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Light transmission through the material of composite scintillator

N.L.Karavaeva, E.V.Martynenko,
V.D.Panikarskaya, O.A.Tarasenko

Institute for Scintillation Materials, STC "Institute for Single Crystals", National
Academy of Sciences of Ukraine, 60 Lenin Ave., 61001 Kharkiv, Ukraine

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Single-layer and multilayer (20 mm high) composite scintillators based on stilbene were studied in the work. For all the scintillators the luminous transmittances on wavelengths of 360 nm, 390 nm, 440 nm and 700 nm have been measured. It was shown that the basic channel of light losses for investigated systems is not absorption by stilbene of its luminescence, but light scattering by grain boundaries. For the multilayer composite scintillators the scintillation response value increased with the grain size growth. This dependence is accounted for decrease of the amount of scattering facets on the path of light to a photodetector.

Исследованы однослойные и многослойные (высотой 20 мм) композиционные сцинтилляторы на основе стильбена. Для всех сцинтилляторов измерены коэффициенты пропускания света на длинах волн 360 нм, 390 нм, 440 нм и 700 нм. Показано, что основным каналом потерь света для исследуемых объектов является не поглощение стильбеном собственной люминесценции, а рассеяние света на границах гранул. Для многослойных композиционных сцинтилляторов наблюдался рост величины сцинтилляционного отклика с увеличением размера гранул, который объясняется уменьшением количества рассеивающих граней на пути света к фотоприёмнику.

1. Introduction

Organic molecular scintillators are very effective for detection of short-range charged particles (α- and β-radiations), and also in problems of spectroscopy of fast neutrons [1]. Recently the new class of organic scintillators, composite scintillators, has been developed [2–8]. The proposed composite material is a mosaic of the scintillation monocrystal grains introduced into the transparent polymeric matrix (a binder). A doubtless advantage of this scintillation system is that it allows to make detectors of any geometrical shape and with unlimitedly large detecting area, what is very important for detection of low fluxes of the most hazardous to human health ionising radiation (α-particles and fast neutrons) [2–8]. Depending on specification of a considered problem it is possible to use single- or multilayer composite scintillators. An increase in number of grain layers of the composite scintillator is necessary for enhancement of detection efficiency of the radiations having a big depth of penetration for the material of the scintillator. At the same time, transmission of light through such optical system will be diffusion in nature that can essentially influence on its coming to a photodetector. Composite scintillators are rather new class of scintillators and processes of light collection and peculiarities of light transmission in these systems have not been investigated yet, but these investigations are important for practical applications of the detector. Composite scintillators can find wide utilization in radioecological researches, radiobiology and medicine. The large area detectors for detection and spectrometry of fast neutrons, which at the same time can be applied as effective detectors of short-range radiations (such as α-particles), can be used for reliability enhancement of monitoring systems of nuclear power plants, enhancement of safety and improvement of a radioecological state of the atomic power stations and for control of unauthorized transportation of fissionable radioactive materials. Understanding of the processes defining properties of composite scintillators is a necessary element of their further development and perfection. Therefore, study of peculiarities of light transmission through the material of composite scintillator is the important task.

2. Experimental procedure

Composite scintillators were produced from crystalline grains, which were obtained by grinding of organic crystals at liquid nitrogen temperature. By means of a set of calibrated sieves crystalline grains were separated into fractions with a different size. The chosen fraction of grains was introduced into the transparent polymeric matrix (Sylgard-527). In the work there were studied: the series of composite scintillators based on stilbene of 30 mm in diameter and 20 mm high (grain size was varied in ranges: 1.0–1.3 mm; 1.3–1.5 mm; 1.5–1.7 mm; 1.7–2.0 mm; 2.0–2.2 mm; 2.2–2.5 mm; 2.5–3.0 mm; 3.0–3.5 mm; 3.5–4.0 mm; 4.0–4.5 mm) and the series of single-layer composite scintillators based on stilbene of 30 mm in diameter, whose height was determined by a size of grains (grain size was varied in ranges: 1.0–1.3 mm; 1.3–1.5 mm; 1.5–1.7 mm; 1.7–2.0 mm; 2.0–2.2 mm; 2.2–2.5 mm; 2.5–3.0 mm).

