Thermoluminescence and optically stimulated luminescence disadvantages of α-Al_2O_3:C crystal grown by the temperature gradient technique*

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Recently, α-Al_2O_3:C crystal with highly sensitive thermoluminescence (TL) and optically stimulated luminescence (OSL) has been successfully grown by the temperature gradient technique. This paper investigates the heating rate dependence of TL sensitivity, light-induced fading of TL signals and thermal stability of OSL of α-Al_2O_3:C crystals. As the heating rate increases, the integral TL response decreases and the dosimetric glow peak shifts to higher temperatures in α-Al_2O_3:C crystals. Light-induced fading of TL increases with the irradiation dose, and TL response decreases as the exposure time increases, especially in the first 15 minutes. With the increasing intensity of the exposure light, the TL fading of α-Al_2O_3:C crystal increases sharply. The OSL response of as-grown α-Al_2O_3:C crystal is quite stable below 373 K and decreases sharply for higher temperatures.

Keywords: α-Al_2O_3:C, thermoluminescence, optically stimulated luminescence, light-induced fading

PACC: 7860K, 7630M

1. Introduction

The α-Al_2O_3:C crystal, which was first grown by Akselrod *et al.* in the 1990s,[1] has been widely used as a material for thermoluminescence dosimetry (TLD) and recently for optically stimulated luminescence dosimetry (OSLD). The major advantage of α-Al_2O_3:C crystal is that it possesses a high thermoluminescence (TL) sensitivity to α, β and γ-rays, which is 40–60 times higher than that of LiF (TLD-100). Additionally, α-Al_2O_3:C crystal also shows other advantages, such as a single glow peak at ∼460 K, low background and dose threshold (0.1 μGy with nitrogen flow), low fading during storage in the dark (less than 5% per year), wide linear dose response from 10^{-7} to 10 Gy and a simple emission spectrum with a maximum at 420 nm which corresponds to the sensitivity of low noise high sensitivity photomultiplier tubes, and so on.[2] All the above merits make α-Al_2O_3:C crystal an attractive candidate for low dose and prompt measurement applications, such as personal and environmental dosimetry.

However, with regard to TL or OSL materials none of them is perfect. Two main disadvantages have been reported in α-Al_2O_3:C crystal so far. Firstly, the TL sensitivity depends strongly on the heating rate,[1] and secondly its sensitivity is related to light exposure.[3,4] The TL sensitivity decreases as the heating rate increases; this effect could be a disadvantage in routine dosimetry when fast heating readers are often used. In α-Al_2O_3:C crystal, the light sensitivity manifests itself in three ways:[5] (i) a light-induced fading of the TL signal; (ii) the generation of TL signal in unirradiated samples; (iii) the phototransfer of charge from deep state to shallower states giving rise to a phototransferred (PTTL) signal. However, the light-induced fading of the TL signal from α-Al_2O_3:C and the subsequent PTTL into shallow traps indicate the potential of the crystal as a sensitive OSL dosimeter. The precision of the α-Al_2O_3:C detector decreases heavily because of the light-induced fading of the TL signal.

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Recently, α-Al_2O_3:C crystal with highly sensitive TL and OSL was successfully grown by the temperature gradient technique (TGT).[6] For practical application, more work needs to be done. The purpose of this paper is to evaluate the performance of α-Al_2O_3:C crystal grown by the TGT method, especially the TL and OSL disadvantages of the crystal, which can provide some useful suggestions for future applications. In the present paper, the influence of the heating rate on TL sensitivity, the light-induced fading of the TL signal and the thermal stability of OSL in α-Al_2O_3:C crystal grown by the TGT method are reported.

