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Production of large-size polystyrene based plastic scintillators with uniform optical properties

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The purpose of this work was to determine the nature of the appearance of striae in the volume of plastic scintillators obtained by bulk polymerization. A model was built and the calculation of the expected character of the convective flow of the styrene reaction mass at different widths of ampoules for block polymerization was carried out. The optimal size of the ampoule width for block polymerization was determined. A comparison of the characteristics of plastic scintillators with and without striae was made. For a scintillator without striae, the TAL is 10–25 % better. The minimum detecting activity under irradiation with an Am241 source is 2 times better than that of a scintillator with striae.
Keywords: plastic scintillator, striae, convection.

Виробництво великогабаритних пластмасових сцинтиляторів на основі полістиролу з однорідними оптичними властивостями. *А.Ю.Бояринцев, А.В.Колесніков, С.М.Ковальчук, І.С.Невлюдов*

Вивчено характер виникнення свілей в об'ємі пластмасових сцинтиляторів, отриманих полімеризацією у масі. Побудовано модель і здійснено обчислення передбачуваного характеру конвективного перебігу реакційної маси стиролу при різних ширинах ампул для блокової полімеризації. Визначено оптимальний розмір ширини ампули для блокової полімеризації. Проведено порівняння характеристик пластмасових сцинтиляторів зі свілями та без свілей. Для сцинтилятора без свілей отримано TAL краще на 10–25 %. Мінімальна детектуюча активність при опроміненні джерелом Am241 краще вдвічі у порівнянні зі сцинтилятором зі свілями.

Исследован характер возникновения свилей в объеме пластмассовых сцинтилляторов, полученных полимеризацией в массе. Построена модель и произведен расчет предполагаемого характера конвективного течения реакционной массы стирола при различных ширинах ампул для блочной полимеризации. Определен оптимальный размер ширины ампулы для блочной полимеризации. Проведено сравнение характеристик пластмассовых сцинтилляторов со свилями и без свилей. Для сцинтиллятора без свилей получен TAL лучше на 10–25 %. Минимальная детектирующая активность при облучении источником Am241 лучше в 2 раза по сравнению со сцинтиллятором со свилями.

1. Introduction

Large-area plastic scintillators are used to register gamma quantum in customs and metallurgical portals, as well as in melt level control systems. Plastic scintillators have a low price and allow covering large areas of registration where it is necessary [1, 2]. The main and important requirement for a plastic scintillator having large area and linear dimensions (more than 500 mm), is high transparency to its intrinsic emission. One of the problems that impair the transparency of the scintillator is optical inhomogeneity, i.e. the presence of optical striae in the volume of the scintillator. Such effects are observed in large-size polystyrene based plastic scintillators produced by block polymerization from a styrene monomer in an aluminum ampoule [1].

In this paper, we consider a method for producing large-size polystyrene based plastic scintillators with improved transparency. The nature of optical striae formation during the block polymerization of polystyrene was studied.

The article discusses a method for reducing the striae when producing large-size plastic scintillators in the process of thermally initiated polymerization of styrene in bulk.

Comparative characteristics of plastic scintillators with striae and without them were obtained. As the results of this work showed, the TAL (Technical Light Attenuation Length) [3] of polystyrene scintillators with a diameter of 50 mm and a length of 1000 mm without striae is higher by 30–70 % if compared with scintillators with optical striae.

2. Calculation part

The nature of convective flows is determined by thermo-gravitational convection. This process is accompanied by a complex current of descending and ascending flows depending on the thermophysical characteristics of the liquid, the temperature gradient, the shape of the ampoule and its geometric dimensions [4].

At the initial stage of polymerization, the viscosity of styrene at the temperature of 60 degrees is equal to $\mu = 4.53 \cdot 10^{-4}$ Pa·s [5]. During the heating of styrene, as well as over time, the viscosity of the reaction mass changes. At the beginning of the process, as the temperature grows, the viscosity decreases. The viscosity of the reaction mass increases strongly with the conversion depth. By the end of the polymerization process, the viscosity can reach $\mu = 1 \cdot 10^3 – 1 \cdot 10^4$ Pa·s [6, 7]. It is hard to describe mathematically such a complex process due to the difficult process of controlling the viscosity at each stage of the polymerization process. In the initial phase of polymerization, convection does not work on the formation of striae. At this stage, the styrene boils violently and mixes with abundant heat release [1]. At the final stage of polymerization, the formed polystyrene has a high viscosity and does not cause convective flows. Thus, the calculation was made for the intermediate stage of the styrene polymerization process.

