Chemische Fabrik, Darmstadt, Germany.

0168-9002/01/$ - see front matter
PII: S0168-9002(01)00177-2

of WLS fiber readout of plastic scintillator made the requirement for a long attenuation in the scintillator less important. At this time, we were considering a new neutrino experiment [6] at Fermilab that would require a very large active detector. Conventional cast scintillator plate was considered, but was quickly rejected due to cost. Although this experiment never proceeded beyond the proposal stage, we started research within the Particle Detector Group at Fermilab investigating the possibility of using commercial polystyrene pellets as the base material for extrudable plastic scintillators.

2. Extruded plastic scintillators

Although several factors contribute to the high cost of cast plastic scintillating sheets, the main one is the labor-intensive nature of the manufacturing processes. The raw materials, such as styrene, vinyltoluene and dopants, need to be highly pure. Cleaning and assembly of the molds for the polymerization process is a detail-oriented operation that greatly adds to the overall effort. The polymerization cycle lasts for 3–5 days, consisting of a high temperature treatment to induce full conversion from monomer to polymer and a controlled ramp-down to room temperature to achieve a stress-free material. Finally, there is machining of the raw sheets that can significantly add to the final detector cost.

In order to significantly lower the cost of plastic scintillators, we considered the use of extruded plastic scintillation materials. 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, the extrusion process can manufacture essentially any shape. There are, however, some important disadvantages: Generally the extruded plastic scintillator will have a poorer optical quality than the cast material. This is mainly because of the high particulate matter content in the polystyrene pellets. (General purpose polystyrene pellets are used in numerous products but none of these products has strict optical requirements.) Another reason for the inferior optical quality of extruded material is that the rapid cool-down cycle in this technique leaves the final material stressed. This stress can lead to non-absorptive optical distortions in the material that degrade the attenuation length. A way to bypass the short attenuation length problem is to extrude a scintillator shape and use wavelength shifting (WLS) fiber readout.

Our first approach was a two-step process: First we added dopants to commercial polystyrene pellets (compounding) to produce scintillating polystyrene pellets. Then we used these pellets to extrude a scintillator profile with a hole in the middle for a WLS fiber (Fig. 1). The goal in the first step was to prepare scintillating pellets of good optical quality in a factory environment. After selecting the raw materials, our main concern was a possible discoloration of the scintillating pellets. This could result either from residues present in the equipment or from degradation of the polymer pellets or dopants in the processing device. Polymer and dopant degradation can be induced by the presence of oxygen during the high temperature and pressure processing conditions typically present in the extruder.

2.1. Scintillating polystyrene pellet manufacture

We chose a conventional industrial process, compounding, to incorporate organic dopants into commercial grade polystyrene pellets in order to produce a scintillating plastic. With the goal of reducing costs, a factory production environment was dictated, using standard equipment and personnel, but with some simple and important precautions. In our initial discussions with many "compounding houses," we explained our need for exceptionally clean equipment and continuous avoidance of cross-contamination from other, nearby, product lines. We stressed that more than normal attention to detail, especially regarding heat profiles and residence times, would be essential. We encouraged high throughput rates as beneficial to both cost and quality.

Prior to the compounding run,[2] polystyrene pellets were purged for several days with an inert

[2] Work performed at Chroma Corporation, McHenry, IL.

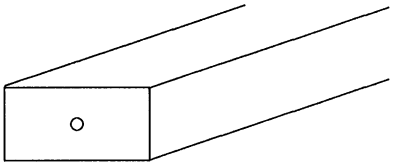

Fig. 1. Rectangular scintillator profile with a hole in the middle for a WLS fiber.

gas, generally argon, to remove dissolved oxygen and moisture. The compounding step was a batch process where polystyrene pellets and dopants were tumble-mixed for 15 min and then added to the hopper of the extruder. Each batch prepared 45 kg of mixture. We used a silicone oil as a coating aid to achieve better distribution of the dopants on the pellets. We also required that the material be processed under an inert gas blanket at the machine entrance and exit. An argon flow was added to the hopper of the extruder in order to provide this inert gas blanket. In addition, we recommended that a vacuum degassing section be included in the extruder. Fig. 2 shows the die at the extruder head generating several strings of material. These strings were then re-pelletized yielding the scintillating pellets. The extruder incorporated a screw with a length to diameter ratio greater than 30 : 1 and with a special screw mixing section. This was done to assure uniform mixing of the dopant into the polymer. Finally, the newly compounded scintillating pellets were classified for uniform size and then post-blended. At this stage the pellets were ready for further extrusion or injection molding into a final profile. Throughout the compounding/pelletizing process, we obtained quality control samples and recorded machine parameters.

