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Fluorescence and optical properties of Eu^{3+}-doped borate glasses

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ABSTRACT

The glasses with chemical composition of $25Na_2O$:23CaO:$6P_2O_5$:$44B_2O_3$:$2ZrO_2$: xEu_2O_3 for extra different Eu_2O_3 concentrations of $x = 0.25$, 0.5, 0.75, 1.0 and 1.25 mol% were synthesized by the conventional melt quenching technique. The glass structure, physical property, optical and luminescence characteristics of the synthesized glasses were characterized by FT-IR spectroscopy, X-ray powder diffraction and photoluminescence (PL) spectra, respectively. The results indicated that the density and molar volume of the glasses were found to increase with Eu_2O_3 concentration. The glasses were composed of [PO_4], [BO_3] and [BO_4] basic structural units. Under 393 nm wavelength excitation, five luminescence bands were observed at 578 nm, 592 nm, 612 nm, 653 nm and 701 nm, which corresponds to $^5D_0 \rightarrow {}^7F_J$ (J = 0, 1, 2, 3, 4) transitions. And the most intense red emission was found at 612 nm. Moreover, the color coordinate and color correlated temperatures (CCTs) changed with Eu_2O_3 contents. This work shows the potential application of europium-doped glass samples in UV converted white light-emitting diodes (WLEDs) and other optical devices.

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1. Introduction

Recently, luminescence materials have emerged as a fascinating field of research since its characteristics encompasses a wide variety of scientific applications including the design and development of new optical devices such as solid state lasers, white light emitting diodes (WLEDs), optical telecommunication, sensor and other flat panel technologies like plasma display panels (PDP) [1–6]. Among these luminescence materials, rare earth (RE) ions doped glass is considered to be a promising host material due to its homogeneous distinct visible emission, simpler manufacture procedure, lower production cost, free of halo effect, and better thermal stability, and it can be used for various optical devices such as lasers, WLEDs, stimulated phosphors and optical amplifiers [1,7–9]. It is well known that the luminescence and absorption properties of RE ions in glasses depend on the chemical composition, structure and nature of bonds of the host glass [10]. Borate-based glasses are good hosts for RE ions and having flexibility for both size and composition of the materials [11]. The Eu^{3+} ion having a red-emitting nature of $^5D_0 \rightarrow {}^7F_2$ transition around 613 nm is frequently used in field emission technology, spectroscopic probe, display and WLEDs [12–14]. Na_2O-CaO-P_2O_5-B_2O_3-ZrO_2 (NCPBZ) glass systems are expected to be a new kind of forming glass because of their low melting temperature, low cost, good physical and chemical properties. To our best knowledge, few researches on the Eu^{3+} doped-NCPBZ glasses have been reported.

In this work, the Eu^{3+} doped-NCPBZ glasses were prepared via melting-quenching technique. The effects of Eu^{3+} on the densities, molar volume, FT-IR transmittance, absorption, photoluminescence and decay time in the as-prepared NCPBZ glasses were studied systematically. Moreover, the color coordinate and color correlated temperatures (CCTs) were also investigated. It is expected that the investigated glasses might be a promising candidate for solid-state lighting as well as optical display devices.

2. Experimental procedures

Powders of analytical reagent grade, comprising Na_2CO_3, $CaCO_3$, $NH_4H_2PO_4$, H_3BO_3, ZrO_2 (≥99.5%), and high purity Eu_2O_3 (≥99.99%), were used as the starting materials. Glass samples with the formula $25Na_2O$-23CaO-$6P_2O_5$-$44B_2O_3$-$2ZrO_2$-xEu_2O_3 (mol %), where $x = 0.25$, 0.5, 0.75, 1.0 and 1.25. The concentration percentage of Eu^{3+} ions (x) is according to the total mol of all raw materials except for Eu_2O_3, namely, extra mole percentage. Corresponding samples are labeled as NCPBZ: 0.25Eu, NCPBZ: 0.5Eu, NCPBZ: 0.75Eu, NCPBZ: 1.0Eu and NCPBZ: 1.25Eu. $SrCO_3$ and $CaCO_3$ were dried at 180 °C except for $NH_4H_2PO_4$ and H_3BO_3 while Eu_2O_3 and ZrO_2 were dried at 900 °C overnight to remove the moisture prior to weighing. The mixtures were calcined at 500 °C for 60 min to remove vapor and then were melted in a corundum crucible at 1250 °C in air for 2 h in an electric furnace. The glass melts were poured onto a preheated copper mold for quenching and annealed in a muffle furnace at 450 °C for 3 h to release thermal stress and then cooled to room temperature. The glass samples were then cut and polished into $10 \times 10 \times 1$ mm^3 shape for optical measurements.

