

Factors which define the α/γ ratio in CsI:Tl crystals

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Abstract

Dependences of light yield and α/γ ratio on the Tl concentration have been studied within a wide range of shaping times. It is shown that the α/γ ratio essentially depends on the Tl concentration. Proper combination of the Tl concentration and optimum shaping time in electronics allows to obtain detectors with rather high light output for γ -rays, α -particles and α/γ ratio values. It has been shown that both the light yield at α -excitation and the α/γ -ratio depend on the time of crystal storage after polishing. On the basis of the idea of the formation of deformation-induced point defects in a thin surface-adjacent layer, the causes of the temporary increasing in light yield for α -particle are explained. An explanation is given of the results obtained by Gwin and Murray concerning the fact that the α/γ ratio is practically independent on Tl concentration.

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Keywords: Scintillator; CsI:Tl; Light yield; Activator concentration; Scintillation process

1. Introduction

As is known, in doped alkali halide crystals the value of light yield (L) per 1 MeV of absorbed energy (E) is lower for α -particles than for γ -rays. To take this fact into account, the definition of α/γ ratio is used in scintillation method. For the detectors with α -reference point the value of this ratio should be as close to unity as possible.

Decrease of the specific L/E light yield for particles with large linear energy loss (dE/dx) in the track has been attributed to “saturation of activator centers”. This term expresses depletion of available activator sites by a particle of high dE/dx . A phenomenological model of scintillation process taking into account conception of saturation of the available activator centers was proposed by Murray and Meyer [1]. This saturation model (SM) predicts that the L/E vs. dE/dx curve would depend on the Tl content (C_{Tl}).

The SM was experimentally verified by Gwin and Murray [2] for CsI:Tl. According to their data, the shape of the curve L/E as a function of dE/dx

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is practically independent of C_{Tl} . Their results do not agree with the work of Tsirlin [3]. Tsirlin's data showed a distinct decline in the α/γ ratio with the decrease in the C_{Tl} . Analysis of known dependences of γ - and α -light yield on the C_{Tl} as for NaI:Tl [4,5] as CsI:Tl [5–7] allows to conclude that maximum light yield for α -particles is achieved at a higher C_{Tl} than for γ -rays, i.e. α/γ ratio must depend on the Tl content. Nevertheless, as recently pointed out by Kubota [4], in view of such a discrepancy of the results, the problem of the Tl concentration dependence seems to be still open for further investigation, especially for CsI:Tl.

CsI:Tl is a relatively slow scintillator: its average decay time is of the order of microsecond for γ -rays. The decay is described at least by two components. The fast component has a value of about 0.6 μs , whereas the slow one runs into 3.5 μs at γ -excitation. CsI:Tl crystals are distinguished by the fact that the response of both α - and γ -scintillations strongly depends on the electronics shaping time (t_{RC}), and this also predetermines the dependence of α/γ ratio on the t_{RC} as it shown in Ref. [8–10].

The goal of the present research was to study some factors influencing the value of α/γ ratio (in particular, Tl content, shaping time, state of the surface) in order to find out the origin of the discrepancy of the results obtained by different authors.

2. Experimental

CsI:Tl crystals were grown by the Stockbarger method. As a raw material, we used the so-called "CsI for low background crystal"; this salt was additionally purified from homological impurities to remove Rb impurity. Crystallization was realized in hermetically sealed quartz ampoules under low partial oxygen pressure whose value depended on particular crystal growth conditions and was determined experimentally. The choice of the growth method was bound up with the fact that such conditions were optimum for the obtaining of rather large (100 \times 120 mm) transparent crystals with elevated (up to 0.35 mol%)

content of thallium and retaining their color in daylight. The Tl concentration in the crystals was determined by atomic absorption analysis. This value is presented further in the text in mole% or, in some specified cases, as the number of Tl^+ ions per cm^{-3} . The sections of the crystalline ingot in which the difference in the values of C_{Tl} did not exceed 10%, were used to obtain the detectors measuring $\varnothing 30 \times 25$ and $\varnothing 30 \times 5$ mm. As the quality of entrance surface after polishing strongly depends on the crystallographic orientation, we did not use those ingots whose axes coincided with the direction $\langle 100 \rangle$, since their deformed surface-adjacent layer had unpredictable depth [11]. The samples' entrance and end surfaces were polished, their side surface was matted.

The light output was measured for γ -rays (L_γ) from ^{137}Cs ($E_\gamma = 662$ keV) and α -particles (L_α) from collimated ^{239}Pu source ($E_\alpha = 5.15$ MeV). The crystal was mounted onto the photocathode of Hamamatsu R1307 photomultiplier with optical contact, the α -source was placed on the opposite face of the sample, the γ -source was fixed over the crystal at a distance of 30 mm from it. The side sample surface was covered with a Teflon reflector, and the collimator of α -particles served as a top reflector, as it was proposed by Lyubynskiy et al. [12]. We used BUS 2–94 preamplifier, BUS 2–97 amplifier and AMA 03F analyzer. The amplifier allowed to vary the shaping time from 0.2 to 12.8 μs . The measurements were carried out by comparing the detector and primary standard signals. This method is described in detail by Sysoeva et al. in Ref. [13].

The light output L_α is sensitive to C_{Tl} , especially in the interval of low Tl concentrations, therefore its values were measured on two opposite faces. For 5 mm thick crystals the values of L_α coincided in almost all the cases, for 25 mm thick an essential difference was observed in some cases. If the difference did not exceed 10%, the obtained data were averaged; the crystals which showed a difference higher than 10% were excluded from consideration.

All the measurements were realized on the samples subjected to aging. For the newly prepared samples we discovered instability of the position of the total absorption peak of

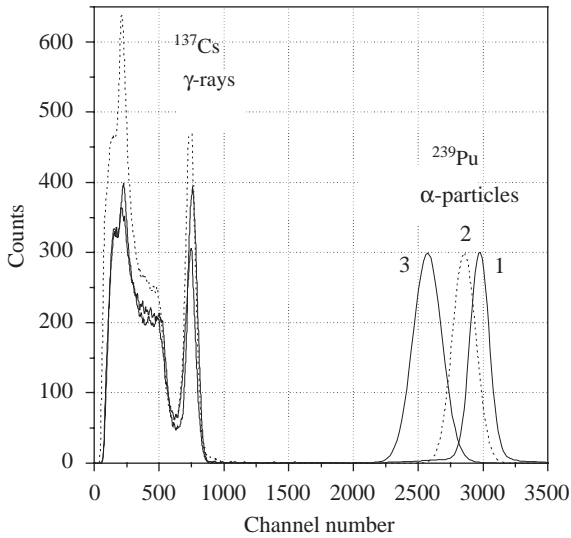


Fig. 1. Pulse height spectra for 662 keV γ -rays and 5.15 MeV α -particles both absorbed in CsI:Tl crystal with 0.02 mol% of Tl. Curve 1 corresponds to an hour after polishing, 2–6 h, 3–8 days of the aging.

