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Optical, scintillation and dosimeter properties of MgO:Tb translucent ceramics synthesized by the SPS method

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ABSTRACT

MgO translucent ceramics doped with different concentrations of Tb (0.01, 0.05, 0.1, 0.5%) were prepared by the Spark Plasma Sintering (SPS) method. Further, the optical, scintillation, dosimeter properties of were evaluated systematically. In the photoluminescence (PL) and scintillation spectra, sharp emission peaks due to the 4f-4f transitions of Tb^{3+} were observed. In the PL and scintillation decay curves, the decay time constants were a few ms which were on a typical order of the 4f-4f transitions of Tb^{3+}. The thermally-stimulated luminescence (TSL) glow curves exhibited glow peaks around 80, 160 °C after X ray irradiation of 10 mGy. The intensity of TSL peak at 160 °C exhibited a linear response against X-ray dose over a dose range of 0.1−10 mGy. The optically-stimulated luminescence (OSL) under 590 nm stimulation exhibited strong emissions due to Tb^{3+} around 385−550 nm after X-ray irradiation. As in TSL, the intensity of OSL peak showed a linear response to X-ray dose, and the dynamic range confirmed was 0.1−1000 mGy.

1. Introduction

Dosimeters have been used for decades to determine radiation dose, energy and spatial distribution. Dosimeters are required in various applications of radiation detection, such as personnel monitoring [1–3], environmental dosimetry [4] and retrospective accidental dosimetry [5]. Dosimeters store and accumulate incident radiation energy in a form of carrier trapping at localized centers. The absorbed energy can be read out as a form of photon emissions by external stimulation, which de-capture the trapped charges followed by recombination. When the stimulation is light, the resultant light emission is so-called optically-stimulated luminescence (OSL), whereas emitted light stimulated by heat is so-called thermally-stimulated luminescence (TSL). The use of TSL for radiation dosimetry has been established for many decades. Up to now, many kinds of TSL materials such as $Li_2B_4O_7$:Cu, Mg_2SiO_4:Tb, $CaSO_4$:Dy and $CaSO_4$:Tm, and LiF:Mg, Cu, P have been developed [6]. Recently, OSL has been developed for one of alternative methods for TSL. OSL have several advantages over TSL for radiation dosimetry. The nature of optical readout process does not involve problems of blackbody radiation and thermal quenching which are unavoidable during TSL readout. In addition, a precise delivery of stimulation light enables multiple readings with the same precision and with minimal depletion of traps. The use of OSL for radiation dosimetry was first suggested by Antonov-Romanovskii [7]. After that, many kinds of materials such as Al_2O_3:C, MgO:Tb, $NaMgF_3$:Eu and Mg_2SiO_4:Tb have been developed for OSL dosimetry [6].

Recently, fabrication of optically transparent ceramics has been developed with the advancement of ceramic fabrication techniques especially in the laser field. In general, optically transparent ceramics are fabricated by hot-isostatic pressing or vacuum sintering at very high temperatures using ultrapure powders. Owing to the high sinterability of nanocrystalline powers with the rapid densification rates of spark plasma sintering (SPS), the SPS technique has been widely promoted as a novel method for the fabrication of transparent polycrystalline oxides [8]. In general, the SPS is performed in a highly reductive atmosphere; therefore, oxygen vacancies are effectively created, and the dosimeter properties are expected to be enhanced. In our previous studies, many kinds of transparent ceramics were fabricated by the SPS method in order to enhance dosimeter properties of the ceramics [9–16].

In this study, we focused on MgO ceramic doped with Tb as a potential dosimeter material. MgO ceramics have excellent thermal and mechanical properties with a high melting point (2800 °C). Up to now, MgO ceramics doped with several different transition and rare earth elements have been studied for dosimeters [17–21].

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Previously, it has been reported that MgO:Tb ceramics show notable OSL properties with a dynamic range of 0.1–10,000 mGy [6,22]. However, dosimeter properties of MgO:Tb translucent ceramics fabricated by the SPS method have not been investigated yet. In this study, MgO translucent ceramics doped with Tb ion (0.005, 0.01, 0.05 and 0.1%) were fabricated by the SPS method. After the synthesis, in-line transmittance spectra of non-doped MgO and obtained MgO:Tb ceramics were compared. Further, the photoluminescence (PL) and scintillation spectra and their decay time profiles were characterized. As dosimeter applications, TSL and OSL were also demonstrated.

2. Experimental

MgO:Tb translucent ceramics were fabricated by the SPS method using Sinter Land LabX-100. Stoichiometric quantities of MgO (99.99%) and Tb_4O_7 (99.99%) were mixed by using a mortar and pestle for 30 min. The mixture was next loaded in a graphite die and held between two graphite punches. The temperature was increased from 600 °C to 1500 °C with a heating rate of 17 °C/min and held for 1 h while applying a pressure of 80 MPa. After these synthesis procedures, the surfaces of the obtained ceramic sample were mechanically polished.

