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Emission Analysis of Dy^{3+} Doped Fluoroborate Optical Glasses

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ABSTRACT

Trivalent rare earth ion Dy^{3+} doped with various concentrations (0.1, 0.2 & 0.3 mol) of Zinc magnesium lithium fluoro borate (ZMLB) optical glass were synthesized through traditional melt quenching method and their emission spectra were observed under an excitation with 386 nm, it exhibits four emission bands were positioned at 481, 575, 662 and 753 nm corresponding to $^4F_{5/2}\rightarrow{}^6H_{15/2}$, $^4F_{5/2}\rightarrow{}^4F_{13/2}$, $^4F_{5/2}\rightarrow{}^4F_{11/2}$ and $^4F_{5/2}\rightarrow{}^4F_{9/2}$ transitions of Dy^{3+} ion, respectively. The decay lifetimes were determined and the lifetimes were decreased with the rise of Dy^{3+} doping concentration. The CIE coordinate values were estimated from emission spectra, which positioned at white light emission region. Based on all these results, Dy^{3+} ions doped ZMLB optical glasses are highly desirable for white light emitting applications.

Keywords: Dy ion, ZMLB glasses, emission analysis, WLED, CIE.

INTRODUCTION

Luminescent materials activated with rare earth (RE) ions are well favourable alternate to vapour discharge and fluorescent lamps [1]. RE ions are having specific photoluminescence properties in the region from visible to NIR, which is due to their 4f-4f and 4f-5d electronic intra-configurational transitions when excited with suitable excitation source [2]. The oxide glass samples possessing intrinsic chemical and thermal durability compared to polycrystalline powders materials, hence oxide glasses are well suitable host materials for activating with RE ions [3]. Oxide glasses doped with RE ions are inexpensive to synthesize and expansible to various dimensions and shapes for scintillators, optical amplifiers, dosimeters, solid-state lasers and light emitting applications [4].

The RE ions fascinating photoemission properties are generally based on the activated ion content and the host matrix chemical composition. In different oxide glass materials, borate possessing low atomic number, huge transparency in UV-Vis region, melting at low temperature and elevate RE ion soluble when compared to silicate glass matrix [5-6] Researchers ultimately to overcome the disadvantages of borate glass material by incorporation the alkali and heavy metal ions in the borate glass material as a network modifier. The MgO and LiF presence in the borate glass material reduce the melting temperature, which owing to disparity of glass-phase and phonon energy by changing from BO_3 to BO_4 unit cell [7]. The introducing of ZnO material (huge energy gap) to the borate glass-material enhances the UV-Vis transparency and chemical stability of borate glass materials [8]. The alkali LiF (network modifier) and mixture of MgO, ZnO (network intermediates) with B_2O_3 as a network developer glass host composition can be used for efficient optical utilizations.

Dysprosium (Dy^{3+}) ions exhibit the prominent luminescence properties among all the RE's ions in the borate glass materials with network modifier ions and they acquired research concern for a white light production with low cost and mercury free. The Dy^{3+} doped glass matrix depicts several excitation bands in blue and n-UV region and it has high intense emission bands in the region blue and yellow of visible spectrum [9]. These emission bands are ultimately favourable in the white light generation and lasers in yellow region discover the applications in the areas of headlights and architectural lighting [10]. Currently, an ionizing radiation impacts investigation on RE-doped heavy oxide metal glasses is attaining importance for uses in food industry, dosimetry and nuclear engineering [11]. In addition, the compact Dy^{3+}-doped borate-alkali metal glasses are best replacement to guard the radioisotopes [12]. To the best of our knowledge and according to literature, Dy^{3+} ions doped ZMLB glasses thermal, structural and photoluminescence properties are not reported so far. This work concerned with various concentrations of Dy^{3+} (0.1, 0.2 and 0.3 mol) activated ZMLB glass matrix synthesized by melt-quench method and thermal, structural photoluminescence and decay lifetimes properties were studied in detailed.

Experimental section

Glasses of chemical composition 10ZnO-10MgO-20LiF-(60-x)B_2O_3-xDy_2O_3 (where x= 0, 0.1, 0.2 and 0.3 mol) taken for glass preparation via a melt quench method. The 10-gram weighted analytical grade chemicals were taken with respect to the glass composition calculation and then these chemicals were fine grounded by using agate mortar and pestle. Completely mingled chemicals gathered in a porcelain crucible and placed in the furnace with control temperature for melting at 950 ^{0}C for 1 hour in order to homogeneous melts. Then these melts were quenched in between two smooth surfaced brass plates to obtain good optical quality glasses.

