

SOME ASPECTS OF THE ENERGY EXCHANGE IN AN IONIZING PARTICLE TRACK FOR ORGANIC SOLID DETECTORS

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(Received May 16, 2014)

This work studies the radioluminescence energy yield of organic solid scintillators. The energy that is necessary to produce a scintillation photon is calculated for different ionizing radiations. The analysis of the results is based on the study of the physical processes proceeding during the early stages of the ionizing particle energy exchange. It bases on the concept of a decisive influence of the polarization, which appears in an organic molecular medium, on the recombination of hot charge carriers that results in reduction of the radioluminescence energy yield. The possible reasons of the energy losses are also analyzed.

PACS: 29.40.Mc, 72.20.Jv, 77.22.Ej

1. INTRODUCTION

When employing scintillation detectors, it is important to know their response to ionizing radiations with different specific energy loss (dE/dx). This problem becomes particularly topical for organic scintillators, which are efficient detectors of ionizing radiations with high values of dE/dx (alpha particles, protons, heavy ions) [1, 2, 3, 4]. These types of radiations create high densities of radiation excitation in an organic scintillator and are the most dangerous to human life [5].

Since the 60-ies of the XX century, it was known that the conversion efficiency of organic scintillators, or in other words the radioluminescence energy yield decreases with increasing dE/dx of an ionizing particle [1, 6, 7]. This process characterized by the energy losses became known as the "specific quenching" [1] due to a misapprehension of the mechanisms which cause it. We studied this problem since 2008. Firstly we investigated the dependence of the scintillation response of organic scintillators against dE/dx -values of ionizing radiations [2, 3, 4, 8, 9, 10, 11]. These results allow us to propose the general description (the one-step model) of the ionizing particle energy exchange that takes into account the polarization process as the main factor in recombination of hot charge states in tracks and spurs generated by a primary particle [8, 9, 10]. In this model, as a "step" we assumed a single act of a hop of a charge carrier, in other words, the particle track expansion on the one intermolecular distance. The initial recombination of hot charge states, initiated by the polarization, was considered as a faster process. Actually, a

sudden decrease of pairs of hot charge states in the "frozen" track was investigated. It should be noted that the parameters of this description that were obtained by fitting the experimental data quite accurately describe the quenching process in the ionizing particle track.

In this paper we discuss the physical processes that can determine the energy losses in the track regions for the ionizing radiations with low and high values of dE/dx . Our assumptions are based on the detail analysis of the radioluminescence energy yield of organic scintillators and on the concept of the determining influence of polarization interactions on the recombination of charge states in the particle track.

Organic single crystals of anthracene, stilbene and *p*-terphenyl, and also the plastic scintillator on the base of polystyrene are the objects of the investigation. It should be noted that recently we have developed the new types of organic scintillation detectors, namely, organic polycrystals and composite scintillators [2, 3, 4]. Microcrystalline grains are scintillating material in these types of detectors. It was shown that their scintillation efficiency depends on a size of these grains. Such a detector is effective when the track length of an ionizing particle (in the case of a fast neutron, e.g., it is the track of a recoil proton) does not exceed the linear dimensions of a single grain [4, 9]. A generality of dielectric, optical and scintillation properties of the new types of detectors and classical organic single crystals makes it possible to extend the physical processes under consideration on a wider range of organic scintillators.

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2. THEORY

The ionizing radiations with low values of dE/dx ($< 10^1$ MeV/cm) such as photons of gamma radiation, electrons with energies above 100 eV create separate local regions of high activation density. Such regions are called spurs. A typical spur may consist of one or several pairs of charge states, several excited states. The ionizing radiations with values of $dE/dx \geq 10^1$ MeV/cm (protons, alpha particles, heavy ions) generate the spurs those overlap and form a single region of high activation density, which is known as the primary particle track [2, 10, 12].

During the excitation by an ionizing particle in each a single event of interaction with a medium this particle mainly transmits such a portion of its energy, which exceeds the ionization potential of molecules. The interaction of an ionizing particle with a medium can be described in terms of successive "collisions" of the particle with molecules of the medium [2, 10, 12]. The direct knockout of an electron occurs during the knock-on activation of a molecule. The kinetic energy of such an electron is sufficient for ionization and excitation. Secondary high-energy electrons (100 eV and above [2, 12]) called δ -electrons form short tracks. During the glancing activations of the molecules of a medium the events of ionization proceed through a rapid ($10^{-16} \dots 10^{-15}$ s) intermediate stage of a formation of short-lived superexcited [13] and plasmon states [2, 9, 10, 14].

U. Fano [15] advanced the theory of the existence of plasmons as delocalized many-particle states. He considered organic molecular systems as the most obvious example of systems in which the generation of plasma oscillations was possible. The experimental evidence of the existence of plasmons as quanta of excited valence electrons is based on the measurements of the spectra of characteristic energy loss of fast electrons in organic polymers and single crystals [16, 17]. According to the experimental data the average energy of plasmons in organic condensed media is estimated as $\langle E_p \rangle \sim 20$ eV [2, 10, 14, 15].

