Functional Materials 14, No.2 (2007)

Study of the spectrum of scintillation amplitude generated by fast neutron in an organic scintillator

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Received November 27, 2006

The features of the organic scintillator light yield as a function of the ionizing radiation energy at high specific energy losses (dE/dx) taking fast neutrons as an example have been considered. The dependences of light yield on the neutron energy have been obtained in experiment for stilbene single crystal and the liquid scintillator (diisopropyl naphthalene + BPO, 5 g/l) under irradiation by ^{239}Pu–Be neutron source. A method of neutron spectra reconstruction basing on recoil proton spectra has been described and the obtained spectra for ^{239}Pu-Be source have been presented.

Рассматриваются особенности зависимости светового выхода органического сцинтиллятора от энергии ионизирующего излучения с большими удельными энергетическими потерями dE/dx на примере быстрых нейтронов. Экспериментально получены зависимости светового выхода монокристалла стильбена и жидкого сцинтиллятора (диизопропилнафталин + 5 г/л BPO) от энергии нейтрона при облучении нейтронами от источника ^{239}Pu–Be. Описана методика восстановления нейтронных спектров из спектров протонов отдачи и приведены полученные спектры источника ^{239}Pu–Be.

The hydrogen containing media and thus, first of all, organic scintillators, are the most efficient means for the fast neutron spectrometry [1, 2]. The fast neutrons generate in organic scintillators the recoil protons causing the scintillation pulses. When an organic scintillator is excited by ionizing particles with high specific energy losses (dE/dx), such as protons, its light yield depends non-linearly on the particle energy. This non-linearity of the scintillator light yield is due to the presence of quenching energy loss in the particle track [1, 2]. It is Birks who has proposed a semi-empirical expression for the relationship between the particle energy E and the number of scintillation photons L generated by such a particle. This description is based on assumption of unimolecular quenching character within the track [1]. In [3], a $L(E)$ dependence was obtained by Wright assuming the bimolecular quenching caused by interaction of neighboring ionized or excited molecules. However, it has been shown [4] that the Wright formula was derived incorrectly and thus cannot be used. Our results [5, 6] testify to bimolecular quenching for ionizing radiation with high specific energy losses (dE/dx), thus, the unimolecular approximation is inapplicable. That is why the semi-empirical relation by Birks for $L(E)$ is invalid, too. Thus, the problem of the scintillation response dependence on the ionizing particle energy requires a further study.

It is to note that there are essentially no separate sources emitting any specific single type of ionizing radiation, therefore, when a neutron source is used in experiment,the scintillator detects both neutrons and accompanying background γ photons. In [7], we have described the experimental

setup of our design and technique for selective detection of fast neutrons and γ radiation. The scintillation amplitude spectra for stilbene single crystal presented below have been obtained using that setup. As the scintillation amplitude spectra obtained under neutron irradiation of organic scintillating media are the recoil proton ones, the neutron spectrum is to be reconstructed prior to its study.

Let the function $\varphi(E_n)$ describes a spectrum of neutrons with energy values E_n. When neutrons are scattered elastically in a substance, the recoil nuclei are formed, the energy E_r thereof being related to the initial neutron energy E_n as [1, 8]

$$E_r(E_n) = \alpha \cdot E_n \cos^2\theta, \qquad (1)$$

where $\alpha = 4mM_r/(m + M_r)^2$; m is the neutron mass; M_r, the recoil nucleus mass; θ is the scatter angle; $0 \le \cos^2\theta \le 1$. It follows from Eq.(1) that for monoenergy neutrons, the spectrum of recoil nuclei is in proportion to

$$u(E_r) = \alpha \cdot \cos^2\theta. \qquad (2)$$

Differentiation of Eq.(1) gives us

$$dE_r = E_n \frac{du(E_r)}{dE_r} dE_r,$$

or

$$du(E_r) = \frac{dE_r}{E_n}. \qquad (3)$$

If the scintillator active substance is hydrogen, then $\alpha = 1$ and the recoil proton spectrum is formed (with energy values E_p). For hydrogen, the scattering in the mass center system is substantially isotropic up to $E \approx 20$ MeV [1, 8]. Taking into account that for isotropic scattering (for $E_n < 20$ MeV), the following expression is valid:

$$du(E_r) = u(E_r)dE_r,$$

we can get from (3):

$$u(E_p)dE_p = \begin{cases} \dfrac{dE_p}{E_n}, & E_p \le E_n; \\ 0, & E_p > E_n \end{cases}. \qquad (4)$$

