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Spectroscopic properties of alumina borosilicate glasses with alkaline oxides doped with Sm_2O_3 for display laser emission

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Abstract

This research investigates the influence of the concentration of Sm^{+3} ions on the features of a new borosilicate glass system with the composition $50\text{B}_2\text{O}_3$ – 14SiO_2 – $20\text{Li}_2\text{O}$ – $15\text{Na}_2\text{O}$ – $1.0\text{Al}_2\text{O}_3$ – $x(\text{Sm}_2\text{O}_3)$, where $x= 100, 200, 300, 400$, and 500 ppm. The glasses are characterized by XRD, FTIR, UV- Vis- NIR, and photoluminescence spectra. The XRD results proved that all prepared glass has amorphous nature. Moreover, it was found that the value of density, ρ , and refractive index, n , increase with increasing of Sm^{+3} content. Otherwise, the value of molar volume, optical energy gap, and boron-boron distance decrease by increasing sm^{+3} concentration. The structure of glasses studied was investigated by computing Internuclear (r_i), Polaron radius (r_p), field strength (F) and the deconvolution of FTIR spectra. Judd-Ofelt theory applied to clarify the structural changes and nature of the bonds of prepared glasses. Moreover, A spectroscopic quality factor, the branching ratio (β_r), and lifetime (τ) are calculated via the parameters of Judd-Ofelt. The color chromaticity of the present glasses evidenced that the emission was in the white-reddish orange region under 402nm excitation depended on the concentration of Sm^{3+} ions incorporated into the host glasses. From photoluminescence emission, stimulated emission cross-section, and CIE chromaticity leads to these glasses are promising for light-emitting diode (LED) as; laser material in a wide range.

Keywords: Borosilicate glasses; UV-Vis-NIR spectra; Physical parameters; Spectroscopic properties; Laser emission.

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1. Introduction

Borosilicate glass has attracted the researcher's attention due to its superior properties compared to ordinary glass. It has a very weak thermal expansion coefficient equals to $\sim 3 \times 10^{-6} \text{ K}^{-1}$ at 20°C ^[1] such glass doesn't suffer from thermal shock. It can be used in reflecting telescope manufacturing.^[2] Also, this glass helps to fabricate precise lenses. Borosilicate glass has a vital contribution to immobilizing and disposal of radioactive waste because it is distinguished by its chemical resistance.^[3] Borosilicate glass consists of different oxides such as B_2O_3 , SiO_2 , Na_2O , Li_2O , and Al_2O_3 . The main former of borosilicate glass is B_2O_3 . Glass which is containing borates has special attention because of its promising linear and nonlinear optical properties^[4]. The increase of B_2O_3 content enhances chemical resistance and improves transparency. Since borate glasses own high phonon energies which may hold back the efficiency of their luminescence during various relaxations^[5]

The addition of silica to borate glass reduces the thermal expansion coefficient and enhances the melting point and the working point. The acid stability of the glass decreases as the silica concentration decreases. Another component of borosilicate glass is Alkali metal oxide. It cooperates with silica to reduce the melting temperature and enhancement of low thermal expansion.^[6] Alkali Metal oxide has an important role in the crystallization process, combining two metal oxide enhanced the resistance toward unwanted crystallization.^[7] Therefore, alkaline earth cations can be added to the borate glass to upgrade their optical properties, impact the network connectivity, arranging of modifier cations and modify chain bonding in glasses by replacing the bridging oxygen with non-bridging oxygen^[5]. Li^+ ions improve the acid-resistance of glass by forming bonds with glass networks^[7]. Other oxides such as aluminum trioxide as a modifier of glass (Al_2O_3), have an important role in the fluidity of the glass as increasing of alumina decreasing the fluidity of glass^[7]. Alumina also enhances the temperature of glass processing^[8], enhancing the glasses' stability^[9], affect the lasing and thermo-mechanical properties of the products in which they have been associated^[9]. Interestingly, borate glasses act as suitable host materials for exploring both the nature and structure of the luminescence and paramagnetic centers^[10]. In the recent three decades, doped materials with trivalent rare earth (RE^{3+}) ions are extensively used for the upgrading of several optical devices, e.g. waveguides, up-conversion lasers, optical fibers, detection of infrared radiation, optical amplifiers, display devices, sensors, and light converters.^[11-12]. The optically active f-f electronic transitions of lanthanides ions generate sharp emission lines when integrated into different glass matrices^[11-5]. Owing to high thermal stability, promising optical, structural, and electrical properties low melting point, good RE^{3+} ion solubility, and

high transparency, ^[11]. The importance of Glass containing Sm^{+3} is delivered from its visible emissions through different channels of emission ^[9]. The luminescence spectrum of Sm^{+3} ions in glasses and crystals shows distinct green, orange, and red emission bands, which refer to the $^4\text{G}_{5/2} \rightarrow ^6\text{H}_{5/2}$, $^4\text{G}_{5/2} \rightarrow ^6\text{H}_{7/2}$, and $^4\text{G}_{5/2} \rightarrow ^6\text{H}_{9/2}$ emission transitions, respectively that can be used in fluorescent display devices, visible lasers, new light sources, and UV sensor ^[10]. In this work, borosilicate glass which was doped with Sm^{3+} has been prepared with the chemical formula $(50\text{B}_2\text{O}_3-14\text{SiO}_2-20\text{Li}_2\text{O}-15\text{Na}_2\text{O}-1\text{Al}_2\text{O}_3-x\text{Sm}_2\text{O}_3)$, where $x=100, 200, 300, 400$, and 500 ppm. The objectives of this work are (i) To synthesis borosilicate glasses doped with Sm^{3+} with different concentration of Sm_2O_3 by usual melt quenching technique, (ii) To characterize the glass samples by XRD, FTIR, UV, and PL, (iii) To compute and measure the optical and physical parameters with different concentration of Sm_2O_3 . (iv) To investigate optical properties which include the absorption and luminescence spectra. By the use of emission spectra of Sm^{3+} ions the color coordinates were evaluated and were presented in the CIE color diagram.

