

## Density and Elastic Behaviour of Ternary Tellurite Glass Systems – A Short Review

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**Abstract:** This short review paper presents the elastic properties of various ternary tellurite glass systems prepared using the conventional melt-quenching technique. Their density ( $\rho$ ), molar volume ( $V_m$ ) values were calculated from the characterizations data obtained for the interpretation of physical, and elastic properties of the glass system under study. Ultrasonic pulse-echo technique carried out at room temperature, and at resonating frequency of 5 MHz was used for the determination of the ultrasonic velocities (longitudinal and shear). Elastic (longitudinal, shear, bulk and Young's) moduli, Poisson's ratio, Debye temperature, and a softening temperature of all glass samples have been estimated using the measured ultrasonic velocities and densities. Experimental data shows that most of the elastic moduli of the ternary glasses were found to be composition dependent.

**Keywords:** Density, elastic behaviour, ternary tellurite system

Tellurium oxide ( $\text{TeO}_2$ ) based glasses have been given both technical and scientific interest due to their many unique characteristics which include high dielectric constants, high index of refraction, low temperatures of melting, low phonon energies excellent chemical and thermal stabilities [1-3].

The glasses have wide applications in the fields of nuclear, electronics, acoustic-optics devices and solar energy technologies [4]. Tellurium oxide is considered a conditional glass former that requires a modifier addition for easy glassy state formation [5].

Figure 1 shows the variation of density,  $\rho$  and molar volume,  $V_M$  values of  $\text{TeO}_2$ – $\text{WO}_3$ – $\text{Li}_2\text{O}$  glasses as a function of a) equimolar substitution of  $\text{TeO}_2$  by  $\text{WO}_3 + \text{Li}_2\text{O}$  b) increasing  $\text{Li}_2\text{O}$  at constant  $\text{TeO}_2$  [3].

The high infrared transmission (up to 6  $\mu\text{m}$ ) of tellurite based glasses is due to their low phonon energies, which is also responsible for their low rates of multi-phonon relaxation for excited states of rare-earth [1]. The third-order non-linear optical susceptibility ( $\chi$ ), of these glasses, was reported to be orders of magnitude larger than that of the conventional silicate and borate glasses with values up to 100 times [2].

The addition of metal oxides into the tellurite network not only stabilizes the random network but also provides additional oxygen ions for modifying Te-O coordination number, and improves the third order non-linear optical properties [6].