Eq.(4) describes the differential spectrum of the recoil protons obtainable by measurements. If a fast neutron flow with the spectral energy distribution $\varphi(E_n)$ enters a hydrogen-containing medium with the neutron detection $\varepsilon(E_n)$, then, according to (4), we get for an infinitesimal energy range dE_n

$$u(E_p)dE_n = \frac{\varepsilon(E_n)\varphi(E_n)}{E_n} dE_n. \qquad (5)$$

By integrating Eq.(5) between E_p and E_{max}, we get

$$u(E_p) = \text{const} \int_{E_p}^{E_{\max}} \frac{\varepsilon(E_n)}{E_n} \varphi(E_n) dE_n. \qquad (6)$$

To reconstruct the neutron spectrum, Eq.(6) is to be differentiated with respect to E_p. In (6), the function under the integral sign as well as its upper limit are independent of E_p. It is only the derivative of the lower limit with respect to E_p that is nonzero:

$$du(E_p) = -\frac{\varepsilon(E_n)}{E_n} \varphi(E_n) dE_p \Big|_{E_p = E_n}$$

or the differential neutron spectrum can be written as

$$\varphi(E_n) = -\frac{E_n}{\varepsilon(E_n)} \times \frac{du(E_p)}{dE_p}\Big|_{E_p = E_n}. \qquad (7)$$

As noted above, the scintillation pulse amplitude L is a nonlinear function of the recoil proton energy [1,5]. The expression (7) does not contain items of the form $L(E_p)$ that should characterize the relationship between the recoil proton energy E_p and the scintillation response L caused by such a recoil proton. Therefore, the information on the scintillation response nonlinearity is comprised in $\varphi(E_n)$ (7). Thus, if a scintillator is irradiated by a neutron source having a spectrum $E_n(k)$ consisting of k known energies, then it is just the expression (7) that provides information on the $E_n(k)$ spectrum under account for the scintillation response nonlinearity. In other words, in the approach proposed, a neutron source with an at least partly known spectrum should be used to study the excitation features of the scintillation response to neutrons. It is just such an approach that provides the initial experimental data, the approximation of which makes it possible to obtain directly the information on the scintillation response nonlinearity.

The light yield is known to be a linear function of the gamma radiation energy E_γ. Therefore, if the analyzer scale is calibrated

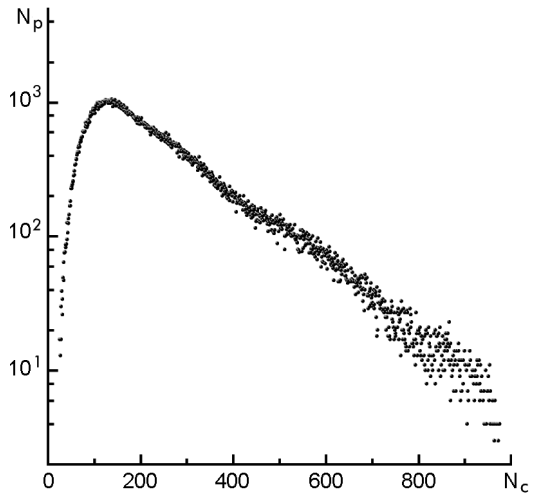

Fig. 1. A recoil proton spectrum for stilbene single crystal (dia. 40 mm, height 40 mm) under neutron irradiation from ^{239}Pu–Be source.

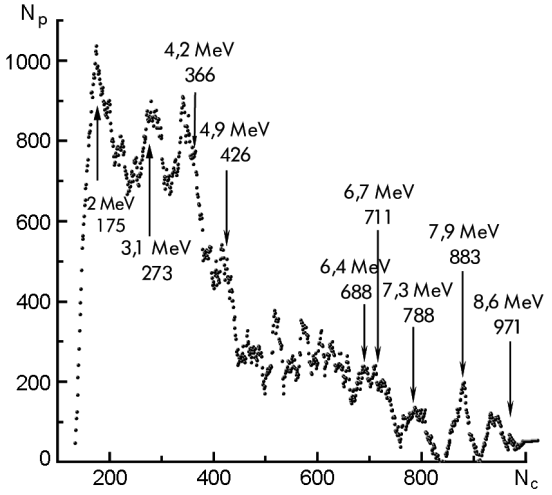

Fig. 2. The neutron spectrum for stilbene single crystal (dia. 40 mm, height 40 mm) reconstructed according to (7) for neutron irradiation from ^{239}Pu–Be source.

using a set of γ energies from separate radionuclide sources, the linear dependence obtained makes it possible to confront any channel number of the analyzer scale with a corresponding γ radiation energy. Using an attested scintillator with the known light yield in photons per 1 MeV of γ energy, the energy scale of the device can be calibrated in terms of scintillation light photons. If the apparatus conditions (the irradiation conditions, voltage, etc.) are the same for neutron measurements and γ radiation ones, the photon calibration of the scale obtained under γ irradiation will be valid for the neutron excitation, too.