2. Experimental

The glass samples with composition of $50\text{B}_2\text{O}_3-14\text{SiO}_2-20\text{Li}_2\text{O}-15\text{Na}_2\text{O}-1.0\text{Al}_2\text{O}_3$ sample code (BSLNA0), $50\text{B}_2\text{O}_3-14\text{SiO}_2-20\text{Li}_2\text{O}-15\text{Na}_2\text{O}-1.0\text{Al}_2\text{O}_3-100\text{ppm}$ Sm_2O_3 , sample code (BSLNA1), $50\text{B}_2\text{O}_3-14\text{SiO}_2-20\text{Li}_2\text{O}-15\text{Na}_2\text{O}-1.0\text{Al}_2\text{O}_3-200\text{ppm}$ Sm_2O_3 , sample code (BSLNA2), $50\text{B}_2\text{O}_3-14\text{SiO}_2-20\text{Li}_2\text{O}-15\text{Na}_2\text{O}-1.0\text{Al}_2\text{O}_3-300\text{ppm}$ Sm_2O_3 , sample code (BSLNA3), $50\text{B}_2\text{O}_3-14\text{SiO}_2-20\text{Li}_2\text{O}-15\text{Na}_2\text{O}-1.0\text{Al}_2\text{O}_3-400\text{ppm}$ Sm_2O_3 , sample code (BSLNA4) and $50\text{B}_2\text{O}_3-14\text{SiO}_2-20\text{Li}_2\text{O}-15\text{Na}_2\text{O}-1.0\text{Al}_2\text{O}_3-500\text{ppm}$ Sm_2O_3 , sample code (BSLNA5). These glasses were synthesized and prepared by standard melt quenching technique. High-quality powders of lithium carbonate Li_2CO_3 (99.5%, EUROMEDEX), sodium carbonate Na_2CO_3 (99%, Chemajet), boric acid H_3BO_3 (99.98%, Sigma-Aldrich), acid-washed quartz sands SiO_2 (99.9 %, Sigma-Aldrich), aluminum oxide Al_2O_3 (99%, Sigma-Aldrich) and Samarium Oxide Sm_2O_3 (purity 99.9%, Sigma-Aldrich) were weighed by using a digital balance and mixed well, then charged into platinum crucible followed by melting in an electric furnace at 1050°C for 20 minutes. Then we poured melts onto a steel plate and annealed them in a furnace at 600°C for 3 hr to get rid of the thermal strains. The prepared glasses are named BSLNA0, BSLNA1, BSLNA2, BSLNA3, BSLNA4, and BSLNA5, respectively as found in Table 1. XRD was carried out to investigate the amorphous network of the prepared glass samples. BRUKER X-Ray Powder Diffraction

system was used in this test. The range of investigation was (0-100) 2θ with a radiation source of Cu ($\lambda=1.5406^\circ\text{A}$). An optical glass surface was obtained after polishing each sample surface with fine Al_2O_3 powder. The thickness of each sample was measured by a micrometer. Then the UV–vis-NIR absorption spectra were measured in the range of 200-2500 nm at ambient condition by spectrophotometer JASCO Corp, v-570, Rel-00 Japan with the double beam. The Photoluminescence spectra were recorded by using fluorescence spectrometer Ls 55 Perkin Elmer in the range of 300–900 nm at room temperature. FTIR was used to study the structural changes of each sample of glass after doping with different concentrations of Sm_2O_3 . FTIR measurement in the range of 350–4000 cm^{-1} was occurred by BX Perkin Elmer equipment using KBr pellets.

3. Results and Discussion

Figure 1 demonstrates the XRD data of the prepared glass sample from BSLNA0 to BSLNA5. The XRD data confirmed the amorphous nature of glass by exhibiting broad diffused peaks. Moreover, no significant sharp peaks appeared in the data and the atoms of glass showed random distribution in the glass network, furthermore large variation in interatomic distances [9].

The corresponding glass samples density were measured at ambient condition via Archimedes concept. The density of glass calculated by using Eq. [1],

$$\rho = \frac{W_{air}}{W_{air}-W_{liquid}} \cdot \rho_{liquid} \quad (\text{g/cm}^3) \quad [13] \quad [1]$$

Where, ρ , is a glass sample density (g/cm^3), W_{air} is the weight of prepared glass in the air, W_{liquid} is the weight of glass in toluene and ρ_{liquid} is a density of toluene (0.864 g/cm^3). The molar volume of glass samples was obtained by implementing the Eq. [2],

$$V_m = \frac{M_{wt \text{ sample}}}{\rho_{sample}} \quad (\text{cm}^3/\text{mol}) \quad [13] \quad [2]$$

Since V_m is the molar volume of the glass sample (cm^3/mol) and M_{wt} is the molecular weight of the sample. The M_{wt} of the sample (g/mol) could be obtained by Eq. [3], as well as oxygen packing density (OPD) (g.atom/L) could be obtained by applying for the Eq. [4],

$$M_{wt} = \frac{50\text{B}_2\text{O}_3 + 14\text{SiO}_2 + 20\text{Li}_2\text{O} + 15\text{Na}_2\text{O} + 1\text{Al}_2\text{O}_3 + x\text{Sm}_2\text{O}_3}{100} \quad (\text{g/mol}) \quad [3]$$

$$OPD = \frac{[\rho \cdot C]}{M_{wt}} \cdot 1000 \quad (\text{g. atom/L})^{[13]} \quad [4]$$

Since, C, is the oxygen atoms number which is existing in the structure of each glass sample. As found in Table 2, the density of glass samples was enhanced from 2.397 to 2.625 g/cm³ by increasing the Sm₂O₃ concentration from 0 to 500 ppm. The increase in density as a result of increasing OPD from 78.3 to 85.61 g.atom/L. Thus, the glass samples become denser and more compact ^[14], another factor that increased the density of glass samples is the alteration of [BO₃]⁻³ triangles into [BO₄]⁻⁴ tetrahedral by the gradual increase of Sm³⁺ ions in glass samples. ^[15]

As a result of Sm₂O₃ concentration increasing in glass sample, the molar volume decreased from 27.58 to 25.24 (cm³/mol), we ascribed this observation due to the decrease which occurs for both interatomic distances and the length of bonds between doped glass atoms. Such results enhanced the compactness of the structure. ^[9] The average boron-boron separation is also calculated by Eq. 5 to clarify the effect of modifier concentration on the glass system.

$$d^{(B-B)} = (V_M^B/N_A)^{1/3} \quad (5)$$

where;

$$V_M^B = V_M/(1 - X_B)$$

Where V_M^B and X_B is molar volume and mole fraction of B₂O₃ respectively.

