Comparative Study on Optically Stimulated Luminescence Properties of BaCl$_2$:Ce and BaCl$_2$:Eu Transparent Ceramics

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0.1% BaCl$_2$:Ce and BaCl$_2$:Eu transparent ceramics were synthesized by the spark plasma sintering method, and the optical and dosimetric properties of BaCl$_2$:Ce and BaCl$_2$:Eu were compared. BaCl$_2$:Ce and BaCl$_2$:Eu transparent ceramics showed photoluminescence (PL) and optically stimulated luminescence (OSL) peaks at around 380 nm due to the 5d–4f transition of Ce^{3+} and Eu^{2+}. The PL quantum yields of BaCl$_2$:Ce and BaCl$_2$:Eu transparent ceramics were 3.8 and 25.6%, respectively. The PL decay times of BaCl$_2$:Ce and BaCl$_2$:Eu were 21 and 400 ns, respectively. In OSL dose responses, the BaCl$_2$:Eu sample showed better OSL intensity than BaCl$_2$:Ce, and the OSL signal was detectable from 0.01 mGy. In terms of detection sensitivity, this value was comparable to those of some commercially available materials.

1. Introduction

Phosphor-based radiation detectors are classified into two categories, namely, scintillation detectors[1–3] and dosimeters with storage phosphors.[4–6] They are widely applied to many fields such as medicine,[7–9] environmental monitoring,[10–12] security,[13–15] and astrophysics.[16–18] Dosimeters with storage phosphors have the function of storing the energy of ionizing radiation in a form of trapped electrons and holes at localized sites. The trapped carriers can be released by external stimulation, which eventually causes the recombination of electrons and holes with the emission of photons. If the stimulation is optical, the process is called optically stimulated luminescence (OSL).[19–25] OSL materials are mainly used in radiation measurements such as personal radiation dosimeters[26–28] and imaging plates (IPs).[29–33] In particular, the required properties of OSL materials for IPs include high intensity, wide dynamic range, low fading, and high effective atomic number.

Conventionally, storage phosphors such as BaFBr:Eu and CsBr:Eu have been developed and used for commercially available IPs.[34–37] These phosphors show a high OSL intensity and a wide dynamic range. Moreover, the OSL wavelengths of BaFBr:Eu and CsBr:Eu are 390 and 440

nm, respectively, which can be effectively detected by conventional photodetectors.[38,39] Recently, our group has developed transparent ceramics of BaFBr:Eu and CsBr:Eu and has evaluated their OSL properties.[40,41] Transparent ceramics have attracted attention as new optical materials because of their superior heat resistance and mechanical strength, as well as their advantages of shape flexibility and mass production compared with single crystals.[42–47]

In this study, we focused on $BaCl_2$ as a host material of transparent ceramics. $BaCl_2$ has a high effective atomic number (46.8) as do BaFBr (47.8) and CsBr (51.9). In addition to this characteristic, $BaCl_2$ has almost no hygroscopicity, which is also an advantage in applying $BaCl_2$ to OSL materials. Up to now, single crystals of $BaCl_2$:Ce[48] and $BaCl_2$:Eu[49] have been reported to have OSL properties; however, the potential of $BaCl_2$:Ce and $BaCl_2$:Eu transparent ceramics for use in OSL dosimeters remains unclear. Therefore, the OSL properties of $BaCl_2$:Ce and $BaCl_2$:Eu transparent ceramics were investigated and compared. In addition, their photoluminescence (PL) properties were also evaluated to specify the difference in OSL properties between $BaCl_2$:Ce and $BaCl_2$:Eu.

2.   Experimental Methods

$BaCl_2$:Ce and $BaCl_2$:Eu transparent ceramics were synthesized in a vacuum by spark plasma sintering (SPS) (LABOX-300, Sinterland). Raw powders of $BaCl_2$ (4N, Rare Metals) and $CeCl_3 \cdot xH_2O$ (3N, Kojundo Chemical) or $EuCl_3 \cdot 6H_2O$ (3N, Furuuchi Chemical) were homogeneously mixed in a molar ratio of 99.9:0.1 respectively. The mixture of $BaCl_2$ and $CeCl_3 \cdot xH_2O$ was dried at 270 °C for 4 h using a small electric furnace under vacuum. On the other hand, the mixture of $BaCl_2$ and $EuCl_3 \cdot 6H_2O$ was dried at 400 °C for 2 h using the same electric furnace under vacuum. Subsequently, the mixtures were enclosed with a cylindrical tungsten carbide die and a pair of tungsten carbide punches. The sintering temperature of $BaCl_2$:Ce was raised from 20 to 300 °C over 20 min, while the pressure was raised from 0 to 300 MPa for 20 min. In contrast, $BaCl_2$:Eu was sintered at a temperature of 350 °C. These temperatures and pressure (350 or 300 °C and 300 MPa) were held for 10 min, and the sintering was completed. The surfaces of the fabricated samples were polished with sandpaper.

