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Research paper



Structural and Optical Properties of Nd^{3+} Doped Lead Borotellurite Glass Containing Silver

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Abstract

Nd^{3+} doped lead borotellurite glasses which contained silver had been prepared successfully by using conventional melt-quenching method with the chemical composition of (69.5-*x*)TeO_2-20B_2O_3-10PbO-0.5Nd_2O_3-*x*$AgNO_3$ (where *x* = 0, 0.5, 1.0, 1.5, 2.0 and 2.5 mol%) and varied silver nitrate content. The structural properties had been studied through X-ray diffraction (XRD) analysis and Fourier Transform Infrared (FTIR) spectroscopy. Meanwhile, the optical properties of the glass samples had been characterised by Shimadzu UV-1800 UV-Vis spectrophotometer in the range of 190 – 1100 nm where the absorption peaks were obtained and band assignments of each peak were determined from the absorption spectra. The XRD pattern had been used to confirm the amorphous nature of the glass samples. There was no sharp peak being observed in XRD pattern of the glass samples which had confirmed the amorphous nature of the glass. FTIR spectra were used to analyse the functional groups that existed in the glass samples. They had revealed the presence of Te - O - Te or O - Te - O , Ag - O , Te - O - Pb , BO_4 and the characteristics of the hydrogen bond in the prepared glass samples.

Keywords: *Borotellurite glasses, Neodymium, XRD, FTIR.*

1. Introduction

Glasses that contain metallic nanoparticles (NPs) and rare-earths ions (REIs) are remarkable since the NPs could improve the nonlinear properties and change the luminescent characteristic of the material. Potential energy transfer among NPs and REI grows by the enhancement of local field that acts on the REI placed in the locality of the NPs [1]. The luminescent enhancement of Eu^{3+} in borate glasses which contain silver NPs has been reported by O.L. Malta et al. [2]. Several reports on the REI luminescent enhancement due to silver or gold NPs in glasses were recorded and reported by numerous authors [3-5].

In order to obtain large luminescence from REI hosted in glasses, heavy metal oxide glasses are well thought as a promising candidate, since it can exhibit low cut-off phonon energy. Therefore, glasses which are based on tellurium oxide TeO_2 have recently been used as host materials due to their low phonon energy (~700 cm^{-1}) which are helpful to obtain efficient luminescence, good optical quality with high refractive index, extended infrared transmittance (0.35 – 6.0 µm), slow crystallization rate, durability, medium processing temperature (800-900°C), high mechanical strength and large corrosion resistance as compared to the other glass hosts. This type of glass deserves great attention since it is a promising material for photonic applications [6]. For the purpose of forming a glass, tellurite oxide needs the transition of metal oxide or the addition of glass former oxide. Thus, borate oxide which is a common glass former oxide was introduced into the tellurite glass to increase its system ability. In addition, the incorporation of zinc oxide might also improve the glass forming ability and decrease the crystallization rates in a borotellurite glass matrix [7].

To study on the influence of silver NPs on the optical absorption of rare-earth in tellurite glass, Neodymium (Nd^{3+}) had been chosen due to its sufficient lasing emissions in the visible and infrared regions since it exhibited prominent emission at around 1060 nm corresponding to $^4F_{3/2} \rightarrow {}^4T_{11/2}$ transition [8]. Along with these, Nd^{3+} ions could give lasing emission at 1800 nm, 1350 nm and 880 nm. The efficient laser required long fluorescence lifetime for $^4F_{3/2}$ level of Nd^{3+} ion which was influenced by non-radiative decay due to multiphonon relaxation and energy transfer process i.e. self-quenching of emission due to Nd^{3+} ions pair interaction [6].

Until now, various studies have been made on the luminescent properties of Nd^{3+} ions in a variety of glass systems, including silicate, phosphate, borate, tellurite, fluoride and chalcogenide glasses. In the present work, Nd^{3+} ions doped lead borotellurite glass were prepared and extensively studied on the effects of silver nanoparticle on the structural and optical properties of TeO_2-B_2O_3-PbO-Nd_2O_3 glass system doped with Nd^{3+} using X-ray diffraction, Fourier transform infrared (FTIR) spectroscopy and UV-Vis spectrophotometer.

