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Dependence of neutron-sensitive plastic scintillator's n/γ-discrimination capability on the shifter's concentration

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A number of polystyrene based plastic scintillators containing 30.0 wt. % 2,5-diphenyloxazole and various concentration of the shifter 9,10-diphenylanthracene (0.02–2.0 wt. %) was obtained. The spectral-luminescent and scintillation characteristics of the obtained scintillators are investigated. The threshold character of the dependence of the n/γ-discrimination parameter FOM on the concentration of 9,10-diphenylanthracene molecules in the plastic scintillator is shown. The value of the triplet excitation energy diffusion coefficient is estimated as $D \approx 7.6 \cdot 10^{-7}$ cm^2/s. A criterion for the optimal content of the shifter in a neutron-sensitive plastic scintillator is proposed.

Keywords: plastic scintillator, shifter, light yield, n/γ-discrimination.

Залежність n/γ-розділяючої здатності нейтроночутливого пластмасового сцинтилятору від концентрації шифтеру. *П.М.Жмурін, Д.А.Єлісєєв, О.В.Єлісєєва, В.Д.Алексєєв, Ю.О.Гуркаленко*

Отримано ряд пластмасових сцинтиляторів на основі полістиролу, що містить 30,0 мас. % 2,5-дифенілоксазолу і різну концентрацію шифтеру 9,10-дифенілантрацену (0,02–2,0 мас.%). Досліджено спектрально-люмінесцентні та сцинтиляційні характеристики отриманих зразків. Показано граничний характер залежності параметра n/γ-розділення FOM від концентрації молекул 9,10-дифенілантрацену в пластмасовому сцинтиляторі. Оцінено значення коефіцієнта дифузії енергії триплетного збудження $D \approx 7.6 \cdot 10^{-7}$ cm^2/s. Запропоновано критерій оптимального вмісту шифтеру в нейтроночутливому пластмасовому сцинтиляторі.

1. Introduction

The creation of detectors for fast neutrons registration remains one of the urgent tasks in the field of experiments in high energy physics. Since neutrons are electrically neutral, the main method of their registration is based on the registration of recoil protons, which arise when the energy of neutrons is scattered by hydrogen atoms. Therefore, organic media are best suited for fast neutrons registration due to the significant specific content of hydrogen atoms. But any neutron radiation is almost always accompanied by gamma radiation, which also contributes to the registered signal. Therefore, there is a problem to discriminate the signals created by neutrons and gamma quanta.

Methods of discrimination by the shape of the scintillation pulse have been proposed long ago and are successfully used in the creation of detectors based on liquid or crystalline organic media [1–3]. But liquid and crystalline media severely limit the range of possible detecting devices due to stringent fire and environmental requirements and the geometry factor of the detec-

tor. An organic medium that is able to satisfy these requirements is a plastic scintillator (PS). But traditional PSs based on polystyrene or polyvinyltoluene do not make it possible to discriminate the scintillation signal by the pulse shape. This is due to the fact that excited triplet states in a polystyrene base are effectively quenched by various types of dimers (as a rule, these are excimers), which are formed in a polymer medium [4, 5]. In systems with phenyl chromophores, such as benzene, toluene, xylenes, as well as polystyrene, for individual molecules (at a low concentration in solution), relatively long lifetimes of triplet states can be observed. However, with an increase in the concentration of chromophores due to mutual quenching, the lifetimes of triplet states are significantly reduced, sometimes up to complete disappearance of triplet radiation [6, 7]. Therefore, the lifetimes of the triplet states of polystyrene are comparable to the lifetimes of the singlet states, and no slow luminescence is observed in such system [8, 9].

In order to observe slow luminescence in a polymer medium, various kinds of additives are introduced into it, in which the lifetimes of triplet states significantly exceed the lifetimes of singlet excited states. Practice has shown that 2,5-diphenyloxazole (PPO) molecules are the most convenient for introduction into the polymer base of a plastic scintillator [10]. Their solubility in a polystyrene base reaches 40 wt. %. The triplet excited states created under the action of high-energy irradiation can migrate along PPO molecules, which leads to the possibility of triplet-triplet annihilation (TTA) with the conversion of the molecule to an excited singlet state and, as a consequence, to slow luminescence. Since the penetration depth of neutrons in a polymer medium is several centimeters, samples of appropriate sizes are required for effective neutron detection. In order to avoid reabsorption of its own radiation at such distances, shifter molecules are introduced into the composition of the PS. As a rule, 9,10-diphenylanthracene (DPA) is chosen as a shifter for neutron-sensitive PS [11, 12]. However, DPA molecules triplet states energy is lower than the triplet states of PPO and, in this case, Dexter energy transfer can proceed. In this way DPA can act as a quencher for the triplet excitation energy of PPO. This leads to a significant decrease in the discrimination properties of PS, which was demonstrated on example of liquid scintillators [13]. A slight increase in the concentration of shifter molecules led to a significant decrease in the n/γ-discrimination properties, but at the same time increased the light yield.

