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PECULIARITIES OF THE FORMATION OF SCINCILLATION RESPONSE IN ORGANIC MATERIALS WITH STOCHASTIC CHARACTER OF LIGHT PROPAGATION

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The paper examines the possibility of using an organic polycrystal as the "opaque" scintillator. Polycrystals are produced by pressing crystalline grains. When light propagates through a polycrystal, it is repeatedly reflected and refracted at the boundaries of the grains. This makes its propagation difficult. We studied the light output and optical transmittance of stilbene and *p*-terphenyl polycrystals with different fractions of crystalline grain: from 0.06…0.1 to 2.0…2.5 mm (the samples 20 mm in diameter and 2 mm in height) was conducted. Modelling of light propagation in polycrystalline samples of stilbene and *p*-terphenyl was carried out and the values of the light collection coefficients were calculated. It was found that in order to obtain the polycrystalline samples with sufficiently high light output and high efficiency of detection of local sites of interaction of ionizing radiations, grains in the range of 0.4…0.8 mm should be used.

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INTRODUCTION

In the vast majority of applied problems, transparent scintillators are used. Transparency is an important characteristic of the scintillator, its decrease leads to a decrease in the intensity of light reaching the photodetector, that is, to a decrease in the value of the technical light output. Recently, the idea of using precisely the scintillators with low transparency ("opaque" scintillators) to localize the local place of scintillation pulse was arisen [1]. Complicating the propagation of scintillation light through such an "opaque" scintillator and the use of lightguides located at a certain distance from each other in the volume of the scintillator allow solving this problem. Liquid scintillators are proposed to be used as "opaque" scintillation materials [1]. However, liquid scintillators are known to be inconvenient when used in practice due to their fire hazard, thermal expansion, leakage hazard, toxicity, etc.

The use of organic polycrystals as an "opaque" scintillator, as an alternative to liquid scintillators, seems promising. Polycrystalline scintillators are made by pressing crystal grains or plates. Therefore, when light propagates through a polycrystal, it is repeatedly reflected and refracted at the boundaries of grains or plates, that makes its propagation difficult. Note, that polycrystalline scintillators retain most of the advantages of singlecrystalline scintillators (high decay time, high efficiency of detecting short-range and neutron radiation, the ability to separately detection an ionizing radiation, etc.), but at the same time have a significantly lower cost. In addition, polycrystalline scintillators are quite resistant to the appearance of cracks during mechanical processing, that allows the integration of lightguides into their volume. Changing the conditions for obtaining polycrystals (temperature, pressure, grain size) allows to vary their scintillation characteristics (for example, light output), as well as the degree of their transparency. At the same time, the goal of previous research was to achieve the maximum values of light output and transparency of polycrystals.

Determining the conditions for obtaining polycrystalline scintillators, which are characterized at the same time by high light output, high uniformity of light output and a given degree of scattering, is an urgent task.

1. EXPERIMENTAL

1.1. SAMPLES PREPARATION

To obtain trans-stilbene and *p*-terphenyl grains, quasi-crystalline ingots obtained after zone melting of trans-stilbene and *p*-terphenyl raw materials were used. The resulting grains were sifted through calibrated sieves and divided into fractions. The sizes of the holes in the calibrated sieves we selected were: 0.06, 0.1, 0.3, 0.5, 1.0, 1.5, 2.0, and 2.5 mm. Samples of polycrystalline scintillators were made from crystalline grains by the method of hot pressing [2–5]. Fig. 1 presents the photo of polycrystalline *p*-terphenyl scintillators obtained from grains of different fractions.

Fig. 1. Photo of polycrystalline scintillators of p-terphenyl obtained from grains of different fractions

1.2. EXPERIMENTAL METHODS

The relative light output of polycrystalline scintillators was determined by the standard method using scintillation amplitude spectra. The photomultiplier 9208A (Electron Tubes Ltd.) was used as a photodetector, which has a dark current of $6.8 \cdot 10^{-11}$ A at an anode sensitivity of 50 A/Lm [6]. The amplitude distribution of scintillator pulses was obtained using an amplitude-to-digital converter. The

relative value of the light output value was calculated according to (1):

$$L = \left(J \middle/ J_{ref} \right) \cdot 100\,\%, \quad (1)$$

where J is the value of the amplitude that corresponds to the center of gravity of the peak in the spectrum of the scintillator under investigation, J_{ref} is the value of the amplitude that corresponds to the center of gravity of the peak in the spectrum of the reference scintillator. Single crystals of stilbene and *p*-terphenyl were used as reference scintillators.