The excitation, luminescence and decay lifetimes of Dy^{3+} activated ZMLB glasses were measured by using SPEX Flurolog-3 spectrophotometer, included with an Xe-arc lamp (450 W) as the pumping source.

RESULTS AND DISCUSSIONS

Photoluminescence properties

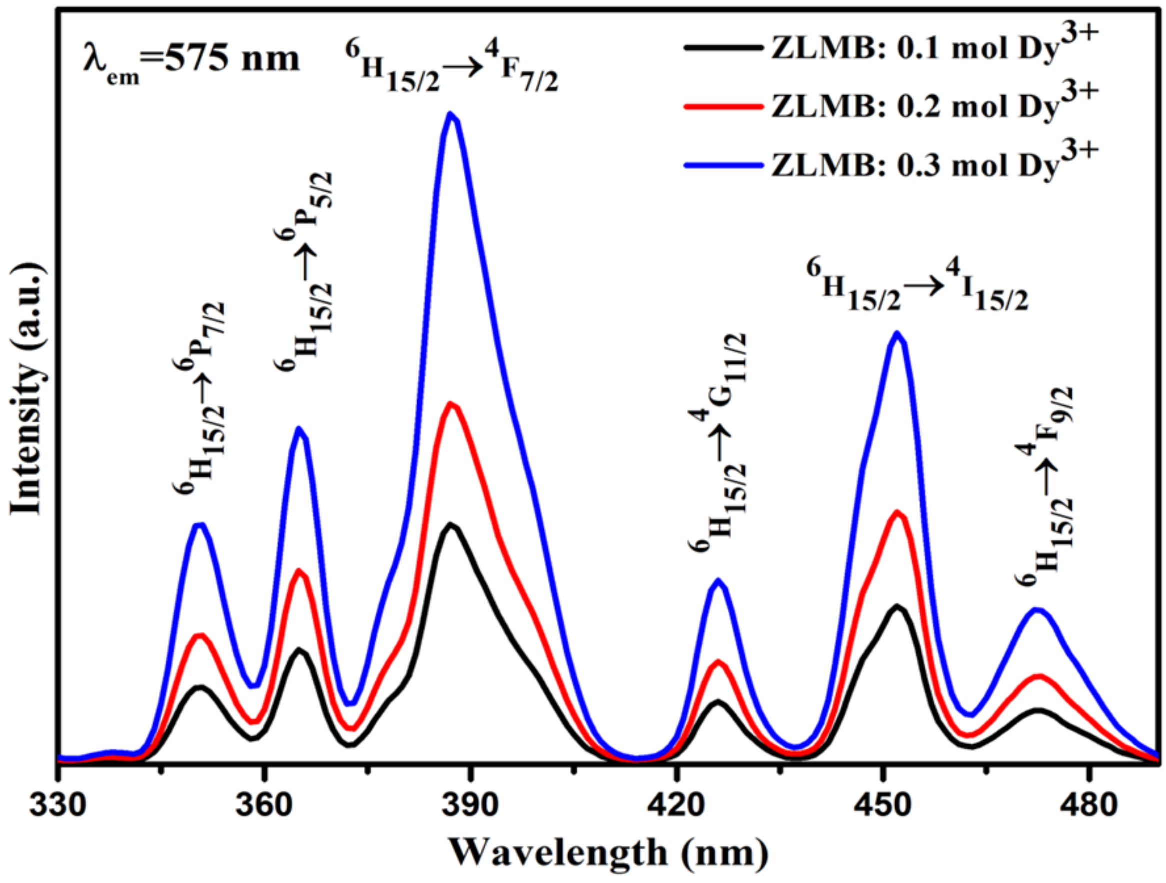

Fig. 1. Excitation spectra of ZMLB: xDy^{3+} (x= 0, 0.1, 0.2 and 0.3 mol) glasses.