As a rule, for a standard PS the maximum light yield is a sufficient criterion for determining the optimal content of the shifter. However, for a neutron sensitive PS the n/γ-discrimination parameter and the light yield don't correlate with each other. For such scintillators the maximum discrimination capability is more important. Therefore, the aim of the work was to study the dependence of the PS's n/γ-discrimination capability on the shifter?s content.

2. Experimental

PS samples were obtained by the method of thermally initiated polymerization of styrene in bulk. For this, 0.5 wt. % of 1,6-hexanediol dimethacrylate, 30.0 wt. % of PPO and X wt. % of 9,10-diphenylanthracene (X = 0.02, 0.05, 0.1, 0.5, 1.0, 1.5 and 2.0 wt. %) were placed in glass ampoules. The contents of the ampoules were filled with an appropriate amount of styrene and purged with argon for 6 minutes. The polymerization was carried out at a temperature of 85°C for 96 hours and 48 hours at 125°C. Samples in the form of cylinders 16 mm in diameter and 10 mm in height were cut from the obtained blanks, after which they were polished to optical transparency.

The spectral-luminescence characteristics of the obtained samples were investigated at room temperature on a FluoroMax-4 spectrofluorometer (Horiba Jobin Yvon — Edison, USA).

The light yield of polystyrene scintillators was measured on a scintillation spectrometer. The device consisted of a Hamamatsu R1307 photomultiplier and a LeCrou 2249A charge converter. To measure the light yield, PS samples were mounted directly on the surface of the photomultiplier tube photocathode (optical contact was provided by means of an immersion liquid) and irradiated with a monoenergetic source of conversion electrons. Radionuclide Bi–207 was used as a source of electrons. The light yield was determined from the position of the maximum of the total absorption peak of conversion electrons with an energy of 976 keV. The measurement error in this case was 5–10 %.

The parameter of n/γ-discrimination FOM was determined by comparing the total

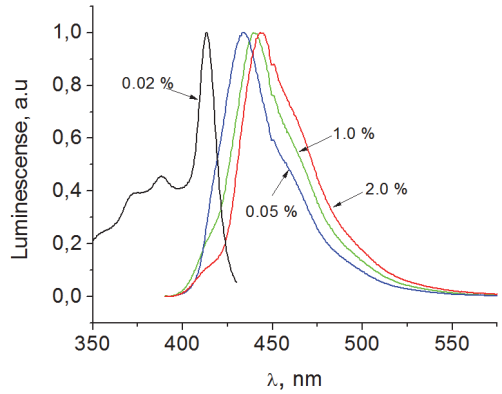

Fig. 1. Luminescence specter's of neutron-sensitive PS with different concentrations of DPA molecules.

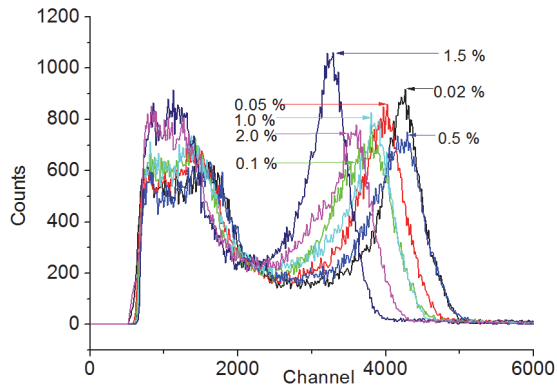

Fig. 2. Scintillation amplitude specter's of neutron-sensitive PS with different concentrations of DPA molecules.

charge of the signal (Q_{total}) and its ratio to the slow component (Q_{total}/Q_{slow}). For this PS's samples were irradiated with fast neutrons and γ-quanta from a plutonium-beryllium source (^{239}Pu–Be) through a lead plate 20 mm thick. After digital processing, the shape of the pulse and the area of the slow component (Q_{slow}) in the range from 50–70 to 200 ns and the entire pulse (Q_{total}) were stored in the source file for each detected pulse. The choice of the initial integration time point for Q_{slow} calculation was selected individually for each sample. Then, for each event, the ratio $D = Q_{slow}/Q_{total}$ was calculated and the distribution of the number of events over D was built, from which the FOM was determined. More detailed method described in the work [14].

3. Results and discussion

In the series of obtained PSs, a sample with the minimum concentration of the shifter (0.02 wt. %) has a luminescence spectrum specific for 9,10-diphenylanthracene with a maximum at 413 nm (Fig. 1). With an increase of DPA molecules concentration, the luminescence band maximum shifts to longer wavelengths diapason, which may indicate some aggregation of DPA molecules.

Table 1. Relative light yield (LY) and n/γ-discrimination parameter (FOM) of PSs with different contents of DPA

Concentration of DPA, wt. %	LY, %	FOM
UPS 923A [15]	100	–
0.02	99	1.31
0.05	94	1.313
0.1	88	1.25
0.5	100	1.36
1.0	91	1.365
1.5	77	1.13
2.0	84	1.04

However, light yield of the obtained PSs did not change significantly with a change in the concentration of DPA molecules (Fig. 2, Table 1). This suggests that even with an increase in concentration of DPA molecules by almost two orders of magnitude (taking into account the possibility of the formation of various types of aggregates), they perform their function of a shifter.