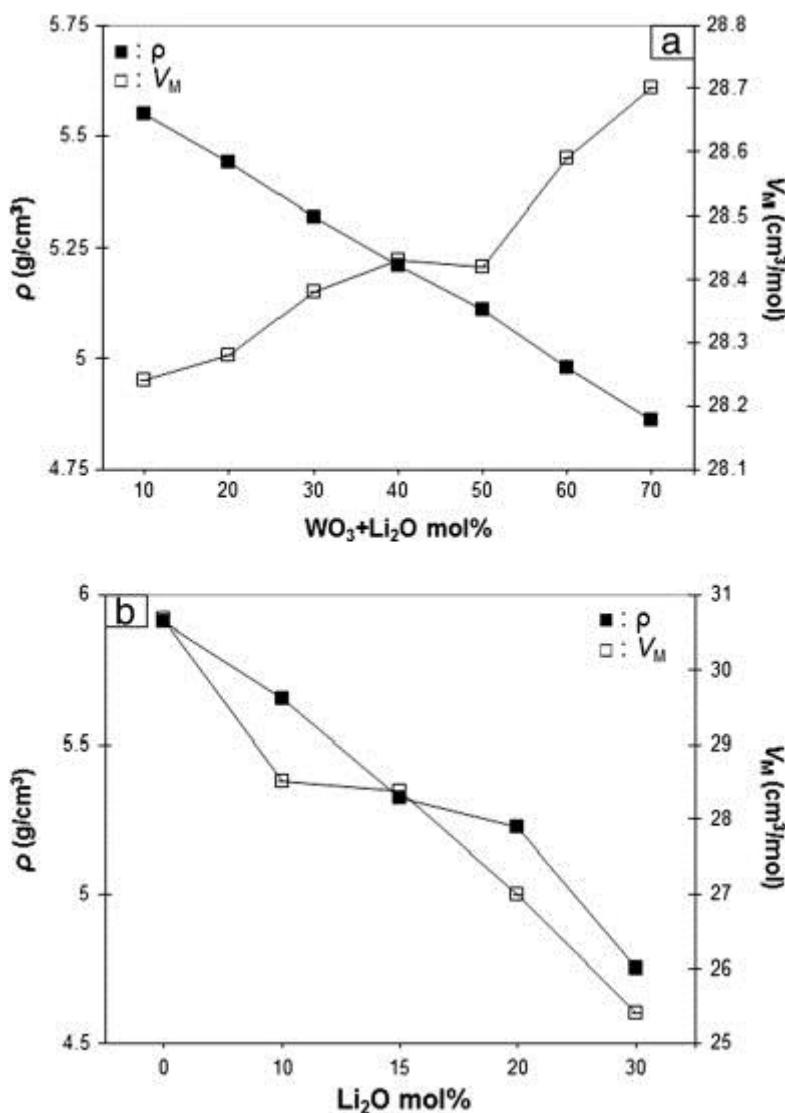


Fig. 1. Variation of density,  $\rho$  and molar volume,  $V_M$  values of  $\text{TeO}_2$ - $\text{WO}_3$ - $\text{Li}_2\text{O}$  glasses as a function of a) equimolar substitution of  $\text{TeO}_2$  by  $\text{WO}_3 + \text{Li}_2\text{O}$  b) increasing  $\text{Li}_2\text{O}$  at constant  $\text{TeO}_2$  [3].

The addition of metal oxides into the tellurite network not only stabilizes the random network but also provides additional oxygen ions for modifying Te-O coordination number, and improves the third order non-linear optical properties [6]. For the acoustic properties (which include ultrasonic velocities, elastic moduli, microhardness, ultrasonic attenuation coefficient, Poisson ration and Debye temperature) study, the technique of ultrasonic pulse-echo has been considered to be among the best tool has been found to be one of the best tools employed [7]. Golding (1976) found that methods of ultrasonic technique are the most powerful way of understanding the amorphous solid materials' unusual properties at low temperatures [8].

The elastic and structural properties of glasses are influenced primarily by the chemical, electrical and physical properties of the constituent materials and their compositions [9]. The strength of non-crystalline solid materials increase as their elastic moduli increases, and thus possibility is there to assess the strength indirectly from their elastic properties. The elastic constant studies of glass materials give the structural information of the non-crystalline

material's structure; this is due to the connections between the elastic constant and interatomic potentials and forces [7]. They also provide information about both the network formers and the modifiers' structural behavior in the glass system under study.

Elazoumi et al, has studied a system of  $[ZnO]_x [(TeO_2)_{0.7}-PbO]_{0.3} ]_{1-x}$  glasses with  $x= 0.15, 0.17, 0.20, 0.22$  and  $0.25$  mol% was fabricated the technique melt quenching [10]. The fabricated glass samples were homogeneous, lime green color and transparent. Density and calculated molar volume values and oxygen packing density are reported in Table 1. It decreases in the values was observed with increasing concentration of ZnO in glass system. Figure 2 presents density and molar volume variation for studied glasses with ZnO mol%. The measured values of the glasses' densities are high as shown in Table 1; it can be seen that the density decreases gradually as the weight fraction of ZnO.

