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Effect of Tungsten on Physical and Optical Properties of Niobate Tellurite Glasses

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Abstract

In the present work, three optically transparent tungsten niobate tellurite glasses with compositions of (90-x)TeO2-10Nb2O5-xWO3 (x = 0, 6, 12 mol%) were synthesized by conventional melt quenching technique. For all these three glass samples, the structural and the compositional dependence of different physical parameters such as density, molar volume and ionic packing density have been analyzed. It been observed that the amorphous nature of the samples has been confirm by X-ray diffraction analysis. The physical properties of the glasses were evaluated and the change in density ( ), molar volume ( ) and ionic packing density ( ) in these glasses indicates the effect of WO3 different content registered on the glasses structure. The highest refractive indexes value 2.171 at 632.8 nm was measured for 78TeO2–10Nb2O5–12WO3 glass. It is found that the addition of WO3 concentration to TeO2-Nb2O5 glass system increase the value of density, molar volume as well as ionic packing density. The refractive index value of the glass increase with tendency for small optical energy band gap.

Keywords: *Physical, Refractive Index, Structure*

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INTRODUCTION

In the past two decade, tellurium oxide (TeO2) based glass have received considerable interest for their promising potential in photonics and optoelectronics field due to its unique physical, chemical and optical properties compare to other host materials such as silicate, phosphate and borate glasses. This because TeO2 offer large infrared transparency up to 6µm, high refractive index, high nonlinear refractive indices, low phonon energy, low melting temperatures and excellent as host matrix for active dopant (Gayathri Pavani, Sadhana and Chandra Mouli, 2011; Selvaraju and Marimuthu, 2013; Maheshvaran et al., 2014). These outstanding properties make tellurite glasses are most favored use in application relating to solid state lasers, optical amplifiers, non-linear optical devices and optical data storage (Lezal et al., 2001; Ersundu et al., 2011; Zamyatin et al., 2016; Sayyed et al., 2017). Unlike other glass formers, pure TeO2 does not have ability to form glass under normal condition easily without addition of another oxide which acts as modifier in the system. The present of modifiers like alkali, alkaline earth and transition metal oxide/heavy metal oxide into TeO2 networks is crucial to initiate the formation of a glass from TeO2 powder (El-Mallawany RA, 2002; Nayar et al., 2016). Furthermore, combination of different oxide also improve the linkage between Te-O chains which lead to increase in glass forming ability (Manikandan, Ryasnyanskiy and Toulouse, 2012). Recently, growing interest among researcher are focused on producing new tellurite glass of improved optical performance with high refractive index and an excellent nonlinear properties (Hayakawa et al., 2010; Tan et al., 2013; Miedzinski, Fuks-Janczarek and Said, 2016; Sadhu et al., 2017) Such interest because these glass have been demonstrated to exhibit third order nonlinear optical susceptibility ($\chi 3$) higher magnitude compare to silicate glass (Stepien et al., 2010; Manning, Ebendorff-Heidepriem and Monro, 2012). Since studies showed various properties of glassy materials can be observed by modifying their composition, it was reported that, the addition of transition/heavy metal oxide (Nb2O5, MoO3, Bi2O3, WO3, TiO2, BaO) into tellurite glass can enhance the third order nonlinear properties considering large number of polarized metal ions taking part in the network (Kim and Yoko, 1995; Berthereau et al., 1996; Yousef, Hotzel and Rüssel, 2007; Xu et al., 2011a; Chen et al., 2014). This can be explained due to incorporation between high polarizability of Te4+-ions with other heavy metal ions (Pb2+, Bi3+) that can be easily polarized or transition metal ions (Ti4+, Nb5+, W6+) with empty d orbital (Villegas and Navarro, 2007; Wang et al., 2009; Chen et al., 2010). Among several metal oxide, tellurite glass containing niobic oxide Nb2O5 or tungsten oxide WO3 is being studied extensively because such addition seem to show remarkable changes in both physical and optical properties of these glasses. According to the literature, the insertion of Nb2O5 not only stabilizes the random network by connect Te-O chains in the structure but also improves vitrification and optical nonlinearity of the tellurite glass (Lin et al., 2004, 2009; Umair and Yahya, 2015; Elkhoshkhany et al., 2018). Also, the addition of WO3 into tellurite glass show significant improvement in term of thermal, chemical stability and devitrification resistance (Ersundu et al., 2010; Çelikbilek et al., 2011b, 2011a; Çelikbilek, Erçin Ersundu and Aydin, 2013; Zaki et al., 2018). However, the enhancement of optical properties is greatly influence by the composition and the network structure of the glasses. Keeping the above fact in focus, it is important to study and understand whether the nonlinear optical and other related properties that are critical for a glass such as structure and physical properties can be increased for tellurite glasses when they contain both Nb2O5 and WO3. Therefore, we have prepared ternary glass system within composition of TeO2-Nb2O5-WO3. The present study aims to determine the density, molar volume, ionic packing density, hardness, linear optical refractive index and optical band gap by varying the concentration of tungsten oxide in niobate tellurite glass. Afterwards, some of such glasses will be selected as the best for a further study focused on their nonlinear optical properties.