We used the following sources of ionizing radiation: ^{239}Pu (alpha particles with energy $E_\alpha = 5.15$ MeV, specific energy loss $dE/dx \sim 10^3$ MeV/cm); ^{137}Cs (conversion electrons with energy $E_\beta = 0.6225$ MeV, specific energy loss $dE/dx \sim 10^{-1}$ MeV/cm).

The optical transmittance of the samples was determined using a Shimadzu UV 2450 spectrophotometer [7] using an integrating sphere. Measurements were performed in the wavelength range from 200 to 750 nm. The value of optical transmittance T was calculated as follows:

$$T = \left(L \middle/ L_0 \right) \cdot 100\,\%, \quad (2)$$

where L_0 is the flux of light falling on the sample, L is the flux of light that propagates through the sample. In fact, the value of T (2) is the relative light transmittance, where $T = 100\%$ is the light transmittance of air at room temperature.

1.3. MODELLING OF LIGHT PROPAGATION IN HETEROGENEOUS MATERIAL

Computer modeling of scintillation light propagation in a heterogeneous medium was used to determine the light collection coefficients. In the modeling program, the heterogeneous scintillator is represented as a dispersed phase and a dispersed medium, i.e., a system with many boundaries separating two phases with different physical properties. In a number of works [8, 9], the heterogeneous scintillator is presented in the form of densely packed balls with a dispersed medium that fills the voids (Fig. 2).

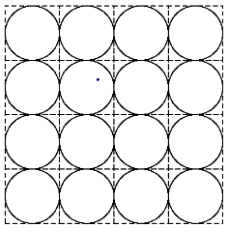

Fig. 2. A model of a heterogeneous scintillator in the form of spheres with dense volume-centered packing

In the geometric model of the heterogeneous scintillator that we used, its volume is also divided into simulated cells. Each cell contains a portion of the dispersed phase of a given shape (with fixed or random size and location). Such a geometric model allows obtaining different variants of modeling heterogeneous scintillators. For the dispersed phase in the form of spheres, a volume filling factor of 0.52 can be obtained (dense volume-centered packing). This approach allows representing systems with a random location in cells of particles of different or the same size. The approach used in this work to model the propagation of light in heterogeneous scintillators was described in [10].

The simulation program uses the Monte Carlo method. At various stages the specific parameters of the simulation (locations and directions of radiation, absorption in the volume and refraction/reflection at the boundaries of the media, etc.) are randomly selected ("randomized") based on the respective coefficients or distributions. The MeshTarReflector program was used for modeling.

2. RESULTS OF INVESTIGATION AND THEIR ANALYSIS

2.1. LIGHT OUTPUT

The paper investigated the relative light output of samples of stilbene and *p*-terphenyl polycrystals obtained from 7 fractions of crystalline grains of different sizes, namely: 0.06…0.1, 0.1…0.3, 0.3…0.5, 0.5…1.0, 1.0…1.5, 1.5…2.0, 2.0…2.5 mm. Sample thickness $h = 2$ mm, diameter $d = 20$ mm. Corresponding single crystals with a thickness of $h = 5$ mm and a diameter of $d = 30$ mm were used as reference detectors.

As an example, Fig. 3 presents the results of calculations of the relative light output of stilbene polycrystals with different sizes of crystal grains under alpha excitation, and Fig. 4 presents the results of calculations of the relative light output of *p*-terphenyl polycrystals with different sizes of crystal grains upon excitation by conversion electrons.