The decrease in the density value from  $6.257$  to  $6.078$   $g\ cm^{-3}$  with ZnO concentration increase is associated with lower molecular weight ( $81.38$  g/mol) ZnO replacement at  $TeO_2$  and PbO expense with  $159.598$  and  $223.1994$  g/mol molecular weight respectively. The molar volume value decreased from  $26.22$  to  $25.40$   $cm^3/mol$  with ZnO concentration increase. Figure 2 shows the density ( $\rho$ ) and molar volume ( $V_m$ ) of the  $[ZnO]_x [(TeO_2)_{0.7}-PbO]_{0.3} ]_{1-x}$  glasses. Decrease in the molar volume might be related to the substitution of larger sized atoms of Te ( $138$  pm), and PbO ( $146$  pm) with smaller atoms of Zn ( $122$  pm).

Table 1: Experimental values of density ( $\rho$ ), molar volume ( $V_m$ ) and oxygen packing density (OPD) of ternary  $[ZnO]_x [(TeO_2)_{0.7}-PbO]_{0.3} ]_{1-x}$  glass system (Elazoumi et al, 2017).

$x$ (mol %)	Density $\rho$ ( $kgm^{-3}$ )	Molar Volume $V_m$ ( $cm^3.mol^{-1}$ )	(O.P.D)
15	6257	26.22	60.822
17	6230	26.03	60.748
20	6191	25.72	60.568
22	6144	25.60	60.395
25	6078	25.40	60.050

The calculated values of the longitudinal (L), shear (G), bulk (K) and Young's (E) moduli of the studied glasses are presented in Figure 3. The results showed an increase in the values of the elastic moduli with increase in the ZnO composition from 15 to 25%. Longitudinal, shear, Young's and bulk moduli increased from  $47.09$  to  $50.61$  GPa,  $15.05$  to  $16.94$  GPa,  $38.09$  to  $42.30$  GPa, and  $27.01$  to  $28.01$  GPa have been observed. Figure 3 illustrates the elastic moduli variation data for lead tellurite glasses with varying composition of ZnO; it is evident that the moduli of elasticity values increase were gradual with the different ZnO mol%. The addition of zinc oxide to the  $[ZnO]_x [(TeO_2)_{0.7}-PbO]_{0.3} ]_{1-x}$  network increases both shear and longitudinal ultrasonic velocities, which therefore led to increase in the glass rigidity, which in turn increased the elastic moduli.