The following parameters were used to calculate the reaction mass motion by the finite volume method:

— reaction mass density $p_0 = 1.05$ kg/dm^3;

— reaction mass temperature $T_0 = 440$ K;

— thermal expansion coefficient $\beta = 6.2 \cdot 10^{-4}$ 1/K;

— styrene heat capacity $C_v = 1100$ J/(kg·s);

— dynamic viscosity $\mu = 1$ Pa·s;

— Prandtl number Pr = $1 \cdot 10^3$.

The calculation volume is divided into cells by a uniform rectangular grid with dimensions 500×50×30 mm^3.

The calculation was made for an ampoule with a length of 2000 mm, styrene height of 400 mm and different width of 600 mm, 300 mm, 150 mm, 100 mm, and 60 mm. The variation of the ampoule width was made to determine the geometry at which the convection would be suppressed.

The nonstationary solver buoyantPimpleFoam from the open OpenFOAM CFD code was used to calculate the fluid motion [8]. It implements the finite volume method. In this paper, the polystyrene solution is considered to be an incompressible and homogeneous liquid. To model the melt convection, the Boussinesq approximation is used, where the melt density is a linear function of temperature.

$$\rho = \rho_0(1 - \beta T). \quad (1)$$

The PISO-SIMPLE algorithm is used to solve the Navier-Stokes equations [9].

Due to low temperatures, radiation heat transfer was not taken into account.

The calculation area is represented by the actual volume of polystyrene without the ampoule walls. Thermal boundary conditions are represented as boundary conditions "fixedValue"

$$T(x) = F(x). \quad (2)$$

Specific values of these boundary conditions were obtained experimentally using thermocouple and pyrometric measurements.

For the velocity field at the boundary with the ampoule walls, it is assumed

$$ux = uy = uz = 0 \quad (3)$$

boundary condition.

3. Experimental

To determine the nature of the convective flows movement and the formation of optical striae during block polymerization, a model experiment was carried out. For this purpose, a block of styrene monomer with dimensions of 600×400×2000 mm was produced. To load styrene, an aluminum ampoule for polymerization [1] with dimensions of 600×2300×2000 mm was used. After preheating to 80°C in the dissolution units, the styrene was unloaded into an aluminum ampoule. After filling the ampoule, the styrene was heated to 165°C at the rate of 5°C/h. After boiling, the styrene was kept at 165°C for 48 h. Next, the mass was cooled to room temperature at the rate of 0.75°C/h [10, 11]. The temperature of the reaction mass was controlled with two thermocouples installed at the ends of the aluminum ampoule.

To visualize convective flows during polymerization (boiling) at high temperatures, the styrene was colored due to oxidative processes [1].

To compare the characteristics of the plastic scintillator, block polymerizations of scintillation plastic equivalent to UPS-923A

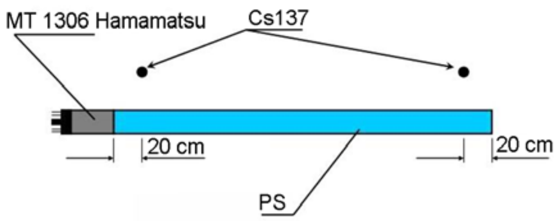


Fig. 1. Scheme of the light output measurements for the ∅50×1000 mm cylinders. Reflector is PTFE.

