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

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In this paper we studied the influence of ionization loss of a heavy charged particle on change in the distance between the charge pairs, which are generated in an organic solid scintillator, and on recombination processes between them. The specific energy loss dE/dx and the ranges of alpha particles and protons were calculated by computer simulation using SRIM-2011 program. The values of a correction factor for the revised calculation of the distance between the charge pairs were obtained. In contrast to the previous studies, we took into account the change of dE/dx-value of a charged particle passing through an organic medium.

В работе изучается влияние ионизационных потерь тяжелой заряженной частицы на изменение расстояния между зарядовыми парами, которые генерируются в органическом твердотельном сцинтилляторе, и на рекомбинационные процессы между ними. Расчет удельных энергетических потерь dE/dx и пробегов альфа-частиц и протонов проведен методами компьютерного моделирования с использованием программы SRIM-2011. Получены значения поправочных коэффициентов для уточнённого расчета расстояния между зарядовыми парами. Данные коэффициенты, в отличие от предыдущих исследований, учитывают изменение dE/dx заряженной частицы, проходящей через слой вещества.

1. Introduction

In [1–4] we studied recombination processes of the charge states, generated in an organic solid scintillator by ionizing radiations of various types, as well as the influence of the polarization mechanisms on this process. Organic single crystals of stilbene, *p*-terphenyl, anthracene, and the polystyrene plastic scintillator were used as the objects of the research. Comparative analysis of the estimated value of an average distance d between the centers of charge pairs (M_+, M_-) with the estimated value of a radius of the stable polarization environment r_c [2, 5], created by an excess charge carrier in an organic condensed medium, was carried out. It was shown that in the case of excitation by an alpha particle with the energy E_α in a range from 0.4 to 10 MeV (it corresponds to the specific energy loss $dE/dx \approx 10^3$ MeV/cm [2]) the value of the average distance d between pairs is always less than r_c. This fact testified to the mechanism of rapid recombination of the charge states, accelerated by the polarization interactions in organic condensed media. In the case of excitation by protons with energies E_p from 0.1 to 10 MeV ($dE/dx \approx 10^1$–10^2 MeV/cm [2]), the d-value could be less, equal or even greater than r_c. In the last case, the slower process of charge state transport follows the process of their recombination [1, 2].

The calculations of the distance d between the centers of charge pairs (M_+, M_-) were performed in [1–4] for the case when their density in the whole track volume is

assumed to be the same. However, it is well known that a value of the specific energy loss dE/dx of a heavy charged particle increases as its velocity decreases. Hence, the number of ionization events per unit interval of the way increases and the Bragg's curve describes the ionization loss of a particle [6]. This paper examines the possible influence of change in the specific energy loss dE/dx of a particle on an average distance d between the charge pairs, and therefore, on the recombination processes in the track of a heavy charged particle.

2. Objects and parameters for estimations

As in our previous paper [1], organic single crystals of *trans*-stilbene ($C_{14}H_{12}$, density $\rho = 1.22$ g/cm^3), *p*-terphenyl ($C_{18}H_{14}$, $\rho = 1.23$ g/cm^3), anthracene ($C_{14}H_{10}$, $\rho = 1.25$ g/cm^3), and plastic scintillator on the base of polystyrene ($[-CH_2-C(C_6H_5)H-]_n$, $\rho = 1.05$ g/cm^3) became the objects of investigation. The results of previous studies have shown that process of the charge state energy exchange in the track of an ionizing particle is general for all solid organic scintillators under investigation [1–4]. Therefore, this paper clearly presents the results of the calculations only for the single crystal of stilbene, but for other organic scintillation materials it gives such information only in a finite form to preserve generality.