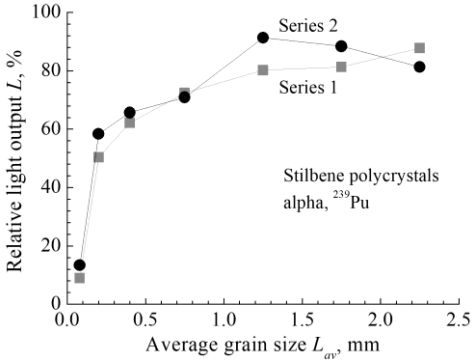

Fig. 3. Relative light output L of stilbene polycrystals with different sizes of crystal grains under alpha excitation

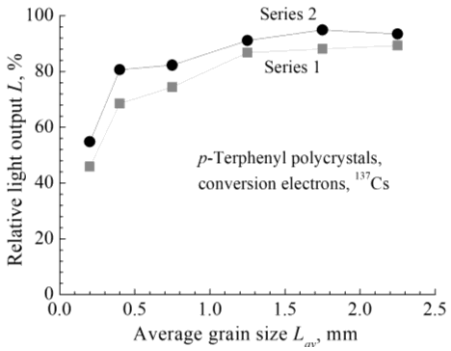

Fig. 4. Relative light output L of p-terphenyl polycrystals with different sizes of crystal grains upon excitation by conversion electrons

2.2. OPTICAL TRANSMITTANCE OF SAMPLES

Figs. 5 and 6 present the dependences of optical transmittance T of a series of polycrystals of stilbene and *p*-terphenyl, respectively, on the average size of crystalline grains L_{av}. The T-values are given for the luminescence wavelength (λ = 400 nm) and for the scintillator transparency region (λ = 700 nm).

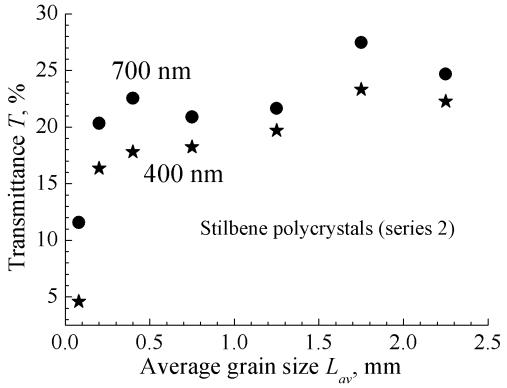

Fig. 5. The value of the optical transmittance T of stilbene polycrystals at the wavelength of luminescence (λ = 400 nm) and in the region of transparency (λ = 700 nm)

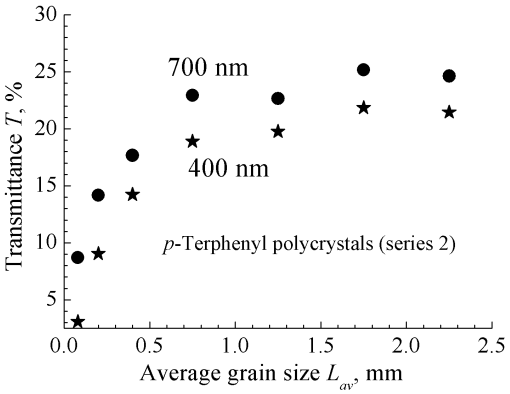

Fig. 6. The value of the optical transmittance T of p-terphenyl polycrystals at the wavelength of luminescence (λ = 400 nm) and in the region of transparency (λ = 700 nm)

2.3. RESULTS OF SIMULATION OF LIGHT PROPAGATION IN POLYCRYSTALLINE SAMPLES OF STILBENE AND *P*-TERPHENYL SCINTILLATORS

The propagation of light in polycrystalline samples of stilbene and *p*-terphenyl scintillators obtained by the hot pressing method was simulated. Both the size of the samples with a diameter of 20 mm and a height of 2 mm, and the size of the grains were specified. It was assumed that in the process of pressing, the sizes of the initial scintillator grains change slightly, except for the largest ones. For the size series of the original real grains 2.25, 1.75, 1.25, 0.75, 0.4, 0.2, and 0.08 mm, practically the same series of virtual cells was used: 2×2.5, 1.75, 1.25, 0.75, and 0.4 mm. The "gap" (boundary layer) between the particles was set to 0.01 mm. Refractive indices and light absorption coefficients for the scintillator and boundary layer were also specified. An external reflector was not used during the simulation.

