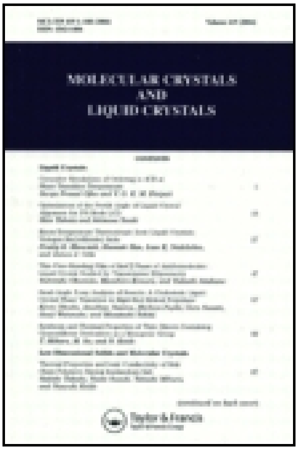

Molecular Crystals and Liquid Crystals

Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl20

Organic luminescent polycrystalsas novel materials for detectingionizing radiations

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Version of record first published: 18 Oct 2010

To cite this article: Sergey V. Budakovsky, Nikolai Z. Galunov, Andrey Yu. Rybalkooleg, A. Tarasenko & Vladimir V. Yarychkin (2002): Organic luminescent polycrystalsas novel materials for detectingionizing radiations, Molecular Crystals and Liquid Crystals, 385:1, 71-77

To link to this article: http://dx.doi.org/10.1080/713738800

Mol. Cryst. Liq. Cryst., Vol. 385, pp. [191]/71–[197]/77
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1058-725X/02 $12.00 + .00
DOI: 10.1080/10587250290113088

ORGANIC LUMINESCENT POLYCRYSTALS AS NOVEL MATERIALS FOR DETECTING IONIZING RADIATIONS

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Organic molecular polycrystals obtained by the pressing technique of our design and their scintillation characteristics are discussed. The analysis of the dependence between structural features and radioluminescent characteristics of polycrystalline systems is based on the results of investigation of their light yield and pulse shape.

Keywords: organic polycrystals; radioluminescence; ionizing radiation

INTRODUCTION

In contrast with organic single crystals, plastic and liquid scintillators organic polycrystals are novel progressive type of scintillation materials. Scintillators with large diameters are necessary for radioecological and medicinal investigations, in which short-range charged particles of low intensity and ultraviolet radiations are detected. Technologies of organic polycrystals creation are in progress now. Therefore the analysis of their radioluminescent properties is very important both for understanding the dependence between structural features and radioluminescent characteristics of polycrystalline systems and for designing and creating new effective detectors. Besides that, the organic polycrystals are effective material for light shifters included in complicate systems for light conversion from ultraviolet to red wavelength range.

In our previous papers [1–2] it has been shown that in contrast to the photoluminescence process features of generation, transport and recombination of dynamic charge-states [3] of the polarization origin cause the radioluminescence process. For description of a scintillation pulse shape of

The Science and Technology Center in Ukraine supported this work by grant No. 1733.

organic systems it is necessary to take into account duration of the processes preceding an excitation of the lowest singlet excited states. For organic solids the additional delay of the moments of photon emission is observed. Gaussian form of function describing this delay is due to a plurality of acts of generation and recombination of charge-states, i.e. the processes result in excitation of organic solids molecules [4]. In this paper we examine the radioluminescence of organic polycrystals.

EXPERIMENTAL

Organic polycrystals were obtained by the pressing technique from a fine-grained powder under pressures of $(1\ldots8)10^2$ MPa at the room temperature [5]. The powder was obtained by cryogenic grinding of the melt-grown single crystals, and then single-axial pressing in a nodeformed mould pressed it. The recrystallization process took place during an annealing of a polycrystal at the melting temperature and hydrostatic compression of $10\ldots20$ MPa. It allowed obtaining the samples of polycrystals with good transparency and thickness up to 3 mm. Using this technology we obtained *p*-terphenyl polycrystals doped with diphenylbutadiene molecules, stilbene polycrystals. Scintillation properties of some scintillators and detectors on theirs base are discussed below.