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The phase analysis of the glass samples was identified by X-ray powder diffractometer (Model D8-Advance, BRUKER, Germany) with Cu Kα radiation at room temperature. The densities (ρ) were measured by Archimedes method using distilled water as an immersion liquid. Infrared spectra of the glass samples were recorded at room temperature in the 400–1800 cm^{-1} by Fourier transform infrared spectrometer (Model Tensor 27 FT-IR, BRUKER, Germany). The optical absorption spectra were recorded in the wavelength range 200–900 nm using a UV-3600 Shimadzu UV–Vis–NIR spectrophotometer. The emission, excitation spectra, temperature dependent emission spectra and decay curves of the Eu^{3+} doped glass samples were recorded on an Edinburgh Instruments FS5 spectrofluorometer equipped with both continuous (150 W) and pulsed xenon lamps as well as a homemade temperature controlling stage.

3. Results and discussion

XRD patterns of all the glass samples are shown in Fig. 1. The result of XRD pattern indicates that all the glass samples exhibit similar XRD patterns. And all of the glass samples are structurally amorphous characterized by diffused humps and the absence of sharp peaks, which confirms that these samples are all glasses and there is no any crystallization.

The densities and molar volumes of the investigated NCPBZ glasses are shown in Fig. 2. It is found that both the density and molar volume of the glass samples increase with increasing Eu_2O_3 concentration. The values of density are in the range of 2.52–2.60 g/cm^3 while their molar volumes (VM) are in the range of 28.13–28.58 cm^3/mol. The increase in density of the glass with increasing Eu^{3+} doping content is due to a higher molecular weight of Eu_2O_3 as compared to other components in the glasses. While the increase in the molar volume, indicates the increasing of inter-atomic spacing in the glass network. Eu_2O_3 acts as a network modifier and produces more non-bridging oxygen (NBOs) in glass matrices [15].

The infrared spectra of Eu^{3+} doped NCPBZ glasses in a range of 400–1600 cm^{-1} are displayed in Fig. 3. It is observed that the main structures of all the glasses are almost identical. There are three groups of absorption bands due to different structural units of P_2O_5 and B_2O_3. The detail FTIR analysis of the Eu^{3+} doped NCPBZ glasses are listed in Table 1. The band observed at 496 cm^{-1} can be assigned to the bending vibrations of O—P—O [16]. The band located at 558 cm^{-1} is due to the bending vibration of P—O—B chains and PO_4 units [17]. The broad band centered at 736 cm^{-1} is assigned to the bending vibrations of B—O linkages in the borate network [18, 19]. The absorption band near 885 cm^{-1} is attributed to the stretching vibrations B—O of BO_4 groups [20,21]. The band located at 1065 cm^{-1} is due to the symmetric stretching P—O—B link between BO_4 and PO_4 units. [17,20–22]. The band at 1324 cm^{-1} comes from the stretching vibrations of the B—O of trigonal BO_3 units and double bonded P═O linkages [17,20–23].

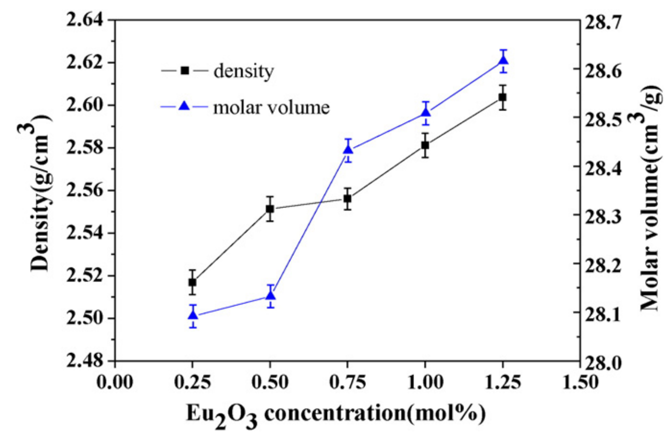

Fig. 2. Densities and molar volumes of NCPBZ glass samples with different Eu_2O_3 concentrations.