2. Experimental

The Nd^{3+} doped lead borotellurite glass sample was prepared by using the melt-quenching technique with composition of (69.5-*x*)TeO_2-20B_2O_3-10PbO-0.5Nd_2O_3-*x*$AgNO_3$ (*x* = 0, 0.5, 1.0, 1.5, 2.0 and 2.5 mol%). Appropriate amount of chemical powder were weighted by using digital weighing scale. The powder was mixed thoroughly about one hour to obtain 10 g of mixed-powder. The

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mixture was melted in an alumina crucible inside a preheated electric furnace at 1000°C for 30 min. The melt was then poured into a stainless steel mould and annealed for 3 hours at 400°C to remove the mechanical and thermal stress until the good qualities of transparent glasses were obtained. Samples were then being allowed to cool down slowly to room temperature in the furnace. The samples of each composition were prepared and well-polished with various types of sand papers to perform detailed spectroscopic studies.

The density of glass sample was measured at room temperature by using the Archimedes principle and distilled water was used as the immersion liquid. The weight of the glass in air and in water was recorded. All measurements were repeated three times and the average density was recorded. The accuracy of the results in duplicate measurements was 0.001 g/cm^3. The corresponding molar volumes (V_m) were calculated by using relations $V_m=M/\rho$, where M was the molecular weight and ρ, the density of the corresponding glass samples.

The structure of the glass was investigated using X-ray diffraction (XRD) and Fourier Transform Infrared Spectroscopy (FTIR) using Perkin-Elmer Spectrum 100 FT-IR Spectrometer system with a resolution of 4 cm^{-1} in the 400-4000 cm^{-1} range. The optical absorption of Nd^{3+} doped borotellurite glasses were recorded in the range of 190 – 1100 nm intervals using Shimadzu 3101 UV-VIS NIR scanning spectrophotometer.

3. Result and Discussion

3.1. Density and Molar Volume

Table 1 shows the variation of densities and molar volume with respect to silver content. As tabulated in Table 1, the density shows the increasing trend up to 1.0 mol% from 4.969 gcm^{-3} to 5.053 gcm^{-3}. Then, it suddenly dropped at 1.5 mol% of $AgNO_3$ to 5.028 gcm^{-3}. By increasing some amount of $AgNO_3$, the formation of non-bridging oxygen (NBO) took place and caused the modification of glass network to bring extra compactness with higher density but shrinkage in free volume [9]. However, the decreasing value of density at 1.5 mol% might be due to the fact that an occupation of Ag between 1.0 to 1.5 mol% in the free space region in the network structure caused the compact reduction of the glass network [10]. Nevertheless the density of glass had shown an increment as it increased from 5.031 gcm^{-3} at 2.0 mol% to 5.071 gcm^{-3} at 2.5 mol% of $AgNO_3$. The molar volume was found to be decreasing from 29.642 cm^3/mol to 29.169 cm^3/mol with the increasing concentration of $AgNO_3$ from 0.0 mol% to 1.0 mol% before it started to increase to 29.324 cm^3/mol at 1.5 mol%. These results might be due to incorporation of Ag of radii 1.65 pm to replace Te of radii 1.23 pm which had the tendency to modify the glass network [13]. The similar trend of results has been observed by Yusoff et al. [11].

Table 1: Density and molar volume of Nd^{3+} doped borotellurite glass

Sample	Density (g/cm^3)	Molar Volume (cm^3/mol)
S1 (0.0 mol%)	4.969	29.642
S2 (0.5 mol%)	4.980	29.586
S3 (1.0 mol%)	5.053	29.169
S4 (1.5 mol%)	5.028	29.324
S5 (2.0 mol%)	5.031	29.317
S6 (2.5 mol%)	5.071	29.096

3.2. X-Ray Diffraction

The XRD analysis was done to confirm the amorphous or crystalline state of the materials. The X-ray diffraction pattern of Neodymium (Nd^{3+}) ions doped lead borotellurite glass was recorded in the range $10°\leq\theta\leq90°$. As seen in Figure 2, the XRD patterns showed broad humps without any sharp peak which confirmed the amorphous structure of the samples. The absence of sharp peaks from the XRD spectra agreed on the non-existence of crystalline phase. Broad diffusion at lower scattering angles had indicated the presence of long range structural disorder which was the characteristic of an amorphous angle [12]. Therefore, this sample could be classified as pure amorphous which showed a good agreement with previous researches [13].