Superexcited states arise from a decay of plasmons [13]. In its turn, they decay into pairs of charge carriers. Further recombination of charge pairs can lead to molecular excitation and then, to a luminescence of molecules of an organic scintillator [2, 14].

In a molecular organic system, which consists of initially neutral molecules, the polarization energy that is necessary to produce a molecular polaron by a factor of 10^2 is greater than the energy of intermolecular interaction. The time of the electronic, molecular and in some cases of the lattice polarization formation is less than the time of electronic excitation energy transfer in an organic crystal [2, 18]. Therefore ionizing radiations generate quasi-free electrons in an organic molecular medium [1]. Such the electron rapidly localizes on any one molecule and produces a negative quasi-ion M_- . The molecule, which has lost an electron, in its turn, becomes a positive quasi-ion M_+ . The polarization time of the electron orbitals of the molecules of an organic crystal is esti-

mated as $\tau_e \approx 10^{-16} \dots 10^{-15}$ s [18, 19]. The molecular π -orbitals of the neighbouring nonionizing molecules have a negative charge. They attract to the positive quasi-ion M_+ , but their skeletons repel from it. The polaron M_p^+ is formed. In the case of the negative quasi-ion M_- the molecular π -orbitals of the neighbouring molecules repel from it, and the skeletons of these molecules attract to it. The polaron M_p^- is formed.

According to [18], the polarization surrounding, which originates around a quasi-ion, includes about 10^3 molecules. For example, for the organic crystals of polyacenes the radius of the stable polarization surrounding r_c around a surplus charge is equal to 13...16 nm [19]. The value of r_c is defined as [2, 19]:

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

where $\langle\epsilon\rangle$ is an average relative permittivity of a crystal, k is the Boltzmann constant, T is a temperature, and ϵ_0 is the dielectric constant. In the case when the distance between the polarons M_p^- and M_p^+ is less than r_c these polarons form the molecular-polaron pair, or bipolaron (M_p^-, M_p^+) [2, 18, 19].

For a very strong external electrostatic field ($E \approx 10^6$ V/cm) the scattering time τ of a charge carrier becomes very small value, estimated as 10^{-14} s. For example, according to the results of the direct calculations of the drift velocity of charge carriers in an anthracene crystal based on the experimental data, it was obtained $\tau = 8 \cdot 10^{-14}$ s [19]. The value of τ is comparable with the time of molecular polaron formation [18]. In [20] it was shown that the carrier, whose initial motion is opposite to the field of the parent ion, is thermalized in the time no less than $2 \cdot 10^{-13}$ s. Therefore, the processes occurring after generation of charge carriers in a time comparable or shorter than 10^{-13} s are the processes involving "hot" charge states [2, 14, 19].

The action of the external electric field on the transport and recombination of excess hot states is equivalent to the action of the internal local field created by a bipolaron (M_p^-, M_p^+). The value of this field depends on the initial distance R_0 between M_p^- and M_p^+ . Table 1 shows the calculated values of the local field strength E_{loc} created by the pair of polaron states M_p^- and M_p^+ , which are located on the lattice sites of anthracene crystal. The value of $\epsilon=3.2$ [18] was used. Table 1 shows that the values E_{loc} correspond to the situation of strong external fields, which promote a major decrease in the charge carrier scattering time τ .

Thus, the polarization mechanisms are able to accelerate the process of transport and recombination of hot charge states. They should be taken into account when describing the energy exchange processes, especially for the case of high densities of radiation excitation.

Table 1. Calculated values of the local field strength E_{loc} created by the pair of polaron states M_p^- and M_p^+ in anthracene crystal

Position of M_p^- in a lattice ^{1,2}	$R_0, \text{Å}^1$	$E_{loc}, 10^6 \text{ V/cm}$	Position of M_p^- in a lattice ^{1,2}	$R_0, \text{Å}^1$	$E_{loc}, 10^6 \text{ V/cm}$
(1/2,1/2,0)	5.24	32.4	(0,2,0)	12.07	6.1
(1/2,-1/2,0)	5.24	32.4	(3/2,3/2,0)	15.72	3.6
(0,1,0)	6.04	24.4	(3/2,-3/2,0)	15.72	3.6
(1,0,0)	8.56	12.2	(2,0,0)	17.12	3.0
(1,1,0)	10.48	8.1	(2,2,0)	20.95	2.0
(1,-1,0)	10.48	8.1	(2,-2,0)	20.95	2.0
(0,0,1)	11.18	7.1	(0,0,2)	22.37	1.8

¹ – according to [21],

² – polaron M_p^+ is on (0,0,0) lattice site.

