STM JOURNALS

Journal of
Polymer & Composites

ISSN: 2321-2810 (Online)
ISSN: 2321-8525 (Print)
Volume 12, Special, Issue 6, 2024
DOI (Journal): 10.37591/JoPC

http://journals.stmjournals.com/JOPC

Research JoPC

Synthesis and Study of Photoluminescence properties of S_m^{3+} activated $7TiO_2 + 3Al_2O_3 + (60\text{-}x)B_2O_3 + xY + 5LiCO_3 + 25SrCO_3$ Borate Glass for SSL Applications

U.A. Nage[1,*], C.S. Khade[2]

Abstract

Melt quenching was used to successfully create the raddish orange light-emitting Sm3+activated $7TiO_2$ + $3Al_2O_3$+ (60-x) B_2O_3+xY+ $5LiCO_3$+$25SrCO_3$ (X=0.001, 0.002, 0.005, 0.01 and 0.02, Y=S_m^{3+}) borate glass. XRD examination confirmed the phase of the prepared Un-doped and Doped $7TiO_2$ + $3Al_2O_3$+ (60-x) B_2O_3+xY+ $5LiCO_3$+$25SrCO_3$ glass. Its PL characteristics under ultraviolet (UV) light (302-468 nm) were later studied. At 602 nm, the excitation was detected, and at 402 nm, the emission. The emission spectra have two prominent peaks at 564 nm and 612 nm, with one faint peak at 647 nm. The excitation spectra are made up of a string of sharp lines at 310, 363, 377, and 393 nm, which correspond to the 7F0◊6H5/2 and 7F0◊4L13/2 transitions, respectively. The $^7F_0 \rightarrow ^6H_{5/2}$ magnetic dipole is emitted at 564 nm, the $^5F_0 \rightarrow ^7D_2$ (electric dipole) at 612 nm, and the faint emission at 647 nm is associated with the $^5F_0 \rightarrow ^7D_3$ transition of S_m^{3+} ions. After analysis, the produced glass's CIE (Commission International de l′Eclairage) coordinates were determined to be x=0.0.6358, y= 0.3636. For solid state lighting and light-emitting diode (LED) applications, $7TiO_2$ + $3Al_2O_3$+ (60-x) B_2O_3+xEu3+ + $5LiCO_3$+$25SrCO_3$ has the potential to be extremely useful. The synthetic Sm^{3+}-doped borate glass's amorphous nature and thermal stability were verified by X-ray diffraction (XRD) patterns and differential thermal analysis (DTA), respectively. S_m^{3+} ions in the glass matrix have a long luminous lifetime, according to the decay curve analysis, which makes it a good choice for persistent luminescence applications.

Keywords: borate glass, melt quenching method, LED, CIE band red emitting

INTRODUCTION

Rare-earth (RE) ion-doped glasses are important components for optical amplifiers, waveguide lasers, and fiber optics. When exposed to appropriate excitation wavelengths, they radiate intensely in the visible, near-infrared, and infrared spectrums [1–3]. Because of their large inhomogeneous bandwidths, low production costs, strong thermal stability, ease of manufacture, and great doping capability, RE doped glasses are often superior than crystalline materials [4]. The oxide, borates glasses have been a matter of great interest for several researchers as they are considered to be the finest glass formers because of their low melting point, high transparency, high thermal stability and good RE ion solubility.

However, in addition to these features, borate glasses possess high phonon energies (1300cm-1) that suppress radiative emissions. Lead oxide

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Received Date: May 14, 2024
Accepted Date: July 13, 2024
Published Date: September 23, 2024

Citation: U.A. Nage, C.S. Khade. Synthesis and Study of Photoluminescence properties of S_m^{3+} activated $7TiO_2 + 3Al_2O_3$ + (60-x)B_2 O_3 +xY+ $5LiCO_3$ +$25SrCO_3$ Borate Glass for SSL Applications. Journal of Polymer & Composites. 2024; 12(6): S55–S60p.

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(PbO), a heavy metal oxide (HMO), can be added to borate glasses to boost their capacity for radiative emission. Zinc oxide (ZnO) can be added to lead borate glasses to act as a network forming or modifier, which helps to achieve low glass transition temperature and better chemical stability. A borate glass's chemical strength, emission characteristics, and mechanical strength can all be improved by adding aluminum trioxide (Al_2O_3). We are driven to synthesize a new borate glassy system doped with RE ions in order to better comprehend their potential applications in luminous devices like lasers and w-LEDs by the aforementioned diverse scientific patronages provided by B_2O_3, PbO, Al_2O_3, and ZnO [5,6].

