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GSO: Ce^{3+} scintillator with a high energy resolution

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Abstract. It is known that in case of registration of the scintillation light using photomultiplier energy resolution of the scintillation detector can be written as sum of three components, each of them is statistically independent of others: R_{in} – the value describing the dispersion of the light quantity which is formed in the process of radioluminescence and depends on the scintillator material; R_{ph} – the value describing statistic variations of the number of photoelectrons knocked out from the photocathode by the scintillation photons, it is reversely proportional to the scintillator light output; R_t – the value corresponding to the non-uniformity of the light collection coefficient in different parts of the scintillator volume. In this paper the authors made an attempt to improve energy resolution of GSO(Ce) detector owing to lowering the latter two terms by varying the bulk optical properties and reflection characteristics of the surface by means of annealing. Energy resolution 8.5% by γ-line 662 keV from ^{137}Cs was achieved due to improvement of transparency and especially selected surface roughness of the cylindrical Gd_2SiO_5:Ce scintillator 27 mm in diameter and 90 mm long.

Keywords: scintillator, energy resolution, radioluminescence.

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1. Annealing

It is difficult to get scintillators with high light output and energy resolution using a conventional growth method [1-4]. A significant contribution to the deterioration of energy resolution is made by the non-uniformity of the light collection coefficient over the scintillator volume. This non-uniformity is explained by a significant absorption of the intrinsic luminescence emission due to the activator-vacancy complexes formed during the crystal growth; the complexes are formed on the basis of cation vacancies of gadolinium and cerium ions. Decomposition of such complexes and the corresponding decrease of undesirable absorption is possible when annealing the crystals in atmosphere with certain chemical composition. It has been already observed in oxide crystals doped with transitional ions [5].

The study was carried out with GSO(Ce) crystals (0.6 wt.% of Ce) 27 mm diameter and 0.9 mm thick for measurements in the UV part of the spectrum; crystals of 90 mm in length were taken for measurements in the region of $\lambda > 400$ nm. The absorption spectra before and after annealing were registered using a spectral complex KSVU-23. The annealing was carried out in a furnace with a hermetic operational space. Chemical potential of the atmosphere was about 40 kJ/mol. It was this value that corresponded to the best results. The annealing time was defined by sizes of samples, i.e. time of vacancy diffusion. Prior to measuring the spectra the end surfaces of the samples were polished to the roughness value R_z = = 0.05 μm. After annealing, the transmission of the samples became better both in the region of radioluminescence (Fig. 1) and in the UV part of the spectrum (Fig. 2).

This being so, the wavelength of the best transmission (λ = 430 nm, Fig.1) practically coincides with the maximum of the luminescence spectrum [6]. The emergence of the well-structured band at λ = 250 nm, a band near 200 nm as well as significant decrease of absorption at $\lambda > 210$ nm (Fig. 2) give evidence to a structural reconstruction of color centers in the UV part of the spectrum. Similar phenomena occur in high temperature oxides [7]; they are related to structural changes in activator-vacancy complexes. Improvement of transmission (in the long-wave range) only in the wavelength range of luminescence of Ce activator ions confirms this point of view (Fig. 1). A detailed study of the mechanism of structural reconstruction of activator-vacancy complexes under annealing in the controlled medium is of particular interest and requires special investigations.

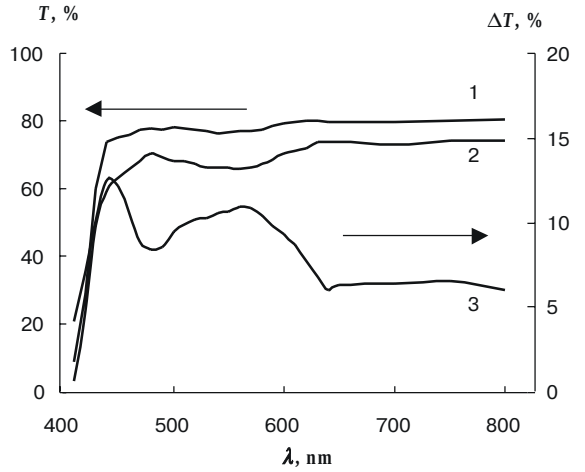

Fig. 1. Transmission spectrum of GSO(Ce) crystal: 1 – before annealing; 2 – after annealing; 3 – an extracted difference between the transmission spectra of the annealed and non-annealed samples.

2. Surface treatment of a scintillator

The effect of the scintillator transparency and character of its surface microrelief on energy resolution of the detector was studied using a numeric model of the light collection process [8]. This model is based on the Monte-Carlo method operating with the light beams as unit events. In the calculation of light scattering by the detector surface a conception of it being a smooth plane covered with randomly oriented grooves was used. This conception allowed to model the process of light collection in the detector which has no optical coupling between the reflecting coating and scintillator surface. The following values of GSO(Ce) characteristics were used for the calculation: refraction index $n = 1.9$, light absorption co-efficient in the luminescence band $K = 0.02\ldots0.08$ cm^{-1}. The output window (end) of the scintillator in the form of a cylinder was considered to have not a full coupling with the photoreceiver through the immersion medium with n=1.5. The coefficient of light reflection by the coating was $R = 0.85$.

Table 1. Calculated data on the mean light collection coefficient $\bar{\tau}$ and degree of its non-uniformity R_τ over the cylindrical scintillator Ø27×90 mm.

K, cm^{-1}	0.08	0.06	0.04	0.03	0.02
$\bar{\tau}$	0.167	0.199	0.241	0.268	0.301
R_τ	32.6	20.1	9.6	6.2	4.7

It is known [1, 8] that for achieving the best uniformity of the light collection coefficient τ over the length of the protruded cylinder all its surfaces should be mirror smooth, and the absorption coefficient should be minimal. Table 1 illustrates the degree of the effect of the scintillator transparency on resolution and light output of the detector.

