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ORGANIC COMPOSITE DETECTORS FOR SELECTIVE DETECTION OF ALPHA AND GAMMA RADIATIONS

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We have studied the alpha-gamma discrimination capability ($FOM_{\alpha\gamma}$) of organic composite scintillators based on stilbene and *p*-terphenyl. Two series of composite detector samples with varying fractions of crystalline grains in the range of average grain size from 0.2 to 1.9 mm, with a thickness of 4 mm and a diameter of 15 mm, were studied. It was shown that for composite samples based on both stilbene and *p*-terphenyl, the $FOM_{\alpha\gamma}$-values were significantly higher compared to reference samples of the corresponding single crystals. A trend towards a slight increase in the $FOM_{\alpha\gamma}$-value was observed for samples with the largest L_{av}-values.

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INTRODUCTION

The issue of detecting low-intensity ionizing radiation fluxes is of paramount importance in contemporary environmental, biological, and medical tasks, as well as in preventing the unauthorized transport of nuclear fuel and other radioactive materials.

Organic luminescent materials (single crystals, liquid phosphor solutions, and polymers) have proven effective in many detection and spectrometry tasks for ionizing radiation, particularly in the separate detection of ionizing radiations. This primarily concerns the issue of fast neutron spectrometry in the presence of background gamma radiation [1-7]. The physical basis for such separation lies in the difference in the pulse shapes of radioluminescence generated by ionizing radiations with different specific energy loss values (*dE/dx*) [2, 6, 8, 9]. As is well known, the radioluminescence pulse of an organic scintillator contains both fast and slow components. When excited by ionizing radiation with high *dE/dx*-values (alpha particles, fast neutrons), the contribution of the slow component is greater than when excited by radiation with low *dE/dx*-values (electrons, gamma photons). This criterion enables the identification of radiation type in real experiments [8, 9].

Heterogeneous organic scintillators are produced from single crystalline grains. This approach avoids the stage of growing structurally perfect single crystals, significantly reducing the cost of producing organic scintillation grains and scintillators based on them. They consist of a mass of single crystalline grains that are bonded together either by sintering under pressure (polycrystals) [1, 10], or using a transparent non-scintillating matrix (composite scintillators) [1, 3, 5, 7, 10]. The main advantage of composite scintillators is the ability to create scintillation detectors of large size and arbitrary shapes.

In our previous studies [11], we investigated the light output of polycrystalline and composite scintillators based on crystalline grains of stilbene and *p*-terphenyl, depending on the average grain size L_{av} used to prepare the sample. Additionally, the neutron-gamma discrimination capability of the samples was studied when irradiated with a combined source of fast neutrons and photons of gamma radiation. It was found that for composite samples based on both stilbene and *p*-terphenyl, the $FOM_{n\gamma}$-values (the quality of discrimination capability), firstly, do not have a certain dependence on the size of the grains (in the studied range). Secondly, composite samples have sufficiently high $FOM_{n\gamma}$-values in comparison with the single crystals: for stilbene composite detectors it is on average 76% of the corresponding single crystal and for *p*-terphenyl composite scintillators it is about 70% [11]. This work is dedicated to the study of alpha-gamma discrimination capability of organic composite scintillators based on stilbene and *p*-terphenyl.

1. SAMPLE PREPARATION

We obtained two series of experimental samples of composite scintillators based on stilbene and pure *p*-terphenyl (without fluorescent additional agents). The grains were obtained by crushing crystalline ingots of previously grown structurally perfect single crystals. The grains were sieved to produce grains of different fractions. For each series, we used the following 7 grain fractions: 0.1…0.3, 0.3…0.5, 0.5…1.0, 1.0…1.2, 1.2…1.4, 1.4…1.8, and 1.8…2.0 mm. Grains of each fraction were placed in a non-scintillating gel composition, SYLGARD-184. The samples were then left to polymerize for 24 hours. A more detailed description of the technology for producing organic composite scintillation detectors can be found in our previous works [1, 3, 5, 7, 10]. The samples had a thickness of 4 mm and a diameter of 15 mm.

We determine the average size L_{av} of grain of the specific fraction as the mean between the smaller and the larger mesh sizes of our sieves, which were used to select grain fraction. It can be represented as $L_{av} = (L_i + L_{i+1})/2$, where L_{av} is the average grain size, L_i is the smaller mesh size and L_{i+1} is the larger mesh size used to create the sample [12].

The reference detectors used were corresponding single crystals of stilbene and pure *p*-terphenyl, grown by the Bridgman-Stokbarger method [13]. The single crystals had a diameter of 25 mm and a height of 5 mm.

