

МАТЕРІАЛИ ЕЛЕКТРОННОЇ ТЕХНІКИ

УДК 535.373

THERMALLY STIMULATED AND TUNNEL LUMINESCENCE IN Y₂O₃ AND Sc₂O₃ CERAMICS

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Thermally stimulated luminescence (TSL) and tunnel luminescence (TL) in Y₂O₃, Y₂O₃:Eu and Sc₂O₃ at X-ray irradiation was investigated. The Alentsev-Fock method for separate TSL curve into elementary components with additional experimental control by method of thermally clearing was used. Based on analysis of forms of elementary bands, which formed TSL curve, was made a conclusion that the recombination processes in TSL bands with maxima at 110, 122, 176, 185, 200 and 247 K in Y₂O₃ ceramics and at 121, 136, 200, 215 and 221 K in Sc₂O₃ ceramics was described of linear kinetics. The TSL in these ceramics connected with recombination processes in structural complex YO₆⁹⁻ (or ScO₆⁹⁻). In Y₂O₃:Eu ceramics TSL spectra consist in the main activators bands. Account that Eu³⁺ ions were seek to stability half-filled 4f-shell and effective trapped of electrons, proposed, that the TSL light was caused by recombination of holes with Eu²⁺-centers. The energy activation and frequency factors of local traps centers, which exhibited in TSL bands, were calculated with number methods. Recombination processes at vacated local traps centers in Y₂O₃ and Sc₂O₃ ceramics was caused diffusion-controlled tunnel recombination at the expense of thermally stimulated migration of holes, located on V_k-centre, was proposed. Exist of TL and his investigated in Y₂O₃ and Sc₂O₃ ceramics was carry out. The recombination processes were described models which observed recombination with complex interaction, based analysis of decay kinetics was established. The TL take placed in pairs with electron components in centers, which thermally vacated at 176 K in Y₂O₃ ceramics and 200 K in Sc₂O₃ ceramics. Holes components of tunnel pairs were caused with centers, which thermally vacated at 110 and 122 K in ceramics based on Y₂O₃ and 121 and 146 K in Sc₂O₃ ceramics.

Key words: thermally stimulated luminescence, tunnel luminescence, yttrium oxide, scandium oxide.

1. Introduction. Yttrium and scandium oxides due to chemical, thermal and radiation stability are perspective materials for registration of radiation [1 – 4]. The Y₂O₃ and Sc₂O₃

ceramic scintillators are applied for individual and technology dosimetry in the certain fields of experimental nuclear physics, because their scintillation efficiency is higher than in monocystals [1].

The investigation of trap centres in scintillator ceramics is vital, since these centres determine a lot of the material parameters, particularly constant of the de-excitation time. The spatial distribution of point defects in many cases is playing the important role in the spectral and kinetic characteristic of luminescence. The tunnel recombination of spatial separated charge carriers that produce tunnel pairs is frequently accompanied by tunnel luminescence (TL). The temperature independence of decay glow kinetic parameters and temporal shift of glow spectra in long-wave region is typical for TL and was revealed in Y_2O_3 crystals [5]. The studies of TL, give possibility to obtain the information in mechanisms of the electron-hole interaction and about the relative positioning of trap centres (TC) in the energy gap.

This paper is devoted to studies of TC of charge carriers and determination of their parameters by thermoactivation methods in Y_2O_3 , $Y_2O_3:Eu$ and Sc_2O_3 ceramics.

2. Experiment. The ceramic samples obtained by a baking of a pressed powder on the air at 1100 °C were studied. The initial raw material was Y_2O_3 of ItO-I marks, luminophor $Y_2O_3:Eu$ and Sc_2O_3 of E marks.

The samples were placed in a vacuum cryostat where the temperature was changed within 80–400 K range during the studies of the thermostimulated luminescence (TSL) and TL.