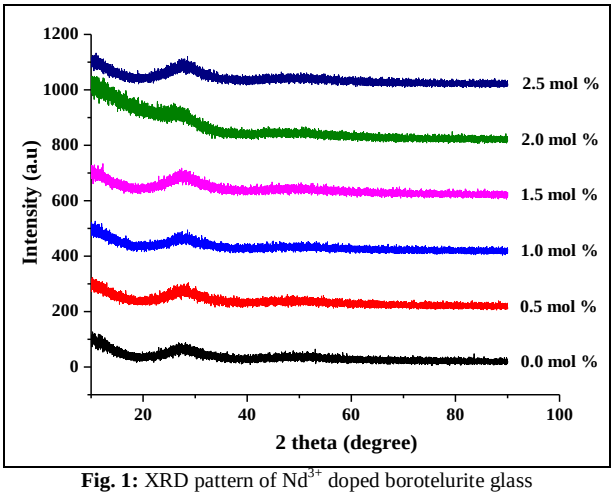


Fig. 1: XRD pattern of Nd^{3+} doped borotelurite glass

3.3. Photoluminescence Spectra

With the Nd_2O_3 concentration fixed at 0.5 mol% and by varying the concentration of silver NPs, a fluorescence spectrum was obtained. Figure 2 shows the absorption spectra of Nd^{3+} doped borotellurite glass with and without silver NPs. From the spectra, it exhibited nine emission transitions under 406 nm light excitation. The emission bands found were centred at 431, 471, 521, 587, 628, 684, 749, 810, 879 nm which corresponded to the $^4I_{9/2} \rightarrow {}^2P_{1/2}$, $^4I_{9/2} \rightarrow {}^2K_{15/2}$, $^2G_{9/2}$, $^2D_{3/2}$, $^4I_{9/2} \rightarrow {}^4G_{9/2}$, $^4I_{9/2} \rightarrow {}^4G_{7/2}$, $^4I_{9/2} \rightarrow {}^4G_{5/2}$, $^2G_{7/2}$, $^4I_{9/2} \rightarrow {}^2H_{11/2}$, $^4I_{9/2} \rightarrow {}^4F_{9/2}$, $^4I_{9/2} \rightarrow {}^4F_{7/2}$, $^4S_{3/2}$, $^4I_{9/2} \rightarrow {}^4F_{5/2}$, $^2H_{9/2}$ and $^4I_{9/2} \rightarrow {}^4F_{3/2}$ transitions. The intensities of all absorption peaks were observed to be slightly affected by the addition of $AgNO_3$. Among the transition, the emission band which corresponded to $^4I_{9/2} \rightarrow {}^4G_{9/2}$ (587nm) transition possessed the highest intensity and this band was enhanced significantly. This similar observation was also reported by N.A. Azmi et al. [14]. Interestingly, the absorption spectra showed an enhancement as the concentration of Ag NPs increased. It might be possibly due to the lower rigidity of the glass system resulted from higher $AgNO_3$ concentration.

As the emission was at 587 nm, associated with $^4I_{9/2} \rightarrow {}^4G_{5/2}$, $^2G_{7/2}$ had shown the largest luminescent enhancement for most of the transitions observed in the visible region and the transition was more affected by the NPs, thus it was selected for the measurement emission spectrum of Nd^{3+} doped lead borotellurite glass.

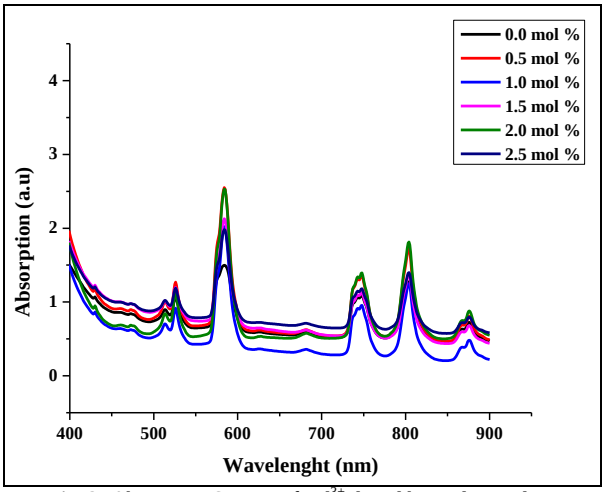


Fig. 2: Absorption Spectra of Nd^{3+} doped borotelurite glass

3.4. FTIR

The information on the arrangement of the glass structure was discovered by the FTIR spectroscopy. The glass samples were characterized in the wave number ranging from 400 cm^{-1} to 4000 cm^{-1} in order to obtain the FTIR spectra as presented in Figure 3. The spectra consisted strong absorption bands at 460-476 cm^{-1}, 552-575 cm^{-1}, 1177-1209 cm^{-1}, 1350-1371 cm^{-1}, 2872-2880 cm^{-1} and 2940-2960 cm^{-1}. The observed IR bands and the band assignments were presented in Figure 3 and Table 2, respectively. These observed bands were assigned based on the available literature on borotellurite glasses [13,15-19].

