



# Accumulation features and TL of TLD-500 detectors in a wide temperature range at pulsed and continuous high-dose irradiation



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## HIGHLIGHTS

- Using the 580 and 830 K TL peaks allow expand range of the recorded doses to 1 kGy.
- Under pulsed irradiation the upper limit can be expanded to several MGy.
- Ultimate irradiation dose rate of TLD-500 detectors is  $10^{10}$  Gy/s.
- In the spectrum of the 830 K TL peak an UV emission with  $h\nu = 4.1$  eV dominates.
- The maximum of the optical depletion spectrum of TL peak at 830 K is around 5.2 eV.

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## ABSTRACT

The results of a comparative research of thermoluminescence (TL) of TLD-500 detectors based on anion-defective corundum irradiated with continuous and pulsed X-ray and pulsed electron beams in a range of doses of  $0.3 \div 10^7$  Gy, dose rates of  $0.02\text{--}2.6 \cdot 10^{11}$  Gy/s, and in a temperature range of 300–950 K are presented. It is found that, in contrast to continuous and pulsed irradiation, upon pulsed irradiation with a duration of 10 ns and dose rate of  $P_p \geq 5 \cdot 10^6$  Gy/s, the first linear region of dose dependences for TL peaks at 450, 580 and 830 K is, instead of saturation, followed by a second one with a smaller slope at doses near 2, 200 and  $10^3$  Gy. Moreover, the slope of the second region increases with growing  $P_p$ . It was also found that dose dependence for the peak at 830 K in the area of the first linear region at  $10\text{--}10^3$  Gy remains invariable at  $P_p \leq 10^{10}$  Gy/s. It is shown that the upper limit of doses registered by TLD-500 detectors can be increased to  $2 \cdot 10^3$  and  $6 \cdot 10^6$  Gy for continuous and pulsed irradiation, respectively. New broadband UV luminescence with a maximum  $h\nu = 4.1$  eV and half width  $H = 0.85$  eV was registered within the TL peak spectrum at 830 K. Besides, the optical depletion spectrum in which a single band with  $h\nu = 5.2$  eV and  $H = 1.6$  eV is observed was investigated for a trap causing a peak at 830 K.

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## 1. Introduction

Intensive pulsed beams of ionizing radiations are increasingly employed in radiation technologies, nondestructive control and medicine. The range of doses is  $10^{-5}$ – $10$  Gy for personal dosimetry problems and  $10\text{--}10^7$  Gy for radiation technologies problems. Unfortunately, most conventional methods based on optical and paramagnetic spectroscopy or chemical reactions fail to provide accurate dosimetry at elevated temperatures (Schönbacher et al.,

2009). Extra difficulty is associated with measurements of doses in pulsed radiation fields due to strong electromagnetic interference and considerable pulsed dose rates  $P_p$  amounting to  $10^6\text{--}10^{11}$  Gy/s (Vavilov and Gorbunov, 1985). A possible way to solve this problem is detection, study and practical use of the relatively understudied phenomenon of high-temperature thermoluminescence (TL) of TLD-500 detectors based on anion-defective corundum (Lo et al., 2006; Mil'man et al., 2008; Surdo et al., 2014). For example, in (Lo et al., 2006), dose dependences for TL peaks at 590, 730, 780 and 920 K are investigated while irradiating a powdered form of TLD-500 with beta particles of a  $^{90}\text{Sr}/^{90}\text{Y}$  source over a dose range of 1–100 Gy. In Mil'man et al. (2008), the occurrence of a high-temperature TL peak at 830 K was registered when X-raying TLD-500 detectors at 700 K. Its dose

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dependence is shown to be linear within the range  $D = 10^4\text{--}6 \cdot 10^6$  Gy when irradiating TLD-500 detectors with a pulsed electron beam (Surdo et al., 2014). Yet, TL features of TLD-500 detectors remain understudied in the main and higher-temperature peaks under pulsed and continuous irradiation in a wide range of doses and dose rates including areas typical to personal, emergency and technological dosimetry. Hence the objective of this paper is comparative research of features of accumulation and highlighting of dosimetric information provided by TLD-500 detectors when registering continuous and pulsed X-ray and electron radiations over a broad range of doses, dose rates, and temperatures.

