

Features of charge pairs recombination in the track regions of organic solid scintillators

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Some features of the recombination of charge states generated in organic solid scintillators for different densities of radiation excitation, as well as the effect of the polarization mechanisms on this process are studied. The estimations are made for single crystals of stilbene and *p*-terphenyl, and a plastic scintillator. The influence of anisotropy of the dielectric characteristics on the processes under investigation is considered for an anthracene single crystal. The estimated values of the average distance d between the centers of charge pairs (M_+ , M_-), generated in a charged particle track, are compared with the value of the radius of stable polarization environment r_c , created by an excess charge carrier in an organic condensed medium. It is shown that an influence of the polarization on the recombination mechanism of the charge state pairs, which is produced by the ionizing radiations with different specific energy loss dE/dx , grows for high local density in the track volume. The estimations that we obtained are in a good agreement with the results of the light yield measurements of organic solid scintillators.

Рассмотрены особенности рекомбинации зарядовых состояний, генерируемых в органическом твердотельном сцинтилляторе, при различных плотностях радиационного возбуждения, а также влияние поляризационных механизмов на этот процесс. Оценки проведены для монокристаллов стибена, *n*-терфенила, пластмассового сцинтиллятора. Влияние анизотропии диэлектрических характеристик на изучаемые процессы рассмотрено для монокристалла антрацена. Проведены оценки среднего расстояния d между центрами зарядовых пар (M_+ , M_-), генерируемых в треке заряженной частицы, и сравнения значения d с расчетным значением радиуса устойчивого поляризационного окружения r_c , создаваемого избыточным носителем заряда в органической конденсированной среде. Показано, что степень влияния поляризации на специфику рекомбинации пар зарядовых состояний, создаваемых ионизирующими излучениями с разными удельными энергетическими потерями dE/dx , определяется их локальной плотностью в объеме трека. Вышеприведенные оценки находятся в хорошем соответствии с результатами исследований светового выхода органических твердотельных сцинтилляторов.

1. Introduction

Previously [1], we examined the aspects of generation and energy exchange of charge states which arise as the result of interaction of an ionizing radiation with an organic molecular system. The estimations were carried out for the stilbene single crystals excited by the ionizing radiations with low (gamma photons) and high (alpha particles) specific energy loss dE/dx . The analysis was based on a comparison of the aver-

age distance d between charge pairs generated in the ionizing particle track and the radius of stable polarization environment r_c . Strong polarization interactions increase the speed of the charge state recombination when $d \leq r_c$. In this paper we extended the range of objects. We have considered the single crystals of anthracene, stilbene and *p*-terphenyl, the polystyrene plastic scintillator. Additional analysis for the anthracene crystal allowed us to take into account

the anisotropy of the dielectric and optical properties and to estimate its contribution to the mechanism of energy exchange of charge states generated in an organic solid under ionizing radiations of various types.

We carried out estimations for electrons with the energy $E_e = 0.622$ MeV ($dE/dx \approx 10^{-1}$ MeV/cm), for protons in the range of E_p from 1 to 10 MeV ($dE/dx \approx 10^1-10^2$ MeV/cm), for alpha particles in the range of E_α from 0.5 to 10 MeV ($dE/dx \approx 10^3$ MeV/cm) [2]. Such a choice of types and energies of ionizing radiations is not accidental. It is well known [2, 3], that organic scintillators are effective detectors both short-range ionizing radiations and fast neutrons (the edge of the recoil proton spectrum is analyzed [2]). In other words, organic scintillators are mainly used for detection those types of ionizing radiations, for which the processes of generation and energy exchange of charge states in the particle track have a determinate effect on the radioluminescence energy yield [2, 4]. In addition, such a range of dE/dx of an ionizing particle gives a possibility to compare the data of estimates with the results of light yield measurements of solid organic scintillators for practicable laboratory sources of ionizing radiations [2].

