QUENCHING PROCESS IN AN IONIZING PARTICLE TRACK FOR ORGANIC CRYSTALLINE SCINTILLATION DETECTORS

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The processes of primary energy exchange of ionizing radiation in track regions of organic molecular scintillation crystalline materials are reviewed. The semi-empirical description of the processes of primary energy exchange of ionizing radiation, which takes into account quenching in a track for organic crystalline scintillators, is discussed. Parameters of this description characterize the primary quenching effect in a track. The revised model of the scintillation process was obtained due to taking into account the influence of polarization interaction between excess charge states and molecules of a substance in the track area.

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1. INTRODUCTION

It is generally known that organic scintillators and detectors based on these scintillators are the most effective for detecting of short-range ionizing radiation (alpha and beta particles), as well as for spectrometry of fast neutrons [1-3]. It means that organic scintillators are the most widely used for detecting the ionizing radiations those produce tracks in such a scintillation material. So, the study of the processes of energy exchange in the track regions, which cause energy loss and determine eventually the scintillation response, is a topical task.

As far back as fifties of the XX century it was known that the energy conversion efficiency of an organic scintillator decreases with specific energy loss dE/dr increase. It is considerably lower under excitation by ionizing radiations with high dE/dr (e.g. $10^1...10^3$ MeV/cm, for fast neutrons or alpha particles) than under excitation by radiations with low dE/dr (for photons of gamma radiation of middle energies $dE/dr \leq 10^{-1} MeV/cm$) [1, 3]. The nature of the process causing such loss was not clear and this effect had been called the "specific quenching" [1, 4, 5]. It was considered that the main part of this loss (quenching) has to take place in a track area (regions of high activation density) at the initial stage of scintillation processes (is less than $50 ns$ after excitation) [1-7]. This statement was based on the results of the measurements of kinetics of the slow component of scintillation pulse. It was shown that if the time after excitation is larger than $50 ns$ then

the fast component of scintillation pulse does not distort it slow component kinetics, and it is described by a diffusion approximation. It was observed the very slow time variation of the concentration of triplet-excited molecules, determined by diffusion expansion of a track but not by quenching [3, 6, 7]. Therefore the existence of energy loss indicates that some effective quenching has to forego the stage of molecular excitation energy exchange, when the diffusion approximation is applicable. Besides, the existence of appreciable additional energy loss ("specific quenching") have to be the result of primary processes of the energy exchange in the ionizing particle track area, but not in surrounding regions of low activation density [1, 3-5, 8, 9].

It was observed that for ionizing radiations with high dE/dr the dependence of the scintillation response of an organic scintillator versus ionizing radiation energy is non-linear [1, 3-5, 8, 9]. Many researchers studied quenching in a track and the dependence of the scintillation response of an organic scintillator versus specific energy loss dE/dr [1-6, 10-12]. The hypotheses of monomolecular quenching in a track of an ionizing particle [10], bimolecular quenching [6, 11], quenching resulting from interaction of m molecules ($m > 1$) [12] and corresponding relationships of the scintillation response and specific energy loss dE/dr were studied. One can find the review of early works on the subject in [1] and the review of this material before 1997 in [3]. The semi-empirical relationship giving the dependence of specific fluores-

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cence dL/dr versus specific energy loss dE/dr proposed by Birks [1, 10] is most widely used in scientific literature. It was based on the assumption of "unimolecular" quenching in a track of an ionizing particle. According to Birks [1]:

$$dL/dr = \frac{S\,dE/dr}{1 + k\,dE/dr}\,,\qquad(1)$$

S is absolute scintillation efficiency, k is a quenching parameter, $B\,dE/dr$ is the specific density of ionized and excited molecules along the particle track, B is a constant. For small dE/dr (fast electrons) equation (1) gives [1]:

$$dL/dr \approx S\,dE/dr\,.$$

At large dE/dr (e.g. alpha particles with energies less than $6\,MeV$) equation (1) becomes [1]:

$$dL/dr \approx S/kB = const\,.$$

Unfortunately, the experimental data contradict this theory. Let P denote the scintillation response caused by alpha particle excitation and R denote the range of an alpha particle with energy E_α in corresponding scintillation substance. Using the data of calculated R-values cited in [13, 14] and the P-values measured in [13-16] we can obtain P/R values versus alpha particle energy E_α. Such a result was obtained for a number of stilbene and p-terphenyl single crystals. Fig. 1 demonstrates the typical example of the dependence of P/R values versus E_α. One can see that the P/R-ratio is not a constant. Thus, the description of the quenching in a track proposed by Birks is easy to use, but it does not fit experimental data well.
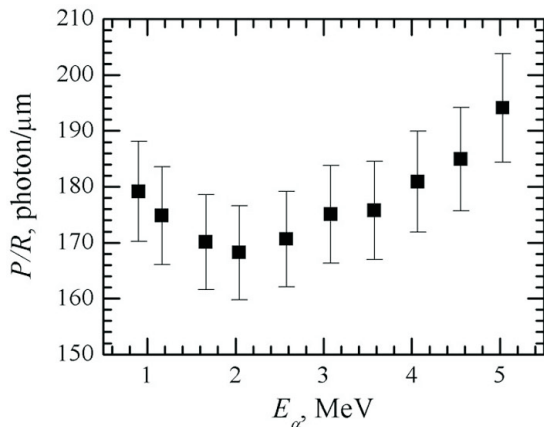