The calculations show that there is no radial non-uniformity of τ at the given design of the detector. Deterioration of τ and R_τ is caused only by axial non-uniformity of τ (see Fig. 3). The source of such deterioration is the increased absorption of the scintillation light. Thus, improvement of the scintillator transparency has a double effect: decrease of R_{ph} connected with the rise of $\bar{\tau}$ and light output; decrease of R_τ connected with the rise of the light collection coefficient from the parts of the scintillator remote from the photoreceiver.

There is one more way of improving the energy resolution of the detector. It is connected with the change of

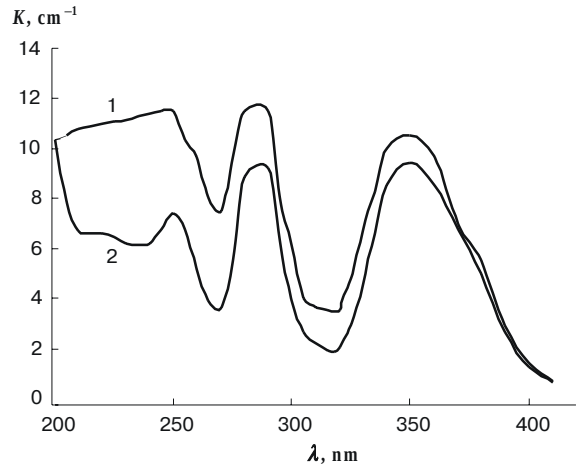

Fig. 2. Absorption of GSO(Ce) samples: 1 – before thermal annealing; 2 – after thermal annealing.

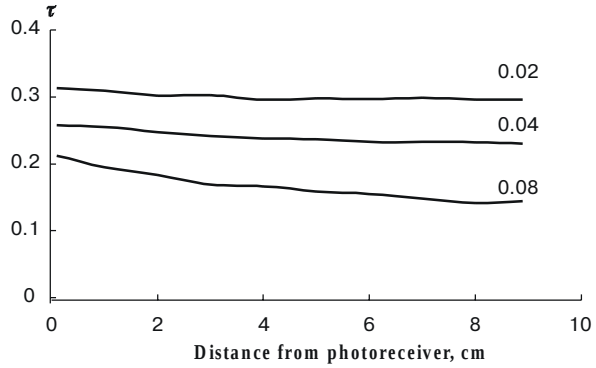

Fig. 3. Calculated axial distributions of the light collection coefficient over the length of the GSO(Ce) cylinder Ø27×90 mm. The numbers near the curves denote transparency of a scintillator K, cm^{-1}.

Table 2. The effect of annealing and surface roughness on energy resolution of GSO(Ce) scintillator having the form of a cylinder, ∅27×90 mm by γ-line 662 keV from ^{137}Cs.

Treatment of the sample surfaces	Energy resolution R, %	
	Reflecting coating Tyvek	Reflecting coating fluoroplast film
Side surface-rough matting; both ends of cylinder are polished	24.4	23.8
Side surface and photosensitive end – polished, free end-rough grinding; the sample has been annealed	13.8	13.1
Same surface roughness, except: free end – fine grinding; the sample has been annealed	12.0	11.4

light collection conditions in the sample. At matting (making the upper end rough) the light collection coefficient becomes higher in the regions of the scintillator adjacent to this end. Its increase depends on the degree of the end roughness (Fig. 4). As it is seen from the presented data there exists a certain degree of roughness at which deviation of τ from the mean value is minimal. This degree can be determined by experimental selection. It was this thing that done in this paper. With account of the calculated tendencies a series of measurements of the light output and energy resolution has been conducted. The results are given in Table 2.

Data on energy resolution of the sample given in Table 2 were obtained using a photomultiplier FEU-173. A sample treated according to the last line of the Table 2 and used with the Hamamatsu R1307 PM showed the energy resolution 8.5% on the line 662 keV.

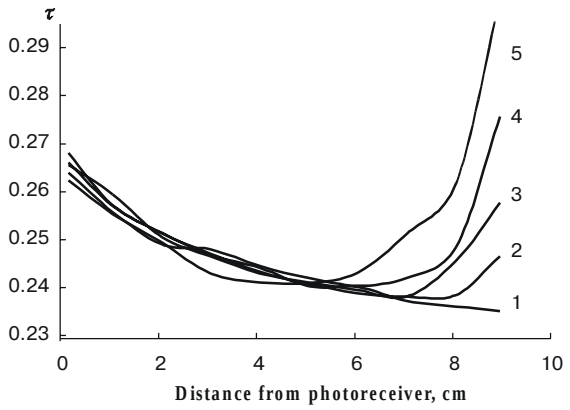

Fig. 4. Calculated axial distributions of the light collection coefficient over the length of the GSO(Ce) cylinder ∅27×90 mm with different degree of roughness of the end opposite to the photoreceiver. The curves are enumerated in the order of the roughness degree increase: 1 – polished end (R_z = 0.05 mm); 5 – maximum rough end.

Conclusions

Increase of transparency of GSO(Ce) scintillation crystal by annealing in the atmosphere with the chemical potential ~40 kJ/mol and lowering of non-uniformity of the light collection coefficient mechanical treatment of the surface allows to achieve extremely high energy resolution of the detector. As a result of the change of surface and bulk characteristics of cylindrical Gd_2SiO_5:Ce scintillator with a diameter 27 mm and length 90 mm coupled with PM Hamamatsu R1307 the energy resolution 8.5% by γ-line 662 keV from ^{137}Cs was obtained.

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