The band around 460-476 cm^{-1} was assigned to anti-symmetric vibrations of Te-O-Te linkages constructed by two inequivalent Te-O bonds in trigonal pyramid of TeO_3 units associated with the non-bridging oxygen, which was the characteristic peak of TeO_2 based glasses [13,20]. The band observed in the region 552-575 cm^{-1} was assigned to the symmetrical bending vibrations of Ag-O bond [13,21]. The band observed in IR of 1177-1209 cm^{-1} and 1350-1371 cm^{-1} corresponded to the Te-O-Pb stretching vibrations and the vibration of the B-O bond in BO_3 groups respectively. Similar results were reported by Fudzi, F.M et al [7] and Pawar et al [22]. The range between 1350-1371 had proven the formation of non-bridging oxygen (NBO) in the borotellurite glass system [7]. Meanwhile, the last peaks positioned at 2872-2880 cm^{-1} and 2940-2960 cm^{-1} were due to the characteristic of the hydrogen bond in the glass samples [13,23].

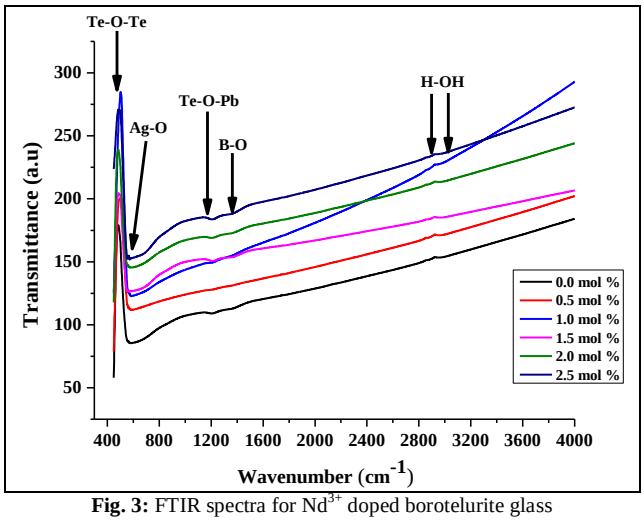


Fig. 3: FTIR spectra for Nd^{3+} doped borotelurite glass

Table 2: Assignments of the FTIR band for Nd^{3+} doped borotelurite glass

Wavenumber (cm^{-1})	Assignments
460–476	Stretching vibrations modes of Te - O - Te or O - Te - O
552–575	Bending vibrations of Ag - O
1177–1209	Streching vibrations of Te - O - Pb
1350–1371	B-O stretching vibration in BO_3
2872–2880	Hydrogen bond
2940–2960	Hydrogen bond

4. Conclusion

A series of Nd^{3+} doped borotellurite glass which contained Ag NPs was prepared by using melt quenching technique where the concentration of Nd_2O_3 was fixed while the concentration of silver nanoparticles was varied from 0.5 mol% to 2.5 mol%. The glassy nature of the prepared glass has been proven by the presence of broad hump in the XRD pattern. The effects of embedding NPs to the luminescent properties of the glass had been investigated through photoluminescence measurements at 406 nm excitation wavelength. The photoluminescence spectra showed nine emission bands centred at 431, 471, 521, 587, 628, 684, 749, 810, 879 nm had corresponded to the $^4I_{9/2} \rightarrow {}^2P_{1/2}$, $^4I_{9/2} \rightarrow {}^2K_{15/2}$, $^2G_{9/2}$, $^2D_{3/2}$, $^4I_{9/2} \rightarrow {}^4G_{9/2}$, $^4I_{9/2} \rightarrow {}^4G_{7/2}$, $^4I_{9/2} \rightarrow {}^4G_{5/2}$, $^2G_{7/2}$, $^4I_{9/2} \rightarrow {}^2H_{11/2}$, $^4I_{9/2} \rightarrow {}^4F_{9/2}$, $^4I_{9/2} \rightarrow {}^4F_{7/2}$, $^4S_{3/2}$, $^4I_{9/2} \rightarrow {}^4F_{5/2}$, $^2H_{9/2}$ and $^4I_{9/2} \rightarrow {}^4F_{3/2}$ transitions respectively. Incorporation of silver nanoparticles enhances the intensity of these emission bands and the enhancement increases with an increment of silver NPs content. In general, it can be said that the optical properties of the borotellurite glass has been enhanced with the addition of silver nanoparticles in the glass matrix. The glass structure has been well studied as the FTIR results revealed the existence of bending and stretching vibration of the TeO_3 group.

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