Fig.1. *P/R-values versus alpha particle energy E_α for a $\oslash40\,mm \times 10\,mm$ stilbene single crystal*

Another weak point of all known descriptions of the quenching in a track was that they did not take into account the influence of electronic polarization of molecules of a scintillator by charge states. Really, the charge states generation is one the main stages of the radioluminescence pulse formation [1-5, 17-19]. All effective organic scintillators can be referred to dielectrics or wide-gap semiconductors. The polarization primarily influences on the processes of

charge state formation, transport and recombination in them [2, 3, 20-24]. It becomes evident even from the fact that for organic molecular crystals the energy of polarization interactions is by two orders of magnitude greater than the energy of Van der Waals' interactions, that holds molecules in the lattice of a molecular crystal [2, 3, 21, 22, 24]. The average time that is necessary for a random hop of a charge carrier from one molecule to other in organic crystals is about $10^{-12}\,s$. The time that is necessary for electronic polarization of neighbouring molecules varies from 10^{-16} to $10^{-15}\,s$ [21, 22, 24]. If a molecule loses an electron and becomes a positive molecular quasi-ion M_{p+}, it means that this electron has to localize on some other molecule and a negative molecular quasi-ion M_{p-} arises. The M_{p+} and M_{p-} molecular quasi-ions are surrounded with unionized molecules. Molecular π-orbitals of neighbouring molecules have negative charge; therefore they are attracted to the M_{p+} and are repelled from the M_{p-}. So, the generation of charge states by an ionizing radiation is accompanied by polarization interactions between a surplus charge carrier localized on a molecule and its surroundings. The polarons arises. According to the calculations cited in [21, 24], about 7000 molecules around such the quasi-ion are polarized and the diameter of polarization surrounding r_C varies from 13 to $16\,nm$ for different organic crystals. If M_{p+} and M_{p-} are on a some distance $r \le r_C$ then a polaron pair is formed. The polaron pair formation is accompanied with appearance of the strong electrostatic field between M_{p+} and M_{p-} states (can reach $10^7\,V/cm$). This field will promote the recombination of M_{p+} and M_{p-} states, which results in molecular excitation and therefore in the radioluminescence or in non-radiative recombination (quenching) [21, 22, 24]. According to [23], in a very strong field when electric field strength $E \sim 10^6\,V/cm$, which is comparable with the local field strength inside the polaron pair, the time of charge carrier scattering τ becomes an extremely small value and can be estimated as $10^{-14}\,s$. For instance, the direct calculation of drift velocity for anthracene made on the base of experimental data gives $\tau = 8 \times 10^{-14}\,s$ [23]. In [25] it was shown that the time, which is necessary to a charge state to thermalize, exceeds $10^{-13}\,s$. The τ-value gives the estimation of an average transport time of a charge carrier in the process of ordered motion of electron and hole (belongs to the same polaron pair) to recombine. It is the transport of charge carriers with no additional relaxation on each a molecule on their way to each other. Of cause if M_{p+} and M_{p-} are on a distance $r > r_C$, the random walk of polarons have to forego the polaron pair formation.

Thus, the polarization effect can dramatically change the speed of the processes of recombination of charge states in a particle track. The comparison of the radius of track cross-section r0 for a $5.5\,MeV$ alpha particle ($50\,nm$) [3] with the diameter of polarization surrounding r_C (from 13 to $16\,nm$) had shown that we have to estimate the r_C as a large value [13].

It means that recombination of polarons arising on a distance $r \leq r_C r$ and forming polaron pairs without random walk can be an appreciable fraction of the process of charge states recombination in a track.

The above-mentioned analysis substantiated a necessity of a complete revision of the results obtained in works of the 50th...90th years, where the "specific quenching" was studied ignoring the influence of the polarization of scintillator molecules by charge states in a particle track.

2. EXPERIMENTAL

In our experiments [7, 13, 14, 16, 26-29] we used a $^{239}Pu - Be$ radionuclide source of fast neutrons and gamma radiation, ^{239}Pu and ^{241}Am sources of alpha particles, as well as ^{22}Na, ^{57}Co, ^{60}Co, ^{137}Cs, and ^{152}Eu sources of gamma radiation. To obtain the different energies E_α of alpha excitation we used the effect of moderation of alpha particles in air [13]. The method of selective detection of fast neutron in presence of gamma-background has been reported in [28, 30, 31] and the method of reconstruction of a neutron spectrum from the corresponding recoil proton spectrum was presented in [16, 26-28, 31].

In our experiments we used organic single crystals, polycrystals and composite scintillators. Polycrystals and composite scintillators are rather new type of organic materials that we have recently studied [13, 14, 16, 27, 29]. They were prepared from different grain size fractions obtained by grinding a single crystal at low temperatures followed by sieving. It should be noted that the structure ordering of organic single crystals, polycrystals and composite scintillators based on the same scintillation material differs dramatically [3, 14, 27].