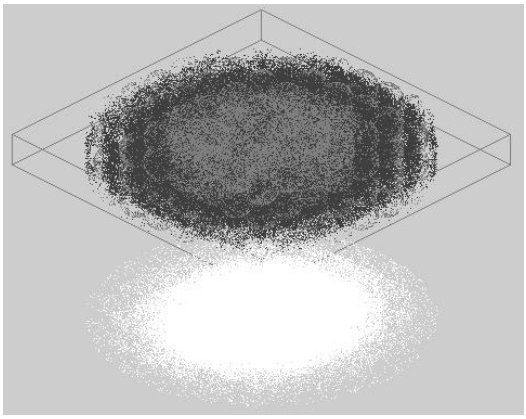

Fig. 7. Light propagation in a heterostructured scintillator with a grain size of 2 mm

During the operation of the program, the trajectories (lines) or the places of changes in the trajectories (points) of the rays were displayed. Screenshots of examples of displaying the places of changes in the trajectories (points) of the rays are shown in Figs. 7–9.

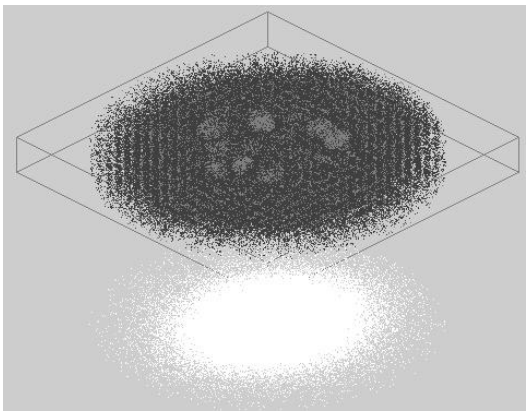

Fig. 8. Light propagation in a heterostructured scintillator with a grain size of 0.75 mm

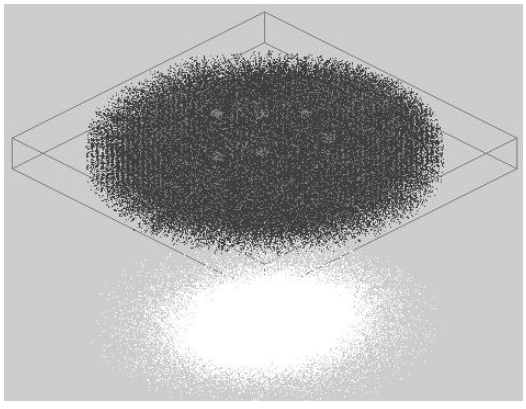

Fig. 9. Light propagation in a heterostructured scintillator with a grain size of 0.4 mm

It can be seen that the positions of the points demonstrate the structure of the model object, that is, the places of refraction/reflection at the phase boundaries. The results of calculations of light collection coefficients τ are given in Table.

In all cases the light collection coefficient decreases with decreasing a grain size, which is explained by an increase in the number of reflections/refractions and the path length.

The value of light collection coefficients τ of polycrystalline samples

Average grain size, mm	Light collection coefficients τ			
	Stilbene		*p*-Terphenyl	
	Alpha	Electrons	Alpha	Electrons
0.08	0.191	0.315	0.117	0.303
0.2	0.239	0.379	0.183	0.366
0.4	0.280	0.433	0.231	0.436
0.75	0.281	0.445	0.276	0.445
1.25	0.327	0.514	0.282	0.510
1.75	0.389	0.597	0.324	0.594
2.25	0.379	0.602	0.383	0.597

This increases the probability of absorption in the volume or exiting the volume boundaries. When modeling the propagation of light from alpha particles (the emergence of light in the surface layer of 0.35 μm), the light collection coefficients are smaller than in the case of internal conversion electrons (the range in the considered substances is more than 2 mm). In the latter case, the average path of the rays to the output window of the photodetector will be smaller than in the first case. The corresponding light collection coefficients for stilbene and *p*-terphenyl are approximately the same, which is consistent with experimental measurements of the optical transmittance of the samples and the relative light output. This correspondence also indicates the main role of the light collection process in the formation of the light signal at the output of the scintillator.