2.2. Intrinsic light yield and radiation resistance of scintillating pellets

During our first trials at producing scintillating polystyrene pellets, we used optical grade polystyrene pellets from Dow Chemical, XU70251 and XU70252. After producing the first batch of scintillating pellets, we then prepared samples for light yield and radiation degradation studies. Samples of standard cast scintillators, such as BC404 and BC408, and samples prepared through

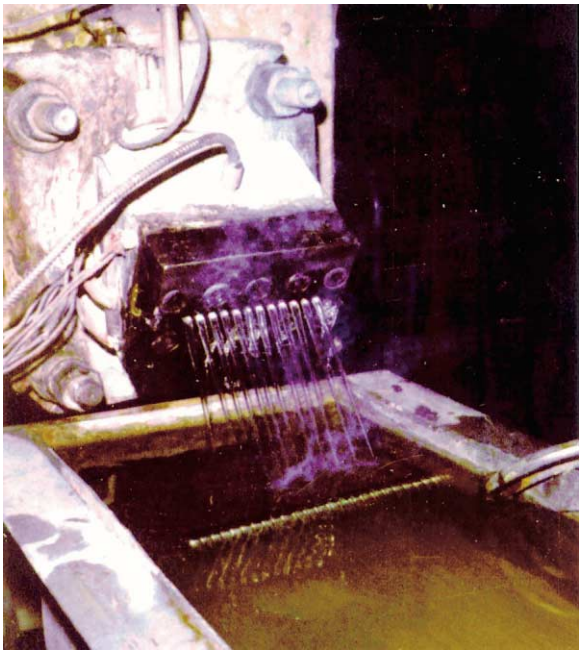

Fig. 2. View of the extruder head showing strings of scintillator.

Table 1
Relative light yield of samples with similar compositions but from different manufacturing processes

Scintillator	Bicron[a]	Extruded	Bulk polymerized
404	1.0	0.80	0.78
408	1.0	0.85	0.77

[a] Bicron scintillator has a poly(vinyltoluene) matrix which yields 20% more light than a polystyrene one [1].

bulk polymerization at Fermilab were also included in the studies. All the samples had similar dopant composition and were cut into 2-cm cubes. The light yield measurements were performed using a ^{207}Bi source (1 MeV electrons). The light yield results (Table 1) showed no significant difference between the extruded and bulk polymerized material. The Bicron samples do show a 20% increase in light output over the other samples, but this is accounted for by the fact that BC404 and BC408 use poly(vinyltoluene) instead of polystyrene as the base polymer [1]. Samples for radiation damage studies were placed in stainless steel cans and then evacuated for 2 weeks. The cans were back-filled with nitrogen and irradiated

with a ^{60}Co 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 10 kGy. After irradiation and annealing, the extruded scintillator cubes showed a 5% decrease in light yield. This is similar to the losses observed in our cast material and in commercial scintillator of this composition. Based on these tests, we conclude that the intrinsic light yield and radiation resistance of our extruded scintillator pellets are equal to that of conventional cast plastic scintillator. This material was then used to produce extruded scintillator of different profiles with a hole in the middle for WLS fiber.

