Kinetic Features of Thermoluminescence in Oxygen – Deficient Crystals of Aluminum Oxide[1]

Orozbek uulu Askar, I.A. Weinstein, A.I. Slesarev, V.S. Kortov

Ural State Technical University – UPI, Ekaterinburg, Russian Federation
E- mail: askar@dpt.ustu.ru

Abstract – The study deals with the effect of the irradiation dose on thermoluminescence (TL) of F^+– centers (3.8 eV) at temperatures of 300 – 900 K in anion – defective crystals of aluminum oxide. An intense TL peak at 835 K was detected at high doses. It was found that the intensity of the TL dosimetric peak (3.8 eV) at 450 K was largely affected by trapping centers of impurity chromium ions and deep traps at 730 K and 835 K.

1. Introduction

Dosimetric and kinetic properties of ionizing radiation TL detectors type TLD – 500 based on anion – defective single crystals of aluminum oxide are being studied intensively because the TL peak at 450 K is used as the source of information during dosimetric measurements. Therefore, it is necessary to ascertain the origin of active centers and determine the mechanism of TL in this peak.

It is known that the dosimetric peak is complex and luminescence spectra include bands corresponding to F– and F^+–centers (oxygen vacancies with two and one trapped electrons respectively) [1]. There is evidence that aluminum oxide crystals contain, in addition to active centers produced by intrinsic defects, impurity – induced centers, which can considerably influence the spectrum composition and the intensity of the dosimetric peak by competing in trapping of charge carriers [2].

The dosimetric TL peak dominates at low doses, while the luminescence of impurity centers and deep traps can hardly be measured. One may think that at high irradiation doses, when the dosimetric peak is saturated, charge carriers will be captured on impurity and deep traps. Then it will be possible to measure the corresponding TL peaks and watch their variation dynamics with growing radiation dose.

The objective of this study was to analyze how kinetic parameters of the TL peaks, which are due to the luminescence of F^+ – centers, change at temperatures of 300 – 900 K depending on the irradiation dose of anion – defective crystals of aluminum oxide.

2. Samples and Experimental Technique

The subjects of study were anion – defective single crystals of aluminum oxide in the form of pellets 5 mm in diameter and 1 mm thick. They were grown by the method of oriented crystallization in strongly reducing conditions [3]. The samples were irradiated from a β – source based on the $^{90}Sr/^{90}Y$ isotope. The radiation dose rate at the location of the samples was 0.032 Gy per minute.

TL curves were recorded using a MUM grating monochromator and a FEU–39A photomultiplier. The monochromator separated the luminescence band of F^+ – centers at 3.8 eV. For depletion of dosimetric and deep traps, the samples were annealed up to 920 K, irradiated from the β – source, and heated linearly up to 900 K at a rate of 2 K/s. The β – radiation dose was 0.064 Gy to 76.8 Gy.

3. Results and Discussion

Figure 1,*a* presents TL curves for the crystals under study exposed to different irradiation doses. As the dose increases up to 2 Gy, the intensity of the dosimetric peak at 450 K is enhanced and the TL maximum (T_m) shifts to the region of low temperatures. Note that at the dose of about 4 Gy the dosimetric peak is saturated and a weak luminescence at 570 K, which is due to impurity chromium ions, appears [4]. It is known that the capture of charge carriers on traps is followed by the change of the valence of Cr^{3+} ions: Cr^{2+} and Cr^{4+} centers are formed when electrons and holes are trapped respectively. As the dose grows further, the TL intensity of the dosimetric peak begins decreasing, but T_m changes insignificantly and the luminescence of Cr^{3+} centers is enhanced simultaneously. At the dose of 30.7 Gy new TL peaks appear at about 390 K and 835 K. The peak at 390 K is probably due to the luminescence of titanium ions [2, 4]. Two states of titanium Ti^{3+} and Ti^{4+} are possible in the aluminum oxide lattice. Triply charged ions produce additional trapping levels in the forbidden band of aluminum oxide and serve as efficient centers for recombination of secondary electron – hole pairs [5]. Oppositely, Ti^{4+} ions are electron trapping centers and, therefore, facilitate the accumulation of holes on chromium traps. At still higher doses, the TL maximums at 390, 450 and 570 K are saturated, but the intensity of the TL peak at 835 K, which is due to deep trapping centers, is considerably enhanced.

The saturation of the intensity of the dosimetric peak (450 K) at the dose of about 4 Gy and the decrease of the peak with growing dose can be explained by the capture of charge carriers on competing traps produced by Cr^{3+} impurities. This supposition is con-