In the present work, by increasing Sm₂O₃ concentration, the boron-boron separation, $d^{(B-B)}$, value decreased as shown in Table 2. Noticeably, the previous results confirmed that, by increasing the concentration of Sm₂O₃, the molar volume and the interatomic spacing decrease which enhanced the compactness of glass. ^[9]

Ultraviolet-visible-near infra-red (UV-vis-NIR) optical absorption of Sm³⁺-doped BSLNA at ambient condition shows nine absorption peaks as shown in Fig. 2. These peaks result from electronic transitions from the ⁶H_{5/2} level to the further excited states of the Sm³⁺ ion. These absorption peaks appear at 435, 484, 950, 1094, 1253, 1402, 1505, 1536, and 1563nm due to transitions ⁶H_{5/2} to ⁴F_{7/2}, ⁴I_{13/2}, ⁶F_{11/2}, ⁴F_{9/2}, ⁶F_{7/2}, ⁶F_{5/2}, ⁶F_{3/2}, ⁶H_{15/2} and ⁶F_{1/2} respectively ^[16-19].

The value of indirect optical band gap, E_{opt}^{in} , of prepared glasses can be calculated from the following Eq. [6] ^[20].

$$\alpha(\nu) = \frac{\alpha_o (h\nu - E_{opt}^{in})^m}{h\nu} \quad [6]$$

Where, $\alpha(v)$, is the optical absorption coefficient ^[22]. By employing the Eq. [7] absorption coefficient (α) can be calculated by Eq. [7].^[12]

$$[7]\alpha(v) = \frac{1}{t} \ln \frac{I_0}{I_t}$$

The thickness of the glass samples is t and the intensity of incident light is I_0 , while the intensity of transmitted light is I_t . Light energy is $h\nu$, α_0 is an energy-independent constant, $m=2$ for the allowed indirect transition. Tauc's plot is shown in Fig 3. for indirectly allowed transitions. E_{opt}^{in} can be determined by deriving a linear portion of the curve to the X_{axis} at $Y_{axis}=0$. The values are monitored and found in Table 3. As is evident, E_{opt}^{in} , values were reduced from 3.61 to 2.0 eV by increasing Sm_2O_3 concentration from 0 to 500ppm ^[20]. If the oxygen bonding (BO) in the glass network, such as non-bridging oxygen forming (NBO), modifies the absorption properties, an increase in the concentration of Sm^{3+} ions can occur. It leads to a high non-bridging oxygen concentration owing to a change in the binding of oxygen. Concerning this increase in the concentration of Sm^{3+} ions, the degree of electron delocalization in the compactness of the glasses is decreased, decreasing the optical energy band difference. This describes why, when Sm_2O_3 content increases, the optical band distance decreases. Where, ΔE , an Urbach energy can be represented as the width of a localized state in the typically forbidden gap. Increasing ΔE values indicate a higher tendency of the disturbance system to change weak bonds into defects. It is calculated by Eq. [8] ^[12].

$$\alpha(v) = \alpha_0 \exp\left(\frac{h\nu}{\Delta E}\right) \quad [8]$$

Where $\alpha(v)$ is the optical absorption coefficient and α_0 is constant ^[13-20]. The values of ΔE can be computed from $\ln(\alpha)$ versus photon energy, ΔE , see Fig. (4), which can be affected by extensions of the valence and conduction bands to the bandgap region.

The glass samples which doped with a different concentration of Sm_2O_3 exhibited increasing ΔE values from 0.36 to 1.37 eV by increasing the concentration of Sm_2O_3 . The present glass sample doped by 500ppm of Sm_2O_3 showed ΔE equaled 1.37 eV, higher than the un-doped glass sample which exhibited ΔE equaled to 0.36 eV. A higher concentration of Sm_2O_3 increases the degree of disordered which results in higher values of ΔE , also the degree of disordered ^[13].

The physical parameters such as; r_i , r_p , F , n , R_m , and α_m confirm the glass structure which is determined by the following Eq. [8],[9],[10],[11]^[12-20]

$$\text{Concentration of ions } N = RE \text{ mol}\% \cdot \frac{N_A \cdot \rho}{\text{glass average molecular wt}} \quad [9]$$

$$\text{Internuclear distance } (r_i) = \left(\frac{1}{N}\right)^{1/3} \quad [10]$$

$$\text{Polaron radius } (r_p) = \frac{1}{2} \left(\frac{\pi}{6N}\right)^{1/3} \quad [11]$$

$$\text{Field strength } (F) = \frac{Z}{r_p^2} \quad [12]$$

Where (N) concentration of Sm^{3+} ions, N_A is Avogadro's number, ρ is the density of glass sample and Z is the atomic mass of samarium ion $Z_{\text{Sm}} = 150.36 \text{ g}\cdot\text{mol}^{-1}$ ^[22], (r_i) mean distance between the Sm^{3+} ions, (r_p) polaron radius and (F) field strength of Sm-O bond in doped BSLNA glass. The values have been calculated and placed in Table 4.

