Interaction of Traps in Anion-Defective Aluminum Oxide[1]

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Abstract – It was experimentally found that the intensity of the TL peak at 350 K could change considerably in crystals of anion-defective aluminum oxide having the same dosimetric sensitivity. Dose dependences of the TL intensity in the peaks at 450 and 350 K were analyzed. Experimental evidence was adduced to the presence of the competitive interaction between traps of the dosimetric peak and shallower trapping centers responsible for the TL peak at 350 K.

1. Introduction

Crystals of anion-defective aluminum oxide, which are grown in reducing conditions, are highly sensitive to radiation. Therefore, this material can be used as the thermoluminescence dosimeter of both UV light and ionizing radiation [1]. Thermoluminescence of anion-defective α-Al_2O_3 is related to intrinsic defects and impurities, which are formed under certain conditions, including those of the crystal growth. The thermoluminescence curve of anion-defective α-Al_2O_3, which is exposed to irradiation doses of up to 1 Gy at room temperature, contains two thermoluminescence (TL) peaks at 450 K and 350 K [2]. The TL maximum at 450 K is used for dosimetric measurements. The peak at 350 K arises from shallow trapping levels. The interest shown to shallow traps is explained by the fact that they can compete with dosimetric centers and distort dosimetric information.

The present study deals with processes underlying the interaction of shallow and dosimetric traps.

2. Samples and Experimental Technique

The subjects of study were samples of nominally pure anion-defective single crystals of α-Al_2O_3, which were grown in reducing conditions by the Stepanov method. The samples were 5 mm in diameter and 1 mm thick. A β-source based on the $^{90}Sr/^{90}Y$ isotope was used for thermoluminescence excitation. The radiation dose rate at the location of the samples was 0.032 Gy per minute. Because the intensity of the TL peak at 350 K decreased immediately after irradiation, the samples were held in darkness for 20 seconds before each TL measurement so as to obtain reproducible result. The TL was measured by the standard technique in the luminescence band of F-centers (420 nm) at the heating rate of 5 K/s. A light filter was used for separation of the luminescence band of F-centers. The samples were exposed to X-rays at 80 K from a URS-55 A apparatus (I = 15 mA, U = 40 kV, Co anode). A FEU-106 photomultiplier served as the radiation detector.

3. Results and Discussion

Figure 1 presents TL curves of anion-defective aluminum oxide exposed to X-rays at 80 K. According to the literature data, the weak TL peak at 220 K is due to the release of holes from $\{Mg\}^0$-centers (which represent the isomorphous replacement of Al^{3+} by Mg^{2+} ions), while the peak at 260 K results from the decay of an electron trapping center [3–4]. The origin of the trapping center responsible for the TL peak at 310–320 K has not been identified reliably. Some researchers hold to the opinion that it is related to both hole and electron traps. It is suggested with respect to the origin of the TL of the dosimetric peak (450 K) that this peak is due to a complex defect. This defect consists of closely spaced vacancies in different charge states and Cr^+ ions, which act simultaneously as a charge-carrier trap and a luminescence center [5].

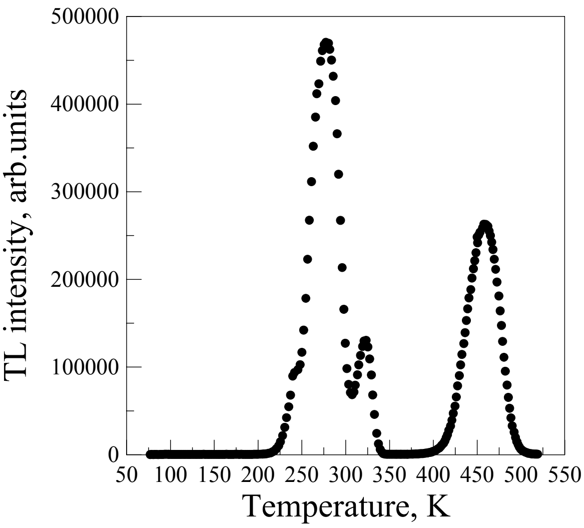

Fig. 1. Thermoluminescence curve of anion-defective aluminum oxide exposed to X-rays at 80 K

During excitation by the test dose (0.032 Gy) of β-radiation at room temperature, the TL maximum, which is observed at 320 K under low-temperature irradiation, shows up near 350 K. The study demonstrated that samples, which had a nearly equal light sum of the dosimetric peak, could be characterized by considerably different (more than one order of magni-

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tude) intensities of the TL at 350 K. This discrepancy was explained by different concentrations of shallow traps. However, the temperature position and the shape of the TL peak at 350 K remained unchanged in different samples. This observation pointed to identity of the TL kinetic parameters in different samples [2].