α -particles, especially within first several hours after polishing. Fig. 1 shows the pulse height spectra of both γ -rays and α -particles for the crystal with $C_{\text{Tl}} = 0.02\%$. As is seen, the position and shape of the photopeak of γ -rays are stable, whereas the position of the α -peak is shifted towards lower energies and the resolution worsens. After two weeks of storage the position and shape of this peak become reproducible.

For the crystals with $C_{\text{Tl}} > 0.1\%$ the difference between the values of L_{α} for polished and aged crystals is less than the measurement error, though the resolution shows reproducible values only after several days of storage.

3. Results

Fig. 2 represents the dependences of the light output on the shaping time for CsI:Tl with $C_{\text{Tl}} = 0.11\%$. Such a concentration is optimum for detection of both γ -quanta and α -particles [6,7]. As is seen, the dependence of the L on t_{RC} is more strong for the electrons with low dE/dx than for α -particles with high dE/dx . Similar dependences for CsI:Tl crystals of another origin were

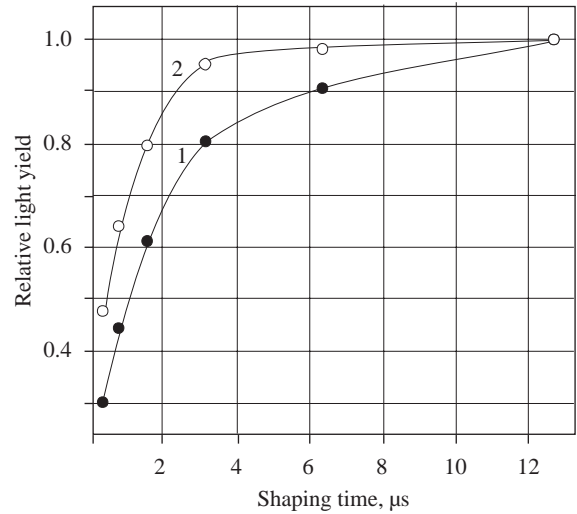


Fig. 2. Dependences of the light output on the shaping time for CsI:Tl crystal with $C_{\text{Tl}} = 0.11\%$ excited by γ -rays (1) and α -particles (2).

presented in Refs. [9,14] for optimum C_{Tl} . The data of Fig. 2 show that the α/γ ratio must decrease with the increase of t_{RC} since the L_{γ} grows in the interval where the $L_{\alpha} \approx \text{const}$.

The dependences of the light output on the activator concentration at α - and γ -excitation are presented in Fig. 3 for $t_{\text{RC}} = 6.4 \mu\text{s}$. Such shaping time allows to obtain minimum values of energy resolution for γ -rays. As seen from the data of Fig. 3, the dependence $L_{\gamma}(C_{\text{Tl}})$ reaches its plateau at the $C_{\text{Tl}} \geq 0.05\%$, whereas the $L_{\alpha}(C_{\text{Tl}})$ one achieves saturation only at $C_{\text{Tl}} \geq 0.15\%$.

It should be noted that for the crystals of another origin, e.g., those considered in Refs. [7,8], these two points correspond to lower C_{Tl} values: 0.04 and 0.11%, respectively.

However, the general conclusion is to be the following: for achievement of the highest light yield for α -particles, the crystals with elevated Tl content are required. Moreover, as Lyapidevsky [8] has shown, position of the initial point of the plateau for the L_{γ} and L_{α} depends on the chosen shaping time, especially if the $t_{\text{RC}} < 1.0 \mu\text{s}$. The shape of the $L_{\alpha}(C_{\text{Tl}})$ curve in Fig. 3 is in a good agreement with the data known from the literature and summarized by Goriletsky et al. [7]. However,

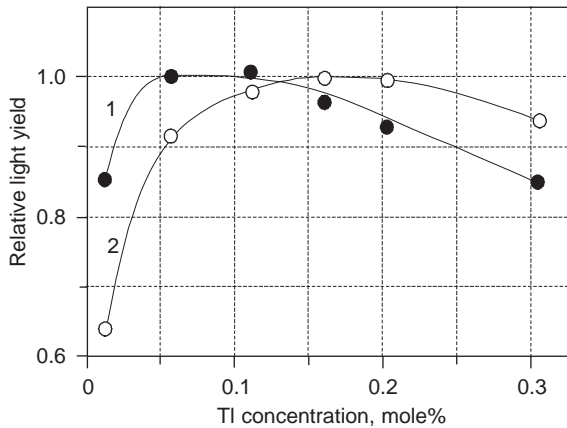


Fig. 3. Dependences of the light output on the Tl concentration for $t_{RC} = 6.4 \mu s$ at γ - (1) and α -excitation (2).

our results concerning the $L_\gamma(C_{Tl})$ dependence essentially differ from those presented in [2]. Such a discrepancy is observed for the interval of high Tl concentrations and consists in the fact that L_γ decreases at $C_{Tl} > 0.12\%$ in the case when a “blue” photomultiplier is used [7,8]. The so-called “concentration quenching” does not take place with “red” photoreceivers [7]. The paradox lies in the fact that Gwin and Murray [2] used a “blue” photomultiplier with S-20 photocathode response.

Dependences of the α/γ ratio on the shaping time are shown in Fig. 4 for the crystals with different Tl concentrations.

Similar dependence was earlier obtained by Kilgus et al. [9] for $C_{Tl} = 0.1\%$ only as well as in Refs. [10,14] where the Tl concentrations were not specified. For comparison, the data of Ref. [9] are presented on Curve 5.

As is seen, our results for $C_{Tl} = 0.11\%$ are in a good agreement with the literature data. It should be noted that our results span a wider interval of C_{Tl} . The data presented in Fig. 4 show that there exists a simple way to increase the α/γ ratio by diminishing the shaping time. Though in this case the light output decreases to some extent and the energy resolution raises, proper choice of particular t_{RC} values allow to increase the α/γ ratio, the value of light output remaining rather high.