2. EXPERIMENTAL

A measurement system for studying the discrimination capability of organic scintillation crystals and calculating the *FOM*-value was developed and assembled. The system includes the following components (Fig. 1): the photomultiplier with power supply units, the pulse signal amplifier-shaper, the signal control block for the time-to-amplitude converter; the time-to-amplitude converter, the multichannel pulse amplitude analyzer, and the computer. The signals from the photomultiplier are integrated and differentiated in the amplifier-shaper and are then sent to the signal control block for the time-to-amplitude converter. The generated signal is simultaneously directed to the inputs of the level discriminator and zero-crossing detector. The signal from the discriminator is first delayed and then sent to the pulse shaping circuit "start" for the time-to-amplitude converter, while the signal from the zero-crossing detector is directed to the "stop" pulse shaping circuit. The "start" and "stop" pulses control the time-to-amplitude converter. The amplitude spectrum is built using the signals obtained via the multichannel pulse amplitude analyzer and software, which is then used to calculate the parameters of organic single crystal and heterogeneous scintillators.

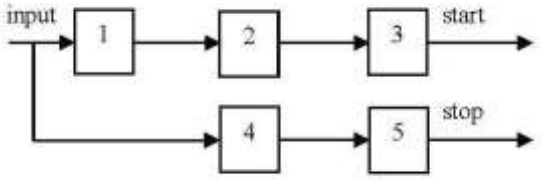


Fig. 1. The measurement setup for studying the discrimination capability (the FOM-value) of organic scintillation materials: 1 is a level discriminator, 2 is a delay circuit, 3 is a pulse shaper "start", 4 is a zero-crossing detector, and 5 is a pulse shaper "stop"

3. RESULTS AND DISCUSSION

The $FOM_{\alpha\gamma}$-value (the parameter that determines the discrimination capability of a detector) was investigated for samples with different fractions of crystalline grain in the range of the average grain size L_{av} from 0.2 to 1.9 mm. The values of L_{av} were: 0.2, 0.4, 0.75, 1.1, 1.3, 1.6, and 1.9 mm.

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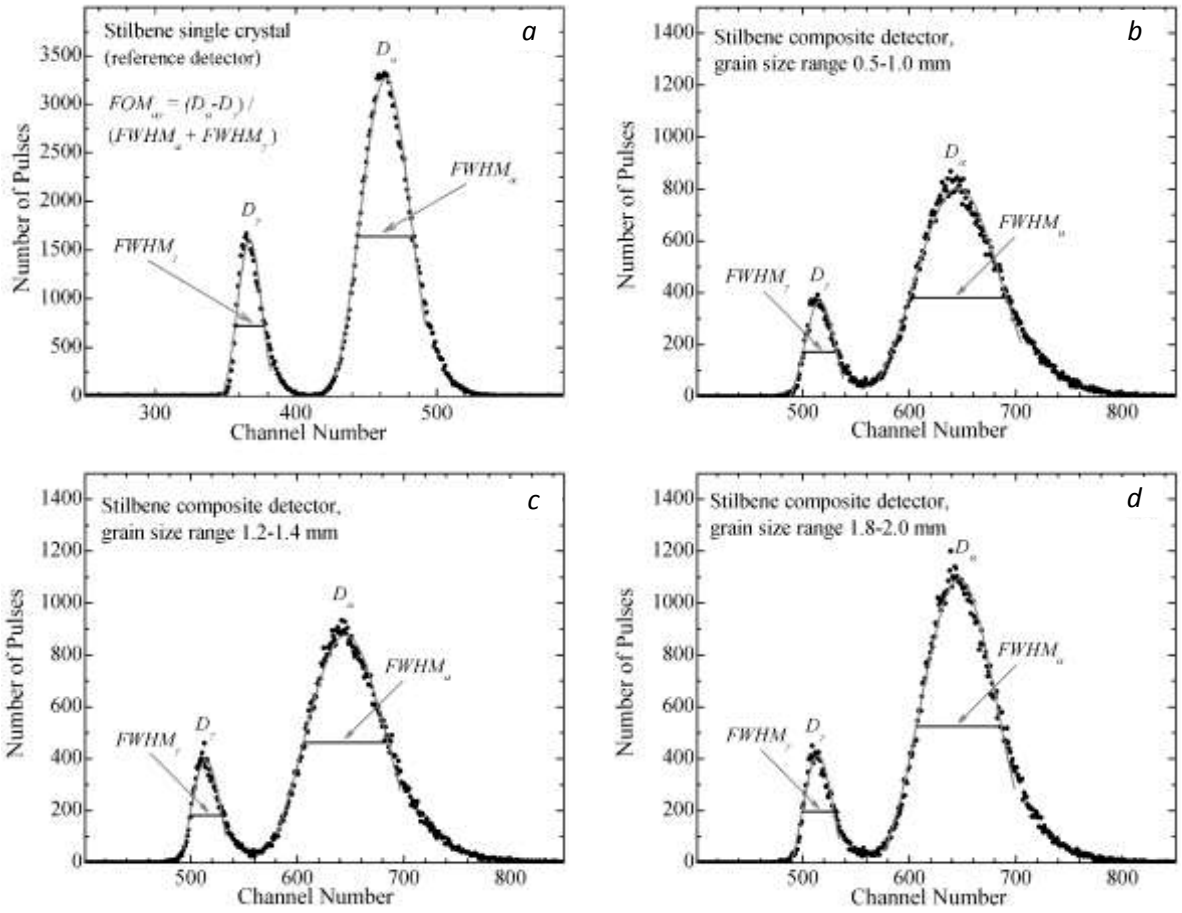