There were studied $\oslash 25\,mm \times 20\,mm$ and $\oslash 50\,mm \times 5\,mm$ stilbene single crystals (unary system); a $\oslash 25\,mm \times 15\,mm$ p-terphenyl single crystal doped by $0.1\,wt.\%$ 1,4-diphenyl-1,3-butadiene (binary system); $\oslash 20\,mm \times 5\,mm$ p-terphenyl single crystals doped by 0.05, 0.1, 0.3, 0.5%$wt.\%$ of 1,4-diphenyl-1,3-butadiene (binary system); 7 polycrystals obtained from grains of stilbene (unary system); 10 composite scintillators obtained from grains of stilbene (unary system); 10 composite scintillators obtained from grains of doped p-terphenyl (binary system). The grain size values L changed in the following ten ranges: between i) 1 and 1.3, ii) 1.3 and 1.5, iii) 1.5 and 1.7, iv) 1.7 and 2.0, v) 2.0 and 2.2, vi) 2.2 and 2.5, vii) 2.5 and 3, viii) 3 and 3.5, ix) 3.5 and 4.0, x) 4.0 and 4.5 mm. In the case of the polycrystals, the crystalline grains with the size values L of ranges i) - vii) were used. To compare the properties of unary and binary p-terphenyl scintillators we used the composite scintillators based on scintillation grains with size L changed in the following three ranges: between i) 0.5 and 1, ii) 1 and 2, iii) more than 2 mm. All the composite scintillators were $\oslash 30\,mm \times 20\,mm$. All the polycrystals were $\oslash 30\,mm \times 5\,mm$.

3. RESULTS AND DISCUSSION

Unfortunately, measuring the shape of the scintillation pulse could not give enough information to investigate of the processes of primary energy exchange in track those cause the energy loss. Really, the fast component of the scintillation pulse, which is generated in the regions of low activation density (beyond the track), masks the initial part of the slow component i.e. that part, which contain the information about quenching in a track, during $50\,ns$ after excitation [1, 3-5]. In the same time, the intensity of the slow component of a scintillation pulse is a several orders of magnitude less than the fast component intensity for these early moments after excitation [3]. For time $t > 50\,ns$ the influence of the processes of primary energy exchange and quenching in a track is negligible. The investigations of a scintillation response were more promising way. It was mentioned in Section I that the non-linear dependence of the scintillation response of an organic scintillator versus the energy of ionizing radiations with the high dE/dr values is caused by the losses on a quenching in a track. So, the dependence of the scintillation response versus ionizing radiation energy can give the initial information for investigation of the processes occurring in a track. The comparison of scintillation responses of one the same scintillator for the case of excitation when track is formed (high dE/dr) and for the case of excitation when track is not formed (low dE/dr) allows to make the direct estimation of the influence of quenching in a track on the scintillation response and to separate the information concerning the quenching in a track.

The following value was used to estimate the effect of additional loss (quenching) in the track of i-type ionizing radiation [3, 13, 14, 26]:

$$\zeta_{i\gamma} = \left(\frac{P_i}{E_i}\right) \Big/ \left(\frac{P_\gamma}{E_\gamma}\right) = \frac{E_\gamma}{E_i}\Big|_{P_i = P_\gamma}, \qquad (2)$$

where P_i and P_γ are measured amplitudes of the scintillation response generated by i-type of radiation and by gamma radiation with energies E_i and E_γ, respectively. The $\zeta_{i\gamma}$-value (2) gives the ratio between the energy E_γ, which is necessary to produce P scintillation photons in a given scintillator, and the energy E_i, which is necessary not only to produce the same number P of scintillation photons in the same scintillator but also to compensate the energy loss in a track of the particle of i-type with energy E_i. Therefore, value (2) has to be always less then one and decreases with quenching effect increase [14].

We had obtained the $\varsigma_{i\gamma}$-values (2) for alpha and neutron excitations ($i = \alpha, n$) for the above-mentioned experimental samples. The obtained results were typical for all scintillators and for some experimental samples they are presented in [13, 14, 16, 26]. Table presents the $\varsigma_{\alpha\gamma}$- values obtained for single crystals of p-terphenyl doped with different concentration of the 1,4-diphenyl-1,3-butadiene.

The $\varsigma_{\alpha\gamma}$-values obtained for single crystals of p-terphenyl doped with different concentrations C of 1,4-diphenyl-1,3-butadiene

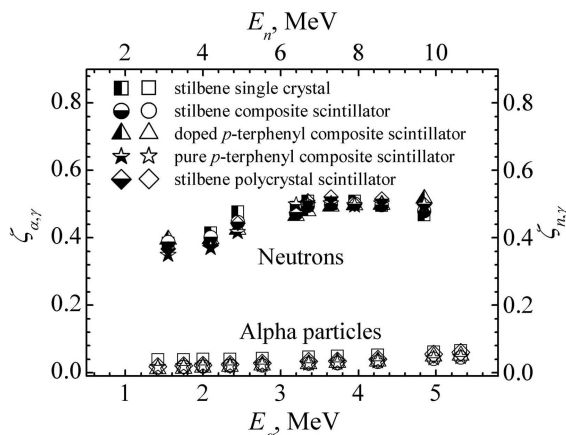| C, wt. %	$\varsigma_{\alpha\gamma}$
0.05	0.093
0.1	0.097
0.3	0.096
0.5	0.095