According to [2, 5], a radius of the stable polarization environment r_c, created by an excess charge carrier in an organic condensed medium will define as:

$$r_c = \frac{e^2}{4\pi\langle\varepsilon\rangle\varepsilon_0 kT}, \qquad (1)$$

where $\langle\varepsilon\rangle$ is an average relative permittivity of a material, k is the 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 Coulomb field which holds the charges [2, 5]. The mechanism of creation of a molecular polaron pair and subsequent recombination of such pair has been described in detail in [1, 2]. It should be only mentioned, that the process of 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 the charge pairs (and hence, on the specific energy loss dE/dx of a particle), and on the dielectric properties of a substance (see (1)). Review of the data on relative permittivity ε of organic materials which are widely used in scintillation technique was presented in [1].

The values of ranges of alpha particles R_α and protons R_p with the energies E_α and E_p, correspondingly, as well as the values of the specific energy loss dE/dx of these particles were simulated by SRIM-2011 program (The Stopping and Range of Ions in Matter) [7]. The particle range R is determined by the value of the specific energy loss dE/dx (or by the stopping power of a medium) as [2]:

$$R = \int_{E_0}^{E_1} \left(-\frac{dE}{dx}\right)^{-1} dE. \qquad (2)$$

The particle range R is obtained by (numerical) integration with respect to dE/dx. Integration is limited in the lower limit, since the description of the stopping power is not reliable at low energies and, in any case, nuclear collisions take place of the electronic stopping at low velocities [2]. The specific lower limit is not so important if the energy of the incident particle is large. The value of E_0 corresponds to the energy of 1 keV in SRIM-2011 program [7].

3. Estimations

As in [1–4], let us assume that the most probable energy Ω to create a single pair of charge states (M_+, M_-) is about 20 eV for organic crystals and plastics (see the review of the papers in [2]). Then the average number of pairs N_{av}, appearing on the unit interval of a path Δx in the unit area perpendicular to a movement of an ionizing particle, can be presented as:

$$N_{av} = \frac{\langle dE/dx\rangle}{\Omega}\Delta x = \frac{E}{R\times\Omega}\Delta x, \qquad (3)$$

where E is the particle energy in eV, R is the particle range, an $\langle dE/dx\rangle$ range is the specific energy loss which is averaged over the maximum particle range.

A single pair in the track occupies some volume V. Its average value can be estimated as the result of division of the track volume by the number of pairs. Then an average distance d between the centers of

pairs on the unit interval of path Δx is equal to:

$$d = V^{1/3} = \left(\frac{\pi r_0^2 \times \Delta x}{N_{av}}\right)^{1/3} = \left(\frac{\pi r_0^2 \times R \times \Omega}{E}\right)^{1/3}, \quad (4)$$

where r_0 is a value of the cylindrical track radius. This approach [1–4] ignores changes in the ionization loss of a charged particle which penetrates a medium. To take into account this factor let us describe the number of the charge state pairs N_{pair}, which appear in the layer with number i along the particle path (all the layers are Δx in thick), as follows:

$$(N_{pair})_i = \left(\frac{dE}{dx}\right)_i \times \frac{\Delta x}{\Omega}. \quad (5)$$

In this approach we use the value of $(dE/dx)_i$ for i-layer of the ionizing particle path.

Let us introduce a correction factor:

$$\mu_i = \frac{(N_{pair})_i}{N_{av}} = \frac{(dE/dx)_i}{\langle dE/dx \rangle} = \frac{(dE/dx)_i}{E/R}. \quad (6)$$

The correction factor μ_i (6) shows a deviation of the estimated value of the number of charge state pairs $(N_{pair})_i$ (5) on the penetration depth of a particle from l_i to $l_i + \Delta x$. It takes into account real changes in the specific energy loss $(dE/dx)_i$ of the particle from the value of the number of charge state pairs N_{av} (3), obtained for the case when their density throughout the track volume is averaged. Using (6), one can obtain correction for Eq.(4) in a form:

$$(d_{corr})_i = \left(\frac{\pi r_0^2 \Delta x}{N_{pair}}\right)^{1/3} = \left(\frac{\pi r_0^2 \Delta x}{N_{av}\mu_i}\right)^{1/3}. \quad (7)$$