The Dy^{3+} ions show excitation peaks in the n-UV to blue range of the visible region, commercially available blue and UV LED chips also possess their luminescent bands in visible region. A peculiar excitation peak of Dy^{3+} ion was chosen for excitation of the glass materials to analyse the relevant emission characteristics. In a manner to identify prominent excitation source wavelength, the excitation peaks of Dy^{3+}-activated glass materials were collected by examining the analogous emission wavelength at 575 nm. Dy^{3+} ions doped ZMLB glasses excitation spectra are depicted in Fig. 1. The spectra depicts six peaks relating to $^6H_{15/2}\rightarrow{}^6P_{7/2}$, $^6P_{5/2}$, $^4F_{7/2}$, $^4G_{11/2}$, $^4I_{15/2}$ and $^4F_{9/2}$ electronic transitions at 351, 364, 386, 426, 452 and 472 nm, respectively [13]. Among these transitions, the excitation band noticed at 386 was detected to be most prominent band.

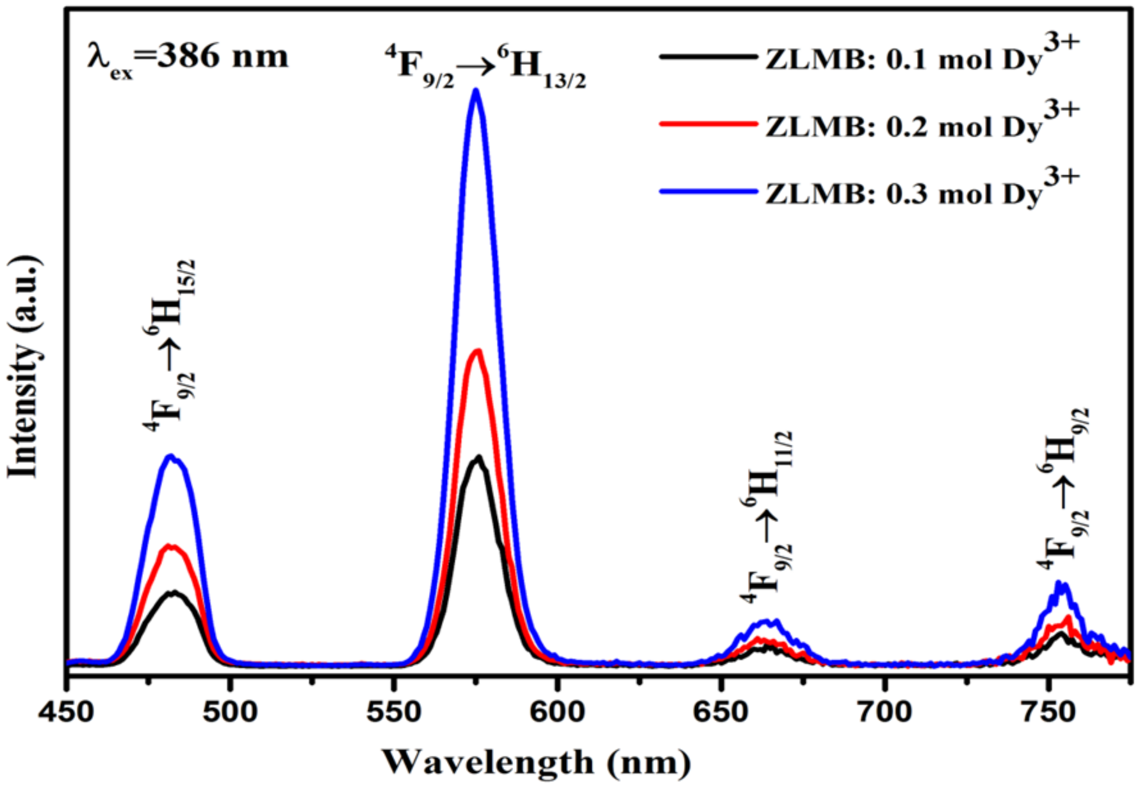

Fig.2 Emission spectra of ZMLB: xDy^{3+} (x= 0, 0.1, 0.2 and 0.3 mol) glasses.