The dependences of the α/γ ratio on the C_{Tl} for different shaping times are presented in Fig. 5. These data permit to conclude that the α/γ ratio

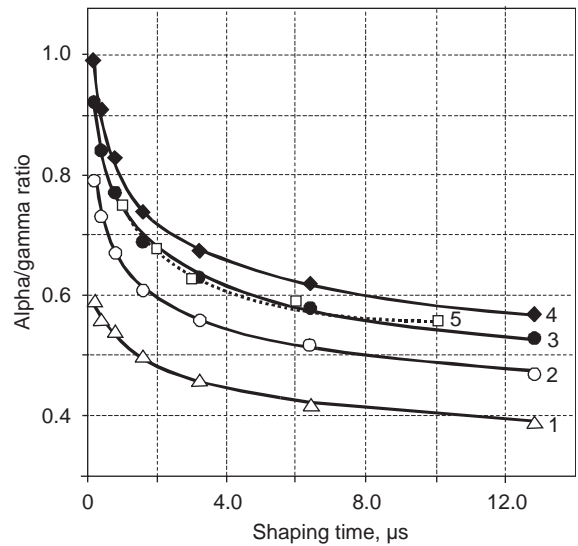


Fig. 4. Dependences of the α/γ -ratio on the shaping time for the crystals with different C_{Tl} : 1–0.013; 2–0.06; 3–0.11; 4–0.3%. Curve 5 represents data from Ref. [9] for the $C_{Tl} = 0.1\%$.

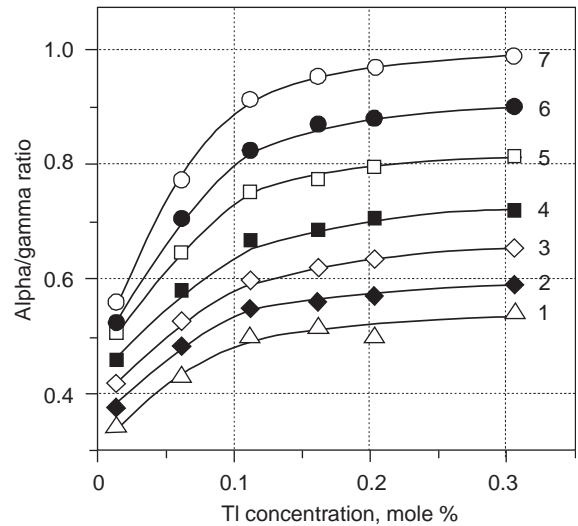


Fig. 5. Dependences of the α/γ -ratio on the C_{Tl} for the different shaping times: 1–12.8 μs ; 2–6.4; 3–3.2; 4–1.6; 5–0.8; 6–0.4; 7–0.2 μs .

grows with the increase of the C_{Tl} at any t_{RC} . Similar dependence was obtained in Ref. [3] for CsI:Tl crystals grown in vacuum for the $t_{RC} = 1.0 \mu s$ only. Thus, our experimental results confirm the dependences obtained in Ref. [3] and

totally disprove the conclusions made by Gwin and Murray [2].

4. Discussion

The results presented above as well as the literature data [3,5–7] allow to come to the conclusion that for the obtaining of both α -detectors with maximum light output and reference-point detectors with better values of the α/γ ratio, CsI:Tl crystals with elevated activator contents are required. Such a conclusion is general enough since analogous results for NaI:Tl are contained in the earlier papers [4,12]. Analysis of the concentration dependences of the L_α and L_γ for CsI:Na obtained by Panova et al. [15] also confirms this statement.

First of all, let us discuss the shape of the curves $L(C_{\text{Tl}})$ for different types of excitation. Here we shall use the term C^* which denotes the Tl concentration at the initial point of the plateau in dependence $L(C_{\text{Tl}})$ which corresponds to the minimum content of Tl within the interval of its optimum values. The fact that $C_\alpha^* > C_\gamma^*$ signifies the difference in the dE/dx for electrons and α -particles. The simplest explanation of this fact gives the SM. Recombination of electrons and holes (e–h pairs) in the host lattice comes to a fast ($\tau \approx 10$ ns) exciton luminescence ($\lambda = 310$ nm in CsI) with an efficiency of about 3000 MeV^{-1} , relatively low in comparison with activator luminescence in CsI:Tl ($\sim 50\,000 \text{ MeV}^{-1}$, [16]). For the used photomultipliers with glass windows such low light yield in the UV region of the spectrum may be neglected. The $C_{\text{Tl}} = 0.05\%$ is sufficient for suppression of inefficient recombination of e–h pairs in the track of electron ($dE/dx \sim 1 \text{ keV cm}^2/\text{mg}$) though too low for the track of α -particles ($dE/dx \sim 450 \text{ keV cm}^2/\text{mg}$) [1,4]. Thus, the SM concept is necessary to explain the behavior of the $L_\gamma(C_{\text{Tl}})$ and $L_\alpha(C_{\text{Tl}})$ curves [1,5]. As far as we know, there does not exist another proper explanation of it. Just these results concerning the change in CsI:Tl luminescence spectrum with the growth of the C_{Tl} were obtained by Gwin and Murray in the second part of their well-known paper [2].

Our results for CsI:Tl crystals as well as the results of Kubota et al. [4] for NaI:Tl, confirm the SM validity. Actually, by increasing the Tl concentration the difference between the values of L/E for γ -rays and α -particles may be partially leveled. As the SM uses the total luminescence yield, but not of light output, for verification of the SM the α/γ value is to be taken at the highest t_{RC} . From the data of Fig. 5 it follows that for $t_{\text{RC}} = 12.8 \mu\text{s}$ the α/γ ratio increases from 0.4 to 0.6 as the C_{Tl} grows.

We have already mentioned that our results are in a good agreement with the other literature data. There exists only one paper—[2]—the conclusions of which totally contradict our data concerning both the influence of C_{Tl} on the α/γ ratio and the character of the dependence $L_\gamma(C_{\text{Tl}})$ in the region of concentration quenching.

