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Scintillation and dosimetric properties of Ce-doped strontium aluminoborate glasses

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ABSTRACT

Strontium aluminoborate glasses with different concentrations of Ce (0, 0.01, 0.05, 0.1 and 0.5 mol%) were synthesized by the conventional melt-quenching technique. After the synthesis, photoluminescence (PL), scintillation and dosimetric properties of the samples were evaluated systematically. In the photoluminescence and scintillation spectra, a sharp peak around 370 nm due to the 5d-4f transitions of Ce was observed. The 0.05% Ce-doped sample showed the highest intensity in PL and scintillation among the samples investigated. The 0.01–0.1% Ce-doped samples also showed thermally-stimulated luminescence (TSL) with a glow peak of around 90 °C after X-ray irradiation. The TSL response of 0.01–0.05% Ce-doped samples increased monotonically with X-ray dose over a dose range of 10^{-1} – 10^4 mGy.

1. Introduction

Ionizing radiation detections have attracted much attention for over the past decades due to a wide range of application fields such as border security [1], nuclear medicine [2], oil-dwelling [3] and individual radiation monitoring [4]. Those radiation detectors often utilize solid state phosphor materials, and the detectors are categorized into two types; scintillation detectors and dosimeters. The former type uses scintillators, which immediately convert a single quantum of high energy (keV–GeV) to thousands of low energy photons such as UV/Vis/NIR light via energy transfer from the host to luminescence centers [5]. On the other hand, the latter type uses storage type phosphors, in which the incident radiation energy is stored as a form of locally captured carries and then emit photons by external stimulation [6]. In general, luminescent phenomena involved in dosimeter materials are categorized into three types: thermally-stimulated luminescence (TSL) [7], optically-stimulated luminescence (OSL) [8], and radio-photoluminescence (RPL) [9]. In the past studies, many researchers have developed ionizing radiation detectors using various kinds of material forms such as single crystals [10], ceramics [11], crystalline powder [12,13] and glasses [14].

Among the various material forms, glass materials have many advantages such as low cost, large productivities, easy handling and flexible chemical compositions. One of the examples of glass materials as scintillator is Li-glass, which is known as GS20 (Saint Gobain) [15]. The Li-glass consists of silica glass including ⁶Li and small amount of

Ce³⁺. The ⁶Li (enriched 95 mol%) is incorporated into the glass host in order to detect thermal neutrons [16] since ⁶Li has a high cross-section against neutrons. In addition, Ag-doped phosphate glasses (NaPO₃-Al(PO₃)₃) show RPL and are commonly used for individual radiation monitoring, commercialized by Chiyoda Technol Corp. In this glass system, the RPL is understood to be due to valence changes of Ag ion induced by ionizing radiation ($\text{Ag}^+ \rightarrow \text{Ag}^0 + \text{Ag}^{2+}$) [9]. The generated Ag⁰ and Ag²⁺ act as luminescent center and emit in blue and orange, respectively. The emission intensity is very stable at room temperature and excitation UV light, so the signal can be read out multiple times without fading.

In addition to these examples, borate glasses are of great interest due to the low melting temperature, high accommodation capability with rare earth elements and low effective atomic number (Z_{eff}). For individual radiation monitoring, the effective atomic number of dosimeter materials is preferred to be close to that of biological tissue ($Z_{\text{eff}} = 7.35$ – 7.65) [17], and many researchers have studied dosimetric properties of borate glasses [18–19]. For example, strontium tetraborate (SrB₄O₇) exhibits TSL with glow peaks at 110 and 360 °C, and it has a sensitivity equivalent to that of the commercial dosimeter (TLD-700) [20]. Li₂B₄O₇ doped with copper is known to exhibit the TSL sensitivity nearly eight times higher than that of TLD-100 [21–22]. Further, scintillation properties of borate glasses have also been investigated in the past studies. The scintillation light yields of orthoborates glasses such as YBO₃:Ce³⁺, GdBO₃:Ce³⁺ and YAl₃B₄O₁₂:Ce³⁺ are reported to be of the order of 1.0×10^4 photons/MeV [23]. Moreover,

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our research group demonstrates that Ce-doped magnesium aluminoborate glasses show efficient photoluminescence (PL) with the quantum efficiency (QY) reaching about 50%, scintillation as well as TSL properties [24]. As these examples, borate glass is one of candidate materials for scintillator and dosimeter applications.

In this study, we synthesized strontium aluminoborate glasses doped with various concentrations of Ce as luminescent center by the conventional melt-quenching technique. Few studies reported the scintillation and dosimetric properties of strontium aluminoborate glasses so far while dosimetric properties of alkaline earth borate compounds have been reported [23–24]. These glasses doped with Ce^{3+} exhibit efficient PL, scintillation and storage luminescence for dosimeter application due to the 5d-4f transitions of Ce^{3+} . The synthesized glass samples were characterized for radiation measurement applications, and the characterizations included optical in-line transmittance, PL, scintillation, TSL and OSL.

2. Experimental

Ce-doped strontium aluminoborate ($50\text{B}_2\text{O}_3\text{-}15\text{Al}_2\text{O}_3\text{-}35\text{SrO-xCeO}_2$, $x = 0\text{--}0.5$) glasses doped with different concentrations of Ce were synthesized by the conventional melt-quenching technique. Stoichiometric ratio of CeO_2 (99.99%, Rare Metallic), SrCO_3 (99.99%, Furuuchi Chemical), Al_2O_3 (99.99%, High Purity Chemicals) and B_2O_3 (99.99%, Furuuchi Chemical) powders were homogeneously mixed first, and the mixture was melted in an alumina crucible inside an electric furnace heated at 1200°C for 30 min under ambient atmosphere. The melt was then quenched on a preheated stainless plate at 350°C . The obtained glass samples were mechanically polished before characterizations.