In an effort to create innovative, tunable solid-state lasers, fiber optic communication devices, solar energy converters, and white light emitting diodes (w-LEDs), several researchers have documented the luminescence properties of doped crystalline and non-crystalline materials with RE^{3+} ions [7–16]. The structural and photoluminescence properties of lithium borate glasses based on various modifier oxides and doped with Dy3+ were reported by Balakrishna et al. [17]. The broad band 2.84 mm luminescence properties of Dy^{3+} doped ZrF_4–BaF_2–LiF_3–AlF_3 glass were recently reported by Tian et al. [18]. Through optical and thermal analysis of Dy^{3+} doped sodium fluoroborate glasses, Rayappanet al. were reported [19]. Pisarska*et al.* reported visible luminescence of dysprosium ions in oxyhalidelead borate glasses [20].

In this work, we have reported the preparation and characterization of $7TiO_2 + 3Al_2O_3$+ (60-x) B_2O_3+xY+ $5LiCO_3$+$25SrCO_3$ (x=0.001,0.002,0.005,0.01,0.02,Y=Eu^{3+}) glass synthesized using melt quenching method. The study also attempted to examine and investigate the concentration quenching effect and test the color purity of glass by CIE co-ordinates witnessing the prepared glass as main candidate for SSL and LED based applications.

Experimental

The Eu activated Borate glasses with composition $7TiO_2 + 3Al_2O_3$+ (60-x)B_2O_3+xY+ $5LiCO_3$+$25SrCO_3$ (x=0.001,0.002,0.005,0.01,0.02,Y=Eu^{3+}) were successfully prepared by melt quenching method.

Analar grade quality materials have been used in the present work. In this process, the materials were mixed up in an agate mortar and then melted in a silica crucible at 1000ºC in a furnace for 5 hours. In order to form the glass samples of uniform thickness, the melts were quenched between two preheated brass plates. To remove thermal strains, the as-prepared glasses were annealed for a period of 2 hours in an electrical furnace at a temperature of 300ºC.

The phase purity of prepared phopshors were analysed and evaluated by means of X-ray diffraction (PXRD) using a Rigakuminiflex II diffractometer with Cu Kα (λ = 1.5405 Å) operated at 5 kV. A Scanning Electronic Microscope (SEM) was used to study the structural and morphological characteristics i.e., particle size and shape of the sample. The measurements were carried out at Deparment of Physics RTM Univesisty Nagpur using a ZEISS EVO/18 Research. The measurements of PL and PL excitation (PLE) spectra were taken on (Hitachi F-7000) fluorescence spectrophotometer with a 450W xenon lamp.

RESULTS AND DISCUSSION

X-Ray Diffraction (XRD) Pattern and Surface Morphology

The XRD pattern of the RE activated $7TiO_2 + 3Al_2O_3$+ (60-x)B_2O_3+xY+ $5LiCO_3$+$25SrCO_3$ borate glass as shown in Figure 1 From Figure 1, it is clearly observed that the spectra do not show any sharp peaks. This indicates the amorphous nature of glass samples.

Figure 1 shows the SEM image of RE activated $7TiO_2 + 3Al_2O_3$+ (60-x) B_2O_3+x+ $5LiCO_3$+$25SrCO_3$ Borate Glass. From the above figure, it is observed that the microstructure of the glass

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Journal of Polymer & Composites
Volume 12, Special Issue 6
ISSN: 2321-2810 (Online), ISSN: 2321-8525 (Print)

comprises of irregular grains with heavy agglomeration. The average size of prepared glass is in submicron range.

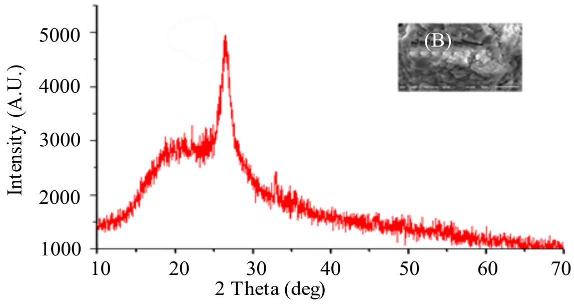

Figure 1. X-ray diffraction pattern of $7TiO_2 + 3Al_2O_3 + (60\text{-}x)B_2O_3 + xY + 5LiCO_3 + 25SrCO_3$ (x=0.001, Y=Eu^{3+}) borate glass and (B) Surface morphology of prepared glass.

