

Semi-empirical description of quenching processes in a track of ionizing particles for organic crystalline scintillators

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The semi-empirical description of the process of quenching in the track that takes into account the influence of polarization of molecules in the track by charge states is proposed. The values of light yield were measured for alpha-, neutron- and gamma-excitation of $\varnothing 25\text{mm}\times 20\text{mm}$ stilbene single crystal and the $\varnothing 30\text{mm}\times 20\text{mm}$ composite scintillators on the base of stilbene (unary system), and *p*-terphenyl doped by 0.1% 1,4-diphenylbutadiene-1,3 of (binary system). The description obtained is in a good agreement with results of light yield experimental data.

Предлагается полуэмпирическое описание процесса тушения в треке, учитывающее возможность эффекта поляризации зарядовыми состояниями молекул в треке. Для монокристалла стиблена $\varnothing 25\text{мм}\times 20\text{мм}$ и композиционных сцинтилляторов $\varnothing 30\text{мм}\times 20\text{мм}$ на основе стиблена (однокомпонентная система) и *n*-терфенила, активированного 0.1% 1,4-дифенилбутадиеном-1,3 (двухкомпонентная система) измерены величины светового выхода при альфа-, нейтронном- и гамма- возбуждении. Полученное описание хорошо согласуется с экспериментальными данными по световому выходу.

The back scattering of charge particles from organic materials due to low effective atomic number of these materials is negligible. Organic scintillators are hydrogen-bearing materials and therefore give the best results in fast neutron spectroscopy. A fast neutron produces recoil protons in such an organic material with its highest energy equal to that of the neutron. That is why the organic scintillators and detectors based thereon are the most effective for detecting short-range (α - and β -particles) ionizing radiation, as well as for spectrometry of fast neutrons [1].

A proton or alpha particle loses its energy within its track to produce the high concentration of plasmons ν_0 , which results in generation of charge states [2]. The recombination of charge states may result in (i) molecular excitation and therefore in luminescence, or in (ii) non-radiative recombination. It is well known from 1960's that the quenching losses in the track of an *i*-type ionizing particle with energy E_i causes non-linear dependence of scintillation light yield (LY) vs the E_i value, and that the LY vs E_i dependences are not the same for different *i* [1, 3, 4]. This effect is referred to as "specific quenching". To describe the "specific quenching", Birks had proposed a hypothesis of "unimolecular" quenching in an ionizing particle track [1]. This hypothesis, however, does not explain the "specific quenching" process in organic scintillators (see details in [5]). If a high value of ν_0 causes the "specific quenching" effect and therefore results in non-linear scintillation response, then the decrease of ν_0 can result from the following three causes. Those are (i) the

exchange interaction between charge states (the recombination), (ii) diffusion expansion of the particle track, and (iii) trapping of the charge states at some distance from the track in a material with reduced transport of charge states. To date, there is even no answer on the question. What the process mainly influence the ν_0 decrease? Thus, such an analysis is important and topical question.

In our previous works [6-8], we have described the composite scintillators based on stilbene and *p*-terphenyl as a new type of organic scintillation systems. The composite scintillators were prepared from crystalline grains obtained by grinding of a single crystal with high structural perfection at low temperature followed by sieving. These grains were introduced into an optically transparent glue. The organic single crystals and composite scintillators based on the same scintillation material but differ dramatically in their structure perfection are the good objects to investigate the effect of scintillator structure perfection on the “specific quenching”. In this work, considered is the semi-empirical description of LY_i vs ν_{i0} under alpha or fast neutron excitation ($i = \alpha, n$) of organic crystal scintillators and comparison of that description with results of light yield measurements.

Plasmons are the primary states resulting from action of ionizing radiation on organic molecular solid scintillators. The energy of such a plasmon for organic crystals is $\Omega \sim 20$ eV [2]. These primary states will dissociate into a substantially the same number of coupled charge states [9]. Therefore, as the particle energy E rises, the number of primary states will increase as E/Ω . To obtain the concentration of primary states in the track, it is necessary to divide E/Ω by the track volume V . For the instant case, a track of a proton or an alpha particle may be approximated by zones with cylindrical symmetry with effective cross-section radius r_0 and height that is equal to the particle path length [2-5, 9, 10]. We took the r_0 values from the experimental data cited in [9]. The data on the path of a particle with energy E_i in a solid mixed material containing hydrogen (H) and carbon (C) (e.g. stilbene (C₁₄H₁₂) and *p*-terphenyl (C₁₈H₁₄)) are taken from reference book [11]. If N is the number of the molecules per cubic centimeter, then Eq. (1) gives the concentration of primary states being a small value as compared to unity:

