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Features of thermoluminescence in anion-defective alumina single crystals after highdose irradiation

V.S. Kortov*, S.V. Zvonarev, V.A. Pustovarov, A.I. Slesarev

Physics & Engineering Institute, Ural Federal University, 620002 Yekaterinburg, Russia

HIGHLIGHTS

- In TLD-500 detectors the dosimetric glow curve does not change with dose increase.
- Sublinear dose response of the main TL peak was found in the range 80–800 kGy.
- In PL spectra of irradiated detectors there appears a band with $\lambda_{max} = 550$ nm.
- PL intensity in a new spectrum band depends on the dose.
- F_2-type centers are responsible for the change in PL spectra and TL yield.

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ABSTRACT

The effect of high-dose irradiation by electron beam with nanosecond duration and by gamma-rays on thermoluminescence (TL) yield of anion-defective dosimetric Al_2O_3:C crystals is studied. It is shown that in a wide dose range up to 10 kGy no significant changes in the TL curve shape and the temperature position of the main dosimetric peak ($T = 460$ K) are observed. The TL yield of this peak is in saturation in the high-dose range 5–80 kGy. Then anomalous increase in TL yield is registered at the dose growth up to 800 kGy. With that an intensive band appears in the green spectrum region in the photoluminescence spectrum. The role of aggregate defects forming F_2-type centers with the increase of TL yield in Al_2O_3:C crystals under high-dose irradiation is discussed.

1. Introduction

Anion-defective α-Al_2O_3:C single crystals are well-known phosphors widely used in personal dosimetry (Akselrod et al., 1990; McKeever et al., 1995; Kortov, 2007). The high-sensitive detectors TLD-500 are developed on the basis of these crystals. The oxygen vacancies formed in single crystals trap one or two electrons giving rise to F^+ and F − centers correspondingly. These centers with impurities form complex defect which is a trap of free electrons resulting from irradiation. Thermal ionization of such traps at the sample heating with the following recombination of delocalized electrons causes the main dosimetric thermoluminescent (TL) peak at 460 K. The peak intensity is proportional to the dose of X-ray, gamma- and beta-radiations in the range of 10^{-7}–10 Gy. With the dose increase higher than 10 Gy, the TL intensity of the main peak is saturated. This does not allow using it for high dose measurements. Therefore, TL and dosimetric properties of the main peak at high-dose irradiation have not been studied until recently. However, there is a growing interest in the design of high-dose TL detectors due to development of radiation technologies and necessity to test material radiation resistance. Some examples which illustrate the use of a number of crystalline and nanostructured detectors for high-dose measurements are available (Bilski et al., 2010; Salah, 2011; Kortov and Ustyantsev, 2013).

The objective of the paper is to study TL of the main dosimetric peak at high-dose irradiation of anion-defective Al_2O_3:C single crystals.

2. Materials and methods

The samples under study were the discs 5 mm in diameter and 1 mm in thickness. These were made of α-Al_2O_3:C single crystal grown in highly reducing atmosphere with the presence of carbon. According to the optical absorption data, the concentration of oxygen vacancies in single crystals was $1.1 \cdot 10^{17}$ cm^{-3}.

* Corresponding author. Tel.: +7 343 375 44 43; fax: +7 343 375 44 15.
E-mail addresses: vskortov@mail.ru (V.S. Kortov), s.v.zvonarev@urfu.ru (S. V. Zvonarev), vpustovarov@bk.ru (V.A. Pustovarov), a.i.slesarev@ustu.ru (A. I. Slesarev).

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To irradiate the crystals, the radiation sources of three types were employed. The source of high-dose radiation was a pulse electron accelerator with the following parameters: pulse length is 2 ns, medium electron energy is 130 keV, and current density is 60 A/cm^2. The absorbed dose from one accelerator pulse was preliminary calculated and then experimentally proved by the optical absorption method with the use of film dosimeters SORD (F) R-5/50. The medium absorbed dose (in the water) was found to be 1.5 kGy/pulse at 10 cm distance (Afanasjev et al., 2005). In this case the temperature change of the sample does not exceed 1–2 K. The due irradiation dose can be obtained by increasing a pulse number. High-dose gamma-irradiation was carried out by means of industrial ^{60}Co-gun with the dose rate of 1.3 Gy/h. For low-dose irradiation ^{90}Y/^{90}Sr beta-radiation source with 1.92 Gy/h dose rate was used.

