3D PRINTING OF SCINTILLATORS USING LOW-TEMPERATURE PLASTIC MATRICES

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3D-printing of scintillators is a new approach in detector manufacturing. It has found specific applications in the tracking area, including voxel-type and opacity-controlled light propagation. The latter method involves using composite filaments with predetermined opacity. The use of grains of organic scintillators for this purpose is highly beneficial, as they exhibit pulse-shape discrimination features related to triplet exciton reflection. However, this has not been studied before, primarily due to the low melting temperature of organic scintillators. In this work, we created a series of plastic scintillator samples with varying *p*-terphenyl grain content and demonstrated their transparency and light output dependencies.

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INTRODUCTION

3D printing technology opens up a wealth of possibilities for creating complex structures. While there are several basic printing methods, in this work, we will focus on one of the most common: FDM, or fused deposition modeling. This technology is already being successfully used in the production of scintillation detectors.

Currently, the task of determining the position of a scintillation pulse is very popular in high-energy physics. For example, when printing cubic pixels (voxels), it is possible to create their non-transparent walls and go-through foramens for light guides that will transmit the scintillation flash to individual detectors and allow their position to be determined [1]. In this case, the light is isolated in a separate cell, so that the signal emanates only from this cell, thanks to its walls. Another technology involves limiting the propagation of light within the scintillator volume by controlled reduction of the transmittance of the composite material from which the scintillator is composed, specifically by varying the content and size of scintillator grains in the matrix [2]. In this work, we chose the second method.

Using organic grains as scintillators can increase the amount of information obtained about detected events. We previously demonstrated that they possess unique properties that significantly improve pulse shape discrimination [3]. However, FDM technology involves melting plastic filaments, which often have melting points above 200 °C. This temperature is higher than the melting point of *trans*-stilbene and similar to that of *p*-terphenyl. Therefore, the choice of matrix and filament production parameters remains critical.

1. EXPERIMENTAL

1.1. OBTAINING OF CRYSTALLINE GRAINS

The production of organic scintillator grains is described in detail in [4]. We will briefly describe this process.

The main source of grains in this work are *p*-terphenyl single crystals grown using the Bridgman-Stockbarger method. They are immersed in liquid nitrogen, where they crack along the defects. The required fractions are then collected using calibrated sieves. For this stage of studies, we chose 1.2…1.4 mm fraction.

1.2. COMPOSITES CREATION

First, we needed to create a plastic matrix with a melting point that would allow us to embed *p*-terphenyl grains without the risk of them melting. For this, we chose polystyrene (PS) modified with 10% diphenyloxazole. This allowed us to lower its melting point from 230 to 95 °C.

When determining the position of a scintillation pulse, the dependence of scintillation and optical characteristics on grain concentration is critical. This required creating a series of composite samples with varying grain content. In this work, we accomplished this in two ways.

Since the chosen matrix's melting point allowed heating in a water bath, we initially used this method. However, this led to severe, uncontrollable clouding of the composite due to the inclusion of water vapor in the mixture.

The second method we used was a laboratory heating oven. Creating a specialized heating, holding, and cooling profile allowed us to obtain samples suitable for further study. To analyze the dependence of the parameters on grain concentration, we selected composites with grain contents of 17, 33, and 50 wt.%. The geometry of the samples was chosen to minimize the difference in light collection conditions compared to the reference samples, namely: 30 mm diameter and 5 mm thickness. Photographs of these composites are shown below in Fig. 1.

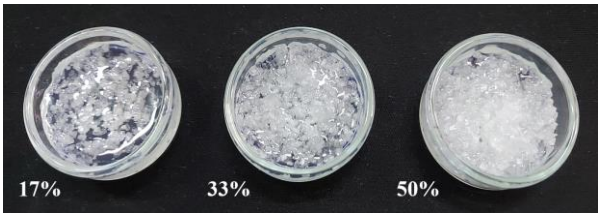

Fig. 1. Photo of the composites with different content of p-terphenyl grains (17, 33, and 50 wt.%, respectively)

1.3. EXPERIMENTAL METHODS

Light output measurements were performed using the standard method [3], analyzing scintillation amplitude spectra. The well-known UPS-923A polystyrene scintillator was used as a reference.

Optical transmittance measurements were performed using a Shimadzu UV2450 spectrophotometer with an integrating sphere. The reference beam passed through the air. Therefore, the final transmittance value in % can be calculated as $T=(L_{samp}/L_{air})\times100\%$, where L_{samp} is the value in the sample channel and L_{air} is the value in the reference channel.

2. RESULTS OF STUDIES AND THEIR ANALYSIS

As noted in Section 1.2, for the purposes of this work, we used samples with a diameter of 30 mm and a thickness of 5 mm. We varied the amount of *p*-terphenyl grains in the modified PS matrix from 17 to 50 wt.%, with a grain size of 1.2…1.4 mm. The sample with the same geometry but without *p*-terphenyl grains was used as a reference.

The figures below show the scintillation amplitude spectra for these samples. Irradiation was performed using the ^{90}Sr and ^{137}Cs isotopes.