The kinetic parameters of the TL peak at 350 K were analyzed using different techniques. Values of the activation energy E, the frequency factor S and the kinetics order b, which were calculated from TL measurements performed by different methods, had good convergence. It was assumed therefore that the TL peak at 350 K was due to a monoenergy trap and the TL kinetics was of the first order [2].

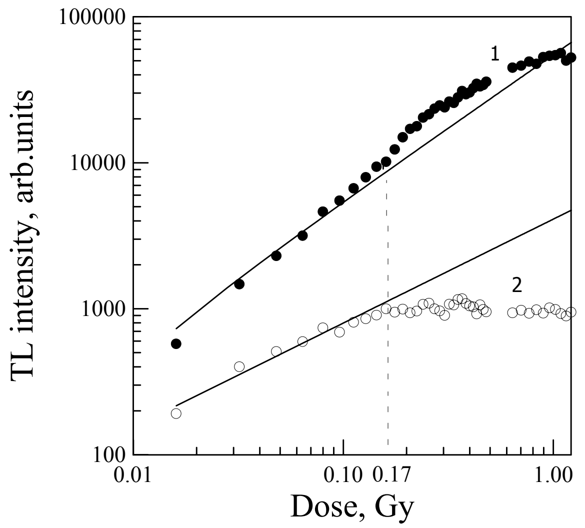

Fig. 2. Fragment of the dose dependence of the TL yield in the peaks at 450 K (1) and 350 K (2)

The analysis of dose dependences demonstrated that the increase in the irradiation dose was followed by the growth of the TL intensity in both the dosimetric maximum and the peak related to shallow traps. The temperature position and the shape of the peak at 350 K remained almost unchanged whatever the irradiation dose. This fact provided one more proof of the first-order kinetics of processes responsible for the said peak. Figure 2 presents the dose dependence of the TL intensity in the peaks at 450 K and 350 K. It is seen that the amplitude of the peak at 350 K was saturated at the dose of about 0.17 Gy, which corresponded to the beginning of the superlinear growth of the dosimetric maximum. It may be assumed that one of reasons for this correspondence was the interaction of shallow and dosimetric traps. To get a deeper insight into processes of the interaction between shallow and dosimetric traps, we analyzed the dependence of the intensity of the TL peak at 350 K on the occupancy of dosimetric traps. For this purpose, the sample was exposed to the test dose and then the TL of shallow traps was measured. The irradiation dose of the sample increased in steps of 0.032 Gy, leading to smooth filling of dosimetric traps. After each irradiation step, the sample was annealed to 373 K so as to deplete shallow traps, while dosimetric traps remained occupied. Then the sample was exposed repeatedly to the test dose and was heated to 373 K. The intensity of the peak at 350 K was measured. By this algorithm, it was possible to measure the intensity of the TL at 350 K after the exposure to the same (test) dose, while the occupancy of dosimetric traps was different. The occupancy was assumed to be proportional to the dose in the linear section of the dose characteristic, which corresponded to the TL peak at 450 K (Fig. 3).

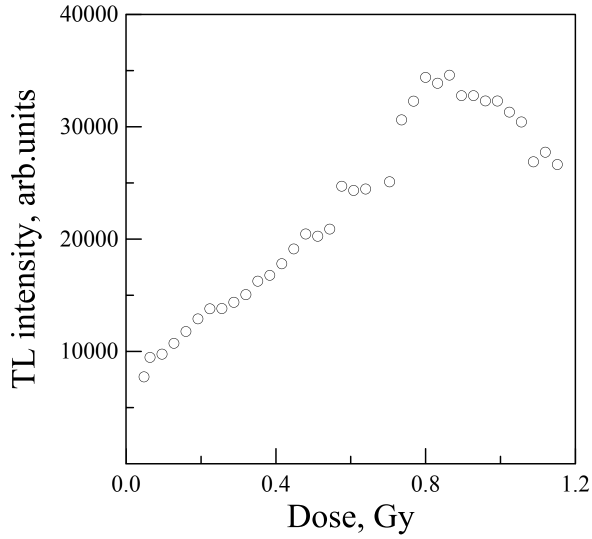

Fig. 3. Dependence of the TL intensity in the peak at 350 K on the dose that causes filling of dosimetric traps