## 2. Experiment

The objects of the research were samples of TLD-500 detectors (Ural Federal University, Russia) 5 mm in diameter and 1 mm in thickness with medium sensitivity to irradiation, made of single crystals of anion-defective corundum ( $\alpha\text{-Al}_2\text{O}_3\text{-}\delta$ ) with a concentration of  $\text{F}^+$  and  $\text{F}$  centers (anion vacancies with one and two electrons respectively)  $\sim 10^{17}$   $\text{cm}^{-3}$ . Sources of continuous and pulsed X-ray radiation were devices URS-55 (Cu anode,  $U_H = 55$  kV,  $I_{\text{max}} = 18$  mA,  $P = 0.3$  Gy/s) and Arina-3 (IMA5-320D tube,  $U_H = 200$  kV,  $f = 5$  Hz,  $P_p \leq 2.6 \cdot 10^7$  Gy/s), respectively. Thickness of the half-attenuation layer in  $\alpha\text{-Al}_2\text{O}_3$  is 11.5 mm for radiation of the Arina-3 device and 2.7 mm for URS-55. The values of  $D$  for continuous and pulsed X-ray irradiation were determined from the readings of water-calibrated TLD-100 detectors a feature of which is independence of TL yield from dose rates up to  $1.7 \cdot 10^8$  Gy/s (Chen and McKeever, 1997). The  $P_p$  of pulsed X-ray radiation was determined as the ratio of the average dose per pulse as measured by the TLD-100 to pulse duration. Irradiation with pulsed electron beams was provided by the Arina-02 device (IMA2-150E tube,  $E_e = 130$  keV,  $\tau_p = 10$  ns,  $f = 10$  Hz,  $P_p \leq 2.6 \cdot 10^{11}$  Gy/s) and by linear accelerator UELV-10-10S ( $E_e = 10$  MeV,  $I_{a,\text{max}} = 1$  mA,  $\tau_p = 15$   $\mu\text{s}$ ,  $f = 210$  Hz,  $P_p = 3 \cdot 10^6$  Gy/s).  $D$  (in water) at pulsed electron irradiation was determined from the response of dosimetric colored films SO PD (F)R-5/50 (VNIIFTRI, Russia) and the dose rate, as in the measurement of  $P_p$  of pulsed X-ray radiation, while using TLD-100. The capability of dose measurement at  $P_p \leq 10^{12}$  Gy/s using the above mentioned film was shown by Afanas'ev et al. (2005).

TL curves were measured on an automated installation at a heating rate of 2 K/s. The TL signal was registered by FEU-142 photomultiplier (Gran Company, Russia) with reduced sensitivity to thermal radiation of the heater whose maximum temperature was 1200 K. Photon energy dependence of TL response (Chen and McKeever, 1997) was considered when comparing TL yields for samples exposed to X-ray radiation with different energies. The ranges of electrons with differing energies and correspondingly effective thicknesses of luminescing layers were estimated under electron irradiation. Samples under study were annealed at 1200 K during 10 s prior to each irradiation for depletion of the main and deep traps.

TL spectra of irradiated samples were registered within a temperature range  $T = 300\text{--}950$  K by a Cary Eclipse spectrofluorimeter in the mode of continuous cyclic scanning over a range of 250–750 nm (1.6–5 eV) at a speed of 90 nm/s and with an entrance slit spectral width of 10 nm. A technique based on analyzing residual TL curves was used to determine the optical depletion spectrum.

## 3. Results and discussion

Fig. 1(a, b) gives TL curves and dose dependences of light sums  $S_{450}$ ,  $S_{580}$  and  $S_{830}$  in the most intensive peaks at 450, 580 and 830 K

for a TLD-500 detector irradiated with continuous X-rays with doses from 0.2 to 2000 Gy at  $P = 0.3$  Gy/s. It can be seen (Fig. 1a, curves 1–3, Fig. 1b, curves 1 and 2) that  $S_{450}$  dependence begins to be saturated at  $D \geq 8$  Gy and to be accompanied by increasing intensity of the so-called “chromium” peak at 580 K caused by the chromium impurity ever present in  $\alpha\text{-Al}_2\text{O}_3$  crystals at a trace level (Bessonova et al., 1977). The beginning of the dependence saturation area  $S_{580}$  is accompanied by the emergence of little-studied high-temperature peaks at 710 and 830 K (Fig. 1a, curves 4–6) (Mil'man et al., 2008; Surdo et al., 2014). As can be seen (Fig. 1b, curve 3), the dependence  $S_{830}$  begins to be saturated at  $D \geq 1 \cdot 10^3$  Gy. Besides, the occurrence of a peak at 830 K is accompanied by the emergence of a peak near 390–400 K that is also saturated at  $D \geq 1 \cdot 10^3$  Gy.