EXPERIMENTAL

The WO3 doped glasses of the form (90-x)TeO2- 10Nb2O5-xWO3 with x = 0; 6, 12 mol % and 0 ≤ x ≤ 12 mol % as shown in Table 1 and all the glasses were prepared using the conventional melt-quenching technique. The starting materials using high purity chemical of TeO2 (purity >99%), Nb2O5 (purity 99.99%) and WO3 (purity >99%) are commercially provided in the powder forms. An appropriate amount of raw powdered were weighed in stoichiometric ratio in 10 g batch each separately and thoroughly milled within 20 minutes to obtain homogeneous chemical mixtures. Each batch were then placed inside a platinum crucible before being heated in an electrical furnace at a temperature of 900 °C for 30 minutes. After obtaining the required viscosity, the molten fluid were subsequently poured onto stainless steel metal plate for annealing process in under 3 hours at 300 °C before being slowly cooled down until reach room temperature to released internal stress induced in the glass during melt quenching that could cause glass to crack. Further, each transparent with slight yellowish color formed glasses were carefully checked for any bubbles in them to avoid from defects prior being cut to 20 mm × 20 mm × 2.0 mm size by using low speed saw machine and mechanically polished to a mirror finish using SiC/water.

Table 1. Composition of TNW Glass System**.**

Sample	Nominal Composition (mol%)		
	TeO_2	Nb_2O_5	WO_3
TNW1	90	10	0
TNW2	84	10	6
TNW3	78	10	12

X-ray Diffraction (XRD) analysis is performed by using Siemens Diffractometer D5000 using Cu-Kα radiations (λ≈1.54 Å) at 40 kV and 100 mA, with scanning angle of 2θ ranges between 10-80°. The glass density (in g/ cm3) is determined by using the Archimedes method with water is used as an immersion liquid using distilled water, = 1.000 g/cm3 as the reference liquid with single pan balance. The density, ρ is calculated using equation (1) (Shelby, 2007)

$$\rho = \frac{W_a}{W_a - W_b}\rho_x \qquad (1)$$

where is the weight of glass sample in air and is the weight of the sample when immersed in distilled water.

The glass composition depends on the variation of molar volume which indicates the free excess volume formation according to the size of interstices in the glass structure as molar volume shown the information of glass modification (Sanad et al., 1985). The molar volume, is determined by equation (2) (El-Diasty, Abdel Wahab and Abdel-Baki, 2006)

$$V_m = \frac{M_{av}}{\rho} \qquad (2)$$

where is the molecular weight of the sample.

The ionic packing density (vt) is determined via equation (3) (Gowda, 2013)

$$v_t = \left(\frac{1}{V_m}\right)\sum(v_i x_i) \qquad (3)$$

and

$$v_i = \left(\frac{4\pi N_A}{3}\right)[x r_m^3 + y r_o^3] \qquad (4)$$

where xi is the mole fraction (mol%).

The refractive indices of the glasses were measured by the Brewster's angle technique using Helium-Neon Diode Laser at 632 nm wavelength as a source. This technique can be express in equation (5) (Mallur et al., 2015)

$$\tan\theta_b = \left(\frac{n_1}{n_2}\right) \qquad (5)$$

where = refractive index of the glass medium, = refractive index of the air and = angle where reflected incident beam of unpolarized light is at fully polarized.

Glass refractive index (n) in terms of optical band gap ( ) yields: (Gupta et al., 2017)

$$\frac{(n^2-1)}{(n^2+2)} = 1 - \sqrt{\frac{E_{opt}}{20}} \qquad (6)$$

RESULT AND DISCUSSION

XRD Analysis

Figure 1 depicts the typical XRD pattern for sample TNW1, TWN2 and TWN3 with 0, 6 and 12 mol% of WO3. The diffraction pattern in the presence of a broad peak around 15 to 35° without any discrete or continuous sharp peaks confirms the amorphous nature or structural disorder of prepared glass (Surendra Babu et al., 2010). It clearly indicates the absence of long range atomic arrangement and the lack of periodicity of three dimensional networks, where no well-defined planes in the structure on or around which the constituent's atoms are regularly arranged (Sidek et al., 2009). Furthermore, the presence of W6+ ions does not alter significantly the physical appearance of the glasses.