The following ionising radiation sources were used for investigation of the value of a scintillation response: $^{239}{}_{94}$Pu (α-particles with energy E_a = 5.15 MeV), $^{137}{}_{55}$Cs (photons of γ-radiation with energy E_γ = 0.662 MeV). The value of a scintillation response was determined from spectra of amplitudes of scintillations for all samples; the measurement error did not exceed 5 %. The scintillation amplitude spectra were obtained by means of multi-channel pulse-height analyzer AMA-03F. For calibration of the analyzer scale there were used: γ-radiation sources $^{137}{}_{55}$Cs (E_γ = 0.662 MeV), $^{60}{}_{27}$Co (E_γ = 1.173 MeV, E_γ = 1.333 MeV), $^{22}{}_{11}$Na (E_γ = 0.511 MeV, E_γ = 1.275 MeV) and the reference stilbene single crystal, light yield of which is known in photons per 1 MeV of γ-radiation energy.

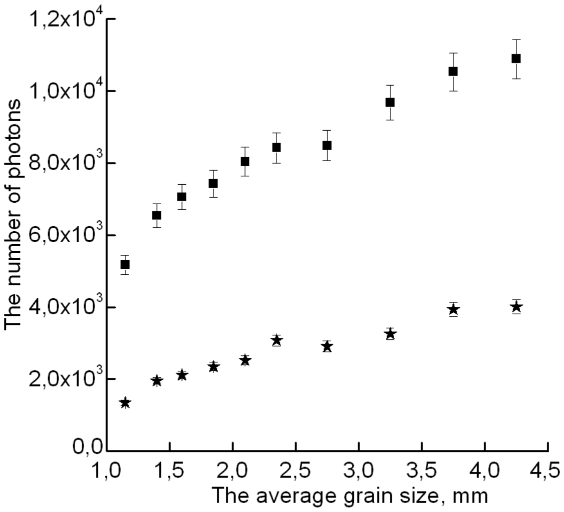

Fig. 1. Scintillation response of the multilayer (20 mm high) composite scintillators versus the average grain size. Stars – excitation by α-particles with energy E_γ = 4.97 MeV ($^{239}{}_{94}$Pu), squares — excitation by photons of γ-radiation with energy E_γ = 0.662 MeV ($^{137}{}_{55}$Cs).

It should be noted, that the source of α-particles $^{239}{}_{94}$Pu was in the collimator. Therefore, α-particles passed through a layer of air of width equal to the collimator width before hit a scintillator. As a result their energy was decreased. It has been calculated that for the collimator width used in the work energy of α-particles hitting a sample was 4.97 MeV.

Measurements of luminous transmittances were made by means of spectrometer "Hitachi 330" with an integrating sphere of 150 mm in diameter. The effective range of the sphere was 350–750 nm. The measurement error was 0.5 %.

3. Results and discussion

Fig. 1 shows dependence of the value of scintillation signal versus the average grain size for multilayer (20 mm high) composite scintillators. Measurements have been made for cases of excitation by α-particles and photons of γ-radiation and, as one can see from the figure, in the second case the scintillation signal was significantly higher. One of the reasons of such result may be that α-radiation is short-range. According to [1, 9], for α-particle with energy about 5 MeV the track length in organic single crystals is close to 30 μm.

We have calculated ranges of α-particle in stilbene for different energies of α-particle E_α in the range of $0.05 \text{ MeV} \le E_\alpha \le 10 \text{ MeV}$ in [10, 11]. The formula for range of a particle in substance with the complex atomic composition and data on ranges of α-particle in hydrogen and carbon, presented in the handbook [12], were used for the calculation. Dependence of value of α-particle range versus energy E_α, obtained as a result, is presented in [11]. The results of the calculations have shown that in the considered energy range (from 0.05 MeV to 10 MeV) the value of α-particle range in stilbene varies approximately from 0.001 mm to 0.09 mm. This value 1:3 orders of magnitude less than the size of crystalline grains from which the composite scintillators were made. According to the calculation results for energy of α-particle $E_\alpha = 4.97$ MeV used in the work the value of range in stilbene makes about 29 μm, that is in a good agreement with the data presented in [1, 9].

Thus, it is possible to state that under excitation of investigated composite scintillators by α-radiation with energies up to 10 MeV scintillation flashes always arise in the top layer of crystalline grains. Light have to pass a scintillator throughout its height to reach a photodetector what can lead to the strong attenuation of a scintillation signal. Under excitation by photons of γ-radiation, penetrating power of which is considerably higher, scintillation flashes arise over all volume of the scintillator.