Diffuse transmittance spectra, PL quantum yields (*QY*s), and PL decay curves were measured using a spectrophotometer (SolidSpec-3700, Shimadzu), Quantaurus-QY (C11347, Hamamatsu Photonics), and Quantaurus-τ (C11367, Hamamatsu Photonics), respectively. OSL spectra were measured using a spectrofluorometer (FP-8600, JASCO). The sample was irradiated with X-rays with a dose in the range from 0.01 to 100 mGy, and the dose response curves were obtained. An X-ray generator (XRBOP&N200X4550, Spellman) was used for X-ray irradiation.

3.   Results and Discussion

Figure 1(a) shows the appearance of $BaCl_2$:Ce and $BaCl_2$:Eu transparent ceramics. The samples appeared transparent and showed blue luminescence under UV light. Figure 1(b) indicates the diffuse transmittance spectra of $BaCl_2$:Ce and $BaCl_2$:Eu transparent ceramics. An absorption band was detected at 250–400 nm for both samples. This absorption band is due to
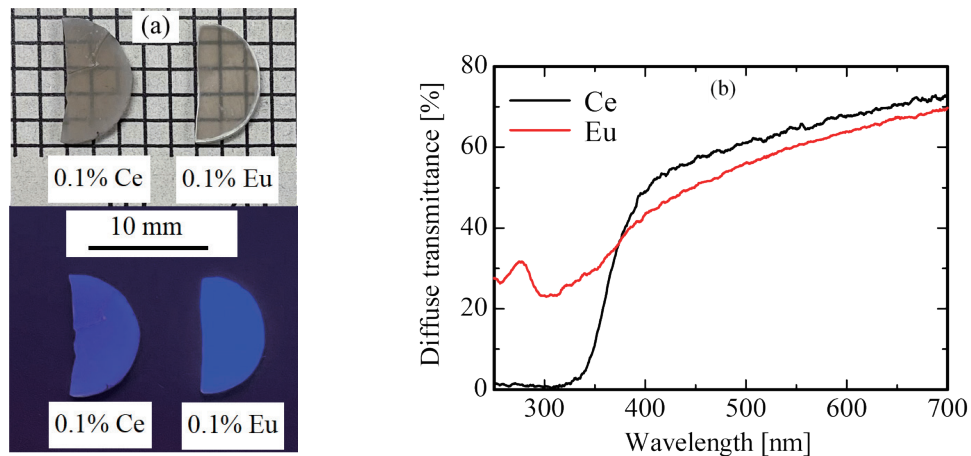
Fig. 1.   (Color online) (a) Synthesized $BaCl_2$:Ce and $BaCl_2$:Eu transparent ceramics under room light and UV (302 nm) light. (b) Diffuse transmittance spectra of $BaCl_2$:Ce and $BaCl_2$:Eu transparent ceramics.

the 4f–5d transitions of Ce^{3+}[50] and Eu^{2+}.[51,52] The diffuse transmittance of BaFBr:Eu and CsBr:Eu transparent ceramics fabricated by our group is ~50% in the visible range,[40,41] and the samples fabricated in this study showed comparable transmittance.

Figure 2 shows the PL excitation and emission maps of $BaCl_2$:Ce and $BaCl_2$:Eu transparent ceramics. An emission peak was observed at 380 nm under 330 nm excitation in $BaCl_2$:Ce and at 400 nm under 325 nm excitation in $BaCl_2$:Eu. The excitation and emission wavelengths are well matched to previously reported ones of $BaCl_2$:Ce and $BaCl_2$:Eu;[48,51] thus, the luminescence origin is the 5d–4f transitions of Ce^{3+} and Eu^{2+}. The PL *QY*s of $BaCl_2$:Ce and $BaCl_2$:Eu transparent ceramics were 3.8 and 25.6%, respectively, and the PL *QY* of $BaCl_2$:Eu was higher than that of $BaCl_2$:Ce. The inset of Fig. 2 shows their PL decay curves. The emission wavelengths were monitored at 380 nm ($BaCl_2$:Ce) and 400 nm ($BaCl_2$:Eu) under 340 nm excitation, which were chosen from data of Fig. 2. The decay curves of both samples were approximated by a single exponential decay function. The PL decay times obtained by fitting analysis were 21 and 400 ns for $BaCl_2$:Ce and $BaCl_2$:Eu, respectively. Compared with the previous reports,[51,53,54] these decay time constants were reasonable values for 5d–4f transitions of Ce^{3+} and Eu^{2+} in $BaCl_2$.