The typical excitation spectrum of NCPBZ: 0.25Eu obtained by monitoring the emission with 612 nm is shown in Fig. 4(a). The spectrum contains four bands at 362, 383, 393 and 413 nm which are attributed to the transitions of the Eu^{3+} ions: $^7F_0 \rightarrow {}^5D_4$, $^5L_7$, $^5L_6$ and $^5D_3$, respectively. The typical absorption spectra of the NCPBZ: 0.25Eu are shown in Fig. 4(b). It is observed that Eu^{3+} ions-doped glass sample exhibits absorption levels at 362, 383, 393 and 413 nm, and the prominent levels observed are assigned to the appropriate electronic transitions, as follows: $^7F_0 \rightarrow {}^5D_4$, $^5L_7$, $^5L_6$ and $^5D_3$, which notices that the wavelength of excitation light matches with the wavelength of absorption peak of the sample. It indicates that the Eu^{3+} doped glasses can be efficiently excited by UV light. Therefore, the strongest excitation wavelength of 393 nm is suitably chosen as the exciting wavelength in the following experiments.

The emission spectrum of NCPBZ: 0.25Eu under 393 nm is shown in Fig. 5 (a). The spectra exhibit five emission bands corresponding to the $^5D_0 \rightarrow {}^7F_0$ (578 nm; yellow), $^5D_0 \rightarrow {}^7F_1$ (592 nm; orange), $^5D_0 \rightarrow {}^7F_2$ (612 nm; red), $^5D_0 \rightarrow {}^7F_3$ (653 nm; red) and $^5D_0 \rightarrow {}^7F_4$ (701 nm; deep-red) transitions. Fig. 5(b) shows the emission spectra

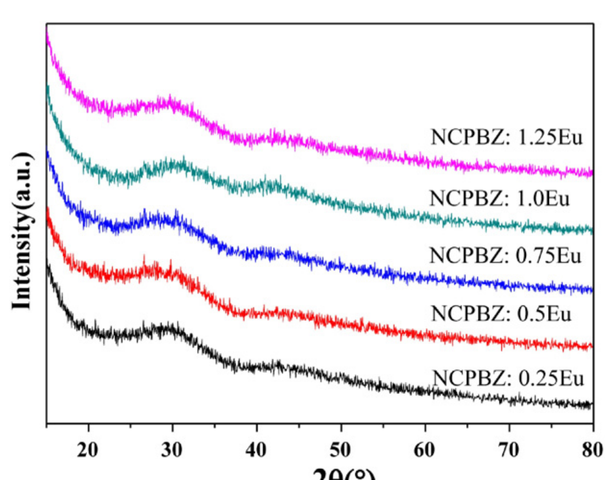

Fig. 1. XRD patterns of all the glass samples.

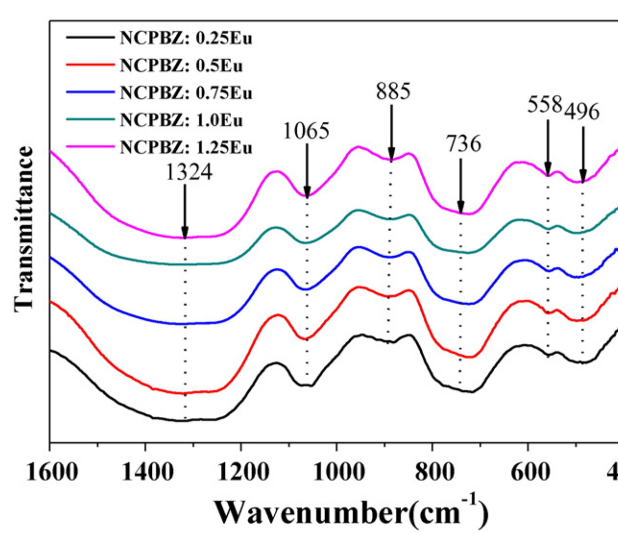

Fig. 3. FT-IR spectra of the glass samples from NCPBZ: 0.25Eu to NCPBZ: 1.25Eu.

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