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Absorption and emission analysis of zinc borotellurite glass doped with dysprosium oxide nanoparticles for generation of white light

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ABSTRACT

A series of zinc borotellurite glass doped with Dy_2O_3 nanoparticles with chemical formula $\{[(TeO_2)_{0.7}(B_2O_3)_{0.3}]_{0.7}(ZnO)_{0.3}\}_{1-x}(Dy_2O_3NP)_x$ (where x = 0.01, 0.02, 0.03, 0.04 and 0.05 M fraction) has been fabricated by using conventional melt-quenching method. In this work, absorption and emission analysis of the glass systems have been done. The absorption data are obtained from the UV–Vis spectroscopy while the emission spectrum of the glass systems is recorded by using the Luminescence spectrometer. From the absorption data, the direct and indirect optical band gaps for the glass systems are calculated. Both of the band gaps are found to decrease as opposed to the concentration of Dy_2O_3 nanoparticles. The decrement of the optical band gaps leads to the increment of the refractive index of the glass systems. From the emission spectra, two transition bands are observed, which represent the transition from $^4F_{9/2}$ to $^6H_{15/2}$ and $^6H_{13/2}$. Other than that, the *x* and *y* CIE chromaticity coordinates are determined from the emission spectra and are found in the region of white light. The values of the correlated colour temperature for this glass systems are in between 4104.22 and 4886.17 K and is in the neutral white light region.

1. Introduction

In recent times, extensive research has been done on glasses as the materials for optical devices applications [1–3]. The main goal of the study is to search for a new glass system with a better technological parameter. Tellurite based glass is one of the most promising glass and is widely used as the main host to achieve good optical and dielectric properties. The tellurite based glass has a high quality of glass forming ability [4], good mechanical strength and chemical durability, low melting point, high refractive index and good infrared transmission [5,6]. This characteristic of tellurite glass has made the tellurite glass as a good candidate for the further development of the optical systems. Addition of borate oxide into the tellurite glass systems able to improve the properties of the glass and provide moderate stability and durability to the glass systems. Furthermore, an addition of zinc oxide into the glass network helps to increase the glass forming ability and able to lower the crystallization rates of the glass [7].

According to Kityk et al. [8], doping rare earth element into the glass systems is important to enhance the quality of the glass. Rare earth not only acts as an optically active ion but also can influence the structure and properties of the multicomponent glass systems. Besides, the addition of rare earth helps to improve the glass stability and enhances the resistance to crystallization [9].

Among the series of rare earth elements, Dy^{3+} ion is identified as an active luminescence center. The Dy^{3+} ion is recognized as *f-f* localized trap-creating ion, which is found to increase the afterglow for a protracted time [10]. Furthermore, Dy^{3+} ion has been well incorporated into several glasses in order to obtain two primary colors, yellow and blue luminescence materials [11] thus can be considered as promising materials in the solid state lighting (SSL) technology. Other than that, the Dy^{3+} ion can act as a well-known activated ion, which leads to light emission in the visible range and offers an excellent possibility for white light application because it has strong excitation bands around 454 nm that perfectly matches with the emission spectrum of GaN-based LED [12]. Moreover, Dy^{3+} ion doped glass systems are a promising luminescent material in the blue-yellow region, thus, by adjusting to a suitable intensity of yellow to blue ratio (Y/B), the chromaticity coordinates of the glass that contained Dy^{3+} ion can generate white light [13].

Vigorous research has been done on the rare earth doped glass systems, however, to the best of authors' knowledge, there is still insufficient research that focuses on the generation of white light by using the rare earth nanoparticles. This current research highlighted the absorption and emission analysis by providing information and insight regarding the optical properties of zinc borotellurite glass systems at various concentrations of Dy^{3+} nanoparticles for the generation of white light.

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2. Experimental

A series of zinc borotellurite glass doped with dysprosium oxide nanoparticles (NP) with chemical formula $\{[(TeO_2)_{0.7}(B_2O_3)_{0.3}]_{0.7}(ZnO)_{0.3}\}_{1-x}(Dy_2O_3NP)_x$ (where x = 0.01, 0.02, 0.03, 0.04 and 0.05 M fraction) were fabricated by using conventional melt-quenching method. All the glass samples were synthesized from high purity chemicals of tellurium (IV) oxide, TeO_2 (99.9%, metal basis, Alfa Aesar), boron oxide, B_2O_3 (98.5%, Alfa Aesar), zinc oxide, ZnO (99.9%, metal basis, Alfa Aesar), dysprosium (III) oxide nanopowder, Dy_2O_3 (99.9%, < 100 nm, Nanostructure & Amorphous Materials Inc.)

All the chemicals with a composition of 5 g batches were weighted by using the digital weighing machine with an accuracy of ± 0.001 g and then were mixed and stirred thoroughly in the alumina crucible for 30 min to obtain a homogeneous mixture. The homogeneous mixture was then placed in the first electric furnace for the preheating process at 400 °C for 1 h as to remove the water molecules trapped in the mixture. The preheated mixture was transferred into the second furnace for the melting process. The melting process was done at 900 °C for 2 h. The molten mixture was immediately quenched into preheated cylindrical stainless steel mould to avoid solidification process from taking place. The fabricated glass samples were subsequently annealed in the first furnace at 400 °C for the period of 1 h. The purpose of the annealing process is to remove thermal strain, avoid the formation of cracks and air bubbles and to enhance the mechanical strength of the samples. The furnace was then switched off and the samples were left inside the furnace to cool down to room temperature. The prepared glass samples were polished on both sides to obtain clean, parallel and clear surfaces.