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

Fig. 1 shows the calculated ν_{i0} values vs E_i for stilbene and *p*-terphenyl single crystals excited with protons and alpha-particles. The ν_{i0} value in (1) decreases with E_i increase (see Fig. 1) because of fast growth of the particle path (and therefore the increasing track volume V) [11].

Silinsh et al. [12] were the first who have shown a prime influence of polarization effects on the charge state formation, transport, and recombination in organic molecular crystals. According to them, the average time necessary for charge carriers to make the random “jump” from one molecule to another in organic crystals is about 10^{-12} s. This value of charge state random walk can be considered as the majorization of the time necessary for a particle track to expand on one molecular layer in the diffusion process. The time necessary for electronic polarization of neighboring molecules varies from 10^{-16} to 10^{-15} s. If a certain molecule loses an electron and becomes M_{p+} state, then this electron has to be localized on another molecule of a molecular dielectric such as an organic scin-

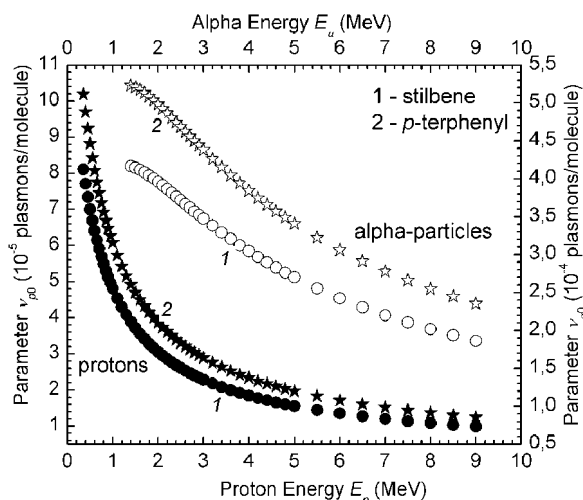


Fig. 1. Calculated primary concentration ν_{i0} (1) of plasmons vs the particle energy E_i for stilbene and *p*-terphenyl single crystals excited with protons and alpha-particles.

tillation crystal. This results in the M_{p^-} formation. The M_{p^+} state and M_{p^-} state are surrounded by unionized molecules, which concentration is $1 - v_{i0}$ at the moment of excitation ($t = 0$). The molecular π -orbitals of neighboring molecules have negative charge. Therefore, the molecular quasi-ion for the case of M_{p^+} state attracts those while the molecular quasi-ion of the M_{p^-} state repels. A pair of polarons arises. The calculations made in [12] showed that about 7000 molecules around such a molecular quasi-ion are polarized. Therefore, up to a distance r_C , which is about 13-16 nm for organic crystals [12], these two polaron states behave like a bipolaron (so-called CP-state), i.e. the state that precedes and promotes the recombination, which results in molecular excitation or quenching. The average radius of a 5.5 MeV alpha-particle track is about 50 nm [9]. This means that we have to estimate the r_C as a large value. It should also be stated that for organic molecular crystals, the energy of polarization interactions ($\sim 10^{-1}$ eV) exceeds by two orders of magnitude the energy of Van der Waals interactions ($\sim 10^{-3}$ eV), which hold the molecules in crystal [2, 9, 12]. Thus, the primary quenching seems to be a rapid and almost single-step process.

So, in contrast to the works of the 60s and 70s [1], we have to take into account the polarization effects to investigate the energy exchange processes in a particle track in organic condensed media. Polarons can localize at a small distance from each other and form CP- states. Polarons can localize at a large distance when the secondary electron that leaves a molecule has enough energy. In the latter case, the random path of polarons foregoes the CP- state formation.