2.3. 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 companies offer optical quality polystyrene pellets. Needless to say, there is a substantial difference in price among the different grades of polystyrene pellets. Nonetheless, we prepared the first plastic scintillating pellets using optical grades of polystyrene from Dow Chemical labeled XU70251 and XU70252. These were later superseded by XU70262 (Dow 262). The price for Dow 262 is about $ 4.5 per kg. After our initial tests confirmed that high quality extruded plastic scintillators were feasible, we began a search for a general purpose material that could replace this costly optical grade pellet. We obtained samples of different polystyrene grades from Dow, Fina, Nova, BASF and Huntsman. 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 in. long. Transmittance measurements were performed using a Hewlett-Packard 8452 spectrophotometer. We compared all tested materials to samples of Dow 262. Often, polystyrene contained additives that absorbed at critical wavelengths, such as that shown in the Fina pellets illustrated in Fig. 3. The Fina polymer referenced to DOW 262 showed increased absorption in the wavelength region between 350 and 400 nm (which can effect coupling between primary and secondary dopants) and again at long wavelength (> 450 nm). Polymers containing absorption of this type yield scintillators with lower effective light yield. We also observed long absorption tails and haziness in some of the polymers. This was caused by either additives or debris in the pellets. Two of the studied materials were put to repeated tests and showed high clarity and lack of excessive absorption relative to DOW 262. Finally, we chose Dow Styron 663 (Dow 663) 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.

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 bulk scintillator is typically used at a concentration of 0.01–0.03% by weight. Our goal was to prepare a blue-emitting scintillator that could be readout with a green WLS fiber. Most green fibers are doped with K27 thus the emission of the scintillator had to match, as best as possible, the absorption of the K27 in the fiber. We based the selection of dopants on these spectroscopic requirements as well as price and ease of manufacture. p-Terphenyl (PT) ($ 200–225/kg) and 2,5-diphenyloxazole (PPO) ($ 100–160/kg) were considered as primary dopants. 1,4-bis(5-Phenyloxazole-2-yl)benzene (POPOP) and 4-bis(2-Methylstyryl)benzene (bis-MSB) (both at $ 0.5–1/g) were tested as secondary dopants. The final choice for extruded plastic scintillator in most of our work was PPO and POPOP in Dow 663. Fig. 4 plots the transmittance spectrum of an extruded scintillator sample through a 1 cm thickness. The data below 325 nm in this plot should be ignored, however. The reference in this measurement was polystyrene. Since polystyrene does not transmit well below roughly 325 nm, the data show an artificially high transmittance in this region.

2.4. Manufacturing techniques for scintillating profile extrusion

The majority of our extruded scintillator has been prepared through Method 1 [7], a two-step

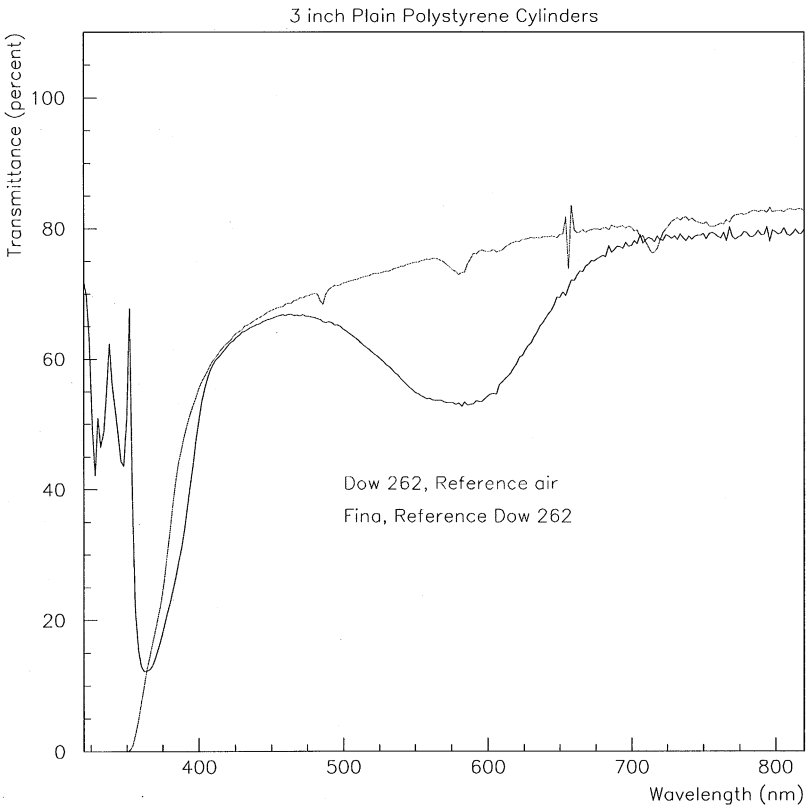

Fig. 3. Transmittance data of commercial polystyrene pellets.

