Study of scintillation and optical properties of polycrystalline stilbene

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Received March 23, 2011

Scintillation properties of the stilbene polycrystals, which were made by the hot-pressure method from the powders of different sizes are studied. Influence of grain sizes of powders on light yield and optical transparency of polycrystals is investigated. It was found that grain size L of 2.2–3 mm is optimal for detection of short range radiation, such as alpha particles. The light yield in polycrystalline scintillators with optimal configuration was in the range of 50–75 % with respect to the reference stilbene single-crystal. It is also shown that decreasing of optical transparency is a basic factor that affects scintillation characteristics of crystals based on small grains.

Работа посвящена исследованию сцинтилляционных характеристик поликристаллов стильбена, полученных методом горячего прессования из порошков различной дисперсности. Изучено влияние размера зерен порошков на световой выход и оптическую прозрачность поликристаллов. Показано, что размер зерна порядка 2,2–3 мм является оптимальным для регистрации короткопробежных излучений при выбранной толщине образца. При этом удается получить световой выход поликристаллического сцинтиллятора 50–75 % относительно монокристалла. Также показано, что основным фактором, ухудшающим сцинтилляционные характеристики при относительно малых размерах зерен, является снижение оптической прозрачности.

1. Introduction

Organic scintillation materials are used in nuclear physics, radioecology, radiobiology, geology, etc. An extensive application of organic luminescent materials and detectors on their base for detection of ionizing radiations constantly stimulate a great interest in this problem. Organic scintillators are usually used as doped and pure single crystals, or as objects with amorphous structure (i.e. plastics and liquid scintillators). Radiation sensitive organic luminescent materials have to be fast and highly transparent for their own fluorescence. As a result of small atomic effective number organic molecular systems get the advantage over inorganic ones by detection of the short-range charged particles (alpha and beta particles). Organic polycrystals are very attractive systems for development of big in diameter detectors for detection of the most dangerous for human organism types of ionizing particles. The modern technologies of organic single crystal growth do not permit to create systems with large diametrical sizes (more than 100 mm) and high degree of structure perfection. Polycrystalline scintillators made by hot pressing of organic substances (e.g. stilbene) are promising materials for wide spectrum of applications [1–3].

An investigation of scintillation and spectral characteristics of organic polycrystals is very important not only for designing and creating new effective detectors but for understanding the influence of structural features of crystalline systems on their radioluminescence properties.

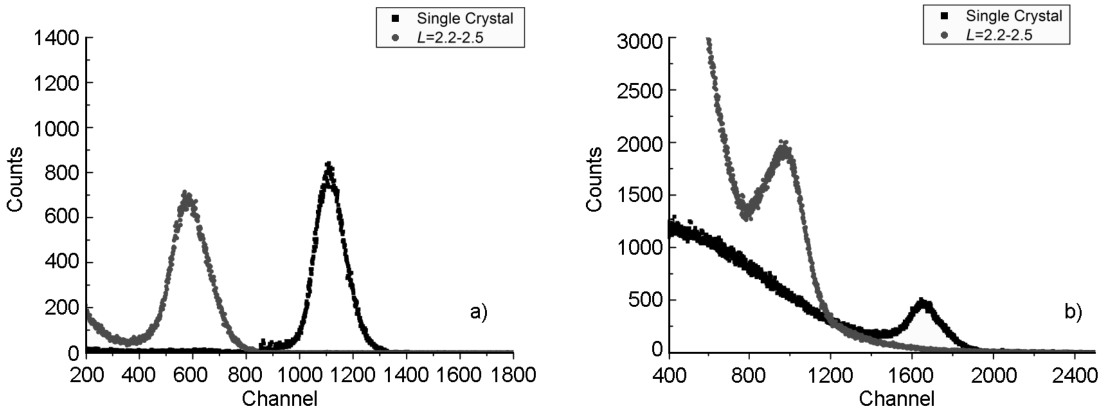

Fig. 1. Scintillation amplitude spectra for a stilbene single crystal and for a stilbene polycrystalline scintillator with grain size 2.2–2.5 mm excited by (a) alpha particles with energy E_α = 5.15 MeV (^{239}Pu radionuclide source) and (b) by beta particles with E_β = 0.624 MeV (^{137}Cs — radionuclide source).

