THE INFLUENCE OF ACTIVATOR CONCENTRATION ON PULSE SHAPE DISCRIMINATION ABILITY OF *p*-TERPHENYL SINGLE CRYSTALS DOPED WITH 1,4-DIPHENYL-1,3-BUTADIENE

A.V. Krech, I.V. Lazarev, S.V. Makhota, E.V. Martynenko, O.A. Tarasenko
Institute for Scintillation Materials, NAS of Ukraine, Kharkiv, Ukraine

The influence of dopant concentration on pulse shape discrimination ability of the *p*-terphenyl single crystals doped with 1,4-diphenyl-1,3-butadiene was investigated. The figure of merit (FOM) values were obtained under mixed alpha+gamma radiation for *p*-terphenyl single crystals containing different concentrations of 1,4-diphenyl-1,3-butadiene. Charge-time transformation method was used. The content of 1,4-diphenyl-1,3-butadiene in *p*-terphenyl crystals varied from 0 (pure *p*-terphenyl) to approximately 0.8 mass%. Obtained results has shown, that with increase of introduced 1,4-diphenyl-1,3-butadiene concentration the pulse shape discrimination ability of *p*-terphenyl single crystals is getting worse in general.

PACS: 32.50.+d; 78.60.−b; 87.53

INTRODUCTION

p-Terphenyl is a widely known organic scintillation material which is successfully used in many tasks due to short decay time, high efficiency of detection of short-range radiations and also ability to selective detection of ionizing radiations by a pulse shape. The pulse shape discrimination ability of ionizing radiations of different types is especially important feature for scintillators because the effect on medium differs significantly for radiations of different types and natural isotopes are mostly sources of mixed radiation fields [1].

A scintillation pulse of an organic scintillator consists of the fast and the slow components. Pulses caused by the ionizing radiations with high specific energy losses *dE/dx* (alpha-particles, recoil protons, etc.) have more intensive slow component than those caused by ionizing radiations with low *dE/dx* (for instance, gamma-radiation, conversion electrons). Thus, it is possible to identify the type of radiation that caused the scintillation pulse by intensity of the slow component and its relative contribution to the total scintillation pulse.

The fast component is caused by direct transition of singlet-excited states to the ground state, that is $S_1 \rightarrow S_0$ transition. The slow component is formed through the singlet channel of the intermediate process of triplet-triplet annihilation [1]:

$$T_1 + T_1 \rightarrow \begin{cases} S_1 + S_0 \rightarrow h\nu + 2S_0 \\ T_1 + S_0 \\ Q + S_0 \\ \ldots \end{cases} \qquad (1)$$

An efficiency of the processes of transport and recombination of triplet states determines the intensity of the slow component of a scintillation pulse and, as a consequence, the selective ability of a scintillator.

It is known, that doping of single crystal of *p*-terphenyl with 1,4-diphenyl-1,3-butadiene results in the growth of light output values, that is increasing the number of photons generated by the same type of excitation [2–4].

Increasing of light output values for doped single crystals of *p*-terphenyl (PTP) when doped with 1,4-diphenyl-1,3-butadiene (DPB) caused by the effective transfer of electronic excitation energy from the host molecules (PTP) to the molecules of dopant (DPB). For this reason, the doped crystals of PTP are more commercially demanded.

On the other hand, adding the dopant an amount that exceeds certain concentration leads to decreasing of the light output due to the concentration quenching, to loss in transparency of a scintillator and also it can significantly reduce its pulse shape discrimination ability [4–6].

The influence of DPB concentration on the light output values for scintillators of PTP doped with DFB was studied in [1, 4, 6]. At this work we investigated the influence of DPB concentration on the selective ability of PTP single crystals doped with it.

EXPERIMENTAL SAMPLES AND METHODS

The single crystal sample of pure PTP and 3 series of single crystal samples of PTP doped with DFB, containing different concentrations of DFB, were investigated. Samples that belong to the same series were obtained by sawing of the single crystal boule of PTP doped with DFB that was grown from the melt with given concentration of introduced DFB. DFB was added to the melt in the following initial concentrations: 0.05, 0.1, and 0.3 mass%. Each series was consisted of 6 single crystal samples 2 mm thick, which were numbered from 1 to 6. Number 1 corresponds to the sample from the beginning part of the single crystal boule and number 6 – to its end part according to crystal growth direction. One can find more details about sample preparation in our previous work [6].