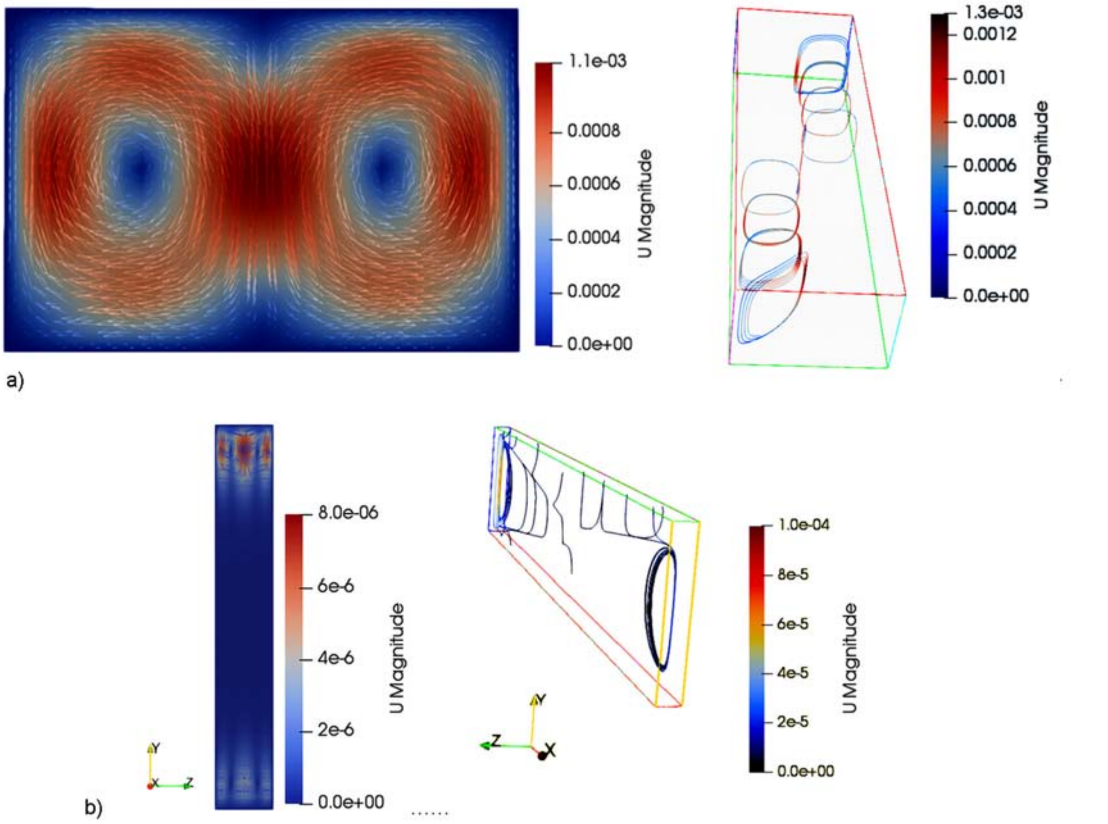


Fig. 2. Convective flows motion at different dimensions of the reaction mass. a) Convective flows motion at dimensions of the reaction mass 2000×400×600 mm. b) Convective flows motion at dimensions of the reaction mass 2000×400×60 mm.

were carried out in a standard configuration with the 600 mm and the 60 mm ampoule width. The polymerization modes were the same as described above.

Plastic scintillators with dimensions of 50×250×1000 mm and of ∅50×1000 mm were made from the blocks produced in two configurations. Further the striae testing was made. The striae were controlled visually against the light, against the background of a black-and-white grid. A sheet of white paper with a black grid printed on it was used to find striae. The size of the grid cell is approximately 10×10 mm, the thickness of the grid line is 1 mm. To determine the presence of striae, the grid was placed close to the polished surface of the plastic scintillator. The inspection was carried out on the side of the scintillator which is opposite to the grid.

For scintillators with dimensions of ∅·1000 mm, the minimum detectable activity (MDA) [12] (testing was carried out under irradiation with an Am241 source), technical light attenuation length (TAL) were compared. The TAL measurements were carried out using a Sr-90 + Y-90 source according to method 1 described in the standard [13]. The light output was also compared. The measurements were carried out using a Cs137 source placed in a collimator. The source was located at a distance of 20 cm from the front and rear ends of the cylindrical scintillator. A PMT R1306 was used as a photodetector. The measurement scheme is shown in Fig. 1.

4. Results and discussion

4.1. Calculation

As a result of the calculations, visual patterns of the flows in the polymerization process were obtained for the ampoule with the width of 600 mm and 60 mm (Fig. 2). The cross sections are vertical, the view from the end of the ampoule. The vectors show the direction of the particles flow, and the velocity magnitude is shown in color.

The maximum flow rate of the reaction mass was also calculated for ampoules with different width (Fig. 3).