batch process conducted at two separate facilities. Fig. 5 depicts the flow chart for this method. The first step was carried out at a compounding company whose function was to add the dopants to the polystyrene pellets. At the end of the run, the scintillating pellets collected during the run were blended to homogenize the material. These pellets could then be used to produce plastic scintillators through several procedures—namely extrusion, casting, and injection molding. In our work the scintillating pellets were taken to an extrusion company to extrude the various scintillator profiles. Fig. 6 shows a triangular extrusion exiting the die of the extruder head.

Using this batch process, there is also the possibility of directly extruding the scintillator profile and thus by-passing the pelletizing step. This variation of Method 1 can reduce costs since all the work is done at one facility. It also reduces the heat history of the product by removing its exposure to another high temperature cycle and eliminates an additional chance for scintillator degradation. The drawbacks to this batch work are twofold: The polymer and the dopants still need to be accurately weighed and proportioned for proper dopant concentration. Second, the tumble-mixing step is susceptible to contamination and prone to errors. Fig. 7 shows a picture of some of the extrusions we have made using Method 1. This picture shows 1×2 cm, 5 mm $\times$ 5 mm, and 5 mm triangular extrusions along with some of the scintillating pellets.

An alternative to these operations is given by Method 2 which is summarized in Fig. 8. Method 2 is a continuous in-line compounding and extrusion process. It emphasizes the most direct pathway from polystyrene pellets to a particular scintillator profile with the least amount of handling of the raw

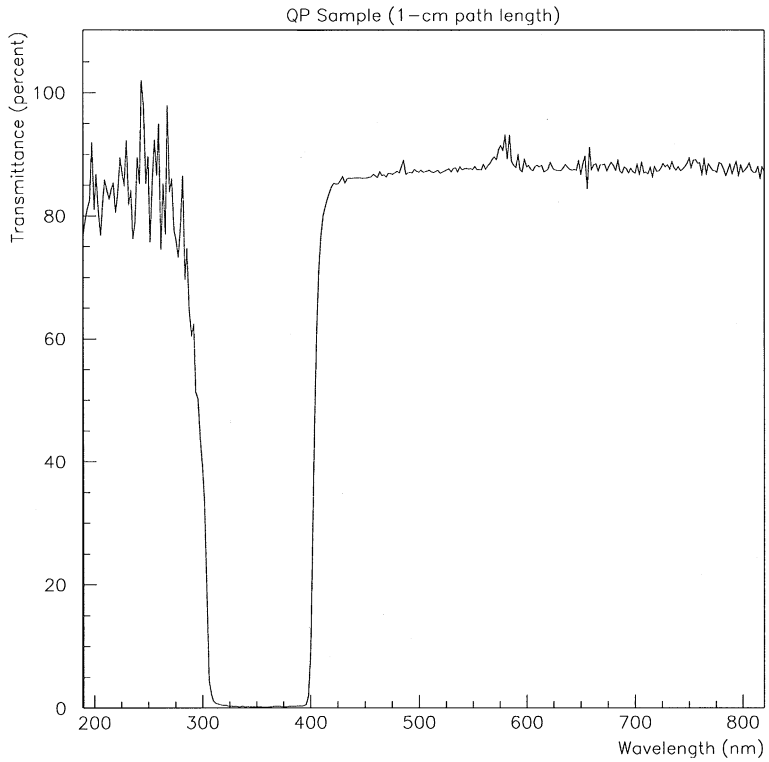

Fig. 4. Transmittance data of extruded plastic scintillator.