During the growth process of the PTP single crystal doped with DFB from a melt, the dopant is rejected by the freezing solid and is accumulated in the melt. Thus, DFB concentration in different parts of the grown single crystal boule differs, as we had showed in [6], it increases along crystal growth direction. That is, for every series of investigated samples the concentration of contained DFB increases with sample number from 1 to 6. DFB content also increases for all samples of the series with increase of the initial amount of DFB added to the melt (Table 1). The method of gas chromatography mass spectrometry was used to determine DFB content in the samples of PTP single crystals [6]. In this work the pulse shape discrimination ability of the above-mentioned single crystal scintillators was determined for the case of mixed irradiation with alpha-particles and photons of gamma radiation using charge-time transformation method. This method allows to separate pulses caused by photons of gamma radiation and alpha-particles using differences in shapes of scintillation pulses generated by these types of radiation. This method is described in detail in [1, 7, 8]. Data were analyzed autonomously by charge comparison method (CCM) [9, 10]. Pu^{239} (alpha-particles + gamma radiation) and Cs^{137} (gamma radiation) sources were used for excitation of the samples. Cs^{137} source was in the metal container to avoid conversion electrons hit a scintillator.

Table 1

DFB content in the samples of PTP single crystals for different initial concentrations of DFB added in melt [6]

Sample number	PTF+0.05 mass% DFB	PTF+0.1 mass% DFB	PTF+0.3 mass% DFB
1	0.0094	0.0169	0.0544
2	0.0087	0.0184	0.0685
3	0.0107	0.0178	0.0859
4	0.0161	0.0332	0.1581
5	0.0219	0.0517	0.2608
6	0.0764	0.232	0.7979

According to the above-mentioned, the scintillation pulses caused by alpha-particles have more intensive slow component than those caused by gamma-quanta. So, it is possible to identify the type of radiation that generated scintillation pulse by the relative contribution of the "tail" part of the pulse into the total pulse.

RESULTS

Figure demonstrates, as an instance, spectra of relation of "tail" part area of the pulse to the total area of the pulse (tail-to-total relation) for pure PTP, sample 1 of series PTP+0,1 mass% DFB, sample 5 of series PTP+0,3 mass% DFB. They are typical spectra obtained with charge comparison method. The peak on the left corresponds to events caused by gamma-radiation, for pulses generated by it have lower contribution of the slow component. The peak on the right corresponds to events caused by alpha-particles. Curves similar to those presented at Figure were obtained for all investigated samples.

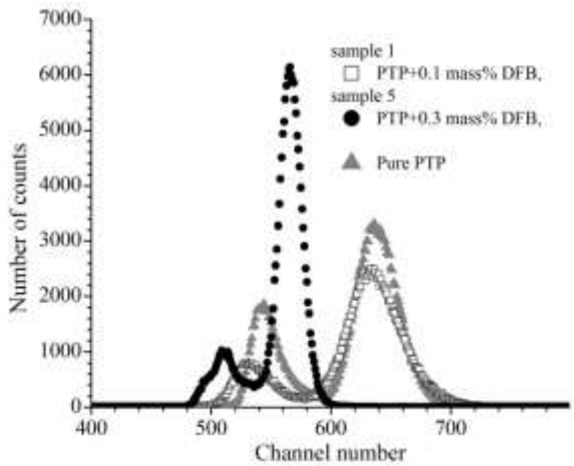
The pulse shape discrimination ability of the scintillator was characterized using the FOM-value that is determined by:

$$FOM = \frac{\Delta A}{H_1 + H_2}, \quad (2)$$

where ΔA – is difference of maximums of alpha- and gamma peaks, H_1 and H_2 – are full widths at half maximum of alpha- and gamma peaks, respectively.

Using obtained spectra, the FOM values were calculated for all samples by formula (2). The results of these calculations are presented in the Table 2.

As one can see from the data presented in Tables 1and 2, for the majority of samples (with the exception of 3 of them) FOM values for doped PTP single crystals were smaller than for pure one. Despite the fact that a clear dependency of FOM-values versus DFB content cannot be distinguished, the general tendency to reducing of FOM-value (that is to worsening of selective ability of PTP crystals) with DFB concentration growth is observed. For sample 6 of series PTP+0,3 mass% DFB, which has the maximal content of DFB among all samples under investigation, it was unable to obtain FOM value because peaks of spectrum of relation of "tail" part area of the pulse to the total area of the pulse were not resolved.