2. Theory

2.1. Polarization and recombination of charge states

Suppose that as the result of ionization of a molecule an electron has thermalize at a distance r_0 from it. In such a case the probability that the electron leaves his geminate molecular ion and its polarization surrounding, and as the result of this the recombination of this geminate pair does not occur, is proportional to $\exp(-r_0/r_c)$, where the critical Coulomb radius r_c [2, 5]:

$$r_c = \frac{e^2}{4\pi\epsilon\epsilon_0 kT}. \quad (1)$$

In (1) $\hat{\epsilon}$ is the average relative permittivity of a material, k is a Boltzmann constant, T is a temperature, ϵ_0 is a dielectric constant. The value of r_c (1) is determined by equality of the energy of thermal motion in a medium and the energy of the Coulomb field which holds the charges [5, 6]. This parameter determines the radius of the stable polarization environment for the excess charge. In a molecular matrix a quasi-free electron rapidly localizes on any molecule,

forming a negative quasi-ion M_- . A molecule that has lost an electron becomes a positive quasi-ion M_+ . During the time $\tau_e \approx 10^{-16}-10^{-15}$ s the process of electronic polarization of the molecular π -orbitals of the neighboring unionized molecules by excess charge carriers takes place. According to [7], about 7000 molecules around the molecular quasi-ion are polarized, that is are involved in its polarization environment. Polarons M_p^+ and M_p^- are formed and separated by some average distance from each other. If this distance is less than the radius of the stable polarization environment r_c (1), then a molecular-polaron pair, or a bipolaron (M_p^+, M_p^-) is created [2, 5].

Polarons can occur around quasi-ions M_+ and M_- , which locate on small (less than r_c) distances from each other. The charge-transfer (CT) state is forming [2, 5]. At further approach of the bound states with high vibrational energy the excess vibrational energy is lost, and the total energy of the CT-state becomes smaller than the width of the energy gap. It prevents a dissociation of the pair. Then, the CT-state decays to a vibrationally excited Frenkel exciton and a molecule in a vibrationally excited ground state. After that, these states lose their vibrational energy and transfer to the ground state. The bound CT-state can also use the excess energy, resulting in the nuclear relaxation, for further separation. Further separation of charges can takes place when the CT-state relaxes to the CP-state (the state of a charge pair [2, 5]).

Thus, the recombination of polaron pairs should occur through the formation of the bipolaron, which is the CT-, or the CP-state. It follows thence, in particular, that the generation of charge states by an ionizing particle should be accompanied by the appearance of the polarization interactions between the excess charge carrier localized on the molecule and its environment [2]. According to [6], the probability of separation of a thermalized electron-hole pair in an isotropic medium with $\epsilon = 3.02$ weakly depends on the direction of the external field, that does not exceed 10^3 V·cm⁻¹. With field strength increasing up to 10^5 V·cm⁻¹ the probabilities of thermalized electron-hole pair separation along the field and in the direction opposite to the applied field, are approximately as 1 to 6. For very strong fields ($E \approx 10^6$ V/cm), reaching the values of the local field strength inside the polaron pair [5], the charge carrier scattering time

Table 1. The relative permittivity ϵ of solid organic scintillators and the calculated value of the radius of stable polarization environment of an excess charge r_c for these systems

