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Optically stimulated luminescence of α-Al_2O_3:C by the vertical gradient freezing (VGF) method

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ABSTRACT

A highly sensitive α-Al_2O_3:C crystal was directly grown by the vertical gradient freezing (VGF) method, which combines the advantages of the crystal growth in the highly reducing atmosphere in the presence of graphite and the crystal directly grown by the using Al_2O_3 and graphite powder as raw materials. The main OSL characteristics of the detectors as grown were investigated. The OSL signal originates from the main TL peak, the responsible electron trap can be emptied basically by OSL readout. The mean OSL sensitivity per unit mass of the VGF α-Al_2O_3:C is 1.6 times higher than that of the TLD-500K. The OSL detection thresholds of the VGF α-Al_2O_3:C and the TLD-500K are 0.21 μGy and 0.51 μGy respectively. The OSL signals for the two kinds of detectors are reproducible within a coefficient of variation of less than 2.5% over ten re-use cycles without systematic decrease. The VGF α-Al_2O_3:C detectors shown an excellent linear response within the dose range from 1 μGy to 15 Gy. The dose can be independently re-analysed multiple times. The VGF α-Al_2O_3:C can be used to measure the very low dose in environmental monitoring and personnel dosimetry.

1. Introduction

Al_2O_3 doped with carbon (α-Al_2O_3:C) was first prepared in Russia [1,2]. The major advantages of this detector are its high sensitivity to radiation and low fading property [3–6]. The drawbacks of α-Al_2O_3:C as a thermoluminescent (TL) detector are its sensitivity to visible light and thermal quenching [3–6]. The material can be applied as a high sensitive optically stimulated luminescence (OSL) detector by using light to stimulate the luminescence signal [7–9]. At present, α-Al_2O_3:C is one of the only two kinds of OSL materials commercially available [4–10].

Now there are several methods to prepare α-Al_2O_3:C OSL detectors. α-Al_2O_3:C crystals were first grown by Czochralski (Cz) method in the highly reducing atmosphere in the presence of graphite [2]. However, the characteristics of α-Al_2O_3:C exhibits significant variations with slight changes in growth conditions [11]. Dosimetric grade α-Al_2O_3:C detectors can be prepared by heating an Al_2O_3 single crystal in the presence of graphite under vacuum [12], and by melting the alumina powder in graphite environment [13]. But the sensitivity of the α-Al_2O_3:C detectors prepared by both methods is lower than that of crystal grown by Cz method [1,12,13]. The α-Al_2O_3:C crystal was directly grown by the temperature gradient technique (TGT) [14] and Edge-Defined, Film-Fed technique (EFG) [15] with Al_2O_3 and graphite powder as raw material. The α-Al_2O_3:C crystal grown by TGT had a high sensitivity [14]. However, the TGT crystal growth system used by Yang et al. [14] has been discarded and cannot be used again.

Vertical gradient freezing (VGF) method is proposed for producing high optical quality crystals [16], which has been used to grow $LiCaAlF_6$ [16], CdZnTe [17] crystals etc. In the present work, a highly sensitive α-Al_2O_3:C crystal was directly grown by VGF method using Al_2O_3 and graphite powder as the raw materials, which combines the advantages of the α-Al_2O_3:C crystal growth in the highly reducing atmosphere in the presence of graphite [1,2] and the α-Al_2O_3:C crystal directly grown by the using Al_2O_3 and graphite powder as raw materials [14,15]. The main OSL characteristics of the α-Al_2O_3:C detectors as grown were investigated.

2. Materials and methods

Al_2O_3 (99.999%) and graphite powder (99.999%) were used as raw materials. Graphite powder was mixed with Al_2O_3 powder in 5000 ppm proportion in an agate ball mill for 48 h. The mixture thus obtained was cold pressed in the form of a block (Dia.50 mm). The α-Al_2O_3:C crystal was grown in a tapered molybdenum crucible with an oriented seed. The VGF furnace was loaded for the growth process, evacuated to 10^{-3} Pa, heated to 1200 °C. The high pure Ar was input, and then the furnace heated to 2050 °C at rate of 60 °C/h and kept in the molten state for 30 min. After molten completely, crystallization was started by slow

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cooling (1 °C/h) with a high precision temperature program controller. α-Al_2O_3:C detectors with the size of 4 × 4 × 0.8 mm^3 (abbreviated as VGF α-Al_2O_3:C) were cut from the as-grown crystal. Single-crystal α-Al_2O_3:C detectors 5 mm in diameter with a thickness of 0.9 mm (TLD-500K) kindly provided by Urals Polytechnical Institute in Russia were employed for a comparison.