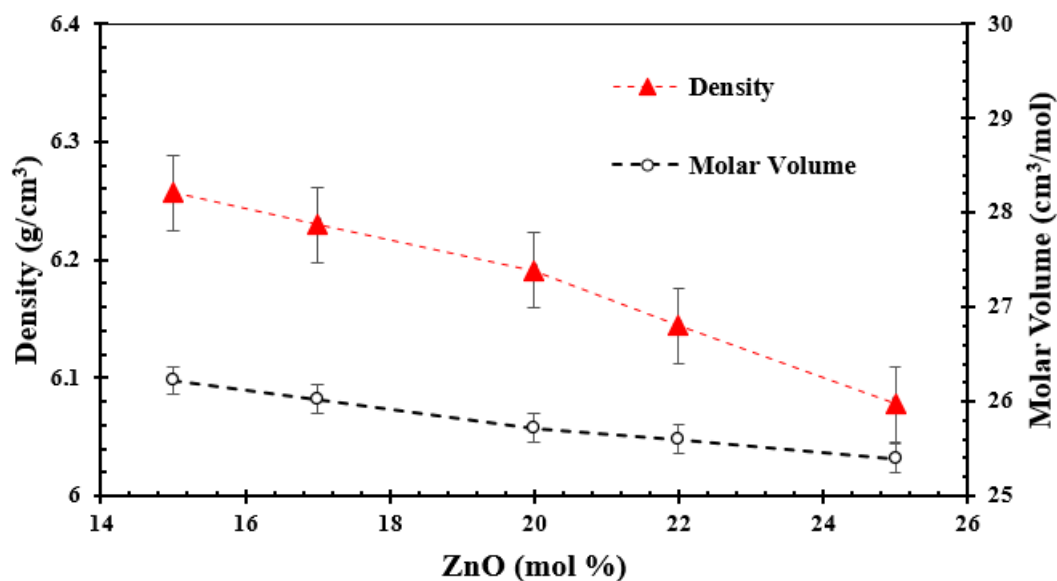


Figure 2. Density ( $\rho$ ) and molar volume ( $V_m$ ) of the  $[ZnO]_x [(TeO_2)_{0.7}PbO]_{0.3}]_{1-x}$  glasses (Elazoumi et al, 2017).

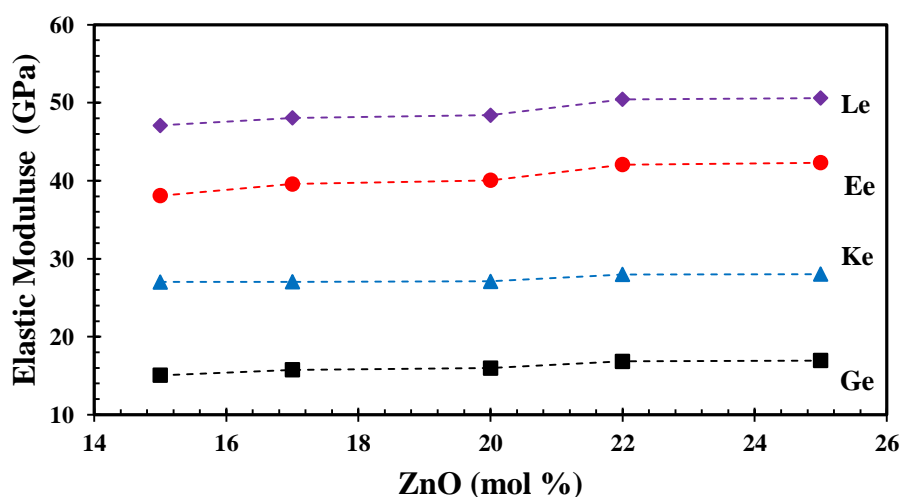


Figure 3: Elastic moduli longitudinal (L), shear (G), bulk modulus (K) and Young's (E) vs. the mole percentage of (ZnO) for  $[ZnO]_x [(TeO_2)_{0.7}PbO]_{0.3}]_{1-x}$  tellurite glasses [10].

Meanwhile the ultrasonic wave velocities measurements for  $50TeO_2-(50-x)V_2O_5-xAg_2O$  where  $x = 5, 10, 15, 20, 22.5, 25$  and  $27.5$  wt% ternary tellurite glasses were carried out at room temperature by El-Mallawany et al. [11]. The elastic moduli and Debye temperature were calculated from the ultrasonic velocities and density data. The physical parameters measured and calculated were found to be  $Ag_2O$  (wt%) percentage sensitive in the glass system.

Sidkey et al. studied the variation of ultrasonic velocity with the composition of  $TeO_2-V_2O_5-Sm_2O_3$  glasses at room temperature and 4MHz frequency using the pulse-echo technique [12]. It was found that the stability and material compactness of the studied glasses increased with increase in  $Sm_2O_3$  concentration. This might be due to increase in the network

bonds per unit volume number and average atomic ring size decrease in the network which consequently increased the elastic moduli the glasses.

Meanwhile Afifi and Marzouk measured the transverse and longitudinal ultrasonic waves velocities in  $20\text{WO}_3-(80-x)\text{TeO}_2-x\text{PbO}$  glass system through use of the pulse-echo technique at room temperature and 5MHz frequency [13]. Generally, this kinds of glasses possess high density and the density increase with PbO mole percentage increase was ascribed higher atomic size and weight of the constituting oxides. The ultrasonic velocity value increase in the glasses studied with the addition of PbO showed that inside the materials; the ultrasonic waves finds it more easier to travel. The results indicated that addition of more PbO in the glass system causes an increase in the material compactness and bond tightness which led to the increases in the elastic moduli, Poisson's ratio, Debye temperature, softening temperature and microhardness while 10 to 20 mol% increase was made of PbO content.