Material	Formula	T, K	Frequency, Hz	ϵ	Notes	r_c , nm
Pyrene	C ₁₆ H ₁₀	290	8·10 ⁵	3.21	[10]	17.7
Phenanthrene	C ₁₄ H ₁₀	290	8·10 ⁵	3.0		18.59
Cyclohexane	C ₆ H ₁₂	168	1·10 ⁵	2.52		22.13
Polystyrene	$[-\text{CH}_2-\text{C}(\text{C}_6\text{H}_5)\text{H}-]_n$	–	–	2.6	[11]	21.45
Anthracene	C ₁₄ H ₁₀	290	8·10 ⁵	3.46	[10]	16.11
		–	5.5·10 ¹⁴	3.55	[7] ($n_g = 2.22$; $n_m = 1.816$; $n_p = 1.557$)	15.71
		–	5.1·10 ¹⁴	3.24	[7] ($n_g = 2.04 \pm 0.08$; $n_m = 1.78 \pm 0.01$; $n_p = 1.55 \pm 0.01$)	17.21
		–	–	3.2	[7]	17.43
		–	–	3.23	[7]	17.26
Naphthalene	C ₁₀ H ₈	300	1·10 ³	2.85	[10]	19.57
		–	5.5·10 ¹⁴	3.02	[7] ($n_g = 1.945$; $n_m = 1.722$; $n_p = 1.525$)	18.46
<i>p</i> -Terphenyl	C ₁₈ H ₁₄	300	7.5·10 ¹⁴	2.86	[12] ($n_a = 1.655$; $n_b = 1.727$)	19.50
		–	–	3.2	[13] ($\epsilon_r(\infty)$)	17.43
trans-Stilbene	C ₁₄ H ₁₂	300	7.5·10 ¹⁴	3.62	[12] ($n_a = 2.018$; $n_b = 1.778$)	15.40

τ is a very small value, estimated as 10^{-14} s [8]. This value is comparable with the time τ of molecular polaron formation [7].

The above will show that the ionizing radiation exchange energy in organic molecular systems proceeds through the stage of rapid ($\leq 10^{-13}$ s) primary recombination of charge pairs accelerated by the polarization interactions. Features of this process depend both on the initial concentration of charge pairs (and hence, on the specific energy loss of a particle) and on the dielectric properties of a substance (see (1)).

2.2. The relative permittivity of organic solid scintillators

The relative permittivity ϵ of the medium is a dimensionless physical quantity characterizing the properties of the insulating (dielectric) medium. It is associated with the effect of polarization of an insulator under an electric field. The force of interaction between two electric charges in a medium is ϵ times as many than in the vacuum. The relative permittivity ϵ much depends on a frequency of electromagnetic

field [9]. It should always be considered because reference tables usually contain data for a static field or low frequencies up to several kHz. At the same time, there are optical methods to determine the relative permittivity of the medium by a refractive index. The value of ϵ obtained by the optical method (frequency of 10^{14} – 10^{15} Hz) may differ significantly from standard reference data.

Table 1 gives the overview of the experimental data on the magnitude of the relative permittivity ϵ of solid scintillation materials [7, 10–13]. In addition, the ϵ -value is the function both of a temperature T of the sample and of the applied pressure. The notes in Table 1, in addition to the references, show the values of the refractive indexes of anisotropic crystals. These values are used for the direct calculation of the average value of the relative permittivity ϵ_{av} using the formula:

$$\epsilon_{av} = \frac{n_g^2 + n_m^2 + n_p^2}{3}, \quad (2)$$

where $\epsilon_{av} = n^2$ is the average relative permittivity of a crystalline material; n_g^2 , n_m^2 , n_p^2

Table 2. The principal components of the dielectric tensor (according to [14]) and the calculated value of the radius of stable polarization environment of an excess charge r_c for anthracene single crystal

	ϵ_1	ϵ_2	ϵ_3	Notes
		2.62 ± 0.03	2.94 ± 0.03	
$r_c, \text{ nm}$	21.28 ± 0.24	18.97 +BR RA III 0.20	13.67 ± 0.26	$T = 300^\circ\text{K}$, atm. pressure