3. Results and discussions

3.1. Density and molar volume

Density is mainly determined by the chemical composition of the glass systems [14]. The density of the glass is one of the important parameters used to analyse the changes in the structure of the glass systems [15]. The density of the glasses firmly depends on the amount of network modifiers, their atomic weight and their atomic radius [16]. In this study, the variation of the density, ρ is shown in Fig. 1. It is seen from the figure that the density increases as the concentration of the Dy^{3+} NPs increases. The increasing value of the density of the glass systems is because of the addition of Dy_2O_3 NP which has larger molecular weight compare to other elements in the glass systems [17]. The increasing of the density also indicates that the glass system is more compact as the addition of the dopant increases.

Generally, the variation of the density and molar volume show an opposite behaviour. The molar volume, V_M which is defined as the volume occupied by one mole of glass, is calculated using the equation $V_M = M_T/\rho$, where M_T is the total molecular weight of all the elements [16]. The trend of the molar volume for the prepared glass samples is depicted in Fig. 2. It can be observed from the figure that the molar

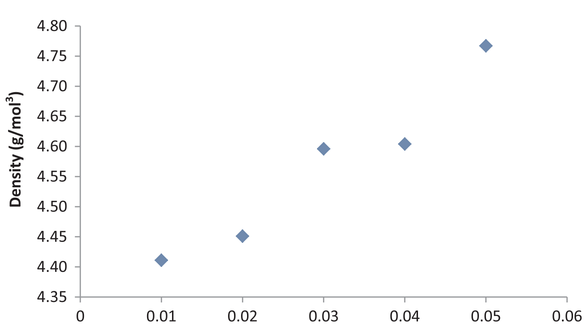

Fig. 1. Density of the glass systems at different concentration of Dy^{3+} NP.

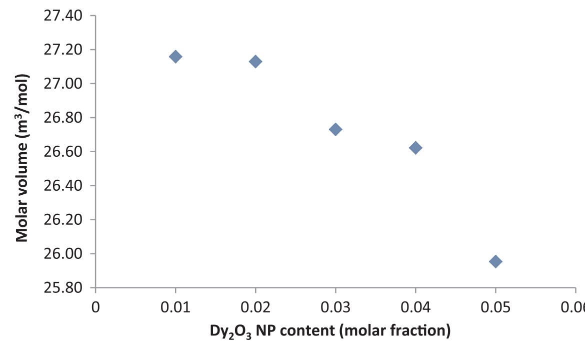

Fig. 2. Molar volume of zinc borotellurite glass systems against molar fraction.

volume of the glass systems decreases as the addition of Dy_2O_3 NP. The decrement trend of the molar volume of the glass systems gives an opposite trend as expected since the variation of the molar volume is mainly influenced by the rate of change of density and the molecular weight [18].

From the result of the density, the other related physical properties can be determined such as rare earth (RE) ion concentration, polaron radius, inter nuclear distance and also the field strength. All of these parameters can be calculated through the equation listed as below [19]:

Ion concentration, N:

$$N\ (\text{ion/cm}^3) = (\%mol\ of\ RE)\frac{(Avogadro's\ number)(glass\ density)}{(glass\ average\ molecular\ weight)} \quad (1)$$

Polaron radius, r_p:

$$r_p\ (\text{Å}) = \left(\frac{1}{2}\right)\left(\frac{\pi}{6N}\right)^{1/3} \quad (2)$$

Internuclear distance, r_i:

$$r_i\ (\text{Å}) = \left(\frac{1}{N}\right)^{1/3} \quad (3)$$

Field strength, F:

$$F\ (\text{cm}^{-2}) = \left(\frac{Z}{r_p^2}\right) \quad (4)$$

As listed in Table 1, the value of the RE ion concentration increases with the mol fraction of Dy_2O_3 NP, whereas the polaron radius and internuclear distance of the Dy^{3+} ions are found to decrease. The increment value of RE ion concentration indicates that the RE ions (Dy^{3+}) are spread within the glass matrices [20]. Since the addition of Dy^{3+} ions into the glass systems cause the glass network to become crowded with dysprosium interstices, the internuclear distance between the ions will become closer hence producing more compact glass systems. The decrease in the internuclear distance eventually will lead to the stronger field strength within the glass network [10,21].

3.2. Transmission Electron Microscopy (TEM)

TEM measurement is used to confirm the existence of the nanoparticles in the glass samples. Fig. 3 demonstrates the TEM image for dysprosium oxide NP doped zinc borotellurite glass systems after the formation of the glass. The average size of the nanoparticle is found in the range of 40.41 nm.

3.3. X-ray Diffraction (XRD)

XRD analysis has been done in order to confirm the nature of the glass systems. In this research, the XRD pattern of the zinc borotellurite glass doped with dysprosium oxide nanoparticles is recorded in the range of $20° \le \theta \le 80°$. The XRD pattern of the proposed glass is shown

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