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Key Engineering Materials Vols. 675-676 (2016) pp 424-429 Submitted: 2015-06-11 © (2016) Trans Tech Publications, Switzerland Revised: 2015-08-10 doi:10.4028/www.scientific.net/KEM.675-676.424 Accepted: 2015-08-18 All rights reserved. No part of contents of this paper may be reproduced or transmitted in any form or by any means without the written permission of Trans Tech Publications, www.ttp.net.

(ID: 114.79.12.33-16/10/15,19:43:07) Key Engineering Materials Vols. 675-676 425 426 Applied Physics and Material Applications II Key Engineering Materials Vols. 675-676 427 428 Applied Physics and Material Applications II Key Engineering Materials Vols. 675-676 429 Keywords: Optical properties, Nd3+, Borate glass Abstract. The present paper reports the structural (XRD and FTIR spectra), physical (like density, molar volume, refractive index etc.), optical absorption and emission properties of Nd3+ doped sodium lead zinc lithium borate glass.

The glasses with chemical formula (65-x)B2O3-15Na2O- 10PbO-5ZnO-5Li2O -xNd2O3 (where x = 0, 0.05, 0.1, 0.5, 1.0, 2.0 and 4.0) have been prepared using the melt-quenching method. XRD characterization shows that the glasses are amorphous nature. The values of measured absorption transitions obtained ten transitions and the hypersensitive transition at 4I9/2 ? 2G7/2+4G5/2.

To excite photon from ground state to upper state, used two different wavelength spectra at 582 and 804 nm. The maximum emission intensity of the 4F3/2 ? 4I11/2 transition obtained when Nd3+ ion content in the borate glass is 1.0 mol %. It is indicating that Nd3+ doped borate glass have self-quenching or concentration quenching.

Introduction Investigation of the physical and optical properties of borate glass doped rare earth (RE) ions have been widely reported at the past several years. Neodymium (Nd3+) is one of the rare-earth ions for generate of the hypersensitive light emission at 1060 nm corresponding to the 4F3/2 ?4I11/2 transition [1-2]. The Nd3+ ion doped glasses such as phosphate, borate, silicate and tellurite can be applicated for optical amplifier, solid state laser, optical communication, optical waveguide and storage data optically [3,4].

Borate glass as a host matrix of the Nd3+ ion has been widely employed since high transparency, high density, appropriate bandwidth, suitable for infrared transmission, high mechanical stability, corrosion resistivity and can be doped by Nd3+ ions with high concentrations [3,5]. The optical properties like absorption and luminescence spectra of the Nd3+ ions are influenced by ingredients present in the glasses.

On the other hand, a higher Nd3+ concentration is needed to obtain the amplification of the emission light [3]. Therefore, in this work the dopant Nd3+ ions are investigated up to 4.0 mol% to obtain energy transfer process caused by the interaction of the Nd3+ ions. The addition of sodium oxide in the glasses compositions can increase the intensity absorption due to the optical density in the ultraviolet wavelength longer than the absorption edge [6].

Moreover, alkali ion serves as network modifier for forming of non bridging oxygen (NBO) so that the amounts of the cation coordination in the network are increase[7]. Emission properties and non-radiative transition of the rare earth ion mostly affected by lead oxide glasses because Pb2+ ions have a role to change the RE environment network [8].

Besides to increase the refractive index, borate glasses containing heavy metal oxides (HMO) also reduce phonon energy therefore the non-radiative transition decreases. In addition, borate and HMO Key Engineering Materials Vols. 675-676 (2016) pp 424-429 Submitted: 2015-06-11 © (2016) Trans Tech Publications, Switzerland Revised: 2015-08-10 doi:10.4028/www.scientific.net/KEM.675-676.424 Accepted: 2015-08-18 All rights reserved.

No part of contents of this paper may be reproduced or transmitted in any form or by any means without the written permission of Trans Tech Publications, www.ttp.net. (ID: 114.79.12.33-16/10/15,19:43:07) glasses doped Nd3+ ion are offer a higher quantum efficiency because of the larger radiative transition probabilities and smaller multiphonon decay rates [9,10].

As a comparison, the optical properties of the borate glasses doped Nd3+ ions have been reported such as BINLAB glasses [3], BBS glasses [11], BZBNd glasses [12], lithium cesium mixed alkali borate glasses [13] and also in Bi2O3-BaO-B2O3 glass system [14]. In view of this work, we investigate the physical and optical properties of Nd3+ ions in sodium lead zinc lithium borate glasses.