Optical in-line transmittance spectra were measured by using JASCO V670 spectrophotometer over a spectral range of 200–2700 nm with 1 nm intervals. PL spectra were measured by using Quantaurus-QY (Hamamatsu Photonics). PL decay time profiles were evaluated by using Quantaurus-τ (Hamamatsu Photonics). In these measurements, the excitation wavelength was 340 nm, and the monitoring wavelength in PL decay was 385 nm. X-ray induced scintillation spectra were measured by using our original setup [23]. The excitation source was an X-ray generator equipped with a tungsten anode target (XRB80P&N200 × 4550, Spellman), and it was operated with a tube voltage of 40 kV and current of 5.2 mA. The scintillation photons from the sample were collected and guided to the spectrometer (Andor DU-420-BU2 CCD and Shamrock 163 monochromator) through a 2.0 m optical fiber. X-ray induced scintillation decay profiles were evaluated by using an afterglow characterization system equipped with a pulse X-ray source [24]. The X-ray source is equipped with a tungsten anode target with a Be window, and the applied voltage to the X-ray source was 30 kV. Therefore, the mean X-ray energy was around 20 keV. The time resolution of this system was a few ns, and the equipped photomultiplier tube had an effective sensitivity across 160–650 nm. TSL glow curves were measured by using a TSL reader (TL2000, Nanogray Inc.) after X-ray irradiation [25]. The heating rate was 1 °C/s, and the measurement temperature range was from 50 to 500 °C. OSL spectra were measured under 590 nm stimulation by using Quantaurus-τ (C11367-25, Hamamatsu Photonics). Further, TSL spectrum was measured using an Andor CCD-based spectrometer while the sample was heated by an electric heater at a constant temperature. OSL spectrum was measured under 630 nm stimulation by using Quantaurus-τ (Hamamatsu Photonics).

3. Results and discussion

3.1. Sample

Fig. 1 shows a photograph of non-doped MgO and MgO:Tb ceramic samples with different concentrations of Tb under room light. The thickness of the samples was about 1.1 mm after polishing. On one hand, the stripe pattern on the back of non-doped MgO, MgO:0.01%Tb and MgO:0.05%Tb can be seen through. On the other hand, MgO:0.1%Tb and MgO:0.5%Tb were not translucent.

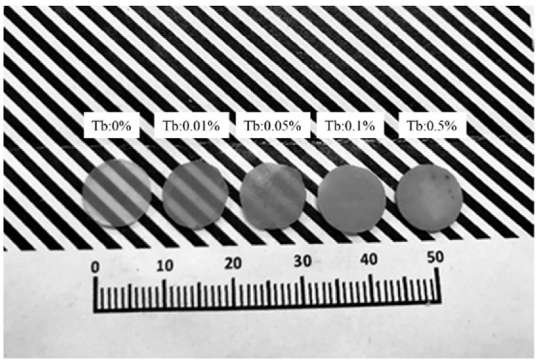

Fig. 1. Photograph of the un-doped MgO and MgO:Tb ceramics.

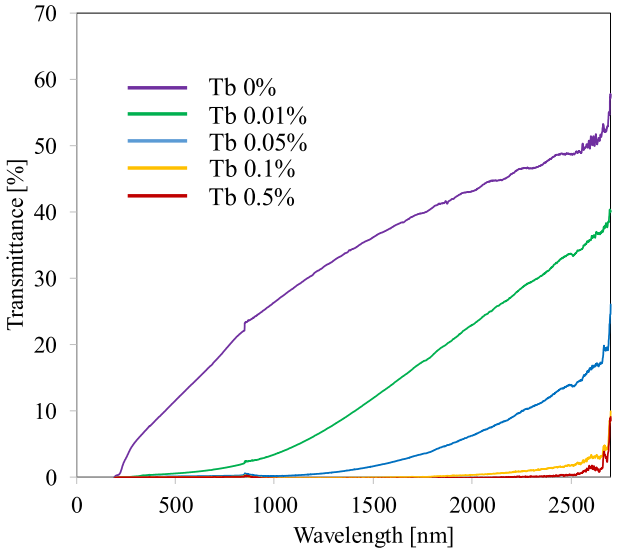

Fig. 2. Transmittance spectra of un-doped MgO and MgO:Tb ceramics.

Fig. 2 shows in-line transmittance spectra of the non-doped MgO and MgO:Tb ceramic samples. The transmittance decreased with increasing the concentration of Tb over the wavelengths of 200–2700 nm. The transmittance was close to zero at and shorter wavelengths than 200 nm for all the samples. The latter wavelength was longer than the wavelength corresponding to the band gap energy of MgO (~7.8 eV = 159 nm) [26].

3.2. Photoluminescence properties

Fig. 3 shows PL spectra of non-doped MgO and MgO:Tb ceramics under excitation wavelength of 280 nm. Several emission peaks were observed over the wavelengths of 350–650 nm for MgO:Tb ceramics. The origins of these peaks were attributed to the 4f-4f transitions of Tb^{3+} based on the past studies. Each transition is indicated in Fig. 3 [22,27–29]. Among the ceramic samples prepared, the intensity of MgO:Tb 0.01% was the highest. The PL intensity decreased with increasing the concentration of Tb, and the reason was considered to be due to the concentration quenching. On the other hand, the non-doped MgO did not show measurable signal.

Fig. 4 shows PL decay time profiles of MgO:Tb ceramics. The excitation wavelength was 340 nm while the emission wavelength monitored during the measurements was 385 nm ($^5D_3 \rightarrow {}^7F_6$). Each decay curve was approximated by a first-order exponential decay