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Dosimeter properties of MgO transparent ceramic doped with C

Takumi Kato*, Go Okada, Takayuki Yanagida

Nara Institute of Science and Technology (NAIST), 8916-5, Takayama-cho, Ikoma-shi, Nara, 630-0192, Japan

HIGHLIGHTS

- We have synthesized MgO translucent ceramics doped with C ions by an SPS technique.
- We have characterized the optical, scintillation and dosimeter properties.
- The peak at ~250 °C of 0.1% C-doped sample showed the most high-performance throughout this work.

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ABSTRACT

We have investigated the photoluminescence (PL), scintillation and thermally-stimulated luminescence (TSL) dosimeter properties of MgO ceramic doped with C ions (0.001, 0.01 and 0.1%). The samples were synthesized by a Spark Plasma Sintering (SPS) technique. The PL emission peaks appeared around 400 and 750 nm in all the samples. The PL decay time constants at 400 nm were ~10 and ~100 ns which were on the typical order of F+ center in the undoped MgO. The scintillation emission peaks were detected at 330, 400 and 750 nm under X-ray irradiation. The TSL glow curves showed the ~250 °C peak in 0.1% C-doped sample. The TSL response was confirmed to be linear to the irradiation dose over the dose range from 0.1 to 1000 mGy. As a result, the sensitivity of MgO was improved by C-doping.

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1. Introduction

In ionizing radiation detectors, luminescence materials have been used to visualize invisible radiations by visible photons. Among such luminescence materials, scintillators which convert a single high energy ionizing radiation to thousands of ultraviolet–visible photos immediately (Yanagida, 2013) is the most famous type of the solid state radiation detector and has a wide range of applications such as medical (Yanagida et al., 2010), security (Totsuka et al., 2011). On the other hand, another luminescence type detector, dosimeter storages the absorbed energy for several weeks and release by optical (optically stimulated luminescence, OSL) or thermal (thermally stimulated luminescence, TSL) stimulation. The main application of OSL or TSL device is a personal dosimetry (Mayhugh et al., 1970; McKeever, 2011). Required dosimeter properties depend on applications, but typically suitable sensitivity, dose linearity, energy response and low fading are considered. In addition, if the aim is to measure a radiation dose absorbed in human body, it is desirable that the effective atomic number (Z_{eff}) of the dosimeter material, in the view point of bioequivalence, is close to that of the soft tissue ($Z_{eff} = 7.13$). With such a tissue equivalent detector, no mathematical calibration for energy dependence is required ideally. Therefore, for such dosimeter applications especially in the human radiation monitor, it is preferred for the detector materials with light elements.

Magnesium oxide (MgO) is a wide band-gap insulator ($E_g = 7.8$ eV) with a rock-salt crystal structure (fcc) under ambient pressure, and the Mg ions occupy the octahedral sites within the anion close-packed structure (Klein and Hurlbut, 1999; Roessler and Walker, 1967). The luminescence properties of undoped MgO have been studied in many material forms including powders, films, bulk single crystals and ceramics (Chen et al., 1969; Jeffries et al., 1982; Kappers et al., 1970; Martinez-Boubeta et al., 2011; Rosenblatt et al., 1989; Soma et al., 2015; Summers et al., 1983; Uchino et al., 2009; Uenaka and Uchino, 2011; Yanagisawa and Huzimura, 1984). In MgO crystals, Rosenblatt et al. have demonstrated that there are two primary oxygen vacancies, namely F and F^+ color centers, which capture two and one electrons, respectively.

* Corresponding author.
E-mail address: kato.takumi.ki5@ms.nasit.jp (T. Kato).

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Although absorption bands of the F and F^+ color centers are almost at the same wavelength of ~250 nm, they show different photoluminescence (PL) bands with the peaks at ~500 nm and ~400 nm, respectively (Rosenblatt et al., 1989).

From the view point of MgO for radiation detector uses, in the 1970s, undoped MgO was found to show dosimeter properties. When it was irradiated by X-ray, γ-ray and UV, two TSL glow peaks were found around 90–100 and 140 °C (Nanto et al., 1975; Sibley et al., 1969; Takeuchi et al., 1975). Furthermore, in the later decades, MgO have been intensively studied for dosimeter applications due to the bioequivalency (Dolgov et al., 2002; Las and Stoebe, 1984; Sathyamoorthy and Luthra, 1978; Soliman, 2009; Ziniker et al., 1973). Recently, with advancement of ceramic fabrication techniques, MgO transparent ceramic was developed (Chaim et al., 2004, 2010; Chen et al., 2008; Fang et al., 2004; Wakahara et al., 2013). It shows phosphorescence (or so called long afterglow or persistent luminescence) emission at the wavelength of 390 nm due to the F^+ center acting as a luminescent center (Wakahara et al., 2013). The phosphorescence, in other words, is a form of TSL around room temperature; therefore, the MgO transparent ceramic is potentially applicable for TSL dosimeter if the TSL peak temperature can be somehow increased in order to store the dose signal for a long term without fading. In order to develop efficient TSL device, we have studied transparent ceramic MgO materials (Kato et al., 2016a, 2016b).

