

SCINTILLATION CHARACTERISTICS OF HEAVILY DOPED CsI:Tl,IO₃ CRYSTALS

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The effect of IO₃⁻ ions on spectrometric characteristic of CsI:Tl crystal has been considered. It has been shown that co-doping of CsI:Tl crystal by IO₃⁻ anions permits to obtain clear ingots with increased thallium concentration up to $C_{Tl} \sim 0,9$ mole % in which the concentration quenching of photo- or radioluminescence do not observed. It has been shown that the decay of solid solution in CsI:Tl,IO₃ crystals is not observed, at least up to $C_{Tl} \sim 0.5$ mole %, as evidenced by a good energy resolution ($R = 6.3\%$) of samples with the specified Tl concentration and this also evidenced by a larger segregation coefficient of thallium in CsI:Tl,IO₃ crystals ($k = 0,24$ contrary to $k = 0,19$ in CsI:Tl). An explanation of thallium solid solution stability in CsI matrix has been proposed which based on experimental fact of Tl⁺ ... IO₃⁻ complex formation. Compensation of elastic stresses of opposite sign due to complex formation results in preventing of nucleation for solid solution decay.

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INTRODUCTION

CsI:Tl crystals are widely used in various fields of science and scintillation technique [1]. The obvious advantage of these crystals is the ability not only to register charged particles [2], but also to identify them by the pulse shape [3, 4]. The identification of particles by atomic number Z and mass A predetermines the stringent requirements for the composition of the scintillation material [5] and the stability of its characteristics over time [6]. The need to identify particles arises in such tasks as registration of fission fragments [7] or products of nuclear physical reactions for instance in the FAZIA (for π A and Z identification array) project [8].

The requirements for the composition of the scintillator primarily relate to the activator concentration – C_{Tl} . It is well known that optimal concentration C_{Tl}^* depends on particle type and increase with density of energy loss dE/dx in particle track [3]. The value of C_{Tl}^* for fission fragments is more than an order of magnitude higher than the optimum concentration for detecting γ -rays [9]. Thus, for detection and identification of charged particles including heavy ions, the crystals with $C_{Tl} \sim 1$ mole % are necessary [10].

Detection of charged particles is complicated by the fact that concentration quenching of luminescence is observed in heavily doped CsI:Tl crystals [3, 10]. The light yield decrease (at excitation by gamma quanta from ¹³⁷Cs) for CsI:Tl crystals is observed starting of the $C_{Tl} \geq 0.2$ mole %, at the same time the energy resolution is degraded [5, 11]. It is believed [5] that the spinodal decay of activator solid solution in the CsI matrix cause of the concentration quenching of light yield.

Attempts have been made [11, 12] to obtain crystals with an increased concentration of thallium. Crystals from GB (USA) and SCIONIX (Holland) were studied in [12], it has been shown that heavily doped CsI:Tl crystals are characterized by a sharply inhomogeneous distribution of thallium, both along the growth axis and in the transverse direction. Concentration

inhomogeneity are pronounced at the $C_{Tl} \sim 0,5$ mole % [12] and caused by decay of thallium solid solution [5]. Spinodal decay of solid solution results in degradation of light yield [5, 11] and energy resolution of scintillator especially.

On the basis of literature data and our own experience, it can be assumed that doping of crystal with impurity-stabilizer [13] can prevent decomposition of solid solution at the initial stage of formation of paired and more complex activator centers. Such an approach justified itself for NaI:Tl scintillation material which is doped additionally with IO₃⁻-ions [14], and as a result the thallium concentration limit has been increased threefold. A similar result [13] was obtained for LiI:Eu scintillation material. It has been shown that the introducing of the O²⁻ anions into the crystal lattice permits to increase the limiting concentration of europium significantly.

The aim of present paper consists in obtaining of CsI:Tl,IO₃ crystals with increased thallium concentration and investigation of main spectrometric characteristics of this scintillation material.

EXPERIMENTAL PROCEDURE

The crystals were grown by directional crystallization in evacuated quartz ampoules according to Stockbarger technique. The technique for growing of CsI:Tl crystal without additional co-doping with Na⁺ cations is described in detail in [15].

Grown ingots had the following dimensions: ~ 82 mm in diameter and ~ 120 mm in height. If the activator concentration did not exceed value of the $C_{Tl} \leq 0.2$ %, obtained ingots were transparent and had the correct shape, i.e. convex crystallization front and smooth side surface. In the case of growing crystals with the highest thallium concentration, the temperature in the crystallization zone was decreased by 20°C, and the pulling rate was halved to 1 mm/hour. However, the ingots grown had a concave crystallization front. The funnel-shaped depression in upper part of ingot was filled with a fine-crystalline mixture of individual components.