The irradiation was carried out by X-rays Cu K_{α} -lines (URS-55A, 40 kV, 10 mA). Quantity of X-rays dose was measured by IDMD-1 dosimeter. The exposition dose was changed within the limits of 0,15–2,35 C/kg. The intensity of TSL and TL were studied by FEU-51 photo multiplier and by B1–7 electrometric amplifier the signal of which was recorded by LKD4-003. The spectra of TSL were studied with UM-2 monochromator. The linearity of heating was provided by RIF-101 attemperator. The heating rate was $\beta=0,150\pm0,005$ K/sec, that provided sufficient resolution of the method for the studied samples with good relative aperture.

3. Results and discussion.

3.1 Local trapp centres in Y_2O_3 and Sc_2O_3 ceramics.

The curves of thermostimulated luminescences (TSL) of Y_2O_3 ceramics are presented on fig.1.

The three basic groups of peaks: 90–140, 150–220 and 230–280 K are revealed in the studies temperature range. The group of peaks in the interval 230–280 K range has much greater intensity for $Y_2O_3:Eu$ ceramics, than in Y_2O_3 ceramics. Fig.2. shows TSL curves for Sc_2O_3 ceramics.

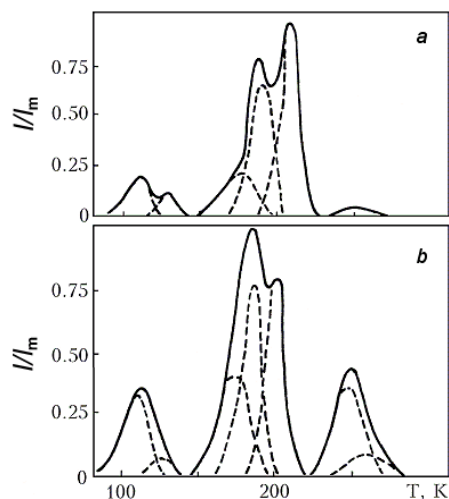


Fig.1. Curves of TSL in Y_2O_3 (a) and $Y_2O_3:Eu$ (b) ceramics at X-ray excitation ($D=1,5$ C/kg, $T=80$ K). Dashed lines show the elemental components.

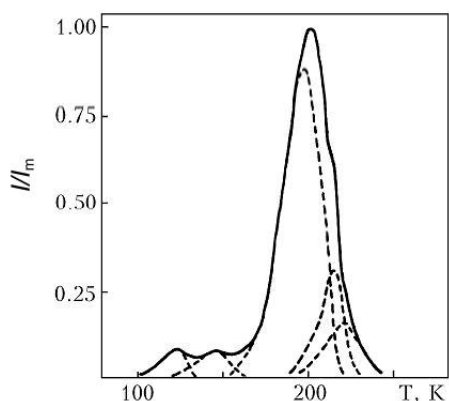


Fig.2. Curves of TSL in Sc_2O_3 ceramics under X-ray excitation ($D=1,5$ C/kg, $T=80$ K). Dashed lines show the elemental components.

The two first group of peaks are shown in the structure of a spectrum and they are characteristic also for Y_2O_3 , however with inappreciable high-temperature shift of curves and change of their relative intensity. The third, most high-temperature group of peaks, owing to small TSL intensity in Sc_2O_3 is not revealed by us.

The method of Alentsev-Fok [6] has been used for partitioning TSL curves on individual components. The method of thermal clearing has been used with the purpose of additional observation checkout of the lead partitioning. This known, that the linear kinetics of a luminescence the low-temperature part of TSL peak has greater slowness, than high-temperature one [7]. Consequently, it has been considered, that recombination processes which shown in oozed TSL peaks are described by the linear kinetics.

The dose dependences of intensity have been studied for all elemental peaks obtain as a result of TSL curves deconvolution. Fig.3. shows the obtained curves for of some TSL peaks in Y_2O_3 ceramics.

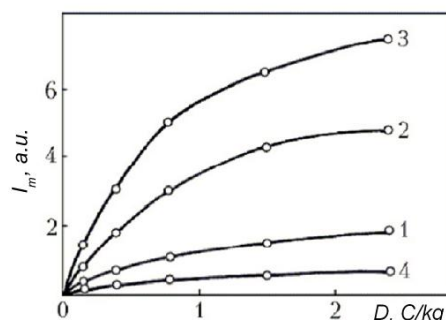


Fig.3. Dose dependences I_m for Y_2O_3 ceramics: $T_{max}=110$ (1), 185 (2), 200 (3) and 248K (4).

