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Fluorescence Properties of VO^{2+} co-doped with Dy^{3+} in Lithium Zinc Borate Glasses for Optoelectronic Applications

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Abstract: In this work, a series of Dy^{3+} and VO^{2+} doped lithium zinc borate (ZLVBDx) glasses with x = 0, 1, 5, 7, 9, 10 mol% was prepared using the melt-quenching approach. These glasses were experimentally investigated for their spectroscopic properties. For this, excitation spectra, emission spectra, Y/B ratio and CIE1931 diagram of these glasses were examined. The excitation spectra were recorded at 575 nm wavelength and the corresponding emission spectra were then observed at 348 nm and 363 nm. The Y/B ratio was observed to show a decreasing trend as the amount of vanadium increased. The CCT values for these glasses were found in the cool white region. In the chromaticity diagram, CIE coordinates fall near the pure white region, suggesting that these glasses are suitable for optoelectronic applications.
Keywords: *Commission Internationale de I'Eclairage, Correlated color temperature, Luminescence, Yellow/Blue Color Intensity ratio.*

1. Introduction

About 19% of the energy used worldwide is used for artificial lighting, which accounts for 10% of global carbon emissions (Van Driel, W. D., Evertz, Zaal, Nápoles, & Yuan, 2013; Van Driel, Jacobs, Watte, & Zhao, 2022; Baumgartner et al., 2012). Light-emitting diodes (LEDs) generate light in visible region using electroluminescence, a process that transforms electricity into light without the need for heat radiation. As a result, LEDs are highly energy- efficient than conventional incandescent lamps and have emerged as a durable, long-lasting, and ecologically friendly substitute (Van Driel et al., 2022; Chen et al., 2019). With the advancement in LED technology, high-power LED packages found applications in many lighting and non-lighting sectors, including data networking (Chew, Karunatilaka, Tan, & Kalavally, 2017; Wang, Wang, Chi, Yu, & Shang, 2013), indoor plant culture (Liang et al., 2018), automotive illumination, healthcare (Yeh, Wu, & Cheng, 2010) and so forth. The response of glassy luminescence materials employed in these LEDs usually depends on their physical, optical and structural features varying with the composition. So, different glass matrices with new compositions are consistently explored to get highly efficient luminescence glasses. Borate glasses are suitable glass formers for various technological applications due to low melting point, distinctive transparency, high thermal stability and for being excellent host for rare-earth (RE) ions (Thabit et al., 2023). Lithium zinc borate glasses have emerged as vital components in optoelectronics due to simple synthesis and diverse optical characteristics. Incorporating

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rare-earth and transition-metal dopants into these glass matrices enables precise modulation of their luminescent behavior, rendering them indispensable for optoelectronic applications (Algradee, Sultan, Samir, & Alwany, 2017), (Arya, Kaur, & Singh, 2016; Dalal et al., 2015; Seema et al., 2015). Among these dopants, vanadium (VO^{2+}) and dysprosium (Dy^{3+}) have gained much popularity due to their distinct optical properties and synergistic impact on the luminescence properties of borate glasses (Bishnoi, Duhan, & Khasa, 2024; Radha, Komaraiah, Sayanna, & Sivakumar, 2022; Manjeet et al., 2023). Vanadium, when doped into semiconducting glasses, exhibits a broad range of optical properties that arise from its variable oxidation states (V^{2+}, V^{3+}, V^{4+} and V^{5+}) and the local glass environment. The ability of vanadium to switch between these oxidation states in response to external stimuli significantly influences its optical absorption, coloration, and non-linear optical behavior. The absorption spectra of vanadium-doped glasses are dominated by broad absorption bands in the visible and Near Infra Red (NIR) regions. This absorption arises from d-d transitions and charge transfer processes between different oxidation states of vanadium (Pothuganti, Bhogi, Kalimi, & Reniguntla, 2020; Abdel-karim, Fayad, El-kashef, & Saleh, 2023). On the other hand, the unique optical properties of Dy^{3+} ion are dominated by its 4f-4f electronic transitions, which are well-shielded by the outer electron orbitals, leading to sharp absorption peaks in the ultraviolet, visible and near-infrared regions. It also exhibits strong luminescence, with emission peaks around 480 nm (blue) and 572 nm (yellow). This characteristic makes dysprosium-doped glasses suitable for use in white light-emitting devices, where a combination of blue and yellow emission can produce a broad-spectrum white light (Ramteke & Gedam, 2015; Bishnoi et al., 2024).