Optical transparency is a significant factor affecting the light output when detecting short-range radiation. Incident radiation (for example alpha particles) is absorbed in a thin layer near the surface while the scintillation light is detected after transmission of the scintillation signal through this material.

This work is aimed to study the relationship between the grain size, optical transparency of the polycrystalline material, and its scintillation properties.

2. Experimental

The polycrystalline samples were obtained using pressing at high temperatures by single axle compression with the following slow cooling to room temperature. As a raw material we used thermally cracked single crystals of stilbene in liquid nitrogen. Different fractions of grains were obtained using the appropriate calibration sieves. By an average grain size in the selected fraction L_{av} we mean averaged characteristic which is equal to the difference between the openings of sieves j and i, where $d_i > d_j$

$$L_{av} = \frac{d_i + d_j}{2}. \quad (1)$$

Hereinafter the size of the grains in a fraction will be described either by this quantity or by a range of L from d_i to d_j.

The hot pressing was carried out at temperatures in the range $2/3T_m < T_p < 4/5T_m$, where T_m is a melting temperature of a raw material and T_p is a temperature that was fixed during pressing process. After heating of the form to the temperature of $2/3kT$, the pressure was slowly increased from normal to 400 MPa during 40 min, and then was held during 20–40 min. At the end of pressuring cycle the pressure was slowly (during 20 min) reduced to the normal. This mode of pressing leads to smoother changes of internal microstresses arising due to thermal or elastic anisotropy in the pressing process.

3. Results

We made comparative investigations of scintillation characteristics of ∅30 mm×5 mm stilbene polycrystals with the different grain size L, which were obtained by the above described method of hot-pressure. In this work we studied the series of 7 polycrystalline scintillators with different grain sizes. Samples had diameter of 30 mm and a height of 5 mm. The following fractions L were used: 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. We used the stilbene of the same dimensions (thickness 5 mm, 50 mm in diameter) as the reference single crystal.

For a comparative analysis of the light output of polycrystals we carried out measurements of their amplitude spectra using the following sources of ionizing radiation: ^{137}Cs (conversion electrons with the energy of 0.624 MeV) and ^{239}Pu (alpha particles with the energy of 5.15 MeV). R1307 Hamamatsu photomultiplier tube was used as a photodetector [5]. The results of the measurements are presented on the Fig. 1.

The relative light output of the samples can be calculated using the results of measurements of the amplitude spectra The results of relative light output J calculation for organic polycrystals are presented on Fig. 2. The measurement error did not ex-

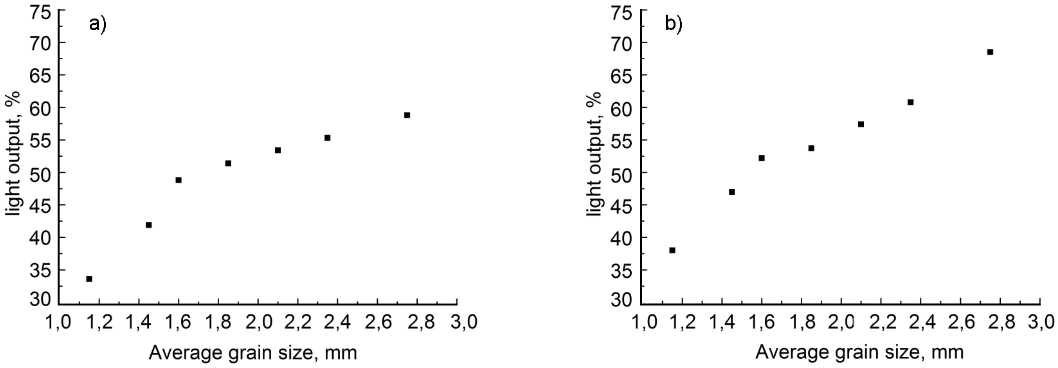

Fig. 2. The light yield versus the grain size for a stilbene polycrystalline scintillators during detection for excitation by (a) alpha particles with energy $E_\alpha = 5.15$ MeV (^{239}Pu — radionuclide source) and (b) beta radiation with $E_\beta = 0.624$ MeV (^{137}Cs — radionuclide source).

ceed the methodical error of light yield, which was equal to 5 % at confidence probability of 0.95.