We used a $\varnothing 25 \times 20$ mm² stilbene single crystal and 10 composite scintillators obtained from grains of stilbene (unary system), as well as 10 composite scintillators obtained from grains of *p*-terphenyl doped with 0.1% 1,4- diphenylbutadiene-1,3 (binary system). All the composite scintillators were $\varnothing 30 \times 20$ mm² size. The investigation included different grain size ranges with *L* values between 0.5 and 4.5 mm. In our experiments, we used a ²³⁹Pu-Be radionuclide source of fast neutrons and gamma radiation, ²³⁹Pu and ²⁴¹Am sources of alpha particles as well as ²²Na, ⁵⁷Co, ⁶⁰Co, ¹³⁷Cs, and ¹⁵²Eu sources of gamma radiation. To obtain different energies E_α of alpha excitation, we used the effect of alpha particle moderation in air. The reconstruction of a neutron spectrum from the corresponding recoil proton spectrum has been reported in [5, 7, 13] in detail.

Semi-empirical description of quenching in particle track. If, according to (1), the total concentration of molecules (ionized and unionized) is equal to 1, then the concentration of unionized molecules is $1 - v(t)$ and $1 - v_0$ for time $t > 0$ and $t = 0$, respectively. If $q(v, t)$ describes quenching in the track and *D* is the diffusion coefficient, then

$$\frac{dv(r, t)}{dt} = D \nabla v(r, t) - \{q(v, t)\} v(r, t) . \quad (2)$$

The quenching processes of *j* polaron pairs in each event define the second term in (2). This quenching may result from (i) recombination of *j* CP- states (rate parameter p_j) and (ii) recombination of *j* random polaron pairs (rate parameter c_j). Thus, the second term in (2) can be written as:

$$\begin{aligned} -\{q(v, t)\} v(r, t) &= -\{c_1 v(r, t) + p_1 [1 - v(r, t)] v(r, t) + c_2 v^2(r, t) + \\ &+ p_2 [1 - v(r, t)] v^2(r, t) + c_3 v^3(r, t) + p_3 [1 - v(r, t)] v^3(r, t) + \dots\} = \\ &= -(c_1 + p_1) \left\{ 1 + \frac{\sum_{j=1}^{\infty} (p_{j+1} + c_{j+1} - p_j) v^j(r, t)}{(c_1 + p_1)} \right\} v(r, t). \end{aligned} \quad (3)$$

So, $q(v, t)$ is represented as some expansion into a power series in terms of $v(t)$. The following cases are possible:

1. The quenching in the track is too weak. The diffusion expansion of the track becomes the main cause of decreasing concentration of excited and charged states therein. According to results of previous studies, we know that this case becomes possible when the primary quenching is practically over, i.e. from 50 ns after excitation on [5, 9].

2. The quenching in the track is not so strong and its influence on the decrease of the primary states concentration is comparable with the influence of the diffusion track expansion. In this case, the quenching process will be accompanied by an increase of the distance between primary states. The quenching process efficiency will decrease with increasing diffusion expansion of the track.

3. The primary quenching in a track is very efficient and rapid. During such a rapid quenching process, the space distribution of ν value will remain almost unchanged and variations in q with time will be negligible (i.e. $q(\nu, t) \approx q_0(\nu_0) = \text{const}$). The concentration of charge states in the track undergoes a “sudden change” due to rapid and effective primary quenching in recombination process of CP- states. This agrees with the above estimate that the time necessary for electron polarization is more than by three orders of magnitude shorter than the time of diffusive track expansion over one molecular layer. The experimental data prove as well that the “sudden change” of charge states concentration in the track is closest approach to the real situation. Really, as it was shown in [5], to estimate the effect of additional losses in the particle track for i -type of radiation, it is expedient to compare the results obtained for such a case with those obtained for the case when track is not formed (e.g. the case of gamma-excitation). The following value describes this effect [5]:

$$\zeta_{i,\gamma} = \left(\frac{M_i}{E_i} \right) \bigg/ \left(\frac{M_\gamma}{E_\gamma} \right) = \frac{E_\gamma}{E_i} \bigg|_{M_i=M_\gamma}, \quad (4)$$

where M_i and M_γ are measured amplitudes of scintillation pulses generated by i -th radiation type and by gamma-radiation with energies E_i and E_γ , respectively.