Typically the intensity of maxima of oozed TSL peaks in studied ceramics tends to saturation with increase of irradiation dose. Such increase of TSL peaks intensity, probably, is caused by increase of concentration of the free charge carriers trapped on existing in ceramics TC.

Besides, during the irradiation the generation of new defects which are TC or centres of recombination, is enough inappreciable or does not occur absolutely. In particular, considering, that spectra of X-ray luminescence of ceramics after the irradiation in the specified doses are not changed, it is possible to conclude, that during the irradiation the generation of new recombination centres does not occur.

The analysis of TSL spectra shows, that for studied TC in Y_2O_3 and Sc_2O_3 ceramics the recombination occurs in the range of 340–370 nanometers. Thus in TSL Y_2O_3 spectra the luminescence with a maximum at 368 nanometers and in TSL Sc_2O_3 ceramics spectra – with a maximum at 350 nanometers, is not observed, which is dominating one in spectra of the stationary luminescence. This confirms exciton nature of such luminescence. $Y_2O_3:Eu$ ceramics TSL spectra contain basically activation bands. Taking into account, that Eu^{3+} ions tend to have stable half-filled 4f-shell and effectively trap electrons [8], it is possible to consider, that TSL corresponds to the recombination of holes with the Eu^{2+} -centers. Thus, the Eu^{3+} -centers in the excited state are created, which relax with quantum emission selfactivation luminescence. Such situation enables to explain, that in $Y_2O_3:Eu$ ceramics the intensity of TSL peaks with maxima nearby 247 and 258 K is relatively higher, than in Y_2O_3 ceramics where the strong temperature luminescence quenching is observed at $T > 200$ K [9], and in $Y_2O_3:Eu$ the temperature quenching of activation luminescence is not observed [10].

The energy of thermal activation E_T is the one of the main TC parameters. We used different methods for E_T calculation in studied ceramics: method of superposition of the theoretical and observational TSL curves, offered by Antonov-Romanovskiy [11], a method which is based on changing of halfwidth of studied TSL peaks [12] and Garlik-Gibbson method [7], where lie depth is defined as $E_T = d(\ln I)/d(kT)^{-1}$.

The second important TC feature – the frequency factor p_0 , which characterizes frequency of the effective collisions, capable to void the localized charges. We defined p_0 for derived peaks as:

$$E_T = \left(\ln \frac{p_0 T_{\max}}{\beta} - \ln \ln \frac{p_0 T_{\max}}{\beta} \right) k T_{\max} ; \quad 1)$$

which obtained in [13] for linear kinetics, which as it was mentioned is typically also for Y_2O_3 and Sc_2O_3 ceramics.

The results of TSL curves processing by different methods are given in table 1 for Y_2O_3 ceramics and table 2 for Sc_2O_3 ceramics.

Table 1.

Energy of thermal activation and TC frequency factor in Y_2O_3 ceramics

T_{\max} , K	E_T , eV			p_0 , s ⁻¹
	Antonov-Romanovskyi method	Lushchik method	Garlik-Gibbson method	
110	0.15	0.15	0.16	$2.9 \cdot 10^4$
122	0.26	0.23	0.25	$2.6 \cdot 10^7$
176	0.34	0.39	0.31	$6.4 \cdot 10^5$
185	0.40	0.37	0.36	$5.2 \cdot 10^6$
200	0.47	0.49	0.47	$5.2 \cdot 10^8$
247*	0.54	0.53	0.49	$6.0 \cdot 10^6$
258*	0.56	0.48	$\begin{cases} 0.81 \\ 0.98 \end{cases}$	$8.3 \cdot 10^6$

*Calculation is lead for $Y_2O_3:Eu$ ceramics.

Table 2.