Using SRIM-2011 program [7], we calculated values of the specific energy loss dE/dx and ranges R for alpha particles and protons. The calculations were performed for alpha particles of the following energies E_α: 0.39, 0.67, 0.9, 1.17, 1.66, 2.04, 2.58, 3.08, 3.57, 4.07, 4.56, 5.03, 6.5 and 7.56 MeV and for protons of the following energies E_p: 0.85, 2.0, 3.1, 4.2, 4.9, 6.4, 6.7, 7.3, 7.9, 8.6 and 9.7 MeV. Such a choice of the energies is not accidental. It allows adapting the obtained estimations to the results of light yield measurements in the real experimental conditions (for more details see [1]).

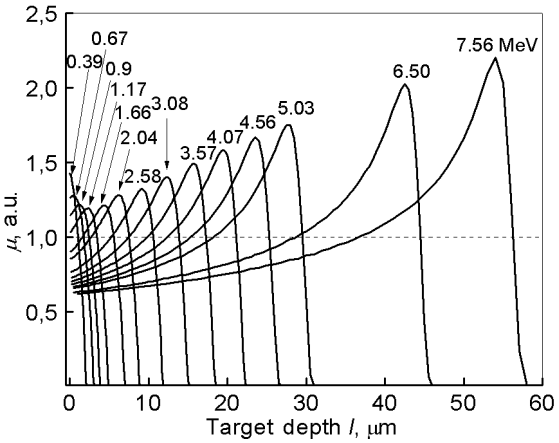

Fig. 1. Values of the correction factor μ (6) versus the target depth l of an alpha particle with the energy E_α for stilbene single crystal. The energies of alpha particles used for calculation are shown in MeV. The dashed line corresponds to the case when $N_{pair}(5) = N_{av}(3)$ ($\mu = 1$).

To improve an accuracy of the calculations we used the following approach. The range of analysis (plot window) r is used for calculations of the specific energy loss of a particle. It was varied depending on the energy and the type of an ionizing particle. In each case it value was chosen to satisfy the inequality $r > R$. Using SRIM-2011 program we calculated the set of the discrete values $(dE/dx)_i$ for the penetration depths of a particle $l_i = i\Delta x$ for i from 1 to m with the step Δx ($m\Delta x = r$). For the chosen value of $m = 100$ an error in determining the values l_i and $(dE/dx)_i$ was equal about 1 %. Thus, the parameters μ_i (6) and d_{corr} (7) also represent the sets of m discrete values with the step of Δx. Hereinafter, the subscript i will be omitted, bearing in mind that both values of l, and the functions of this argument (μ, d_{corr}) are determined discretely.

Using values of the specific energy loss (dE/dx) and ranges R we calculated values of the correction factor μ (6) for alpha particles and protons of the above-mentioned energies. The results of these calculations for the organic single crystal of stilbene are shown in Fig. 1 and 2, correspondingly.

According to calculation of the correction factor μ (6) a value of the distance d_{corr} (7) between the centers of charge pairs (M_+, M_-) was obtained. The d_{corr}-value takes into account change in the charge pair density across the path of a charged particle.

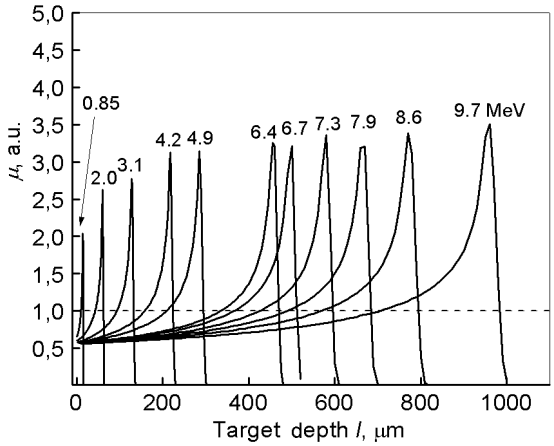

Fig. 2. Values of the correction factor μ (6) versus the target depth l of a proton with the energy E_p for stilbene single crystal. The energies of protons used for calculation are shown in MeV. The dashed line corresponds to the case when $N_{pair}(5) = N_{av}$ (3) (μ = 1).