2. Experimental procedure

The α-Al_2O_3:C crystal doped with 5000 ppm carbon was grown by the TGT method and the growth process was described in Refs. [6] and [7]. Samples with a size of $5\times5\times1$ mm^3 were cut and polished for TL and OSL experiments. All the samples were annealed in air at 773 K for 10 min before any experiments to remove the stresses arising during the growth, cutting and polishing processes. The TL and OSL measurements were performed on a Risø TL/OSL-DA-15 automatic reader (Risø National Laboratory, Denmark) with an integrated ^{90}Sr/^{90}Y beta source. The light detector was an Electron Tubes 9235QB photomultiplier tube. Three Hoya U340 filters of thickness 2.5 mm are used for the light detection. An optical excitation unit consisting of a blue light emitting diode (LED) cluster with wavelength $\lambda = 470\pm30$ nm was used in the OSL measurements.

3. Results and discussion

A generalized model used to describe the TL and OSL processes of α-Al_2O_3:C crystal is shown in Fig. 1. The TL and OSL processes may be described by considering only the levels associated with the main dosimetric trap (MDT) and the recombination centre (RC). Irradiation creates electron-hole pairs (transition 1). The electrons are either trapped at the MDT (transition 2) or recombine with F^+-centres at RC (transition 4), creating an F-centre. The free holes generated during irradiation are captured by the F-centres, increasing the concentration of F^+-centres. The electrons are freed from the MDT and recombine with the F^+-centres, resulting in 410 nm emission by heating or optical stimulation. In order to explain the TL and OSL dose response and sensitivity changes, three other levels, deep electron traps (DETs), deep hole traps (DHT) and shallow traps (ST), are introduced. The free holes can be captured by DHT (transition 5). The electrons can also be trapped by DET (transition 3) and ST (transition 7) or combine with the holes at DHT (transition 6). Two STs have been identified in α-Al_2O_3:C, one at 260 K and another at 310 K, but only the latter has relevance at room temperature.[3] Empty DETs act as competitors, capturing electrons excited to the conduction band during irradiation (or illumination) or electrons stimulated from the MDT during the recombination stage. As the DETs are filled, the competition becomes less important and more electrons can be trapped at the MDT during the irradiation, resulting in increased sensitivity.

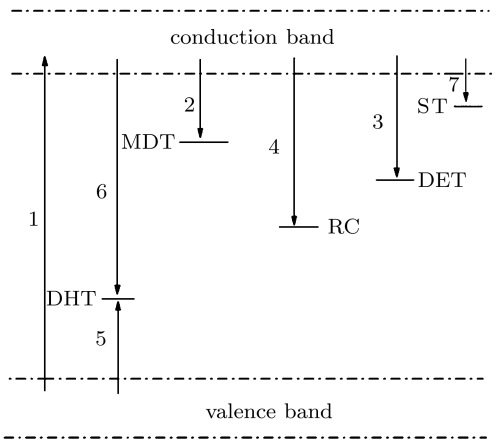


Fig. 1. Energy band model describing charge transitions through the delocalized bands in α-Al_2O_3:C, where MDT = the main dosimetric trap, DET = the deep electron trap, DHT = the deep hole trap, RC = recombination centre (F^+-centres), ST = the shallow traps. The main electron and hole transitions during the irradiation stage with beta rays are depicted: 1 electron–hole pair creation; 2 electron capture by the MDT; 3 electron capture by the DET; 4 electron–hole recombination at RC F^+-centres; 5 hole capture by DHT; 6 electron–hole recombination at DHT; 7 electron capture by the ST.

3.1. Effects of the heating rate on TL

Figure 2 is the TL curves of α-Al_2O_3:C crystal under different heating rates after irradiation with 0.1 Gy ^{90}Sr β-rays, and the integral TL response and glow peak temperature as a function of the heating rate is shown in the inset. With increasing heating rate, the integral TL response of the crystal decreases and glow peak shifts to higher temperatures. According to the