2. Material and Characterization

A glass series (coded as ZLVBDx) consisting of six glass samples have been fabricated using melt quenching method with the following chemical composition: $xV_2O_5 \cdot (10\text{-}x)ZnO \cdot 30Li_2O \cdot 60B_2O_3$ (x = 0, 1, 5, 7, 9, 10) with 1.0 mol% of Dy_2O_3. All the chemical components in the ratio specified above have been combined in a mortar pestle and ground to a smooth powder consistency. After that, the mixture has been heated in an alumina crucible for forty-five minutes at 1050 °C in an electric muffle furnace, by stirring the melt after every fifteen minutes. Two brass plates preheated to 200 °C have been used to pour and quench the melted sample to get glasses of uniform thickness. The Photo Luminescence (PL) emission and excitation spectra have been recorded using a Horiba spectrometer with a spectral slit width of 3 nm (Model: Fluoromax R298P).

3. Results and Discussion

3.1. Photoluminescence properties

For analyzing the photoluminescence characteristics of Dy^{3+} doped glass samples, excitation wavelengths (λ_{exc}) are required. Therefore, the excitation spectra have been recorded at an emission wavelength (λ_{emi}) of 575 nm (J. Dahiya et al., 2022) and has been displayed in Figure 1. The spectrum for samples ZLVBDx with x = 0,1,5,7 mol% comprises of 8 sharp peaks at 323 nm, 348 nm, 363 nm, 386 nm, 424 nm, 452 nm, 472 nm and 525 nm which corresponds to the electronic transitions from ground state (6H15/2) to these excited states respectively: $^6P_{3/2}$, $^6P_{7/2}$, $^6P_{5/2}$, $^4F_{7/2}$, ($^4B_{2g} \rightarrow {}^2A_{1g}$), $^4I_{15/2}$, $^4F_{9/2}$ and $^6F_{3/2}$ (Bishnoi et al., 2024). The peaks centered at 348 nm and 363 nm have been chosen as excitation wavelengths for further exploring the emission spectra.

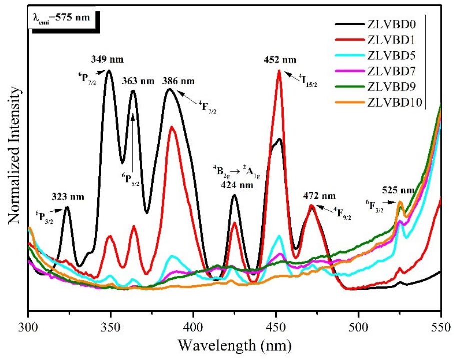

Figure 1: Excitation spectra of Dy^{3+} and VO^{2+} doped ZLVBDx glass system for λ_{emi} = 575 nm.

Figure 2 exhibits the emission spectra recorded for emission at an excitation wavelengths of 348 nm and 363 nm. It represents two intense bands for blue and yellow emissions respectively at 482 nm ($^4F_{9/2} \rightarrow {}^6H_{15/2}$), 575 nm ($^4F_{9/2} \rightarrow {}^6H_{13/2}$) and one feeble band for red emission at 664 nm ($^4F_{9/2} \rightarrow {}^6H_{11/2}$) (Dahiya, Hooda, Agarwal, & Khasa, 2022a, 2023).

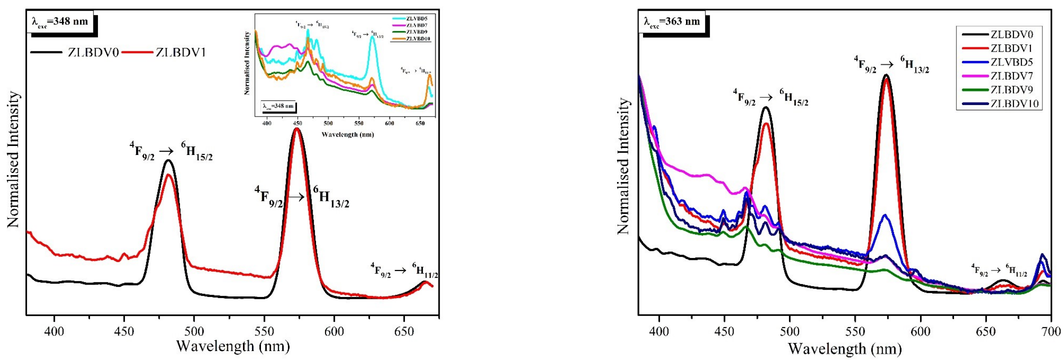

Figure 2: Emission spectra of Dy^{3+} and VO^{2+} doped ZLVBDx glass system for λ_{exc} (a, Left) 348nm; (b, Right) 363nm.