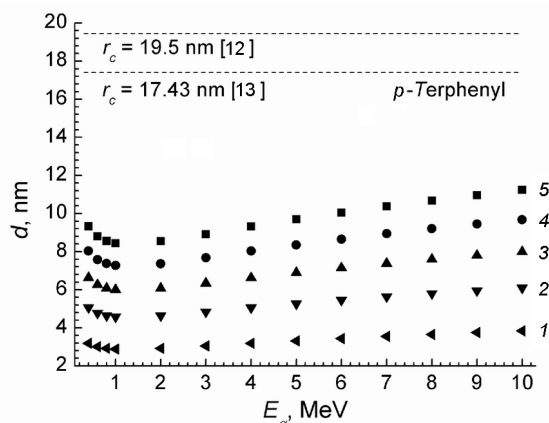


Fig. 1. The calculated average distance d between the centers of charge pairs (M_+ , M_-) in the track of the alpha particle with energy E_α for a p -terphenyl single crystal. The following values of cylindrical track radius r_0 are used: 1 — $r_0 = 10 \text{ nm}$, 2 — $r_0 = 20 \text{ nm}$, 3 — $r_0 = 30 \text{ nm}$, 4 — $r_0 = 40 \text{ nm}$, 5 — $r_0 = 50 \text{ nm}$.

are the principal components of the dielectric tensor along the direction of optical indicatrix axes g , m and p [7].

The results obtained by Munn with co-authors [14] are extremely important in the context of our studies. The complete dielectric tensor of anthracene single crystals has been measured in [14] by a low-frequency capacitance technique at temperatures between 235 and 335 K and at pressures between 1 and 3000 bar. In the range studied, the temperature reduces the anisotropy of the dielectric tensor, individual components changing by amounts of the order of 2 % per 100°K. Table 2 shows the principal components of this tensor measured at 300°K and used in our estimations.

3. Estimations

To study the aspects of the recombination of charge states arising in the tracks of ionizing particles, and the effect of polarization mechanisms on this process we use in this paper the approach developed in [1, 15]. According to this approach, the track

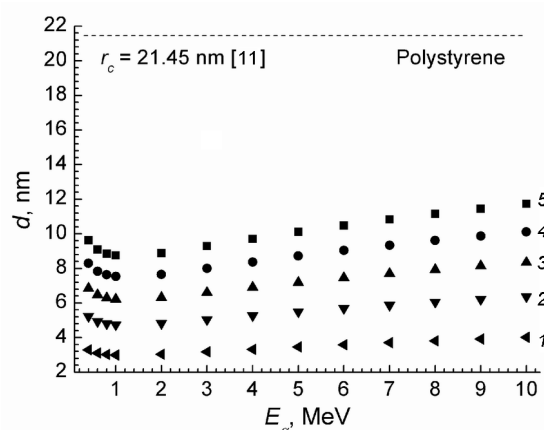


Fig. 2. The calculated average distance d between the centers of charge pairs (M_+ , M_-) in the track of the alpha particle with energy E_α for a polystyrene scintillator. The symbols are the same as in Fig. 1.

has the form of a cylinder whose length is equal to the range of the particle with a definite value of dE/dx . The value of dE/dx is supposed to be constant throughout the particle range. The various methods in allied sciences used for studying the tracks of ionizing particles and their structure, in particular, give appreciably different estimations of track cross-sections (see the analysis of literature data in [1]). Therefore, in our calculations we vary the radius of the cylindrical track r_0 within the range of the above-mentioned estimations.

For organic crystals and plastics the most probable energy of plasmon creation E_{plasm} is usually assumed to be about 20 eV (see the review of papers in [2]). The number of pairs of charge states (M_+ , M_-) finally arising by a decay of plasmons is assumed to be equal to the number of the initial plasmons $N = E/E_{plasm}$. The volume V , occupied by one pair in the track, is calculated as the track volume divided by the number of pairs N . The average distance d between the centers of pairs is estimated as $V^{1/3}$. This value is directly compared with the calculated value of the radius of the stable polarization environment r_c (1),