The concentration of thallium in heavily doped samples was determined by a specially developed technique described in [16]. Hereinafter, the Tl concentration is given in molar fractions of percent.

A distinctive feature of heavily doped CsI:Tl crystals was the non-uniform distribution of activator, both in height of ingot and in cross section. The C_{Tl} values could differ twice in adjacent parts of ingot for example, at a distance of ~ 1.5 mm. The local scatter of C_{Tl} values was studied by point-to-point chemical analysis using laser surface evaporation [17]. The size of the evaporation crater was approximately 0.5 mm, and the neighboring craters were separated from each other by a distance of 2 mm. At an average C_{Tl} of about 0.5 %, the spread of the concentration values exceeded 100 % in the cross section of ingot, where the activator concentration should be the same. Our results on the inhomogeneous distribution of the activator at distances of about 1 mm are fully confirmed by the data of [12]. Due to the large scatter of data on C_{Tl} , the thallium segregation coefficient in heavily doped ingots was not determined. Samples cut from the transparent part of such ingots had unsatisfactory spectrometric characteristics.

Transparent crystals with $C_{Tl} > 0.2$ % which had a standard shape and regular thallium distribution over the height of the ingot could be obtained by a similar method of growing, but in a reaction gas atmosphere. The gas atmosphere consisted of argon and oxygen, the pressure in the ampoule was about 10.1 kPa.

Grown crystals usually contained some amount of CsIO₃ impurity. In Fig. 1 the IR transmission spectrum is presented for CsI:Tl,IO₃ crystal at room temperature. As it seen from data of figure there are two overlapping absorption bands located at 804 and 799 cm⁻¹ which are characteristic for stretch vibration of IO₃⁻ ions [18].

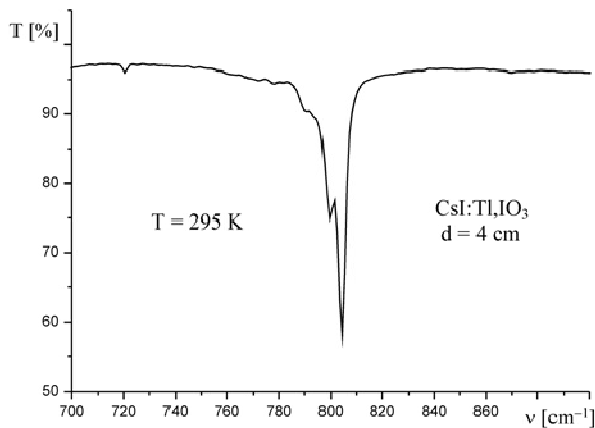


Fig. 1 – Infra red transmission spectrum of **CsI:IO₃** crystal with $C_i = 2 \cdot 10^{-4}$ mole % at room temperature.

According to chemical analysis, the content of IO₃⁻ ions in this sample is equal to the $C_i = 2 \cdot 10^{-4}$ %. Knowing the absorption coefficient k_{804} in the most intense band of 804 cm⁻¹, one can determine the concentration of iodate ions by the empirical relation:

$$C_i = 1,47 \cdot 10^{-3} k_{804},$$

where the coefficient k_{804} is expressed in cm⁻¹, and the C_i in mole %. The proportionality coefficient ($\xi =$

$1,47 \cdot 10^{-3}$) found was close to the known one [14] for determining the concentration of iodate ions in NaI crystals ($\xi = 1,52 \cdot 10^{-3}$).

Fig. 2 shows an example of the activator distribution by ingot volume in the process of directional crystallization in CsI:Tl (curve 1) and CsI:Tl,IO₃ crystals (curve 2). It can be seen that in the case of a relatively small activator concentration in the melt ($C_0 = 0.3$ %), the distribution of C_{Tl} along the volume of ingot is described by smooth curves with gradually increasing thallium concentration.