Using the expression (7) let the neutron spectrum be reconstructed from the recoil proton one. Knowing the neutron peak energy, the channel number can be confronted with it and thus the number of photons in a scintillation pulse. This makes it possible to determine the set of $\varphi(E_n^j)$ values for $j = 1...k$. (Here, j is the neutron peak number in the spectrum; E_n^j, the energy corresponding to that peak). Thus, a correlation is obtained between the light signal and the neutron energy for k energy values. Since the law describing the light yield nonlinearity is unknown and is a quantity to be sought in the procedure under consideration, the values for intermediate energies between the known k ones are not defined uniquely. The corresponding light response in photons can be determined only for the known energies of neutron peaks. Using the calibration curve for γ radiation, it is possible to determine the γ radiation energy values E_γ corresponding to light signals identical to E_n^j neutron energies. Thus, one can find the corresponding E_γ and E_n energy values resulting in scintillation pulses of the same amplitudes. This correlation is just the scintillation response nonlinearity characteristic under excitation by recoil protons that is obtained directly from the experiment.

The following radionuclides were used in this work: ^{239}Pu-Be as the source of neutrons and γ radiation as well as ^{137}Cs, ^{60}Co, ^{152}Eu, and ^{22}Na as γ radiation sources. The studies were carried out using the above-mentioned setup and measurement procedure [7], taking a stilbene single crystal and a liquid scintillator as examples of two limiting cases, namely, a stable ordered structure and a structure where diffusion processes prevail [2]. The 40 mm high stilbene single crystal was 40 mm in diameter. The liquid scintillator on the diisopropyl naphthalene was deoxygenated and sealed in 40 mm high glass container of 30 mm in diameter. The recoil proton spectra were obtained for both scintillators under irradiation by fast neutrons from ^{239}Pu-Be source. Then, using the expression (7), the neutron spectra of ^{239}Pu-Be were reconstructed under account for the above note on the function (7) nonlinearity. Then, the experimental conditions remaining unchanged, the amplitude spectra were obtained for the same scintillators under γ irradiation from ^{137}Cs, ^{60}Co, ^{152}Eu, and

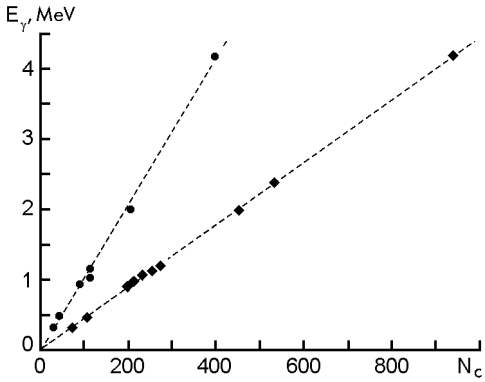

Fig. 3. Dependence of energy E_γ of γ radiation on the analyzer channel number N_c. Stars refer to the stilbene single crystal (dia. 40 mm, height 40 mm) and circles to the liquid scintillator (diisopropyl naphthalene + 5 g/l BPO sealed in a 40 mm high glass container of 30 mm in diameter)

^{22}Na sources. Using the obtained amplitude spectra, the channel numbers were found corresponding to the Compton edge energies. Knowing the later and the channel numbers corresponding thereto, the analyzer scale was calibrated. In the ^{239}Pu–Be spectrum, the neutron energy values were selected that have been studied well both theoretically and in experiment. Fig. 1 demonstrates the recoil proton spectrum obtained for the stilbene single crystal under irradiation by fast neutrons from ^{239}Pu-Be radionuclide source. Fig. 2 shows the reconstructed neutron spectrum for that source and the scintillator. Similar results have been obtained for the liquid scintillator. The multichannel analyzer channel width calibration for γ scintillations of the stilbene crystal and the liquid scintillator are shown in Fig. 3.

Then, the correspondence between E_γ and E_n energy values resulting in scintillation pulses with equal amplitudes in the same scintillator has been found. Fig. 4 shows the dependence of E_n on E_γ for the stilbene single crystal and the liquid scintillator. The only data concern E_n (E_γ) dependence for organic crystals, which were known before this paper, were the results of [11] by Broek and Anderson. In [11] one can find four pairs of values of E_n and E_γ. Each pair of E_n and E_γ values gives the energies of neutron and gamma excitation, which have to result in the equal scintillation amplitudes in a stilbene single crystal. These data are cited in many books (see e.g. [1,8]) and remain thus far unique. The stars on Fig. 4 show

