



Excitation induced spectroscopic study and quenching effect in cerium samarium codoped lithium aluminoborate glasses



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ARTICLE INFO

Article history:

Received 16 December 2015

Received in revised form

17 March 2016

Accepted 16 April 2016

Available online 19 April 2016

Keywords:

Borate glass

Melt quench

Optical absorption

Fluorescence

Concentration quenching

Energy transfer

ABSTRACT

Lithium aluminium borate host has been codoped with cerium and samarium to prepare glass by conventional melt quench technique. Their structural and spectroscopic investigation has been carried out using XRD, FTIR and density measurements. The UV-Vis absorption spectra and fluorescence spectra ($\lambda_{exc.} = 380$ nm and 400 nm) have been studied for spectroscopic analysis. The amorphous nature of the prepared samples is shown by XRD. The density is increasing with addition of cerium at the expense of aluminium, keeping other components constant. FTIR study also shows the presence of compact and stable tetrahedral BO_4 units thus supporting the density results. The UV-Vis absorption spectra show a shift of optical absorption edge towards longer wavelength along with an increase in intensity of peaks with rising samarium concentration. The fluorescence spectra show a blue shift and subsequent suppression of cerium peaks with addition of samarium.

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1. Introduction

Rare earth ions are extensively examined in various crystals and glasses and play a prominent role in the development of various optoelectronic devices such as lasers, sensors, light converters, hole burning high density memories, optical fibres and amplifiers [1–3]. Glasses have now become the promising host materials for lasing rare earth ions due to their flexibility of shape and size, transparency and ease of fabrication by different methods which are useful in the class of photonic materials due to several inherent advantages over their crystalline competitors. Borate glass, among them, is a suitable optical material having high transparency, thermal stability, low melting temperature and good solubility of rare-earth ions [4,5].

Samarium and cerium play a vital role as dopants/components in glasses. The spectroscopic studies of $Sm^{3+}(4f^5)$ ions have shown that their optical properties can be influenced by varying the glass composition [6,7] which makes these glasses an appropriate host medium due to the presence of sharp and clear bands in their absorption and luminescence spectra. Sm^{3+} ions emit reddish-orange light due to $^4G_{5/2} \rightarrow ^6H_J$ transitions ($J = 5/2, 7/2, 9/2, 11/2$) [8]. Cerium incorporated glasses possess very important optical and

lasing properties [9–11] and are useful as laser media, scintillating materials and protector of fibre optic materials against radiation [12,13]. Moreover, the presence of rare earth oxides significantly reduces the viscosity of the melt but slightly affect the thermal expansion, chemical durability and mechanical properties of the glass [14]. Aluminium oxide improves the physical properties and chemical stability by changing the glass structure. It affects thermo-mechanical properties as well as laser properties [15]. The presence of aluminium also prevents the concentration quenching effect due to clustering of rare earth ions [16]. The addition of alkali oxide like lithium oxide may help to reduce the melt viscosity thereby lowering the melting temperature [17].

We have already studied the cerium and samarium codoped lithium aluminium borate host keeping samarium content constant and varying cerium concentration and found the applicability of those glass compositions in white light emitting devices [18]. Since the literature on cerium-samarium codoped systems is scarce [19] and needs investigation, so in the present study we have studied the same host and same rare earths for codoping but now we have kept cerium content constant and varied samarium concentration in order to find some novel aspects of this glass composition for laser devices and in the field of luminescence based lighting. The results have also been compared with the previous ones.

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2. Experimental procedure and characterization

Glass samples having chemical composition $x\text{Sm}_2\text{O}_3-(9.5-x)\text{Al}_2\text{O}_3-0.5\text{CeO}_2-10\text{Li}_2\text{CO}_3-80\text{B}_2\text{O}_3$ with x varying from 0% to 1.5% mole fraction are prepared by using conventional melt quench technique. The experimental procedure and characterization techniques are same as described in our previous study [18]. Table 1 shows the nominal composition of prepared glass samples.

3. Results and discussion

3.1. X-ray diffraction

X-ray diffraction pattern (Fig. 1) does not contain any continuous or distinct sharp peaks and comprises of diffused bands only which clearly indicate the amorphous/glassy nature of the prepared glass samples.

3.2. Density

It is shown in Table 1 that as the Sm_2O_3 concentration is increased at the cost of Al_2O_3 ; there is an increase in density of the prepared glass samples. This increase is sharp when 0.5 mol% samarium oxide is added in sample CS0.0 and then this increase is gradual. Since the molecular weight and density (8.347 g/cm^3) of Sm_2O_3 is more than any other component of the given glass system, so this increase is quite obvious as samarium concentration is being increased.

A corresponding decrease in molar volume is observed which supports the above discussed density change and shows a quite expected harmony between these two physical parameters as it is also clear from Fig. 2.

3.3. FTIR

Fig. 3 shows the room temperature FTIR spectra of the prepared glass samples. It provides the information regarding various structural groups present and helps to know the changes occurring in their arrangements with compositional variation in the glass samples. The borate spectra have been classified as follows: (i) $600-800 \text{ cm}^{-1}$ is attributed to bending vibrations of various B-O-B segments, (ii) $800-1200 \text{ cm}^{-1}$ is for stretching vibrations of BO_4 groups and (iii) $1200-1600 \text{ cm}^{-1}$ shows the B-O stretching vibrations of BO_3 groups. The other bands from 2300 to 4000 cm^{-1} are due to O-H vibrations of water groups. The band at 806 cm^{-1} has been assigned to boroxol rings which is absent in our glass system depicting the presence of BO_3 and BO_4 groups only [20,21].

