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# Effect of Zinc on the Physical Properties of Tellurite Glass

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**Abstract:** A series of binary tellurite glass samples containing zinc oxide, ranging from 10 to 40 mol%, were successfully prepared. Longitudinal and shear ultrasonic wave velocities were measured at room temperature and were taken at 5 MHz. Elastic properties, Poison's ratio, microhardness, Debye temperature and fractal bond connectivity have been calculated from the measured density and ultrasonic wave velocities. Results from the studies show that the densities of all glasses increase from 5098 to 5283 kg $m^{-3}$, molar volumes decrease from 29.773 to 24.287 $m^3$ $mol^{-1}$ and the transition temperature, $T_g$ decreases from 381.17 to 360.52°C with the substitution of $TeO_2$ by ZnO content. Both the longitudinal and shear wave velocities decrease with increase of ZnO composition. The experimental results indicated that the elastic constants depend upon the composition of the glasses. The role of the ZnO inside the glass network is also discussed. Quantitative analysis based on bond compression model has been carried out, in order to obtain more information about the structure of these glasses.

**Key words:** Tellurite glasses, ultrasonic velocity, elastic properties, cross-link density, bond compression model

## INTRODUCTION

Vitreous tellurite with tellurium dioxide as the main glass-former, which under normal condition, does not have the ability to transform into the glassy state without a modifier (Bahgat *et al*., 1987). It has superior physical properties such as low melting temperature, high refractive index, high dielectric constant and good infrared transmission (Rajendran, 2000). The distinguish factor about the matrix of this glass is that the tellurium atoms have unshared pairs of electrons which do not take part in bonding. Their relatively low temperature of crystallization and melting makes these types of glass an active candidate for CD memory devices (Sabry and El-Samanoudy, 1995).

Elastic properties are important parameters for understanding the structural characteristics of glass network. The measurement of ultrasonic parameters such as velocity and attenuation as a function of composition, temperature and frequency is of great interest in glass. The variation of ultrasonic parameters, besides density and molar volumes, are closely related to the changes occurred in the structure of glass network (Mallawany *et al*., 1994; Sidkey *et al*., 1997). Such properties are associated with the inter-atomic forces and potentials in the lattice structure (Saddeek, 2004). In general, strength of amorphous materials increases with their elastic moduli; therefore, it is possible to asses the strength indirectly from their elastic properties (Saddeek, 2005).

The objectives of this research are to study the elastic moduli of zinc tellurite glasses and to investigate the structural changes and their physical properties of the glasses.

## MATERIALS AND METHODS

This study was started at 2004 until now. It was conducted in Ultrasonic Research Laboratory at Universiti Putra Malaysia, Malaysia. The binary tellurite glasses, $(TeO_2)_{1-x}(ZnO)_x$ were prepared by mixing together specific weights of tellurium dioxide (technical grade) and zinc oxide (purity 99%) in alumina crucible. The x percentage was ranging from 10 to 40 mol %. The samples were prepared by melt quenching method. The 20 g batches were then preheated in a first furnace to 400°C for 30 min to reduce tendency of volatilization. The crucible was then transferred to a second furnace at 750-800°C for 1 h, depending on the percentage of ZnO.

The melt was then removed from the second furnace before it was poured in a stainless steel cylindrical shaped split mould which had been preheated to 400°C followed by annealing in the first furnace at 350°C for 1 h, after which the furnace was switched off and the glass was

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allowed to cool in situ for 24 h. This was done to remove residual stresses before cutting procedures. The prepared samples were then cut into required dimension for density and ultrasonic measurement and a fine glass powder was used for transition temperature measurement. Detailed information on glass preparation can be found elsewhere (Sidek *et al*., 2004).

The glass transition temperatures (Tg) were determined by differential thermal analyzer SETARAM (DTA). The densities of the glasses were determined by the Archimedes method using acetone as immersion liquid. The X-ray diffraction pattern of the glass samples were examined using diffractometer (model X'Pert Pro Panalytical) by employing Cr-Kα radiation. For measurement of ultrasonic velocity in the glass samples, MATEC MBS 8000 was used. All measurements were taken at 5 MHz frequency and at room temperature.