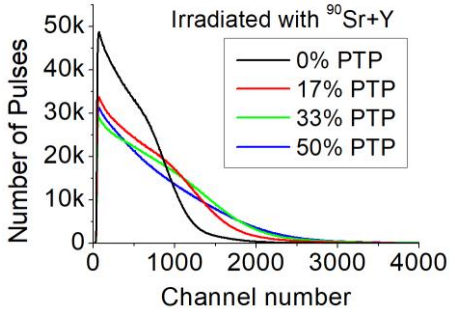

Fig. 2. Scintillation amplitude spectra of the composite samples and reference sample. Irradiated with ^{90}Sr source

As shown in Fig. 2, all samples exhibit relatively high light output when irradiated with ^{90}Sr beta-particles. The inflection point shifts toward higher channels as the grain content increases (this indicates that light output increases with increasing grain concentration). However, it cannot be calculated directly due to the low energy resolution.

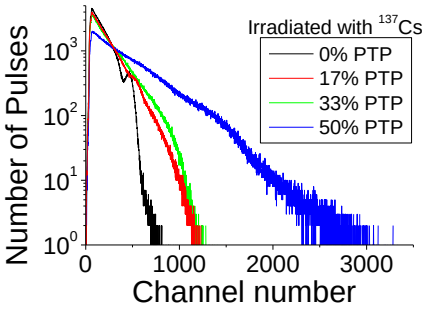

Fig. 3. Scintillation amplitude spectra of the composite samples and reference sample. Irradiated with ^{137}Cs source

As shown in Fig. 3, the tendency of increasing light output with increasing grain concentration is even more pronounced when irradiating samples with ^{137}Cs gamma rays and beta particles. However, the problem of low energy resolution remains.

Let us move on to optical studies. For this point, we measured the light transparency of our samples using the method described in 1.3. The light transparency was measured in the range from 190 to 800 nm as shown in Fig. 4.

Let us move on to the optical studies. For this, we measured the optical transmittance of our samples using the method described in Section 1.3. Optical transmittance was measured in the range from 190 to 800 nm, as shown in Fig. 4.

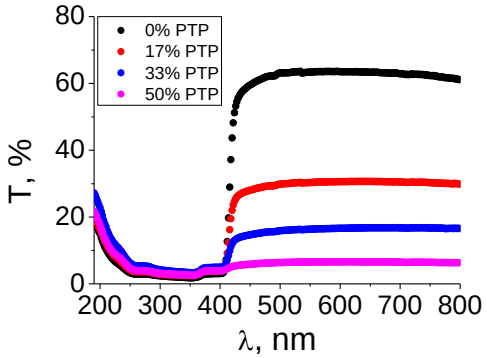

Fig. 4. Optical transmittance of the studied samples. The grain concentrations are indicated in the figure legend

As shown in Fig. 4, optical transmittance decreases with increasing grain content. To estimate this effect, the approximation shown in Fig. 5 was used.

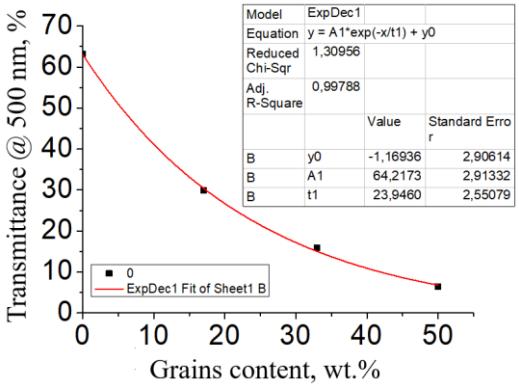

Fig. 5. Dependence of optical transmittance at a 500 nm wavelength on the grain content

To approximate the optical transmittance decrease, we used Origin 8.5's built-in "One-phase exponential decay function with a time constant parameter". It can be described by the following function: $T=T_0+C\cdot\exp(-N/D_c)$, where T is the transmittance, N is the grain concentration in wt.%, T_0 is the transmittance offset, C is the amplitude and D_c is the decay constant. For the studied compound, these constant parameters were calculated as T_0=-1.17, C=64.22, and D_c=23.94. All experimental points follow the approximation curve with high accuracy.

CONCLUSIONS

1. For the first time, it has been demonstrated that single-crystal organic scintillator grains can be used to create composite scintillators with a plastic matrix. This opens the possibility of creating corresponding filaments for FDM 3D printing.
2. The light output of the created compounds shows a strong dependence on the grain content, increasing with the grain concentration.
3. The optical transmittance of the created composite samples gradually decreases with increasing grains concentration and can be accurately described by an exponential decay with constants calculated in this work.

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3D-ДРУК СЦИНТИЛЯТОРІВ З ВИКОРИСТАННЯМ НИЗЬКОТЕМПЕРАТУРНИХ ПЛАСТИКОВИХ МАТРИЦЬ

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3D-друк сцинтиляторів – це новий підхід у виробництві детекторів. Він знайшов специфічне застосування в галузі трекінгу, включаючи воксельне та контрольоване за непрозорістю поширення світла. Останній метод передбачає використання композитних філаментів із заданою непрозорістю. Використання гранул органічних сцинтиляторів для цієї мети є дуже вигідним, оскільки вони демонструють особливість розділення за формою імпульсу, пов’язану з відбиттям триплетних екситонів. Однак, такі філаменти раніше не вивчалися, головним чином через низьку температуру плавлення органічних сцинтиляторів. У цій роботі ми створили серію зразків пластикових сцинтиляторів з різним вмістом зерен *n*-терфенілу та продемонстрували залежності їх прозорості та світлового виходу.