2.4. ANALYSIS OF THE EXPERIMENTAL RESULTS

We analyzed the above experimental results, namely, 1) the results of measuring the relative light output of samples of stilbene and *p*-terphenyl polycrystals obtained from 7 fractions of crystalline grains of different sizes, namely: 0.06…0.1, 0.1…0.3, 0.3…0.5, 0.5…1.0, 1.0…1.5, 1.5…2.0, and 2.0…2.5 mm; 2) the results of the study of the optical transmittance values T of the above-mentioned polycrystal samples, and 3) the results of the calculations of the light collection coefficients τ for these samples. The results of this analysis are combined in Figs. 10–13, where the normalized values of the relative light output L, optical transmittance T, and light collection coefficients τ are presented as a function of the average size L_{av} of grains of these polycrystalline samples. Each of these parameters is obtained by normalization to its maximum values obtained in separate calculations. This made it possible to visually combine the values of various parameters in one figure.

Figs. 10 and 11 combine the results of calculations of light collection coefficients τ, transmittance T and relative light output L for stilbene polycrystals, respectively, upon excitation by ^{239}Pu alpha particles and conversion electrons of the ^{137}Cs source. Of course, the value of optical transmittance T (at a wavelength of λ = 400 nm) is the same as in Figs. 10 and 11. Figs. 12 and 13 present similar normalized values of light collection coefficients τ, transmittance T and relative light output L for *p*-terphenyl polycrystals.

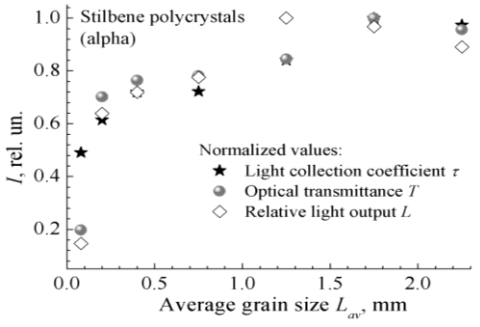

Fig. 10. Normalized values of light collection coefficient τ, optical transmittance T (λ = 400 nm) and relative light output L for stilbene polycrystals excited by alpha particles of ^{239}Pu

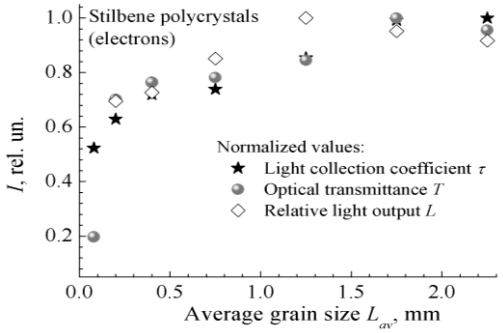

Fig. 11. Normalized values of light collection coefficient τ, optical transmittance T (λ = 400 nm) and relative light output L for stilbene polycrystals excited by conversion electrons of ^{137}Cs

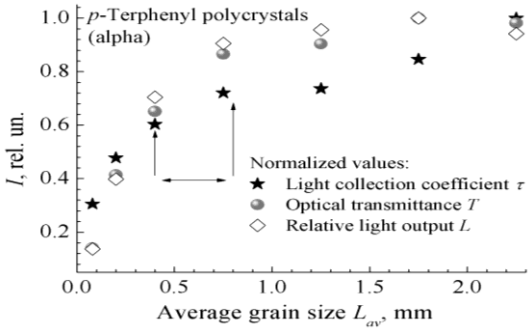

Fig. 12. Normalized values of light collection coefficient τ, optical transmittance T (λ = 400 nm) and relative light output L for p-terphenyl polycrystals excited by alpha particles of ^{239}Pu

On the one hand, the scintillation a detector must have a sufficiently high light output and a sufficiently high transparency. But on the other hand, for its possible use as an "opaque" scintillation detector for effective detection of local interaction sites of ionizing radiations, its transparency and propagation of scintillation photons should be sufficiently limited. The analysis of the data presented above (see Figs. 10–13) shows that polycrystalline samples obtained from fractions of grains 0.06…0.1 and 0.1…0.3 mm have extremely low values of the of light collection coefficients τ, transmittance T (λ = 400 nm) and the relative light output L.