## RESULTS AND DISCUSSION

**Density and molar volume:** The X-ray diffraction patterns of the studied glass system reveal the absence of any discrete or continuous sharp crystalline peaks but show homogeneous glassy characters. The experimental values of the density and the molar volume of the studied glasses are shown in Table 1. The density of all glasses increase from 5098 to 5283 kg $m^{-3}$ with the substitution of $TeO_2$ by ZnO, while molar volume decrease from 29.773 to 24.287 $m^3$ $mol^{-1}$. The similar behavior has been found by Mallawany (1993). However, the present values are somewhat lower than those found by Mallawany, perhaps due to different preparation technique.

Addition of ZnO to the tellurite network causes some type of structural rearrangement of the atoms as found by Hoppe *et al*. (2004). Therefore, there is a possibility for the modification of the glass microstructure upon substitution of $TeO_2$ by ZnO.

Apparently, while substituting $TeO_2$ by ZnO, the density should decrease because of the lower molecular weight of ZnO ($M_{Zn}$ = 65.37 g $mol^{-1}$) than that of $TeO_2$ ($M_{Tc}$ = 127.6 g $mol^{-1}$). The increase of density indicates that the zinc ions reticule the network of the glass. This was confirmed by the results of the molar volume where it shows a decrease values.

Table 1: Glass compositions, density and molar volume

| | Composition (mol %) | | | |
|---|---|---|---|---|
| Sample | $TeO_2$ | ZnO | Density (± 1 kg $m^{-3}$) | Molar volume (±0.01 $cm^3$ $mol^{-1}$) |
| $T_1$ | 90 | 10 | 5098 | 29.77 |
| $T_2$ | 85 | 15 | 5102 | 28.98 |
| $T_3$ | 80 | 20 | 5136 | 28.03 |
| $T_4$ | 75 | 25 | 5194 | 26.96 |
| $T_5$ | 70 | 30 | 5211 | 26.13 |
| $T_6$ | 65 | 35 | 5280 | 25.05 |
| $T_7$ | 60 | 40 | 5283 | 24.29 |

As reported earlier by Hoppe *et al*. (2004), Lambson *et al*. (1984), Neov *et al*. (1979) and Khozukharov *et al*. (1986), the structure of the tellurite-rich glasses is a laminar network consisting of the structural units $TeO_4$ trigonal bipyramids (tbp) and $TeO_3$ trigonal pyramids (tp). There is 4-coordination of Te in the tetragonal form, the nearest-neighbours being arranged at four of the vertices of a trigonal bipyramid, suggesting considerable covalent character of the Te-O bonds.

An oxygen atom introduced into glasses with high $TeO_2$ contents opens a Te-O-Te bridge and changes two $TeO_4$ into $TeO_3$ units. Each of these $TeO_3$ units has two terminal Te-O bonds with partial double bond character (Hoppe *et al*., 2004). The increase in density is due to glass structural change of which influence of $Zn^{2+}$ on breaking tellurium-oxygen networks. In this case, all oxygen atoms from ZnO are used for rupture of Te-O-Te bridges which accompanied with a change of nearly all participating $TeO_4$ to $TeO_3$ groups (Kozhukharov *et al*., 1986).

In general, the decrease in the molar volume indicates a decrease in the interatomic distances. Therefore, the compactness of the glass will increase and more bridging oxygen's (BOs) will be created which increase the rigidity of the glass (Saddeek, 2005).

**Ultrasonic velocity and elastic moduli**

**The determination of the experimental and theoretical derived parameters:** The ultrasonic wave velocities measured in this work are found to be sensitive with the glass composition. The elastic properties of the binary zinc-tellurite glasses with different percentage of x, such as, the longitudinal modulus L, the shear modulus G, the bulk modulus K, the Young's modulus E and the Poisson's ratio σ will be calculated using the following equations:

$$L = v_l^2 \rho (GPa) \quad (1)$$

$$G = v_s^2 \rho (GPa) \quad (2)$$

$$K = L - \left(\frac{3}{4}\right) G (GPa) \quad (3)$$

$$E = 2(1+\sigma) G \ (GPa) \quad (4)$$

$$\sigma = \frac{L - 2G}{2(L - G)} \quad (5)$$

Where:

ρ = The density

$v_l$ and $v_s$ = The measured longitudinal and shear ultrasonic velocities, respectively

The micro-hardness H and the Debye temperature $\theta_D$ were calculated using the following equations:

$$H = \frac{(1-2\sigma)E}{6(1+\sigma)}(GPa) \quad (6)$$

$$\theta_D = \frac{h}{k}\left(\frac{9N}{4\pi V}\right)^{1/3} vm(K) \quad (7)$$

Where:

h = The Planck's constant
k = The Boltzman constant
$v_m$ = The mean ultrasonic velocity
(N/V) = The No. of vibrating atoms per unit volume

$$(N/V) = PN_A$$

Where:

P = The No. of atoms in the chemical formula
$N_A$ = The Avogadro number

The composition dependence of longitudinal and shear wave velocities is shown in Fig. 1. Both the longitudinal and shear wave velocities ($v_l$ and $v_s$, respectively) decrease gradually with the concentration of ZnO. The decrease in ultrasonic velocity of the studied glass reveal the fact that, adding ZnO content to the tellurite network causes a difficult movement from the ultrasonic wave inside the network of the glass structure and hence the tendency of the ultrasonic velocity to decrease as the mole percentage of ZnO increases.

It can be seen that the values of $v_l$ are higher than $v_s$. For most materials, ultrasonic wave velocities will increase as the density increases. The similar behavior has been found by Mallawany (1993) for the same glass systems. But in this study, the unique is that it shows the opposite phenomena, where the ultrasonic velocities decrease as the density increases. The difference of this behavior may be due to the difference of preparation technique as mentioned earlier. The $Zn^{2+}$ ions in ZnO give rise to the non-bridging oxygen bond (single bonded oxygen ion).

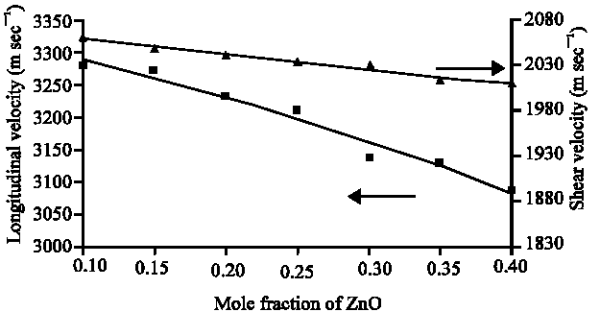


Fig. 1: Ultrasonic velocities of $(TeO_2)_{1-x}(ZnO)_x$ glasses

It was inferred from a study on binary zinc tellurite glasses performed by Hoppe *et al.* (2004) that the average coordination number of $TeO_2$ decreases from 4 to 3 as the ZnO content increases. Therefore, the structural unit $TeO_4$ tbps, will be converted into the structural unit $TeO_3$ tp, which in its turn is accompanied by the creation of non-bridging oxygens (NBOs).

Table 2 shows the variation of elastic moduli of different values of ZnO mol percentage content. The results indicate that the elastic moduli decrease with increasing of ZnO mole percentage content, while the values for bulk modulus, K increase. In comparison, the longitudinal modulus, L, is always higher than the shear modulus, G. This means that the glasses can withstand longitudinal stress better than shear stress. Young's modulus is the ratio of the linear stress to the linear strain. The decreasing Young's modulus implies the weakening of the overall bonding strength.

Comparing the bulk modulus to the Young's modulus, the values of bulk modulus are relatively lower. This concludes that the glass samples can tolerate stress in one direction better than stress acting in all directions; they are tougher at certain axes (Sidek *et al.*, 2004).

Rajendran *et al.* (2003) reported that Poisson's ratio is affected by the changes in the cross-link density of the glass network. In the studied glass system, the value of Poison's ratio is varied from 0.20 to 0.24 (Table 2). The increasing trend of Poisson's ratio suggests that if the same amount of stress is applied over the whole range of the glass composition, the lateral strain will gradually level out (Sidek *et al.*, 2004).