Energy of thermal activation and TC frequency factor in Sc_2O_3 ceramics

T_{\max} , K	E_T , eV			p_0 , c ⁻¹
	Antonov-Romanovskyi method	Lushchik method	Garlik-Gibbson method	
121	0.20	0.21	0.19	$2.3 \cdot 10^6$
136	0.25	0.25	0.27	$2.2 \cdot 10^7$
200	0.40	0.31	0.35	$8.1 \cdot 10^6$
215	0.51	0.47	0.45	$4.9 \cdot 10^8$
221	0.53	0.52	0.51	$6.0 \cdot 10^9$

Considering, that the method [11] have the certain error caused by theoretical simplifications, the error of the Garlik-Gibbson method is equal to 2-3 %, and the calculation error by Lushchik method is equal to 7–15 % [12], could be concluded, that E_T value, obtained by means of different methods for the majority of TSL peaks, practically coincide. It enables to approve, that studied TSL peaks for Y_2O_3 ceramics with maxima at 110, 122, 176, 185, 200 and 247 K and for Sc_2O_3 ceramics with maxima at 121, 136, 200, 215 and 221 K are caused by separate traps which energetically are not overlapped with other traps. From the given data it

could be seen that E_T value, obtained by Garlik-Gibbson method for TSL peak $Y_2O_3:Eu$ ceramics with a maximum at 258 K, considerably differs from obtained by other methods and has enough great value. The E_T value break up on two subgroups during processing of above specified peak which shows on nonelementary not only of recombination processes, but also of the centre. Such situation testifies, that the given peak is caused by existence quasimonotonous TC energy distribution.

Fig.4. shows the temperature dependences of E_T values for studies ceramics, obtained by Garlik-Gibbson method that owns the least error.

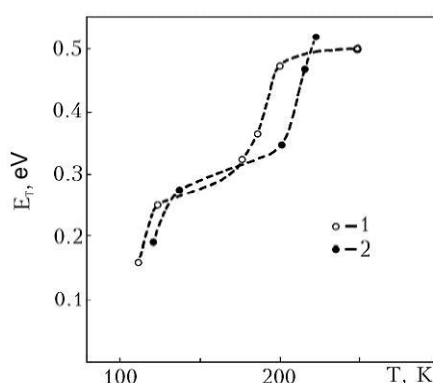


Fig.4. Temperature dependence of E_T value in Y_2O_3 (1) and Sc_2O_3 (2) ceramics defined by Garlik-Gibbson method.

3.2. Tunnel luminescence of Y_2O_3 and Sc_2O_3 ceramics .

The complex of studies of tunnel processes in viewed ceramics have been conducted for finding-out of nature TC of charge carriers in Y_2O_3 and Sc_2O_3 ceramics.

It is known, that dependence of TL intensity on time of signal attenuation (time after the termination of excitation) $I(t)$ is well featured by the Becquerel law [14]:

$$I(t) = \frac{I(0)}{(1+bt)^\alpha}; \quad (2)$$

where b and α – parameters of approximation.

Fig.5 shows in double logarithmic coordinates the kinetics of signal attenuation of TL intensity for Sc_2O_3 ceramics at different doses of an irradiation. Similar dependences are obtained and for Y_2O_3 and $Y_2O_3:Eu$ ceramics.

The kinetics of TL signal attenuation contains the information on a spatial distribution of tunneling centres [14, 15]. The two theoretical models are used for the description of a real kinetics: model of the complex recombination interaction and model which considers a recombination in the isolated genetic pairs.

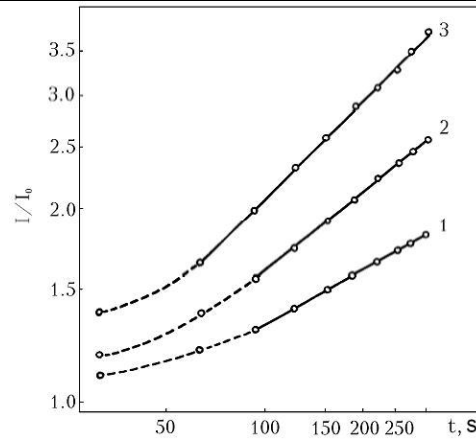


Fig.5. Kinetics of signal attenuation TL in ceramics Sc_2O_3 in double logarithmic coordinates at temperature of X-ray irradiation 150 K and recordings 80 K. Dose of an irradiation 2,30 (1), 1,55 (2) и 0,77 C/kg (3).