Fig. 2. *The $\varsigma_{\alpha\gamma}$- and $\varsigma_{n\gamma}$-values (lower and upper family of curves) as a function of the energy of alpha (E_α) and neutron (E_n) excitation, respectively, for a $\oslash 25\,mm \times 20\,mm$ stilbene single crystal, a stilbene polycrystal (L is from 1.7 to 2.0 mm) and for composite scintillators on the base of: stilbene grains (L is from 1.5 to 1.7 mm), doped p-terphenyl grains (L is from 1.0 to 2.0 mm), and pure p-terphenyl grains (L is from 1.0 to 2.0 mm)*

Fig. 2 shows $\varsigma_{\alpha\gamma}$- and $\varsigma_{n\gamma}$-values obtained for stilbene and p-terphenyl based scintillators. The $\varsigma_{\alpha\gamma}$- values are always lower than $\varsigma_{n\gamma}$-values, so the quenching effect for alpha excitation is higher than for neutron excitation. The selection of experimental samples, that we had made, allowed us to compare the influence on the quenching in a track of: i) the type of a scintillation material; ii) structure ordering of a scintillator; iii) a presence of molecules of the addition agent and it concentration C; and iv) the type of ionizing radiation (i.e. density of excitation in a particle track). The obtained that $\varsigma_{i\gamma}$- values for all concentrations C of 1,4-diphenyl-1,3-butadiene are very close (Table). It denotes that the influence of the concentration of the molecules of the addition agent on the "specific quenching" is negligible. Thus, in our early researches we used scintillators based on p-terphenyl doped by 1,4-diphenyl-1,3-butadiene only with $C = 0.1\,wt.\%$ as the binary system.

As one can see (Fig. 2), the difference in $\varsigma_{i\gamma}$-values for the same type of excitation for different scintillators (the single crystal, the polycrystal and the composite scintillators based on stilbene, pure and doped

p-terphenyl) is negligible in comparison with difference in $\varsigma_{\alpha\gamma}$- and $\varsigma_{n\gamma}$-values for the same scintillator. It means that the influence on the "specific quenching" of the type of scintillation material, it structural ordering and presence of luminescent addition agents is negligible in comparison with the influence of the type of excitation [13, 14].

3.1. Semi-empirical Description of the Process of Primary Energy Exchange in a Particle Track

Recently [13], we have obtained the semi-empirical description of the processes of primary energy exchange of ionizing radiation and quenching in a track for organic crystalline scintillators, which takes into account the influence of polarization interaction. The basic points of this description are presented below.

An action of ionizing radiation on organic molecular solid scintillators results in formation of the primary states, plasmons [2, 3]. The energy of such plasmon for organic crystals is $\Omega \sim 20\,eV$ [2, 3]. The concentration of plasmons per 1 molecule ν_{i0} for excitation of i type is [13]:

$$\nu_i(t)|_{t=0} \equiv \nu_{i0} = \frac{1}{N} \times \frac{E_i}{\Omega} \times \frac{1}{V_i}, \qquad (3)$$

where N is the number of molecules in a cubic centimetre, E is an energy of the particle of i type, V is the volume of a track. Equation (3) gives some value ν_{i0} averaged over a track volume and does not contain the information about the space distribution of the primary states in a track. It was mentioned above, that the scintillation response is the experimental data that we used as the initial information for investigation of the processes in a track. The value of the scintillation response is obtained as an averaged characteristic of accumulated signals from single measurements (each of which is caused by a single particle). For a single measurement the signal is obtained as the result of summation of the signals from photons those emitted in scintillation pulse and originate in different points of the scintillator. The information about the coordinate of a single scintillation photon is not saved. Therefore such approach, when we used average n0-value, which does not contain the information about the space distribution of charged states, is reasonable.

The calculation of ν_{i0}-values versus E_i for stilbene and p-terphenyl single crystals excited by protons and alpha particles was made in [13]. The results showed that the ν_{i0}-value decreases with E_i increase because the ratio between E_i and the range of particle (and therefore the value E_i/V_i) decreases with E_i increase.

The primary states (plasmons) dissociate on, practically, the same number of the pairs of charge states [2, 3, 18]. In (3) $\nu(t)$ is normalized value and the total relative concentration of all molecules (ionized and unionized) is equal to one. According to (3), the average relative concentration of pairs of molecular ions in a track $\nu(t)$ is dimensionless quantity that for zero time ($t = 0$) is equal to the initial

concentration of the primary states n0. Thus, the average relative concentration of unionized molecules in a track is $1 - \nu(t)$ and $1 - \nu_0$ for time $t > 0$ and $t = 0$, respectively. The decrease of concentration of pairs of charged states (or pairs of polarons) ν_0 can be a result of i) the recombination of charge states, ii) diffusion expansion of a particle track [13, 14]. If $q(\nu, t)$ defines the recombination of charged states (polarons) in a track and D is the diffusion coefficient of the charged states, then in the general case:

$$\frac{d\nu(r,t)}{dt} = D\nabla\nu(r,t) - \{q(\nu,t)\}\,\nu(r,t)\,. \qquad (4)$$