Fig. 2. Typical spectra obtained using the "zero-crossing" method for the stilbene single crystal and stilbene composite scintillators when excited by an alpha-particles and photons of gamma radiation from ^{239}Pu source: $FWHM_{\gamma}$ – full width at half maximum of the gamma peak (in channels), $FWHM_{\alpha}$ – full width at half maximum of the alpha peak (in channels), $D_{\alpha\gamma}$ – arithmetic difference between the position of the alpha peak and the gamma peak (in channels)., $FOM_{\alpha\gamma}$ – alhpa-gamma discrimination capability. The grain sizes of the composite scintillators are shown in the figure

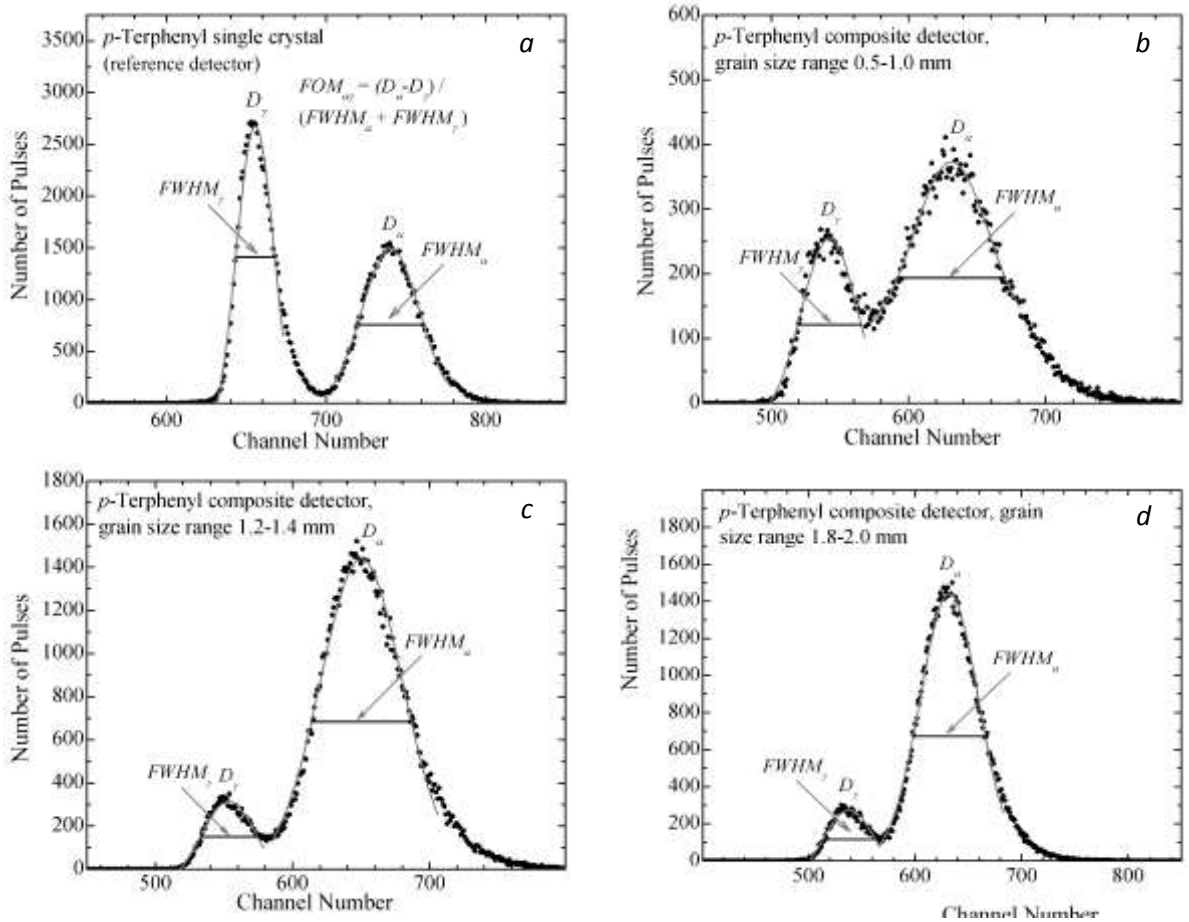


Fig. 3. Typical spectra obtained using the "zero-crossing" method for the p-terphenyl single crystal and p-terphenyl composite scintillators when excited by an alpha-particles and photons of gamma radiation from ^{239}Pu source. Other symbols are the same as in Fig. 2

Fig. 2 shows typical spectra obtained using the "zero crossing" technique for the stilbene single crystal and composite stilbene scintillators when excited by an alpha-particles (5 MeV) and photons of gamma radiation (60 keV) from ^{239}Pu. Fig. 3 presents similar results for *p*-terphenyl detectors. For demonstration, three samples from grains of different fractions were arbitrarily selected. Figs. 2 and 3 present composite detectors as examples, featuring L_{av} grain sizes of 0.75, 1.3, and 1.9 mm.

The $FOM_{\alpha\gamma}$-value was calculated using the formula [11]:

$$FOM_{\alpha\gamma} = \frac{D_{\alpha\gamma}}{FWHM_{\alpha} + FWHM_{\gamma}}, \quad (1)$$

where $FWHM_{\gamma}$ – full width at half maximum of the gamma peak (in channels); $FWHM_{\alpha}$ – full width at half maximum of the alpha peak (in channels); $D_{\alpha\gamma}$ – arithmetic difference between the position of the alpha peak and the gamma peak (in channels).