3. EXPERIMENTAL

We investigated the single crystals of stilbene, anthracene and *p*-terphenyl, the plastic scintillator on the base of polysterene. The single crystals of *p*-terphenyl containing 1,4-diphenyl-1,3-butadiene (0.1% in the melt) as an addition agent were studied as well. The cylindrical samples had a thickness of 5 mm. To obtain a wide range of dE/dx we used the medium energy photons of gamma radiation (^{22}Na , ^{60}Co , ^{137}Cs , and ^{152}Eu radionuclide sources; $E_\gamma \sim 10^5 \dots 10^6$ eV), conversion electrons from a ^{137}Cs source ($E_e = 0.622$ MeV), alpha particles in the energy range from 0.7 to 7.5 MeV, fast neutrons from a ^{239}Pu -Be source. The amplitude scintillation spectra of the samples were measured by a multichannel amplitude analyzer AMA-03F. To separate the spectrum of recoil protons generated by fast neutrons from a ^{239}Pu -Be radionuclide source in an organic scintillator, we used the method of discrimination of an ionizing radiation by a scintillation pulse shape [2].

The photoluminescence energy yield Y_{ph} is defined as [22]:

$$Y_{ph} = \Phi_{ph} \times \frac{\lambda_{ex}}{\lambda_{em}^{av}}, \quad (2)$$

where Φ_{ph} is the absolute photofluorescence quantum yield, λ_{ex} and λ_{em}^{av} are the excitation wavelength and the average wavelength of an emission spectrum, respectively.

The average value of the radioluminescence energy yield for a particle with the initial energy E_0 crossing the maximum range in a scintillator is equal to [22]:

$$\langle Y_r \rangle = \frac{1}{E_0} \int_0^{E_0} Y_r(E) dE = \frac{L}{E_0}, \quad (3)$$

where L is the total energy of the radioluminescence photons.

Let us use the following notations: $E_{ph}(\lambda_{em}^{av})$ is the average energy of a luminescence photon, E_i is the excitation energy, Y_i is the luminescence energy yield, and δ_i is the average energy that is necessary to produce a luminescence photon. The suffix i means the excitation of i type. Below by ph, γ, e, n , and α we

denote the following types of excitation i : light photons, photons of gamma radiation, conversion electrons, neutrons and alpha particles, respectively. The symbol of averaging will be omitted.

The value of δ_i is equal to

$$\delta_i = \frac{E_{ph}(\lambda_{em}^{av})}{Y_i}. \quad (4)$$

According to (3) and (4) it is necessary to measure the total number of photons in a scintillation pulse to obtain the values of Y_i and δ_i for i type of excitation with the energy E_i .

The aspects of measuring the amplitude scintillation spectra, calculating the light yield of the organic scintillators and the number of photons in a scintillation pulse for i type of excitation with the energy E_i were previously described [11, 22]. The values of F_{ph} and $E_{ph}(\lambda_{em}^{av})$ were calculated in [22] as well.

Table 2 shows the calculated values of $E_{ph}(\lambda_{em}^{av})$, F_{ph} , Y_i , and δ_i for the photoexcitation and for the excitation by the ionizing radiations with low dE/dx (photons of gamma radiation and conversion electrons).

According to Table 2, even in the case of the excitation by the ionizing radiations with low dE/dx the values of the energy yield Y_γ and Y_e are 10...20 times larger, or, respectively, the values of δ_γ and δ_e are 10...20 times smaller than in the case of the excitation by light photons of the visible range. It should be noted that the values Y_γ and Y_e (δ_γ and δ_e) for the same type of a scintillator are not practically differ.

According to Eq. (3), the values of the radioluminescence energy yield for the cases of excitation by fast neutrons and alpha-particles were described as $Y_n = P_n \times E_{ph}(\lambda_{em}^{av})/E_n$ and $Y_\alpha = P_\alpha \times E_{ph}(\lambda_{em}^{av})/E_\alpha$, where P_n and P_α , respectively, the number of scintillation photons for the neutron and alpha-particle excitations. The values of Y_n for the organic solid scintillators under investigation are in the range from 0.012 to 0.040, and the values of Y_α ranges from 0.0012 to 0.0095. Moreover the scintillation response for excitation by the ionizing radiations with high values of dE/dx is nonlinear.

Table 2. Values of $E_{ph}(\lambda_{em}^{av})$, Φ_{ph} , Y_i , and δ_i (for $i = ph, \gamma$ and e)

Scintillator	$E_{ph}(\lambda_{em}^{av})$, eV	Φ_{ph}	Y_{ph}	δ_{ph} , eV	Y_{γ}	δ_{γ} , eV	Y_e	δ_e , eV
Stilbene	3.08	0.65	0.55	5.60	0.045	68.03	0.042	72.93
p-Terphenyl (undoped)	3.13	0.48	0.41	7.63	0.053	58.82	0.052	59.83
p-Terphenyl (0.1% of 1,4-diphenyl-1,3-butadiene) butadiene)	2.88	0.97	0.77	3.74	0.072	40.16	0.066	43.52
Anthracene	2.71	0.55	0.50	5.42	0.079	34.48	0.073	37.02
Polystyrene (1.5% of <i>p</i> -terphenyl+0.02% of POPOP)	2.84	0.77	0.49	5.80	0.026	108.70	0.025	113.42

Fig. 1 summarizes the data of the average energy δ_i that is necessary to produce a scintillation photon for i type of excitation as a function of dE/dx . The values of dE/dx were calculated using the online programs ESTAR, PSTAR, and ASTAR of the National Institute of Standards and Technology (NIST) [23]. The ranges of the δ_{ph} -values and the δ_{γ} -values (see Table 2) are indicated as well. In the case of neutron excitation we calculated the dE/dx -values for the maximum energy of recoil protons, which were generated by the corresponding fast neutrons with a set of energies E_n . The complicated dependence of Y_{α} for high values of dE/dx (for $E_{\alpha} \cong 1$ MeV) is caused by the effect of an ion recharge [2, 11, 24].