Randall–Wilkins first-order kinetics model,[8] the TL intensity I can be described as

$$I = n_0 \times \frac{s}{\beta} \times \exp[-E/kT] \times \exp\left[-\frac{s}{\beta}\int_{T_0}^{T} \exp(-E/kT')\,\mathrm{d}T'\right], \quad (1)$$

where n_0 is the total number of trapped electrons in MDT, s is the frequency factor, β is the heating rate, E is the activation energy of MDT, T is the temperature and k is the Boltzmann constant. When other parameters are kept constant, it is obvious to see that the glow peak T shifts to higher temperatures and the TL intensity I decreases as the heating rate β increases. When the heating rate increases from 1 to 10 K/s, the total light output decreases about 4.5 times in our samples. X. Yang *et al.*[7] have reported that the TL response decrease rate increases with an increasing amount of carbon doped in α-Al_2O_3:C crystals.

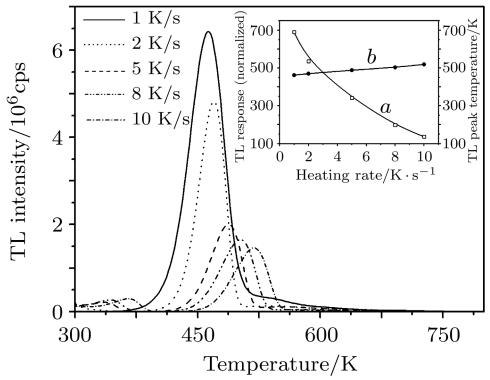


Fig. 2. The TL curves of α-Al_2O_3:C crystal under different heating rates, the inset is the integral TL response (curve a), glow-peak temperature (curve b) as a function of the heating rate.

Such changes are not expected from a consideration of the kinetics of TL production in TLD materials, in which the total light is expected to be independent of the heating rate.[9] Two mechanisms have been proposed to explain that the TL response depends on the heating rates in α-Al_2O_3:C crystal. Akselrod *et al.*[10] suggested that the decrease of integral TL response can be attributed to the thermal quenching of the F centre luminescence efficiency. However, Milman *et al.*[5] proposed a temperature-dependent competition process in which the RC competes with DETs for the localization of electrons during TL readout. In their suggestion the DETs compete more effectively at higher temperatures and, thus, as the sample is heated during readout, more trapping at the DETs takes place. Since the TL process shifts to a higher temperature range as the heating rate increases, a reduced TL output is observed at higher heating rates due to this competition effect. We think that both processes have effects on the decrease of integral TL output, and thermal quenching of the F centre emission may be the primary factor according to the present results.[10–12] This is one disadvantage of the α-Al_2O_3:C detectors which should be taken into account when using them in TL readers with various heating rates.

3.2. Light-induced fading of TL

The α-Al_2O_3:C crystal exhibits high light sensitivity, which leads to the light-induced fading of α-Al_2O_3:C detectors. Figure 3 is the changes of TL response in α-Al_2O_3:C crystal after irradiation with different ^{90}Sr beta doses and then exposure in the room centre on a sunny day (∼200 lx) for various times. Each datum presents the average integral TL response of three samples. The TL response decreases as the exposure time increases, especially in the first 15 minutes. The irradiation dose also has an effect on the light-induced fading of α-Al_2O_3:C. With the increase of the irradiation dose, the light-induced fading of TL becomes faster. The TL response of the sample irradiated with 0.05 Gy decreases about 60% after 15 mins exposure, whereas with 30 Gy the loss is 90%. Izak-Brian *et al.*[3] reported that the loss of the TL signal following 10 minutes of exposure to 5000 lx of incandescent light was ∼83% for irradiation with 10 Gy gamma rays, and they also found that light-induced fading of TL in α-Al_2O_3:C crystal increased with irradiation dose.