Especially for optical properties, we focus on absorption and fluorescence properties of the Nd3+ glasses. Furthermore, these results were compared with the earlier paper. Experimental Glasses with the chemical composition (in mol%) of (65-x)B2O3-15Na2O-10PbO-5ZnO- 5Li2O -xNd2O3 (where x = 0, 0.05, 0.1, 0.5, 1.0, 2.0 and 4.0) are powders form and high purity (? 99.5 %). Appropriate amounts of compounds are weighed and systematically mixed in alumina crucible.

The glasses medium are prepared by melting quenching method at the temperature 1000 oC for 3 h in an electric furnace. The homogeneous of the ingredients were achieved by shaken of the crucible. The molten form of the glass was quenched on the stainless steel mold plate for desired shape.

Concurrently, the glass material was annealed in the other furnace at 450oC during 3 h. Subsequently, for decreasing thermal stresses inside of the glass, the sample cooled to room temperature with the rate of reduction is 64oC per hour. Finally, the glasses were cut in proper size 20 x 10 x 3 mm3 and polished for optical characterization.

The absorption spectra of these glasses were measured in the wavelength range from 300 to 1000 nm using a Shimadzu UV-Vis NIR Spectrophotometer with UV-3600 model. Fluorescence spectra were recorded in the range 800–1600 nm by a Quanta Master (QM-500) NIR system from Photon Technology International (PTI) with 582 nm and 804 nm excitation wavelength selected.

Light source from 75 W Arc Xenon lamp was dispersed by 1200 line/mm excitation monochromator and emission light dispersed by 600 line/mm with 300 mm focal length respectivelly. The Emission signal were detected by InGaAs detector equiped with the TE-cooled. All the instruments setup and measurements were carried out at room temperature.

Results and discussion The studies of X-ray diffraction were carried out by using LabX XRD-6100 Shimadzu diffractometer operating with radiation source from a Cu-target tube (V = 40.0 kV and I = 30.0 mA) at room temperature. The spectrum patterns of powder Nd3+ ion doped borate glasses system are shown in Fig.1a. Fig. 1. Structure

pattern of B2O3-Na2O-PbO-ZnO-Li2O -Nd2O3 observed by (a) XRD and (b) FTIR The patterns were not contained any sharp peaks confirms the amorphous and absence of crystalline nature of the prepared glasses.

The observed spectrums are very similar to each other of the glasses and there are no appear any discrete peak except presence of broad humps around 2?=26o. The 20 40 60 80 Intensity (a.u) 2?(deg) BNPZLiN1 BNPZLiN2 BNPZLiN3 BNPZLiN4 BNPZLiN5 BNPZLiN6 1400 1200 1000 800 600 Transmittance (a.u) Wavenumber (cm-1) BNPZLiN1 BNPZLiN2 BNPZLiN3 BNPZLiN4 BNPZLiN5 BNPZLiN6 (a) (b) Key Engineering Materials Vols.

675-676 425 patterns of diffraction spectrum in this study were similar to that reported earlier work [15,16]. Fig. 1b shows the infrared transmittance spectra in the range 600 – 1400 cm-1 for BNPZLiN glasses. As seen from the Fig.1b, the FTIR spectra of the BNPZLIN1 to BNPZLIN6 consists of several peaks which are broad in bandwidth.

The main active absorption spectra in infrared range of sodium borate network are defined to consist of three disparate regions. The first region is absorption band at wavenumber 1200-1700 cm-1 attributed to the presence of bending vibration due to BO3 groups. The second absorption groups from 800 to 1200 region cm-1 attributed to the B-O bond stretching due to tetrahedral (BO4) groups.

The next region around 400 to 800 cm-1 attributed to the stretching vibration due to B-O-B (BO3) linkages in borate groups. In the presents glass system have three broad band vibrations in the IR spectra regions are centered at 837–864 cm-1, 1021–1061 cm-1 and at 1179–1192 cm-1. BNPZLiN3 glass shows peak at 1313 cm-1 which is attributed to the B(3)-OB(4) stretching vibrations in the glass system.

The broad band peak around 1021 to 1061 cm-1 in all present glasses due to stretching vibration of tetrahedral BO4 units present in the glass network [17]. The absorption band centered at wavenumber 837-864 cm-1 in the present glasses are appear from a few and limited concentration of Nd2O3 [18]. Absorption spectra The optical absorption spectra of Nd3+ doped borate glass referred as BNPZLi, BNPZLiN1, BNPZLiN2, BNPZLiN3, BNPZLiN4, BNPZLiN5 and BNPZLiN6 recorded at room temperature in the wavelength range 300-1000 nm is shown in Fig. 2a.

The absorption spectra exhibit ten sensitive bands derived from the ground state 4I9/2 to various excited states. The absorption peaks centered at 354 nm, 430 nm, 475 nm, 525 nm, 582 nm, 626 nm, 680 nm, 751 nm, 804 nm and 875 nm which are assigned to 2D5/2, 2P1/2, 2G9/2, 4G7/2, 2G7/2+4G5/2, 2H11/2, 4F9/2, 4F7/2, 4F5/2, 4F3/2

transitions respectively.