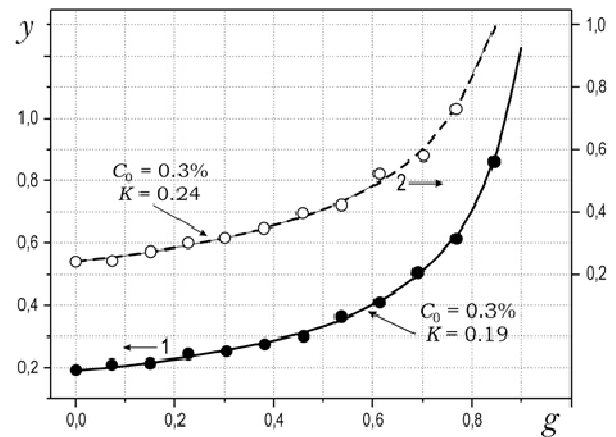


Fig. 2 – Thallium distribution curves during directional crystallization for **CsI:TI** (1) scintillation material and **CsI:TI,IO₃** (2). The points represent experimental data and the solid lines are according to Pfann's law. Надпись

Determination of segregation coefficient K carried out in " $\ln y$ vs $\ln g$ " coordinates, where $y = C_{Tl}/C_0$ is a relative Tl concentration, and $g = m/m_0$ denotes the proportion of crystallized melt. The calculation gave the value $K = 0.19$ in the case of CsI:Tl material crystallization ($C_0 = 0.3$ %, which corresponds to the initial $C_{Tl} \sim 0.06$ %). The obtained value of K within the limits of measurement error corresponds to the well-known equilibrium segregation coefficient $K = 0.18 \pm 0,01$, which is given in [19]. In the case of CsI:Tl,IO₃ material growing, a markedly larger segregation coefficient was obtained: $K = 0.24$.

To determine the spectrometric characteristics of scintillator the samples measuring $10 \times 10 \times 10$ mm were made for light output determination and $\varnothing 20 \times 4$ mm for measuring the luminescence. The polishing of the samples was carried out taking into account the recommendations of works [20, 21] to prevent the formation of a dead layer. Measurements of the light yield were carried out on a spectrometric bench with a Hamamatsu S3590-08 photodiode relative to the standard. The accuracy of light yield determining by this method was $\pm 3.2\%$ [15].

RESULTS

As noted in the introduction, the optimal concentration of thallium for detection of light ions differs from that for γ -rays. Therefore, for a more complete characterization of the scintillation material let us firstly consider the crystals with $C_{Tl} \sim 0,1$ % which is enough for γ -

rays detection and then with $C_{Tl} \sim 0,5 \%$ (more suitable amount for light ions registration).

Crystals with $C_{Tl} \sim 0,1\%$. First of all let us consider the luminescence properties of codoped crystal. Curve 2 on Fig. 3 presents the radioluminescence spectrum of CsI:Tl,IO₃ crystal. Curve 1 corresponds to emission spectrum of CsI:Tl standard with the same activator concentration ($C_{Tl} = 9,5 \cdot 10^{-2}\%$). It is seen from the date of Fig. 3 that two curves are very similar on the shape as well as position of maxima. However, two noticeable differences are found. First, despite the same C_{Tl} in two crystals under investigation the maximum of emission is located at low wavelength for CsI:Tl,IO₃ material. It is known [10] that maximum of emission spectrum shifts to longer wavelengths with C_{Tl} increasing. According to [10] the maximum of peak on curve 2 corresponds to the apparent $C_{Tl} \sim 7 \cdot 10^{-2}\%$ instead indicated thallium concentration $9,5 \cdot 10^{-2}\%$ in studied CsI:Tl,IO₃ crystal.

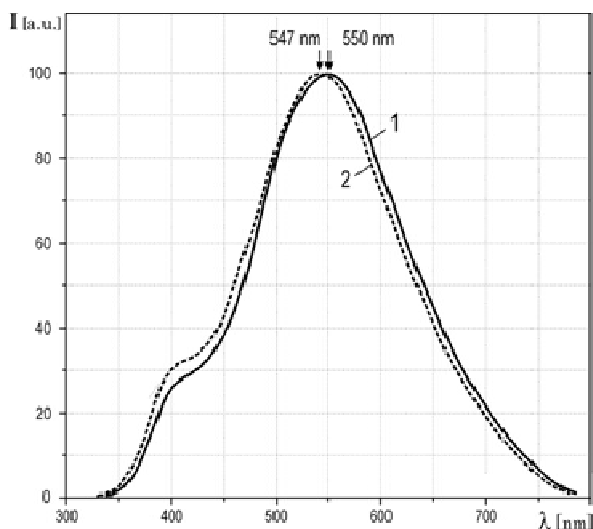


Fig. 3. Spectra of radioluminescence for CsI: Tl (1) and CsI: Tl, IO₃ (2) crystals with the same thallium concentration. T = 295K.