The thermoluminescence curves were measured at heating rate of 2 K/s by means of photomultiplier FEU-142. Photoluminescence (PL) spectra at UV-excitation were measured by means of the 400 W gas-discharge deuterium lamp, the prismatic double monochromator DMR-4 and photoelectron multiplier R-6358-10 (Hamamatsu).

3. Results

The electron nanosecond pulse effect on the samples under study is an effective express method of high-dose irradiation. First of all, it was necessary to find out whether high-dose irradiation affected the temperature position and shape of the main dosimetric peak TL. Fig. 1 shows the given peak TL curves at low-dose and high-dose irradiation of the crystals. It can be seen that the TL curves do not noticeably change their shape when the dose increases by more than 7 orders. The band half-width remains practically constant. The temperature position of TL peaks varies slightly depending on the dose. The data obtained show that high-dose irradiation does not destroy luminescence centers present in the crystal which are responsible for the main dosimetric TL peak.

Fig. 2 presents the results of the research into dose dependence of TL yield after irradiation by pulse electron beam. One can see that saturation of the main dosimetric peak TL is observed in a wide range of doses from 5 kGy to 80 kGy. One of the possible reasons for saturation of the peak TL intensity at high-dose irradiation is known to be the limited capacity of the traps capable of capturing free electrons arising from irradiation (Chen et al., 2006). Moreover, Fig. 2 also shows that after the saturation region a linear TL yield is registered in the dose range from 80 kGy to 800 kGy. Then, TL intensity saturation and its subsequent decrease follow again.

One can assume that anomalous increase in TL yield in the given high dose range is possibly caused by the formation of new luminescent centers when the dose of 70–80 kGy is reached. The results of PL spectra measurements prove this assumption. Fig. 3 gives PL

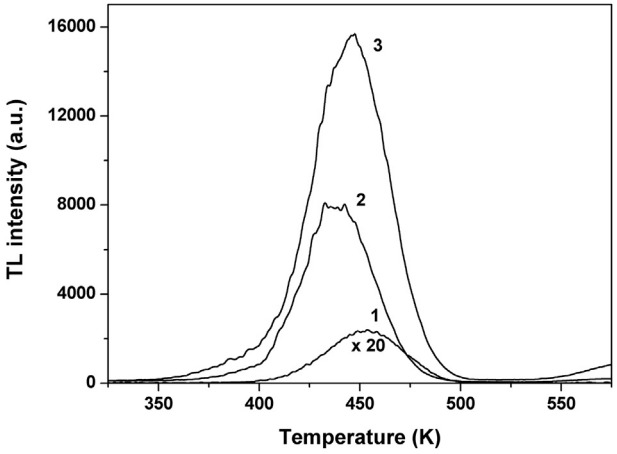

Fig. 1. TL curves in single crystals Al_2O_3:C after β-irradiation with the dose 32 mGy (1) and after electron beam irradiation with the doses 1.5 kGy (2) and 300 kGy (3).

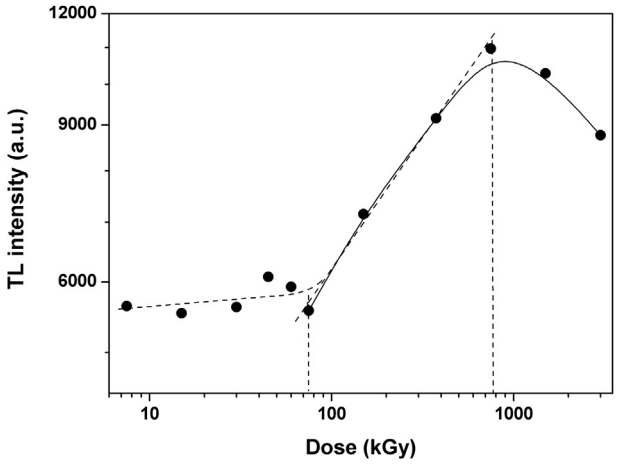

Fig. 2. TL yield dependence in the main dosimetric peak at ultra-high-dose irradiation of the Al_2O_3:C single crystal by an electron beam.

