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Cerium luminescence in borate glass and effect of aluminium on blue green emission of cerium ions



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ABSTRACT

CeO₂ doped lead borate (CE) and lead alumino borate (CEA) glasses are prepared by melt quench method at high temperature. The main luminescence band of 5d–4f transition of Ce³⁺ ions with maxima at around 489 nm of Ce³⁺ ions in these glasses has been observed, along with red shift and larger stokes shift, which shows that the covalency of the rare earth to oxygen bond increases with the increase in CeO₂ content at the expense of Al₂O₃. Shifting of UV absorption edge towards longer wavelength and a decrease in band gap with increase in CeO₂ concentration in both the glass systems has been observed. Moreover densification and stabilization of glass network has been observed which is due to conversion of BO₃ units to more compact and stable BO₄ units. This covalency effect and the formation of BO₄ groups with addition of CeO₂ and incorporation of Al₂O₃ content are responsible for clear effect on luminescence of the present glass system. Moreover the optical basicity values were theoretically determined along with density and molar volume.

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1. Introduction

Lead borate glasses are mainly classified as non-crystalline materials which are favorable to be investigated by spectroscopic methods. These glasses show wide transmission range of light, low phonon energy, large ion polarizabilities resulting in large values of refractive indices and higher covalency which make them suitable host lattices for rare earth (RE) ion incorporation [1,2]. Moreover heavy metal glass (HMG) helps to increase radiative or quantum efficiencies of rare earth ions [3,4].

Oxide materials which consist of heavy metal oxides are most promising for photonics and optoelectronics as they have stable glass structure with weak ionicity and lower phonon energy which are supportive in improving radiation hardness of scintillators and small non-emissive losses during luminescence.

Spectroscopic properties of rare earth ions are mainly associated with 4f transitions and are governed by chemical composition of glass systems. In our present study, we are mainly concerned about cerium doped lead metal borate glasses. A wide range of glasses containing cerium oxide has been developed by many researchers [5–8].

Cerium doped oxide glasses are very important for their optical as well as lasing properties, due to strong optical absorption and fluorescence in near UV or Visible and infrared region [9–11].

Trivalent cerium (4f¹) possesses parity allowed 4f–5d electronic transitions and due to the large energy gaps, non-radiative decay is less likely to occur and 5d emission can be observed [12] whereas in case of tetravalent [4f⁰] cerium, absorption belongs to character of charge transfer and has no luminescence [13]. Spectroscopic properties of Ce³⁺ ions in various crystals and glasses have been an active area of research due to its fast decay time and relatively high light output, so it is used in various applications such as scintillators for applications in high-energy physics, for the detection of X-rays, γ -rays or neutrons and tunable solid-state lasers operating in the near UV, violet, and blue regions [14–16]. However concentration quenching effect is one of the major setbacks in the luminescence yield of Ce³⁺ due to clustering tendency of rare earth ions [17].

Addition of Al₂O₃ in borate and borosilicate glasses enhances glass formation ability and simultaneously reducing non bridging oxygens [18,19]. Al₂O₃ has received significant attention as it increases solubility of rare earth ions by inducing structural changes around rare earth ions and helps to limit concentration quenching effects on photoluminescence thereby improving the optical response of the material [20,21]. Moreover, the presence of Al or B helps to decrease the Ce⁴⁺ ions and increase the Ce³⁺ ions [22].

So for the search of new and efficient host-ion composition that has potential in luminescence for optoelectronic devices. We, in the present study, have made a comparative study of two glass systems CeO₂–PbO–B₂O₃ (CE) and CeO₂–Al₂O₃–PbO–B₂O₃ (CEA).

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Table 1
The nominal composition of prepared glass samples (mol%).

Glass code	CeO ₂ (%)	PbO (%)	B ₂ O ₃ (%)	Al ₂ O ₃ (%)
CE1	0	20	80	0
CE2	0.5	20	79.5	0
CE3	1	20	79	0
CE4	1.5	20	78.5	0
CE5	2	20	78	0
CEA1	0	20	70	10
CEA2	0.5	20	70	9.5
CEA3	1	20	70	9
CEA4	1.5	20	70	8.5
CEA5	2	20	70	8

2. Experimental procedure

2.1. Sample preparation

Glass samples $x\text{CeO}_2\text{--}20\text{PbO}\text{--}(80\text{--}x)\text{B}_2\text{O}_3$, (CE) and $x\text{CeO}_2\text{--}(10\text{--}x)\text{Al}_2\text{O}_3\text{--}20\text{PbO}\text{--}70\text{B}_2\text{O}_3$ (CEA) with x varying from 0 to 2 mol% are prepared by the conventional melt quench technique. The raw materials lead oxide (PbO), cerium oxide (CeO₂), aluminium oxide (Al₂O₃) and boric oxide (B₂O₃) in appropriate proportions are weighed by using an electric balance and ground together to form 15 g batch. Thoroughly ground mixture is melted and has been held at 1050 °C for 60 min in an alumina crucible until a bubble free liquid is formed. The homogenized melt is then quenched in air by pouring it onto preheated steel mold and annealed at temperature of 380 °C for 1 h to avoid breaking of the sample by residual internal thermal strains. The obtained samples are polished with cerium oxide in order to obtain maximum flatness. The nominal composition of the prepared glasses is given in Table 1.