Figure 3 shows the OSL spectra of $BaCl_2$:Ce and $BaCl_2$:Eu transparent ceramics. Under stimulation of 520 nm, $BaCl_2$:Ce and $BaCl_2$:Eu showed an OSL emission peak at around 380 nm. The spectral features are consistent with those of PL; thus, the OSL peak is due to the 5d–4f transition of Ce^{3+} and Eu^{2+}. The stimulation spectra had a broad tail feature, and the intensity increased with a decrease in stimulation wavelength. No stimulation peak was observed since the peak position seems to be close to the emission peak (or maybe at even shorter wavelengths than that of the emission). According to previous reports,[48,55–57] the trapping centers in this case are F centers.

Figure 4 shows the OSL decay curves of $BaCl_2$:Ce and $BaCl_2$:Eu transparent ceramics during a constant stimulation at 520 nm after X-ray irradiation of 10 mGy. The monitored wavelengths
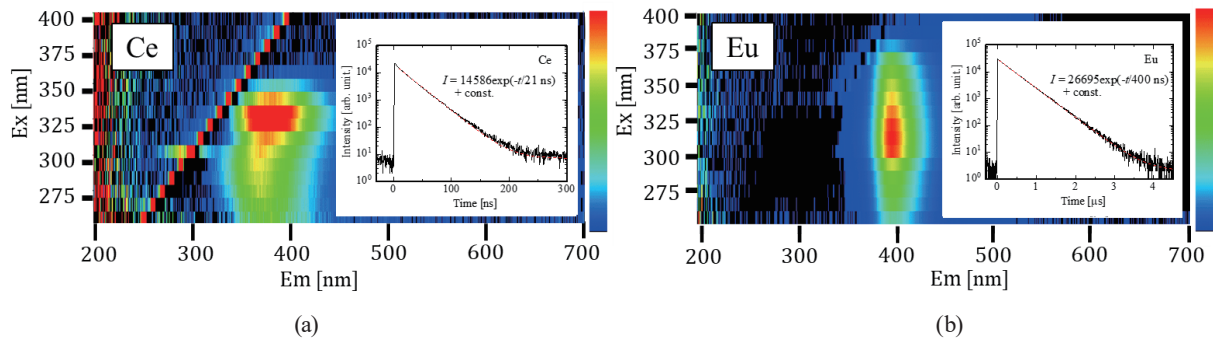
Fig. 2. (Color online) PL excitation and emission maps of (a) BaCl$_2$:Ce and (b) BaCl$_2$:Eu transparent ceramics. The inset shows PL decay curves of (a) BaCl$_2$:Ce and (b) BaCl$_2$:Eu transparent ceramics, where the black solid and red dashed lines show the decay curves and fitted curves, respectively.

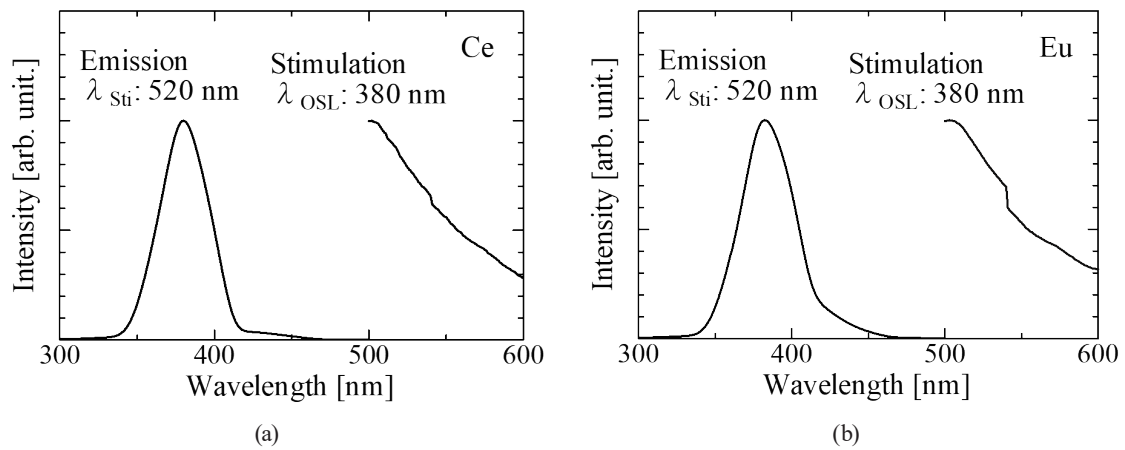Fig. 3. OSL spectra of (a) BaCl$_2$:Ce and (b) BaCl$_2$:Eu transparent ceramics after 1000 mGy X-ray exposure.
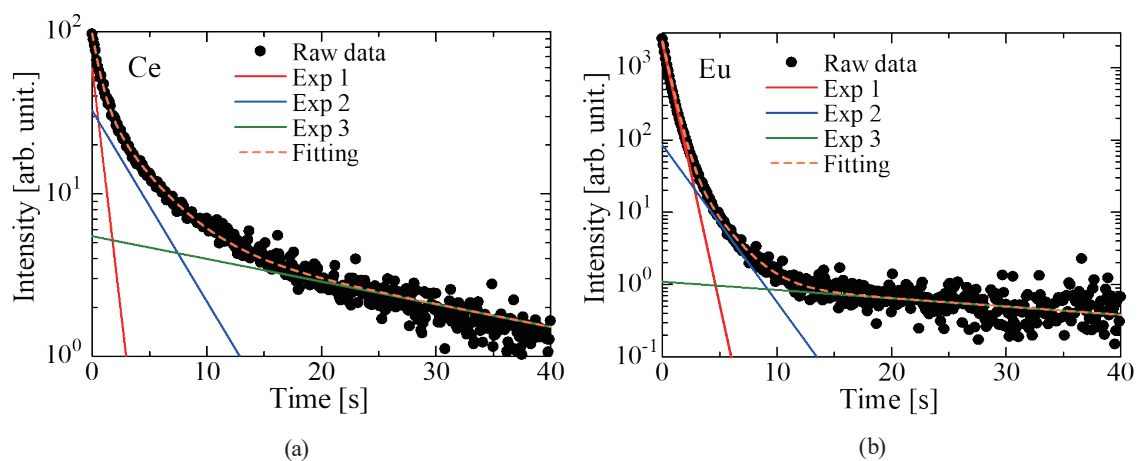