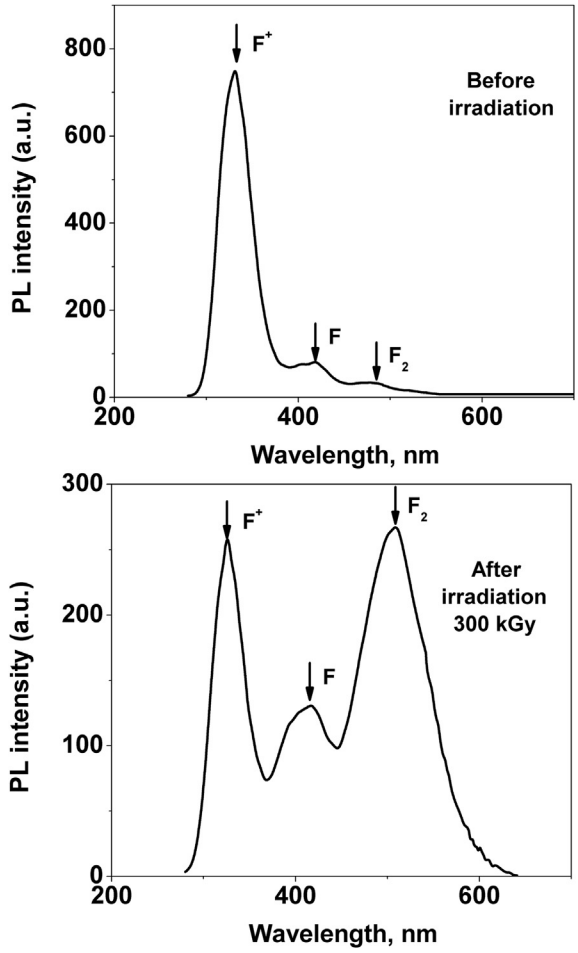

Fig. 3. PL spectra in Al_2O_3:C single crystal before and after high-dose irradiation by an electron beam. E_{exc} = 4.8 eV, T = 295 K.

spectra of the non-irradiated sample and the ones being irradiated by pulse electron beam of 300 kGy. PL spectra were excited into the absorption band of F^+-centers ($\lambda_{exc} = 255$ nm). Fig. 3a shows that in the non-irradiated samples simultaneously with the F^+-centers emission in PL spectrum, low intensity bands of F-centers ($\lambda_{emis} = 420$ nm) and F_2-type centers ($\lambda_{emis} = 550$ nm) are registered. Neutral F_2-centers are known to be formed by aggregate defects made up of two oxygen vacancies which trapped four electrons. Alongside with the neutral F_2-centers, charged F_2^+ and F_2^{2+}-centers form as well. Fig. 3b illustrates that after high-dose irradiation the band intensity of F_2-type centers sharply increases showing the growth of their concentration in the crystal.

Formation of F_2-centers in various charge states is also found at irradiation of alumina crystals with intensive beams of fast neutrons and heavy ions (Izerrouken and Benuahia, 2010; Song et al., 2007). This illustrates a common characteristic connected with the creation of aggregate defects at the initial stages of crystal disordering under the influence of intensive radiation.

PL spectrum change with the dose increase was studied in more detail after irradiation of Al_2O_3:C crystals by gamma-radiation ^{60}Co. Similarly to the previous case, the irradiated samples were excited into the absorption band of F^+-centers (255 nm). Fig. 4 shows that in PL spectrum the band intensity of F^+-centers changes as the irradiation dose grows. F^+-center emission is high at the doses not over 10 Gy. Further increase of the dose up to 100 Gy causes a drop (nearly three times) in the band intensity of F^+-centers indicating the decrease of their concentration. The subsequent dose increase by over 2 orders (up to 72.8 kGy) does not significantly affect emission intensity of F^+-centers. Similar decrease in F^+-center concentration was found in Al_2O_3:C single crystal after high-dose irradiation by means of optical absorption method (Yukihara et al., 2003). It is of interest to note that high-dose irradiation practically does not affect F-centers concentration in the crystals under study. When the F-centers were excited into the optical absorption band (205 nm), the PL band intensity ($\lambda_{emis} = 420$ nm) varied slightly with the two-order dose change.