In addition to analyse the emission behaviour, the Dy^{3+} : ZMLB optical glasses were excited with 386 nm and the related luminescence spectra were measured in the range 450-775 nm. The Dy^{3+} activated ZMLB glasses emission spectra are exhibited in the Fig. 2. The luminescence peaks of Dy^{3+}-activated glasses comprises of four bands cantered at 481 nm, 575 nm, 662 and 753 nm owing to $^4F_{9/2}\rightarrow{}^6H_{15/2}$ magnetic-dipole and $^4F_{9/2}\rightarrow{}^6H_{13/2}$, $^4F_{9/2}\rightarrow{}^6H_{11/2}$ and

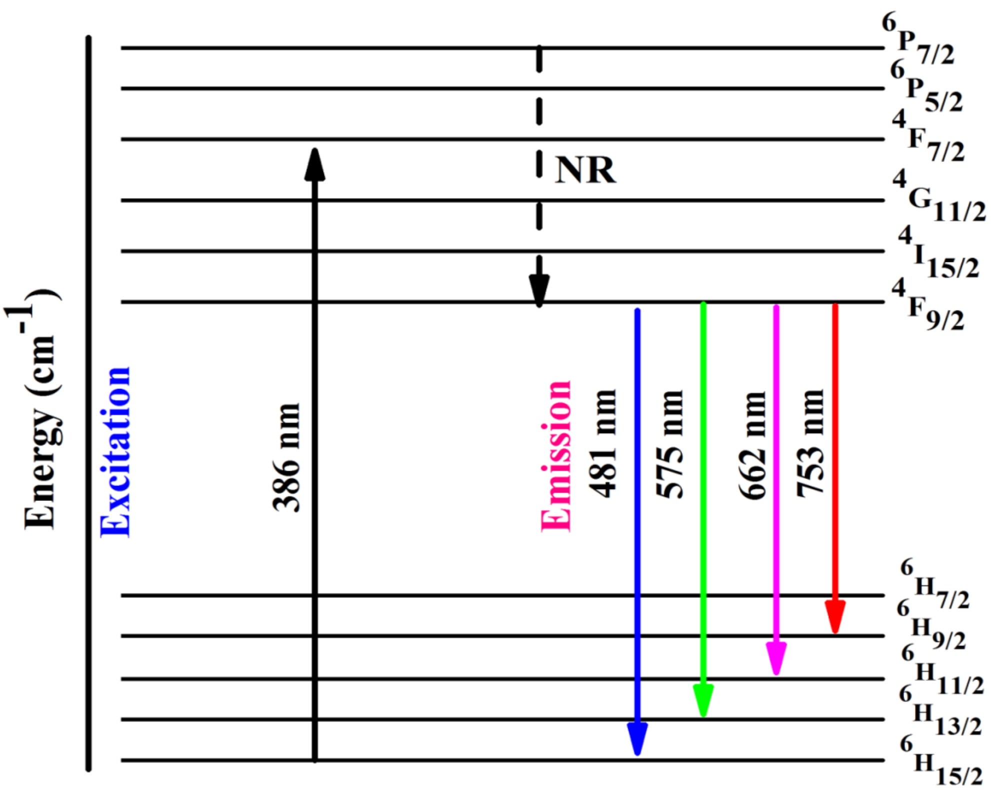

Fig.3. Energy level diagram of Dy^{3+} ions doped ZMLB glasses.

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$^4F_{9/2}\rightarrow{}^6H_{9/2}$ electric-dipole transitions [14]. From the emission spectra it can be observed four emission peaks and the intense band observed at yellow colour ($^4F_{9/2}\rightarrow{}^6H_{13/2}$) region, moderate in blue colour ($^4F_{9/2}\rightarrow{}^6H_{15/2}$) region and low in red colour region of Dy^{3+} doped ZMLB glasses. The luminescence proportion of electric dipole to magnetic dipole transition exhibits the position asymmetry surrounding the Dy^{3+} [15]. The estimated intensity proportions of yellow to blue (Y/B) peaks were at 2.25, 3.06 and 3.44 for ZMLB: xDy^{3+} (x= 0.1, 0.2 and 0.3 mol), respectively. The huge value of Y/B concludes, the Dy^{3+} ions were placed at high asymmetry position in the zinc magnesium lithium fluoro borate glass material and emitted strong yellow colour then blue colour. Fig. 3. shows the energy level diagram of radiative and non-radiative transitions of Dy^{3+} doped ZMLB glasses.

Decay curve analysis

The luminescent components it is know that, the radiative and non-radiative lifetime decay values of excited levels rely on their observable decay lifetimes. At RE positions, the variations in the surrounding crystal nature influence the lifetime values of radiative transitions [16]. The PL decay lifetime characterization performed for $^4F_{9/2}$ excited state of Dy^{3+} in ZMLB glasses were under the excitation with 386 nm wavelength and exhibited in Fig. 4. From Fig. 8, it is noticed the decay curve characteristics are exhibiting bi-exponential. Those bi-exponential decays can be study using the succeeding relation [17].