Considering literature data [16], TL processes in the studied ceramics can be featured, applying model of the complex recombination interaction, as TL intensity is not proportional to tunnel pairs concentration and slope angle of dependence $\ln I$ from $\ln t$ (parameter α) depends on a dose of an irradiation (Table 3). It specifies the redistribution of the tunnel pairs components behind distances in a direction of chaotic distribution.

The value of parameter a in the Becquerel law for the description of TL kinetics in Y_2O_3 , $\text{Y}_2\text{O}_3:\text{Eu}$ and Sc_2O_3 ceramics at different doses of an irradiation.

Table 3

Dose of irradiation, C/kg	Parameter α		
	Sc_2O_3	Y_2O_3	$\text{Y}_2\text{O}_3:\text{Eu}$
0.77	0.528	0.372	0.715
1.55	0.399	0.347	0.657
2.30	0.272	0.274	0.593

The obtained experimental results enable to evaluate the initial distribution function of pairs $n(r, 0)$ [14] behind distances between their components. For this purpose light emission summ $S_a(t)$ are measured at intervals of time Δt_a , which follow one another. The interval of time Δt_a begins at t/a and finished at at . Here a - the any positive number, $a > 1$. At processing of the experimental values quantity do not change at transition between time intervals.

Accepting, that at centres of radiation all electronic transitions are the emissive, we have:

$$S_a(t) = \int_0^{\infty} [n(r, t/a) - n(r, at)] dr \quad (3)$$

Considering, that

$$n(r, t) = n(r, 0) \exp[-W_0 t \exp(r/r_0)] \quad (4)$$

where r – distance between tunneling centres, W_0 and r_0 – constants, and accepting, that near $r(t)$ – the distance between elements of pairs, at which at present moment the rate of tunnel pair decay is maximal, the function $n(r, 0)$ slowly depends on r , we obtain:

$$S_a(t) \approx n(r, 0) 2 \ln a \quad (5)$$

The light emission summ $S_a(t)$ we measured in time intervals from 10 up to 300 sec, and parameter a took equal 1.5. The results of calculations show, that dependences $S_a(t)$, and, hence, and $n(r, 0)$ for Y_2O_3 , $Y_2O_3:Eu$ and Sc_2O_3 ceramics are similar (see fig.6).

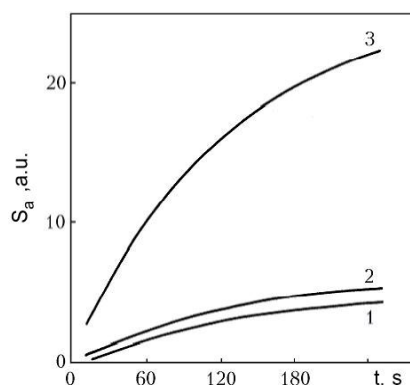


Fig.6. Dependence partial light emission summ S_a from time in Y_2O_3 (1), Sc_2O_3 (2) и $Y_2O_3:Eu$ (3) ceramics. Dose of irradiation 0,77 C/kg.

It shows that in these ceramics the mechanisms of TL are identical. Besides $S_a(t)$ dependence shows, that the majority of radiative defects are remote on distances which exceed radius of effective tunnel transitions with time constant of 10–100 sec. The distances between defects decrease in $Y_2O_3:Eu$ ceramics. Besides, the structural defects created by the activator, in the mechanism of diminishing of distances between radiative defects in the pair the trapping of electronic component of the pair by Eu^{3+} ions gives the contribution.

For definition of the nature of the centres between which the tunnel recombination occurs – a composition of tunnel pairs – we take into account that TL intensity tends to zero if the sample is excited at temperatures which exceed temperature of thermal destruction of one of the components of the studied pair. With this purpose we measured dependence of luminescent emission on temperature of excitation, i.e., efficiency of light emission summ accumulation at different temperatures.

The ceramics, preliminary heated up to 400 K, excited at constant dose at different temperatures, then cooled down to 80 K and the kinetics of their luminescence was measured. The intensity of luminescence at the certain moment of time after the excitation termination was accepted as a measure standard of the relative concentration of the tunnel pairs created at the given temperature of excitation.