These spectra were identified by comparison with other found for Nd3+ doped glass systems [11-15]. The transitions which are rare earth ions sensitive to the environment called as hypersensitive transitions. From the optical absorption spectra in Fig. 2(a) absorption band 4I9/2 ? 2G7/2+4G5/2 centered at 582 nm is the hypersensitive transition. Fig.

2(a) Absorption spectra and (b) 4I9/2 ? 2G7/2+4G5/2 hypersensitive transition for Borate glasses system doped with variation of Nd3+ ion (mol%) The absorption profiles of the 4I9/2 ? 2G7/2+4G5/2 hypersensitive transition in various Nd3+ ion concentration is shown in Fig. 2b. It is observed that absorption peaks intensity increase with increasing Nd3+ ion content in the glass composition.

The absorption intensity maximum occur at BNPZLiN6 glass which Nd3+ ion concentration is 4.0 mol%. Among the absorption spectra, 2G7/2+4G5/2 (582 nm) and 4F5/2 (804 nm) transitions are more intense and they are used as excitation BNPZLi BNPZLiN1 BNPZLiN2 BNPZLiN3 BNPZLiN4 BNPZLiN5 BNPZLiN6 0.0 0.2 0.4 0.6 0.8 1.0 Absorption Int. (a.u) Glasses initial hypersensitive transition (a) (b) 300 400 500 600 700 800 900 0 5 10 15 20 Absorption cross-section (10-22 cm2) wavelength (nm) BNPZLiN1 BNPZLiN2 BNPZLiN3 BNPZLiN4 BNPZLiN5 BNPZLiN6 2 D5/2 4 I9/2? 2 P1/2 2 G9/2 4 G7/2 2 G7/2, 4 G5/2 2 H11/2 4F9/2 4 F7/2 4 F5/2 4 F3/2 426 Applied Physics and Material Applications II source to observe the emission spectra for all the prepared glasses.

The optical absorption edge of nonmetallic materials is interpreted in terms of indirect transitions across an optical band gap. The absorption coefficient ?(?) as a function of photon energy incident (h?) for an amorphous material was proposed by Mott and Davis and given by [19]: ( )n v hv = hv ? Eg 0 ? ( ) ? (1) Where Eg is the optical energy band gap.

The exponent index n = ½ and n = 2 corresponding to an allowed direct transition and indirect transition respectively. ?0 is a constant related to the band tailing parameter. The optical band gap values were obtained by extrapolating from the linear line of the (hv) plots with (?hv)1/2 = 0 for indirect transitions and (?hv)2 = 0 for direct transitions. The Eg magnitudes for indirect transition were estimated by Tauc's plot and shown in Fig. 3.

As can be seen that the optical band gap values for indirect transition almost the same for the first three glasses i.e., BNPZLi, BNPZLiN1 and BNPZLiN2 (around 2.8 eV). Whereas

for BNPZLiN3, BNPZLiN4, BNPZLiN5 and BNPZLiN6 glasses have the optical band gap around 3.2 eV. The left inset at Fig. 3 shows that the indirect band gap (Eg) increase from BNPZLiN2 glass upto BNPZLiN6 glass with decrease in B2O3 content in the host.

Furthermore, for the direct band gap shows increase from BNPZLi to BNPZLiN1 and then decrease upto BNPZLi6. Fig. 3. Plot of optical energy band gap for indirect allow transition with various Nd3+ doped borate glasses. Luminescence spectra Fig.4a and b shows the NIR emission spectra (800-1600 range) for all the glass samples were recorded at room temperature.

The glasses were excited using two different wavelengths of 582 nm and 804 nm by a xenon light source. From these emissions spectra, the emission peak wavelengths (?p) were obtained at 904 nm, 1064 nm, 1336 nm corresponding to the band 4F3/2 ? 4I9/2; 4F3/2 ? 4I11/2 and 4F3/2 ? 4I13/2 respectively. The main peak centered at 1064nm assigned to the 4F3/2 ? 4I11/2 transition is the same with some literature [3,9,12].