OSL measurements were performed at room temperature by a Risø TL/OSL-DA-15 reader, equipped with a bi-alkali EMI 9235QA photomultiplier tube and Hoya U-340 filters of 2.5 mm of thickness (transmission band centered at 340 nm and a full width at half maximum of ~80 nm) to block the blue light. The optical excitation unit consists of a blue light-emitting diode (LED) cluster with wavelength $\lambda = 470 \pm 30$ nm. Unless otherwise stated all measurements were carried out with a delay of at least 15 min after the irradiation and the readout time is 100 s, the data sampling interval is 0.1 s, and the intensity of the stimulation light is 90%. The OSL signal S was the area under the OSL curve, with a constant background subtracted:

$$S = \sum_{i=1}^{n} Ii - n\left(\frac{1}{m}\sum_{n+1}^{n+m} Ii\right) \tag{1}$$

In Eq. (1), I_i is the signal intensity, n is the number of data points of the OSL curve to calculate the OSL sum, and m are the number of data points at the end of the OSL decay curve used to estimate the background. Unless mentioned otherwise, the OSL signal in this study was calculated using Eq. (1) for a 100 s stimulation time, using the last 40 s as the background. Therefore, if the integration time was 0.1 s, n = 600 and m = 400.

Both kinds of α-Al_2O_3:C detectors were first annealed at 900 °C for 30 min [4,18]. Ten VGF α-Al_2O_3:C and ten TLD-500K were read out immediately after annealed at 900 °C for 30 min for calculating the detection threshold. In order to investigate the OSL signals, 20 VGF α-Al_2O_3:C detectors and 20 TLD-500K were irradiated with the same dose free in air of 1 mGy ^{60}Co gamma radiation at a dose rate of 20 mGy/h, 10 VGF α-Al_2O_3:C and 10 TLD-500K were first read out by OSL and then by TL, the others were readout by TL. In the TL case, half detectors were read out at a heating rate of 1 °C s^{-1}, and half at a heating rate of 10 °C s^{-1}. In order to investigated the repeatability, ten VGF α-Al_2O_3:C and ten TLD-500K were irradiated for 3 s to 205.8 mGy at a dose rate of 246.96 Gy/h using an integrated ^{90}Sr/^{90}Y beta source of the Risø TL/OSL-DA-15 reader and subjected to ten readout-annealing-exposure cycles. In this experiment, the detectors read out were annealed at 300 °C for 20 s by Risø TL/OSL-DA-15 reader. To test the dose response, 12 groups of the α-Al_2O_3:C detectors were exposed to ^{60}Co doses ranging from 1 μGy to 20 Gy at various dose rates, respectively. Each group includes 10 VGF α-Al_2O_3:C detectors and 10 TLD-500K detectors. For the multiple re-analyse, 10 VGF α-Al_2O_3:C and 10 TLD-500K α-Al_2O_3:C were irradiated with the same dose of 10 mGy ^{60}Co gamma radiation free in air at a dose rate of 100 mGy/h. The readout time is only 1 s, the data sampling interval is 0.01 s, and the intensity of the stimulation light is 1%.

3. Results

3.1. OSL signal

The typical continuous wave OSL (CW-OSL) decay curves of the α-Al_2O_3:C grown by the VGF method and the TLD-500K are shown in Fig. 1. The shape of the OSL decay curve of the VGF α-Al_2O_3:C is similar to that of the TLD-500K. The initial intensity (normalized to mass) for the VGF α-Al_2O_3:C is much higher than that for the TLD-500K. The mean OSL sensitivity per unit mass of the VGF α-Al_2O_3:C is 1.6 times higher than that of the TLD-500K. The relative standard deviations (SD) of the two kinds of α-Al_2O_3:C were < 5%. It was reported [19] that the total sum for the OSL of the TLD-500K is approximately two times higher than that for TL when using heating rates in the range 1–2 °C s^{-1}.