Secondly, the luminescence intensity for CsI:Tl,IO₃ crystals is lower and this decrease is greater, the greater the concentration of IO₃⁻ ions in crystal lattice. At dopant concentration of $C_i \sim 2 \cdot 10^{-4}\%$ the luminosity η_{RL} is lower by 8-10% relative to the yield of CsI:Tl crystal. Value of the η_{RL} has been determined as a square under curve 2 on Fig. 3 before its normalization if the luminescence is presented as J vs $h\nu$ (emission intensity on photon energy).

Analogical decreasing of the yield is observed for photoluminescence too. At excitation in A-band of activator absorption ($\lambda_{ex} = 299$ nm) the luminosity η_{ph} is also lower by 8-10 % relative to the reference.

For the convenience of comparing crystals of different impurity composition data on the values of η_{RL} and η_{ph} are summarized in Table 1 as well as the results on light yield and energy resolution.

Spectrometric characteristics. Pulse height spectra are presented in Fig. 4 for crystals of two types. As it seen from the data of curve 2 in this figure the position of full absorption peak is shifted to low energy side for

CsI: Tl, IO₃ crystal in compare with standard. Light yield decreasing is about 8-10% for crystal doped by IO₃⁻ ions and this value coincides with degradation of photo- and radio luminescence. Considering the lower light output, it can be concluded that the energy resolution of CsI: Tl, IO₃ crystal should be slightly worse than that of the reference. However, this is not the case, despite a noticeable decrease in light yield the R value does not deteriorate, which indicates that the crystals are homogeneous.

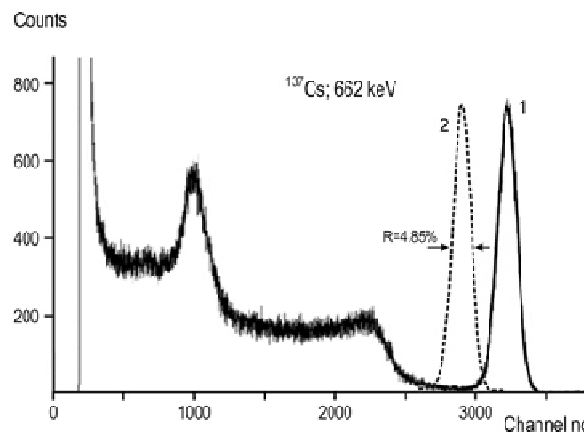


Fig. 4. Pulse height spectra of CsI: Tl (1) and CsI: Tl, IO₃ (2) crystals coupled to photodiode Hamamatsu S3590-08. Size of scintillators are 10×10×10 MM

Table 1. Scintillation and luminescent parameters for crystals of different composition

Parameter	CsI: Tl	CsI: Tl, CO ₃	CsI: Tl, IO ₃	Note
$C_{Tl} = 9 \cdot 10^{-2}\%$				
$\eta_{RL}, \%$	100	100	91	
$\eta_{PL}, \%$	100	100	92	
$L, \%$	100	100	92	PMT
	100	100	90	PD
$R, \%$	5,8	5,8	5,8	PMT
	4,8	4,7	4,8	PD
$C_{Tl} = 4,9 \cdot 10^{-1}\%$				
$\eta_{RL}, \%$	100	100	91	
$L, \%$	100	100	92	PMT
	100	100	90	PD
$R, \%$	5,8	5,8	5,8	PMT
	4,8	4,7	4,8	PD
α/γ	0,69	0,72	0,77	PMT

Radiation hardness. First of all it should be noted that CsI: Tl, IO₃ crystals are photo-stable, i.e. when stored in the day light pink color does not occur. Radiation hardness of CsI: Tl, IO₃ crystal was studied on samples of 2.5×2.5×4 cm³ size relative to the reference. For this test the samples were cut from initial part of ingot where the C_{Tl} slightly increase from 0.096 to 0.134 %. As a reference, the CsI: Tl, CO₃ scintillation material was chosen because this crystal is the most accessible, its radiation hardness satisfies most technical applications [22, 23], and the material homogeneity allows one to make long samples. Data on radiation hardness are shown in Table 2. As it seen, the irradiation with doses

of 10 and 100 Gy results in the negligible decrease of light yield $\Delta L/L_0$ for the CsI:Tl,IO₃ scintillation material. Such decrease even less than for the reference. Taking into account that formal requirement [23] to the light yield degradation is ($\Delta L/L_0 \leq 20\%$), it can be concluded that radiation hardness of CsI:Tl,IO₃ crystal is quite satisfactory and exceeds the γ -irradiation dose of 100 Gy.