It was found that filling of dosimetric traps was followed by the increase in the intensity of the TL in the peak at 350 K under exposure to the test dose, i.e. the radiation sensitivity improved over the temperature interval of the peak studied. This result is a direct experimental proof of the competitive interaction between shallow traps and trapping centers responsible for the dosimetric peak. One may think that the growth of the radiation sensitivity in the peak at 350 K was due to weakening of the competition of dosimetric traps as they were being filled. The higher was the occupancy of dosimetric traps, the larger was the number of free carriers, which were formed under irradiation at room temperature, that were captured on shallow traps and participated in recombination processes accompanying the TL at 350 K. It should be noted that the radiation sensitivity of the TL peak at 350 K no longer improved at the dose of about 0.8 Gy, which corresponded to the saturation of the dose characteristic of the TL maximum at 450 K (see Fig. 3). The saturation of the dosimetric peak amplitude at large irradiation doses was due to the high occupancy of traps responsible for this peak and the progressing processes of concentration quenching of the

TL. It is not improbable that these processes influenced the probability of recombinations involving shallow traps. As a result, the radiation sensitivity in the TL peak at 350 K was impaired at doses corresponding to the dosimetric peak saturation.

To obtain more evidence to the interaction of shallow and dosimetric traps, we analyzed the dependence of the TL intensity in the peak at 350 K as dosimetric traps were depleted. For this purpose, the sample was exposed to the test dose at room temperature and the TL with the peak at 350 K was measured upon subsequent heating. The sample was heated to 420 K, which was the start temperature of the dosimetric maximum, and then the heating temperature increased in 10 K steps for smooth depletion of dosimetric traps. The experimental results shown in Fig. 4 demonstrate that the depletion of dosimetric traps was followed by the drop of the TL intensity in the peak, which was due to shallow trapping levels. This situation took place because unoccupied dosimetric traps went into vigorous competition in charge-carrier trapping processes. Thus, the interactive relation between shallow and dosimetric traps was observed both upon filling and depletion of trapping centers responsible for the dosimetric peak.

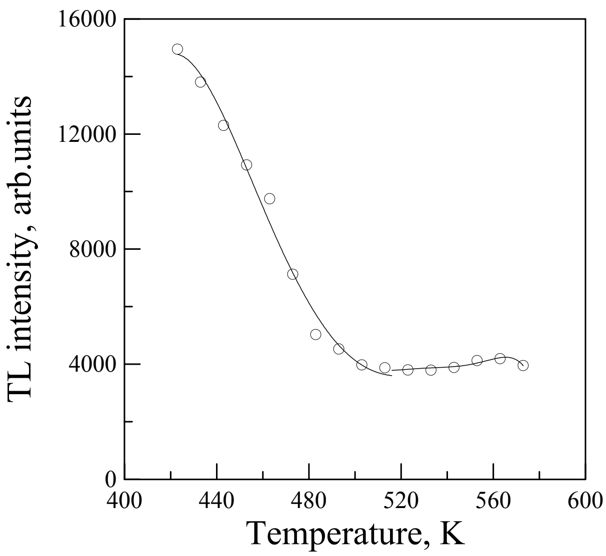

Fig. 4. Dependence of the TL intensity in the peak at 350 K on the depletion temperature of dosimetric traps

It should be noted also that the TL intensities in the peak at hand, which were measured before filling of dosimetric traps and after their depletion, were considerably different. Moreover, the TL sensitivity of the peak at 350 K decreased with respect to the initial sensitivity after depletion of dosimetric traps. It may be thought that the observed effects were due to the competitive action of deeper traps (730 K and 880 K) in the charge-carrier trapping.