Table 1 presents the values of $FWHM_{\gamma}$, $FWHM_{\alpha}$, $D_{\alpha\gamma}$, as well as the calculated value of $FOM_{\alpha\gamma}$ (1) for the reference single crystal of stilbene and for the entire series of composite scintillator samples based on stilbene, obtained from 7 grain fractions with different L_{av}-values.

Table 1

Values of $FWHM_{\gamma}$, $FWHM_{\alpha}$, $D_{\alpha\gamma}$, and $FOM_{\alpha\gamma}$ for stilbene composite detectors and the single crystal

	$FWHM_{\gamma}$, ch	$FWHM_{\alpha}$, ch	$D_{\alpha\gamma}$, ch	$FOM_{\alpha\gamma}$
Single crystal	20	39	98	1.66
Composite detectors				
L_{av}, mm				
0.2	32	–	–	–
0.4	29	78	136	1.27
0.75	31	83	125	1.09
1.1	29	79	135	1.25
1.3	26	79	129	1.23
1.6	28	74	133	1.31
1.9	28	72	130	1.30

Table 2 presents similar parameters for detectors based on pure *p*-terphenyl. It should be noted that for the samples with the lowest L_{av}-value of 0.2 mm, the values of $FWHM_{\alpha}$, $D_{\alpha\gamma}$ и $FOM_{\alpha\gamma}$ are not provided due to the extremely low efficiency of alpha particle detection

from the smallest grain size for a detector with a thickness of 4 mm.

Table 2

Values of $FWHM_\gamma$, $FWHM_\alpha$, $D_{\alpha\gamma}$, and $FOM_{\alpha\gamma}$ for pure *p*-terphenyl composite detectors and the reference single crystal

	$FWHM_\gamma$, ch	$FWHM_\alpha$, ch	$D_{\alpha\gamma}$, ch	$FOM_{\alpha\gamma}$
Single crystal	28	43	85	1.35
Composite detectors				
L_{av}, mm				
0.2	42	–	–	–
0.4	45	68	97	0.86
0.75	48	73	91	0.75
1.1	44	69	93	0.82
1.3	39	69	93	0.86
1.6	42	65	94	0.88
1.9	41	63	104	1.00

Fig. 4 shows the $FOM_{\alpha\gamma}$-values for stilbene and *p*-terphenyl composite scintillators as the function of the average grain size $L_{av.}$ The lines on the figure indicate the $FOM_{\alpha\gamma}$-values for the corresponding reference single crystals.