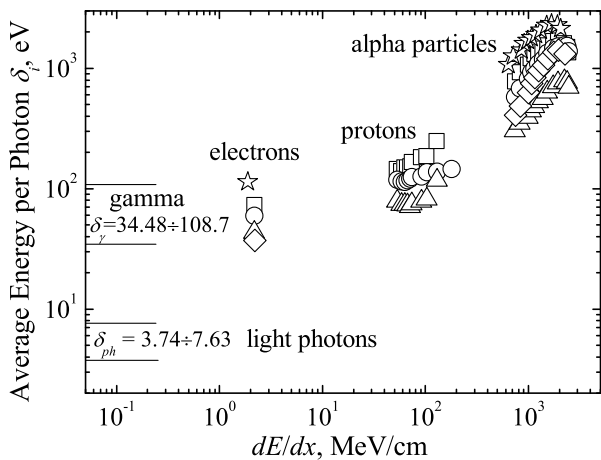


Fig.1. The values of δ_i as a function of dE/dx of the ionizing radiation. Squares, circles, triangles and rhombus represent δ_i -values for the single crystals of stilbene, *p*-terphenyl (undoped), *p*-terphenyl containing 0.1% of 1,4-diphenyl-1,3-butadiene, and anthracene, respectively. Stars denote the polystyrene scintillator

The data presented in Fig. 1 indicate that further increase in dE/dx from protons to alpha-particles causes growing the δ_i -value by a factor of 15-30. At first sight, this increase of the energy losses, or δ_i , with growing dE/dx , seems paradoxical. Indeed, as the value of dE/dx increases, the separate local regions of the high activation density, or spurs, more and more overlap and charge states, which are created, unite in the single region of excitation. The increase in the probability of recombination should

lead to the increase in the number of excited states, and hence, to the increase in the total number of photons in a scintillation pulse. However, we can see from the experiment that the situation is radically opposite.

4. DISCUSSION

A multi-stage nature of the conversion of the primary energy of an ionizing particle to the total energy of photons of a scintillation pulse leads to much less value of the radioluminescence energy yield in contrast the photoluminescence one. It is evident from the above-mentioned results, these losses increase with dE/dx of an ionizing particle. Let us consider the possible physical processes proceeding during the early stages of the ionizing particle exchange energy that may be responsible for these losses for the cases of low and high values of dE/dx , separately.

4.1. Low dE/dx

Ionizing radiations with low dE/dx create in a medium separate local spurs, which consist of one or several charge pairs [2, 12]. A secondary high-energy electron (δ -electron), which is created in such a spur, have a sufficient kinetic energy to escape from this spur. Neighbouring spurs for the ionizing radiations with low dE/dx do not overlap. Therefore, the probability that the electron, which has escaped from one spur, reaches another one is extremely small. Most likely, it will thermalize. This electron will not participate in the process of charge state recombination, and in a subsequent molecular luminescence. So, it may be concluded that the electron escape from a single spur can be a reason for the energy losses for the ionizing radiations with low dE/dx . We estimate the probability of this process on the example of the polystyrene scintillator.

In the general case the total number of δ -electrons in the energy range from E_{min} to E_{max} is equal to [2]:

$$(N_{\delta})_{total} = \frac{2\pi n_e e^4 z^2}{m_e v^2} \left[\frac{1}{E_{min}} - \frac{1}{E_{max}} \right], \quad (5)$$

where n_e is an electron density in a medium, e is an

electron charge, ze and v are the charge and velocity of a primary particle, respectively.

In the absence of an external electric field, δ -electrons whose range is more than the radius of the stable polarization surrounding r_c (1) are able to escape from a spur. For polystyrene r_c is equal to 15.4 nm [25]. According to Ashley [26], an electron with the energy $E_r=450$ eV has such the range in polystyrene. Let us take $E_r=450$ eV as the lower boundary of the electrons, which will escape from the spur with the radius $r_c=15.4$ nm. The ratio of the number of δ -electrons with the energy $E_{esc} \geq E_r$ to the total number of δ -electrons range from E_{min} to E_{max} gives a probability of the δ -electron escape Ξ . From (5) it is easy to obtain that

$$\Xi = \frac{E_{max} - E_r}{E_{max} - E_{min}} \times \frac{E_{min}}{E_r}. \quad (6)$$

Let us calculate the Ξ -value for the case of excitation of an organic scintillator by conversion electrons with the energy $E_e=622$ keV. This value corresponds to the experiment. For the electron excitation we have $E_{max} = E_e = 622$ keV. Fig. 2 presents the results of calculation for the case when E_{min} is varied from 10 eV to $E_r=450$ eV.