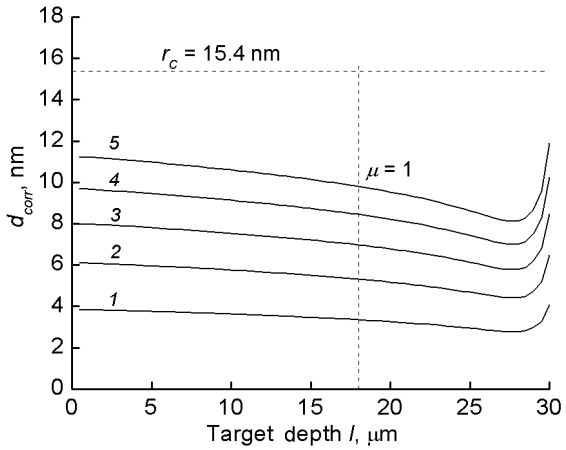

Fig. 3. Values d_{corr} (7) versus the target depth l of an alpha particle with the energy E_α = 5.03 MeV for stilbene single crystal. Values of the cylindrical track radius r_0 used for calculation are the following: *1* — 10 nm, *2* — 20 nm, *3* — 30 nm, *4* — 40 nm and *5* — 50 nm. The horizontal dashed line shows the value r_c = 15.4 nm [1], the cross-points of the vertical dashed line with calculated curves demonstrate the values of d_{corr} (7) for μ = 1.

Figs. 3 and 4 show the results of such calculations for alpha particle with the energy E_α = 5.03 MeV and for proton with the energy E_p = 4.9 MeV, correspondingly. Here, as in [1–4], the calculations are carried out for several values of the cylindrical track radius r_0, namely, 10, 20, 30, 40 and 50 nm for alpha particles; 15, 25, 35, 45, 55 and 65 nm for protons. The value of d_{corr} (7) is compared with the radius of stable polarization environment r_c = 15.4 nm, which has been calculated in [1] according to [8].

For all values of the cylindrical track radius r_0 the distance d_{corr} (7) between the centers of charge pairs varies depending on the target depth l as follows: up to 17 % of its average value (μ = 1) for the case of alpha particle with the energy E_α = 5.03 MeV, up to 32 % for the case of proton with the energy E_p = 4.9 MeV. This deviation is practically does not depend on the r_0-value.

In the case of alpha particle with the energy E_α = 5.03 MeV a value of the distance d_{corr} (7) between the centers of charge pairs is always less than the radius of stable polarization environment r_c (1) for all values of l (see Fig. 3). Thus, $d_{corr} < r_c$ for any one of the values of target depth l, and hence recombination process of the generated charge states is speeded up by strong polarization interactions.

Fig. 4 shows that for proton excitation the situation is more complicated than for alpha excitation (Fig. 3). The ratio between the values d_{corr} and r_c depends both on the value of a radius of the cylindrical track r_0, that we choose for the calculations, and on the penetration depth l of a proton in the medium. This fact is more illustrative for r_0 = 25 nm. In this situation, $d_{corr} > r_c$ for proton with the initial energy E_p = 4.9 MeV and for the values of $l \leq 100$ μm. The d_{corr}-value becomes less than r_c (for $l > 100$ μm) as the proton energy is reduced and its specific energy loss is increased. Thus, for the case of proton excitation the recombination of the charge states is more intricate: when $d_{corr} < r_c$, as in the case of alpha excitation, recombination of the charge state pairs is initiated by strong polarization interactions in an organic condensed medium, but when $d_{corr} > r_c$, the process of charge state transport may precede the process of their fast recombination. In this context, the analysis of changes in the ionization loss of a particle that we consider in this work is more important for the case of proton excitation than for the case of alpha particle excitation.