Table 2.
Radiation hardness for crystals of different composition

Scintillation material		$\Delta L/L_0, \%$		
type	C_{Tl}	Before	10 Gy	100 Gy
Samples of $2.5 \times 2.5 \times 4 \text{ cm}^3$ size				
CsI:Tl	0.094 ± 0.015	0	7	12
CsI:Tl,CO ₃	0.11	0	8	15
CsI:Tl,IO ₃	0.115 ± 0.015	0	6	12
CsI:Tl,CO ₃	0.35	0	14	34
CsI:Tl,IO ₃	0.25 – 0.49	0	10	18
Sample of $\varnothing 6.3 \times 20 \text{ cm}^3$ size				
CsI:Tl,IO ₃	0.11 - 0,38	0	7.5	15

Crystals with $C_{Tl} \sim 0.49\%$. For heavily doped CsI:Tl,IO₃ crystals, first of all, the radioluminescence spectra were studied. It turned out that for such crystals, regularities similar to those described above are observed. The maximum of emission spectrum for CsI:Tl,IO₃ crystals is shifted towards short wavelengths, similarly to the data in Fig. 3. The intensity of emission and the luminosity η_{RL} for CsI:Tl,IO₃ material is 8-10% lower compared to CsI:Tl crystal, see data in Table 1. The luminosity η_{RL} remains constant with C_{Tl} increasing up to 1% for CsI:Tl,IO₃ crystals with the $C_i \approx 2 \cdot 10^{-4}\%$.

The spectrometric characteristics were measured on a sample of $2.5 \times 2.5 \times 4 \text{ cm}^3$, this size corresponds to prototype in FAZIA project [8]. Polymer paper Tyvek served as a reflector, the immersion layer was created with SKTN rubber, and a "red" PMT Hamamatsu R669 was used as a photoreceiver. Pulse height spectrum of CsI:Tl,IO₃ crystal with $C_{Tl} \sim 0.49\%$ is presented on Fig. 4 as a curve 2. Curve 1 corresponds to standard of the same size. As it seen from the data of figure the position of full absorption peak is shifted to low energy side for CsI:Tl,IO₃ crystal relative to the reference. The light yield decrease for heavily doped crystals consists about 8%. Despite the smaller light yield, the energy resolution does not deteriorate and amounts to the same 6.3% as for a reference sample of a similar size.

Alpha to gamma ratio. Quality of scintillation material to detecting the charged particles usually is characterized by the α/γ -ratio [24], that means specific light yield of alpha-particle divided on gamma-ray one. Experimental technique for α/γ -ratio measurement is described in detail in Ref. [24]. During sample preparation the recommendations of [6, 24] regarding the relaxation of the surface damaged layer were taken into account. It turned out that the α/γ -ratio for CsI:Tl,IO₃ crystals with $C_{Tl} \sim 0.49\%$ corresponds to the best values given in [24]. It is well known that the

α/γ -ratio increases with the growth of the C_{Tl} , therefore, the obtained result at first glance seems obvious. It should be noted, however, that the distinctive feature of CsI:Tl,IO₃ crystals is a good energy resolution at γ -ray detection as well as α -particle.

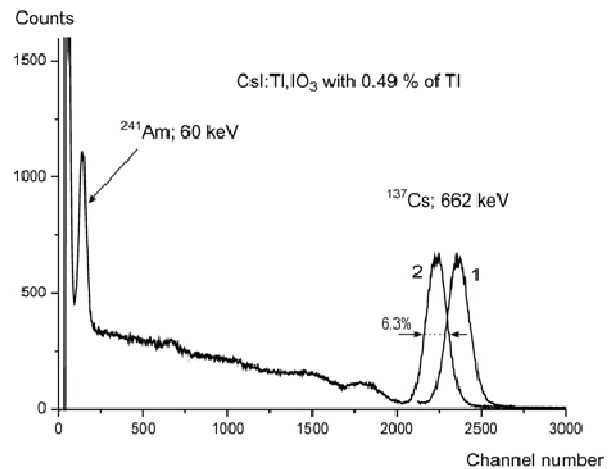


Fig. 5. Pulse height spectrum of CsI:Tl crystal (1) and CsI:Tl,IO₃ (2) with $C_{Tl} = 0.49\%$ coupled to Hamamatsu R669 photomultiplier. Size of scintillator is $2.5 \times 2.5 \times 4 \text{ cm}$.