For the method of light yield measurements [6] we used radionuclide sources ^{241}Am (α-particles with the energy $E_\alpha = 5.4$ MeV) and ^{137}Cs (conversion electrons with the energy $E_\beta = 0.6225$ MeV). To registrate the scintillation pulses we used two types of photoelectronic multipliers: 9208 (Electron Tubes Ltd, England) and FEU-173-1 (Russia). To estimate the results obtained for organic polycrystals we compared these results with analogous ones for organic single crystals. We have used the single-photon technique to investigate a radioluminescence pulse shape. The measurements were carried out on the set-up with subnanosecond resolution. The main principles of the single-photon counting technique application for precise studying of radioluminescence pulse kinetics as well as possibilities to improve a time resolution of the set-up have been considered in the previous papers (see [7], for example).

RESULTS AND DISCUSSION

Table I shows the values of light yield, J and resolution, R for organic detectors on a base of *p*-terphenyl organic single crystal and *p*-terphenyl polycrystal which contain 0.1% of diphenylbutadiene molecules as luminescent addition agents, as well as for detector on a base of stilbene

TABLE I Scintillation Characteristics of the Organic Detectors Studied

	Detector sizes, mm		α-particles (^{241}Am)		Conversion electrons (^{137}Cs)	
Scintillation base	Diameter	Height	J_α, Rel. units	R_α, %	J_β, Rel. units	R_β, %
p-terphenyl single crystal	40	14	1.00	14.8	1.00	9.5
p-terphenyl polycrystal	40	3	1.00	18.3	0.96	11.2
Stilbene polycrystal	40	3	0.33	30.1	0.54	22.7

polycrystal. Measurements were carried both for the case of α-excitation (J_α- and R_α-values) and for excitation by conversion electrons (J_β- and R_β-values). The dimensions of detectors are presented too. We used the detector on a base *p*-terphenyl organic single crystal as the reference one. For this detector the value J_β is equal to 2.45 photons/MeV. To compare the experimental data its values J_α and J_β were taken as a unit.

According to Table I the values J_α and J_β are very close for the detector on a base of *p*-terphenyl single crystal and for the detector on a base of *p*-terphenyl polycrystal. An increase of a transparency of stilbene scintillators to their own luminescence as the result of the recrystallization process can cause to an improvement of their scintillation parameters.

The first study of prompt radioluminescence of stilbene organic polycrystals, which is discussed here, we were carried out using the single-photon technique. Figure 1 demonstrates the radioluminescence kinetics curve for organic polycrystal of stilbene irradiated by a source of β-particles (^{90}Sr + ^{90}Y). The form of radioluminescence pulse fast component is described as,

$$i(t) = f_G(t) * \exp(-t/\tau) \equiv \int_0^t f_G(t-x) \cdot \exp(-x/\tau)dx. \tag{1}$$

In Eq. (1)

$$f_G(t) = \{const/(\sqrt{2\pi}\sigma)\} \cdot \{\exp[-(t-\Delta)^2/2\sigma^2]\}, \tag{2}$$

where σ and τ are time constants, $\Delta \approx 3\sigma$, t is the time after excitation. Function $f_G(t)$ describes the additional delay of the moments of photon emission. Evaluated values for decay time constant τ for stilbene polycrystal -3.3 ns and for time constant $\sigma(2) - 0.4$ ns.

We have studied scintillation characteristics of thin (1 mm) *p*-terphenyl organic polycrystals containing diphenylbutadiene molecules as the guest ones. To investigate an influence of a structure we varied the concentration