Fig. 2 shows $\zeta_{\alpha,\gamma}$ and $\zeta_{n,\gamma}$ values obtained for some stilbene and p -terphenyl based scintillators. These results are typical of all other scintillators. The difference in $\zeta_{i,\gamma}$ values for the same type of excitation for composite scintillators and the single crystal is negligible as compared to difference in $\zeta_{\alpha,\gamma}$ and $\zeta_{n,\gamma}$ values for the same scintillator. This shows that the influence of the scintillation material type and its structural perfection (therefore, the difference in D value), light collection, and reabsorption on the scintillation light yield is not as important as the influence of the primary quenching processes inside the ionizing particle track. Thus, the primary quenching in a particle track has to be a very efficient and rapid.

For “sudden change” of the charge states concentration in the track, it is reasonable to use instead of (2) the Eq. (5) which neglects the influence of track expansion on the space distribution of ν value and gives expansion into power series not for the function $q(\nu, t)$ but for $q_0(\nu_0) = \text{const}$:

$$\frac{d\nu(t)}{dt} \approx -\{q_0(\nu_0)\} \nu(t) \approx -A \left\{ 1 + B\nu_0 + C\nu_0^2 + F\nu_0^3 + \dots \right\} \nu(t). \quad (5)$$

It follows from (3) that due to polarization effect, all these constants (B, C, F , etc.) except $A > 0$ can be both positive and negative. For initial condition (1), the solution of (5) is:

$$\nu(t) = \nu_0 \exp(-q_0 t). \quad (6)$$

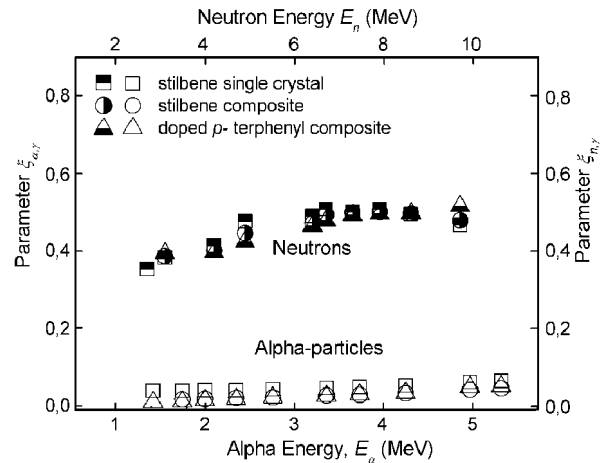


Fig. 2. $\zeta_{\alpha,\gamma}$ and $\zeta_{n,\gamma}$ values (lower and upper family of curves, respectively) as a function of the alpha and neutron excitation energy, respectively, for stilbene single crystal and for composite scintillators on the base of stilbene grains (L 1.5 to 1.7 mm) and doped p -terphenyl grains (L 1.0 to 2.0 mm).

Let us consider a binary crystalline scintillator that contains the base molecules of X type (donors of excitation energy) and the dopant molecules of Y type (acceptors of excitation energy). Let $\theta_{XY}(t)$ and κ denote the probabilities of non-radiative and radiative transport of the excitation energy from X to Y molecules, respectively. For molecules of l type (where l is X, or Y), let λ_l denote the rate constant of luminescence; C_l , the molecular concentration; $\xi_l(t)$, the concentration of excited molecules; $\eta_l \equiv \lambda_l + \chi_l$, the total (radiative plus non-radiative) losses of a molecule. For organic molecular scintillation crystals (and therefore for composite scintillators based on single crystal grains), the concentration of dopant molecules in melt does usually not exceed 1.0%, and their fraction that enters the crystal structure is about 0.1 [9]. Therefore, C_Y has to be about 10^{-5} to $10^{-3} C_X$. This allows us to neglect the direct excitation of Y molecules in the charge states recombination process or with ionizing radiation [2, 9]. Thus, we can consider that with constant rate ω the X molecules are excited in result of recombination of charge states, and Y molecules are excited in result of radiative and non - radiative transfer of excitation energy of X molecules. In addition, the low value of C_Y means that $\theta_{XY}(t) = \theta_{XY} = \text{const}$. Really, for organic solids, the number of the excitation energy donors changes according to the decay law [14]

$$n(t) = n_0 \exp\left(-\frac{t}{\tau_0} - 2q\sqrt{\frac{t}{\tau_0}}\right), \quad (7)$$