It was found also that the TL intensity in the peak at 350 K, which was due to shallow traps, correlated with the FWHM of the dosimetric maximum at 450 K. The obtained results are shown in Fig. 5. It is seen that the TL intensity at 350 K in samples with the dosimetric peak having a small FWHM (35–45 K) could change over broad limits depending on the concentration of shallow traps. However, in samples with a wide dosimetric peak (the FWHM being over 45 K) the intensity of the TL at 350 K was small and changed insignificantly in different samples.

It is known that the variation of the FWHM of the main peak may be caused by the presence of impurities, specifically tetravalent titanium, in the aluminum oxide lattice [6]. If the concentration of the Ti^{4+} impurity is large, the contribution from processes of the charge-carrier transport between shallow traps and impurity titanium ions, which are responsible for widening of the main peak, becomes apparent. This charge transport competes with the recombination of carriers on luminescence centers, leading to the attenuation of the TL intensity in the peak at 350 K.

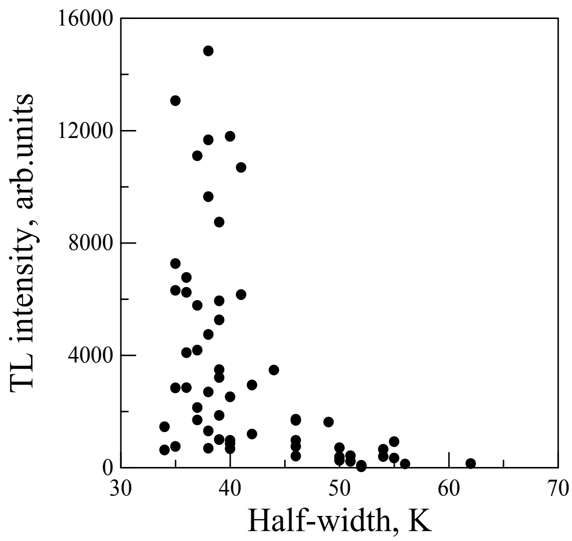

Fig. 5. Dependence of the TL intensity in the peak at 350 K on the FWHM of the dosimetric peak

It was already noted in the foregoing that deep traps responsible for the TL peaks at 730 and 880 K considerably influence TL processes in the dosimetric maximum at 450 K. Dosimetric traps also represent deep traps with respect to trapping centers responsible for the TL peak at 350 K and, as it is shown in this study, actively interact with shallow traps. It may be assumed therefore that the interactive interaction of deep and shallow traps is a general feature that determines TL processes in crystals of anion-defective aluminum oxide.

4. Conclusions

1. It was found that the intensity of the TL peak at 350 K can change considerably in crystals of anion-defective aluminum oxide having the same dosimetric sensitivity.

2. A method was proposed and implemented for evaluation of the radiation sensitivity of the TL peak at 350 K during stepwise filling and depletion of dosimetric traps. As a result, it was possible to prove experimentally the presence of the competitive interaction between traps responsible for the dosimetric peak and shallower trapping centers that give rise to the TL peak at 350 K.
3. It was found that the dose dependence of the TL maximum at 350 K flattens out at doses corresponding to the superlinear section of the dose characteristic of the peak at 450 K. Then one of reasons for the superlinearity may be weakening of the competition by shallow traps for the capture of free charge carriers.
4. The obtained experimental evidence to the presence of the competition for the capture of charge carriers by shallow traps is the manifestation of a general regular feature of the interactive interaction between deep and shallow traps in anion-defective crystals of aluminum oxide.

References

[1] V.S. Kortov, A.I. Surdo, F.F. Sharafutdinov, ZhPS, **67**, No. 7. 72–76 (1997).
[2] V.S.Kortov, S.V.Nikiforov, E.Z.Sadykova, Functional Materials, 232–236 (2005).
[3] M.S. Akselrod, V.S. Kortov, I.I. Milman, UFZh, **28**, No. 7, 1053–1056 (1983).
[4] Bessonova, T.I. Gimadova, I.A. Tale, L.A. Avakumova, L.A. Litvinov, ZhPS, **54**, No. 3, 433–437 (1991).
[5] V.S. Kortov, I.I. Milman, Izv. VUZov, Fizika, **39**, No. 11, 145–161 (1996).
[6] V.S. Kortov, I.I. Milman, S.V. Nikiforov, FTT, **39**, No. 9, 1538 (1997).