The optical in-line transmittance spectrum was measured by using a spectrophotometer (V670, JASCO), which covered the spectral region from 190 to 750 nm. The PL quantum yield (QY) was measured by using Quantaurus QY (C11347, Hamamatsu). PL decay curves were measured by using Quantaurus τ (C11367, Hamamatsu). In these measurements, the excitation wavelength was 280 nm which was the shortest excitation wavelength available in the instrument, and the monitoring wavelength was 380 nm. As a scintillation property, X-ray-induced luminescence spectrum was measured by utilizing our original setup [25]. For irradiation, the X-ray tube was operated by applying the bias voltage of 40 kV and tube current of 5.2 mA. Scintillation decay time profiles were measured by afterglow characterization system, equipped with a pulse X-ray source [26]. The spectral sensitivity of PMT used in this measurement was from 160 to 650 nm. In order to investigate TSL dosimetric properties, the TSL glow curve was measured by using TL-2000 (Nanogray) with a heating rate of 1°C/s over a temperature range from 50 to 490°C [27]. Further, TSL spectrum was measured using an Andor CCD-based spectrometer while the sample was heated by an electric heater (SCR-SHQ-A, Sakaguchi) at a rate of 1°C/s .

3. Results and discussion

3.1. Sample

Fig. 1 illustrates a photograph of non-doped and Ce-doped strontium aluminoborate glass samples under room light. The thickness of the samples was controlled by hand-polishing to be about 1 mm. The stripe pattern on the back of the samples was clearly seen through. The sample color gradually changed from colorless to brown with increasing the concentrations of Ce.

Fig. 2 represents the in-line transmittance spectra of the non-doped and Ce-doped glass samples. All the samples showed high transparency over the 400–750 nm range. In the non-doped sample, the absorption edge was about 200 nm. On the other hand, a strong absorption band was observed in the Ce-doped samples around 300–400 nm, which was

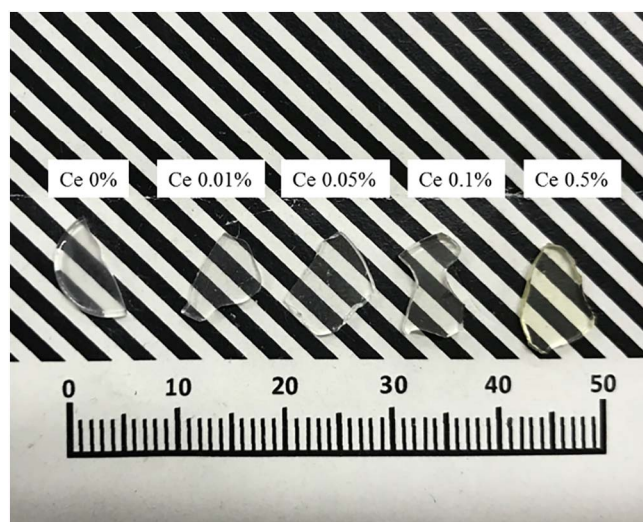


Fig. 1. Photograph of non-doped and Ce-doped strontium aluminoborate glasses.

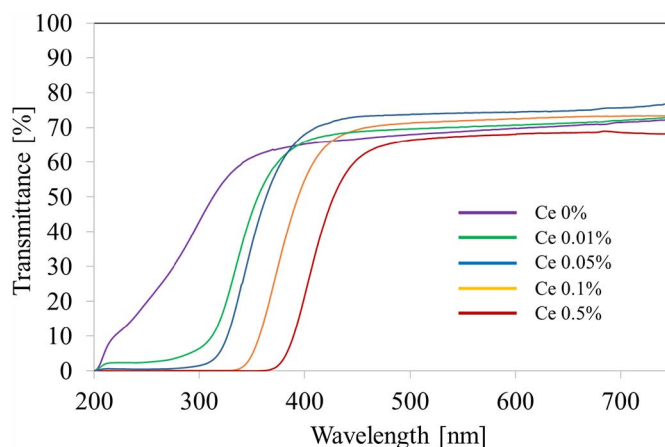


Fig. 2. Transmittance spectra of non-doped and Ce-doped strontium aluminoborate glasses.

a typical absorption feature due to the 5d-4f transitions of Ce^{3+} [28–29]. The optical absorption edge of the Ce-doped samples shifted to lower energy with increasing the concentration of Ce.

3.2. Photoluminescence properties

Fig. 3 shows PL emission and excitation contour graphs of non-doped and Ce-doped strontium aluminoborate glasses. The Ce-doped samples exhibited an emission at 380 nm under excitation wavelengths across 320–410 nm while no measurable signals from the non-doped sample could be detected. The excitation band features coincided with those of the absorption band measured in the in-line transmission spectra in Fig. 2. In addition, the emission wavelength of Ce-doped samples agreed well with reported value for other Ce-doped glasses [28–29]. For this reason, the emission can be attributed to the 5d-4f transitions of Ce^{3+} . The PL peak wavelength seemed to red-shift with increasing the concentration of Ce. The reason was blamed for an increase of absorption intensity due to the 5d-4f transitions of Ce^{3+} located at slightly shorter wavelength than that of emission, which absorbs a high-energy portion of luminescence photons (so-called self-absorption). In addition, the QY values of non-doped and Ce-doped samples were also indicated in Fig. 3. The QY values measured were 0.08 (Ce:0.01%), 0.08 (Ce:0.05%), 0.05 (Ce:0.1%) and 0.01 (Ce:0.5%). The non-doped sample did not show measurable signal. The QY values of 0.01% and 0.05% Ce-doped samples were the highest among the

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