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Scintillation Properties of Er-doped $BaO–B_2O_3$ Glasses

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$BaO–B_2O_3$ glasses doped with different Er concentrations were synthesized by the melt-quenching technique. The Er concentrations were 1, 5, 10, and 15%. All the synthesized samples were transparent and homogeneous and exhibited near-infrared scintillation. The 5% Er-doped sample showed the highest scintillation intensity among all the samples, and the lower detection limit of the X-ray dose rate with our setup was 0.06 Gy/h.

1. Introduction

Scintillators convert ionizing radiation into photons of lower energy.[1–3] Scintillators are used for radiation measurement combined with photodetectors and applied to a variety of fields.[4] Previously, scintillators that emit photons of the ultraviolet–visible (UV–VIS) range have been highly valued as widely used photodetectors have a high sensitivity in that range. Recently, scintillators that emit photons of the near-infrared (NIR) range have started to attract attention as photodetectors with high sensitivity in the NIR range have come into practical use.[5] As applications of NIR-emitting scintillators, monitoring high radiation fields typified by nuclear plants is proposed.[6] In the proposed method, the photons emitted by a scintillator are delivered to a photodetector through an optical fiber so that the photodetector and peripheral equipment can be placed away from the high radiation field to avoid radiation damage. NIR photons are delivered through optical fibers with less loss than UV–VIS photons.[7,8] In addition, NIR photons are easily distinguishable from Cherenkov radiation, which is UV–VIS light generated in high radiation fields;[9] therefore, NIR scintillators are more suitable for monitoring high radiation fields. While scintillators of a variety of material forms, such as single crystals,[10–14] glasses,[15–20] and translucent ceramics,[21–23] have been researched and developed, most of the currently used scintillation materials are single crystals. Glass materials have commercial advantages over single crystals; it is easier to obtain large pieces, reform them into desirable shapes, and obtain pieces of the same quality. Hence, high-performance glass scintillators are highly demanded.

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In this study, we investigated the scintillation properties of $BaO–B_2O_3$ glasses doped with different Er concentrations. Er^{3+} is well known for its luminescence in the NIR range, specifically at around 1500 nm,[24] and has been researched for several applications, such as scintillators,[25] lasers,[26] and optical amplifiers.[27] Barium borate and similar glasses have been well-researched,[28–30] and previous works have revealed that those glasses are promising materials for radiation shielding[31,32] and phosphors,[33–35] for example. In recent years, the optical and NIR photoluminescence properties of Er-doped barium-borate-based glasses have been reported.[36,37] To the best of our knowledge, there is no report on the scintillation properties of Er-doped barium borate glasses; therefore, we decided to investigate the scintillation properties of Er^{3+}-doped $BaO–B_2O_3$ glasses.

2. Materials and Methods

$50BaO–50B_2O_3$ glasses with different Er concentrations were synthesized by the melt-quenching technique with an electric furnace. First, Er_2O_3 (4N), $BaCO_3$ (4N), and B_2O_3 (5N) powders were blended at molar ratios of x/2:50:50 (x = 1, 5, 10, and 15) for the x% Er-doped sample, and the mixtures were ground by hand with a mortar and pestle. Second, the ground mixtures were put into alumina crucibles and melted at 1200 °C for one hour. Third, each melt was poured onto a steel plate and quenched by pressing with another steel plate. The steel plates were preheated at 300 °C. Finally, the obtained pieces were crushed to similar sizes and their surfaces were polished.

To confirm the amorphousness of the samples, X-ray diffraction (XRD) patterns were obtained with MiniFlex 600 (Rigaku). To confirm the transparency of the samples, diffuse transmission spectra were measured using a spectrophotometer (SolidSpec-3700, Shimadzu).

To obtain scintillation spectra, our original setup[38] was used with the X-ray tube voltage 80 kV and the current of 1.2 mA. To detect NIR rays, an InGaAs CCD (DU492A, Andor) was used. X-ray dose rate response functions were measured with our original setup[39] with an X-ray tube voltage of 40 kV.

3. Results and Discussion

Figure 1 shows the appearance and XRD patterns of synthesized $BaO–B_2O_3$ samples with different Er concentrations. All the obtained samples looked transparent and homogeneous. In the XRD patterns, an amorphous halo peak was observed for each sample, suggesting that the obtained samples were glass. Figure 2 shows the transmission spectra of all the samples in the UV–VIS and NIR ranges. Narrow absorption bands were observed at 360, 380, 410, 450, 490, 520, 540, 650, 800, 970, and 1530 nm, which were ascribed to 4f-4f transitions of Er^{3+}, namely, transitions from $^4I_{15/2}$ to $^4G_{9/2}$, $^4G_{11/2}$, $^2G_{9/2}$+$^4H_{9/2}$, $^4F_{5/2}$, $^4F_{7/2}$, $^2H_{11/2}$, $^4S_{3/2}$, $^4F_{9/2}$, $^4I_{9/2}$, $^4I_{11/2}$, and $^4I_{13/2}$, respectively.[36] Those narrow absorption bands became more intense with increasing Er concentration.