Photoluminescence Properties (PL)

Figure 2 shows the combined excitation and emission spectra of $7TiO_2 + 3Al_2O_3 + (60\text{-}x)\ B_2O_3 + xEu^{3+} + 5LiCO_3 + 25SrCO_3$ (x=0.001 to 0.02 moles) borate glass. Monitoring was done at 618 nm for the excitation and 393 nm for the emission. The excitation spectra consists of series of sharp lines at 310, 363, 377 and 393 nm correspond to the $^7F \rightarrow {}^5H_3$, $^7F_0 \rightarrow {}^5D_4$, $^7F_0 \rightarrow {}^5L_7$ and $^7F_0 \rightarrow {}^5L_6$ transitions respectively. Figure 2 shows that among all these peaks, the excitation at 393 nm is the strongest one. On the contrary, emission spectra comprise two sharp peaks 595 nm and 618nm with one weak peak at 653nm. The $^5F_0 \rightarrow {}^7D_1$ (magnetic dipole) is emitted at 595 nm, the $^5F_0 \rightarrow {}^7D_2$ (electric dipole) at 618 nm, and the faint emission at 653 nm is associated with the $^5F_0 \rightarrow {}^7D_3$ transition of Eu3+ ions [21].

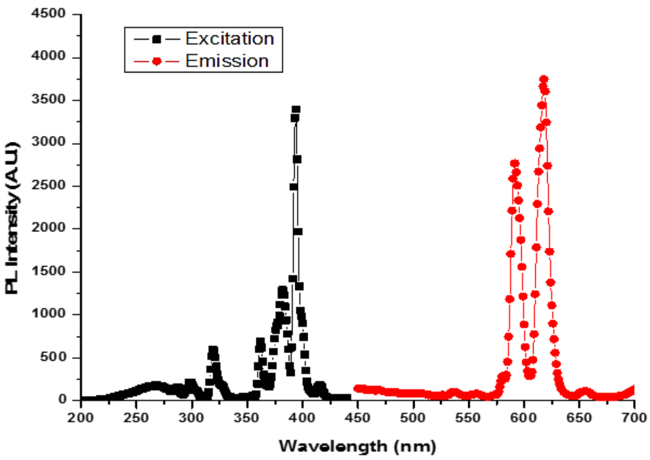

Figure 2. Combine excitation and emission spectra of $7TiO_2 + 3Al_2O_3 + (60\text{-}x)B_2O_3 + xEu^{3+} + 5LiCO_3 + 25SrCO_3$ (x=0.001 to 0.02 moles) Borate Glass

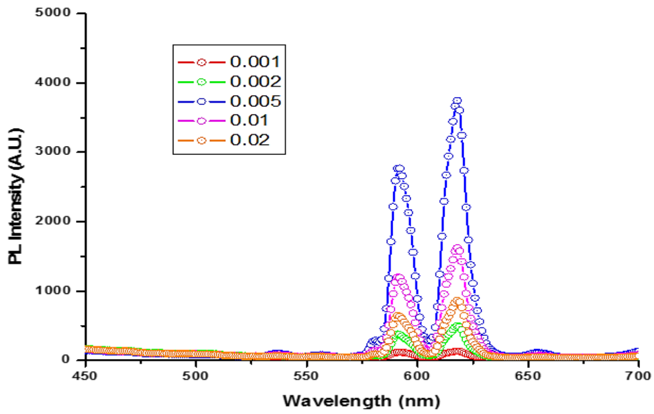

Figure 3. Emission spectra $7TiO_2 + 3Al_2O_3$+ (60-x)B_2O_3+xEu^{3+} + $5LiCO_3$+$25SrCO_3$ (x=0.001 to 0.02 moles)borate glass

Figure 3 illustrates the PL emission intensity as a function of Eu^{3+} ion concentration (0.001 to 0.02 moles). It is evident from Figure 3 that the ideal concentration for the created borate glass is 0.005 mol. When the concentration of Eu^{3+} ions is raised above this optimal value, it is seen that the luminescence intensity decreases because of the unique phenomena known as concentration quenching. Figure 4 shows the energy level scheme to give explanation of the emission process in $7TiO_2 + 3Al_2O_3$+ (60-x)B_2O_3+xEu^{3+} + $5LiCO_3$+$25SrCO_3$glass.