The present glass system exhibits infrared transmission at about $692, 928, 1050, 1224$ and 1391 cm^{-1} . The bands at $1391-1405 \text{ cm}^{-1}$ may be attributed to stretching vibrations of non-bridging oxygens (NBOs) of trigonal BO_3 units in metaborate chains or rings, pyro- and ortho- borate groups [22]. Though these

Table 1

Nominal chemical composition (in mol%), density and molar volume of the glass samples.

S.No.	CeO ₂	Sm ₂ O ₃	Al ₂ O ₃	Li ₂ CO ₃	B ₂ O ₃	Density (g/cm ³)	Molar Volume (cm ³ /mol)
CS0	0.5	0.0	9.5	10	80	2.187	33.669
CS0.5	0.5	0.5	9.0	10	80	3.108	24.088
CS1.0	0.5	1.0	8.5	10	80	3.682	20.668
CS1.5	0.5	1.5	8.0	10	80	3.903	19.814

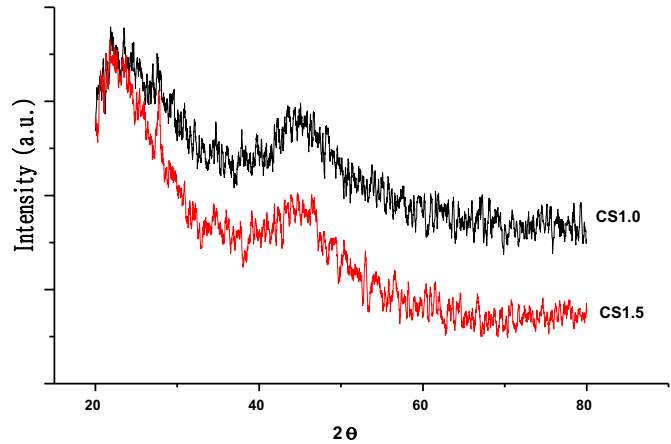


Fig. 1. XRD spectra of $\text{CeO}_2\text{-Sm}_2\text{O}_3\text{-Al}_2\text{O}_3\text{-Li}_2\text{CO}_3\text{-B}_2\text{O}_3$ glasses.

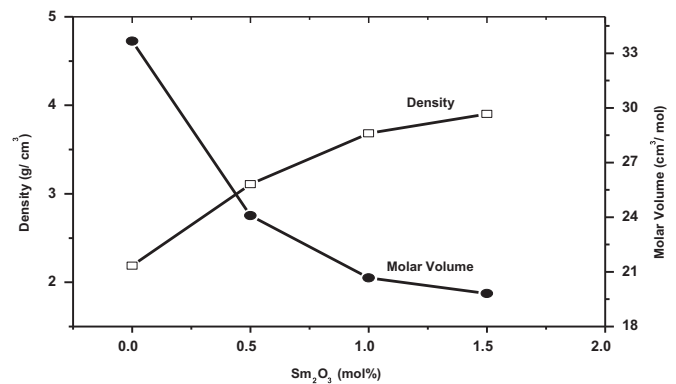


Fig. 2. Density and molar volume of $\text{CeO}_2\text{-Sm}_2\text{O}_3\text{-Al}_2\text{O}_3\text{-Li}_2\text{CO}_3\text{-B}_2\text{O}_3$ glasses.

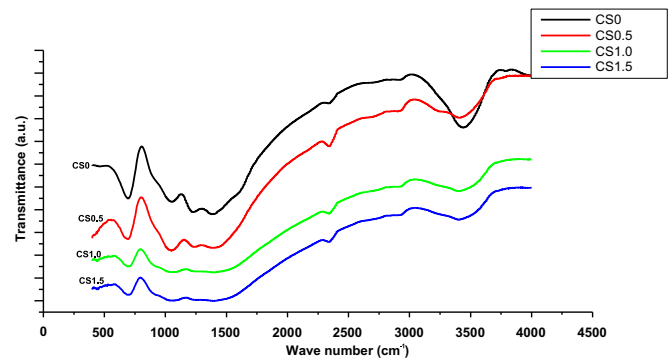


Fig. 3. FTIR spectra of $\text{CeO}_2\text{-Sm}_2\text{O}_3\text{-Al}_2\text{O}_3\text{-Li}_2\text{CO}_3\text{-B}_2\text{O}_3$ glasses.

NBOs' vibrations are present in the sample CS0.0 but with addition of samarium, its intensity decreases continuously and moves towards disappearance in the sample CS1.5 which shows that NBOs are getting extinct. So we can say that samarium helps in reducing the NBOs. This also goes well with density observations that density shows an increasing trend as we move from sample CS0.0 to CS1.5. The IR transmission band at about 1224 cm^{-1} is due to B-O stretching vibrations of trigonal BO_3 units present in meta-, pyro- and ortho- borate groups [23]. There is a reduction in intensity of this band as samarium oxide is increased at the expense of aluminium oxide and it is almost absent in the sample CS1.5 which shows that BO_3 groups are decreasing. The band at 1050 cm^{-1} may be attributed to B-O stretching vibrations of BO_4 units from diborate ($\text{B}_4\text{O}_9^{2-}$), triborate (B_3O_7^-), tetraborate ($\text{B}_4\text{O}_{10}^{2-}$) and pentaborate ($\text{B}_5\text{O}_{10}^-$) groups [22,24–26]. A small band at 928 cm^{-1} may be assigned to stretching

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