Fig. 4. (Color online) OSL decay curves of (a) BaCl$_2$:Ce and (b) BaCl$_2$:Eu transparent ceramics. Solid and dashed lines are raw data and fitted curves, respectively.
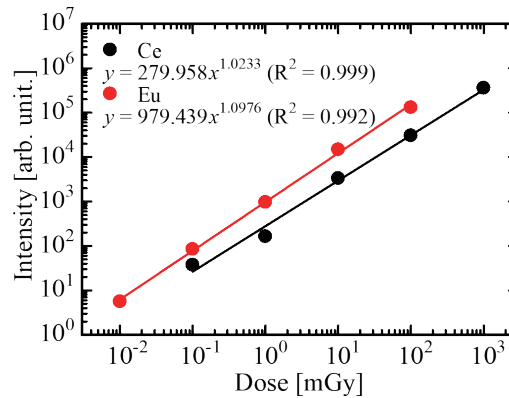
Fig. 5. (Color online) OSL dose response functions of $BaCl_2$:Ce and $BaCl_2$:Eu transparent ceramics.

of $BaCl_2$:Ce and $BaCl_2$:Eu were 380 nm, whereas the stimulation wavelengths were 520 nm. The emission intensities of both samples decreased with continuous stimulation; therefore, the emission is entirely due to the OSL phenomenon. The decay curves were best approximated by a sum of three exponential decay functions. Thus, there must be three different paths for charges photoionized and transferred to recombination centers. Comparison between $BaCl_2$:Ce and $BaCl_2$:Eu shows that their OSL decay time constants were almost the same. Since the number of components and decay times were close regardless of dopant, the host $BaCl_2$ was thought to play a major role in the OSL phenomenon.

Figure 5 shows the OSL dose response of $BaCl_2$:Ce and $BaCl_2$:Eu transparent ceramics. Here, the OSL intensity represents an integrated value of the OSL curves (shown in Fig. 4) at each X-ray dose. The irradiation dose range was from 0.01 to 1000 mGy. The dynamic ranges of $BaCl_2$:Ce and $BaCl_2$:Eu were 0.1–1000 and 0.01–100 mGy, respectively. Note that the upper detection limit of $BaCl_2$:Eu reached the detection limit of the instrument, and the OSL signal of $BaCl_2$:Eu could not be read out correctly at 1000 mGy. When the lower detection limits of $BaCl_2$:Ce and $BaCl_2$:Eu were compared, $BaCl_2$:Eu showed higher sensitivity than $BaCl_2$:Ce. Furthermore, the $BaCl_2$:Eu sample could detect a signal of lower dose than could the commercially available IPs (Fujifilm, HR-BD).[40]

4. Conclusions

The optical and OSL properties of 0.1 mol% $BaCl_2$:Ce and $BaCl_2$:Eu transparent ceramics synthesized by SPS were investigated. The PL excitation and emission maps showed the emission peaks at 380 ($BaCl_2$:Ce) and 400 ($BaCl_2$:Eu) nm. The PL decay time constants of $BaCl_2$:Ce and $BaCl_2$:Eu were 21 and 400 ns, respectively. Furthermore, the OSL spectra of the samples correspond to the results of the PL spectra. The PL *QY* of $BaCl_2$:Eu was higher than that of $BaCl_2$:Ce, which caused the superior lower detection limit of $BaCl_2$:Eu.

Acknowledgments

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