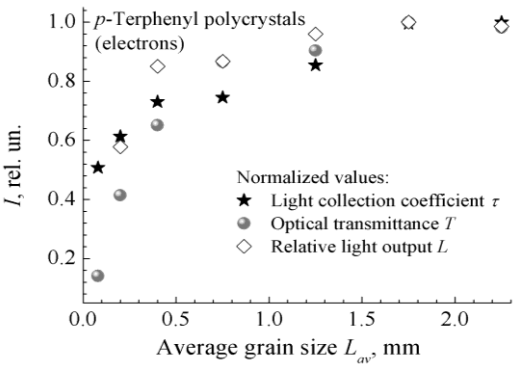

Fig. 13. Normalized values of light collection coefficient τ, optical transmittance T (λ = 400 nm) and relative light output L for p-terphenyl polycrystals excited by conversion electrons of ^{137}Cs

This is characteristic of both polycrystalline stilbene samples and *p*-terphenyl samples. This regularity also actually does not depend on the type of radiation excitation (alpha particles or conversion electrons). Starting with grain fractions of 1.0…1.5, 1.5…2.0, 2.0…2.5 mm, the values of light collection coefficients τ, transmittance T (λ = 400 nm) and the relative light output L stop increasing significantly and tend to saturate. Therefore, to obtain an "opaque" polycrystalline scintillation detector, we consider it optimal to use crystal grains in the range ~ 0.4…0.8 mm (see, for example, Fig. 12).

CONCLUSIONS

The light output and optical transmittance of stilbene and *p*-terphenyl polycrystals with the following fractions of crystalline grains were studied, namely, 0.06…0.1, 0.1…0.3, 0.3…0.5, 0.5…1.0, 1.0…1.5, 1.5…2.0, and 2.0…2.5 mm (the samples 20 mm in diameter and 2 mm in height). Modelling of light transmission in polycrystalline samples of stilbene and *p*-terphenyl scintillators obtained by the hot pressing method was carried out. We calculated the values of light collection coefficients τ of polycrystalline samples both for the case of excitation by alpha particles, and conversion electrons. It was found that in order to obtain polycrystalline samples with sufficiently high light output and high efficiency of detection of local interaction sites of ionizing radiations, crystal grains in the range of 0.4…0.8 mm should be used.

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ОСОБЛИВОСТІ ФОРМУВАННЯ СЦИНТИЛЯЦІЙНОГО ВІДГУКУ В ОРГАНІЧНИХ МАТЕРІАЛАХ ІЗ СТОХАСТИЧНИМ ХАРАКТЕРОМ РОЗПОВСЮДЖЕННЯ СВІТЛА

Я.І. Полупан, І.В. Лазарєв, Є.В. Мартиненко, С.С. Міненко, О.А. Тарасенко, В.О. Тарасов

Досліджується можливість використання органічного полікристала в якості «мутного» сцинтилятора. Полікристали виготовляють методом пресування кристалічних гранул. Світло при розповсюдженні крізь полікристал зазнає багаторазового відбивання та заломлення на границях гранул, що ускладнює його проходження. Проведено дослідження світлового виходу та оптичного пропускання полікристалів стильбену та *n*-терфенілу із різними фракціями кристалічних гранул: від 0,06…0,1 до 2,0…2,5 мм (діаметр зразків 20 мм та висота 2 мм). Проведено моделювання проходження світла в полікристалічних зразках стильбену та *n*-терфенілу, та розраховані значення коефіцієнтів світлозбирання в них. З'ясовано, що для отримання полікристалічних зразків із достатньо високим світловим виходом та високою ефективністю виявлення локальних місць взаємодії іонізуючого випромінювання слід використовувати гранули в діапазоні 0,4…0,8 мм.