firmed by the increase in the intensity of the TL peak at 570 K. As soon as the peaks at 390, 450 and 570 K are saturated, the deep trap enters into the process of competitive trapping and, hence, its intensity is enhanced.
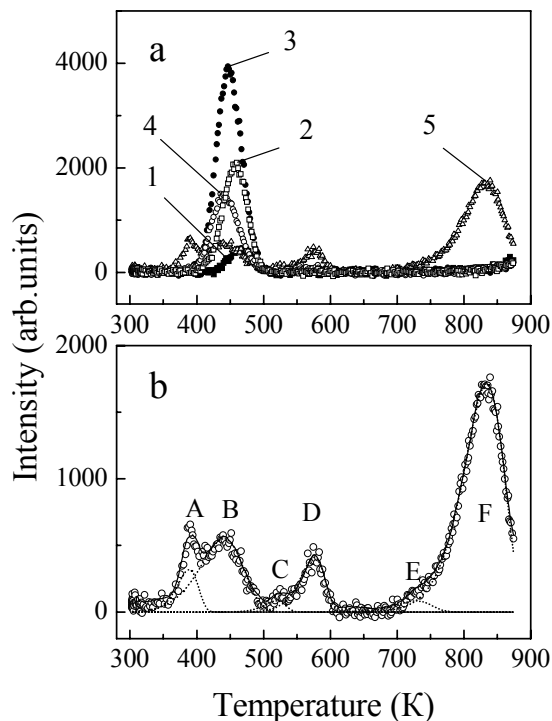

Fig. 1. (*a*) Thermoluminescence curves for $\alpha - Al_2O_3$ single crystals at temperatures of $300 - 900$ K depending on the dose: $1 - 0.06$ Gy, $2 - 0.5$ Gy, $3 - 1.9$ Gy, $4 - 4.8$ Gy, $5 - 76.8$ Gy. (*b*) Example of the resolution of TL peaks and estimates of the kinetic parameters at a dose of 76.8 Gy

The kinetic parameters of complex peaks were evaluated from the general formal kinetics equation for the thermally stimulated luminescence [6]:

$$I(T) = n_0 S \exp\left(-\frac{E}{kT}\right) \cdot \left[1 + \left((b-1)\frac{S}{\beta}\right)\int_{T_0}^{T}\exp\left(-\frac{E}{kT}\right)dT\right]^{-\frac{b}{b-1}} \quad (1)$$

where n_0 is the initial concentration of charge carriers captured on traps, S is the preexponential factor, E is the activation energy, k is the Boltzmann constant, b is the kinetics order, β is the heating rate, and T_0 is the initial temperature.

The calculation procedure is described in detail elsewhere [7]. The resolution of the TL peaks at a dose of 76.8 Gy is shown in Fig. 1,*b*. Six TL peaks are formed at this dose: A, B, C, D, E and F at 390, 440, 520, 570, 730 and 835 K respectively. The dominant peak is the peak at 835 K. The kinetic parameters of these curves over the interval of test doses were evaluated from the equation (1). Table 1 gives calculated

values of T_m, b, E and S for all the TL peaks. It is seen from this table that the kinetics order of the peaks A and B changes with growing dose, while the peaks C, D, E and F are described in terms of molecular kinetics.

Table 1. Kinetic parameters of TL peaks at different irradiation doses (3.8 eV)

Peak	Dose (Gy)	T_m, (K)	b	E, eV	S, (s^{-1})
A	30.7 – 76.8	390 – 392	1.58 – 1.00	1.14 – 0.81	$9.46 \cdot 10^{13}$ – $3.70 \cdot 10^{9}$
B	0.06 – 1.9	467 – 447	1.19 – 2.00	1.32 – 1.30	$2.65 \cdot 10^{13}$ – $2.08 \cdot 10^{14}$
	3.8 – 76.8	442 – 440	2.00 – 1.00	1.28 – 0.49	$1.22 \cdot 10^{13}$ – $2.33 \cdot 10^{4}$
C	76.8	520	1.00	1.5	$4.48 \cdot 10^{13}$
D	4.8 – 76.8	571 – 586	1.00	1.30 – 1.79	$9.97 \cdot 10^{9}$ – $5.31 \cdot 10^{14}$
E	76.8	728	1.00	1,70	$4.38 \cdot 10^{10}$
F	30.7 – 76.8	840 – 835	1.00	1.80 – 2.08	$3.25 \cdot 10^{9}$ – $2.27 \cdot 10^{11}$

Figure 2 presents dose dependences of the kinetic parameters of the main peak (450 K). It was noted in the foregoing that the dosimetric peak is saturated and the TL yield decreases at a dose of 4 Gy.
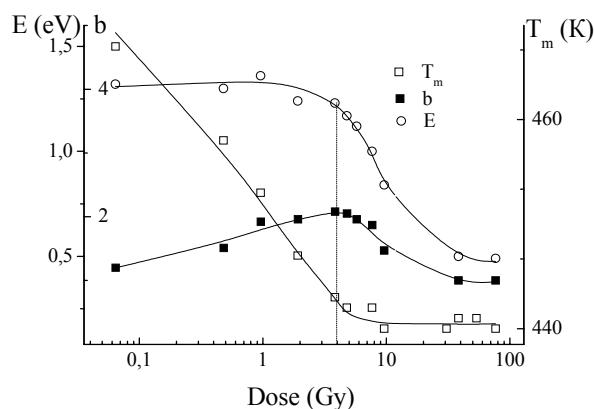

Fig. 2. Dose dependences of the kinetic parameters of the dosimetric peak (the luminescence band at 3.8 eV)

The activation energy of the TL of the main peak decreases from 1.28 eV to 0.5 eV and T_m practically stops changing at the same dose. At doses lower than 4 Gy the T_m value decreases and, correspondingly, the kinetics order changes from 1 to 2, while E remains practically unchanged. Thus, the dose of 4 Gy is critical for the crystals at hand: the kinetics is considerably altered when this dose is reached and increases further. It may be conjectured that this situation is due to the cessation of the interactive interaction between dosimetric and deep traps.