The above-mentioned results have been confirmed by calculating the values of the distance d_{corr} between the charge state pairs generated in the tracks of alpha particles and protons for other organic solid scintillators. So, both for anthracene and *p*-terphenyl single crystals, and for the polystyrene plastic scintillator irradiated by alpha particle with the energy E_α = 5.03 MeV, the maximum deviation of the distance d_{corr} (7)

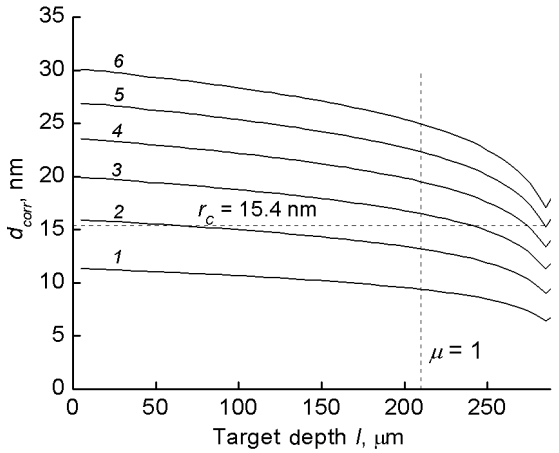

Fig. 4. Values d_{corr} (7) versus the target depth l of a proton with the energy E_p = 4.9 MeV for stilbene single crystal. Values of the cylindrical track radius r_0 used for calculation are the following: *1* — 15 nm, *2* — 25 nm, *3* — 35 nm, *4* — 45 nm, *5* — 55 nm and *6* — 65 nm. The other designations are the same as in Fig. 4.

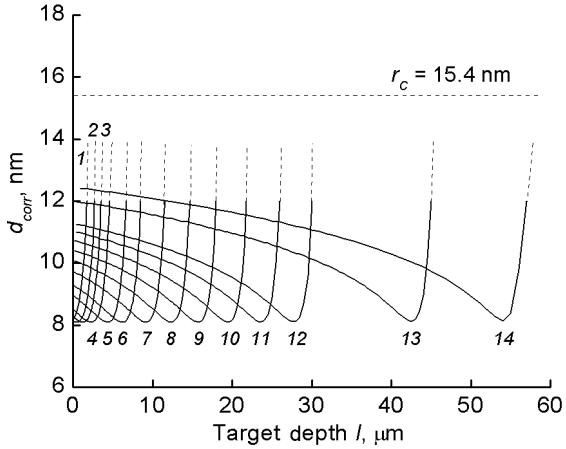

Fig. 5. Values d_{corr} (7) versus the target depth l of an alpha particle with the energy E_α for stilbene single crystal. The value of the cylindrical track radius r_0 = 50 nm is used. The energies E_α of alpha particles used for the calculation are the following: *1* — 0.39, *2* — 0.67, *3* — 0.9, *4* — 1.17, *5* — 1.66, *6* — 2.04, *7* — 2.58, *8* — 3.08, *9* — 3.57, *10* — 4.07, *11* — 4.56, *12* — 5.03, *13* — 6.5 and *14* — 7.56 MeV.

between the centers of charge pairs from its average value (i.e. for μ = 1) did not exceed 17 %. For the case of proton with the energy E_p = 4.9 MeV the maximum deviation of the d_{corr}-value (7), as for the single crystal of stilbene, did not exceed 32 %. This result is explained by the negligible difference in the values of ranges and the specific energy loss of alpha particles and protons for the group of organic scintillation materials under investigation.