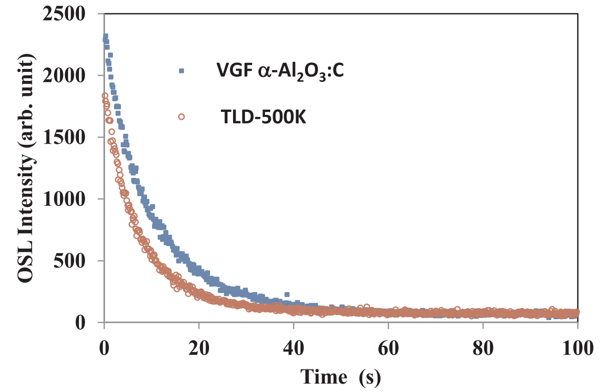

Fig. 1. The typical continuous wave OSL (CW-OSL) decay curves of the VGF α-Al_2O_3:C and the TLD-500K.

Because Al_2O_3:C is already one of the most sensitive TL detectors available, OSL from the TLD-500K α-Al_2O_3:C is a particularly sensitive means to measure radiation dose. It can be deduced that OSL of the VGF α-Al_2O_3:C has also a particularly high sensitivity to radiation.

The typical glow curves (normalized to mass) of the VGF α-Al_2O_3:C and the commercial TLD-500K before and after OSL at a heating rate of 1 °C s^{-1} are shown in Fig. 2. It can be seen that the OSL signal originates from the main TL peak, the responsible electron trap can be emptied basically by OSL readout. Similar results were found at a heating rate of 10 °C s^{-1}, however, the TL responses of both kinds of α-Al_2O_3:C with increasing heating rate reduced significantly. The TL intensity at a heating rate of 10 °C s^{-1} of the VGF α-Al_2O_3:C is about 35% of that at a heating rate of 1 °C s^{-1}. The TL intensity at a heating rate of 10 °C s^{-1} of the TLD-500K is about 30% of that at a heating rate of 1 °C s^{-1}. Reduction in the TL responses of both kinds of α-Al_2O_3:C with increasing heating rate is in agreement with the reported results [20,21]. The changes were ascribed to thermal quenching of the luminescence of the emission centers [22].

3.2. Detection threshold

The test results show that the OSL detection thresholds of the VGF α-Al_2O_3:C and the TLD-500K are 0.21 μGy and 0.51 μGy respectively. The detection threshold of the α-Al_2O_3:C developed by melting the alumina powder in graphite environment was 50 μGy [13]. It was reported [19] that doses lower than 0.5 μGy can easily be measured with good statistics using OSL of α-Al_2O_3:C. The detection thresholds of both kinds of α-Al_2O_3:C exceed the requirements of most personal and environmental dosimetry standards by a large margin. Both kinds of α-Al_2O_3:C can be used to measure the very low dose in environmental monitoring and personnel dosimetry.

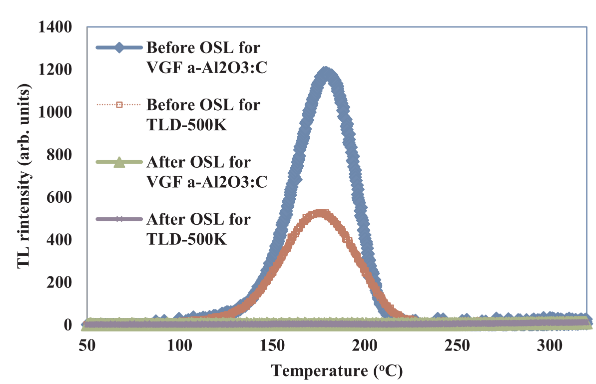

Fig. 2. The typical glow curves (normalized to mass) of the VGF α-Al_2O_3:C and the TLD-500K before and after OSL at a heating rate of 1 °C s^{-1}.