Figure 3 presents dependences characterizing the linear relationship $lg(S)=lg(S_0)+BE$, which is called the compensation effect [8]. This phenomenon was observed earlier during temperature quenching of the pho-

toluminescence of F – centers in aluminum oxide [9]. The parameter S_0 characterizes the ratio of the configuration and oscillation contributions to the total change of the entropy of samples [10]. The coefficient B is the isokinetic temperature $T_i = (ln10 \cdot kB)^{-1}$. It is assumed that processes progress at a constant rate at this temperature.
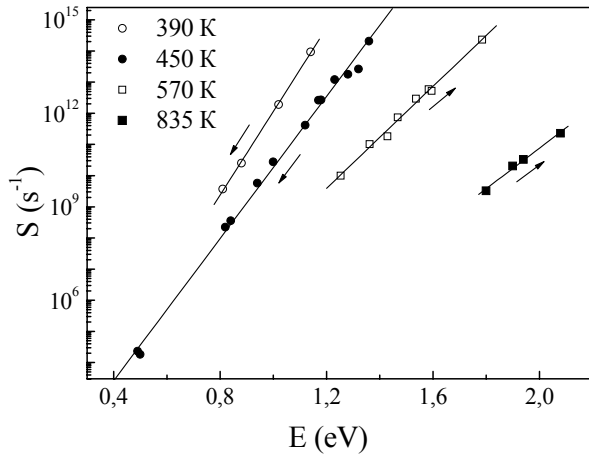

Fig. 3. The relationship between the kinetic parameters of the peaks in aluminum oxide crystals at different doses (the arrows show the direction of the dose variation)

It is seen from this figure that the kinetic parameters of the TL peaks under study are connected by some relationship. As the dose increases, the activation energy and the frequency factor of the TL peaks at 390 K and 450 K decrease linearly. At the same time, the corresponding parameters of the peaks at 570 K and 835 K grow. Calculated values of B, S_0 and T_i are given in Table 2. The isokinetic temperature of the dosimetric peak is 442 K, which corresponds to the temperature position of the peak at its saturation.

Table 2. Values of B, S_0 and T_i as calculated from the data of Fig. 3

T_m (K)	390	450	570	835
B	13.4	11.4	8.2	6.5
S_0	0.05	0.071	0.6	0.0074
T_i (K)	376	442	615	775

4. Conclusion

The analysis of the effect of the irradiation dose on the TL and the kinetic properties of the luminescence band of F^+ – centers (3.8 eV) at temperatures of 300 to 900 K suggests that the mechanism, which determines decrease of the luminescence of F^+ – centers (3.8 eV) in anion – defective crystals of aluminum oxide with increasing irradiation dose, is the competitive capture of charge carriers on impurity centers and deep traps. In this case, the chromium trap at 570 K plays a considerable role. One may think that if the purity of the crystals is improved and their growth conditions are optimized so as to decrease the concentration of impurity and deep traps, the dose characteristic of the main TL peak at 450 K will extend its linear behavior to the region of higher doses. As a result, TL detectors based on anion – defective crystals of aluminum oxide will find more applications.

References

[1] V.S. Kortov, I.I. Milman, Izv. VUZov Fizika. **39**, No. 11. 145 – 161 (1996).

[2] T.S. Bessonova, M.P. Stanislavsky, V.Ya. Khaimov – Malkov, Problems of Nuclear Science and Engineering. Series: Radiation Damage Physics and Radiative Study of Materials. Vol. 1(1). Kharkov: FTI AN USSR, 74 – 80 (1974).

[3] M. Akselrod, V. Kortov, D. Kravetsky, V. Gotlib, Rad. Prot. Dosimetry. **32**, No. 32. 15 (1990).

[4] T.S. Bessonova, M.P. Stanislavsky, V.Ya. Khaimov – Malkov, ZhPS. **30**, No. 5. 829 – 835 (1979).

[5] T.S. Bessonova, M.P. Stanislavsky, V.Ya. Khaimov – Malkov, Opt. i Spektr. **41**, No. 1. 152 – 154 (1976).

[6] R. Chen, S.W.S. McKeever, *Theory of Thermoluminescence and Related Phenomena*, Singapore, World Scientific, (1997) pp.29–35.

[7] A.I. Slesarev, V.S. Kortov, *in Proc. 17th Int. Conf. on Nondestructive Testing and diagnostics*, 2005, p.30.

[8] V.P. Kazakov, UFZh. **15**, No. 10. 1730 – 1731 (1965).

[9] I.A. Weinstein, A.S. Vokhmintsev, V.S. Kortov, Tech. Phys. Lett. **32**, No. 1, 58 – 61 (2006).

[10] V.I. Krysov, FMM. **93**, No. 2. 70 – 74 (2002).