The light emission summ which is accumulated in the studies ceramics, measured at rise of temperature of excitation has two sites of abrupt wane which correlate with peaks of TSL curve (Fig.7). The obtained results enable to draw a conclusion, that TC which correspond to TSL peak with a maximum at 176 K in Y_2O_3 ceramics and at 200 K in Sc_2O_3 ceramics are trap centres of one sign and make one of a builder of tunnel pair. More the low-temperature TSL peaks with maxima at 110 and 122 K in Y_2O_3 ceramics and 121 and 146 K in Sc_2O_3 ceramics

are caused by TC of the carriers of the opposite sign and are correspondingly the second components of tunnel pairs.

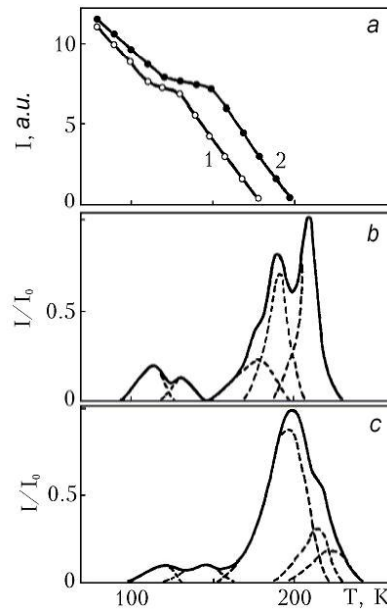


Fig.7. Efficiency of light emission summ attenuation depending on temperature of excitation (a) in Y_2O_3 ceramics (1) and Sc_2O_3 ceramics (2); TSL curves of Y_2O_3 ceramics (b) and Sc_2O_3 (c) ($D=1,5$ C/kg, $T=80$ K). Dashed lines show the elemental compositions

4. Conclusions. The significant diversion of $E_T(T)$ dependence from linearity testifies to presence of the remote tunnel mechanism of a luminescence (Fig.4). Thus, by analogy to related system Al_2O_3 in which has been proved the thermostimulated migration of hole which is delocalized from V_k -centres [17], it is possible to consider, that also in Y_2O_3 and Sc_2O_3 the diffuse-controllable tunnel recombination caused by thermostimulated migration of holes, delocalized from V_k -centres is observed. Diffuse character of migration of electronic excitation with possible intermediate localization proves to be true also by small values of the frequency factor p_0 of the relaxation processes. Apparently from Table 1 and Table 2, in Y_2O_3 and Sc_2O_3 ceramics the frequency factor of relaxation process on some orders is lower than the frequency factor of trap centres ($\sim 10^{12} \text{ c}^{-1}$).

For definition of a sign of charge carriers which trapped on corresponding TC, we used except for the our results, the results of EPR and TSL studies of spectrums yttrium oxide crystals in operations [18, 19]. Our analysis allows to identify TSL peak with a maximum at 176 K in Y_2O_3 as a luminescence of electronic centre. The energy of thermal activation of this centre, defined both for ceramics, and for Y_2O_3 crystals [18], is equal to 0.31 eV. The study of the recharge of this F-center in operation [20] confirms conclusion, that it is electronic centre. The second components of the tunnel pairs which are genetically related with TC which are revealed for ceramics on the basis of Y_2O_3 in TSL peaks at 110 K and 122 K and Sc_2O_3 at 121 and 146 K and are TC of the charge carriers of an opposite sign, make the hole components of tunnel pairs.

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ТЕРМОСТИМУЛЬОВАНА І ТУНЕЛЬНА ЛЮМІНЕСЦЕНЦІЯ В КЕРАМІКАХ Y_2O_3 І Sc_2O_3