The paper [2] seems to be the only investigation in which frames the value of light output were measured on the crystals of different origin: there were used both the crystals obtained from Harshaw Chem Co and those grown by the authors in an available furnace. The quality of CsI:Tl crystals, especially for $C_{\text{Tl}} > 0.1\%$, strongly depends on the growth technology and the personnel's skill. Therefore, the dependence $L_\gamma(C_{\text{Tl}})$ obtained in Ref. [2] reflects not only the change in C_{Tl} , but also the growth method used. Such a circumstance seems to explain the fact that the authors did not observe concentration quenching of L_γ in the region of elevated Tl contents though they used a “blue” photomultiplier. However, this cannot explain why the α/γ ratio is independent of the C_{Tl} .

As for the concentration interval $C_\gamma^* < C_{\text{Tl}} < C_\alpha^*$ where $L_\gamma \approx \text{const}$, the conclusion made in [2] means that, due to some factor, the L_α retains its maximum values with decreasing C_{Tl} (see Fig. 3). In our opinion, the light yield for α -particles increased due to the presence of a sufficient quantity of luminescence centers in the thin surface layer caused by peculiarities of sample preparation technique.

It is well known that plastic deformation sharply increases the number of blue emission centers in CsI [17] and CsI:Na [18,19]. Grinding of the crystals leads to formation of new emission centers

in a thin surface-adjacent layer only [20]. Scintillation properties in the volume of the crystal remain practically unchanged. The nature of additional centers is known: they are the anion vacancies (V_a^+) which form the dipoles $V_a^+ - V_c^-$ [17] resulting from dislocation sliding. In accordance with the data of [21], for sliding of the dislocations existing in CsI, it is sufficient to apply a stress of about $(1-2) \text{ g/mm}^2$ and this threshold is easily exceeded even at “soft” sample polishing. CsI crystals have no cleavage planes, therefore the procedure of sample preparation always implies, at least, the operation of polish.

For pure CsI the effect of polishing is so substantial that near the surface luminescence of self trapped excitons is extremely weak at room temperature. Instead, there is excited slow emission of bound excitons, though in the crystal bulk fast luminescence is induced, as shown by Nishimura et al. [22]. For almost complete suppression of the fast emission in CsI, the concentration of defects in the surface-adjacent layer is to be rather high: $\sim(6-8)10^{17} \text{ cm}^{-3}$. This estimation was obtained from the fact that in CsI:Na crystals the $C_\gamma^* = (9-10)10^{17} \text{ cm}^{-3}$ [15]. If the concentration of sodium is lower than the said value, fast emission at 310 nm is clearly seen in the radioluminescence spectra. If this concentration is higher, then the luminescence of self trapped excitons will be completely suppressed by the activator [23]. According to the data of the second part of Ref. [2] for CsI:Tl, luminescence at 310 nm disappears at $C_{\text{Tl}} > 2 \times 10^{17} \text{ cm}^{-3}$. However, it should be taken into account that the A-band of Tl^+ absorption strongly overlaps the luminescence at 310 nm, and in this case the estimation is undervalued. In CsI: CO_3 scintillator the emission center is a dipole

$\text{CO}_3^{2-} - V_a$ [24], and for suppression of the 310 nm luminescence it is necessary to introduce CO_3^{2-} ions in $(6-8)10^{17} \text{ cm}^{-3}$ quantities. We consider this value to be the most probable quantity of $V_a^+ - V_c^-$ dipoles which arise within the surface-adjacent layer after polishing (C_D).

As is known, plastic deformation noticeably distorts the luminescence spectrum of CsI:Tl [25]. According to our data, this effect manifests itself explicitly if the Tl concentration is approximately equal to C_γ^* or lower. The data presented in Fig. 1 directly demonstrate the role of point defects arising within the surface-adjacent layer under the influence of deformation. As is seen from this figure, position of the α -peak in the pulse height spectrum depends on the time which has passed after the polishing. The values of L_α are stabilized after 6–8 days of crystal aging under the normal conditions. The values of the α/γ ratio determined after polishing the entrance surface and those measured in 14 days of aging, are presented in the table.

As is seen from Table 1 and Fig. 1, polish of the entrance surface influences neither L_γ nor L_α for the crystals with $C_{\text{Tl}} \gg C_D$. Another situation is observed for the crystals with $C_{\text{Tl}} \leq C_D$: in this case the light output of α -particles sharply increases, while that of 662 keV photons (which excite mainly the scintillator's bulk) remains practically the same. Such an effect causes temporary increase of the α/γ ratio. The conclusions made in Ref. [2] seem to be explained by the said fact. In the experimental part of the mentioned paper there is a circumstance which concerns the method of sample preparation. The authors used the samples with a diameter of 25 mm glued to the light guide and then ground to obtain a thickness of 2 mm. If

Table 1
Changes in a α/γ ratio after polishing and in the process of ageing

Tl concentration	α/γ -ratio for $t_{\text{RC}} = 4 \mu\text{s}$			
mol%	cm^{-3}	Before polishing	After polishing	In 14 days
5×10^{-4}	5×10^{16}	0.28	0.511	0.281
6×10^{-3}	6×10^{17}	0.321	0.505	0.32
2×10^{-2}	2×10^{18}	0.512	0.588	0.51
1.5×10^{-1}	1.5×10^{19}	0.602	0.605	0.6

they used the same light guide, then the measurements had to be performed immediately after preparation of the crystals when L_α could not relax to its natural values. In fact the concentration of emission centers was defined by C_D , not by $C_{Ti} \leq C_D$.

The unusual shape of the dependence L/E vs. E_α shown in Fig. 4 of the paper [2] confirms this statement. The value of specific light yield sharply increases for α -particles with $E_\alpha < 1$ MeV, i.e. for the particles absorbed at the crystal surface. While studying analogous dependences for aged CsI:Tl samples excited by protons [26] or α -particles [27] such a situation was not observed. On the contrary, the authors of the mentioned papers came to the conclusion concerning the existence of a layer with a low scintillation efficiency near the surface. In CsI:Tl the depth of this layer is insignificant and runs into fractions of micrometer [27].

Unfortunately, erroneous conclusions from the results obtained in Ref. [2] are widely cited in reviews and reference books devoted to scintillation method [8,28–30]. However, the firms producing the detectors with α -reference point seem to be guided by the conclusions made in Ref. [3], since the catalogues [31,32] advertise the detectors for which the α/γ ratio can be chosen within certain limits at the customer's request.

The value of α/γ ratio also depends on the energy of γ -rays. As recently shown by Kudin et al. [33], the duration of scintillations decreases as the energy of photons falls, and for $E_\gamma = 5.9$ keV the dependence $L_\gamma(t_{RC})$ corresponds rather to Curve 1 in Fig. 2 for α -particles then to Curve 2 for 662 keV photons. Fortunately, in most cases this circumstance must not be taken into account in practice, since the detectors with α -reference point are not used for registration of such low-energy photons.