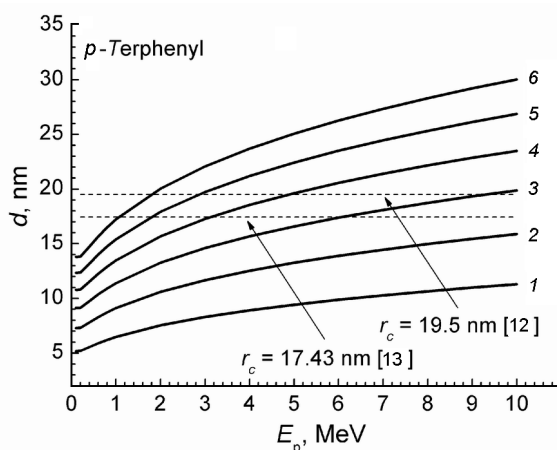


Fig. 3. The calculated average distance d between the centers of charge pairs (M_+ , M_-) in the track of the proton with energy E_p for a p -terphenyl single crystal. The following values of cylindrical track radius r_0 are used: 1 — $r_0 = 15$ nm, 2 — $r_0 = 25$ nm, 3 — $r_0 = 35$ nm, 4 — $r_0 = 45$ nm, 5 — $r_0 = 55$ nm, 6 — $r_0 = 65$ nm.

which an excess carrier creates. The ranges of an alpha particle with energy E_α and a proton with energy E_p are calculated basing on the standard reference database of the National Institute of Standards and Technology (NIST) for the specific energy loss in hydrogen and carbon [16]. After that, the calculations of the ranges for aromatic hydrocarbons under investigation were carried out.

Figs. 1 and 2 show the results of calculations of the average distance d between the centers of charge pairs (M_+ , M_-), which are formed in the track of an alpha particle with energy E_α , for a single-crystal of p -terphenyl and a polystyrene scintillator. Figs. 3 and 4 show similar calculations for the case of the track is a proton with energy E_p . For easy of comparison, the dashed line in the figures denote the values of the radius of the stable polarization environment r_c (1), calculated from the known values of the relative permittivity ϵ of solid organic scintillators (see Table 1).

The results shown in Figs. 1–4 demonstrate the tendencies similar to those that we have obtained earlier for the stilbene single crystal [1, 2]. In the case of the excitation by alpha particles with energies up to 10 MeV for all the varying values of the radius of the particle track r_0 the average distance d between the centers of charge pairs (M_+ , M_-) is always less than the parameter r_c (1). It means that in the alpha particle track ($dE/dx \approx 10^3$ MeV/cm), the polarization environments, both pairs of

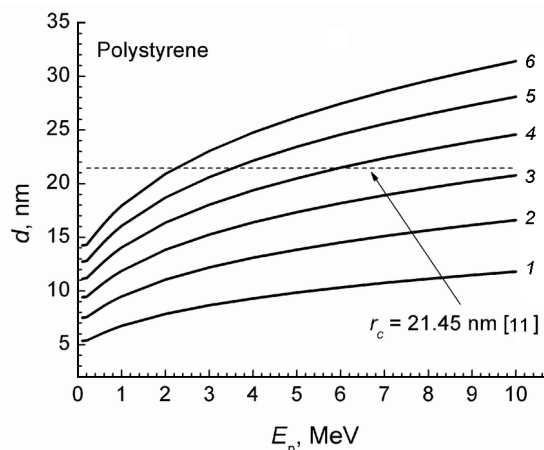


Fig. 4. The calculated average distance d between the centers of charge pairs (M_+ , M_-) in the track of the proton with energy E_p for a polystyrene scintillator. The symbols are the same as in Fig. 3.

charge states, as well as adjacent pairs will effectively overlap. Thus, the polarization interaction will significantly accelerate the fast recombination of charge states in an organic molecular system.

For the case of proton tracks and for the investigated energy range E_p ($dE/dx \approx 10^1$ – 10^2 MeV/cm), the situation is more complicated (see Figs. 3 and 4). Here, the calculated values of the distance d between the centers of charge pairs (M_+ , M_-) strongly depend on the proton energy E_p and the selected values of the track radius r_0 . Therefore, the d -value can be both larger and smaller, than parameter r_c (1). This means that the overlap of the polarization shells of excess charge states arising in the track of a proton is not so dominant and determining mechanism, as in the case of the alpha particle track. In this situation, the recombination of the charge states will proceed through both the polarization interactions (the recombination of hot carriers in the time $\tau \leq 10^{-13}$ s [1, 2, 17]), and the slower process of charge state transport ($\tau > 10^{-12}$ s).