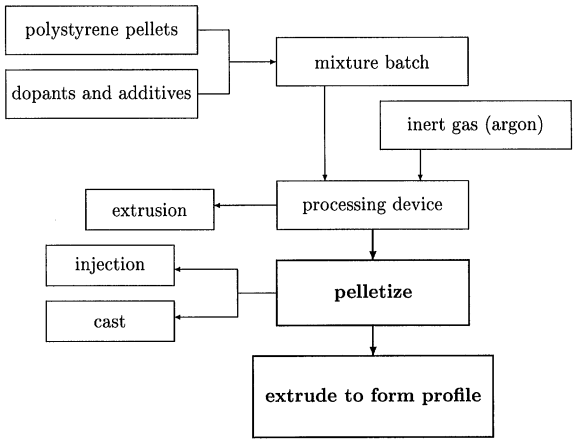

Fig. 5. Two-step process: batch coloring and extrusion (Method 1).

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 can be 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 for accurately metering small quantities of powders such as the dopants and for achieving 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.

3. Light yield of extruded plastic scintillator profiles

Light yield studies have been performed on many samples of extruded plastic scintillator [8]. The measurements reported here have been done

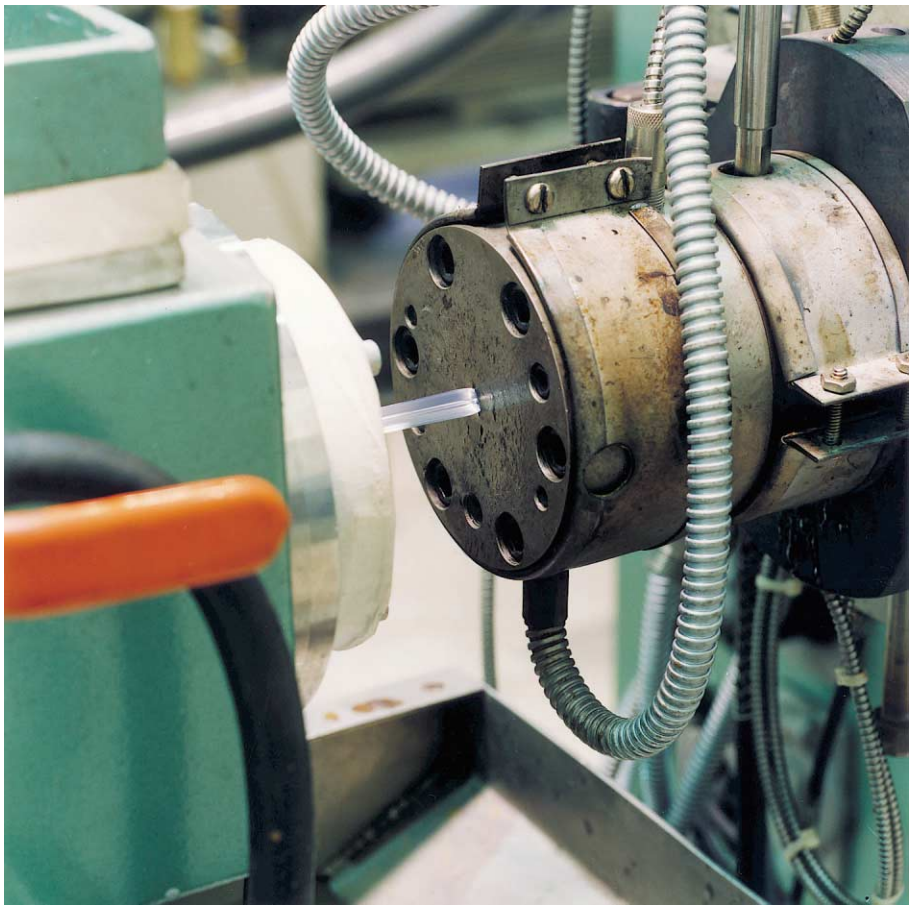

Fig. 6. Picture of triangular extrusions exiting the extruder die.

on 11.5-cm long rectangular extrusions (1 cm × 2 cm) 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 a mirrored end. The light yield test setup uses an electron spectrometer with a ^{106}Ru 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 high single electron resolution. The fiber is held 2 mm from the PMT surface by a fixture. The light yield is determined from the following:

$$\text{Light Yield} = \frac{\text{Mean} - \text{Pedestal}}{\text{Gain}}$$

where the mean and the gain are defined as

$$\text{Mean} = \frac{\sum_i^n v_i x_i}{\sum_i^n v_i}$$

$$\text{Gain} = \text{First Peak} - \text{Pedestal}$$

where v_i is the number of entries for each ADC value, x_i. These data are fit to a multi-Gaussian distribution in order to determine the position of the first and second peaks, and the pedestal. Fig. 9 presents the light yield distribution of a RDN 262 sample[3] and the fit for the first and second electron peaks.