5. Conclusions

Dependences of light yield and α/γ ratio on the Tl concentration have been studied within a wide range of shaping times. It is shown that the α/γ ratio essentially depends on the Tl concentration,

and for the obtaining of high-quality α -detectors and the detectors with α -reference point CsI:Tl crystals with elevated activator concentrations, are required. Proper combination of certain parameters such as the Tl concentration in the crystal and optimum time of shaping in the electronic system allows to obtain detectors with rather high light output for γ -rays, α -particles and α/γ ratio values.

It has been shown that both the light yield at α -excitation and the α/γ -ratio depends on the time of crystal storage after polishing. On the basis of the idea of the formation of deformation-induced point defects in a thin surface-adjacent layer, the causes of the temporary increasing in light yield for α -particle are explained. Given is an explanation of the results obtained by Gwin and Murray [2] concerning the relation between the α/γ ratio and the Tl concentration. In our opinion, the paper [2] cannot be taken into account for experimental verification of theoretical models.

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is practically independent of C_{Tl} . Their results do not agree with the work of Tsirlin [3]. Tsirlin's data showed a distinct decline in the α/γ ratio with the decrease in the C_{Tl} . Analysis of known dependences of γ - and α - light yield on the C_{Tl} as for NaI:Tl [4,5] as CsI:Tl [5–7] allows to conclude that maximum light yield for α -particles is achieved at a higher C_{Tl} than for γ -rays, i.e. α/γ ratio must depend on the Tl content. Nevertheless, as recently pointed out by Kubota [4], in view of such a discrepancy of the results, the problem of the Tl concentration dependence seems to be still open for further investigation, especially for CsI:Tl.

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2. Experimental

CsI:Tl crystals were grown by the Stockbarger method. As a raw material, we used the so-called “CsI for low background crystal”; this salt was additionally purified from homological impurities to remove Rb impurity. Crystallization was realized in hermetically sealed quartz ampoules under low partial oxygen pressure whose value depended on particular crystal growth conditions and was determined experimentally. The choice of the growth method was bound up with the fact that such conditions were optimum for the obtaining of rather large (100 × 120 mm) transparent crystals with elevated (up to 0.35 mol%)

content of thallium and retaining their color in daylight. The Tl concentration in the crystals was determined by atomic absorption analysis. This value is presented further in the text in mole% or, in some specified cases, as the number of Tl^+ ions per cm^{-3} . The sections of the crystalline ingot in which the difference in the values of C_{Tl} did not exceed 10%, were used to obtain the detectors measuring $\varnothing 30 \times 25$ and $\varnothing 30 \times 5$ mm. As the quality of entrance surface after polishing strongly depends on the crystallographic orientation, we did not use those ingots whose axes coincided with the direction $\langle 100 \rangle$, since their deformed surface-adjacent layer had unpredictable depth [11]. The samples' entrance and end surfaces were polished, their side surface was matted.

The light output was measured for γ -rays (L_γ) from ^{137}Cs ($E_\gamma = 662$ keV) and α -particles (L_α) from collimated ^{239}Pu source ($E_\alpha = 5.15$ MeV). The crystal was mounted onto the photocathode of Hamamatsu R1307 photomultiplier with optical contact, the α -source was placed on the opposite face of the sample, the γ -source was fixed over the crystal at a distance of 30 mm from it. The side sample surface was covered with a Teflon reflector, and the collimator of α -particles served as a top reflector, as it was proposed by Lyubynskiy et al. [12]. We used BUS 2–94 preamplifier, BUS 2–97 amplifier and AMA 03F analyzer. The amplifier allowed to vary the shaping time from 0.2 to 12.8 μs . The measurements were carried out by comparing the detector and primary standard signals. This method is described in detail by Sysoeva et al. in Ref. [13].

The light output L_α is sensitive to C_{Tl} , especially in the interval of low Tl concentrations, therefore its values were measured on two opposite faces. For 5 mm thick crystals the values of L_α coincided in almost all the cases, for 25 mm thick an essential difference was observed in some cases. If the difference did not exceed 10%, the obtained data were averaged; the crystals which showed a difference higher than 10% were excluded from consideration.

All the measurements were realized on the samples subjected to aging. For the newly prepared samples we discovered instability of the position of the total absorption peak of

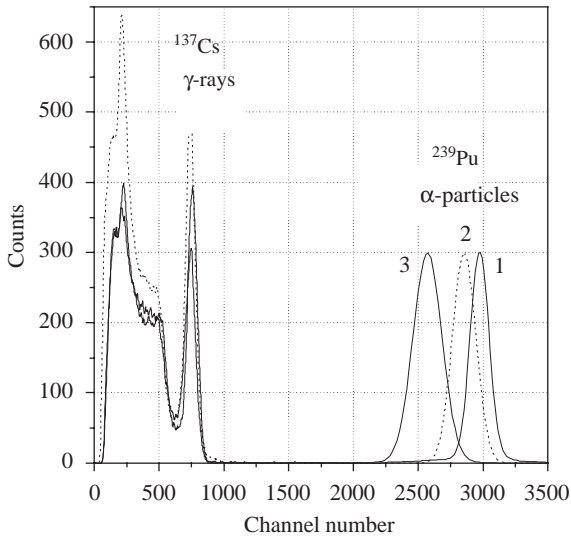


Fig. 1. Pulse height spectra for 662 keV γ -rays and 5.15 MeV α -particles both absorbed in CsI:Tl crystal with 0.02 mol% of Tl. Curve 1 corresponds to an hour after polishing, 2–6 h, 3–8 days of the aging.

α -particles, especially within first several hours after polishing. Fig. 1 shows the pulse height spectra of both γ -rays and α -particles for the crystal with $C_{\text{Tl}} = 0.02\%$. As is seen, the position and shape of the photopeak of γ -rays are stable, whereas the position of the α -peak is shifted towards lower energies and the resolution worsens. After two weeks of storage the position and shape of this peak become reproducible.

For the crystals with $C_{\text{Tl}} > 0.1\%$ the difference between the values of L_{α} for polished and aged crystals is less than the measurement error, though the resolution shows reproducible values only after several days of storage.