To predict the compositional dependence of elastic moduli of polycomponent glasses, the interpretation of the variation in the experimental elastic behavior observed in this study will be based on the bulk compression model proposed by Bridge and Higazy (1986). Formulae

Table 2: Experimental values of glass composition, elastic modulus (L) and (G), bulk modulus (K), fractional bond connectivity (d), Young's modulus (E), Poisson Ratio (σ) and microhardness (H) for $(ZnO)_x(TeO_2)_{1-x}$ glass system

| Sample | L (GPa) | G (GPa) | K (GPa) | d = 4G/K | E (GPa) | σ | H (GPa) |
|---|---|---|---|---|---|---|---|
| $T_1$ | 56.33 | 21.01 | 28.317 | 2.97 | 50.529 | 0.2026 | 4.165 |
| $T_2$ | 55.80 | 20.90 | 27.929 | 2.99 | 50.184 | 0.2005 | 4.173 |
| $T_3$ | 55.79 | 20.44 | 28.540 | 2.86 | 49.505 | 0.2109 | 3.940 |
| $T_4$ | 56.05 | 20.36 | 28.899 | 2.82 | 49.469 | 0.2147 | 3.873 |
| $T_5$ | 56.06 | 19.39 | 30.208 | 2.57 | 47.918 | 0.2356 | 3.418 |
| $T_6$ | 55.94 | 19.46 | 29.989 | 2.60 | 48.007 | 0.2332 | 3.462 |
| $T_7$ | 55.90 | 18.93 | 30.663 | 2.47 | 47.101 | 0.2440 | 3.231 |

Table 3: Bond length (r), first order stretching force constant (F), coordination number ($n_f$) of the oxides $TeO_2$ and ZnO and $n_c$ (Wells, 1975)

| Oxide | r (nm) | F (N m$^{-1}$) | $n_f$ | $n_c$ |
|---|---|---|---|---|
| $TeO_2$ | 0.1990 | 216 | 4 | 2 |
| ZnO | 0.1988 | 219 | 6 | 4 |

Table 4: Average crosslink density ($n_c^-$), Young's modulus (E), shear modulus (G), (E/G) ratio

| Sample | $n_c^-$ | E (GPa) | G (GPa) | E/G |
|---|---|---|---|---|
| $T_1$ | 2.2 | 50.529 | 21.01 | 2.405 |
| $T_2$ | 2.3 | 50.184 | 20.90 | 2.401 |
| $T_3$ | 2.4 | 49.505 | 20.44 | 2.422 |
| $T_4$ | 2.5 | 49.469 | 20.36 | 2.430 |
| $T_5$ | 2.6 | 47.918 | 19.39 | 2.471 |
| $T_6$ | 2.7 | 48.007 | 19.46 | 2.467 |
| $T_7$ | 2.8 | 47.101 | 18.93 | 2.488 |

were developed to express the elastic constants as functions of number of network bonds per unit volume, bond length, first order stretching force constant and crosslink density per cation.

According to bond compression model, quantitatively, Poisson's ratio is inversely proportional to the quantity named the average cross-link density, $n_c^-$. The average number of cross-links per glass formula ($n_c^-$) is calculated according to Bridge and Higazy (1986),

$$n_c^- = \frac{1}{\eta}\sum n_c N_c \quad (8)$$

Where:

$n_c$ = The No. of cross-links per cation (defined as the No. of bridging bonds per cation, $n_f$, minus two)
$N_c$ = The No. of cations per glass formula unit
$\eta$ = The total No. of cations per glass formula unit

Table 3 gives the complete set of parameters adopted from the crystal structure (Wells, 1975) used in this calculation.

The average cross-link density increases from 2.2 to 2.8 when ZnO content increases (Table 4). The increase in cross-link density is due to the increase in dimensionality, since tellurium atom (with coordination number 4) is replaced by zinc atom (with coordination number 6). To isolate another possible variable affecting the Poisson's ratio of the glass, which is the relation of Poisson's ratio with (E/G), applied to the three-chain network (Mallawany *et al.*, 2006). From Fig. 2 and 3, it is clear that the behavior of the Poisson's ratio is like the behavior of E/G ratio with increase of ZnO mol%.