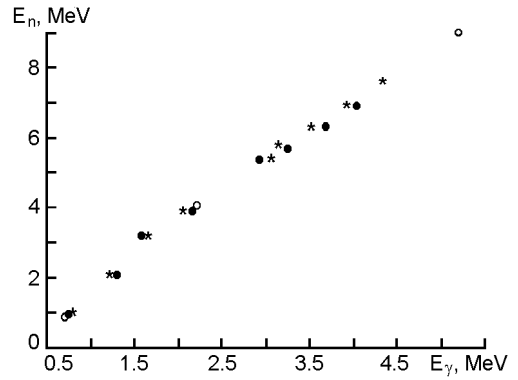

Fig. 4. $E_n(E_\gamma)$ dependence for the same stilbene single crystal (double crosses) and the liquid scintillator (circles) as on Fig. 3. Stars show the $E_n(E_\gamma)$ dependence that was obtained for stilbene single crystal by Broek H.W. and Anderson C.E. [11] (see e.g. [1]).

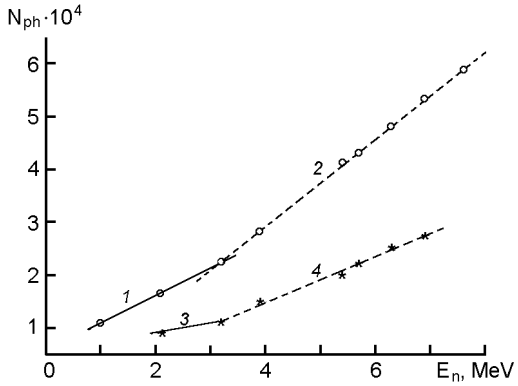

Fig. 5. Scintillator light yield as a function of neutron energy E_n under neutron irradiation from ^{239}Pu-Be source. Circles refer to the stilbene single crystal (dia. 40 mm, height 40 mm); double crosses, to the liquid scintillator (diisopropyl naphthalene + 5 g/l BPO sealed in a 40 mm high glass container of 30 mm in diameter). Points refer to experimental data; lines, to approximation thereof as N = A + B·E_n, where A = 297 photons, B = 5242 photons per MeV (*1*); A = 12319 photons, B = 8296 photons per MeV (*2*); A = 3489 photons, B = 1741 photons per MeV (*3*); A = 7383 photons, B = 4391 photons per MeV (*4*).

the E_n (E_γ) dependence for the stilbene crystal taking from [11]. One can see that our results and results of [11] are in a good agreement. Fig. 5 presents the light yield dependences on the neutron energy E_n under neutron irradiation from the ^{239}Pu–Be source for both scintillators. The experimental points can be approximated by two linear dependences of $N = A + BE_n$ kind. In the con-

text of the problem under consideration in this work, it is only the B values in similar formal approximations that are of importance. Those values provide a qualitative conception of the averaged effect of quenching processes in the tracks of protons generated by neutrons of a specific energy. Increasing B parameter evidences a weakening effect of quenching processes in the tracks for a specific spectrum of recoil protons. The slope of curves *2* and *4* is higher than that of *1* and *3* ones, thus demonstrating clearly the nonlinear increase of the scintillator light yield with increasing neutron energy. Such character of the dependences can be explained by the fact that as the neutron energy decreases (and thus the average energy of the generated recoil protons), the importance of the quenching processes in the track increases, the energy loss for quenching rises and thus the light yield drops.

Thus, a neutron spectrum reconstruction procedure from the experimental recoil proton spectra has been described. Its potential has been demonstrated taking a stilbene single crystal and a liquid scintillator as examples. The obtained dependences of E_n on E_γ and of light yield on E_n provide an initial information to study the nonlinearity of processes occurring in particle tracks in organic scintillators.

Acknowledgments. Author thanks Prof.N.Z.Galunov and Dr.O.A.Tarasenko who assisted in discussion of results as well as Drs. S.V.Budakovsky and L.Sh.Afanasiadi who have offered the samples of stilbene and liquid scintillator.

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Дослідження спектра амплітуд сцинтиляцій, що генеруються швидкими нейтронами в органічному сцинтиляторі

Є.В.Мартиненко

Розглядаються особливості залежності світлового виходу органічного сцинтилятора від енергії іонізуючого випромінювання з великими питомими енергетичними втратами dE/dx на прикладі швидких нейтронів. Експериментально отримано залежності світлового виходу монокристала стильбену та рідкого сцинтилятора (діізопропілнафталін + 5 г/л ВРО) від енергії нейтрону при опроміненні нейтронами від джерела ^{239}Pu–Be. Описано методику відновлення нейтронних спектрів із спектрів протонів віддачі та наведено отримані спектри джерела ^{239}Pu-Be.