3. Results

Fig. 2 represents the dependences of the light output on the shaping time for CsI:Tl with $C_{\text{Tl}} = 0.11\%$. Such a concentration is optimum for detection of both γ -quanta and α -particles [6,7]. As is seen, the dependence of the L on t_{RC} is more strong for the electrons with low dE/dx than for α -particles with high dE/dx . Similar dependences for CsI:Tl crystals of another origin were

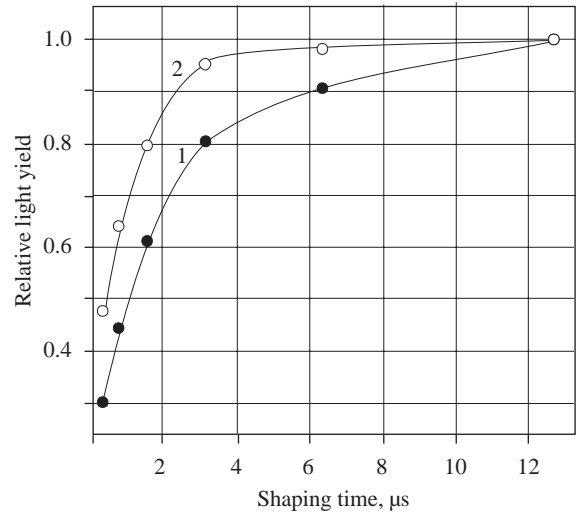


Fig. 2. Dependences of the light output on the shaping time for CsI:Tl crystal with $C_{\text{Tl}} = 0.11\%$ excited by γ -rays (1) and α -particles (2).

presented in Refs. [9,14] for optimum C_{Tl} . The data of Fig. 2 show that the α/γ ratio must decrease with the increase of t_{RC} since the L_{γ} grows in the interval where the $L_{\alpha} \approx \text{const}$.

The dependences of the light output on the activator concentration at α - and γ -excitation are presented in Fig. 3 for $t_{\text{RC}} = 6.4 \mu\text{s}$. Such shaping time allows to obtain minimum values of energy resolution for γ -rays. As seen from the data of Fig. 3, the dependence $L_{\gamma}(C_{\text{Tl}})$ reaches its plateau at the $C_{\text{Tl}} \geq 0.05\%$, whereas the $L_{\alpha}(C_{\text{Tl}})$ one achieves saturation only at $C_{\text{Tl}} \geq 0.15\%$.

It should be noted that for the crystals of another origin, e.g., those considered in Refs. [7,8], these two points correspond to lower C_{Tl} values: 0.04 and 0.11%, respectively.

However, the general conclusion is to be the following: for achievement of the highest light yield for α -particles, the crystals with elevated Tl content are required. Moreover, as Lyapidevsky [8] has shown, position of the initial point of the plateau for the L_{γ} and L_{α} depends on the chosen shaping time, especially if the $t_{\text{RC}} < 1.0 \mu\text{s}$. The shape of the $L_{\alpha}(C_{\text{Tl}})$ curve in Fig. 3 is in a good agreement with the data known from the literature and summarized by Goriletsky et al. [7]. However,

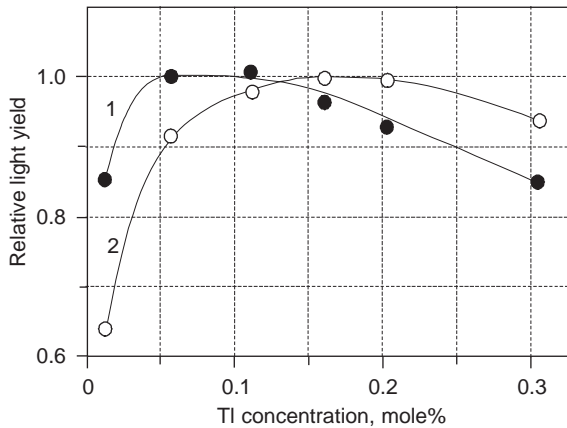


Fig. 3. Dependences of the light output on the Tl concentration for $t_{RC} = 6.4 \mu s$ at γ - (1) and α -excitation (2).

our results concerning the $L_{\gamma}(C_{Tl})$ dependence essentially differ from those presented in [2]. Such a discrepancy is observed for the interval of high Tl concentrations and consists in the fact that L_{γ} decreases at $C_{Tl} > 0.12\%$ in the case when a “blue” photomultiplier is used [7,8]. The so-called “concentration quenching” does not take place with “red” photoreceivers [7]. The paradox lies in the fact that Gwin and Murray [2] used a “blue” photomultiplier with S-20 photocathode response.

Dependences of the α/γ ratio on the shaping time are shown in Fig. 4 for the crystals with different Tl concentrations.

Similar dependence was earlier obtained by Kilgus et al. [9] for $C_{Tl} = 0.1\%$ only as well as in Refs. [10,14] where the Tl concentrations were not specified. For comparison, the data of Ref. [9] are presented on Curve 5.

As is seen, our results for $C_{Tl} = 0.11\%$ are in a good agreement with the literature data. It should be noted that our results span a wider interval of C_{Tl} . The data presented in Fig. 4 show that there exists a simple way to increase the α/γ ratio by diminishing the shaping time. Though in this case the light output decreases to some extent and the energy resolution raises, proper choice of particular t_{RC} values allow to increase the α/γ ratio, the value of light output remaining rather high.

The dependences of the α/γ ratio on the C_{Tl} for different shaping times are presented in Fig. 5. These data permit to conclude that the α/γ ratio

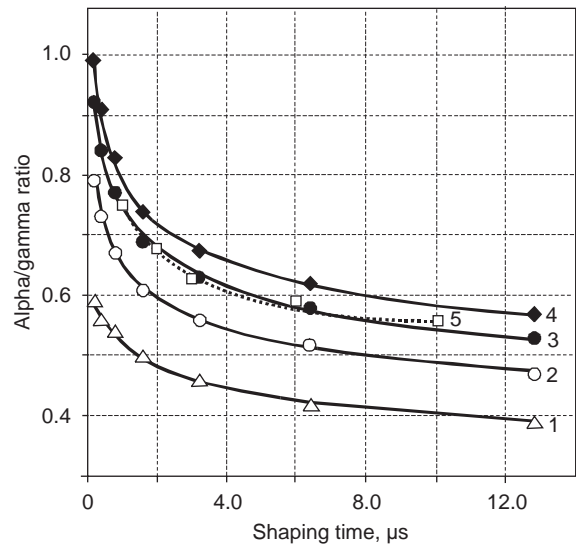


Fig. 4. Dependences of the α/γ -ratio on the shaping time for the crystals with different C_{Tl} : 1–0.013; 2–0.06; 3–0.11; 4–0.3%. Curve 5 represents data from Ref. [9] for the $C_{Tl} = 0.1\%$.