Measurements of the ultrasonic velocities of  $(\text{TeO}_2)_{80}-(\text{WO}_3)_{(20-x)}-(\text{K}_2\text{O})_x$  glass system at 6MHz frequency and room temperature were published elsewhere. The elastic moduli, fugacity, softening temperature and Debye temperature was also calculated from the ultrasonic data. The elastic constants values showed a decreasing pattern with  $\text{K}_2\text{O}$  molar concentration increase, which was ascribed to the increase in the cations number per unit glass formula and crosslink density decrease indicating network structural weakness and breakage in the network linkage [14]

The effect of concurrent removal of ZnO and addition  $\text{Fe}_2\text{O}_3$  on  $80\text{TeO}_2-(2-x)\text{ZnO}-x\text{Fe}_2\text{O}_3$  glass system with  $x=0$  to 0.15 molar fraction, prepared by the method of the melt-quenching method has been studied by Azianty et al. [15]. The FTIR spectroscopy and pulse-echo overlap technique at 5MHz were used in studying the structural and elastic properties respectively. Both transverse and the longitudinal velocities showed increase in large proportion from 3.40% to 4.68% and up to 5% after which the values decreased. Interestingly, the elastic moduli showed a similar pattern with  $\text{Fe}_2\text{O}_3$  concentration increase. The significant increases observed at the initial stage in longitudinal and shear velocities as well as the elastic moduli were suggested to be due to modifications in the glass structures which led to rigidity increase.

Azianty and Yahya also reported a study on the structural and elastic properties  $(80-x)\text{TeO}_2-20\text{PbO}-x\text{WO}_3$  glass system with  $x=0-20$  mol% fabricated using the melt-quenching technique [16]. FTIR spectroscopy and the pulse-echo method were used to study the structural changes and measure the ultrasonic velocities (at % 5 MHz ) respectively. At  $x=5$  mol%, the elastic moduli and microhardness increased slightly which followed by an almost linear and large increase beyond  $x=5$  mol%. The analysis of the FTIR spectra indicated  $\text{TeO}_4$  units increase and concurrent decrease in  $\text{TeO}_3$  structural units for  $x > 5$  mol%, indicating an increase bridging oxygen (BO) number and a decrease in the non-bridging oxygen (NBO) number respectively. The elastic moduli enhancement for  $x > 5$  mol% is ascribed to be due to the increase in BO which caused the increase in rigidity and stiffness of the glass network.

The ternary zinc oxyfluoride tellurite with the composition  $(\text{ZnO})_x-(\text{AlF}_3)_y-(\text{TeO}_2)_z$  with  $60 \leq z \leq 70$ ;  $5 \leq y < 25$ ;  $5 \leq x < 35$  considerations has been performed by Sidek et al. [17]. The microhardness (H), softening temperatures ( $T_s$ ), Acoustic Debye temperature ( $\theta_D$ ) of the studied glass system were calculated from the obtained data of the ultrasonic velocities. With increase in the ZnO concentration, both the microhardness, softening and Debye temperatures were decreased, which confirmed a reasonable change structure of the glasses.

The concurrent of Zn addition and  $\text{AlF}_3$  reduction in the tellurite glass matrix leads to more ions to be opened in the glass system's network. Therefore, the glass network structure weakens, and this led to the glass network rigidity reduction.

In conclusion, this short review paper presents the behavior of elastic (longitudinal, shear, bulk and Young's) moduli, Poisson's ratio, Debye temperature, and a softening temperature of selected ternary tellurite glass samples. As predicted in the hypothesis, the experimental data shows that most of the elastic moduli of the ternary glasses were found to be compositional dependent.

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