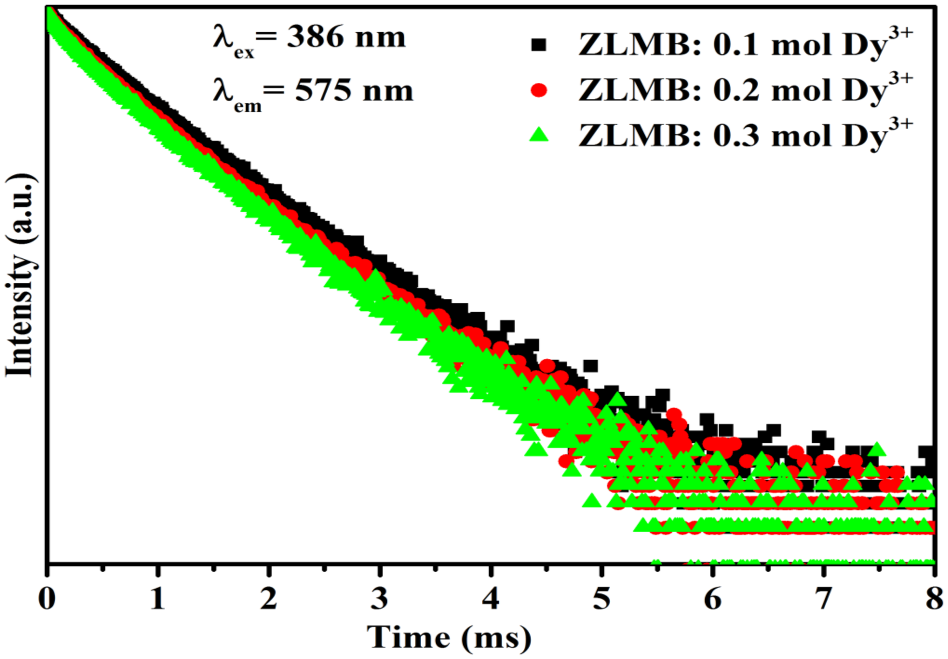

Fig. 4. Decay lifetime plot of ZMLB: xDy^{3+} (x= 0, 0.1, 0.2 and 0.3 mol) glasses.

$$I = A_1 \exp\left(\frac{-t}{\tau_1}\right) + A_2 \exp\left(\frac{-t}{\tau_2}\right) \quad (1)$$

here, I- luminescence intensity. A_1 and A_2- decay constants, τ_1 and τ_2- lifetimes of the both elements are contributing to the decay mechanism. The experimental decay lifetime (τ) values can be estimated by employing the succeeding expression [18]

$$\tau = \frac{A_1\tau_1^2 + A_2\tau_2^2}{A_1\tau_1 + A_2\tau_2} \quad (2)$$

The decay lifetime was determined and were detected to be around 0.75 ms, 0.70 ms and 0.65 ms for ZMLB: xDy^{3+} (x= 0.1, 0.2 and 0.3 mol) optical glasses, respectively. With enhance in Dy^{3+} ion content reduces the inter-atomic length among the Dy^{3+} ions, which makes inhomogeneous spreading of Dy^{3+} entirely the glass material. This nature of non-uniform collision of Dy^{3+} ions produces clusters or agglomerated surrounding environment of the host material. Hence, the energy migration takes place among Dy^{3+} ions centres via resonant energy migration and cross-relaxations. It is noticed that, the experimental decay lifetimes are reduced with the enhance of Dy^{3+} ion content due to the energy migration/cross-relaxation mechanism.