As can be seen from the graph (Fig. 3), the maximum velocities of the reaction mass fall sharply when the ampoule width decreases to 60–100 mm. The calculation result showed that with an ampoule width of 600 mm, the main reaction mass has a flow rate close to the maximum values and is approximately $1\cdot10^{-4}$ mm/s (Fig. 2a). With an ampoule width of 60 mm, the bulk of the reaction mass has a flow rate close to zero. And only in some places it can reach about $6\cdot10^{-6}$ mm/s (Fig. 2b).

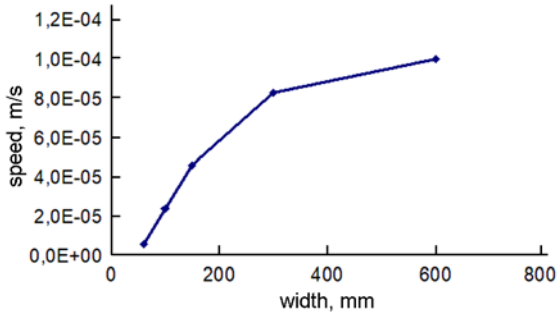


Fig. 3. Convective flows velocity at different ampoule width.

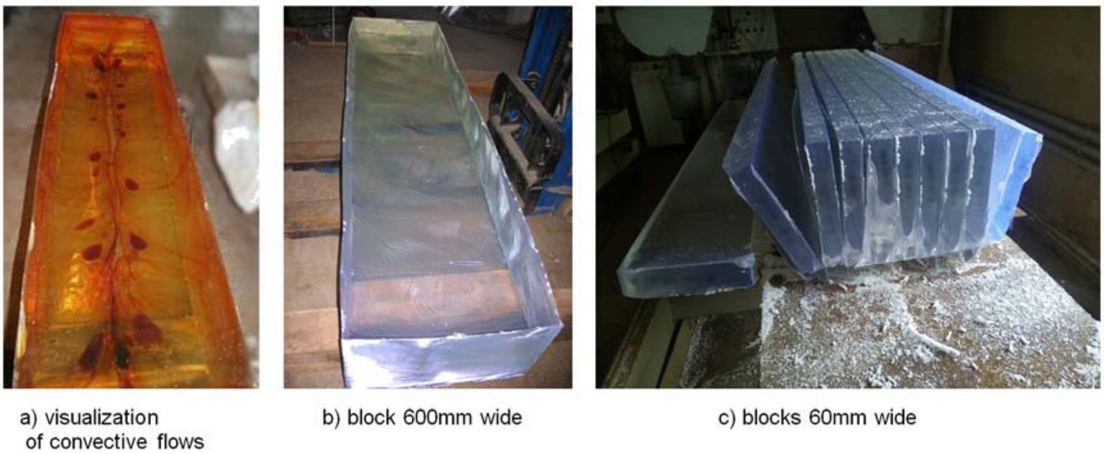
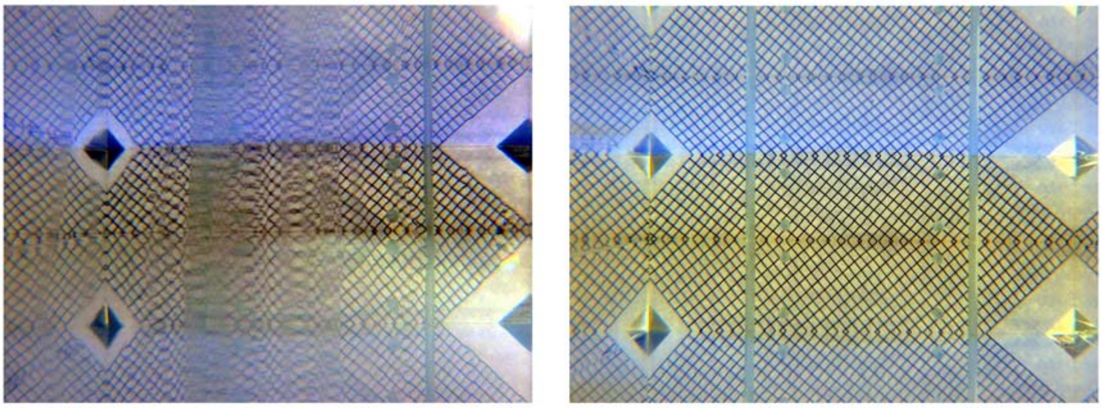
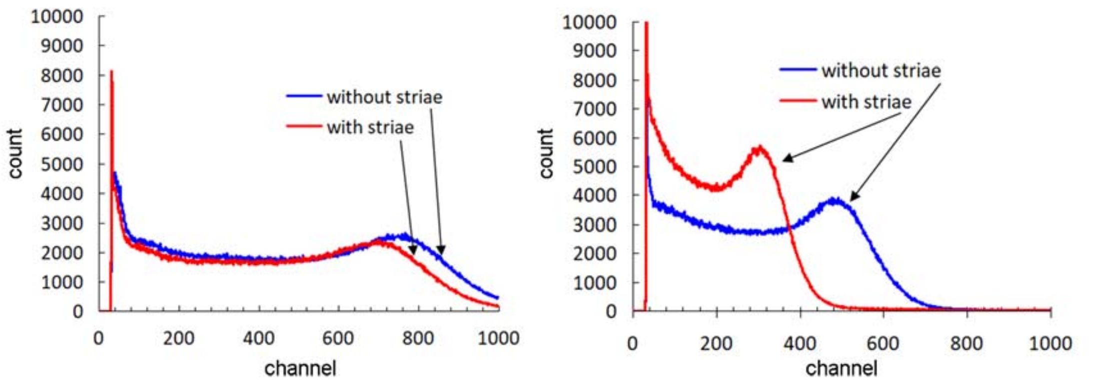