The commonly accepted lower boundary for δ -electrons is equal to 100 eV [2, 12]. Taking into account the polarization mechanisms we obtain the value of $\Xi \cong 22\%$ (see Fig.2) for $E_{min}=100$ eV. This value may be used as the estimation for the analysis of the effect of the δ -electron escape from a spur on the energy losses in the ionizing particle track with low dE/dx . A weak distinction in the densities and electron ranges makes possible extending this estimation to all organic solid scintillators under investigation.

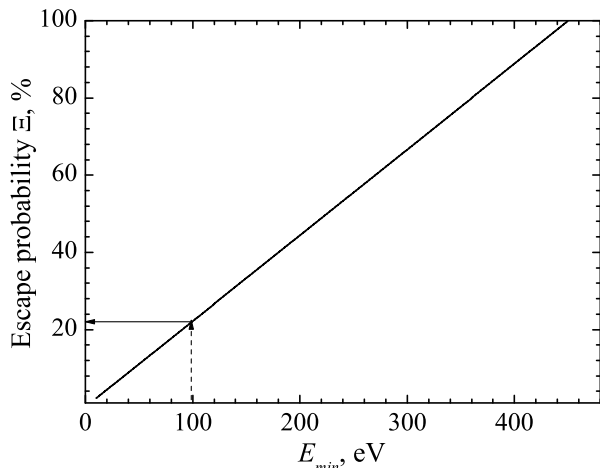


Fig.2. The escape probability Ξ of δ -electrons from a spur. See the text for details

In our opinion, the random recombination of charge state pairs in a single spur is another possible cause of the energy losses occurring in the energy exchange process of an ionizing particle with low dE/dx . In the spur with one pair the geminate recombination takes place when an electron recombines with its parent ion. Such a recombination gives a singlet exciton with a probability close to 100% [2].

The random recombination of charge states prevails in spurs with several pairs [17]. The random recombination gives singlet (S) and triplet (T) states in the ratio 1 to 3. Taking into account the $T - T$ annihilation, the total yield of S -states is approximately equal to 0.4 [27].

We will use the experimental data [28] for the distribution of the energy absorbed in a spur of an electron in water depending on the spur size. According to this data, 56% of the total energy of the electron is used to produce the spurs with one or two pairs of radicals and 44% is used for the spurs containing three and more pairs. Let us extend this result to the case of solid organic scintillators and assume that only the geminate recombination proceeds in spurs with one or two pairs, but the random recombination takes place in spurs with three and more pairs. Considering both the geminate recombination and the random one it is easy to obtain that the yield of S -states is 0.736.

For the case of the excitation by conversion electrons with the energy $E_e=622$ keV we will compare the experimental value of the average energy δ_e that is necessary to produce a radioluminescence photon (see Table 2) and the calculated value δ_{cal} taking into account the above-discussed mechanisms of the energy losses in a single spur. If the energy $\delta_{pair} = \langle E_{pl} \rangle \cong 20$ eV is the average energy that is necessary to create a pair of charge states then approximately 22% of δ -electrons would escape from spurs and will not take part in the recombination process. It means that, actually, the energy necessary for creation a recombining pair is $\delta_{pair}^* = (1/0.78)\delta_{pair}$. Allowing for the yield of S -states (0.736) we can obtain the energy of creation of one S -state: $\delta_S^* = \delta_{pair}^*/0.736$. Dividing the δ_S^* -value by the absolute photofluorescence quantum yield Φ_{ph} we can obtain the calculated value of the energy δ_{cal} that is necessary to produce a radioluminescence photon. Table 3 demonstrates the results of such the calculations. It should be noted that the δ_{cal} -value (see Table 3) is an estimation value. These calculations ignores possible distinctions in the values $\langle E_{pl} \rangle$, Ξ , and in the yield of S -states for each individual scintillator.

Table 3. Values of δ_e and δ_{cal} for organic solid scintillators

Scintillator	δ_e , eV	δ_{cal} , eV
Stilbene	72.93	53.60
p-Terphenyl (undoped)	59.83	72.58
p-Terphenyl (0.1% of 1,4-diphenyl-1,3-butadiene)	43.52	35.92
Anthracene	37.02	63.34
Polystyrene (1.5% of p-terphenyl+0.02% of POPOP)	113.42	45.25

4.2. High dE/dx

An ionizing particle with high dE/dx forms a track in which a density of charge states is high. In such a track region the fast random recombination of charge state pairs dominates. This recombination is

accelerated by the polarization interactions. An influence of the polarization on the process of charge state recombination can be characterized by comparing the radius of the stable polarization surrounding r_c in organic systems under discussion with the actual distance d between charge states generated in the particle track. Such an analysis was performed in [25]. It has been shown that for the case of excitation by alpha particles with the energies $E_\alpha \leq 10$ MeV and for the initial radius r_0 of a cylindrical track (r_0 from 10 to 50 nm) the average distance between pairs d is always less than r_c . It means that the polarization surroundings of the adjacent pairs overlap. Therefore, the polarization effects can initiate a very fast recombination of the charge states in the entire volume of the alpha particle track [25].