Radiation hardness of CsI:Tl,IO₃ crystals was studied on samples of two types. The first type, $2.5 \times 2.5 \times 4 \text{ cm}^3$ in size, corresponds to the supposed detection elements for the prototype in the FAZIA project [8], and the other type – $\varnothing 6.3 \times 20 \text{ cm}^3$ – to the maximum size in our experiments. The samples were cut along the growth axis; therefore, the C_{Tl} distribution on the height was non-uniform, the values of thallium concentration in bottom and the top of the sample are indicated in Tab 2. Degradation of light yield after irradiation with doses of 10 and 100 Gy is presented in Tab 2 for heavily doped crystals. It can be concluded from the data of table that radiation hardness of heavily doped CsI:Tl,IO₃ crystal do not inferior to standard.

DISCUSSION

So, it is shown that activator concentration in the CsI:Tl,IO₃ scintillation material can be increased up to 0.49%. This value is three times the allowable concentration in CsI:Tl crystal. The resulting heavily doped crystals are not inferior to the standard in energy resolution and exceed it in the values of the α/γ -ratio. Note also that the measured spectrometric characteristics are stable over time.

It is well known that the decay of a solid solution can be significantly slowed down by the introduction of stabilizer impurities [13, 14]. An idea of this approach is that part of the activator is bound in complexes with the dopant. In crystals with isovalent impurities, complexes arise due to the elastic interaction of partners. Stable complexes are formed if the elastic stresses around the constituent ions have opposite signs. This situation is typical for Tl⁺ cations and IO₃⁻ anions. The ionic radius of Tl⁺ is equal to 1.47 Å and differs from that of Cs⁺ (1.67 Å) by 12%. The ionic radius of the impurity anion IO₃⁻ is 2.4 Å, which is 8.3% more than the size of the base anion I⁻ (2.2 Å). Therefore, in the case when the

Tl^+ cation and IO_3^- anion occupy the neighboring sites of the crystal lattice, local compensation of the elastic energy occurs. A schematic representation of such a mutual arrangement of impurity ions is shown in Fig. 6.

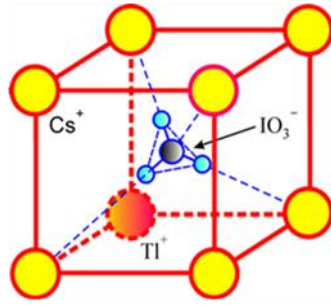


Fig. 6. A schematic representation of interacting ions of the Tl^+ and IO_3^- in the unit cell of CsI crystal lattice.

Experimental confirmation of this statement is shown in Fig. 7, which shows the change in the vibration absorption of the molecular anion IO_3^- in the CsI lattice [25]. At a temperature of 4 K, according to the data of [25], in the IR spectrum of CsI crystal containing IO_3^- ions, two absorption bands are observed at 785 and 800 cm^{-1} , which are associated with symmetric and asymmetrical stretching vibrations of this ion (curve 1 in Fig. 7). This ion looks like a flattened pyramid, in which the I–O bond length is equal to 1.81 Å, and the angle between the I–O bonds is 97°, with point symmetry C_{3v} [18]. If it is embedded in the body centered lattice instead of halogen so that its C_3 axis coincides with the $\langle 1;1;1 \rangle$ direction of the cell, then its local symmetry does not decrease, and the degeneracy is not removed from E type oscillations.

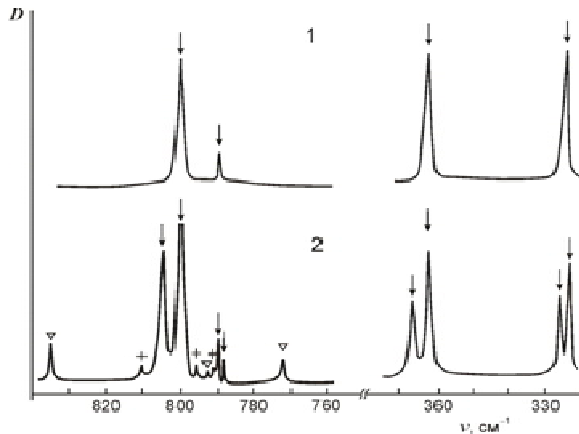


Fig. 7. Infra red absorption spectra of $CsI:IO_3$ (1) crystal and $CsI:Tl,IO_3$ (2) at 4K [25]