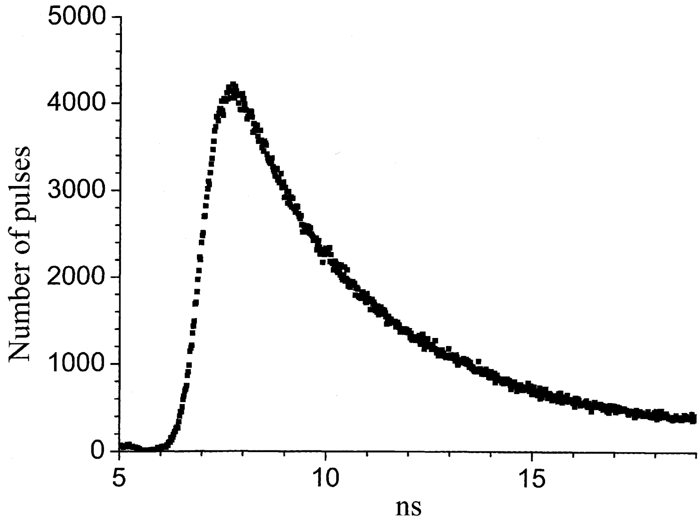

FIGURE 1 The radioluminescence kinetics curve for stilbene polycrystal (diameter –40 mm, height –3 mm) irradiated by a source of β-particles (^{90}Sr + ^{90}Y). Evaluated values for decay time constant τ – 3.3 ns, time constant σ – 0.4 ns.

C of diphenylbutadiene molecules in samples from 0 (pure polycrystal) to 0.5%. Table II shows selected concentrations C of the guest molecules. It should be noted that we have chosen the grains with the mean size of 0.5–1 mm as the material for pressing these polycrystals. For the measurements we used thin (1 mm) *p*-terphenyl organic single crystal with the concentration of diphenylbutadiene molecules C = 0.1% as the reference scintillator. Its light yield J was taken as a unit (J = 1).

We carried out the measurements of amplitude spectra selected *p*-terphenyl organic polycrystals both for α-excitation (isotope ^{241}Am) and for excitation by conversion electrons (isotope ^{137}Cs). Figure 2 illustrates as an example the amplitude spectra for the reference scintillator (curve 1) and for organic *p*-terphenyl polycrystal #3 (see Table II) (curve 2) for

TABLE II The Values C, J_α, J_β and $\tau(1)$ for Thin *p*-Terphenyl Organic Polycrystals

Sample number	1	2	3	4	5	6	7	8	9
C, %	0	0.014	0.07	0.085	0.1	0.2	0.3	0.4	0.5
J_α, Rel. units	0.68	0.74	0.62	0.80	0.94	0.80	0.92	0.74	0.59
J_β, Rel. units	1.14	1.15	1.14	1.20	1.28	1.21	1.24	1.20	1.09
τ	2.10	1.60	1.82	1.84	2.15	2.42	2.51	2.09	2.01

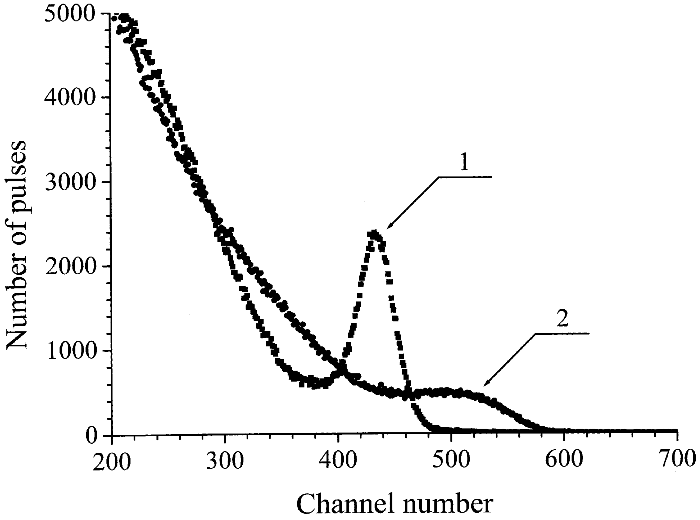

FIGURE 2 Amplitude spectra for the reference scintillator (curve 1) and for organic *p*-terphenyl polycrystal #3 (curve 2) for the case of excitation by conversion electrons (an isotope ^{137}Cs). PMT type: 9208 (Electron Tubes Ltd, England).