[3] Work performed at RDN Equipment Manufacturing Co. Bloomingdale, IL.

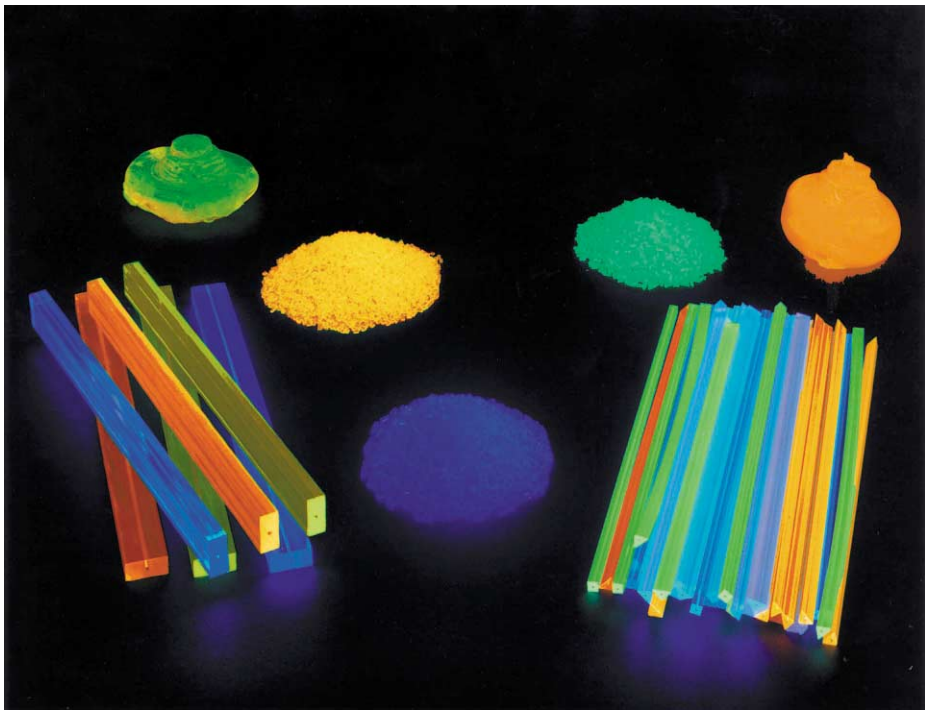

Fig. 7. Examples of extrusions made using method 1.

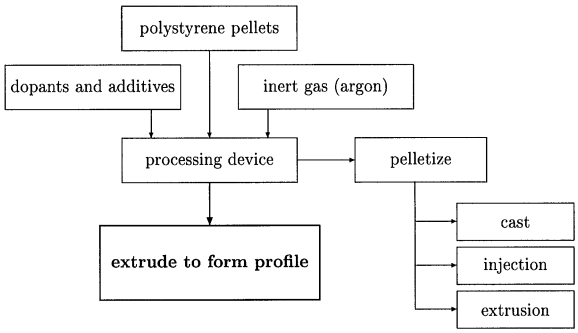

Fig. 8. Continuous in-line coloring and extrusion process (Method 2).

The results from a series of light yield measurements are listed in Table 2. RDN 262 extrusions were prepared by the two-step batch process (Method 1) using Dow 262 optical grade pellets. Leistritz 262 and 262P samples[4] 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 between Method 1 and Method 2. The continuous in-line compounding and extrusion process (Method 2) yields a homogeneous part with the right concentration of dopants. In addition, these numbers confirm that Dow 663 (general purpose polystyrene pellets) can replace the optical grade pellets initially utilized.