The second term in (4) defines a change of ν because of recombination of polarons. It can be simultaneous recombination of $j \geq l$ polaron pairs when:

i) polarons are on a distance $r \leq r_C$ (rate parameter p_j);

ii) polarons are on a distance $r > r_C$ and have to come near each other at first (rate parameter c_j). It means that the second term in (4) can be presented as:

$$- \{q(\nu,t)\}\,\nu(r,t) = - \big\{c_1\nu(r,t) + p_1\left[1 - \nu(r,t)\right]\nu(r,t) + c_2\nu^2(r,t) +$$
$$+ p_2\left[1 - \nu(r,t)\right]\nu^2(r,t) + c_3\nu^3(r,t) + p_3\left[1 - \nu(r,t)\right]\nu^3(r,t) + ...\big\} =$$
$$= -(c_1 + p_1)\left\{1 + \frac{\sum_{j\geq 1}(p_{j+1} + c_{j+1} - p_j)\,\nu^j(r,t)}{c_1 + p_1}\right\}\nu(r,t) \approx$$
$$\approx -a_0(\nu)\left\{1 + a_1(\nu)\nu(r,t) + a_2(\nu)\nu^2(r,t) + ...\right\}\nu(r,t)\,. \qquad (5)$$

So, $q(\nu, t)$ is represented as some expansion into a power series in terms of $\nu(t)$. The charge state transport between molecules of an organic crystal can be described as the intermolecular barrier tunnelling [2, 20-24]. For a single uncompensated charge carrier the polarization interactions in the opposite directions are the same. If the single uncompensated charge carrier is trapped by the trap E' in depth then due to the effect of polarization the real depth of such trap becomes equal to:

$$E = E' + \Delta E\,, \qquad (6)$$

where for the systems under consideration ΔE is about $0.1\,eV$ [3, 24]. Therefore, if dynamic trap formation is initiated even by lattice oscillation modes ($E' = kT = 0.026\,eV$) then the average time that is necessary for a single uncompensated charge carrier to make a random "hop" on a neighbouring molecule $t_D \geq 10^{-12}\,s$ [24]. This value gives the lower bound of the average time that is necessary for a particle track to expand on one molecular layer (i.e. to make one "step" in the process of it expansion) [13].

According to data cited in Section I, the primary recombination of polaron pairs (rate parameter p_j) has to be a very effective and rapid process. It has to proceed at time interval of $10^{-14}...10^{-13}\,s$ after excitation, i.e. faster than a particle track expansion on a distance equal to mean distance between neighboring molecules of a scintillator (on one molecular layer) ($\geq 10^{-12}\,s$). Thus, during such rapid recombination of the main part of polaron pairs the track cross-section and the space distribution of n-value practically will not change. So, the primary recombination of polaron pairs, which result in the primary quenching in a track, seems to be a rapid and almost "one-step" process that determines mainly the changing of the concentration of pairs n of charged states. Experimental data as well does not contradict this.

As it was mentioned above, the dependences of the $\zeta_{i,\gamma}$-value (2) versus E_i ($i = \alpha$, n) obtained by the experiments for the scintillators with dramatically different structure ordering had shown, that the influence of the structural ordering of a scintillator on the total energy loss (quenching) in the track is negligible in comparison with the influence of the type of excitation (i.e. the primary concentration of pairs of charged states ν_0 in a track). This result did not prove the assumption, that the primary recombination is much more effective than diffusion and causes the main losses, but it agreed with it.

"One step model" was formulated to simulate the primary recombination of polaron pairs in a track [13]. In this model time Δt of track expansion at the mean distance between neighbouring molecules is taken as a "one step". The average relative concentration of pairs of charged states at the moment $t_0 = 0$ is equal to ν_0. During such the "one step" the primary concentration of pairs of charge states in a track undergoes a "sudden change" (time $\delta t < \Delta t$) and to the next moment $t_1 = t_0 + \Delta t$ it practically falls to zero. Changing of the concentration of polarons pairs and, in consequence, the mean distance between polarons over the time range shorter than Δt is not examined in the model. That is why such recombination process we have named a "sudden change" [13]. If this process is connected with the recombination probability as a function of the mean distance between polarons, than with ν_0- value growth the fraction of polarons, which recombination can be described within the "one step model", has to increase. It should be noted, that in real situation the relative concentration of pairs of polarons $\nu(t)$ that remains after this primary recombination is not equal to zero. It is determined by the number of charged states that leaves a track during its formation and

by the number of charged states in a track that are far enough from the points of primary recombination and therefore does not take part in it.

For the case of the "one step" model instead of (4) is used (7), which gives expansion into power series not for the function $q(\nu, t)$ but for $q_0(\nu_0) = const$:

$$\frac{d\nu(t)}{dt} \approx \{q_0(\nu_0)\} \nu(t) \approx$$
$$-A\{1 + B\nu_0 + C\nu^2{}_0 + F\nu^3{}_0...\} \nu(t). \quad (7)$$

The constants A, B, C, F, etc. in (7) are effective values that can be obtained only from the approximation of experimental data. They are obtained as a result of averaging of $a_i(\nu)$-values (5) over the ν_0 range, corresponding to different energies of excitation of certain type for the concrete scintillator. From (5) it follows that due to polarization effect all these constants (B, C, F, etc.) except $A > 0$ can be both positive and negative.

