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Light yield non-proportionality of organic and inorganic scintillators exposed to alpha rays of various energy

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The influence of entrance surface for radiation state of CsI:Tl, CsI:Na, stilbene and *p*-terphenyl scintillators on non-proportionality of light yield to the alpha-particle energy has been studied. It has been shown that formation of disrupted near-surface layer results in increasing microheterogeneity of specific light yield over the crystal surface and depth. The organic and alkali-halide scintillator samples show considerable specific light output variations depending on alpha-particle energy. These samples have essential structural disruptions of entrance surface for radiation. A correlation is observed between the non-proportionality and pulse height resolution: the less non-proportionality is, the less is the pulse-height resolution value within the range of used alpha-particle energies.

Исследовано влияние состояния входной для излучения поверхности сцинтилляторов CsI:Tl, CsI:Na, стильбена и *n*-терфенила на непропорциональность светового выхода и энергии альфа-частиц. Показано, что образование приповерхностного нарушенного слоя приводит к увеличению микронеоднородностей удельного светового выхода по поверхности кристаллов и по глубине. Большими изменениями удельного светового выхода в зависимости от энергии альфа-частиц обладают образцы органических и щелочно-галоидных сцинтилляторов, имеющие существенные структурные нарушения входной для излучения поверхности. Между величиной непропорциональности и амплитудным разрешением наблюдается зависимость: чем меньше непропорциональность, тем меньше значение разрешения в диапазоне используемых энергий альфа-частиц.

Organic and alkali-halide scintillation crystals are widely used in production of short-range radiation detectors. These crystals are easy to treat. The necessary shape of crystal samples is provided by grinding and polishing. However, a disrupted layer is formed at the crystal surface during the treatment [1]. The disrupted surface layer thickness is 10 to 20 μm at standard treatment techniques. The disrupted surface layer is characterized usually by high a content of defects and impurities that influence the operating characteristics of the detectors made using these scintillators. This influence is manifested itself in different manners for organic and alkali-halide crystals. The processes resulting in increasing of reserved elastic energy and relaxation processes causing its decreasing are realized simultaneously at any stage of plastic deformation [2]. This factor results in instability of the scintillator treated surface condition. It is possible to increase the service life and work reliability of such scintillators applying the surface coating [3, 4]. However, the surface coating application onto the radiation entrance surface results in worsening of the detector spectrometric characteristics in the case of short-range radiation registration. The property heterogeneity of organic and inorganic scintillators in depth near to the free surface manifests itself, first of all, as non-proportionality of light yield to the registered radiation energy [5, 6]. This, in turn, results

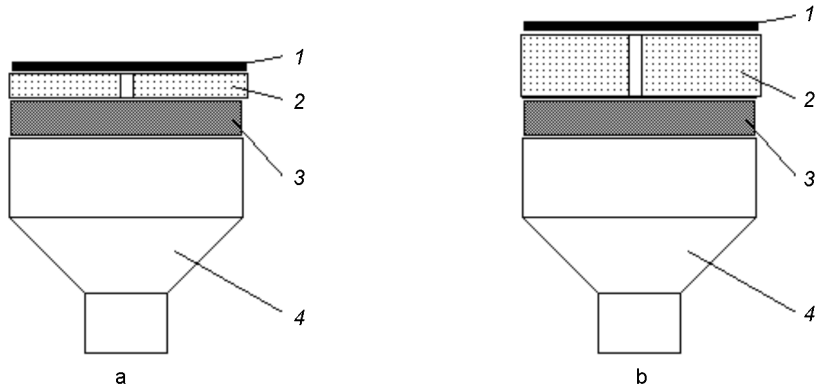

Fig. 1. Experimental setup; *1*–alpha-particle source; *2* – collimator; *3* – scintillator; *4* – PMT.

in pulse-height resolution (R) worsening. The increasing defect number in the surface layer results in increasing slow decay component contribution to the radioluminescence at registration of short range radiation [7, 8]. This causes increasing contribution of the scintillator light output nonproportionality to the pulse-height resolution. The main goal of this work is to study the influence of entrance surface for radiation condition of organic and alkali-halide scintillators on non-proportionality of the light output to the alpha-particle energy.

Alpha-particles with two different energies E_1 = 5 MeV and E_2 = 2 MeV were used in the experiments. Pu-239 radionuclide and collimators of different thickness were used to obtain alpha-particles of those energies. The experiment scheme is demonstrated in Fig. 1. A Hamamatsu R1307 photomultiplier, a BUS-2-94 preamplifier, a BUS-2-97 amplifier, and an AMA-03F pulse analyzer were used. The PMT photocathode sensitivity is in the wavelength range of 300 to 650 nm. The shaping time is 1 μs for organic scintillators and 2 μs for alkali-halide scintillators.