where n_0 is the number of excited molecules for the time $t=0$; τ_0 , the decay time constant of donors without energy transfer to acceptor; $q \approx 3,71R_0^3 N_Y$; R_0 , the Förster radius; and N_Y , the number of Y molecules (excitation energy acceptors) per unit volume. For systems under discussion, the q value in (7) does not exceed

$$q \approx 0.1065. \quad (8)$$

Then we get that for $t > 4q^2 \tau_0 = 1.86 \cdot 10^{-10}$ s, the expression (7) has the form

$$n(t) \approx n_0 \exp\left(-\frac{t}{\tau_0}\right). \quad (9)$$

This means that we can really neglect the time dependence of θ_{XY} to analyze the measurements of a scintillation signal made using a photomultiplier tube with collection time constant about $(1 - 2) \cdot 10^{-6}$ s in it anode integrating network circuit [1, 9].

The concentration of excited molecules is low as compared to the total concentration of molecules [5, 9]. Thus,

$$C_{Y-\xi_Y}(t) \approx C_Y. \quad (10)$$

Let the following α , β , and γ notations be introduced:

$$\alpha \equiv \eta_X + \theta_{XY} C_Y; \quad \beta = (\kappa + \theta_{XY}) C_Y; \quad \gamma \equiv \eta_X + (\kappa + \theta_{XY}) C_Y. \quad (11)$$

The following expressions describe the concentrations of excited molecules:

$$\frac{d\xi_X(t)}{dt} = \omega\nu(t) - \eta_X \xi_X(t) - \theta_{XY}(t) \times [C_Y - \xi_Y(t)] \xi_X(t) = \omega\nu(t) - \alpha \xi_X(t) \quad (12)$$

$$\frac{d\xi_Y(t)}{dt} = [\kappa + \theta_{XY}(t)] \times [C_Y - \xi_Y(t)] \xi_X(t) - \eta_Y \xi_Y(t) = \beta \xi_X(t) - \eta_Y \xi_Y(t) \quad (13)$$

with initial conditions:

$$\xi_X(0) = \xi_Y(0) = 0. \quad (14)$$

It should be noted that when the scintillation photons are taken into account, which X molecules emit and Y molecules absorb, then the concentration ($\tilde{\xi}_X$) of X type excited molecules those cause

the direct detection of their luminescence will be described as

$$\frac{d\tilde{\xi}_X(t)}{dt} = \omega\nu(t) - \eta_X \tilde{\xi}_X(t) - [\kappa + \Theta_{XY}(t)] \times [C_Y - \xi_Y(t)] \tilde{\xi}_X(t) = \omega\nu(t) - \gamma \tilde{\xi}_X(t). \quad (15)$$

Now let us consider expression (12) with initial condition (14) and the additional condition (10). We will use the notations (11). The equation (6) describes the function $\nu(t)$ with q_0 , (5). Therefore, the concentration of all excited X molecules will be

$$\xi_X(t) = \frac{\omega\nu_0}{q_0 - \alpha} \{ \exp(-\alpha t) - \exp(-q_0 t) \}. \quad (16)$$

To obtain the light yield, it is necessary to multiply λ_i by the number of excited molecules, which luminescence forms the scintillation signal. The integration time during which the measuring system collects all the scintillation photons will be taken as infinity [1, 9]. In such a case, the portion of light yield produced directly by molecules X will be equal to

$$LY_X \equiv \lambda_X \int_0^\infty \tilde{\xi}_X(t) dt = \lambda_X \int_0^\infty \left[\frac{\omega\nu_0}{q_0 - \gamma} \{ \exp(-\gamma t) - \exp(-q_0 t) \} \right] dt \quad (17)$$

or

$$LY_X = \frac{\lambda_X}{\gamma} \omega \frac{\nu_0}{q_0} = \frac{\lambda_X}{\eta_X + (\kappa + \Theta_{XY})C_Y} \omega \frac{\nu_0}{q_0} = S_X A \frac{\nu_0}{q_0} \quad (18)$$

where

$$S_X \equiv \frac{\lambda_X}{\eta_X + (\kappa + \Theta_{XY})C_Y} \frac{\omega}{A} \quad (19)$$

and the A value is taken from q_0 (5).