It was considered the solid crystalline substitution solution that contains the base molecules of X-type (donors of excitation energy) and the dope molecules of Y-type (acceptors of excitation energy) with low concentration of Y molecules. Solving the simultaneous equations, which describe the time variation of the number of excited molecules of donors and acceptors, we obtained the expression of the scintillation response for such binary system in a form [13]:

$$P_{XY} = \frac{S_{XY}\nu_0}{1 + B\nu_0 + C\nu^2{}_0 + F\nu^3{}_0...}, \quad (8)$$

$$S_{XY} \equiv \left\{ \frac{\lambda_X}{\eta_X + (\kappa + \theta_{XY})C_\gamma} + \right.$$
$$\left. + \frac{\lambda_\gamma(\kappa + \theta_{XY})C_\gamma}{\eta_\gamma(\eta_X + \theta_{XY}C_\gamma)} \right\} \frac{\omega}{A}, \quad (9)$$

where A-value is taking from (7); ω is the probability of X molecules to be excited in the result of recombination of charge states; $\theta_{XY}(t)$ and κ are the probabilities of non-radiative and radiative transport of excitation energy from X to Y molecules, respectively; $\lambda_X(\lambda_Y)$ is the number of detected photons of the radioluminescence of $X(Y)$-type molecules per one excited $X(Y)$-type molecule; $C_{X(Y)}$ is the molecular concentration; $\eta_{X(Y)}$ is the probability of transition of an excited $X(Y)$-type molecule in the ground state.

3.2. Comparison with the Results of the Experiments

The constants in (8) can be obtained as a result of approximation the experimental values of the scintillation response versus calculated n0 (3) for alpha and neutron excitation. Such approximation was made for all scintillators mentioned in Section 2. Fig. 3 shows an example of the approximation procedure for a stilbene single crystal. For all scintillators the accuracy of approximation for considered ranges of ν_0 was satisfactory when only the coefficients S, B and C in (8) were nonzero.
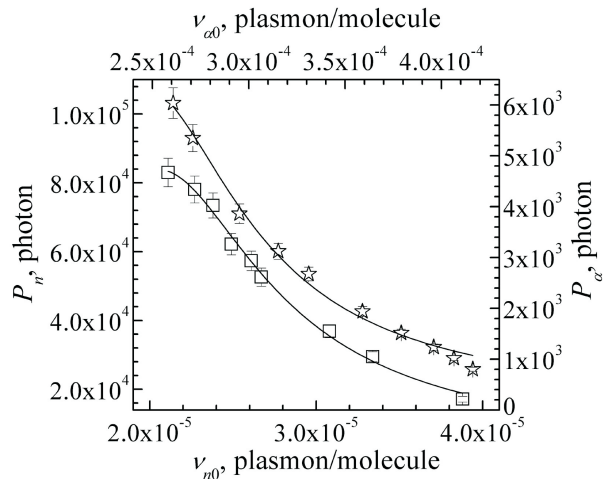
Fig.3. *The approximations (lines) by (8) of experimental values of the scintillation response P for alpha (stars) and neutron (squares) excitation versus calculated ν_0-values (3) for a $\oslash 25\,mm \times 20\,mm$ stilbene single crystal*

The combination of the values $S\nu_0$, $B\nu_0$ and $C\nu_0{}^2$ (8) can provide the information about non-proportionality of the scintillation response connected with quenching effect [13, 29]. The obtained values of $B\nu_0$ and $C\nu_0{}^2$ versus energy of excitation E for some scintillators are cited in [13]. The result had shown that values $S\nu_0$, $B\nu_0$ and $C\nu_0{}^2$ are comparable for stilbene and p-terphenyl based scintillators [13]. The results had also shown that $B\nu_0$ and $C\nu_0{}^2$ values are close. It indicates that square component of quenching is approximately equal to its linear component and can not be neglected while describing the quenching process in a track. We have determined the average volume, which falls at one pair of polarons as $V_{pair} = V/N_0$, where V is the track volume, $N_0 = E_i/\Omega$ is the primary number of pairs of polaron (E_i and Ω are the same as in (3)). The cube root of V_{pair}-value gives the estimation of the mean distance between polarons. Taking into account that a polaron pair is formed on a distances no longer than $r_C \sim 13\,nm$, the value D_{rel} was calculated as:

$$D_{rel} \equiv \sqrt{V_{pair}}/r_c. \quad (10)$$

For $D_{rel} \leq 1$ charge states arise at an average distance required for their prompt effective recombination. For $D_{rel} > 1$ the random walk of polarons has to forego the polaron pair formation and recombination. Figs.4 and 5 demonstrate calculated values of D_{rel} versus ν_0 for stilbene and p-terphenyl scintillators, correspondingly. The range of ν_0-values corresponding to the energies E_α of monochromatic alpha excitation, those were used in our experiments (from 0.96 to 5.32 MeV), is indicated by vertical solid lines. The excitation by the neutron with energy En results in generation of recoil protons with energies E_p in the range from 0 to E_n. For $E_p \leq E_n$ with E_p decrease the ν_0-values increase, according to the above-mentioned. In our studies we used the well-known and verified energies of the neutron spec-

trum of $^{239}Pu - Be$ radionuclide source, namely, 3.1, 4.2, 4.9, 6.4, 6.7, 7.3, 7.9, 8.6 and $9.7\,MeV$. The vertical dotted lines indicate the lowest ν_0-value for excitation with $E_n = 3.1$, 4.2, 4.9 and $9.7\,MeV$. In the case of neutron excitation the possible D_{rel} values are in the hatched areas (Figs.4 and 5).
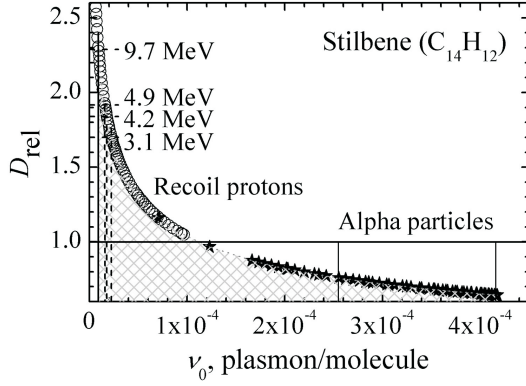