Stilbene and *p*-terphenyl shaped as single crystal disks with of 40 mm diameter and 3 mm thickness as well as disks of the same materials and sizes manufactured by hot-pressing [7] were chosen as the study objects. Crystal powders obtained by mechanical crushing of the optical single crystals grown from the melt are the initial materials for pressing. For comparison, the same investigations were carried out for a plastic scintillator (SP) on polystyrene basis of the same shape. The study objects except for SP were exposed to mechanochemical treatment (grinding and polishing).

Light output (V), pulse-height resolution, relative specific light yield $\eta = V/E$ and non-proportionality between the light output and the alpha-particle energy $\Delta = (V_1/E_1 - V_2/E_2)/(V/E)_{av.}$ ($(V/E)_{av.}$ is the average value of the specific light yield) were determined. The non-proportionality characterizes the deviation extent of a specific light yield from a constant value. After the measurements, the samples, except for SP, were annealed at 80°C for 3 hours. Then the measurements were carried out again.

The pulse height spectra obtained at spectrometric characteristic measurements by detectors on stilbene and *p*-Terphenyl basis are shown in Figs. 2, 3. The average values of specific light yield, pulse-height

Table 1. Specific light yield, pulse-height resolution and light yield non-proportionality values for scintillators on stilbene and *p*-terphenyl basis at alpha-particle registration

Scintillator	Specific light yield, r.u.		Resolution, %		Non-proportionality, %	Non-proportionality after annealing, %
	E_1	E_2	E_1	E_2		
Stilbene	0.69	0.79	10	16	13.5	12
Pressed stilbene	0.64	0.85	15	17	28	27
p-Terphenyl	1.28	1.48	11	15	14.5	18
Pressed *p*-Terphenyl	1.24	1.59	16	18	25	30
Scintillation plastic	0.26	0.26	25	30	–	–

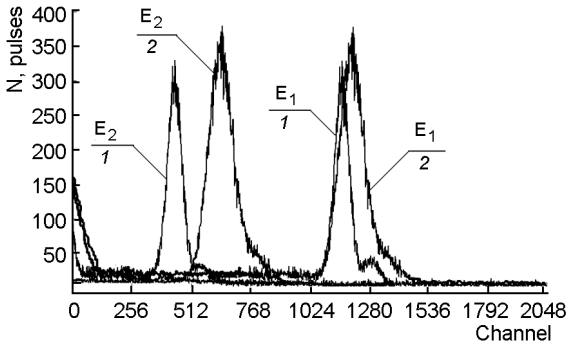

Fig. 2. Pulse-height spectra at irradiation of single crystal (*1*) and hot-pressed (*2*) stilbene samples with alpha-particles of E_1 and E_2 energies.

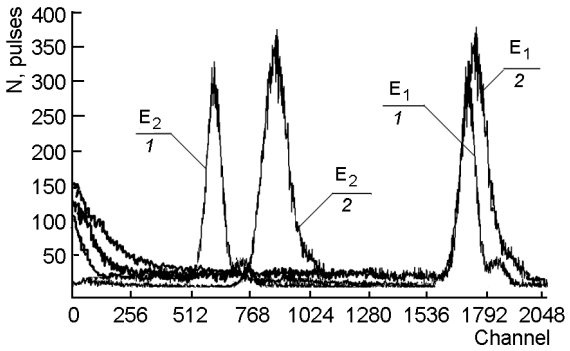

Fig. 3. Pulse-height spectra at irradiation of single crystal (*1*) and hot-pressed (*2*) *p*-terphenyl samples with alpha-particles of E_1 and E_2 energies.

resolution and non-proportionality for several samples of each scintillator kind are presented in Table 1.

Consideration of results presented in the Table shows the specific light yield of studied organic scintillators, except for SP, decreases with increasing alpha-particle energy. A more considerable difference of the specific light yield (V/E) depending on E is observed for the pressed stilbene and *p*-terphenyl samples (about 28 %) than for single crystal samples (about 15 %). Annealing of stilbene samples did not change V/E essentially for both E_1 and E_2 energies, while for *p*-terphenyl, the non-proportionality somewhat increases. High specific light yield non-proportionality of the pressed samples results in worsened resolution as compared to single crystal samples (see Table 1). The alpha/beta-ratio for the pressed samples is higher than for single crystals and amounts 0.1 and 0.01, respectively.