To obtain the description for luminescence of Y type molecules excited due to excitation energy transfer from X (donor) to Y (acceptor) molecules, let us consider expression (13) with initial condition (14) and take into account condition (10). We will use the notations (11). From (13), we get

$$\xi_Y(t) = \beta\omega\nu_0 \left\{ \frac{(q_0 - \alpha)\exp(-\eta_Y t) + (\alpha - \eta_Y)\exp(-q_0 t) - (q_0 - \eta_Y)\exp(-\alpha t)}{(q_0 - \alpha)(\alpha - \eta_Y)(q_0 - \eta_Y)} \right\} \quad (20)$$

and the portion of light yield produced by molecules Y will be equal to

$$LY_Y = \lambda_Y \int_0^\infty \xi_Y(t) dt = \frac{\lambda_Y \beta \omega \nu_0}{(q_0 - \alpha)(\alpha - \eta_Y)(q_0 - \eta_Y)} \int_0^\infty \{ (q_0 - \alpha)\exp(-\eta_Y t) + (\alpha - \eta_Y)\exp(-q_0 t) - (q_0 - \eta_Y)\exp(-\alpha t) \} dt$$

or

$$LY_Y = \lambda_Y \beta \omega \frac{\nu_0}{q_0} \left[\frac{1}{\eta_Y \alpha} \right] = \frac{\lambda_Y \beta}{\eta_Y \alpha} \omega \frac{\nu_0}{q_0} = \frac{\lambda_Y (\kappa + \Theta_{XY})C_Y}{\eta_Y (\eta_X + \Theta_{XY} C_Y)} \omega \frac{\nu_0}{q_0} = S_Y A \frac{\nu_0}{q_0}, \quad (21)$$

where

$$S_Y \equiv \lambda_Y \frac{\omega}{A} \left[\frac{(\kappa + \Theta_{XY})C_Y}{\eta_Y (\eta_X + \Theta_{XY} C_Y)} \right]. \quad (22)$$

Thus, the light yield of binary system with low concentration of Y molecules will be equal to

$$LY = LY_X + LY_Y = (S_X + S_Y) A \frac{v_0}{q_0} \equiv S_{XY} A \frac{v_0}{q_0}, \quad (23)$$

where

$$S_{XY} \equiv \left\{ \frac{\lambda_X}{\eta_X + (\kappa + \Theta_{XY}) C_Y} + \lambda_Y \frac{(\kappa + \Theta_{XY}) C_Y}{\eta_Y (\eta_X + \Theta_{XY} C_Y)} \right\} \frac{\omega}{A}, \quad (24)$$

and the A value is taken from (5). The substitution of q_0 (5) into (23) gives

$$LY_{XY} = \frac{S_{XY} v_0}{1 + B v_0 + C v_0^2 + F v_0^3 + \dots} \quad (25)$$

For excitation with low initial specific energy losses, like excitation with photons of gamma radiation of middle energies, when $v_0 \sim 1.86 \cdot 10^8$ plasmons per molecule, (i.e. when $1 \gg v_0 \gg v_0^2 \gg \dots$), it is easy to obtain from (25) that

$$LY_{XY} \approx S_{XY} v_0. \quad (26)$$

For such small v_0 values, a track does not appear and v_0 is practically linear function of excitation energy [3-5, 9]. Therefore, according to (26), the light yield has to be linear function of the excitation energy.

For unary system, i.e. for $C_Y = 0$, we get instead of (25)

$$S_{XY} = S \equiv \frac{\lambda \omega}{\eta A}. \quad (27)$$

Comparison with experimental results. The dependence of light yield vs Compton edge energy for excitation with gamma photons of middle energies (exceeding the energy of ^{57}Co) was linear. These results agree well with estimation (26). The dependences of light yield on the particle energy for neutron and alpha excitation were non-linear. Fig. 3 shows the results of approximation of experimental scintillation light yield values using (25). The following results are typical of all the scintillators. The accuracy of approximation (25) was lower than the value of corresponding constant in (25) only if $F = 0$. The combination of values Sv_0 , Bv_0 and Cv_0^2 provides the information on quenching effect. Fig. 3 shows the variation in Sv_0 , Bv_0 and Cv_0^2 values with excitation energy for the same scintillators as in Fig. 2. The Bv_0 and Cv_0^2 values show that the quenching effect is comparable for stilbene and p -terphenyl based scintillators. The ratio between Sv_0 values correlates with the light yield ratio for the same scintillators and shows a high quenching effect for alpha excitation as compared to neutron excitation (see Fig. 3).