All obtained PS samples showed n/γ-discrimination properties with different levels of FOM values (Fig. 3, 4).

In contrast to the light yield, the dependence of n/γ-discrimination properties on the concentration of shifter in the obtained PS samples have an obvious threshold character (Fig. 4, Table 1).

Up to a concentration of 1.0 wt. %, with an increase in the content of DPA molecules in PS, a slight increase in FOM is observed. After reaching the DPA concentration threshold of 1.0 wt. %, there is abrupt decrease in the n/γ-discrimination capability of the PS (Fig. 4), which appear itself in a significant decrease in the part of slow luminescence, which is associated with TTA. Such a decrease in the part of slow luminescence in the polymer base of PS is apparently associated with the quenching of triplet excitation by DPA molecules, the excitation energy of triplet levels of which (1.77 eV) is much lower than the energy of excited triplet levels of PPO molecules

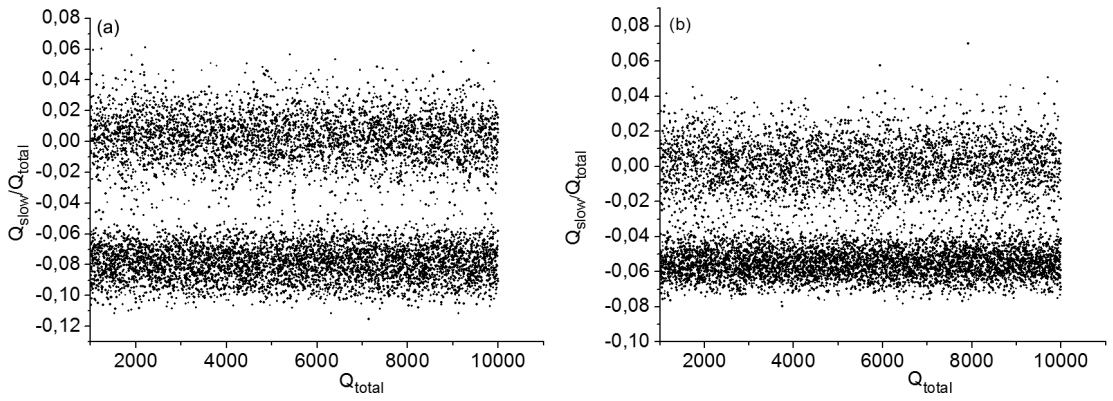

Fig. 3. Neutron/gamma discrimination of the obtained PS samples: (a) at a DPA concentration of 0.05 wt. %; (b) at a DPA concentration of 2 wt. %.

(2.47 eV) [15, 16]. If we assume that the transfer of the triplet excitation energy is possible only due to the exchange mechanism, then, as a consequence, it has the form of a jump from one molecule to another. The diffuse approximation is applicable to this character of the transfer of excitation energy. Taking the distance between DPA molecules at 1.0 wt. % as the limiting distance of transfer of the triplet excitation energy, the value of the diffusion coefficient (D) of the triplet excitation energy can be estimated by the formula $R^2 = D \cdot \tau$. Considering that at a DPA content of 1.0 wt. %, the distance between its molecules is about $R \approx 3.7$ nm, and the time of the observed delayed luminescence is about $\tau \approx 180$ ns, it turns out $D \approx 7.6 \cdot 10^{-7}$ cm^2/s. This value of the diffusion coefficient indicates a low migration rate of triplet excitation in the PS and determines the threshold character of the n/γ-discrimination properties. This threshold of the shifter concentration, at which its quenching properties are not yet manifested, will be the criterion for its optimal content in the PS.

4. Conclusions

The study of the dependence of the n/γ-discrimination properties of PS on the shifter concentration made it possible to study the dynamics of triplet excitation migration and estimate the value of the triplet excitation energy diffusion coefficient. Studies have shown that DPA molecules in a relatively large concentration range (up to 1.0 wt. %) are not quenchers of the triplet excited states in polymer base of neutron-sensitive PS. The threshold character of quenching is manifested due to the low value of the triplet excitation migration rate. The criterion for the optimal content of the shifter in the neutron-sensitive PS is the concentration threshold at which its quenching properties are not yet manifested.

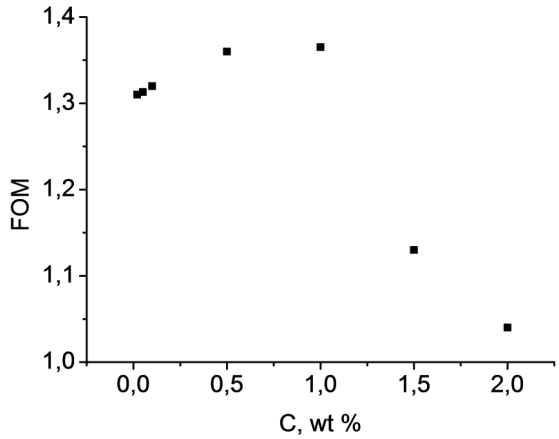

Fig. 4. Dependence of the n/γ-discrimination parameter FOM on the concentration of DPA molecules in PS.

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