Fig. 4 also shows that gamma-irradiation dose increase in the crystals is accompanied by formation in the PL spectrum of the wide band with the maximum emission at 550 nm similar to electron beam irradiation. The high-dose irradiation enhances a probability of aggregate defects creation from F^+-centers. As a result F_2-type centers in different charged states (F^+_2, F^{2+}_2) arise. This situation corresponds to the F^+ -centers concentration decrease observed experimentally. The charged F_2-type centers are the electron traps. The free electrons resulting from irradiation or thermostimulation recombine according to the following reactions:

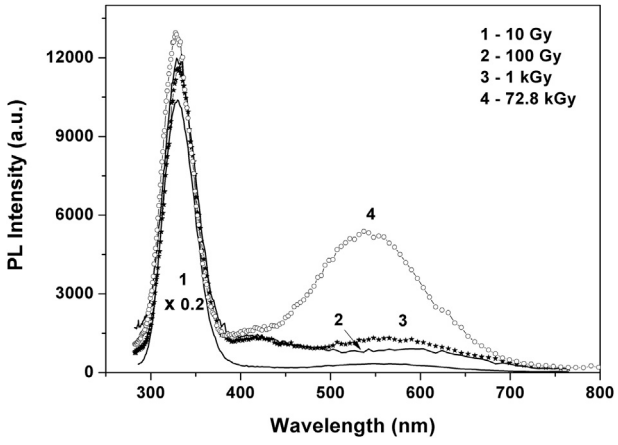

Fig. 4. PL spectra in Al_2O_3:C single crystal after gamma-rays irradiation with various doses: $E_{exc} = 4.8$ eV, $T = 295$ K.

$F_2^+ + e \rightarrow F_2^* \rightarrow F_2$ and $F_2^{2+} + 2e \rightarrow F_2^* \rightarrow F_2$. In both cases transition of the excited F_2^*-state into the ground one is accompanied by photon emission ($\lambda_{emis} = 500$ nm) (Solov'ev et all., 2012). Moreover, aggregate centers can be produced by F^+-centers and impurities, e.g. those of Mg. The studied crystals contained 4ppm of Mg impurity. Therefore, one can expect F^+_2(2 Mg)-centers with $\lambda_{emis} = 517$ nm to form in them (Akselrod and Akselrod, 2006). At the double ionization of F_2-centers the emission band with $\lambda_{emis} = 550$ nm is registered as a result of photostimulation. Superposition of the mentioned above centers emission bands allows us to observe a relatively wide green band in the PL spectrum of the irradiated Al_2O_3:C crystals (see Figs. 3b and 4).

It is necessary to note that accumulation of aggregate F_2-centers in different charge states proceeds in a non-monotonic way. As Fig. 4 shows, gain of intensity in the green region is not significant when the dose increases in the range of $10–10^3$ Gy. Similar results were obtained at the dose of 13.2 kGy (not given in Fig. 4). Considerable intensity increase of this region occurs at the crystal irradiation by gamma-rays dose of 72.8 kGy. We can suppose that the dose (70–80) kGy is a threshold one and further on from it both gamma-photon and electron beam irradiation leads to intensive aggregation of oxygen vacancies in Al_2O_3:C crystals. This process does not depend on radiation type. As it follows from Fig. 2, it is when the dose of 80 kGy is achieved at electron beam irradiation that TL yield increase begins.

4. Conclusions

The obtained results allow us to conclude that the charged centers of F_2-type resulting from high-dose irradiation are the additional electron traps which lead to anomalous TL yield increase in the main dosimetric peak of Al_2O_3:C crystals at a particular irradiation stage. The linear TL yield found in the main dosimetric peak of Al_2O_3:C crystal in the dose range of (80–800) kGy can be used for ultra-high-dose measurements by means of TLD-500 detectors. The charged centers of F_2-type cause appearance and increase of the wide green band in the PL spectrum of the irradiated crystals. In spite of this, TL curves do not change their shape and the crystals are not colored. All the experimental data are evidence of high radiation resistance of the luminescent centers in Al_2O_3:C crystals.

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