It can be easily noticed from the emission spectra that the amplitude of PL emission has decreased with increasing VO^{2+} ions doping concentration. The parameter yellow to blue emission intensity ratio (Y/B) gives an idea about the environment around the Dy^{3+} ions in the host glass. Higher Y/B ratio means higher yellow emission indicating increased asymmetry around the dysprosium ions (Thabit et al., 2023). The computed Y/B ratios are given in the Table 1. The decreasing Y/B ratio with increasing V_2O_5 content indicates that the asymmetry around the Dy^{3+} ions is decreasing. Figure 3 depicts a pictorial comparison between varying Y/B ratio with the increasing concentration of VO^{2+} ions in the glass matrix.

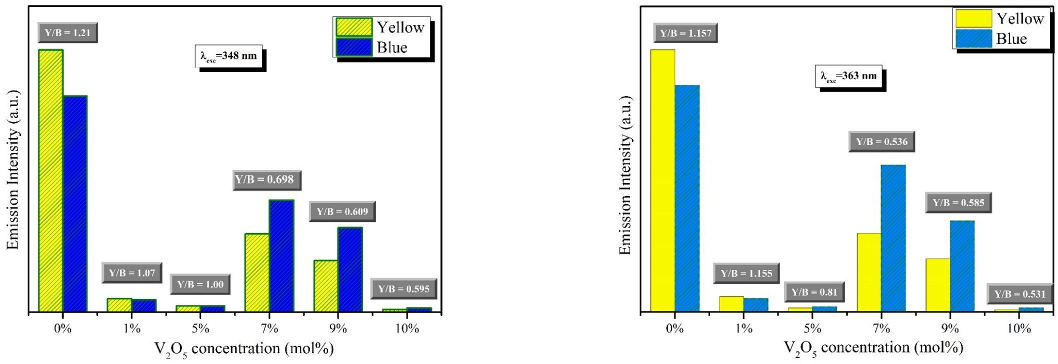

Figure 3: Comparison of Y/B ratio for all the synthesised glass samples of ZLVBDx system with the increasing concentration of VO^{2+} ions at excitation wavelengths of (a, Left) 348nm; (b, Right) 363nm.

3.2. Colorimetric analysis

"Colorimetry" (science of color) is the quantification of the color radiated by a material. It is directly related to the human color vision. Practically, human eye perceives mixture of two colors as single color and is incapable of decoding the actual dichromatic composition of the mixture (Alqarni et al., 2019; Alqarni, Bulus, Danmallam, & Yusof, 2023). Hence, CIE 1931 chromaticity system has been utilised for calculating the color coordinates (x,y) and correlated color temperature (CCT) values to confirm the colorimetry of the synthesised glasses. The values have been compiled in Table 1 and the corresponding CIE 1931 chromaticity curve has been presented in Figure 4. The sample with no vanadium content i.e. ZLVBD0 has parameters in the white light region (0.33,0.33). For VO2+ ions amount from 1 mol% to 5 mol%, the color coordinates lie in the white light zone whereas from 7 mol% to 9 mol%, the parameters shift somewhat towards the blue region and for 10 mol% V2O5 content, the coordinates again lie in the region of white light. Although Y/B ratio for ZLVBD10 is quite low. By using the chromaticity coordinates, CCT value has been calculated through the following McCamy's empirical formula (Kaur et al., 2018):

$$CCT = 499n^3 + 3525n^2 - 6823n + 5520 \quad (1)$$

where, n = (x-x_e)/(y-y_e) and (x_e, y_e) = (0.3320, 0.1858) is the chromaticity epicenter coordinates. It has been found in the previous studies that materials possessing high CCT values have light emission with vision of superior brightness and better acuity. The CCT values for all the samples lie in the cool white light region. These results affirm the prospect of the as-produced glass samples, particularly ZLVBD0, ZLVBD1 and ZLVBD5 for developing White Light Emitting Diodes (WLEDs) devices.