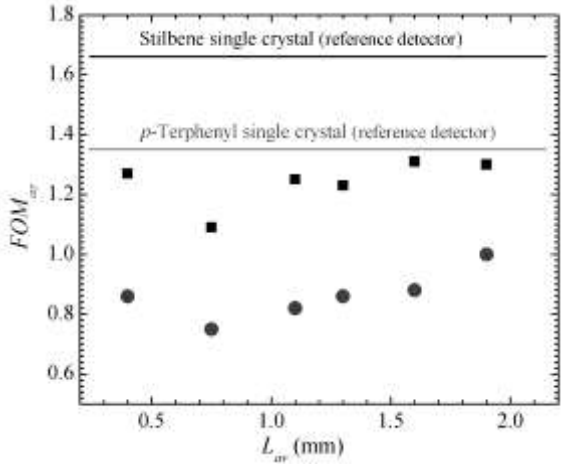


Fig. 4. Values of $FOM_{\alpha\gamma}$ for stilbene composite scintillators (black squares) and p-terphenyl composite scintillators (dark gray circles) as the function of the average grain size $L_{av.}$ The lines on the figure indicate the $FOM_{\alpha\gamma}$-values for the reference single crystals

The analysis of the results presented in Figs. 1 and 2 and in Tables 1 and 2 allows the following conclusions regarding the alpha-gamma discrimination capability ($FOM_{\alpha\gamma}$-values) of organic composite scintillators. For the stilbene composite scintillators, the $FOM_{\alpha\gamma}$-values ranged from 1.09 to 1.31, while for the *p*-terphenyl scintillators, they ranged from 0.75 to 1.0. The highest $FOM_{\alpha\gamma}$-values were obtained for samples with the largest average grain sizes (L_{av}), specifically for L_{av} = 1.6 and 1.9 mm. For the corresponding reference single crystals, the $FOM_{\alpha\gamma}$-values were higher compared to the composite materials, namely 1.66 for stilbene and 1.35 for *p*-terphenyl.

The selection of optimal grain sizes for the creation of composite scintillators depends on the specific practical tasks for which the detector will be used. For example, for tasks in radioecology and the detection of short-range particles, such as alpha particles, an increase in relative light output is necessary. This can be achieved by creating single-layer composite scintillators with grain sizes smaller than 60 μm, where the most significant effect will be the reflection of triplet excitons from the grain surfaces followed by their recombination, as demonstrated by us recently [14].

For the creation of systems, such as those for the selective detection of alpha and gamma radiations, grains larger than 1.4 mm are optimal, as shown by the results presented in this work. This is due to the need for the development of bulk multilayer composite scintillators for more efficient gamma radiation detection. On the other hand, a drawback is that the large number of scintillation layers leads to light scattering at the grain boundaries, which, as a result, reduces the number of photons detected by the photodetector.

CONCLUSIONS

The alpha-gamma discrimination capability ($FOM_{\alpha\gamma}$) of organic composite scintillators based on stilbene and *p*-terphenyl was studied. The results we obtained are the following:

1. The $FOM_{\alpha\gamma}$-values ranged from 1.09 to 1.31 for the stilbene composite scintillators, while for the *p*-terphenyl scintillators, they ranged from 0.75 to 1.0. Thus, the discrimination capability of stilbene composite scintillators is significantly higher than that of *p*-terphenyl scintillators.
2. The highest $FOM_{\alpha\gamma}$-values were obtained for samples with the largest grains, specifically for L_{av} = 1.6 and 1.9 mm, both for stilbene composite scintillators, and *p*-terphenyl ones.
3. For the reference single crystals, the $FOM_{\alpha\gamma}$-values were higher compared to the composite materials, namely 1.66 for stilbene and 1.35 for *p*-terphenyl.

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ОРГАНІЧНІ КОМПОЗИЦІЙНІ ДЕТЕКТОРИ ДЛЯ РОЗДІЛЬНОЇ РЕЄСТРАЦІЇ АЛЬФА- ТА ГАММА-ВИПРОМІНЮВАНЬ

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Проведено дослідження альфа-гамма роздільної здатності ($FOM_{\alpha\gamma}$) органічних композиційних сцинтиляторів на основі стильбену та *p*-терфенілу. Досліджувалися 2 серії зразків композиційних детекторів із різними фракціями кристалічних гранул у діапазоні середнього значення гранул від 0,2 до 1,9 мм товщиною 4 мм та діаметром 15 мм. Було розроблено вимірювальну апаратуру з використанням методики «перетину нуля». Показано, що для композиційних зразків як на основі стильбену, так й *p*-терфенілу $FOM_{\alpha\gamma}$ мають достатньо високі значення, як порівняти з еталонними зразками відповідних монокристалів. Спостерігалася тенденція щодо невеликого збільшення значення $FOM_{\alpha\gamma}$ для зразків із найбільшими значеннями L_{cep}.