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Influence of Eu^{3+} on Physical and Thermal Properties of Borotellurite Glass Containing Manganese Oxides Nanoparticles

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Abstract. The influence of the europium concentration on the physical and thermal properties of manganese nanoparticles (Mn_3O_4 NPs) embedded borotellurite glass are studied. Glasses with composition $(59\text{-}x)TeO_2\text{-}30B_2O_3\text{-}10MgO\text{-}xEu_2O_3\text{-}1Mn_3O_4$ (where x = 0.5, 1.0 and 1.5 mol %) are prepared by melt-quenching method and characterized using X-ray Diffraction (XRD) and Differential Thermal Analysis (DTA). The XRD pattern confirms the amorphous nature of all samples.The physical properties such as density (ρ) and molar volume (V_m) are calculated. The thermal properties of borotellurite glasses show that the glass with 1.0 mol % of Eu_2O_3 possess the highest stability.

1. Introduction

Presently, borotellurite glass represents favorable compromise glass materials for various scientific and technological applications. Burger *et al.* [1] revealed that borotellurite glass system is one of the best hosts because of simple B_2O_3 and TeO_2 structure formation and enhanced stability. Besides they display low phonon energy, superior thermal stability and high chemical durability [2]. Halimah *et al*. [3] found TeO_2 contents dependent significant improvement in the rigidity of borotellurite glass structure with reduced phonon energy loss. The presence of both B_2O_3 and TeO_2 structural units in the glass leads to a complex speciation within the amorphous network [4]. Borotellurite glass is emerged as a favourable host for accommodating amount of rare earth. Among the rare earth family, trivalent Eu^{3+} ion is a well-known activator with simple electronic transitions. The Eu^{3+} ions possess prominent laser emissions in the orange or red region [5] and narrow band emission. The aims of the present study are to investigate the concentration dependent of europium on physical and thermal properties borotellurite glass that containing manganese oxides nanoparticles.

2. Experimental

Starting powder of (TeO_2) (99.9%), Boric acid (H_3BO_3) (99.0%), Magnesium oxide (MgO) (99.0%), Europium oxide (Eu_2O_3)(99.0%) and Mangnese oxide (Mn_3O_4)(99.0%) were used as nominal composition. Magnesium borotellurite glasses of compositions $(59\text{-}x)TeO_2\text{-}30B_2O_3\text{-}10MgO\text{-}xEu_2O_3\text{-}1.0Mn_3O_4$ (where x = 0.5, 1.0 and 1.5 mol %) were prepared by melt quenching technique. The batched mixture around 20 g was placed in an platinum crucible and melted in a furnace at 900 °C for 1 h. The glass samples were annealed for about 3 h at 300 °C to remove the thermal and mechanical strains. Finally, the samples was cut, ground and polished for the measurements. The amorphousity of the sample is identified by X-ray diffraction technique using the Siemens Diffractometer D5000 system operating at 40 kV and 30 mA in the range of 10 º - 90 º. The density of each glass is measured by Archimedes principle using relation [6],

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

where ρ_x is the density of immersion liquid (distilled water ρ_x = 0.9987 g.cm^{-3}), W_a is weight of the glass in air and W_b is the weight of the sample when immersed in distilled water. Consequently the molar volume, V_m of the prepared glasses can be calculated by using relation [6];

$$V_m = \sum_i \frac{xiMi}{\rho} \quad (2)$$

where xi and Mi denote the molar fraction and molecular weight of the ith component respectively. The thermal parameters in term of glass transition (T_g), crystallization temperature (T_c) and melting point (T_m) are determined using Perkin-Elmer pyris Diamond TG/DTA 7 Series. All the measurement is carried out at room temperature.

3. Results and Discussion

Fig. 1 shows the typical XRD pattern of the synthesized glass sample of S3.

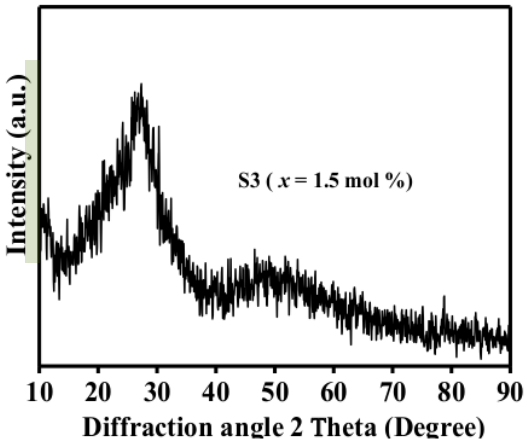

Fig. 1. Typical XRD patterns of S3 glass system.

From Fig.1, the XRD pattern show a broad diffraction band in the range of 25 to 35° which verifies their amorphous nature [7].The obtained glass density and molar volume of the samples are summarized in Table 1.

Table 1. Density (ρ) and molar volume (V_m) of the prepared glass system.

