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Thermoluminescence and optically stimulated luminescence properties of Dy^{3+}-doped $CaO–Al_2O_3–B_2O_3$-based glasses

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ABSTRACT

We developed Dy^{3+}-doped $CaO–Al_2O_3–B_2O_3$ based glasses with Dy concentrations of 0.5, 1.0, and 2.0 mol % using a melt-quenching technique. The as-synthesized glasses were applicable as materials exhibiting thermoluminescence (TL) and optically stimulated luminescence (OSL). The optical and radiation response properties of the glasses were characterized. In the photoluminescence (PL) spectra, two emission bands due to the $^4F_{9/2} \rightarrow {}^6H_{15/2}$ and $^4F_{9/2} \rightarrow {}^6H_{13/2}$ transitions of Dy^{3+} were observed at 480 and 580 nm. In the OSL spectra, the emission band due to the $^4F_{9/2} \rightarrow {}^6H_{15/2}$ transition of Dy^{3+} was observed. Excellent TL and OSL responses were observed for dose ranges of 0.1–90 Gy. In addition, TL fading behavior was better than that of OSL in term of the long-time storage. These results indicate that the Dy^{3+}-doped $CaO–Al_2O_3–B_2O_3$-based glasses are applicable as TL materials.

1. Introduction

Thermoluminescence (TL) and optically stimulated luminescence (OSL) are both emissions that occur when a solid-state inorganic material is heated up and when it is exposed to light following exposure to ionizing radiation, respectively. In the TL and OSL processes, the exposed material absorbs ionizing radiation energy, producing electron–hole pairs. The electrons and holes are separately trapped at defects, creating a metastable state. When these materials are subjected to either thermal or optical stimulation, trapped electrons are released and entrapped in the hole traps, which causes an emission via electron–hole recombination.

Both TL and OSL materials are used to estimate absorbed radiation doses in various applications: radiation therapy, medical imaging, personal monitoring, etc. [1–3]. These materials are generally activated using transition metal or rare-earth metal ions as the luminescent centers. The required properties of these materials are high sensitivity, chemical stability, and linear response to a wide range of the absorbed radiation dose. Currently, Al_2O_3 (sapphire) is used as typical dosimetry materials [4,5]. However, these Al_2O_3 dosimeters have two weak points: weak TL or OSL intensity for a low dose and high production cost.

In this study, we developed dosimetry materials possessing the required properties with the advantage of low-cost fabrication to overcome the drawbacks of the existing dosimetry materials. For example, calcium sulfate doped with dysprosium ($CaSO_4:Dy^{3+}$) is a solid-state TL and OSL material that shows high sensitivity and good dose response [6–8]. Thus, we focused on Dy^{3+} as the dopant and developed Dy^{3+}-doped $CaO–Al_2O_3–B_2O_3$-based glass using the conventional melt-quenching technique. Use of glass matrix leads to low production cost because low-cost raw materials were used for preparing the materials. High TL and OSL intensities and a good response of Ce^{3+}-doped $CaO–Al_2O_3–B_2O_3$ glasses were previously reported by Fujimoto et al. [9]. Using glass as matrix in dosimetry materials is better from the viewpoints of fabrication cost and mold processes than those of single crystals and ceramics. In this study, we evaluated the photoluminescence (PL), TL, and OSL properties to examine the applicability of Dy^{3+}-doped $CaO–Al_2O_3–B_2O_3$-based glasses as TL and OSL materials.

2. Experimental procedure

$3.5CaO–1.5Al_2O_3–5B_2O_3:xDy_2O_3$ glasses were prepared by the previously reported melt-quenching method using an alumina crucible in air atmosphere [10]. Analytical reagent grade of $CaCO_3$ (99.99%, Rare Metallic Co., LTD), Al_2O_3 (99.99%, High Purity Chemicals), H_3BO_3 (99.99%, High Purity Chemicals), and Dy_2O_3 (99.9%, Rare Metallic Co., LTD) were used as starting materials. The Dy^{3+}

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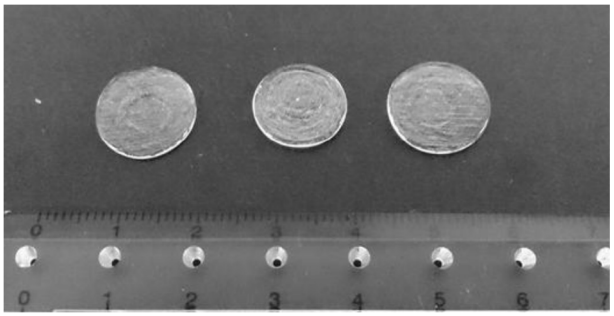

Fig. 1. The photograph of the fabricated Dy^{3+}-doped $CaO–Al_2O_3–B_2O_3$-based glass samples. From left to right, Dy^{3+} concentration is 0.5, 1.0, and 2.0 mol%.

concentrations used were 0.5, 1.0, and 2.0 mol% of $CaCO_3$. Each batch of the mixture in the alumina crucible weighed 2.5 g and was melted at 1100 °C in air for 30 min. Finally, the glass melt was quenched into a stainless mold, which was maintained at a temperature below the glass transition temperature (360 °C). Fig. 1 shows the photograph of the fabricated glass samples.