Dependences of the value of the scintillation signal versus the average grain size for the single-layer and the multilayer (20 mm high) composite scintillators are given in Fig. 2. Scintillators were excited by α-particles with the same energy as during the measurements, results of which are presented in Fig. 1. As one can see from Fig. 1 and Fig. 2, scintillation signals of the multilayer scintillators increased with the average grain size growth. For the single-layer scintillators it was not observed any dependence of the scintillation signal versus the average grain size. The scintillation signals of the single-layer composite scintillators were essentially higher, than those for the multilayer (Fig. 2).

It is possible to explain such results by the fact that in a multilayer scintillator with an average grain size increase the amount of grains on the light path decreases and, hence, the amount of grain surfaces causing light scattering decreases. It should lead to decrease of number of scattering acts. Therefore light losses should be lower and, hence, the scintillation signal should be higher. For single-layer composite scintillators the light having got out a grain hits a photodetector without additional scattering. Therefore the value of scintillation response should be higher than for the multilayer scintillators, and should not depend on the grain size.

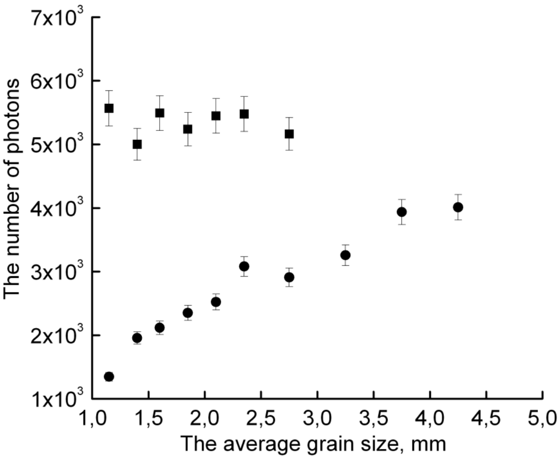

Fig. 2. Scintillation response of the single-layer (squares) and the multilayer (circles) composite scintillators versus the average grain size, measured under excitation by α-particles with energy $E_\alpha = 4.97$ MeV ($^{239}_{94}$Pu).

For all investigated scintillation samples luminous transmittances on wavelengths of 360 nm, 390 nm, 440 nm and 700 nm have been measured. These wavelengths have been chosen on the basis of analysis of excitation and luminescence spectra of stilbene single crystal [1]. In the area of wavelength λ = 360 nm the maximum light absorption should be observed. Wavelengths λ = 390 nm and 440 nm correspond to the maximum and droop of the luminescence spectrum of stilbene and therefore they have been chosen for simulation of transmission of photons of luminescence through a sample. Wavelength λ = 700 nm is in the transparent region and can be used for an estimation of influence of process of light scattering in the sample in absence of light absorption [1].

The results of measurements of luminous transmittances for the single-layer and the multilayer composite scintillators are presented in Fig. 3 and Fig. 4, respectively. The results of measurements of luminous transmittances for stilbene single crystals

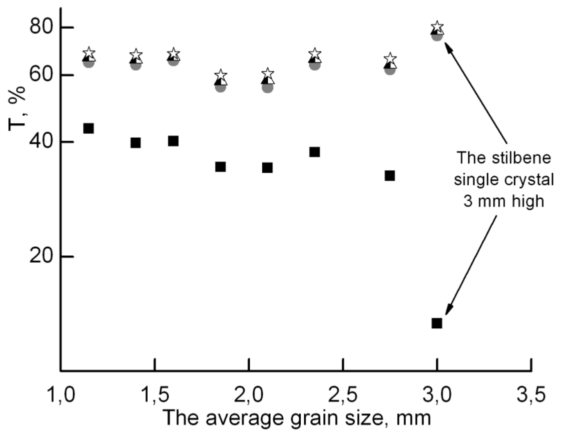

Fig. 3. Dependences of luminous transmittance versus average grain size for wavelengths of 360 nm (squares), 390 nm (circles), 440 nm (triangles), 700 nm (stars). For the single-layer composite scintillators.

of 3 mm and 20 mm high are also given at these figures for comparison.

As one can see from Fig. 3 for the single-layer scintillators on wavelength of 360 nm the value of luminous transmittance varies approximately from 30 % to 40 % while for the other wavelengths this value varies approximately from 60 % to 70 %. For single crystal of 3 mm high the luminous transmittance falls to 13 % on wavelength of 360 nm and reaches values about 80 % for the other wavelengths. The higher values of luminous transmittance on wavelength of 360 nm for single-layer composite scintillators, as compared with single crystal, can be explained as follows. Firstly, height of the single crystal, which has been chosen equal to the maximum grain size (3 mm), was larger than the average heights of the single-layer composite scintillators. Secondly, in the composite scintillators there are areas filled with the transparent polymeric matrix between crystalline grains, where there is no light absorption. Thus, the single-layer composite scintillators are characterized by the value of transmittance comparable with the one for the single crystal over the wavelength range, corresponding to their luminescence.