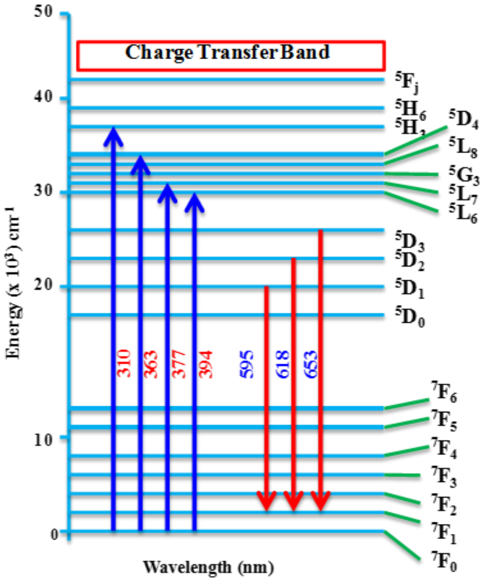

Figure 4. Energy-level scheme of Eu^{3+} ion.

Photometric Characterization

Figure 5 portrays the emission color of $7TiO_2 + 3Al_2O_3$+ (60-x)B_2O_3+xEu^{3+} + $5LiCO_3$+$25SrCO_3$ glass calculated from their PL spectra by observing the 618 nm emission by using Commission Internationale de Eclairage (CIE) diagram.

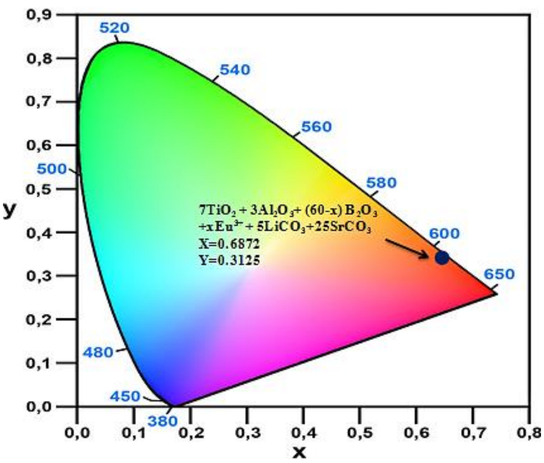

Figure 5. CIE chromaticity diagram of $7TiO_2 + 3Al_2O_3$+ (60-x)B_2O_3+xEu^{3+} + $5LiCO_3$+$25SrCO_3$ borate glass.

The color coordinates of the phosphor are found to be almost similar when it is excited at 398 nm excitation wavelength and correspond to (0.6872, 0.3125) for the phosphor with Eu = 0.005 mol. This indicates that europium activated $7TiO_2 + 3Al_2O_3$+ (60-x) B_2O_3+xEu^{3+} + $5LiCO_3$+$25SrCO_3$ is a promising candidate with red emission when excited in the near UV region. However, it proves to be advantageous to use it at 395 nm excitation, as it is a mercury free lamp.

CONCLUSIONS

The Eu^{3+} activated $7TiO_2$ + $3Al_2O_3$+ (60-x)B_2O_3+xEu^{3+} + $5LiCO_3$+$25SrCO_3$ borate glass were successfully synthesized by melt quenching method. The XRD patterns of Eu activated $7TiO_2$ + $3Al_2O_3$+ (60-x) B_2O_3+xEu+ $5LiCO_3$+$25SrCO_3$ (x=0.001,) borate glass is similar in shape with Un-doped $7TiO_2$ + $3Al_2O_3$+ (60-x) B_2O_3+xY+ $5LiCO_3$+$25SrCO_3$ borate glass. At 618 nm, the PL excitation spectrum was observed, and at 393 nm, the emission was observed. The synthesized borate glass's ideal concentration, according to the PL emission spectra, is 0.005 mol. The glass in its prepared state was determined to have CIE chromaticity coordinates of (0.6872, 0.3125). The results show that $7TiO_2$ + $3Al_2O_3$+ (60-x) B_2O_3+xEu^{3+} + $5LiCO_3$+$25SrCO_3$ borate glass is a prospective and potential candidate for Red-emitting near-UV LEDs.

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