Fig.4. *Values of D_{rel} (10) versus n0 for stilbene*
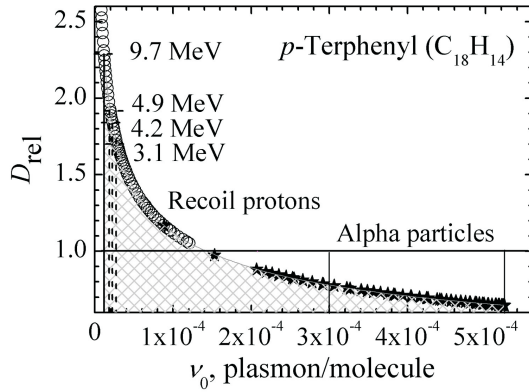

Fig.5. *Values of D_{rel} (10) versus ν_0 for p-terphenyl*

The results of D_{rel} calculation (Figs.4 and 5) demonstrate that for alpha excitation in the considered energy range the instant recombination of polaron pairs have to take place ($D_{rel} < 1$). For neutron excitation situation is mixed: for a fraction of polarons the instant recombination also takes place ($D_{rel} \leq 1$) and for another fraction random walk of polarons foregoes the recombination ($D_{rel} > 1$). So, the different mechanisms of recombination of polaron pairs can be realised for neutron and alpha excitation. It means that A, B, C-values have to be different for alpha and neutron excitation. According to (7), the denominator of (8) multiplied on A characterizes the primary recombination of polarons and the quenching effect connected with it. For the same scintillator excited by i-type of ionizing radiation $S_{XYi} = const/A_i$ (see (9)). The values $B_i\nu_0/S_{XYi}$ and $C_i\nu_0^2/S_{XYi}$Cin02/SXYi take into account the different nature of the processes those for alpha and neutron excitation condition the A, B, C-values and characterize the primary quenching effect in the track. The values $B\nu_0/S_{XY}$ and

$C\nu_0^2/S_{XY}$ versus ν_0 are presented in Figs.6 and 7, respectively. One can see that for alpha excitation values $B\nu_0/S_{XY}$ and $C\nu_0^2/S_{XY}$ is higher than for neutron excitation; it means that quenching for alpha excitation is higher. It is in a good agreement with obtained $\zeta_{\alpha\gamma}$-and $\zeta_{n\gamma}$-values (Fig.2). With ν_0 growth $B\nu_0/S_{XY}$ and $C\nu_0^2/S_{XY}$ increase and, consequently, the quenching effect intensifies. It proves that quenching in a track of an ionizing particle is the concentration-controlled process for organic scintillators. Fig.6 and Fig.7 demonstrates that with ν_0-value increase by an order of magnitude values $B\nu_0/S_{XY}$ and $C\nu_0^2/S_{XY}$ become by two or three orders of magnitude greater. It evidences the nonlinear quenching effect in track with ν_0-value growth.
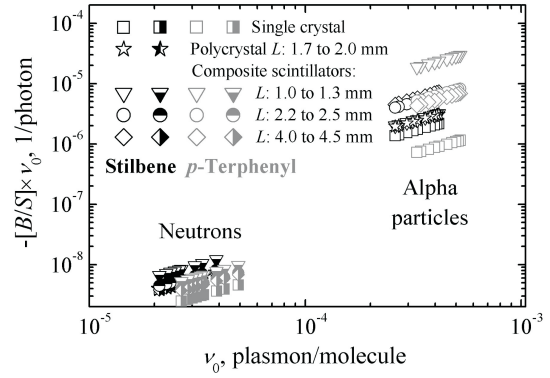

Fig.6. *Values $B\nu_0/S_{XY}$ for stilbene (black symbols) and p-terphenyl (grey symbols) scintillators. Half filled symbols correspond to neutron excitation, while open symbols correspond to alpha excitation*
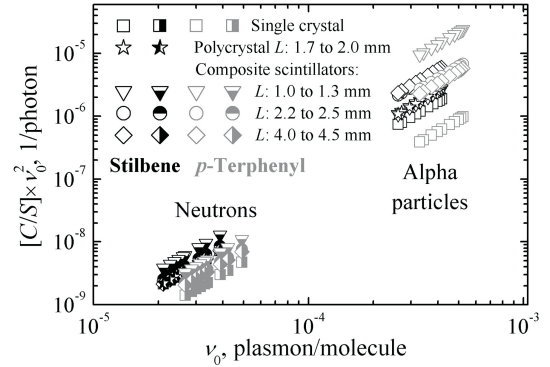

Fig.7. *Values $C\nu_0^2/S_{XY}$ for stilbene and p-terphenyl scintillators. The symbols are the same as in Fig.6*