The nonlinear light yield dependence on the registered radiation energy was studied using CsI:Tl and CsI:Na samples of $\varnothing 30 \times 5$ mm^3 size with equally treated lateral surface and scintillation light output one. The activator concentrations in the samples were $C_{Na} = 5 \cdot 10^{-2}$ mass % and $C_{Tl} = 2 \cdot 10^{-1}$ mass %. The sample surfaces from the radioactive source side were polished by the standard technique. After the measurements, these surfaces were ground using a 0.5 μm grain abrasive without the further polishing and the scintillation characteristics were measured again.

The values of the pulse-height resolution and light yield non-proportionality determined immediately after treatment of CsI:Tl and CsI:Na samples are presented in Table 2. Consideration of those experimental results evidences that the light yield non-proportionality increases by 20 % at the formation of distrupted surface layer. The pulse-height resolution changes insignificantly at registration of alpha-particles with 2 MeV energy and becomes worse by 1 or 2 % in absolute value at registration of 5 MeV alpha-particles. The non-proportionality decreases at polishing of CsI:Tl and CsI:Na crystals entrance surface for radiations due to specific light yield decreasing for alpha-particle energy $E_2 = 2$ MeV as compared to the specific light yield for ground mentioned surfaces. These ratios remain essentially constant for the CsI:Tl crystals and change in time for the CsI:Na ones. At first, the light yield non-proportionality for CsI:Na crystals decreases, and then changes its sign after 20 days of the sample holding

Table 2. The values of light yield non-proportionality to alpha-particle energy and pulse-height resolution at polished or frosted (ground) surfaces of CsI:Tl and CsI:Na crystals

Scintillator and treatment method	Light yield non-proportionality, %	Pulse-height resolution, %	
		E_1	E_2
CsI:Na (polished)	7	4.8	6.1
CsI:Na (ground)	25	5.9	6.4
CsI:Tl (polished)	8	5.3	7.4
CsI:Tl (ground)	33	6.9	7.6

in normal conditions, i.e. the specific light yield becomes substantially lower (30 %) for alpha-particle energy E_2 than for E_1. Such light yield degradation is more pronounced in the crystals with ground entrance surface for radiation. Thus, the condition of that surface influences on the conversion efficiency, which decreases in the depth of CsI:Tl and CsI:Na crystals. This distinction is already appreciable for crystal depth about 4 μm and about 16 μm, corresponding to alpha-particle free path at energy 2 MeV and 5 MeV, respectively.

The increasing of defect concentration in the distrubted surface layer seems to result in increasing luminescence center concentration in the scintillators, thus favoring the decreasing migration loss at the energy transfer in the radioluminescence process. The scintillation characteristic instability of CsI:Na surface layer is connected with higher hygroscopicity of this material as compared to CsI:Tl.

Thus, in the study of light yield proportionality to the registered radiation energy, it is shown that in organic and alkali-halide scintillators with essential structure disruptions of the entrance surface for radiation, the specific light yield changes stronger as a function of energy. It is shown that light yield non-proportionality at registration of short-range radiation is an inherent property of the scintillation material. The surface treatment of the studied scintillators results in increased specific light yield in the near-surface layer as compared to the bulk. A pronounced correlation is observed between the degree of non-proportionality and pulse-height resolution. The resolution is minimum at the minimum non-proportionality in the range of the used alpha-particle energies. Comparing the non-proportionality values for the polished organic and alkali-halide crystals, it is seen that in the latter, the light yield non-proportionality value is substantially lower. No significant changes of spectrometric parameters was observed in time for *p*-terphenyl and CsI:Tl samples with polished surfaces. The stable condition for ground surfaces of the mentioned scintillators, the stable condition was settled after one day.

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Нелінійність світлового виходу при реєстрації альфа-випромінювання різної енергії детекторами на основі органічних і неорганічних сцинтиляторів

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Проведено дослідження впливу стану вхідної для випромінювання поверхні сцинтиляторів CsI:Tl, CsI:Na, стильбену і *n*-терфенілу на непропорційність світлового виходу і енергії альфа-часток. Показано, що утворення приповерхневого порушеного шару призводить до збільшення мікронеоднорідностей питомого світлового виходу вздовж поверхні кристалів і за глибиною. Великі зміни питомого світлового виходу, в залежності від енергії альфа-часток, характерні для зразків органічних і лужно-галоїдних сцинтиляторів, які мають істотні структурні порушення вхідної для випромінювання поверхні. Між величиною непропорційності і амплітудним розділенням спостерігається залежність: чим менше непропорційність, тим менше значення розділення у діапазоні енергій альфа-часток, що використовуються.