The highest values of d_{corr} (7) for alpha particle with the energy E_α = 5.03 MeV correspond to the highest value of parameter r_0 (see Fig. 3). Therefore, in the case of alpha excitation we carried out the additional calculation of d_{corr} (7) for the value of the cylindrical track radius r_0 = 50 nm. Alpha particle energy E_α was varied. Fig. 5 shows the results of these calculations. As in the case of E_α = 5.03 MeV (see Fig. 3), the value of d_{corr} is always less than r_c for the whole energy range under investigation. The deviation of d_{corr} from the average value (for μ = 1) slightly increases with increasing E_α, reaching 23 % for E_α = 7.56 MeV. The dashed lines in Fig. 5 correspond to the end of the particle path, or to the range of the low-energy particles. An accurate calculation of the density of charge pairs, and hence the parameter d_{corr}, is limited by an ambiguity of the description of the specific energy loss for low-energy ionizing particles (see the comments to formula (2)). It should be noted that in the case when the initial energy of a particle is large (that is corresponds to the energy range under consideration) the volume occupied by this area is very small in comparison with the total volume of the particle track. Therefore, the total number of charge state pairs generated in this area is too small, and theirs effect on the processes of charge state energy exchange at high excitation densities have to be neglected.

4. Conclusions

The influence of ionization loss of a heavy charged particle on change in the distance between the charge pairs, generated in organic solid scintillators, and on recombination processes between them was studied.

The specific energy loss dE/dx and ranges of alpha particles and protons were calculated by computer simulation using SRIM-2011 program. The values of correction factor μ (6) for the revised calculation of the distance between the charge pairs were obtained. In contrast to the previous studies [1–4], the change in dE/dx-value of a charged particle passing through an organic medium was taken into account.

The distance d_{corr} (7) between the centers of the charge pairs varies depending on the target depth l as follows: up to 17 % of its average value (μ = 1) for the case of alpha

particle with the energy $E_\alpha = 5.03$ MeV, and up to 32 % for the case of proton with the energy $E_p = 4.9$ MeV. This deviation is practically does not depend on the value of the cylindrical track radius r_0.

The analysis of the results obtained for alpha particles in the energy range E_α from 0.4 to 7.5 MeV and for the r_0-values from 10 to 50 nm shows that the distance d_{corr} (7) between the centers of the charge pairs is always less than the radius of stable polarization environment r_c(1). It is true for any one of the values of target depth l. Therefore, strong polarization interactions always have to make recombination process of the charge states pairs in the alpha particle track more effective and fast.

Such situation seems not to be so univocal for the case of proton excitation. The ratio between the values d_{corr} and r_c depends both on the value of the cylindrical track radius r_0, that we choose for the calculations, and on the penetration depth l of a proton in an organic medium. When $d_{corr} < r_c$, as in the case of alpha excitation, the recombination of the charge state pairs is initiated by strong polarization interactions in an organic condensed medium, but when $d_{corr} > r_c$, the slowly process of charge state transport may precede the process of their fast recombination. In this context, the analysis of changes in the ionization loss of an ionizing particle that we considered in this work is more important for the case of proton excitation than for alpha particle excitation.

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Особливості рекомбінації зарядових пар у трекових ділянках органічних твердотільних сцинтиляторів. Частина II

О.А.Тарасенко

Вивчено вплив іонізаційних втрат важкої зарядженої частинки на зміну відстані між зарядовими парами, які генеруються в органічному твердотільному сцинтиляторі, та на рекомбінаційні процеси між ними. Розрахунок питомих енергетичних втрат dE/dx і пробігів альфа-частинок і протонів проведено методами комп'ютерного моделювання з використанням програми SRIM-2011. Отримано значення поправочних коефіцієнтів для уточненого розрахунку відстані між зарядовими парами. Дані коефіцієнти, на відміну від попередніх досліджень, враховують зміну dE/dx зарядженої частинки, яка проходить через шар речовини.