We also prepared a sample using Bicron BC 404. This sample was machined from cast plate. A square groove was machined in one piece and was then glued to a similar piece to "simulate" a 1×2 cm extrusion with a hole down the middle. Although we expected the BC 404 to have higher yield than the extruded samples (BC 404 is PVT

[4] Work performed at American Leistritz Co. Somerville, NJ.

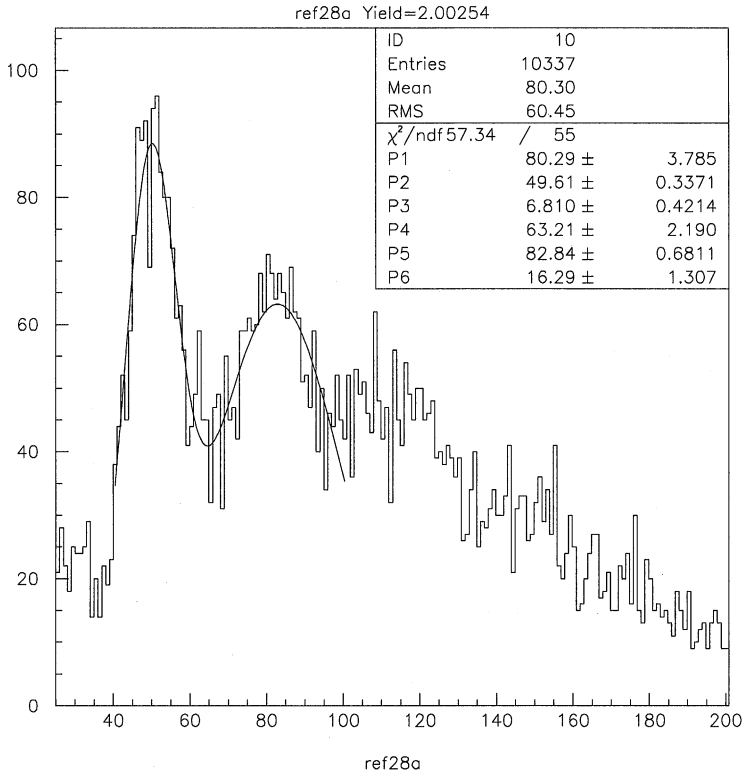

Fig. 9. Light yield distribution of extruded blue plastic scintillator with green WLS fiber.

Table 2
Light yield of extruded plastic scintillator samples

Scintillator	Light yield	Std. Dev.	Characteristics
RDN 262	2.05	0.09	Dow 262, Method 1
Leistritz 262	1.81	0.14	Dow 262, Method 2
Leistritz 262P	2.02	0.10	Dow 262, Method 2
Leistritz 663	2.22	0.07	Dow 663, Method 2
Bicron BC 404	1.83	NA	BC 404 (PVT) machined groove

based, see Table 1), it was lower as indicated in Table 2. This is due to the fact that in these tests the WLS readout fiber was not glued into the hole so that the same fiber could be used for all samples. The surface quality of the machined "hole" is not as good at that in the extruded samples. We believe that this is what caused the lower light yield. In most experimental applications, the WLS is not fully glued into the hole or groove into which it is placed.

4. Experimental applications

The success of our early studies and trial extrusion runs has already led to the use of the technique in one high energy physics experiment and it has been chosen in two others. The D0 experiment has used the two-step process (Method 1) to produce triangular extrusions [9–11] for two preshower detectors. The triangles have a base that is approximately 6 mm wide and are 5.4–6.1 mm in height. A hole down the axis of the extrusion permits the insertion of a WLS fiber for readout. The scintillator used in this application used a p-terphenyl primary with a trans-4,4′-diphenylstilbene (DPS) secondary. The compounding was done at a single facility, while the extrusions were done at two different facilities because of scheduling problems. Nevertheless, the scintillator for the two detectors is extremely uniform, and the light yield from the scintillator used in the two detectors is equal to within 2–3%, even though different

commercial facilities were used in the manufacturing process.