Fig. 4. Obtained polystyrene blocks: a) visualization; b) block 600 mm wide; c) blocks 60 mm wide of convective flows.



a) scintillator with a striae b) scintillator without striae

Fig. 5. Control of optical striae against the background of a black-and-white grid. Inspection along the length of a 50×250×1000 mm scintillation plate: a) scintillator with striae; b) scintillator without striae.



a) the source at 20 cm from the PMT b) the source at 20 cm from the end opposite the PMT

Fig. 6. Scintillation amplitude spectra of ∅50×1000 mm cylinders. Reflector is PTFE: a) the source at 20 cm from the PMT; b) the source at 20 cm from the end opposite the PMT.

4.2. Experiment and measurement results

The result of block polymerization showed visually the nature of the movement of styrene during block polymerization. Figure 4a shows the obtained block.

As the analysis of the experiment results showed, the optical striae in the polystyrene block are due to the formation of convective flows during polymerization.

Further experiments were carried out under standard conditions. A standard block 600 mm wide and 2000 mm long was produced. The standard block is shown in Fig. 4b. Blocks in ampoules with the width of 60 mm were also produced. The blocks with a reduced width are shown in Fig. 4c.

The visual finding of striae against the background of a black-and-white grid is shown in Fig. 5.

Comparative characteristics of plastic scintillators with dimensions of ∅50·1000 mm are listed in Table.

As can be seen from the comparative Table 1, the TAL of scintillators made of scintillation polystyrene without striae is better by 10–25 %. The minimum detection activity during the irradiation with an Am241 source is 2 times better.

The comparative spectrum of the light output measurements of ∅50×1000 mm cylinders at the irradiation with a Cs137 source is shown in Fig. 6.

As can be seen from the obtained spectra, with the source located near the far end from the PMT the light output is about 1.5 times better for a cylinder without striae than for a cylinder with striae.

Table 1. Comparative measurements of ∅50×1000 mm scintillation polystyrene cylinders with and without striae

	With striae	Without striae
Scintillator dimensions	∅50×1000	∅50×1000
TAL	175	192–235
MDA (Am241, 50 cm center), pps-kBq	246	126

5. Conclusions

In this work, the characteristics of polystyrene plastic scintillators made of blocks obtained by bulk polymerization in ampoules of various widths were studied and compared.

A model was constructed and the expected character of the convective flow of the styrene reaction mass at different widths of ampoules for block polymerization was studied. The optimal size of the ampoule width, which is 60–100 mm, is determined.

A method for the comparative visual finding of optical striae in large-size scintillators is proposed.

It was shown that the TAL of plastic scintillators made of scintillation polystyrene with dimensions of ∅50×1000 mm without striae is better by 10–25 %. The minimum detection activity at the irradiation with an Am241 source is 2 times better. It was also shown that with the source located near the far end from the PMT the light output for a ∅50×1000 mm cylinder without striae is about 1.5 times better than for a cylinder with striae.

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