As is clear of calculated values, r_i and r_p values reduce with increasing Sm^{3+} ions. Consequently, the strength of the Sm-O bond increases, so the field strength (F) around Sm^{3+} ions becomes stronger. The combining between these obtained values and the density results confirms the compactness of glass structure with addition Sm^{3+} ions consequently, decreasing in the degree of delocalization of electrons occurs, due to increase in the compactness of glass and then increases the donor centers in the glass matrix and as a result, optical energy band gap decreases.^[20]

The refractive index, n , is one of the most significant properties in optical glasses^[21]. The refractive index values of prepared glass samples were calculated by using Dimitrov and Sakka relation as shown in equation [13]^[22] and obtained in Table 5.

$$\frac{(n^2-1)}{(n^2+2)} = 1 - \sqrt{\frac{E_{opt}^{in}}{20}} \quad [13]$$

The, n , value increases from 2.2497 to 2.736 with the concentration of Sm^{3+} ions increase from 0 to 500ppm in prepared glasses. The value of n depends on ion polarization, electron density, and molar masses. The improvement in the refractive index is due to strong polarizable nonbridging oxygen (NBOs) generation in the glass network. Non-bridging oxygen formation provides greater polarizability over the covalent oxygen bridging bonds by providing a higher refractive index.

The nonlinear response of the materials depends on polarizability. The electronic polarization of the materials causes the optical non-linearity upon exposure to intense light beams [22]. Polarizability is correlated to physical and chemical properties such as optical UV absorption of metal ions and electro-optical effect [23]. The molar refraction is proportionate to molar polarizability, which is associated with the glass sample structure.

Molar refraction (R_m , $\text{cm}^3 \text{mol}^{-1}$) is calculated by applying the following Lorentz–Lorenz Eq. 14:

$$R_m = \frac{n^2-1}{n^2+2} V_m \quad [14]$$

Molar polarizability (α_m) can be calculated by the Eq. [15]:

$$\alpha_m = \frac{R_m}{2.52}$$

The values of polarizability (α_m) and molar refraction (R_m) for samarium doped glass samples are listed in Table 3. Also the dielectric constant (ϵ) and refraction loss (R_L) is computed by employing the following equations: [33]

$$\epsilon = n^2 \quad [16]$$

$$R_L = \left(\frac{n-1}{n+1} \right)^2 \quad [17]$$

The increase in the, R_m , α_m , ϵ and R_L be attributed to the increase of the (NBO) in the glasses network structure with increasing Sm^{3+} ions.

Judd-Ofelt concept has been used to get more information about glass structure and also depends on host material [13]. The parameters of Judd-Ofelt was detected by taking the least square fitting between the calculated and experimental oscillator strength (f_{cal} to f_{exp}) and recorded in Table7. F_{exp} of the absorption band was calculated from the Eq. [18]

$$F_{\text{exp}} = \frac{2.303 mc^2}{N\pi e^2} \int \epsilon(\nu) d\nu \quad [13] \quad [18]$$

where m and e are the electron's mass and charge, respectively. N is Avogadro's constant, c is light velocity, the molar absorptivity (cm^{-1}) is symbolized by $\epsilon(\nu)$, and $\int \epsilon(\nu) d\nu$ is the part which is located under the curve of absorption. F_{cal} was determined from the Eq. [19] [13]

$$F_{\text{cal}} = \frac{8 \pi^2 m c \nu}{3 h (2J+1)} \frac{(n^2+2)^2}{9n} * \sum_{\lambda=2,4,6} \Omega_{\lambda} (\phi \| U^{\lambda} \| \phi' J')^2 \quad [19]$$

where m is the electron mass, the wavenumber (cm^{-1}) is symbolized by ν , c is light velocity, h is Planck's constant, the refractive index is symbolized by n , the Lorentz local electric field correction is equal $(n^2+2)^2/9n$, Judd-Ofelt parameters are symbolized by $\Omega\lambda$ where ($\lambda=2, 4, 6$), the ground state angular momentum is symbolized by J , the excited state is symbolized by ϕ ^ J' , the ground state is symbolized by ϕJ , and $\|U^\lambda\|^2$ refers to squared doubly reduced matrix elements of the tensor operator. Parameters of Judd-Ofelt are significant to know more around rare earth environments, the Internal structure of glass, the nature of bonds and tell us more about the relation between Sm^{3+} ions and the host material ^[13]. Ω_2 symbolizes changes in internal structure, the nature of bonds, asymmetric, and covalent bonds between rare earth ions and surrounding ligands ^[13]. Ω_4 and Ω_6 related to the rigidity and viscosity of prepared glass ^[13]. The higher values of Ω_4 and Ω_6 in our glass system confirm the high rigidity and high viscosity of this glass ^[13]. By calculating the ratio between Ω_4 and Ω_6 parameters the spectroscopic quality factor X will be obtained, which tells us about the quality of prepared glass samples. In our glass system X -factor (> 1) confirms the high quality and stability of this glass, therefore, it can be used in optical devices development ^[13]. It is observed from Table (7) that the parameters of Judd-Ofelt show the trend as $\Omega_4 > \Omega_2 > \Omega_6$ for all prepared glass samples except the BSLNA4 sample it shows the trend $\Omega_4 > \Omega_6 > \Omega_2$. The same trend has been observed in the reported Sm^{3+} doped borate glasses ^[11]. In the two upper cases, the higher magnitudes of Ω_4 confirm the high rigidity of prepared glass samples ^[13]. Lower magnitudes of Ω_2 are due to distortion that results from the change in structure due to the large size of modifier and strong Sm-O bond^[13] which confirmed the high degree of symmetry and strength of ionic bond nature around the Sm^{3+} ions^[13]. Also, we calculated the root mean square (r.m.s) by equation [20]. To estimate the quality of the fit between the two oscillator strengths f_{cal} and f_{exp} .

$$\text{r. m. s.} = \left(\sum_p \frac{(f_{\text{cal}} - f_{\text{exp}})^2}{p-3} \right)^{1/2} \quad [20]$$

Where p is the number of observed transitions on the absorption spectrum.

The lower values of r.m.s. Is evidence that fitting quality Table (5).