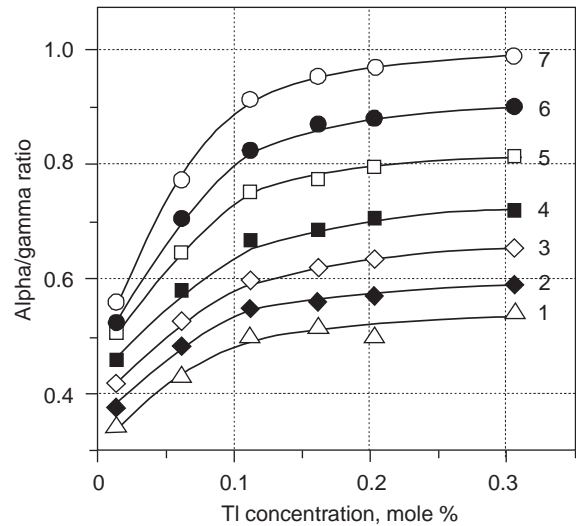


Fig. 5. Dependences of the α/γ -ratio on the C_{Tl} for the different shaping times: 1–12.8 μs ; 2–6.4; 3–3.2; 4–1.6; 5–0.8; 6–0.4; 7–0.2 μs .

grows with the increase of the C_{Tl} at any t_{RC} . Similar dependence was obtained in Ref. [3] for CsI:Tl crystals grown in vacuum for the $t_{RC} = 1.0 \mu s$ only. Thus, our experimental results confirm the dependences obtained in Ref. [3] and

totally disprove the conclusions made by Gwin and Murray [2].

4. Discussion

The results presented above as well as the literature data [3,5–7] allow to come to the conclusion that for the obtaining of both α -detectors with maximum light output and reference-point detectors with better values of the α/γ ratio, CsI:Tl crystals with elevated activator contents are required. Such a conclusion is general enough since analogous results for NaI:Tl are contained in the earlier papers [4,12]. Analysis of the concentration dependences of the L_α and L_γ for CsI:Na obtained by Panova et al. [15] also confirms this statement.

First of all, let us discuss the shape of the curves $L(C_{\text{Tl}})$ for different types of excitation. Here we shall use the term C^* which denotes the Tl concentration at the initial point of the plateau in dependence $L(C_{\text{Tl}})$ which corresponds to the minimum content of Tl within the interval of its optimum values. The fact that $C_\alpha^* > C_\gamma^*$ signifies the difference in the dE/dx for electrons and α -particles. The simplest explanation of this fact gives the SM. Recombination of electrons and holes (e–h pairs) in the host lattice comes to a fast ($\tau \approx 10$ ns) exciton luminescence ($\lambda = 310$ nm in CsI) with an efficiency of about 3000 MeV^{-1} , relatively low in comparison with activator luminescence in CsI:Tl ($\sim 50\,000 \text{ MeV}^{-1}$, [16]). For the used photomultipliers with glass windows such low light yield in the UV region of the spectrum may be neglected. The $C_{\text{Tl}} = 0.05\%$ is sufficient for suppression of inefficient recombination of e–h pairs in the track of electron ($dE/dx \sim 1 \text{ keV cm}^2/\text{mg}$) though too low for the track of α -particles ($dE/dx \sim 450 \text{ keV cm}^2/\text{mg}$) [1,4]. Thus, the SM concept is necessary to explain the behavior of the $L_\gamma(C_{\text{Tl}})$ and $L_\alpha(C_{\text{Tl}})$ curves [1,5]. As far as we know, there does not exist another proper explanation of it. Just these results concerning the change in CsI:Tl luminescence spectrum with the growth of the C_{Tl} were obtained by Gwin and Murray in the second part of their well-known paper [2].

Our results for CsI:Tl crystals as well as the results of Kubota et al. [4] for NaI:Tl, confirm the SM validity. Actually, by increasing the Tl concentration the difference between the values of L/E for γ -rays and α -particles may be partially leveled. As the SM uses the total luminescence yield, but not of light output, for verification of the SM the α/γ value is to be taken at the highest t_{RC} . From the data of Fig. 5 it follows that for $t_{\text{RC}} = 12.8 \mu\text{s}$ the α/γ ratio increases from 0.4 to 0.6 as the C_{Tl} grows.

We have already mentioned that our results are in a good agreement with the other literature data. There exists only one paper—[2]—the conclusions of which totally contradict our data concerning both the influence of C_{Tl} on the α/γ ratio and the character of the dependence $L_\gamma(C_{\text{Tl}})$ in the region of concentration quenching.

The paper [2] seems to be the only investigation in which frames the value of light output were measured on the crystals of different origin: there were used both the crystals obtained from Harshaw Chem Co and those grown by the authors in an available furnace. The quality of CsI:Tl crystals, especially for $C_{\text{Tl}} > 0.1\%$, strongly depends on the growth technology and the personnel's skill. Therefore, the dependence $L_\gamma(C_{\text{Tl}})$ obtained in Ref. [2] reflects not only the change in C_{Tl} , but also the growth method used. Such a circumstance seems to explain the fact that the authors did not observe concentration quenching of L_γ in the region of elevated Tl contents though they used a “blue” photomultiplier. However, this cannot explain why the α/γ ratio is independent of the C_{Tl} .

As for the concentration interval $C_\gamma^* < C_{\text{Tl}} < C_\alpha^*$ where $L_\gamma \approx \text{const}$, the conclusion made in [2] means that, due to some factor, the L_α retains its maximum values with decreasing C_{Tl} (see Fig. 3). In our opinion, the light yield for α -particles increased due to the presence of a sufficient quantity of luminescence centers in the thin surface layer caused by peculiarities of sample preparation technique.