The PL excitation-emission map, the PL quantum yield, and PL decay curves under 340-nm excitation of the glasses before X-ray irradiation were measured at room temperature with the Quantaurus-QY (Hamamatsu) and Quantaurus-τ (Hamamatsu) systems. The monitoring wavelength of the PL decay curves was 580 nm. The TL glow curves were measured with an original setup following X-ray exposure of strengths 0.1, 1, 5, 10, 45, and 90 Gy. The TL photons from the sample were detected with a photomultiplier tube (PMT; H11890-210, Hamamatsu) while the irradiated sample was heated by a heater (SCR-SHQ-A, Sakaguchi). A thermal radiation from the sample was cut with a radiation cut filter. The samples were measured after annealing at 400 °C for 10 min to reduce the thermal stress, at a heating rate of 0.5 °C/s. In the same measurement system, the glow curves of Tm:$CaSO_4$ and $Li_2B_4O_7$:Cu, which are practically used as TL dosimeters, were measured to compare with the Dy^{3+}-doped glass. The OSL spectra were measured by a DeltaFlex 3000U-TMK2 (HORIBA), at room temperature, following annealing at 360 °C for 10 min at 0.1, 1, 5, 10, 45, and 90 Gy X-ray exposure of the samples. All measurements were performed in triplicate at all dosage levels to confirm spectral reproducibility. Emission from an LED at 625 nm was used for stimulation. In the same measurement system, the OSL spectra of C:Al_2O_3, which is an OSL material in practical use, were measured to compare Dy^{3+}-doped glass.

3. Results and discussion

Fig. 2 shows the PL excitation–emission map of the 2.0 mol% Dy^{3+}-doped glass. The PL emission bands were observed at 480 and 580 nm, assigned to the $^4F_{9/2} \rightarrow {}^6H_{15/2}$ and $^4F_{9/2} \rightarrow {}^6H_{13/2}$ transitions of Dy^{3+}, respectively [11]. Fig. 3 shows the PL decay curve of the 2.0 mol% Dy^{3+}-doped glass. The PL decay curves were fitted with a sum of two exponential decay functions. The decay-time constant was 590 μs, which is consistent with a previous report [12]. For the 1.0 mol% and 0.5 mol% samples, the decay-time constants were 669 and 681 μs, respectively, which shows that the decay-time constant increases with decreasing dopant

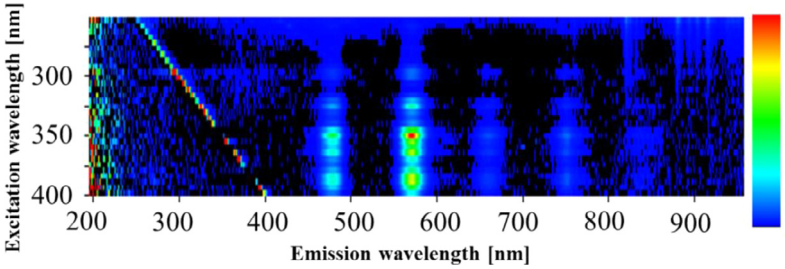

Fig. 2. The PL excitation-emission map of the 2.0 mol% Dy^{3+}-doped glass. The excitation source: Xe lamp.

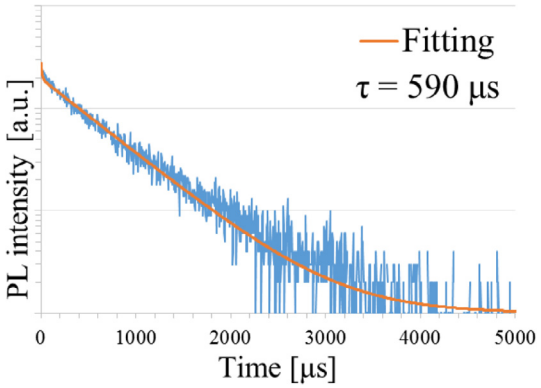

Fig. 3. The PL decay curve of the 2.0 mol% Dy^{3+}-doped glass. Excitation wavelength: 340 nm, monitored wavelength: 580 nm.

Table 1
The QY of the 0.5, 1.0, and 2.0 mol% Dy^{3+}-doped glasses.

Dy^{3+} concentration [mol%]	QY [%]
0.5	14.6
1.0	13.6
2.0	33.3

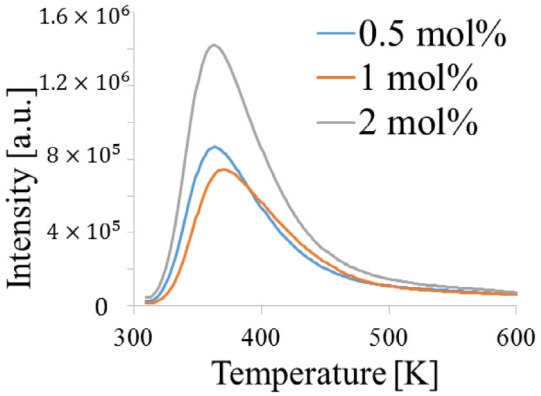

Fig. 4. The glow curves of the 0.5, 1.0, and 2.0 mol% Dy^{3+}-doped glasses irradiated with X-ray at 45 Gy. Heating rate: 1 K/s.