Figs. 5 and 6 present the results of similar calculations of the average distance d between the centers of charge pairs (M_+ , M_-) in the track of alpha particles and protons, respectively, for an anisotropic single crystal of anthracene. The dashed lines in Figs. 5 and 6 represent the values of the parameter r_c , calculated according to (1) for the principal components of the dielectric tensor of anthracene (see Table 2). These results demonstrate weak influence of the an-

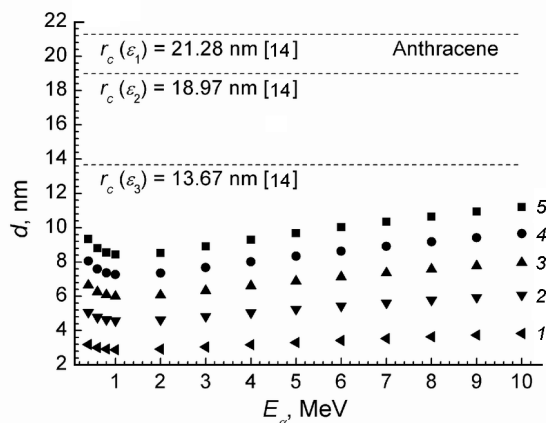


Fig. 5. The calculated average distance d between the centers of charge pairs (M_+ , M_-) in the track of the alpha particle with energy E_α for an anthracene single crystal. The symbols are the same as in Fig. 1.

isotropy of the dielectric properties on the processes occurring in the tracks of ionizing particles with large values of dE/dx .

Comparison of the results presented by Figs. 1–6, as well as the results obtained previously [1], indicates proximity of the nature of charge states energy exchange in the track of an ionizing particle with high specific energy loss dE/dx for all solid organic scintillators under investigation.

It is interesting to compare the results of the estimations of the average distance d between the centers of charge pairs (M_+ , M_-), generated by a particle with high specific energy loss dE/dx , with similar calculations for the case of the radiation with low dE/dx . Let us make such a comparison for an electron with energy $E_e = 0.6$ MeV (this value corresponds to the energy of conversion electrons of a source ^{137}Cs : $E_e = 0.622$ MeV, $dE/dx \approx 10^{-1}$ MeV/cm [2]), using the ESTAR online program of NIST [16]. The continuous slowing down approximation range [2] of an electron with energy $E_e = 0.6$ MeV is equal to 0.189, 0.191, 0.192 and 0.221 cm for anthracene, *p*-terphenyl, stilbene and polystyrene, correspondingly. Assuming, as before in the case of the radiation with high dE/dx , that 20 eV is needed to create a charge state pair (M_+ , M_-), we can estimate the average distance d between the centers of the pairs (M_+ , M_-) for the case of electronic excitation. This calculation gives the d -value in the range from 63 to 74 nm. It is 3–4 times greater than the radius of the stable polarization environment r_c (1) of an excess

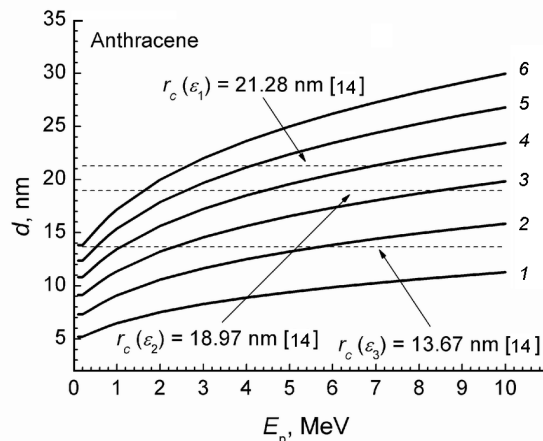


Fig. 6. The calculated average distance d between the centers of charge pairs (M_+ , M_-) in the track of the proton with energy E_p for an anthracene single crystal. The symbols are the same as in Fig. 3.

charge carrier. Thus, in contrast to the above discussed case of the radiations with high dE/dx , in the track of the particle with low dE/dx (an electron with energy $E_e = 0.6$ MeV), the overlap of the polarization environments created by adjacent pairs of the charge states (M_+ , M_-) has to be extremely rare event.