In the case of the neutron excitation, in contrast to the alpha particle one, the situation is more complicated. Here, the average distance d between charge pairs can be less than r_c , comparable with r_c , and greater than r_c according to the value of E_n . In the latter case it means that the polarization surroundings of adjacent pairs do not overlap. Therefore, the charge states of the single pair have a possibility to become widely separated from each other both not to interact, and not to be involved in the polarization surroundings of adjacent pairs. In this case, the influence of the polarization on the fast recombination of hot charge states should not be as strong as in the case of the excitation by an alpha particle [25].

Let us consider the charge pair recombination in the alpha particle track more closely using the results have been obtained in [25]. For this purpose let us normalize the calculated values of the average distance d between the centres of charge pairs (see Fig.1 in [25]) for the *p*-terphenyl single crystal on its minimum value, which corresponds to the energy $E_\alpha = 1$ MeV. The values of d_{norm} as a function of E_α are presented by squares in Fig.3.

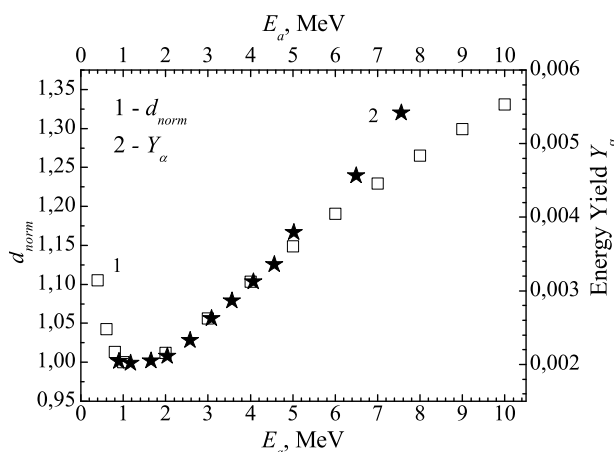


Fig.3. Squares are the normalized values of the average distance between charge pairs in the alpha particle track as a function of E_α for the single crystal of *p*-terphenyl, Stars are the values of Y_α for the single crystal of *p*-terphenyl (undoped)

The experimental values of the energy yield Y_α for

the single crystal of *p*-terphenyl excited by the alpha particles in the range of E_α from 0.9 to 7.6 MeV are presented by stars in Fig.3. From Fig.3 we notice a similar nature of the dependencies of Y_α and d_{norm} in the range of Y_α under investigation. The energy losses decrease with increase in d_{norm} , or with decrease in a density of the recombining pairs in the alpha particle track. It causes increasing the radioluminescence energy yield Y_α . It should be noted that the d_{norm} -value does not depend on the initial radius r_0 of a cylindrical track (see Fig.1 in [25]).

Using the data [25], let us estimate the average number of charge pairs N_r , which are inside the sphere of radius r_c . For the single crystal of *p*-terphenyl r_c is equal to 18.5 nm [25]. If the distance between centres of pairs is d , then the radius of the sphere containing one pair is $d/2$. The maximum coefficient of the dense packing of spheres is about 0.75. Table 4 shows the results of such estimations for the single crystal of *p*-terphenyl excited by an alpha particle with the energy $E_\alpha=5$ MeV.

Table 4. The N_r -value for $E_\alpha=5$ MeV and $r_c=18.5$ nm

r_0 , nm	d , nm	N_r
10	3.32	1038
20	5.26	261
30	6.90	115
40	8.35	65
50	9.69	41

The electrostatic field energy of a quasi-ion necessary for formation a polarization surrounding, E_{pol} , is about 1...1.5 eV [2, 18, 19]. It means that the total energy of a single recombining pair is comparable with the energy of S_1 -state (see. Table 2, the value of $E_{ph}(\lambda_{em}^{av})$). Table 4 shows that within a very small local region of a medium, whose size is determined by the value of r_c , the average number of charge pairs $N_r \gg 1$, and pairs of charge states can recombine simultaneously. In such a situation, an immediate and very strong local heating of a medium becomes very probable. This local heating should lead to a dramatic increase in the nonradiative deactivation of excitation [29]. Such a mechanism of the "temperature quenching" can be a reason for the effective primary quenching of the radioluminescence for the ionizing radiations with high values of dE/dx .

It is reasonable to suggest that the main energy losses in the particle track with high dE/dx occur during the stage of the hot charge state recombination accelerated by the polarization interactions. During the primary recombination ($\tau < 10^{-13}$) the concentration of charge states drastically drops. Their subsequent recombination leads to creation of S - and T -states in the ratio 3 to 1. However, the concentration of S - and T -states in the "cooled" track remains high enough. It makes possible the subsequent secondary stage of the quenching process. Let us name this process as the "concentration" quenching of luminescence [2, 30].

The concentration of S_1 -states, which are the source of the fluorescence in the $S_1 \rightarrow S_0 + h\nu$ transition, can be reduced in the processes of the mutual $S - S$ annihilation, $S - T$ annihilation, singlet exciton fission. In the condition of high activation density the process of $S - S$ annihilation can lead to the autoionization mechanism. The quenching of S_1 -states by doublet states results in decreasing their concentration as well. The contribution from each of the above-mentioned processes to the total energy losses during the stage of energy exchange of excited states will be defined by characteristic rate constants of these processes [30].