Spectra obtained with charge comparison method for the sample of pure PTP, sample 1 of series PTP+0.1 mass% DFB, sample 5 of series PTP+0.3 mass% DFB

Table 2

The FOM-values for the single crystal of pure PTP and the single crystals of PTP grown from melt with adding 0.05, 0.1, and 0.3 mass% of DFB

Sample number	FOM			
	Pure PTP	PTP+0.05mass% DFB	PTP+0.1mass% DFB	PTP+0.3mass% DFB
1		1.22	1.17	1.15
2		1.05	0.88	0.92
3		1.44	1.09	1.18
4		1.45	1.20	1.13
5		1.13	1.32	1.16
6		1.33	1.21	–
	1.33			

Such results previously can be explained by following considerations. As it was mentioned earlier, the greater slow component contribution to total scintillation pulse, the higher the selective ability of scintillator (FOM-value). The slow component forms in process of triplet-triplet annihilation, which, in its turn, is regulated by the processes of transport and diffusion of triplet-excited states. Also, the molecules of dopant (DPB) are close in size to the host molecules (PTP), but are not identical to them. So, it can be supposed, that DFB molecules create inhomogeneities in PTP lattice which are able to influence on transport triplet-excited states and even disturb it at sufficiently high concentrations. It requires carrying out feather studies with higher concentrations of DFB, which was initially added into the melt, to verify this suggestion.

CONCLUSIONS

The FOM-values characterizing the pulse shape discrimination ability of a scintillator were obtained for PTP single crystals, containing different amount of DFB, under mixed alpha+gamma radiation. The content of DFB in PTP crystals varied from 0 (pure PTP) to approximately 0.8 mass.%.

Obtained results show, that with increase of introduced DFB concentration the pulse shape discrimination ability of PTP single crystals is getting worse in general, but do not allow to distinguish a clear dependency. Presumably, such results can be explained by suggestion that DFB molecules create inhomogeneities in PTP lattice which are able to influence on transport and efficiency of recombination of triplet-excited states. However, to proof such suggestion it is necessary to carry out additional investigations with samples, obtained with higher initial concentrations of dopant in the melt.

REFERENCES

1. Н.З. Галунов, В.П. Семиноженко. Теория и применение радиолюминесценции органических конденсированных сред. Наукова думка, Киев, 1997, 280 с.

2. N.Z. Galunov, V.P. Seminozhenko, A.M. Stepanenko. Electronic processes and radioluminescence for crystalline systems with different structure perfection // *Mol. Cryst. Liq. Cryst.* 2001, v. 361, p. 287-292, https://doi.org/10.1080/10587250108025752З.

3. A.M. Stepanenko, O.A. Tarasenko, N.Z. Galunov. Nature of the additional delay of prompt radioluminescence photon emission in organic solids and its description by Gauss function // *Mol. Cryst. Liq. Cryst.* 2001, v. 361, p. 299-304, https://doi.org/10.1080/10587250108025754.

4. S.V. Budakovsky, N.Z. Galunov, A.Y. Rybalko et al. Organic luminescent polycrystals as novel materials for detecting ionizing radiations // *Mol. Cryst. Liq. Cryst.* 2002, v. 385, p. 71-77. https://doi.org/10.1080/713738800.

5. N. Zaitseva, A. Glenn, L. Carman et al. Pulse shape discrimination in impure and mixed single-crystal organic scintillators // *IEEE Trans. Nucl. Sci*. 2011, v.58, p. 3411-3420. https://doi.org/10.1109/TNS.2011.2171363.

6. N.Z. Galunov, I.V. Lazarev, E.V. Martynenko, et al. Distribution coefficient of 1,4-diphenyl-1,3-butadiene in *p*-terphenyl single crystal and its influence on scintillation crystal light output // *Mol. Cryst. Liq. Cryst.* 2015, v. 616, p. 176-186. https://doi.org/10.1080/15421406.2014.991105.

7. В.И. Кухтевич, Л.А. Трыков, О.А. Трыков. *Однокристальный сцинтилляционный спектрометр (с органическим фосфором)*. М., Атомиздат, 1971, 136 с.

8. R. Aryaeinejad, J.K. Hartwell, D.F. Spencer. Comparison between digital and analog pulse shape discrimination techniques for neutron and gamma ray separation // *2005 IEEE Nuclear Science Symposium Conference Record.* 2005, p. 500-505. https://doi.org/10.1109/NSSMIC.2005.1596302.

9. B. Sabbah, A. Suhami. An accurate pulse-shape discriminator for a wide range of energies // *Nucl. Instrum. Meth.* 1968, v. 58, p. 102-110. https://doi.org/10.1016/0029-554X(68)90035-9.

10. F.D. Brooks. Development of organic scintillators // *Nucl. Instrum. Meth.* 1979, v.162, p. 477-505. https://doi.org/10.1016/0029-554X(79)90729-8.