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Досліджено термостимульовану люмінесценцію (ТСЛ) і тунельну люмінесценцію (ТЛ) в кераміках Y_2O_3 , $Y_2O_3:Eu$ і Sc_2O_3 при рентгенівському опроміненні. Для поділу кривих ТСЛ на елементарні складові використано метод Аленцева-Фока з додатковою експериментальною перевіркою методом термічної очистки. На підставі аналізу форми елементарних смуг, які формують криві ТСЛ, зроблено висновок, що рекомбінаційні процеси у смугах ТСЛ з максимумами при 110, 122, 176, 185, 200 і 247 К в кераміках Y_2O_3 і при 121, 136, 200, 215 і 221 К в кераміках Sc_2O_3 описуються лінійною кінетикою. Показано, що ТСЛ в цих кераміках пов'язана з рекомбінаційними процесами у структурних комплексах YO_6^{9-} (або ScO_6^{9-}). Встановлено, що в кераміках $Y_2O_3:Eu$ спектри ТСЛ складаються в основному з активаторних смуг. Різними способами визначено енергії активації і частотні фактори локальних центрів захоплення, які проявляються в кривих ТСЛ. Встановлено, що рекомбінаційні процеси при звільненні центрів захоплення зумовлені дифузною-контрольованою тунельною рекомбінацією, найбільш ймовірно, за рахунок термостимульованої міграції дірок, звільнених з V_K -центрів. Показано наявність і досліджено ТЛ в кераміках Y_2O_3 і Sc_2O_3 . На основі аналізу кінетики загасання ТЛ встановлено, що рекомбінаційні процеси описуються моделлю зі складною взаємодією. ТЛ здійснюється в парах, електронною компонентою яких є центри, які термічно звільняються при 176 К в кераміках Y_2O_3 і 200 К в кераміках Sc_2O_3 . Діркові компоненти тунельних пар пов'язані з центрами, які термічно звільняються при 110 і 122 К в кераміках на основі Y_2O_3 та 121 і 146 К в кераміках Sc_2O_3 .

Ключові слова: термостимульована люмінесценція, тунельна люмінесценція, оксид ітрію, оксид скандію.

ТЕРМОСТИМУЛИРОВАННАЯ И ТУННЕЛЬНАЯ ЛЮМИНЕСЦЕНЦИЯ В КЕРАМИКАХ Y_2O_3 И Sc_2O_3

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Исследована термостимулированная люминесценция (ТСЛ) и туннельная люминесценция (ТЛ) в керамиках Y_2O_3 , $Y_2O_3:Eu$ и Sc_2O_3 при рентгеновском облучении. Для разделения кривых ТСЛ на элементарные компоненты использован метод Аленцева-Фока с дополнительной экспериментальной проверкой методом термической очистки. На основании анализа формы элементарных полос, формирующих кривые ТСЛ, сделан вывод, что рекомбинационных процессов в полосах ТСЛ с максимумами при 110, 122, 176, 185, 200 и 247 К в керамиках Y_2O_3 и при 121, 136, 200, 215 и 221 К в керамиках Sc_2O_3 описываются линейной кинетикой. Показано, что ТСЛ в этих керамиках связана с рекомбинационными процессами в структурных комплексах YO_6^{9-} (или ScO_6^{9-}). Установлено, что в керамиках $Y_2O_3:Eu$ спектры ТСЛ состоят в основном из активаторных полос. Несколькими способами определены энергии активации и частотные факторы локальных центров захвата, которые проявляются в кривых ТСЛ. Установлено, что рекомбинационные процессы при освобождении центров захвата обусловлены диффузно-контролируемой туннельной рекомбинацией, наиболее вероятно, за счет термостимулированной миграции дырок, освобожденных с V_k -центров. Показано наличие и исследовано ТЛ в керамиках Y_2O_3 и Sc_2O_3 . На основе анализа кинетики затухания ТЛ установлено, что рекомбинационные процессы описываются моделью со сложным взаимодействием. ТЛ осуществляется в парах, электронной компонентой которых являются центры, которые термически освобождаются при 176 К в керамиках Y_2O_3 и 200 К в керамиках Sc_2O_3 . Дырочные компоненты туннельных пар связаны с центрами, которые термически освобождаются при 110 и 122 К в керамиках на основе Y_2O_3 и 121 и 146 К в керамики Sc_2O_3 .

Ключевые слова: термостимулированная люминесценция, туннельная люминесценция, окись иттрия, окись скандия.

Стаття надійшла до редколегії 26.05.2011
Прийнята до друку 21.06.2011