Figure 3 shows the X-ray-induced scintillation spectra of all the samples in the NIR range. A scintillation peak was observed at 1540 nm for each sample. The scintillation peak would be due

to the $^4I_{13/2} \rightarrow {}^4I_{15/2}$ transition of Er^{3+}.[36] To evaluate the applicability of the samples in devices, their X-ray dose rate response functions were examined. Figure 4 shows the dose rate response functions of the samples per unit mass. The 5% Er-doped sample showed the highest scintillation intensity of all the samples, according to the approximate functions. The lower detection limit

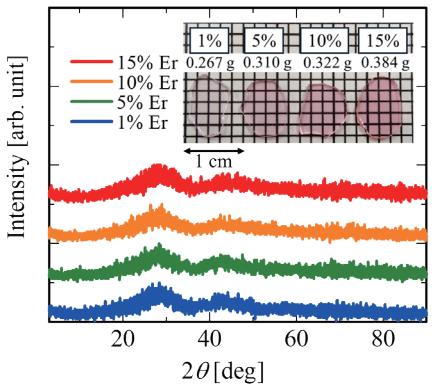

Fig. 1. (Color online) Appearance and XRD patterns of all the samples.

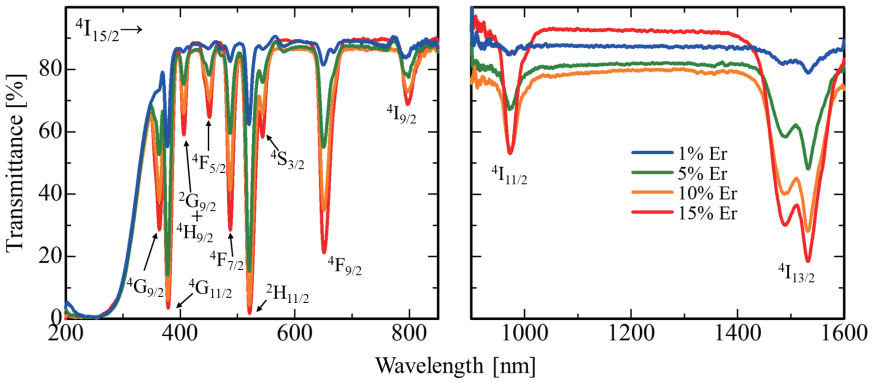

Fig. 2. (Color online) Transmission spectra of all the samples in the UV–VIS (left) and NIR (right) ranges.

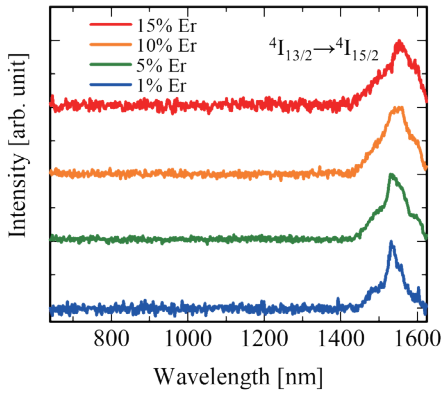

Fig. 3. (Color online) X-ray-induced scintillation spectra of all the samples in the NIR range.

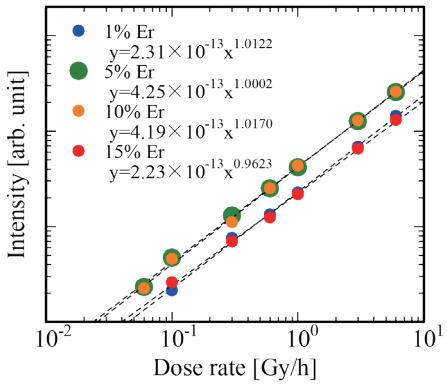

Fig. 4. (Color online) Dose rate response functions of all the samples.

(*LDL*) of the X-ray dose rate was 0.06 Gy/h, which was obtained with the 5% and 10% Er-doped samples. Compared with previous reports on NIR scintillators, this result is mediocre.[25,40–42] The relatively small effective atomic numbers (Z_{eff}s) of Er-doped $BaO–B_2O_3$ glasses could have resulted in low interaction probability with X-rays, leading to inferior LDL. While the Z_{eff} of 5% Er-doped $BaO–B_2O_3$ glass is 52, some NIR scintillators with superior *LDL*s have higher Z_{eff}s, such as Er:$Bi_4Ge_3O_{12}$[40] (*LDL* = 0.006 Gy/h, Z_{eff} = 75), Nd:$LuVO_4$[41] (*LDL* = 0.006 Gy/h, Z_{eff} = 63), and Nd:$Bi_4Ge_3O_{12}$[42] (*LDL* = 0.01 Gy/h, Z_{eff} = 75). To achieve higher scintillation intensity and superior *LDL*, adding heavy metallic oxides would be effective.

4. Conclusions

Er^{3+}-doped $50BaO–50B_2O_3$ glasses were successfully synthesized by the melt-quenching technique. All the obtained samples were transparent and homogeneous. The amorphousness of the samples was confirmed from XRD patterns. Diffuse transmission spectra showed that Er^{3+} ions were contained in the samples. The samples exhibited scintillation in the NIR range, which was due to the 4f-4f transition of Er^{3+}. The 5% Er-doped sample showed the highest scintillation intensity among all the samples, and the *LDL* of the X-ray dose rate was 0.06 Gy/h, which was obtained with the 5% and 10% Er-doped samples. This *LDL* was mediocre compared with those of previously reported samples. To achieve higher scintillation intensity and superior *LDL*, adding heavy metallic oxides would be effective.

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