Both the primary stage of hot charge state recombination and the secondary stage of the energy exchange of excited states are the concentration-controlled processes. Nevertheless the secondary quenching proceeds after the primary one when the concentration of the states is appreciably lower. Therefore the influence of the secondary quenching is not as significant as the influence of the primary one.

5. CONCLUSIONS

Table 5 summarizes the results of this work. The values of the luminescence energy yield Y_i and the energy δ_i that is necessary to produce a luminescence photon for the photoexcitation as well as for the excitation by different types of ionizing radiation are presented.

Table 5. Ranges of the values Y_i and δ_i for organic solid scintillators

Type of excitation	i	Y_i	δ_i , eV
Light photons	ph	0.41-0.77	3.74-7.63
Gamma and conversion electrons	γ, e	0.025-0.079	34.48-113.42
Neutrons	n	0.012-0.040	71.3-250.0
Alpha particles	α	0.0012-0.0095	302.4-2346.2

The main reasons of the energy losses for the ionizing radiations with low values of dE/dx are the following:

1) An electron escape from a spur that decreases the number of charge state pairs, which recombine.

2) The random recombination of charge states in a single spur results in a decrease in the probability of formation of S -states. If one takes into account both the geminate recombination, and the random one, the total yield of S -states is approximately equal to 0.4.

The main reasons of the energy losses for the ionizing radiations with high values of dE/dx are the following:

1) The primary recombination of hot charge states (the "temperature quenching" of luminescence), accelerated by the polarization interactions in an organic molecular medium.

2) The secondary ("concentration") quenching proceeding during the subsequent stage of energy exchange

of excited states. The influence of this process is minor in comparison with the influence of the "temperature quenching" of luminescence.

ACKNOWLEDGEMENTS

This work was supported by the State Fund for Fundamental Research of Ukraine (project No. F58/06, "The effect of large radiation doses on scintillation and optical properties of novel types of organic detectors").

References

1. J.B. Birks. *The Theory and Practice of Scintillation Counting*, London: "Pergamon", 1967, 662 p.
2. N.Z. Galunov, O.A. Tarasenko. *Ionizing Radiation Track Formation in Organic Condensed Media*, Kharkov: "ISMA", 2011, 480 p. [in Russian].
3. S.V. Budakovskiy, N.Z. Galunov, N.L. Karavaeva, et al. New effective organic scintillators for fast neutron and short-range radiation detection // *IEEE Trans. on Nucl. Sci.* 2007, v. 54, N.6, p. 2734-2740.
4. O. Tarasenko, N. Galunov, N. Karavaeva, et al. Stilbene composite scintillators as detectors of fast neutrons emitted by a ^{252}Cf source // *Radiat. Meas.* 2013, v. 58, p. 61-65.
5. The 2007 Recommendations of the International Commission on Radiological Protection. Publication 103 // *Annals of the ICRP.* 2007, v. 37, N.2-4, p. 1-332.
6. J.B. Birks. Scintillation from organic crystals: Specific fluorescence and relative response to different radiations // *Proc. Phys. Soc.* 1951, v. A64, p. 874-877.
7. G.T. Wright. Scintillation response of organic phosphores // *Phys. Rev.* 1953, v. 91, N5, p. 1282-1283.
8. N.Z. Galunov, E.V. Martynenko. Semi-empirical description of quenching processes in a track of ionizing particles for organic crystalline scintillators // *Functional Materials.* 2008, v. 15, N3, p. 380-387.
9. N. Galunov, O. Tarasenko. Primary quenching in a track of ionizing particle for organic scintillation crystalline materials // *Radiat. Meas.* 2010, v. 45, p. 380-382.
10. N.Z. Galunov, V.P. Seminozhenko, E.V. Martynenko, O.A. Tarasenko. Quenching process in an ionizing particle track for organic crystalline scintillation detectors // *Problems of Atomic Science and Technology.* 2013, v. 85, p. 210-219.
11. N.Z. Galunov, O.A. Tarasenko, V.A. Tarasov. Radio-luminescence energy yield of organic solid scintillators excited by ionizing radiations with different specific energy loss // *Journal of Applied Spectroscopy.* 2013, v. 80, N4. p. 550-555.
12. A. Mozumder. *Fundamentals of Radiation Chemistry*, San Diego, London, Boston, New York, Sydney, Tokyo, Toronto: "Academic Press", 1999, 408 p.
13. R.L. Platzman Superexcited states of molecules // *Radiat. Res.* 1962, v. 17, N3, p. 419-425.
14. M. Pope, C.E. Swenberg. *Electronic Processes in Organic Crystals*, New York, Oxford: "Clarendon Press", 1982, 432 p.