Branching ratio describes the probability of stimulated emission for a specific transition, it is a significant factor in laser manufacturer and it has been used to predict the intensities of radiative emission lines that result from an excited state ^[13]. β_r is calculated from the equation [21] ^[13]

$$\beta_r = \frac{A_{\text{Rad}}}{A_T} \quad [21]$$

Where A_{Rad} is the probability of transitions from excited bJ' to lower state aJ for electric dipole spontaneous emission and it is obtained from Eq. [22], A_T symbolizes to total possibilities of all transitions. The transition which $\beta_r \geq 0.50$ is extra distinct for laser radiation [13]. From table 8, it is observed that ${}^6\text{H}_{5/2} \rightarrow {}^6\text{F}_{1/2}$ emission, shows the highest values of branching ratio ($\beta_r \geq 0.50$) so, it can be used for a laser manufacturer. Radiative lifetime τ is not affected by phonon energy of the host matrix but it primarily depends on the total probability of spontaneous transition [13].

$$A_R = \frac{64\pi^4 \nu^3 n(n^2+2)^2 e^2}{27hc^2 (2J+1)} \left[\sum_{\lambda=2,4,6} \Omega_{\lambda} (aJ \| U^{\lambda} \| bJ')^2 \right] \quad [22]$$

The absorption cross-section σ_{ab} of Sm^{+3} doped BSLNA glass obtained by the equation [23] [13]

$$\sigma_{\text{AB}} (\lambda) = \frac{2.303 \text{ OD}(\lambda)}{NL} \quad [23]$$

The cross-section absorbance is symbolized by σ_{ab} , $\text{OD}(\lambda) = \log I_0/I$, which refers to the experimental absorption spectrum optical density, N refers to (Sm^{3+} ions) concentration, and the sample thickness is symbolized by L . The emission cross-section σ_{EM} of Sm^{+3} doped BSLNA glass is obtained by the equation [24] [13]

$$\sigma_{\text{EM}} (\lambda) = \sigma_{\text{AB}} (\lambda) \frac{Z_L}{Z_u} e^{\left(\frac{E_{\text{ZL}} - hc\lambda^{-1}}{K_B T} \right)} \quad [24]$$

Z_u refers to partition functions for upper levels, Z_L refers to partition functions for the lower levels, which are involved in the considered optical transition, E_{ZL} is zero-line energy for the movement that occurs between the lower Stark sublevels of both emitting and receiving multiples [13]. Figure 5 shows the cross-sections of both absorption and emission. The σ_{EM} values for Sm^{+3} -doped glass in the range of 100ppm to 500ppm were about 2.9×10^{-21} , 2.17×10^{-21} , 1.92×10^{-21} , 3.24×10^{-21} , 2.25×10^{-21} , respectively. Also, the gain cross-section was obtained from the absorption and emission cross-section by the Eq. [25] [13].

$$\sigma_{\text{gain}} = P\sigma_{\text{EM}}(\lambda) - (1 - P) \sigma_{\text{AB}}(\lambda) \quad [25]$$

Where σ_{gain} , σ_{EM} , and σ_{AB} are cross-sections of gain, for both emission and absorption. Finally, the population inversion is symbolized by P . As shown in figure 10, at a certain wavelength the gain cross-section has been determined for ($P=0, 0.1, 0.2, 0.3, 0.4, \dots, 1$). It

was observed from Fig. 6 that gain coefficients at 1226nm were 5.01×10^{-21} , 3.7×10^{-21} , 3.2×10^{-21} , 3.4×10^{-21} , 3.7×10^{-21} . The prepared glasses are promising for potential photonic application due to high values of gain coefficient and stimulated emission cross-section.

The measuring of the emission spectra of prepared glass at started by indicating the excitation wavelength 402nm as shown in Figure.7. The spectra of emission of Sm^{3+} doped BSLNA glass show three important peaks centered at 562, 600, and 647 nm, due to transition from $^4\text{G}_{5/2} \rightarrow ^6\text{H}_{5/2}$, $^6\text{H}_{7/2}$, and $^6\text{H}_{9/2}$ transitions of Sm^{3+} ions, respectively [12, 27]. The emission line centered at 600 nm ($^4\text{G}_{5/2} \rightarrow ^6\text{H}_{7/2}$) is the highest intense from all BSLNA glasses doped with Sm^{3+} ions which give orange emission [12] and $^4\text{G}_{5/2} \rightarrow ^6\text{H}_{9/2}$ (647nm) transition gives red emission [12] and shows moderate intensity, the transition at 562 nm $^4\text{G}_{5/2} \rightarrow ^6\text{H}_{5/2}$ shows the lowest intensity between the other two transitions. The glass containing Sm_2O_3 emits bright reddish-orange emission due to the obvious emissions at 562 ($^4\text{G}_{5/2} \rightarrow ^6\text{H}_{5/2}$), 600 ($^4\text{G}_{5/2} \rightarrow ^6\text{H}_{7/2}$), and 647 ($^4\text{G}_{5/2} \rightarrow ^6\text{H}_{9/2}$) nm. As is evident in Figure. 7 the peak intensities enhanced with the increase of Sm_2O_3 concentration from 100 to 400 ppm then the peak intensities decrease at $\text{Sm}_2\text{O}_3 = 500\text{ppm}$. This observable fact is called luminescence quenching. With increasing concentration of Sm_2O_3 the glass matrix becomes crowded by Sm^{3+} ions this reduces the Internuclear distance (r_i) between Sm^{3+} - Sm^{3+} ions also the distance between donor and acceptor centers decreases significantly [5]. Consequently, the excitation energy could be transferred from Sm^{3+} ion to another ion by the interaction between two active ions so the intensity of luminescence decreases [9, 31]. The luminescence intensity of the emission spectral measurements of the prepared glasses has been obtained using 1931 CIE chromaticity diagram. The CIE chromaticity coordinates are found in the range white-reddish- orange range, it is dependent on the concentration of Sm^{3+} ions in the host glass matrix (see Fig. 8).