The obtained results also testify that during the time no longer than $10^{-13}\,s$ the probability of instant ("one step") primary recombination of polaron pairs increases with growing ν_0. In [14] we have obtained that for the energy ranges that we used in our experiments the times of track formation along its full length t are about $10^{-12}\,s$ for alpha particles and $10^{-11}\,s$ for protons. It means that track formation along its full length is considerably longer process than the primary recombination and thermalization

of charge states in the track. The analysis which we carried out has shown that it is reasonable to divide the quenching in a track into primary and secondary one [14]. The process of the primary quenching is determined by the recombination of charges, which arise on a distance $r \leq r_C$ after plasmon dissociation. This primary recombination occurs during the time no longer than $10^{-13}\,s$, i.e. faster than diffusion expansion of a track at the distance comparable with the mean distance between neighbouring molecules and than track formation along its full length. Thus, the primary recombination of "hot" charge pairs, which cause the primary quenching, accompanies the process of track formation and runs in a track with practically constant cross-section. It promotes equalization of charge concentration in the track because it is more effective with the concentration increasing. Simultaneously, it determines the number of high-energy secondary electrons (δ-electrons) those can leave the track volume and form the region of low activation density around it because the probability of δ-electron recombination in the track area increases with growing ν_0. So, it defines the future processes of formation both the fast and the slow component of a scintillation pulse [14]. The results we obtained have shown that the primary quenching is the concentration-controlled process for organic scintillators [13, 14]. The recombination of polarons, which arise on a distance $r > r_C$ after plasmon dissociation and have to approach each other at first by random walk, makes the contribution in the secondary quenching. It occurs when the primary quenching is over. As it follows from the data presented in scientific literature, the secondary quenching is the diffusion-controlled process and runs in the expanding track that has cooled. Experimental results have shown that it is essentially less effective than the primary quenching in the context of the energy loss [14].

4. CONCLUSIONS

We have reviewed the process of primary energy exchange and quenching occurring in an ionizing particle track in early moments after excitation, taking into account the influence of polarization interaction [13, 14, 29]. On the base of data presented in scientific literature and summarising the results of our studies we have formulated the improved model description of the scintillation process in organic condensed media that presented in diagram form in [14]. The fundamental points of the model description are the following. Under interaction of ionizing radiation with a matter of an organic molecular solid scintillator the primary states - plasmons - are generated [2, 3]. For time intervals of $10^{-17}...10^{-16}\,s$ the pairs of charge states result from plasmons dissociation [2, 3, 18]. The polarization surrounding is formed around each charged state for $10^{-16}...10^{-15}\,s$ as a result of electronic polarization of neighbouring molecules, i.e. polarons arise [21, 22, 24]. The recombination of polarons that are formed around the charged states

of opposite signs can bring both to molecular excitation (that can give a quantum of luminescence) or to quenching (energy loss for heat, radiochemical reactions, etc.). The results of our investigations have shown that the "specific quenching" has two stages: the primary (concentration-controlled) quenching, occurring faster than diffusion expansion of a track and its formation along the full length, and the secondary (diffusion-controlled) quenching. With the initial concentration of the primary states ν_0 increase, the fraction of polarons, which take part in the primary recombination, becomes higher and cause more intensive the quenching effect. It remains uncleared why the quenching effect increase with growth of ν_0, i.e. with growth of the fraction of polarons, which take part in the primary recombination. It is going to be the subject of our future investigations.

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ОРГАНИЧЕСКИЕ КРИСТАЛЛИЧЕСКИЕ СЦИНТИЛЛЯЦИОННЫЕ ДЕТЕКТОРЫ: ПРОЦЕССЫ ТУШЕНИЯ В ТРЕКЕ ЧАСТИЦЫ

Н.З. Галунов, В.П. Семиноженко, Е.В. Мартыненко, О.А. Тарасенко

Дан краткий обзор процессов первичного размена энергии ионизирующего излучения в трековых областях для органических молекулярных сцинтилляционных кристаллических материалов. Обсуждается полуэмпирическое описание процессов первичного размена энергии ионизирующего излучения, учитывающее процесс первичного тушения в треке. Параметры этого описания характеризуют эффект первичного тушения. Уточненная модель сцинтилляционного процесса принимает во внимание влияние поляризационного взаимодействия между избыточными носителями заряда и молекулами вещества в треке.

ОРГАНІЧНІ КРИСТАЛІЧНІ СЦИНТИЛЯЦІЙНІ ДЕТЕКТОРИ: ПРОЦЕСИ ГАСІННЯ В ТРЕКУ ЧАСТИНКИ

М.З. Галунов, В.П. Семиноженко, Є.В. Мартиненко, О.А. Тарасенко

Представлено короткий огляд процесів первинного розміну енергії іонізуючого випромінювання в трекових ділянках для органічних молекулярних сцинтиляційних кристалічних матеріалів. Обговорюється напівемпіричний опис процесів первинного розміну енергії іонізуючого випромінювання, що враховує процес первинного гасіння в треку. Параметри цього опису характеризують ефект первинного гасіння. Уточнена модель сцинтиляційного процесу бере до уваги вплив поляризаційної взаємодії між надлишковими носіями заряду та молекулами речовини в треку.