The MINOS experiment [12] has chosen the modified Method 1 process (no pelletization step, Fig. 5) to produce approximately 300,000 kg of scintillator strips for their detector. The material used in this application is a blue-emitting scintillator that utilizes PPO and POPOP as primary and secondary dopants, respectively. The MINOS extrusions have a rectangular profile (10 mm high $\times$41 mm wide) with a 2-mm deep groove in the center of the strip for a WLS fiber for readout.

The STAR experiment [13] will be using extruded scintillator for a shower maximum detector in the electromagnetic end-cap calorimeter. The scintillator extrusions will have a triangular profile with approximately a 10-mm base and a 7-mm height. There will be a hole down the axis of the extrusion for a WLS fiber for readout. These triangles will be produced using the two-step process (Method 1). The scintillator will use p-terphenyl and DPS as dopants. Approximately 150 kg of this material will be needed to build the detector.

5. Conclusions

This research on extruded plastic scintillator was driven by the high cost of cast plastic scintillator. Our goal was to use commercially available polystyrene pellets, in particular from a general purpose grade, and standard extrusion equipment to lower the cost of producing plastic scintillators. Extruded plastic scintillator strips have been manufactured and tested. The intrinsic light yield of our extruded scintillator is equal to that of high quality bulk-polymerized (from monomer) scintillator. Scintillator extrusions read out with WLS fiber give a light output comparable to that obtained with a sample made from Bicron BC 404. The estimated price for extruded scintillator ranges from \$ 3.5/kg to \$ 7/kg. About 50% of the cost is due to the raw materials and the remaining 50% is due to processing. The results indicate that an extruded scintillator profile with WLS fiber readout is a viable system for large-scale scintillation detectors. The technique has already been used successfully to build two preshower detectors for the D0 experiment, and MINOS and STAR have chosen the technique for use in their experiments.

Acknowledgements

We wish to thank Alan Esche of Chroma Corporation for his help with the compounding trials and Michael Pohl of RDN Manufacturing Co. and Augie Machado of American Leistritz Extruder Corporation for their invaluable help during our extrusion runs at their facilities. This work has been supported by the U.S. Department of Energy.

References

[1] J.B. Birks, The Theory and Practice of Scintillation Counting, Pergamon Press, The MacMillan Co., New York, NY, 1964.
[2] A.D. Bross, R.C. Ruchti, M.R. Wayne (Eds.), Proceedings of SCIFI 93: Workshop on Scintillating Fiber Detectors, World Scientific, Singapore, 1995.
[3] A.D. Bross, R.C. Ruchti, M.R. Wayne (Eds.), Proceedings of SCIFI 97: Conference on Scintillating Fiber Detectors, AIP Conference Proceedings 450, American Institute of Physics, Woodbury, New York, 1998.
[4] C. Aurouet, et al., Nucl. Instr. and Meth. A 169 (1980) 93.
[5] J.C. Thevenin, et al., Nucl. Instr. and Meth. A 169 (1980) 53.
[6] M. Murtagh et al., A search for neutrino oscillations using the Fermilab debuncher, Fermilab Proposal P-860, January, 1992.
[7] A.D. Bross, A. Pla-Dalmau, K. Mellott, U.S. Patent 5 968 (1999) 425.
[8] A. Pla-Dalmau, A.D. Bross, K. Mellott, Extruded plastic scintillation detectors, DPF Proceedings (Fermilab-Conf-99/095), January 1999.
[9] M. Adams, et al., A detailed study of plastic scintillating strips with axial wavelength shifting fiber and VLPC readout, Nucl. Instr. and Meth. A 366 (1995) 263.
[10] D. Lincoln (D0 Collaboration), New technologies in the D0 central tracker upgrade, Nucl. Instr. and Meth. A 379 (1996) 424.
[11] P. Baringer et al., Cosmic ray tests of the D0 preshower detector, FERMILAB-Pub-00/159-E, Nucl. Instr. and Meth. A, submitted for publication.
[12] The MINOS detectors, Technical Design Report, NuMI-L-337, 1998.
[13] L.C. Bland et al., An endcap electromagnetic calorimeter for STAR: conceptual design report, STAR Note 401 (1999).