It is well known that plastic deformation sharply increases the number of blue emission centers in CsI [17] and CsI:Na [18,19]. Grinding of the crystals leads to formation of new emission centers

in a thin surface-adjacent layer only [20]. Scintillation properties in the volume of the crystal remain practically unchanged. The nature of additional centers is known: they are the anion vacancies (V_a^+) which form the dipoles $V_a^+ - V_c^-$ [17] resulting from dislocation sliding. In accordance with the data of [21], for sliding of the dislocations existing in CsI, it is sufficient to apply a stress of about $(1-2) \text{ g/mm}^2$ and this threshold is easily exceeded even at “soft” sample polishing. CsI crystals have no cleavage planes, therefore the procedure of sample preparation always implies, at least, the operation of polish.

For pure CsI the effect of polishing is so substantial that near the surface luminescence of self trapped excitons is extremely weak at room temperature. Instead, there is excited slow emission of bound excitons, though in the crystal bulk fast luminescence is induced, as shown by Nishimura et al. [22]. For almost complete suppression of the fast emission in CsI, the concentration of defects in the surface-adjacent layer is to be rather high: $\sim(6-8)10^{17} \text{ cm}^{-3}$. This estimation was obtained from the fact that in CsI:Na crystals the $C_\gamma^* = (9-10)10^{17} \text{ cm}^{-3}$ [15]. If the concentration of sodium is lower than the said value, fast emission at 310 nm is clearly seen in the radioluminescence spectra. If this concentration is higher, then the luminescence of self trapped excitons will be completely suppressed by the activator [23]. According to the data of the second part of Ref. [2] for CsI:Tl, luminescence at 310 nm disappears at $C_{\text{Tl}} > 2 \times 10^{17} \text{ cm}^{-3}$. However, it should be taken into account that the A-band of Tl^+ absorption strongly overlaps the luminescence at 310 nm, and in this case the estimation is undervalued. In CsI:CO₃ scintillator the emission center is a dipole

CO₃²⁻-V_a [24], and for suppression of the 310 nm luminescence it is necessary to introduce CO₃²⁻ ions in $(6-8)10^{17} \text{ cm}^{-3}$ quantities. We consider this value to be the most probable quantity of $V_a^+ - V_c^-$ dipoles which arise within the surface-adjacent layer after polishing (C_D).

As is known, plastic deformation noticeably distorts the luminescence spectrum of CsI:Tl [25]. According to our data, this effect manifests itself explicitly if the Tl concentration is approximately equal to C_γ^* or lower. The data presented in Fig. 1 directly demonstrate the role of point defects arising within the surface-adjacent layer under the influence of deformation. As is seen from this figure, position of the α -peak in the pulse height spectrum depends on the time which has passed after the polishing. The values of L_α are stabilized after 6–8 days of crystal aging under the normal conditions. The values of the α/γ ratio determined after polishing the entrance surface and those measured in 14 days of aging, are presented in the table.

As is seen from Table 1 and Fig. 1, polish of the entrance surface influences neither L_γ nor L_α for the crystals with $C_{\text{Tl}} \gg C_D$. Another situation is observed for the crystals with $C_{\text{Tl}} \leq C_D$: in this case the light output of α -particles sharply increases, while that of 662 keV photons (which excite mainly the scintillator's bulk) remains practically the same. Such an effect causes temporary increase of the α/γ ratio. The conclusions made in Ref. [2] seem to be explained by the said fact. In the experimental part of the mentioned paper there is a circumstance which concerns the method of sample preparation. The authors used the samples with a diameter of 25 mm glued to the light guide and then ground to obtain a thickness of 2 mm. If

Table 1
Changes in a α/γ ratio after polishing and in the process of ageing

Tl concentration	α/γ -ratio for $t_{\text{RC}} = 4 \mu\text{s}$			
mol%	cm^{-3}	Before polishing	After polishing	In 14 days
5×10^{-4}	5×10^{16}	0.28	0.511	0.281
6×10^{-3}	6×10^{17}	0.321	0.505	0.32
2×10^{-2}	2×10^{18}	0.512	0.588	0.51
1.5×10^{-1}	1.5×10^{19}	0.602	0.605	0.6

they used the same light guide, then the measurements had to be performed immediately after preparation of the crystals when L_α could not relax to its natural values. In fact the concentration of emission centers was defined by C_D , not by $C_{Ti} \leq C_D$.

The unusual shape of the dependence L/E vs. E_α shown in Fig. 4 of the paper [2] confirms this statement. The value of specific light yield sharply increases for α -particles with $E_\alpha < 1$ MeV, i.e. for the particles absorbed at the crystal surface. While studying analogous dependences for aged CsI:Tl samples excited by protons [26] or α -particles [27] such a situation was not observed. On the contrary, the authors of the mentioned papers came to the conclusion concerning the existence of a layer with a low scintillation efficiency near the surface. In CsI:Tl the depth of this layer is insignificant and runs into fractions of micrometer [27].

Unfortunately, erroneous conclusions from the results obtained in Ref. [2] are widely cited in reviews and reference books devoted to scintillation method [8,28–30]. However, the firms producing the detectors with α -reference point seem to be guided by the conclusions made in Ref. [3], since the catalogues [31,32] advertise the detectors for which the α/γ ratio can be chosen within certain limits at the customer's request.

The value of α/γ ratio also depends on the energy of γ -rays. As recently shown by Kudin et al. [33], the duration of scintillations decreases as the energy of photons falls, and for $E_\gamma = 5.9$ keV the dependence $L_\gamma(t_{RC})$ corresponds rather to Curve 1 in Fig. 2 for α -particles then to Curve 2 for 662 keV photons. Fortunately, in most cases this circumstance must not be taken into account in practice, since the detectors with α -reference point are not used for registration of such low-energy photons.

5. Conclusions

Dependences of light yield and α/γ ratio on the Tl concentration have been studied within a wide range of shaping times. It is shown that the α/γ ratio essentially depends on the Tl concentration,

and for the obtaining of high-quality α -detectors and the detectors with α -reference point CsI:Tl crystals with elevated activator concentrations, are required. Proper combination of certain parameters such as the Tl concentration in the crystal and optimum time of shaping in the electronic system allows to obtain detectors with rather high light output for γ -rays, α -particles and α/γ ratio values.

It has been shown that both the light yield at α -excitation and the α/γ -ratio depends on the time of crystal storage after polishing. On the basis of the idea of the formation of deformation-induced point defects in a thin surface-adjacent layer, the causes of the temporary increasing in light yield for α -particle are explained. Given is an explanation of the results obtained by Gwin and Murray [2] concerning the relation between the α/γ ratio and the Tl concentration. In our opinion, the paper [2] cannot be taken into account for experimental verification of theoretical models.

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