the case of excitation by conversion electrons. Using the results of the measurements we have analysed an influence of a diphenylbutadiene concentration C on the values of relative light yield. Table II presents the values of relative light yield J_α and J_β for the cases of α-excitation and excitation by conversion electrons, accordingly. The values J_α and J_β for the reference scintillator were taken as a unit. The highest values of a relative light yield correspond to the range of diphenylbutadiene concentration 0.1…0.3%. At low concentrations (<0.1%) of the guest molecules inefficient electron excitation energy transfer from the host molecules (*p*-terphenyl) to the guest ones (diphenylbutadiene) takes place, and for high concentrations (>0.3%) of the guest molecules a concentration quenching of a luminescence is observed. The influence of a transparency of scintillators on results of measurements explains lower relative light yield J for α-excitation, than for β-excitation ($J_\alpha < J_\beta$), i.e. in contrast to the case of the case β-excitation when the luminescent centres are located in a bulk of the scintillator, in the case of α-excitation the scintillation signal is formed in a very little volume near the surface. In the latter case all scintillation photons have to travel through the scintillator. It has to result in decrease of light intensity of scintillation pulses.

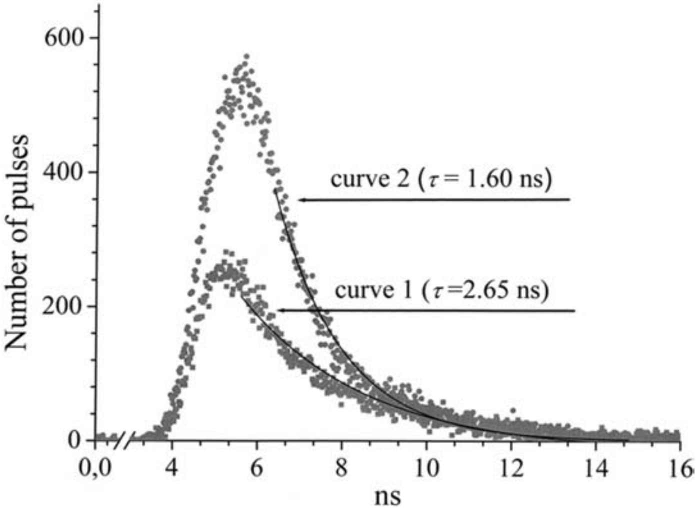

FIGURE 3 The radioluminescence kinetics curves and their decay exponential approximations for the reference scintillator (curve 1) and for thin *p*-terphenyl polycrystal #2 (curve 2) irradiated by a source of β-particles (^{90}Sr + ^{90}Y).

We have studied some aspects of a fast radioluminescence pulse shape formation in thin organic *p*-terphenyl polycrystals. Figure 3 shows the radioluminescence kinetics curves for the reference crystal (curve 1) and for thin *p*-terphenyl polycrystal #2 (see Table II). For all thin organic polycrystals the additional delay of the moments of photon emission was observed and its radioluminescence pulse shape was described best of all in a form (1) with $f_G(t)$ (2). Table II shows the τ-values for polycrystals, which are obtained by experimental data processing. The value τ for the reference scintillator is equal to 2.65 ns.

The dependence between decay time constant τ and the concentration C of the guest molecules in *p*-terphenyl polycrystals correlates with experimental data of light yield measurements (see Table II), Nevertheless, for all polycrystals containing diphenylbutadiene molecules the value of decay time constant τ is smaller than one for the reference organic *p*-terphenyl single crystal with 0.1% of diphenylbutadiene molecules. A small reduction of decay time constant τ for polycrystals in comparison with single crystals can be explained by an increase of number of deep traps for charge carriers and excitons, which have arisen on boundaries of mosaic blocks of polycrystals. More detail analysis of an influence of a structure on parameters of a fast radioluminescence, including studying of aspects of the rising part of a scintillation pulse, will be considered further.

CONCLUSION

We have studied scintillation characteristics of new class of objects – the organic polycrystals obtained by the pressing technique. The fact that scintillation parameters of polycrystals are close to single crystals ones testifies that our technology allows creating samples of polycrystals with enough big size of blocks, i.e. with small concentration of grain boundaries. The difference between structure perfection of organic single crystals and polycrystals obtained according to our technology is not high enough to result in essentially profound contrast of their scintillation characteristics.

The results of the research reported testify to an availability of an application of these systems in modern scintillation engineering, ecology and medicine.

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