4. Conclusion

Dy^{3+} and VO^{2+} ions doped ZLVBDx glasses were prepared using melt quenching method and then characterized to explore the luminescent behaviour. The emission spectra under excitation of 348 m and 363 nm reflect strong blue, yellow and weak red bands. The samples with V2O5 concentration from 1 mol% to 5 mol% exhibited the optimum behaviour. Their CIE color coordinates lie very close to standard white light coordinates in the visible region indicating their aptness for white light emission. These results

Table 1: (x,y) chromaticity coordinates, Y/B ratio and CCT values of ZLVBDx glass system under excitation of 348 nm and 363 nm.

Sample Code	λ_{exc} = 348 nm				λ_{exc} = 363 nm			
	x	y	Y/B	CCT (K)	x	y	Y/B	CCT (K)
ZLVBD0	0.33	0.34	1.21	5765	0.31	0.33	1.157	6511
ZLVBD1	0.31	0.33	1.07	6657	0.3	0.32	1.155	7218
ZLVBD5	0.3	0.32	1	7556	0.29	0.3	0.81	8366
ZLVBD7	0.26	0.26	0.698	16385	0.26	0.25	0.536	18809
ZLVBD9	0.27	0.28	0.609	10941	0.27	0.28	0.585	11815
ZLVBD10	0.3	0.31	0.595	7431	0.29	0.31	0.531	8133

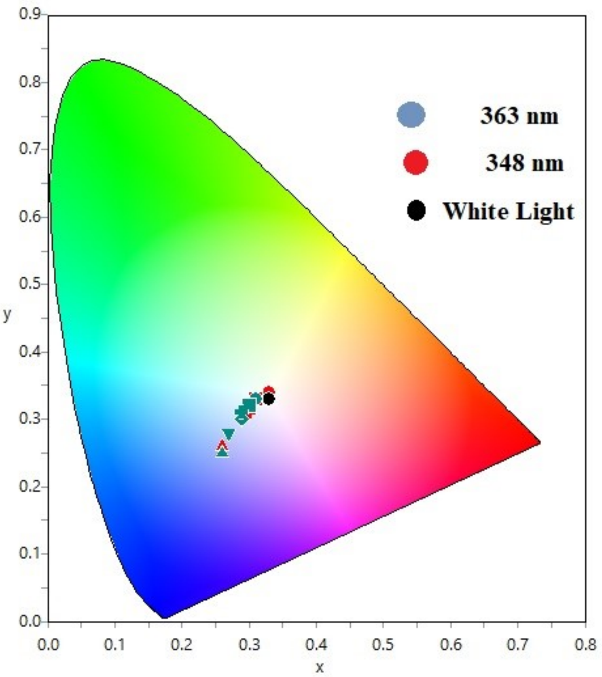

Figure 4: CIE diagram for the color coordinates of white light and ZLVBDx glass system at λ_{exc} = 348 nm and 363 nm.

depict the appropriateness of the synthesised glasses to be used in various photonic applications.

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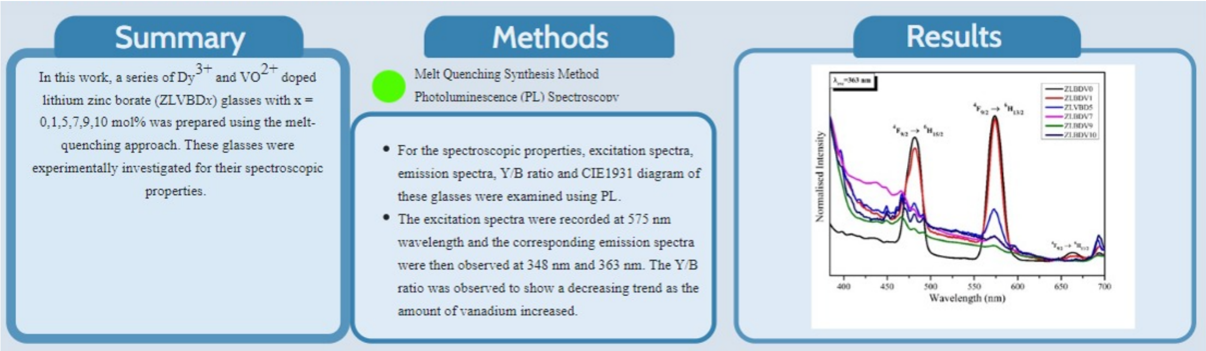
Graphical Abstract



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