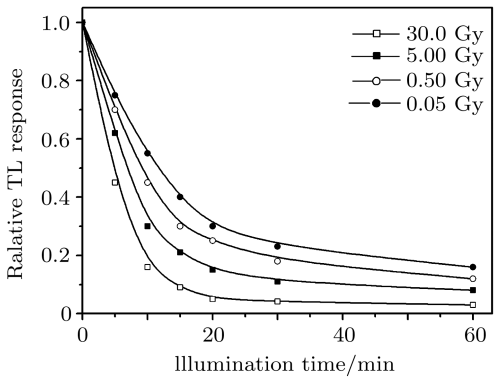


Fig. 3. Light-induced fading of TL in α-Al_2O_3:C crystal under different ^{90}Sr beta doses, the samples were exposed in the centre of the room on a sunny day (∼200 lx) for different times after irradiation.

Figure 4 shows light-induced fading of TL in α-Al_2O_3:C crystal after irradiation with 5.0 Gy beta

dose and then exposure in four typical light conditions. The light-induced fading of TL in α-Al_2O_3:C crystal increases sharply as the light intensity increases. When the sample was exposed outdoors on a sunny day ($\sim 10^5$ lx) for 15 mins, the total loss of the TL signal reached ~95%, whereas in black night ($\sim 10^{-3}$ lx) the loss is less than 3%. The TL signal decreases ~35% after 15 mins exposure in the room centre on a cloudy day. Therefore, a cover which eliminates exposure to light should be used when α-Al_2O_3:C detectors are employed for personal and environmental radiation measurements.

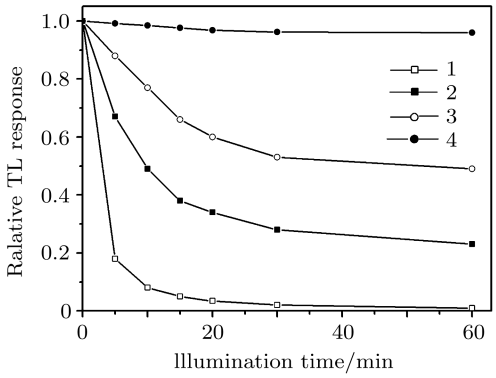


Fig. 4. Light-induced fading of TL in α-Al_2O_3:C crystal after irradiation with 5.0 Gy beta rays and then exposure in different light conditions: 1 outdoors on a sunny day ($\sim 10^5$ lx); 2 the room centre on a sunny day (~200 lx); 3 the room centre on a cloudy day (~25 lx); 4 black night ($\sim 10^{-3}$ lx).

The simplest interpretation of the light-induced fading of TL in α-Al_2O_3:C crystal is that the light is optically stimulating charge into the DETs from the main dosimetric trap (MDT). Two delocalized bands with delocalization temperatures of about 850 K and 1050 K were found in α-Al_2O_3:C crystal by the methods of exoelectron emission, isochronal annealing of the absorption bands and the TL response to light exposure.[1] The proposition is supported by recent PTTL data which clearly show that the main TL peak is the major source of phototransferred charge to low temperature traps.[13] Izak-Brian suggested[3] that the light-induced fading of TL in α-Al_2O_3:C crystal can be reduced by either submitting the dosimeters to an annealing process or pre-irradiation with a high dose. In summary, the light sensitivity is a big problem for α-Al_2O_3:C detectors which should be avoided in TL dosimetry, but is an advantage for α-Al_2O_3:C crystal used as OSL dosimetry.

3.3. Thermal stability of OSL

Thermal stability is one of the essential features of a dosimetric material. All the OSL experiments were done in dark conditions and the effect of light can be neglected. Figure 5 is the integral OSL response from irradiated α-Al_2O_3:C (1 Gy ^{90}Sr beta source) after annealing in various temperatures for 5 minutes. Measurements were done at room temperature after cooling the samples to room temperature. At the temperature below 390 K, the integral OSL response was almost constant, whereas the integral OSL response decreases sharply in the temperature range from 390 K to 473 K. For temperature higher than 473 K, the integral OSL response decreases slowly. Similar results have been found by Markey[14] and Bulur.[15] The TL traps (MDT) at ~460 K are the main source of the OSL emission.[14] So the sharp decrease of the integral OSL response in the temperature range from 390 K to 473 K can be attributed to thermally stimulating charges from the MDT. However, the MDT is not the only source of OSL emission, and some deep traps (DETs) also contribute to the OSL emission in α-Al_2O_3:C crystal. The slow decrease of the integral OSL response is related to thermally stimulating charges from DETs.