The spectrum shapes of emission transitions for all of glass samples (from BNPZLiN1 to BNPZLiN6) are the same, however the emission intensities are different from one glass sample to another glass sample. 0,0 1,0 2,0 3,0 4,0 5,0 6,0 2,5 2,7 2,9 3,1 3,3 3,5 3,7 3,9 (?h?)1/2 (cm-1/2eV1/2) Indirect optical energy band gap, h? (eV) BNPZLi BNPZLiN1 BNPZLiN2 BNPZLiN3 BNPZLiN4 BNPZLiN5 BNPZLiN6 3.3 3.4 3.5 3.6 3.7 BNPZLi BNPZLiN1 BNPZLiN2 BNPZLiN3 BNPZLiN4 BNPZLiN5 BNPZLiN6 2.8 3.0 3.2 3.4 3.6

Direct Optical band gap (hv) Indirect Key Engineering Materials Vols. 675-676 427 Fig.4. Luminescence spectra of Nd3+ ions in borate-based glass with (a) ?exc = 582 nm and (b) ?exc = 804 nm The emission intensity of the 4F3/2 ? 4I11/2 transition increases in the order of BNPZLiN1 < BNPZLiN2 < BNPZLiN3 < BNPZLiN4 and decrease in the order of BNPZLiN4 > BNPZLiN5 > BNPZLiN6.

This phenomenon occurs for both of excitation types, as shown in Fig. 7 and 8. Fig. 9 shows that the intensity increase and achieve a maximum for 1.0 mol% Nd3+ (BNPZLiN4) and drastically decreases with an increase of the Nd3+ ion composition. This result indicates that for the composition of the Nd3+ ion doped B2O3-Na2O-PbO-ZnO-Li2O more than 1.0

mol%, the luminescence will be quenched by energy transfer processes due to interaction among Nd3+ ions [11]. The reduction of luminescence intensity at higher Nd3+ content in glass system can be associated to Nd3+ self-quenching (concentration quenching) due to cross-relaxation processes. It can be improved by the displacement of excitation energy between the active ions [20].

In consequence self-quenching occurs in Nd3+ doped sodium lead zinc lithium borate glasses. Several radiative properties like stimulated emission cross section, fluorescence lifetime and quantum efficiency are needed to determine this glass as a good candidate of laser material. Summary The structural, physical, optical absorption and emission properties of Nd3+ doped borate glass system have been investigated.

FTIR studies shows that the broad band vibrations in the IR spectra centered at 837–864 cm-1, 1021–1061 cm-1 and at 1179–1192 cm-1. The amorphous structure was investigated by X-Ray diffraction and obtained non crystalline profile. The values of measured absorption transitions obtained ten transitions and the hypersensitive transition at 4I9/2 ? 2G7/2+4G5/2.

To excite photon from ground state to upper state used two different wavelength spectra, 582 and 804 nm respectively. The maximum emission intensity of the 4F3/2 ? 4I11/2 transition obtained when Nd3+ ion content in the borate glass is 1.0 mol %. It is indicating that Nd3+ doped borate glass have self-quenching or concentration quenching.

The radiative properties experimental of Nd3+ doped sodium lead zinc lithium borate glasses will determine at future work. Acknowledgement This work was partly supported by Research Grant of Asahi Glass Foundation contract No.1746d/I1.C01/PL/2014. 800 900 1000 1100 1200 1300 1400 1500 0.0 0.5 1.0 1.5 2.0 ? exc = 582 nm 4F 3/2 ? 4 ? 13/2 4F 3/2 ? 4 ? 9/2 Emission Intensity (a.u) Wavelength (nm) BNPZLiN1 BNPZLiN2 BNPZLiN3 BNPZLiN4 BNPZLiN5 BNPZLiN6 4F 3/2 ? 4 ? 11/2 800 1000 1200 1400 1600 0.0 0.1 0.2 0.3 0.4 0.5 0.6 0.7 0.8 0.9

Emission Intensity (a.u) Wavelength (nm) BNPZLiN1 BNPZLiN2 BNPZLiN3 BNPZLiN4 BNPZLiN5 BNPZLiN6 ? exc = 804 nm 4F 3/2 ? 4 ? 9/2 4F 3/2 ? 4 ? 11/2 4F 3/2 ? 4 ? 13/2 0.3 0.6 0.9 1.2 1.5 1.8 2.1 BNPZLiN6 BNPZLiN5 BNPZLiN4 BNPZLiN3 BNPZLiN1 Emission Intensity (a.u) Glass Samples Exc. 582 nm Exc.

804 nm BNPZLiN2 (a) (b) 428 Applied Physics and Material Applications II References [1] M.J. Weber, Science and technology of laser glass, J. Non-Cryst. Solids. 123 (1990) 208–222 [2] J. Li, Y.Z. Mei, C. Gao, F. Ren, A.X. Lu, Variation of luminescence properties of Na2O– CaO–SiO2: Nd3+ glass with crystallinity, J. Non. Cryst. Solids. 357 (2001) 1736–1740 [3] K.V. Kumar, A.S. Kumar, Spectroscopic properties of Nd3+ doped borate glasses. Opt. Mater. (Amst). 35 (2012) 12–17 [4] F.

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