During the studies aimed at establishing a correlation between the parameters of the pressing process and scintillation characteristics of organic polycrystalline stilbene we obtained important preliminary result. It allows to determine the influence of transparency (transmittance in the visible spectrum T,%) on the light output of polycrystalline samples excited by short-range radiation [4]. We used the spectrophotometer "Hitachi-330" to measure the optical transmittance of the polycrystalline stilbene hot pressed scintillators. Measurements were made using an integrating sphere.

Fig. 3 demonstrates the mean values of optical transparency for the polycrystalline stilbene hot pressed scintillators. These data correlate with the above cited results of the light yield measurements under the irradiation by alpha particles (see Fig. 2).

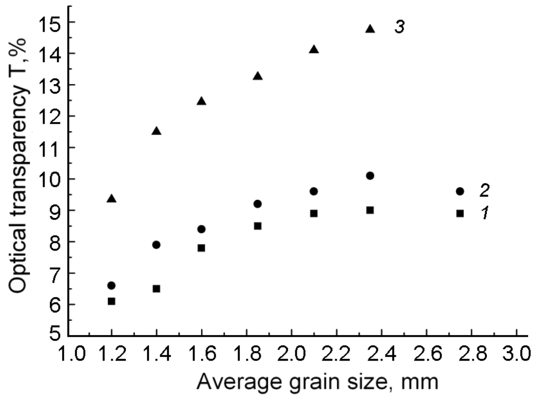

Fig. 3. Dependency of optical transparency on grain size (*1* — 360 nm, 2 — 390 nm, *3* — 700 nm) for a stilbene polycrystalline scintillators.

4. Conclusions

The studies have shown that it is desirable to use polycrystalline scintillators with graing size of gra $L \geq 2$ mm for detection of short-range radiation. The results of optical transparency measurements also indicate a growth of the transmission coefficient T with the grain size increasing for shorter wavelengths. At the same time, the optical transparency exhibits the effect of saturation for $L_{av} \sim 2$ mm. For $L_{av} > 3$ mm it even shows tendency to decrease. Therefore, the optimal grain size is about 2.2–3 mm for polycrystalline stilbene hot pressed scintillators. In this case it is possible to obtain light output of polycrystalline scintillator at 50–75 % relative to the reference stilbene single crystal.

References

1. S.V.Budakovsky, N.Z.Galunov, A.Yu.Rybalko et al., *Mol.Cryst.Liq.Cryst.*, **385**, 71 (2002).
2. N.Z.Galunov, J.H.Baker, S.V.Budakovsky et al., *J. Luminescence*, **102**, 464 (2003).
3. S.V.Budakovsky, N.Z.Galunov, N.L.Karavaeva et al., *IEEE Trans. Nucl. Sci.*, **54**, 2734 (2007).
4. S.S.Minenko, S.V.Budakovsky, L.N.Lisetski et al., *Functional Materials*, **16**, 126 (2009).
5. Hamamatsu Photomultiplier Tubes and Assemblies, Hamamatsu Global Site (2011). http://jp.hamamatsu.com/products/sensoretd/pd002/pd394/index_en.html

Дослідження сцинтиляційних і оптичних властивостей полікристалічного стильбену

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Робота присвячена дослідженню сцинтиляційних характеристик полікристалів стильбену, отриманих методом гарячого пресування з порошків різної дисперсності. Вивчено вплив розміру зерен порошків на світловий вихід і оптичну прозорість полікристалів. Показано, що розмір зерна порядку 2,2–3 мм є оптимальним для реєстрації короткопробіжних випромінювань при вибраній товщині зразка. При цьому вдається отримати світловий вихід полікристалічного сцинтилятора 50–75 % відносно монокристала. Також показано, що основним чинником, що погіршує сцинтиляційні характеристики при відносно малих розмірах зерен, є зниження оптичної прозорості.