CIE chromaticity co-ordinates

The luminescent colour of Dy^{3+}-activated glass materials specified by CIE coordinates values by employing the derivatives named colour corresponding equations and shown in 1931 CIE colour chromaticity image. Fig. 5 exhibits the CIE color chromaticity image of ZMLB: xDy^{3+} (x= 0.1, 0.2 and 0.3 mol) optical glass samples. The CIE coordinate values were

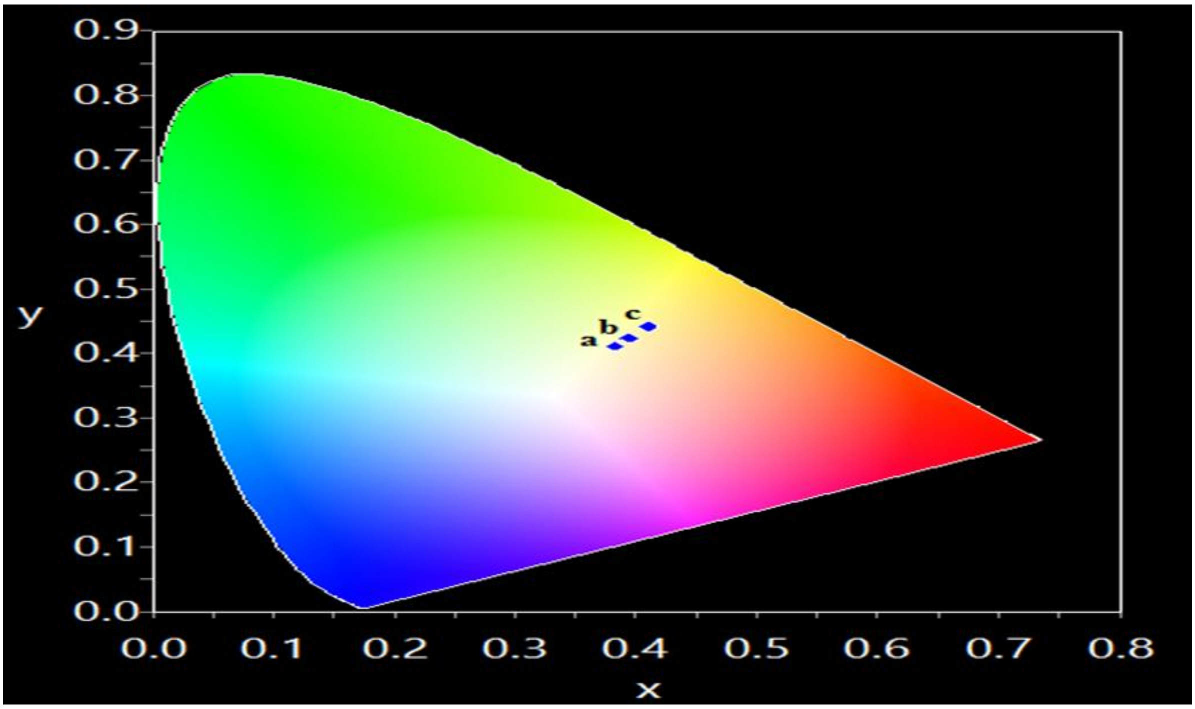

Fig. 5 CIE co-ordinates of ZMLB: xDy^{3+} (x= 0, 0.1, 0.2 and 0.3 mol) glasses.

estimated and were found to be (0.3831, 0.4117), (0.3947, 0.4242) and (0.4104, 0.4417) for ZMLB: xDy^{3+} (x= 0.1, 0.2 and 0.3 mol) optical glasses, respectively. The CIE colour coordinate values are clump about the yellowish white light portion. The strong yellow luminescence from Dy^{3+}-activated glass materials are highly useful in the construction of white light.

CONCLUSION

Dy^{3+}-activated Zinc magnesium lithium fluoro borate optical glasses were synthesized by a melt quench approach and explored their properties for conceivable lighting utilizations. The XRD studies confirmed that all the prepared samples having amorphous nature without any diffraction peaks. From FTIR, IR vibrational bands were confirmed that all metal ions and borate ions present in the samples. The energy band gap values were calculated from absorption spectra and were increased with the increase of Dy^{3+} ion concentration. The photoemission features of the glasses recorded at 386 nm excitation and it consist of emission peaks cantered at 481 nm (blue), 575 nm (yellow), 662 nm (red) and 753 (deep red) nm characteristic transitions of Dy^{3+} ion. The Y/B ratio is confirmed the asymmetric nature of the prepared glass samples. The decay lifetimes were estimated and the lifetimes were decreased with enhance of Dy^{3+} doping content. The strong luminescence from the ZMLB: 0.3 mol Dy^{3+} glass be revealed close to yellowish white light region of the CIE colour chromaticity image x=0.4104, y=0.4417 intimate the probable advantage of the glass material in display devices, backlighting and solid lighting in under excitation with n-UV LED chips.

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