2.2. XRD

The glass formation of the samples is confirmed by X-ray diffraction (XRD) study using an XRD-7000 (Shimadzu, Japan) X-ray diffractometer (Cu K α , $\lambda = 1.54434 \text{ \AA}$) at the scanning rate of 2 deg/min and 2θ is varied from 10–70°.

2.3. FTIR

Infrared spectroscopy is carried out at room temperature in the range of 400–4000 cm⁻¹ using a Varian 660-IR spectrometer. The powder form of each sample is mixed with KBr in an agate mortar in the ratio 1:100 mg (glass powder: KBr) and then pressed to a pressure of 1.470×10^7 Pa to form the pellets which are used for recording the FTIR spectra.

2.4. Density

The density of glass samples at room temperature is measured by using the standard principle of Archimedes using a sensitive microbalance with pure benzene as the immersion fluid using formula (1).

$$D = [W_A / (W_A - W_B)]d, \quad (1)$$

where W_A is the weight of sample in air, W_B is the weight of the sample in benzene, and d is density of the benzene.

The molar volume (V_m) is calculated by using the formula (2):

$$V_m = \sum x_i M_i / d \quad (2)$$

where x_i is the molar fraction of the component, M_i is its molecular weight and d is density of glass sample.

2.5. UV-vis spectroscopy

The optical absorption spectra of polished samples are recorded at room temperature by using a UV-vis spectrophotometer Perkin-Elmer Lambda 35 in the range 200–900 nm.

2.6. Fluorescence

The fluorescence spectra of the samples are recorded with the help of a Perkin-Elmer Fluorescence LS-Spectrometer. The excitation wavelength for the samples is 380 nm.

3. Results and discussion

3.1. X-ray diffraction of CeO₂–PbO–B₂O₃ and CeO₂–Al₂O₃–PbO–B₂O₃ glasses

X-ray diffraction spectra of CE glass samples (Fig. 1) shows no continuous or discrete sharp peaks but consists of diffused halo which reflects the characteristics of amorphous nature of glass samples. However the diffractograms associated with alumino borates show a slight hint of crystallinity but with very low intensity of broad and diffused peaks [23].

3.2. FTIR (Fourier transform infrared spectroscopy)

3.2.1. CeO₂–PbO–B₂O₃ glasses

The FTIR spectra of CeO₂ doped PbO–B₂O₃ have been given in Fig. 2(a). Infrared spectra of borate glasses can be divided in to three regions [24,25].

- (I) 600–800 cm⁻¹ for the B–O–B vibrations.
- (II) 800–1200 cm⁻¹ for BO₄ groups.
- (III) 1200–1600 cm⁻¹ for BO₃ groups.
- (IV) The near infrared bands in region 3000–4000 cm⁻¹ due the presence of water groups' vibration [26].

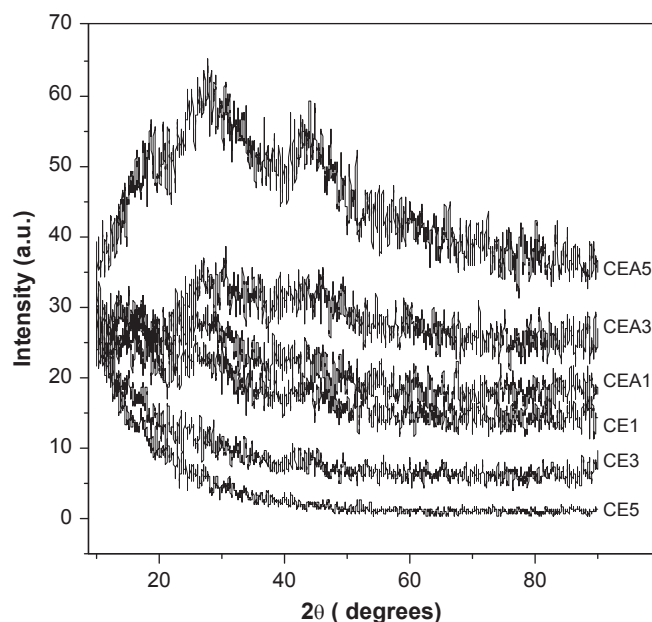


Fig. 1. XRD spectra of CeO₂–PbO–B₂O₃ (CE) and CeO₂–Al₂O₃–PbO–B₂O₃ (CEA) glasses.

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