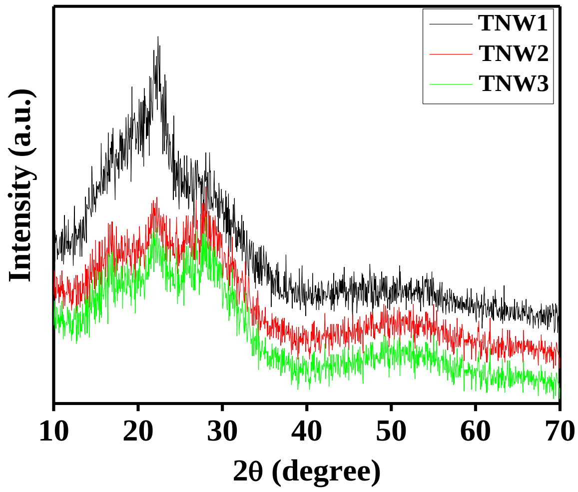

Fig 1: Typical XRD pattern of sample TNW

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Physical Properties

It was found that in the Table 1, both the density and the molar volume increase with the increase in WO3 concentration while in the general the density and the molar volume are inversely related. The increase in density from 5.32 to 5.51 g/cm3 with the increase of WO3 content is attributed to the higher molecular masses of WO3 (231.84 g/mol) than TeO2 (159.60 g/mol) is shown in Figure 2. Besides, the increase in density also is an indication of the formation of Te-O-W linkage which the participation of W6+ in the network formation is favorable because Te and W have comparable electronegativities (2.1 and 2.0 respectively) and can therefore substitute for each other in covalent bonding with oxygen atom (Shaltout et al., 1995; Sokolov et al., 2006). As result, more tightly packed structure is produce and packing density of the glass become higher.

In addition, the increase in molar volume from 32.02 to 32.46 cm3/mol with the increase of WO3 concentration is ascribed to the change in glass networks indicate the formation of non-bridging oxygen (NBO) atoms which opens up the tellurite network. As WO3 concentration increase will also increase the creation of non-bridging oxygen. Consequently, the glass rigidity is decrease. The data trend of both molar volume and density agree with reported earlier by Arshpreet et. al. (Kaur et al., 2013). The source of error for both density and molar volume is due to instrumental with the experimental error ~ 0.02 %.

Table 2. **Physical Properties of The TNW Glasses.**

Samples	Density, ρ ($\pm$0.02 g/cm^3)	Molar Volume, V_m ($\pm$0.02 cm^3/mol)	Ionic Packing Density, V_t ($\pm$0.01)
TNW1	5.32	32.02	0.0050
TNW2	5.42	32.22	0.0051
TNW3	5.51	32.46	0.0052

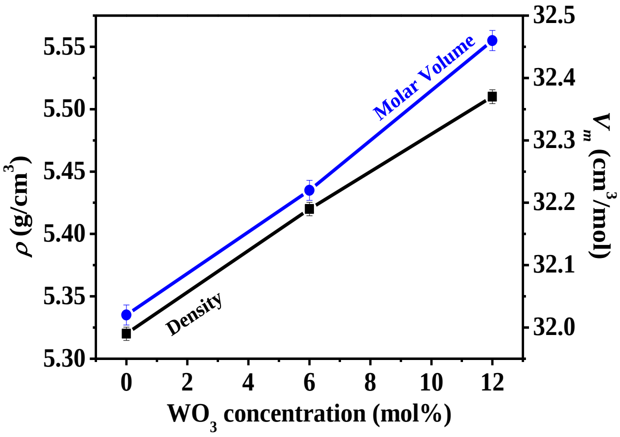

Fig 2. WO3 Concentration Dependent Density and Molar Volume of the Glasses.

The increase in ionic packing density (a measure of glass compactness) from 0.0050 to 0.0052 due to the increase of WO3 concentration as shown in Table 1 is attributed to the slightly smaller ionic radius of W6+ (0.65 Å) than Te4+ (– 0.66 Å) and Nb5+ (– 0.64 Å) ions is displayed in Figure 3 and the source of error due to instrumental with experiment error ~0.01 %. The larger the ionic radius is the higher the creation of excess volume which originates from the misfit into the available glass network (Yusoff and Sahar, 2015). Thus, the compactness of the glass is reduced. Moreover, the increase in ionic packing density (compactness) with the increase of WO3 concentration indicates that W6+ ions have almost filled the available excess volume (Samee, Edukondalu and Rahman, 2012).