It can be seen from analysis of data on Fig. 7 that after the introduction of thallium, the absorption spectrum is modified (curve 2), new bands appear that are associated with the formation of the $Tl^+ \dots IO_3^-$ complexes. Analysis of the spectra and identification of the absorption bands led the authors [25] to conclude that the Tl^+ cation occupy two places relative to the molecular anion. The activator can occupy a position along the C_3 axis (two positions without symmetry breaking, see Fig. 6), or in other neighboring nodes,

which leads to the lifting of the degeneracy due to the symmetry breaking.

Thus, the assumption that the iodate ions form complexes with an activator gets its experimental confirmation. Due to this the elastic energy of the lattice is reduced, solid solution becomes isomorphic, segregation coefficient increases and solid solution of thallium in CsI acquires stability.

Now consider the second feature of $CsI:Tl,IO_3$ crystals: – lower light output. Since the decrease in light output is directly associated with a decrease in luminosity (i.e. the yield of emission at intracenter excitation, see data about η_{ph} и η_{RL} in Table 1) it can be assumed that part of activator centers passes from the excited state to the ground state without emission. It is reasonable to assume that the quenching centers are $Tl^+ \dots IO_3^-$ complexes. Since the number of such complexes should increase with the C_i increasing, the light yield should decrease in proportion to the C_i . Indeed, both the luminosity and the light yield decrease with C_i increasing. However, when growing $CsI:Tl,IO_3$ crystals with different contents of iodate ions, a curious feature was found.

Special experiments have established that the content of IO_3^- ions in crystal lattice depends to a decisive extent on the partial pressure of oxygen above the melt. The dependence of C_i on the partial pressure of O_2 has a threshold character, as can be seen from the course of curve 1 in Fig. 8. Under our crystal growth conditions, the threshold pressure was ~ 2.1 kPa. If the partial pressure of oxygen was less than the specified value, the resulting ingot did not contain IO_3^- ions. For such ingots, the presence of CO_3^{2-} ions was typical. Concentration of carbonate ions was about $(2 - 4) \cdot 10^{-4}\%$ which is characteristic for scintillation material $CsI:Tl,CO_3$ [26]. It is known that such amounts of impurities do not lead to the deterioration of the spectrometric characteristics of activated crystals [27].

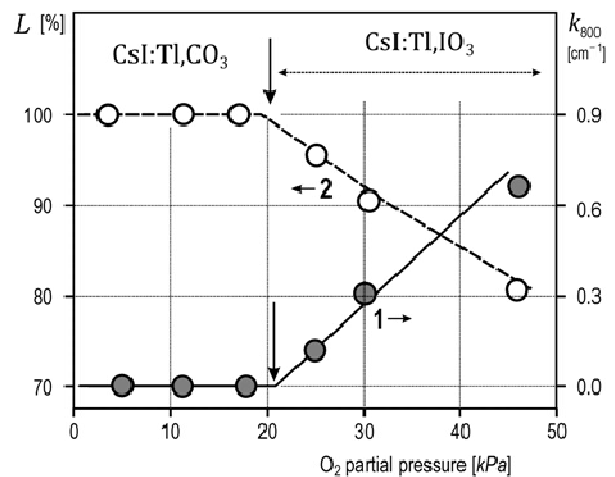


Fig. 8. Concentration of IO_3^- ions (1) in crystal lattice and light yield (2) of grown crystals on oxygen partial pressure under the melt

If the oxygen pressure exceeded the threshold value, the grown ingots were dyed with iodine and contained IO_3^- ions, the concentration of which increased in proportion to the O_2 partial pressure. In the area above the threshold there is a decrease in light yield. From the

data in fig. 8 it follows that there is a clear dependence – the larger the C_i , the smaller the light yield in the region above the threshold.

The existence of a clear correlation between the concentration of iodate ions and the decrease in light yield is a strong argument that the $Tl^+ \dots IO_3^-$ complexes are quenching centers. However, the concentration of such complexes is too low. Thus, thallium concentration usually is equal to 1000 ppm while the concentration of iodate ions (sufficient to drop the light yield by 10%) is 2 ppm. Therefore, we believe that in CsI crystals there are quenching centers, the concentration of which is an order of magnitude greater than the C_i .