Generally, linear regions of dose dependences presented in log–log coordinates can be described by a known relation  $S = A \cdot D^k$  (Chen and McKeever, 1997) where  $A$  and  $k$  are calibration and dose factors. For dependences  $S_{450}$ ,  $S_{580}$  and  $S_{830}$  shown in Fig. 1b,  $k$  values are 1.1, 1.6 and 1.6 respectively.

The data obtained suggest that, upon continuous X-ray irradiation, growth of  $D$  is accompanied by consecutive trap filling causing TL peaks at 450, 580 and 830 K. Another important finding of research into high-temperature TL of TLD-500 detectors irradiated with continuous X-rays is the disclosure of a potential to expand the dose range to  $10^3$  Gy.

The behavior of dose dependences of light sums highlighted in the TL peaks discussed changes dramatically if TLD-500 detectors are exposed to pulsed nanosecond X-ray or electron radiations. For that matter, it should be noted that irradiation with a pulsed beam does not affect the shape of TL curves (Surdo et al., 2014). Fig. 2 shows dose dependences of light sums  $^P S_{450}$  (curves 1–3) measured in the peak at 450 K under pulsed X-ray irradiation (curves 1 and 2) with  $P_p$  values at  $5 \cdot 10^6$  and  $2.6 \cdot 10^7$  Gy/s and under electron irradiation (curve 3) with  $P_p = 1.2 \cdot 10^{10}$  Gy/s. It follows from a comparison of dependences  $^P S_{450}$  and  $S_{450}$  (curve 4 corresponds to curve 1 in Fig. 1b) over a dose range  $D \leq 2$  Gy corresponding to a linear region for continuous irradiation that they are scarcely distinguishable.  $k$  values are  $\sim 1.1$ , as in the case of continuous irradiation. In the range of high doses ( $D \geq 2$  Gy) however, dependences  $^P S_{450}$  are not saturated at  $P_p = 5 \cdot 10^6$  and  $2.6 \cdot 10^7$  Gy/s while acquiring a second linear region, but with a smaller slope. Furthermore, as shown in Fig. 2, dose factors for the second linear region depend on the pulsed dose rate. For example, the  $k$  value increases from 0.4 to 0.8 with  $P_p$  increasing from  $5 \cdot 10^6$  to  $1.2 \cdot 10^{10}$  Gy/s, saturation start shifting to  $\sim 100\text{--}300$  Gy.

Dose dependences  $^P S_{580}$  for the chromium peak at 580 K have a similar character for varying  $P_p$ . Dose factors do not depend on  $P_p$  in the area of the first linear region and equal to 1.6, as in the case of continuous irradiation. In the second linear region,  $k$  value increases from 0.7 to 0.9 as  $P_p$  increases from  $2.6 \cdot 10^7$  to  $1.2 \cdot 10^{10}$  Gy/s, saturation occurring at doses  $\sim 10^3$  Gy.

The results of study of dose dependences  $S_{830}$  and  $^P S_{830}$  are of the greatest interest in terms of a substantially larger limit of traceable doses and possible higher temperatures when irradiating TLD-500 detectors. Fig. 3 shows dependences  $^P S_{830}$  obtained when irradiating a sample with pulsed electron beams with  $\tau_{p1} = 10$  ns (curves 1 and 2) and  $\tau_{p2} = 15$   $\mu\text{s}$  (curve 3) generated by the Arina-02 device and UELV-10-10S accelerator, respectively. Two  $P_p$  values equal to  $1.2 \cdot 10^{10}$  Gy/s (curve 1) and  $2.6 \cdot 10^{11}$  Gy/s (curve 2) were used to measure the  $^P S_{830}$  dependence for  $\tau_{p1} = 10$  ns within a broad dose range from 360 Gy to  $3 \cdot 10^7$  Gy. As can be seen, curves 1 and 2 coincide within a range from  $3 \cdot 10^4$  to  $2 \cdot 10^5$  Gy. Dose dependences  $^P S_{830}$  measured at  $D \leq 2 \cdot 10^3$  Gy have the same appearance ( $k = 1.6$ ) as for continuous irradiation (see Fig. 1b, curve 3). At  $D \geq 2 \cdot 10^3$  Gy, the behavior of dependences  $^P S_{830}$  discussed

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