Sample	x (mol %)	ρ (g.cm^{-3}) ± 0.001	V_m (cm^3.mol^{-1}) ± 0.01
S1	0.5	4.708	25.68
S2	1.0	4.706	25.90
S3	1.5	4.749	25.83

From Table 1, the relationship between density and molar volume against Eu_2O_3 concentration can be plotted and presented in Fig. 2. From Fig. 2, it can clearly seen that the glass density and molar volume exhibits an opposing trend. This is in agreement as reported elsewhere [8].

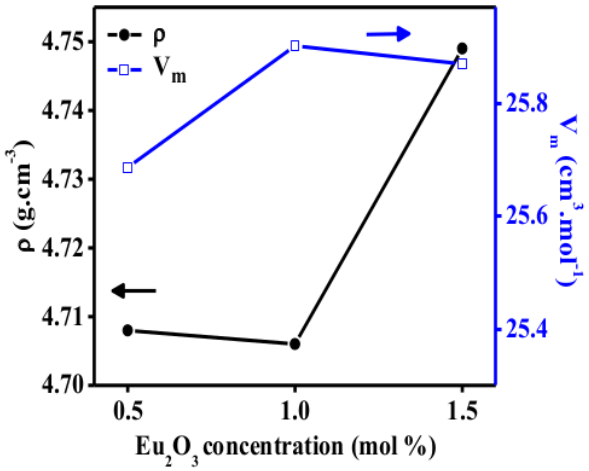

Fig. 2. Density and molar volume as a function of Eu_2O_3 concentration.

The glass density is found to decrease with the increase of Eu_2O_3 concentration up to 1 mol %. This is due to the formation of non-bridging oxygen (NBO) which subsequently reduces the compactness of glass structure. The increasing number of NBO reflected to the increment in molar volume based on the formation of higher space of unoccupied interstitial sites. However, the glass density increases as the concentration of Eu_2O_3 concentration beyond 1.0 mol %. This is associated to the increment of Eu_2O_3 (351.926 g.mol^{-1}) with the expense of TeO_2 (159.60 g.mol^{-1}) which leads to a produce tightly packed glass structure [9].

Fig. 3 illustrates the typical DTA curves for S2. The glass transition temperature, T_g, crystallization temperature, T_c and melting temperature, T_m of S2 have been identified and their value are listed in Table 2.

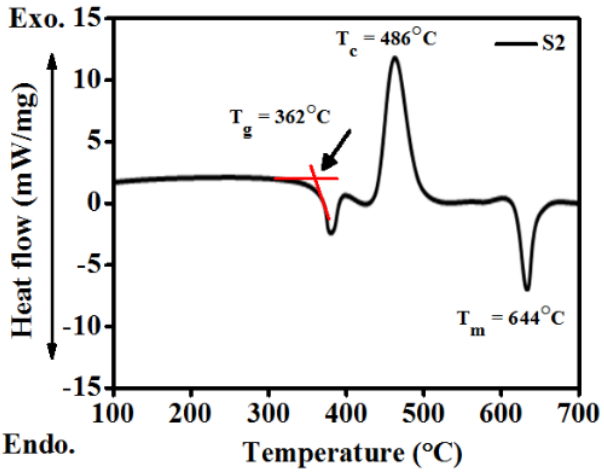

Fig. 3. The typical DTA curves of S2 glass system.

Table 2.Thermal properties of magnesium borotellurite glass system.

Sample	x (mol%)	Temperature (°C) (±1)			$\Delta S = T_c - T_g$	$H_R = \frac{T_c - T_g}{T_m - T_c}$
		T_g	T_c	T_m		
S1	0.5	359	475	648	116	0.67
S2	1.0	362	486	644	124	0.78
S3	1.5	360	478	650	118	0.68

It is clearly seen from the Table 2, that T_g increased from 359 °C to 362 °C as concentration of Eu_2O_3 increases up to 1.0 mol %. The increase in T_g is ascribed to an increase in rigidity of the glass network. The same trend has also been observed for T_c which increases from 475 °C to 486 °C. This result can be explained on the basis of strength of the chemical bonds between components of the considered glass network. However, the decrement part of T_g and T_c are observed beyond 1.0 mol % of Eu_2O_3 which is due to a decrease in the rigidity of the glass network. As a result, NBO atoms in these glasses also increase, thus produce a looser glass structure [10]. Therefore, it requires smaller internal energy to break the glass network which leads to decrease the glass transition temperature, T_g. Meanwhile, the melting temperature, T_m of the glasses is found to be around 650 °C and is comparable to those reported elsewhere [8]. The glass stability ($\Delta S = T_c - T_g$) and Hruby's parameter, $H_R = (T_c - T_g)/(T_m - T_c)$ is a strong indicator to investigate the tendency of glass forming [9]. The calculated glass stability is also listed in Table 2. It is noteworthy to mention that the glass can be considered as a stable glass with good thermal ability if ΔS is greater than 100°C [10].

Conclusion

The XRD pattern confirms the amorphous nature of the prepared glass. The variation density and molar volume due to rupturing lots of BO bonds and creating NBO. The formation of NBO and BO considerably impacted the physical properties of the prepared glass systems. The maximum thermal stability is 124 °C and glass forming tendency is 0.78 at 1.0 mol % concentration of Eu_2O_3.

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