It is known that alkali-halide crystals grown in air usually contain oxide (O^{2-}), peroxide (O_2^{2-}), and superoxide (O_2^-) oxygen ions. Bivalent anions in the CsI lattice manifest themselves by the blue emission of an anion vacancy, however, such emission is not observed in CsI: IO_3 crystals [28]. According to [14], monovalent O_2^- anions can play the role of quenching centers since their characteristic luminescence is not observed in sodium and cesium iodides. This question is the subject of further research.

The disadvantage of the proposed material is that the crystal growth is carried out in the reaction gas atmosphere. When growing such crystals by pulling on the seed, gas evolution at the melt leads to formation of bubbles and the capture of gas inclusions by the crystallization front. In the resulting crystal the gas channels are clearly visible, which leads to defective ingots.

CONCLUSION

The effect of IO_3^- ions on spectrometric characteristic of CsI: Tl crystal has been considered. It has been shown that co-doping of CsI: Tl crystal by IO_3^- anions permits to obtain clear ingots with increased thallium concentration up to $C_{Tl} \sim 0,9$ mole % in which the concentration quenching of photo- or radioluminescence do not observed. It has been shown that the decay of solid solution in CsI: Tl, IO_3 crystals is not observed, at least up to $C_{Tl} \sim 0.5$ mole %, as evidenced by a good energy resolution ($R = 6.3\%$) of samples with the specified Tl concentration and this also evidenced by a larger segregation coefficient of thallium in CsI: Tl, IO_3 crystals ($k = 0,24$ contrary to $k = 0,19$ in CsI: Tl).

An explanation of thallium solid solution stability in CsI matrix has been proposed which based on experimental fact of $Tl^+ \dots IO_3^-$ complex formation. Compensation of elastic stresses of opposite sign due to complex formation results in preventing of nucleation for solid solution decay.

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Сцинтиляційні характеристики сильно легованих кристалів CsI:Tl,IO₃

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Розглянуто вплив аніонів IO_3^- на спектрометричні характеристики кристалів CsI:Tl. Показано, що легування кристалів CsI:Tl іонами IO_3^- дозволяє отримувати прозорі злитки зі збільшеною концентрацією талію (до $C_{\text{Tl}} \sim 0,9\%$), котрі не мають ознак концентраційного гасіння фото- та радіюлюмінесценції. Показано, що розпад твердого розчину в кристалах CsI:Tl,IO₃ не спостерігається, по крайній мірі, до $C_{\text{Tl}} \sim 0,5\%$, про що свідчать високий світловий вихід, хороше енергетичне розділення ($R = 6,3\%$) зразків зі вказаною концентрацією і більший коефіцієнт розподілу талію у кристалах CsI:Tl,IO₃ ($k = 0,24$ проти $k = 0,19$ у CsI:Tl). Запропоновано пояснення більш високої стабільності твердого розчину Tl^+ в матриці CsI, яке базується на формуванні комплексів $\text{Tl}^+ - \text{IO}_3^-$. Компенсація пружних напружень протилежного знаку внаслідок взаємодії катіонів Tl^+ і аніонів IO_3^- запобігає створенню зародків розпаду.

Сцинтилляционные характеристики сильно легированных кристаллов CsI:Tl,IO₃

А.Л. Шпилинская, А.М. Кудин, Л.Н. Трефилова, Д.И. Зосим

Рассмотрено влияние анионов IO_3^- на спектрометрические характеристики кристаллов CsI:Tl. Показано, что легирование кристаллов CsI:Tl ионами IO_3^- позволяет получать прозрачные слитки с увеличенной концентрацией таллия (до $C_{\text{Tl}} \sim 0,9\%$), которые не имеет признаков концентрационного тушения фото- и радиолюминесценции. Показано, что распад твердого раствора в кристаллах CsI:Tl,IO₃ не наблюдается, по крайней мере, до $C_{\text{Tl}} \sim 0,5\%$, о чем свидетельствует высокий световой выход, хорошее энергетическое разрешение ($R = 6,3\%$) образцов с указанной концентрацией и больший коэффициент распределения таллия в кристаллах CsI:Tl,IO₃ ($k = 0,24$ против $k = 0,19$ в CsI:Tl). Предложено объяснение более высокой стабильности твердого раствора таллия в матрице CsI, которое основано на образовании комплексов $\text{Tl}^+ - \text{IO}_3^-$. Компенсация упругих напряжений противоположного знака вследствие взаимодействия катионов Tl^+ и анионов IO_3^- предотвращает образование очагов распада.