15. U. Fano. A common mechanism of collective phenomena // *Rev. Mod. Phys.* 1992, v. 64, p. 313-319.
16. N. Swanson, C.J. Powell. Excitation of π -electrons in polystyrene and similar polymers by 20-keV electrons // *J. Chem. Phys.* 1963, v. 39, N3. p. 630-634.
17. M. Schott. Remarks on the process of carrier generation in the electron-bombarded crystalline anthracene // *Mol. Cryst.* 1969, v. 5, N3, p. 229-243.
18. E.A. Silinsh, M.V. Kurik, V. Capek. *Electronic Processes in Organic Molecular Crystals: Effects of Localization and Polarization*, Riga: "Zinatne", 1988, 329 p [in Russian].
19. E.A. Silinsh, V. Capek. *Organic Molecular Crystals: Interaction, Localization, and Transport Phenomena*, New York: "American Institute of Physics", 1994, 402 p.
20. E.A. Silinsh, A.J. Jurgis, G.A. Shlihta. Charge carrier transport phenomena in polyacene crystals: molecular polaron approach // *Journal of Molecular Electronics.* 1987, v. 3, p. 123-127.
21. P.J. Bounds, W. Siebrand, I. Eisenstein, et al. Calculation and spectroscopic assignment of charge-transfer states in solid anthracene, tetracene and pentacene // *Chem. Phys.* 1985, v. 95, N3, p. 197-212.
22. O.A. Tarasenko, N.Z. Galunov, V.D. Panikarskaya, et al. Luminescence energy yields of organic solid materials excited by photons of light or gamma-radiation // *Functional Materials.* 2012, v. 19, N3, p. 404-409.
23. NIST Standard Reference Database 124. Stopping-Power and Range Tables for Electrons, Protons, and Helium Ions (<http://www.nist.gov/pml/data/star/index.cfm>).
24. U. Fano. Penetration of protons, alpha particles, and mesons // *Ann. Rev. Nuclear Sci.* 1963, v. 13, p. 1-66.
25. O.A. Tarasenko. Features of charge pairs recombination in the track regions of organic solid scintillators // *Functional Materials.* 2012, v. 19, N4, p. 415-420.
26. J.C. Ashley, J.C. Tung, R.H. Ritchie. Inelastic interactions of electrons with polystyrene: calculations of mean free paths, stopping powers, and CSDA ranges // *IEEE Trans. on Nucl. Sci.* 1978, v. NS-25, N6, p. 1566-1570.
27. W. Helfrich, W.G. Schneider. Transients of volume controlled current and of recombination radiation in anthracene // *J. Chem. Phys.* 1966, v. 44, N8, p. 2902-2909.
28. H.A. Schwarz. Application of the spur diffusion model to radiation chemistry of aqueous solutions // *J. Phys. Chem.* 1969, v. 73, N6, p. 1928-1937.
29. I.I. Kucherov, A.N. Faidysh. Transport and transfer of electronic excitation energy in anthracene and naphthalene crystals // *Izv. Akad. Nauk SSSR (Ser. Fiz.)*. 1958, v. 22, N1, p. 29-35 [in Russian].
30. N.E. Geacintov, M. Binder, C.E. Swenberg, M. Pope. Exciton dynamics in α -particle tracks in organic crystals: magnetic field study of the scintillation in tetracene crystals // *Phys. Rev. B.* 1975, v. 12, N10, p. 4113-4134.

ОСОБЕННОСТИ РАЗМЕНА ЭНЕРГИИ В ТРЕКЕ ИОНИЗИРУЮЩЕЙ ЧАСТИЦЫ ДЛЯ ОРГАНИЧЕСКИХ ТВЕРДОТЕЛЬНЫХ ДЕТЕКТОРОВ

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Исследуется энергетический выход радиoluminesценции органических твердотельных сцинтилляторов. Проведен расчет энергии, которая расходуется на создание одного фотона сцинтилляционного импульса при разных видах возбуждения. Анализ результатов основывается на изучении физических процессов, протекающих на ранних стадиях размена энергии ионизирующей частицы. В основе этого анализа лежит концепция об определяющем влиянии поляризации органической молекулярной среды на рекомбинацию горячих носителей заряда, что приводит к уменьшению энергетического выхода радиoluminesценции. Анализируются возможные причины энергетических потерь для ионизирующих излучений разных типов.

ОСОБЛИВОСТІ РОЗМІНУ ЕНЕРГІЇ В ТРЕКУ ІОНІЗУЮЧОЇ ЧАСТИНКИ ДЛЯ ОРГАНІЧНИХ ТВЕРДОТІЛЬНИХ ДЕТЕКТОРІВ

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Досліджується енергетичний вихід радіoluminesценції органічних твердотільних сцинтиляторів. Проведено розрахунок енергії, яка витрачається на створення одного фотона сцинтиляційного імпульсу при різних видах збудження. Аналіз результатів ґрунтується на вивченні фізичних процесів, які відбуваються на ранніх стадіях розміну енергії іонізуючої частинки. В основі цього аналізу лежить концепція про визначальний вплив поляризації органічного молекулярного середовища на рекомбінацію гарячих носіїв заряду, що спричинює зменшення енергетичного виходу радіoluminesценції. Анализуються можливі причини енергетичних втрат для іонізуючих випромінювань різних типів.