FTIR spectroscopy is a precious technique to study different functional groups in the glass network structure. FTIR absorption spectra of the prepared glass samples were recorded in the wavenumber range from 400 cm^{-1} to 3600 cm^{-1} at ambient conditions and depicted in Figure.3.a. Deconvolution was applied on FTIR spectra, fourteen characteristic bands were indicated. These bands belonged to functional groups presented in the glass system as illustrated in Fig. (3.b). Table 3 includes the peak positions and their assignments. The peak in the wavenumber range of $415\text{-}422 \text{ cm}^{-1}$ is ascribed to Li_2O [13, 16]. The peak of $432\text{-}444 \text{ cm}^{-1}$ is due to (Na-O) [17]. The peak around $460\text{-}480 \text{ cm}^{-1}$ is ascribed to Si-O-Si bending vibration [18- 19-20]. The peak around $540\text{-}570$ is attributed to the stretching vibration of Sm_2O_3 [21]. The

peak in the wavenumber range of $650\text{-}663\text{cm}^{-1}$ is attributed to asymmetrical bending vibrations (ν_4) of Si-O-Si ^[12]. The peak in the range of $704\text{-}712\text{ cm}^{-1}$ is due to the bending vibration of $[\text{BO}_3]$ units ^[12, 17-19, 22]. The peak in the wavenumber range of $850\text{-}870\text{ cm}^{-1}$ is due to stretching vibrations of tri-, tetra-, and penta borate groups ^[20]. The peak which is lied between $931\text{-}955\text{ cm}^{-1}$ is attributed to B–O stretch in BO_4 units from di-borate groups ^[12]. The peak which is between $1033\text{-}1045\text{ cm}^{-1}$ is due to Sm^{3+} stretching vibration ^[23] The peak which is between $1070\text{-}1115\text{ cm}^{-1}$ is due to the symmetric stretching vibration of Si–O–Si bonds ^[12]. The peak in the range of $1330\text{-}1345\text{ cm}^{-1}$ is attributed to B – O stretch in BO_3 units from varied types of borate groups ^[12]. The peak in the range of $1414\text{-}1426\text{ cm}^{-1}$ is attributed to B–O stretching vibration of BO_3 units in metaborate, pyroborate& orthoborate groups ^[12]. The peak in the range of $1535\text{-}1549\text{ cm}^{-1}$ is due to stretching modes of Si-OH and B–O stretch in BO_4 units from tetrahedral groups ^[24]. The peak in the range of $1650\text{-}1660\text{cm}^{-1}$ is due to the BO^- bond in the isolated pyroborate group ^[12]. The peak of Al^{3+} wasn't indicated in IR absorbance spectra as shown in Fig.1, because the concentration of Al^{3+} was scanty and its signal of vibration frequency was very weak ^[18]. The groups like di, tri, Penta, meta, and boroxol rings with bridging or non-bridging oxygen ions participate in the formation of multi-component borosilicate glasses ^[25]. In the glass network, the area below the curve of BO_4 and BO_3 was calculated. It was observed that the BO_4 ratio in glass samples equal 0.3121, 0.3770, 0.387254, 0.4289, 0.43720, and 0.464381, respectively, when the concentration of Sm_2O_3 increases from 0.00 to 500 ppm. The presence of bridging oxygen was detected by four coordinated BO_4 boron atoms which are a result of the conversion of three coordinated BO_3 units ^[25], also the peak lies in 1426 cm^{-1} shifted to 1414 cm^{-1} . This shifting to lower wavenumber clarifies that the phonon energy of the glass system decreased because it depends on BO_3 units ^[26] which enhanced the efficiency of fluorescence ^[26].

CONCLUSION

From the results which were obtained in the present studies, it can be concluded that; The glass samples of composition $(50\text{B}_2\text{O}_3\text{-}14\text{SiO}_2\text{-}20\text{Li}_2\text{O-}15\text{Na}_2\text{O-} \text{Al}_2\text{O}_3\text{-} x\text{Sm}_2\text{O}_3)$, where $x= 0.0, 100, 200, 300, 400$, and 500 ppm have been synthesized by the conventional melting and quenching method. The Color of all glass samples prepared was yellowish and transparent. The XRD patterns confirmed the amorphous nature of all glass samples. The density of the prepared glass samples was found to be increased with the increasing content of samarium. Furthermore, the molar volume of these glasses decreased which led to compaction of the BSLNA glass network. The FTIR analysis consisted of several bands which indicated

the characteristic of Si-O and B-O vibrational groups. The optical absorption spectra of these glasses revealed that the fundamental absorption edge shifted to a longer wavelength as the content of Sm_2O_3 increased. The bandgap decreased by increasing Sm_2O_3 content. The refractive index and Urbach energy were found to increase with the increasing content of samarium. It was observed a strong orange-red emission owing to the transition $^4\text{G}_{5/2} / ^6\text{H}_{7/2}$ for BSLNA doped glasses. The PL intensity for this transition was observed to be the maximum for BSLNA4 doped glass sample, then it reduced for glass with BSLNA5 doped glass, due to the luminescence quenching process. Also, CIE color co-ordinates have obtain that in the white- reddish- orange region, the emission of Sm^{3+} ions incorporated in the host glasses has appeared.

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Figure Captions:

Figure 1: X-Ray diffraction patterns of the prepared samples.

Figure 2: Optical absorption spectra for prepared glass samples doped with Sm_2O_3 in the range from 400-1800 nm.

Figure 3: $(ah\nu)^{1/2}$ in $\text{cm}^{-1/2} \text{ eV}^{1/2}$ vs $h\nu$ eV of prepared glass

Figure 4: Variation of $\ln(\alpha)$ against $h\nu$ for all prepared glass samples.

Figure 5: The absorption and emission cross-sections for the prepared glass samples doped with Sm_2O_3 , [A] Sm^{3+} 100ppm, [B] Sm^{3+} 200ppm, [C] Sm^{3+} 300ppm, [D] Sm^{3+} 400ppm, [E] Sm^{3+} 500ppm.