In this study, we have synthesized a MgO transparent ceramic doped with C ions (0.001, 0.01 and 0.1%) by spark plasma sintering (SPS) and studied the dosimeter properties against X-rays. The C-doping is inspired by C-doped Al_2O_3 crystalline powder used for a personal dosimeter (McKeever, 2011). Further, we have characterized the optical and scintillation properties. Recently, it was pointed out that scintillation and dosimeter are complementarily related in some material systems (Yanagida et al., 2014b, 2016) so investigations of both the dosimeter and scintillation properties are important to understand the luminescence phenomena induced by ionizing radiations. In general, SPS is performed in a highly reductive atmosphere; therefore, oxygen vacancies are effectively created, and the dosimeter properties expected to be enhanced.

2. Experiment

MgO transparent ceramic samples were synthesized by an SPS method using Sinter Land LabX-100. Here, a reagent grade of MgO (99.99%) powder and a carbon powder were homogeneously mixed. A carbon powder was made from shattered graphite die by using a ball mill (Premium line P-7, Fritsch). The total mass of the mixture was 0.5 g. The mixture was then loaded in a graphite die and sandwiched by two graphite punches. The sintering temperature was increased from 600 °C to 1500 °C at the rate of 17 °C/min and held for 60 min while applying the pressure of 80 MPa. After the synthesis, the wide surfaces of the ceramic sample were polished. In the course of study, the following measurements were carried out for all the prepared samples.

The in-line transmittance was evaluated by using a JASCO V670 spectrometer in the spectral range from 190 to 2700 nm with 1 nm interval. The PL emission spectrum was measured with a 250 nm excitation using FP-8600 (C002361454, JASCO) and a short-cut filter (LU0300, Asahi Spectra). The PL decay lifetime monitoring at 400 nm was measured with 280 nm excitation using Hamamatsu Quantaurus-τ (C11367-04, Hamamatsu). The X-ray induced scintillation spectrum was measured using our lab-constructed setup. The sample was excited using an X-ray generator in which the applied tube voltage and current were 40 kV and 5.2 mA, respectively. The scintillation emission was guided to Ocean Photonics CCD-based spectrometer (QEPro). The details of the setup was described previously (Yanagida et al., 2013b). Further, the scintillation lifetime by X-ray irradiation was measured using an afterglow characterization system equipped with a pulsed X-ray tube (Yanagida et al., 2014a). The system is commercially available from Hamamatsu as a custom-ordered instrument. The applied voltage to the pulsed X-ray source was 30 kV, and the system offers the timing resolution of ~1 ns?

In order to investigate the properties of MgO ceramic as a TSL dosimeter device, we have measured a TSL glow curve using a Nanogray TL-2000 (Yanagida et al., 2013a) after X-ray irradiations with various doses from 0.1 mGy to 1000 mGy. The heating rate used for all the TSL measurements was fixed to 1 °C/s, and the sample was heated from 50 to 490 °C to measure the glow curve. Further, TSL spectrum was measured using an Andor CCD-based spectrometer as mentioned above while the sample was heated by an electric heater (SCR-SHQ-A, Sakaguchi E.H Voc) at a constant temperature.

3. Results and discussion

3.1. Sample

The synthesized MgO doped with C (0.001, 0.01 and 0.1%) samples are illustrated in Fig. 1. The top photograph shows the samples under the room light while the bottom one shows those under UV (254 nm) light. These samples are visually transparent except for 0.1% C-doped sample. It looks like that the 0.001% C-doped sample emits yellowish green color and the 0.01% C-doped sample emits blue color.

3.2. Optical properties

Fig. 2 shows the in-line transmittance spectra of MgO samples. For the 0.001 and 0.01% C-doped samples, the transmittance is close to zero at 220 nm. These wavelengths are longer than the wavelength of the bandgap of MgO (~7.8 eV = 159 nm) (Roessler and Walker, 1967). No particular absorption bands were observed, and the transmittance became worse by dense C-doping. In addition, transmittance grows with an increasing wavelength from 0 up to 50% in the spectral range 450–2500 nm, and this is a typical tendency of Mie scattering. When the doping concentration increased, the number of scatterers also increased.

Fig. 3 represents the PL spectra of the MgO doped with C (0.001, 0.01 and 0.1%) samples under 250 nm excitation. The emission peaks appeared around 400 and 750 nm, and the origins of these peaks were assigned in previous reports (Mckeever et al., 1995; Rosenblatt et al., 1989; Takeuchi et al., 1975; Uenaka and Uchino, 2011); therefore, we think that 400 nm peak may be due to F^+ center and 600–900 nm peak may be caused by Cr^{3+} ions and Fe^{3+}

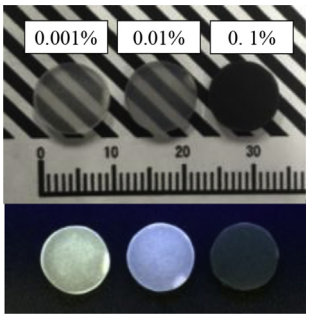

Fig. 1. MgO ceramic doped with C (0.001, 0.01 and 0.1%) samples under room light (top) and UV (254 nm) light (bottom).

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