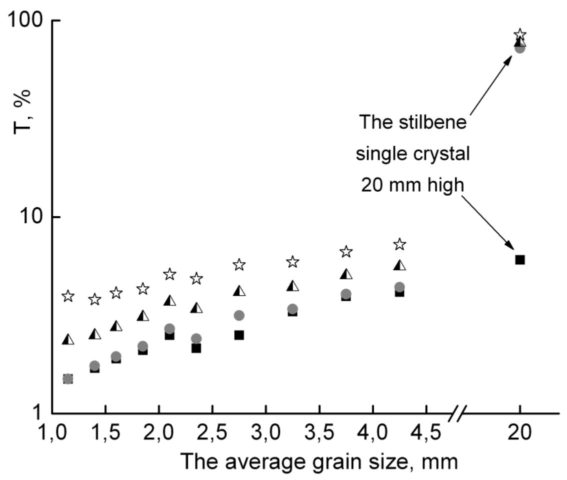

Fig. 4. Dependences of luminous transmittance versus average grain size for wavelengths of 360 nm (squares), 390 nm (circles), 440 nm (triangles), 700 nm (stars). For the multilayer composite scintillators.

The diffusion nature of light transmission through scintillator, predictably, leads to essentially higher losses of light for the multilayer (20 mm high) composite scintillators, than for the single-layer ones (Fig. 4). The luminous transmittances for the multilayer scintillators on all four wavelengths are extremely low and amount to a few percent. For the single crystal of 20 mm high the luminous transmittance on wavelength of 360 nm amount to about 6 %, and for the other wavelengths varies in the interval 72 %–85 %. Thus, the multilayer composite scintillators are characterized by essentially lower values of the luminous transmittances in comparison with a single crystal of the same height over the wavelength range, corresponding to their luminescence.

It is easy to note (Fig. 3 and 4), that for the single-layer scintillators no dependence of the value of luminous transmittance versus the average grain size is observed, while for the multilayer scintillators there is such dependence: with growth of the average grain size the luminous transmittance is also increases. For example, for average grain size of 1.15 mm the luminous transmittance on wavelength of 700 nm is about 4 %, and for the size of 4.25 mm it is about 7 %. Such results are in a good agreement with obtained dependences of the values of scintillation response versus the average grain size presented in Fig. 1, 2.

4. Conclusions

Thus, comparison of the measurement results of the luminous transmittances on wavelengths of 360 nm, 390 nm, 440 nm, 700 nm denote that the basic channel of light losses is not absorption by stilbene of its luminescence, but light scattering by grain boundaries.

Dependence of the value of scintillation response versus the grain size observed for multilayer composite scintillators is explained by growth of the amount of scattering facets on the path of light to a photodetector with grain size decrease. In such systems light scattering can has the defining influence on light coming out of a scintillator and result in essential decrease of scintillation signal. In the case of single-layer composite scintillators, light having got out a grain hits a photodetector without additional acts of scattering. Therefore, scattering, practically, does not influence on light coming to a photodetector and the scintillation signal of single-layer composite scintillators is considerably higher than the one for multilayer scintillators, and does not depend on the size of grains. Increase in the number of grain layers of composite scintillator is necessary for enhancement of detection efficiency of the radiations having a big depth of penetration for the scintillator material. For the detectors developed for detection of short-range radiations the single-layer constructions should be used.

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Проходження світла через матеріал композиційного сцинтилятора

Н.Л.Караваєва, Є.В.Мартиненко,
В.Д.Панікарська, О.А.Тарасенко

Досліджено одношарові і багатошарові (висотою 20 мм) композиційні сцинтилятори на основі стильбену. Для всіх сцинтиляторів виміряно коефіцієнти пропускання світла на довжинах хвиль 360 нм, 390 нм, 440 нм і 700 нм. Показано, що основним каналом втрат світла для об'єктів, що досліджено, є не поглинання стильбеном власної люмінесценції, а розсіювання світла на межах гранул. Для багатошарових композиційних сцинтиляторів спостерігалося зростання величини сцинтиляційного відгуку зі збільшенням розміру гранул, що пояснюється зменшенням кількості розсіюючих граней на шляху світла до фотоприймача.