Refractive Index and Energy Band Gap

The values of linear refractive indexes of glass samples are listed in Table 2. The refractive indexes increase with increasing the WO3 content. Linear refractive indexes with the addition content of WO3 increase from 2.045 to 2.171 as shown in Figure 4. This increase can be related to the structural change in the glass matrix when a lattice modifier is added especially due to the growth of the anionic quantities present in the glass lattice. It is concluded that the WO3 content contributes to increase the refractive index. In fact, Yanfei et. al. (Chen et al., 2008) measured the linear refractive index and verified that the increase in concentration lattice modifiers WO3 can cause an increase in refractive index values.

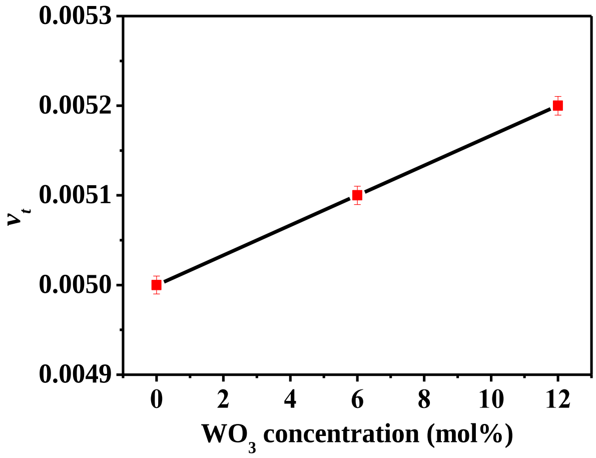

Fig 3. Dependence of ionic packing density on WO3 concentration.

It is well known that energy band gap is an important parameter to describe the nature of the nonlinear refractive index and third order nonlinear susceptibility χ3 (Dimitrov and Sakka, 1996). As show in Table 2, the value of energy band gap is decrease as refractive index increase when the WO3 concentration is increase. The highest value of corresponds to the glass with higher molar proportion of TeO2 (TWN1). For lower TeO2 content, progressively decrease when TeO2 is substituted by WO3. The increasing incorporation of WO3 is mainly responsible for the diminishing. This change is due to the formation of non-bridging oxygen in the glass structure increasing with increasing in the WO3 content that as has been observed for several tellurite glass matrices (Prakash, Rao and Bhatnagar, 2001; Xu et al., 2011b; Elkhoshkhany and El-Mallawany, 2015). Both refractive index and energy band gap data show similar trend in other tellurite glass studies which suggest that the nonlinear optical susceptibility increase with increase in refractive index and decrease in energy band gap (Abdel-Baki, Wahab and El-Diasty, 2006).

Table 3. Linear Refractive Indexes and Optical Energy Band Gap of The TNW Glasses

Samples	Refractive Index, n	Optical Energy Band Gap, E_{opt} (eV)
TNW1	2.045	4.710
TNW2	2.160	4.051
TNW3	2.171	3.994

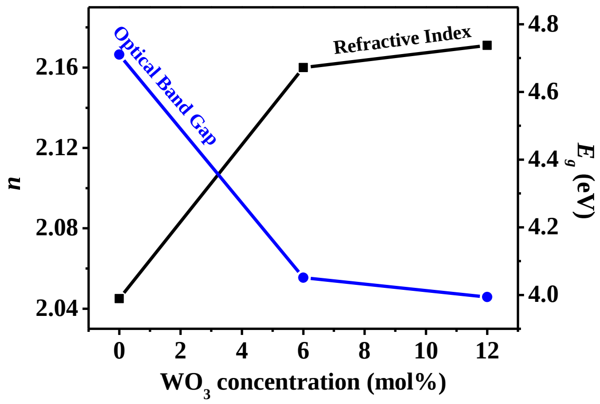

Fig 4. Dependence of Refractive index on WO3 Concentration.

CONCLUSION

Tungsten doped niobate tellurite glasses are prepared using melt-quenching method. The influence of dopants (WO3) concentration to improving the physical properties is examined. The XRD spectra confirmed the amorphous nature of synthesized glasses. Via doping the physical properties and refractive index are considerably improved. Density and molar volume increase with increase in WO_3 concentration due to its higher molecular weight. The maximum refractive index value of 2.171 was obtained for $78TeO_2$–$10Nb_2O_5$–$12WO_3$ glass. The value of optical energy band gap decreases with increase in WO_3 concentration due to the formation of NBO. Our findings suggest that these new niobate tellurite glass compositions are suitable for the fabrication of solid state lasers and amplifiers.

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