Figure 6: Gain coefficient for the prepared glass samples doped with Sm_2O_3 . [A] Sm^{3+} 100 ppm, [B] Sm^{3+} 200ppm, [C] Sm^{3+} 300ppm, [D] Sm^{3+} 400ppm, [E] Sm^{3+} 500ppm.

Figure 7: Emission spectra for the prepared glass samples doped with Sm_2O_3 under excitation wavelength (402nm).

Figure 8: CIE-1931 chromaticity diagram for prepared glass BSLNA under excitation wavelength (402nm)

Figure 9: Deconvolution of FTIR spectra for BSLNA2 glass sample.
 Sm^{3+} 200 ppm

Table (1): Compositions (mol%) and shapes of our prepared glasses system ($50\text{B}_2\text{O}_3\text{--}14\text{SiO}_2\text{--}20\text{Li}_2\text{O--}15\text{Na}_2\text{O--}1\text{Al}_2\text{O}_3\text{--}X\text{Sm}_2\text{O}_3$).

Glass code	Chemical composition	Samples shape
BSLNA0	$50\text{B}_2\text{O}_3\text{--}14\text{SiO}_2\text{--}20\text{Li}_2\text{O--}15\text{Na}_2\text{O--}1\text{Al}_2\text{O}_3\text{--}0\text{Sm}_2\text{O}_3$
BSLNA1	$50\text{B}_2\text{O}_3\text{--}14\text{SiO}_2\text{--}20\text{Li}_2\text{O--}15\text{Na}_2\text{O--}1\text{Al}_2\text{O}_3\text{--}100\text{ppm Sm}_2\text{O}_3$
BSLNA2	$50\text{B}_2\text{O}_3\text{--}14\text{SiO}_2\text{--}20\text{Li}_2\text{O--}15\text{Na}_2\text{O--}1\text{Al}_2\text{O}_3\text{--}200\text{ppm Sm}_2\text{O}_3$
BSLNA3	$50\text{B}_2\text{O}_3\text{--}14\text{SiO}_2\text{--}20\text{Li}_2\text{O--}15\text{Na}_2\text{O--}1\text{Al}_2\text{O}_3\text{--}300\text{ppm Sm}_2\text{O}_3$
BSLNA4	$50\text{B}_2\text{O}_3\text{--}14\text{SiO}_2\text{--}20\text{Li}_2\text{O--}15\text{Na}_2\text{O--}1\text{Al}_2\text{O}_3\text{--}400\text{ppm Sm}_2\text{O}_3$
BSLNA5	$50\text{B}_2\text{O}_3\text{--}14\text{SiO}_2\text{--}20\text{Li}_2\text{O--}15\text{Na}_2\text{O--}1\text{Al}_2\text{O}_3\text{--}500\text{ppm Sm}_2\text{O}_3$

Table (2): The values of the density (g/cm³), molar volume (cm³/mol), and oxygen packing density (g.atom/L) of the prepared glass samples.

Glasses code	Density (g/cm ³)	Molar Volume (cm ³ /mol)	Oxygen packing density (mol/l)	Average boron-boron separation < d ^{B-B} > (nm)
BSLNA0	2.397	27.58	78.31	0.5728
BSLNA1	2.423	27.29	79.14	0.565
BSLNA2	2.5158	26.31	82.133	0.5528
BSLNA3	2.5632	25.83	83.65	0.5445
BSLNA4	2.5961	25.52	84.69	0.5375
BSLNA5	2.62566	25.25	85.62	0.522

Table (3): Indirect optical band gap energy E_{opt}^{in} , Urbach energy ΔE and refractive index, n.

Glasses code	Refractive index (n)	Indirect bandgap $E_{opt}(eV)$	Urbach energy $\Delta E(eV)$	Molar refraction ($R_m, cm^3 mol^{-1}$)	Polarizability(α_m) ($\times 10^{-24}$) (cm^3)	Dielectric constant (ϵ)	Refraction loss (R_L)
BSLNA0	2.2497	3.61	0.36	15.86	6.29	5.061	0.14
BSLNA1	2.2628	3.55	0.45	.158	6.27	5.12	0.149
BSLNA2	2.294	3.41	0.49	15.44	6.12	5.26	0.154
BSLNA3	2.32066	3.3	0.51	15.48	6.14	5.32	0.158
BSLNA4	2.3452	3.2	0.55	15.31	6.072	5.5	0.1617
BSLNA5	2.736	2.0	1.37	17.26	6.85	7.49	0.215

Table (4): Physical properties of prepared glass samples.

Sample glass	Polaron radius of Sm^{3+} ions (A) (± 1.5039)	The internuclear distance of Sm^{3+} ions (A) (± 0.3732)	The field strength of Sm^{3+} ions ($\times 10^{14} \text{ cm}^{-2}$) (± 1.4875)	Sm^{3+} ions concentration ($\times 10^{20} \text{ ion cm}^{-3}$) (± 0.5990)
BSLNA0	0	0	0	0
BSLNA1	6.669	16.550	3.8807	2.20591
BSLNA2	5.2279	12.974	5.5014	4.5783
BSLNA3	4.539	11.2662	7.298	6.993
BSLNA4	4.1075	10.1943	8.912	9.439
BSLNA5	3.79954	9.429	10.415	11.926

Table (5): Experimental (f_{exp}) and calculated (f_{cal}) oscillator strengths along (from the ground state, ${}^6H_{5/2}$), with JO parameters of prepared glass samples.