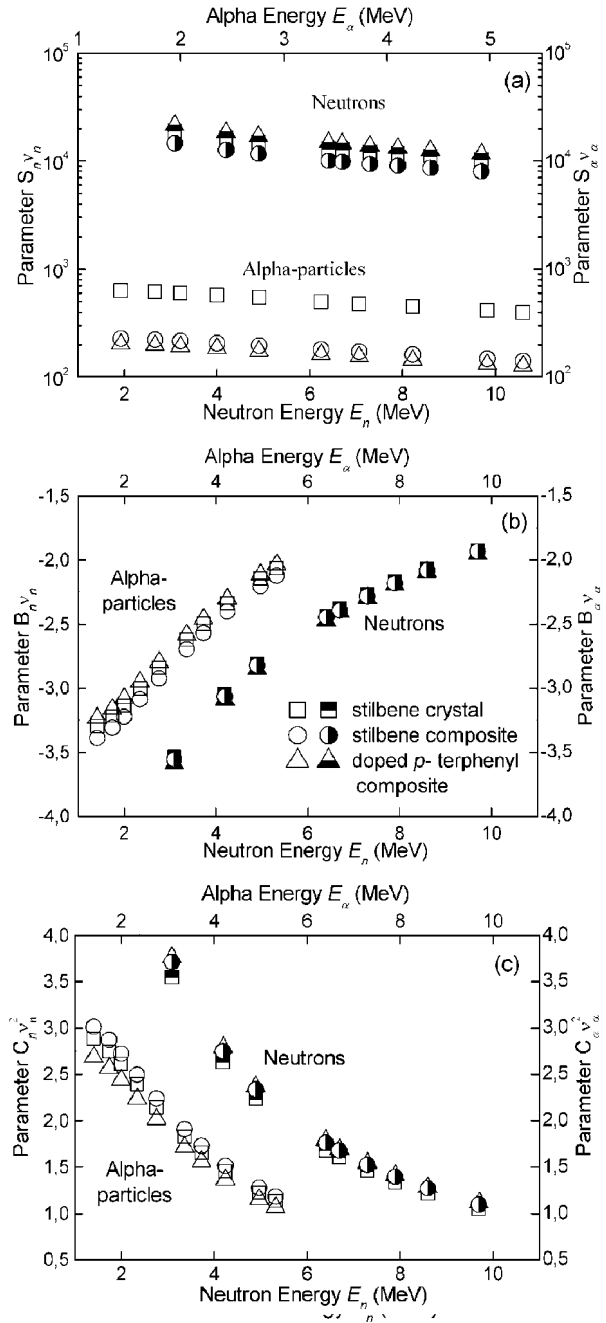


Fig. 3. Values of Sv_0 (a), Bv_0 (b) and Cv_0^2 (c) parameters against the excitation energy for the same organic scintillators as in Fig. 2.

To conclude, the semi-empirical description of the quenching process in the track is proposed. It takes into account the electron polarization processes in particle track promoting the rapid recombination between the charge states, which results either in molecular excitation or in energy losses (quenching). The measured light yield values and calculated excitation density in tracks of particles allow to estimate the quenching effect in the scintillators discussed. The semi-empirical description (25) of the quenching process in the track with nonzero S , B , and C constants forms a base for such the estimation. The results obtained agree well with previous data ([5, 6]) which indicate that the hypothesis of “unimolecular” quenching proposed by Birks [1] in the track of an ionizing particle does not describe the process of “specific quenching” for organic scintillators.

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Напівемпіричний опис процесів гасіння у треку іонізуючих частинок для органічних кристалічних сцинтиляторів

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Пропонується напівемпіричний опис процесу гасіння у треку, який враховує можливість ефекту поляризації зарядовими станами молекул у треку. Для монокристала стилібену $\varnothing 25$ мм \times 20 мм та композиційних сцинтиляторів $\varnothing 30$ мм \times 20 мм на основі стилібену (однокомпонентна система) та *n*-терфенілу, активованого 0.1% 1,4-дифенілбутадієном-1,3 (двокомпонентна система) виміряно величини світлового виходу при альфа-, нейтронному та гамма-збудженні. Отриманий опис добре узгоджується із експериментальними результатами для світлового виходу.