4. Conclusions

In this paper we have estimated the mean distance d between the centers of charge pairs (M_+ , M_-), arising in the track of a charged particle. After that, we compared the d -value with the calculated value of the radius of the stable polarization environment r_c , created by an excess charge carrier in an organic condensed medium. Such the approach allows us to study the specificity of the recombination of the charge states for different densities of radiation excitation and the effect of the polarization mechanisms on the recombination process.

The analysis of the estimations made in this paper, their comparison with the results of earlier studies testifies proximity of the nature of charge states energy exchange in the track of an ionizing particle with the same dE/dx for all solid organic scintillators that we studied. The influence of anisotropy of the dielectric properties on the recombination processes is extremely negligible.

The influence of the polarization effects on the recombination of charge states increases with increasing specific energy loss dE/dx of a particle. Thus, for

electrons with $dE/dx \approx 10^{-1}$ MeV/cm, the probability of overlapping of the polarization environments of adjacent charge pairs (M_+ , M_-) is negligible. This probability become greater in the track of a proton with $dE/dx \approx 10^1-10^2$ MeV/cm, and the polarization interactions will have a significant impact on the recombination of charge pairs. In the case of alpha particles with energies up to 10 MeV ($dE/dx \approx 10^3$ MeV/cm), the average distance d between the centers of charge pairs (M_+ , M_-) is always less than the parameter r_c (1). In this situation, the polarization interaction will be the determinative mechanism of fast recombination of charge states in organic molecular system.

The above estimations are in a good agreement with the results of the light yield measurements of organic solid scintillators excited by ionizing radiations of different types and energies. It is known that the energy yield of luminescence decreases in going from the photoexcitation to the radiation excitation with low specific energy loss dE/dx (gamma photons, electrons, medium and high energy) and then, sequentially, with increasing of dE/dx for protons, alpha particles and heavy ions [1–3, 15, 17]. Thus, the study of the physical processes on the early stages of ionizing radiation exchange energy should take into account the effect of the polarization mechanisms in organic condensed matter. The influence of the polarization on the recombination of charge state pairs, produced by ionizing radiations with different dE/dx , is determined by their local density in the track volume.

The obtained results are intermediate and evaluative in nature, because they do not take into consideration the actual distribution of charge states, both throughout the track volume, and in the isolated excitation regions of the substance. The energy used for the direct production of excited states (bypassing the intermediate charge states) both by the primary particle and by secondary electrons is not taken into account as well.

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Особливості рекомбінації зарядових пар у трекових ділянках органічних твердотільних сцинтиляторів

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Розглянуто особливості рекомбінації зарядових станів, що генеруються в органічному твердотільному сцинтиляторі, при різній густині радіаційного збудження, а також вплив поляризаційних механізмів на цей процес. Оцінки проведено для монокристалів стильбену, *n*-терфенілу, пластмасового сцинтилятора. Вплив анізотропії діелектричних характеристик на процеси, які досліджуються, розглянуто для монокристала антрацену. Проведено оцінки середньої відстані d між центрами зарядових пар (M_+ , M_-), які генеруються у треку зарядженої частинки, і порівняння значення d з розрахунковим значенням радіусу стійкого поляризаційного оточення r_c , яке утворюється надлишковим носієм заряду в органічному конденсованому середовищі. Показано, що ступінь впливу поляризації на специфіку рекомбінації пар зарядових станів, які генеруються іонізуючими випромінюваннями з різними питомими енергетичними втратами dE/dx , визначається їх локальною густиною в об'ємі треку. Вищенаведені оцінки добре відповідають результатам досліджень світлового виходу органічних твердотільних сцинтиляторів.