Transition ${}^6H_{5/2} \rightarrow$	Wavelength (nm)	Energy (cm^{-1})	BSLNA1		BSLNA2		BSLNA3		BSLNA4		BSLNA5	
			f_{exp}	f_{cal}	f_{exp}	f_{cal}	f_{exp}	f_{cal}	f_{exp}	f_{cal}	f_{exp}	f_{cal}
${}^6F_{1/2}$	1563	6397	0.3534	0.7149	0.2598	0.5064	0.2296	0.4403	0.1324	0.3628	0.1798	0.4076
${}^6H_{15/2}$	1536	6510	0.4514	0.0134	0.3398	0.0095	0.3045	0.0083	0.3353	0.0101	0.4049	0.0085
${}^6F_{3/2}$	1505	6644	1.8852	1.3659	1.346	0.9892	1.1732	0.8676	1.1448	0.8024	1.3018	0.9738
${}^6F_{5/2}$	1402	7131	1.725	1.8584	1.2575	1.3615	1.1073	1.1995	1.0284	1.1705	1.3781	1.4657
${}^6F_{7/2}$	1253	7980	1.9952	2.226	1.4642	1.6007	1.2936	1.4059	1.5394	1.5757	1.4118	1.5609
${}^6F_{9/2}$	1094	9135	1.1945	1.1933	0.8676	0.8495	0.7709	0.7447	0.8304	0.8852	0.8443	0.7792
${}^6F_{11/2}$	950	10517	0.2739	0.1648	0.1892	0.1169	0.1636	0.1025	0.1476	0.1238	0.2815	0.1053
${}^4I_{13/2}$	484	20661	3.056	0.096	1.6071	0.0681	1.1917	0.0596	1.1011	0.0724	1.0696	0.061
${}^4F_{7/2}$	435	22988	2.3345	0.0006	0.8993	0.0004	0.7762	0.0004	0.4689	0.0005	0.6955	0.0004
Ω_2 $10^{-20} cm^2$			3.6877		2.6119		2.2711		1.8716		2.1026	
Ω_4 $10^{-20} cm^2$			6.1114		4.488		3.9574		3.9021		4.9141	
Ω_6 $10^{-20} cm^2$			3.1274		2.2181		1.943		2.3582		1.9861	
RMS			0.7540		0.7547		0.76163		0.7510		0.775	

Table (6): Calculated branching ratio (β) and lifetime (τ , ms) of prepared glass samples.

Transition ${}^6\text{H}_{5/2} \rightarrow$	BSLNA1		BSLNA2		BSLNA3		BSLNA4		BSLNA5	
	B	τ (ms)	β	τ (ms)	β	τ (ms)	β	τ (ms)	β	τ (ms)
${}^6\text{F}_{1/2}$	0.5462	3.0156	0.5359	4.0249	0.5323	4.5533	0.4860	4.9740	0.4584	4.0774
${}^6\text{H}_{15/2}$	0.0523	115.68	0.0512	153.82	0.0509	173.085	0.0520	143.484	0.0490	156.947
${}^6\text{F}_{3/2}$	0.5223	2.6317	0.5227	3.5044	0.5228	3.9581	0.5111	4.1247	0.5303	3.4445
${}^6\text{F}_{5/2}$	0.4546	1.9485	0.4611	2.6003	0.4628	2.9338	0.4484	2.8721	0.5058	2.5279
${}^6\text{F}_{7/2}$	0.3804	1.2087	0.3730	1.5899	0.3699	1.7766	0.4077	1.7233	0.3808	1.5869
${}^6\text{F}_{9/2}$	0.2828	1.2172	0.2796	1.6296	0.2800	1.8346	0.3045	1.6545	0.2695	1.6210
${}^6\text{F}_{11/2}$	0.0223	1.1857	0.0224	1.5905	0.0223	1.7906	0.0245	1.5736	0.0218	1.5776

Table (7): Positions and assignments of FTIR bands of the (B₂O₃-SiO₂-Li₂O- Na₂O-Al₂O₃-Sm₂O₃) glass system.

Wavenumber (cm ⁻¹)	Assignment
(a)415-425	Specific vibration of Li ₂ O in the glass network
(b)432-444	Specific vibration of Na-O in the glass network
(c)460-480	bending vibration of Si-O-Si
(d)540-570	Stretching vibration of Sm ₂ O ₃
(e)650-663	Asymmetrical bending vibrations (ν_4) of Si-O-Si
(f)704-712	Bending vibrations B-O-B
(g)850-870	Stretching vibrations of tri-, tetra/ penta borate groups
(h)931-950	B-O stretch in BO ₄ units from di-borate groups
(i)1033-1045	Sm ⁺³ stretching vibration
(j)1070-1115	Symmetric stretching vibration of Si-O-Si bonds
(k)1330-1345	B – O stretch in BO ₃ units from varied types of borate groups
(l)1414-1426	B–O Stretching vibration of BO ₃ units in metaborate, pyroborate & orthoborate groups
(m)1535-1549	Stretching modes of Si–OH and B – O stretch in BO ₄ units from tetrahedral groups
(n)1650-1660	B – O ⁻ bond in isolated pyroborate group

Figure 1: X-ray diffraction patterns of the prepared samples.

Figure 2: Optical absorption spectra for prepared glass doped with Sm_2O_3 in the range from 250- 2000nm.

Figure 3: $(\alpha h\nu)^{1/2}$ in $\text{cm}^{-1/2} \text{eV}^{1/2}$ vs $h\nu$ eV of prepared glass

Figure 4: $\ln \alpha$ (cm^{-1}) vs $h\nu$ eV of prepared glass.

Figure 5: Absorption cross-sections $\sigma_{AB}(\lambda)$ and stimulated emission cross section $\sigma_{SE}(\lambda)$ of doped with Sm_2O_3 . [A] Sm^{3+} 100 ppm, [B] Sm^{3+} 200 ppm, [C] Sm^{3+} 300 ppm, [D] Sm^{3+} 400 ppm, [E] Sm^{3+} 500ppm.

Figure 6: Gain coefficient for transitions of prepared glass samples doped with Sm₂O₃, [A] Sm³⁺ 100 ppm, [B] Sm³⁺ 200ppm, [C] Sm³⁺ 300ppm, [D] Sm³⁺ 400ppm, [E] Sm³⁺ 500ppm.

Figure 7: Emission spectra for the prepared glass samples doped with Sm₂O₃ under excitation wavelength (402nm).

Figure 8: CIE-1931 chromaticity diagram for prepared glass BSLNA under excitation wavelength (402nm)

Figure 9: Deconvolution of FTIR spectra for BSLNA2 glass sample.