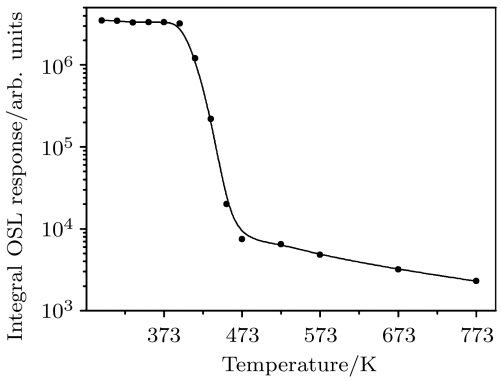


Fig. 5. Integral OSL response from 1 Gy beta ray irradiated α-Al_2O_3:C after annealing in various temperatures for 5 minutes.

Figure 6 is the integral OSL response from irradiated α-Al_2O_3:C (1 Gy ^{90}Sr beta source) after annealing in a given temperature for different times. The OSL decay curves were obtained after cooling the samples to room temperature. The integral OSL response is quite stable below 373 K and begins to decrease as the temperature increases. The longer time annealed, the greater the OSL emission loss. It is obvious that more and more electrons are thermally stimulated from the MDT as the annealing time increases

and fewer electrons are left and recombined with F^+-centres at RC. In conclusion, the integral OSL response of α-Al_2O_3:C crystal is very stable below 373 K and decreases sharply for higher temperatures. The α-Al_2O_3:C detectors would be suitable for personal and environmental dosimetry at room temperature.

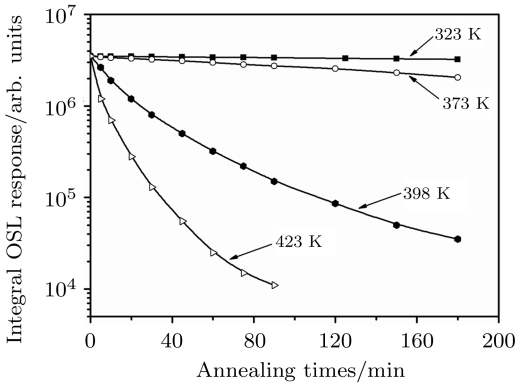


Fig. 6. Integral OSL response from irradiated α-Al_2O_3:C (1 Gy ^{90}Sr beta source) after annealing in a given temperature for different times.

4. Conclusions

The influence of the heating rate on the TL sensitivity, the light-induced fading of TL signals and the thermal stability of OSL in α-Al_2O_3:C crystals grown by the TGT method were studied in this work. With the increase of the heating rates, the integral TL response of the crystal decreases and the main glow peak shifts to higher temperatures. The heating rate dependence of TL sensitivity was mainly caused by the thermal quenching of the F centre luminescence efficiency. This is one of demerits of α-Al_2O_3:C detectors which should be taken into account when using them in TL readers with various heating rates. As-grown α-Al_2O_3:C crystal shows a high TL sensitivity to light. The TL response decreases as the exposure time increases, especially in the first 15 minutes, and the light-induced fading of TL increases as the irradiation dose increases. The light-induced fading of TL increases sharply as the light intensity increases. The loss of the TL signal reached ∼95% when exposing it outdoors on a sunny day for 15 minutes. Therefore, when α-Al_2O_3:C detectors are employed for personal and environmental radiation measurements, light exposure should be avoided. The integral OSL response is quite stable below 373 K and decreases sharply for higher temperature. This can be attributed to thermally stimulating charges from the MDTs at high temperature in α-Al_2O_3:C crystal.

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