For a structure consisting of one type of network bond, the bulk modulus calculated according to bond compression model can be expressed in the form:

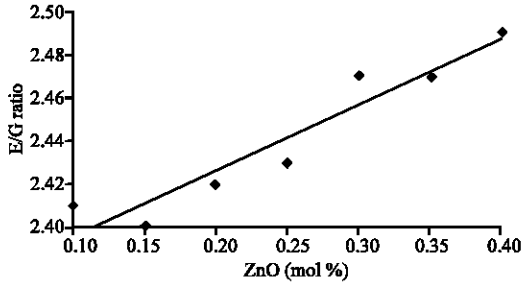


Fig. 2: (E/G) ratio versus ZnO mol% for $TeO_2$-ZnO glasses

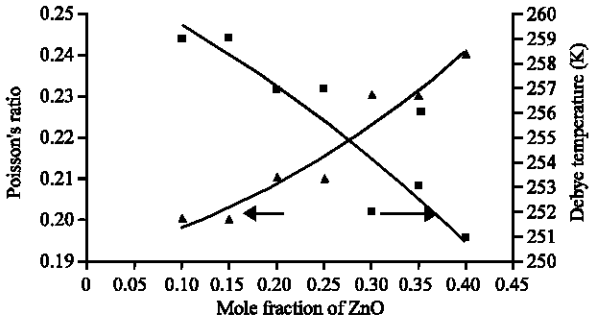


Fig. 3: Poisson's ratio and Debye temperature of $(TeO_2)_{1-x}$ $(ZnO)_x$

$$K_{bc} = N_b r^2 F/9 \quad (9)$$

Where:

r = The bond length
F = The first order stretching force constant
$N_b$ = The No. of network bonds per unit volume and is given:

$$N_b = n_f N_f \quad (10)$$

Where:

$n_f$ = The No. of network bonds per glass formula unit
$N_f$ = The No. of formula units per unit volume

For a polycomponent glass with n different types of network bonds, Eq. 10 can be rewritten as:

$$K_{bc} = \frac{\rho N_A}{9M}\sum_i x_i (n_f) r_i^2 F_i \quad (11)$$

Where:

$x_i$ = The mole fraction of the ith oxide
$N_A$ $\rho$ and M = Avogadro's number, density and molecular weight of the glass, respectively

First stretching force constant is given by the equation:

Table 5: Glass composition, calculated values for bulk modulus ($K_{bc}$), ratio of ($K_{bc}/K_{exp}$), number of bonds per unit volume ($N_b$) and average stretching force constant (F)

| Sample | $K_{bc}$ (GPa) | $K_{bc}/K_{exp}$ | $N_b$ (x$10^{28}$ $m^{-3}$) | F (N $m^{-1}$) |
|---|---|---|---|---|
| $T_1$ | 87.58 | 3.09 | 9.19 | 216.43 |
| $T_2$ | 94.88 | 3.40 | 9.94 | 216.63 |
| $T_3$ | 102.79 | 3.60 | 10.76 | 216.82 |
| $T_4$ | 111.31 | 3.85 | 11.64 | 217.00 |
| $T_5$ | 119.07 | 3.94 | 12.45 | 217.17 |
| $T_6$ | 128.13 | 4.27 | 13.38 | 217.34 |
| $T_7$ | 135.69 | 4.43 | 14.17 | 217.50 |

$$F = 17/r^3 \quad (12)$$

Bridge *et al.* (1983) found that $K_{bc}$ value is usually greater than the experimental value $K_{exp}$ by a factor of 3-10. They concluded that the ratio $K_{bc}/K_{exp}$ is a rough measure of the degree to which bonding processes are involved in isotropic elastic deformation.

It is clear from Table 5 that the bulk modulus $K_{bc}$, increases from 87.58 to 135.69 GPa with the increase of ZnO content from 0.10 to 0.40 mol%. This increase in $K_{bc}$ is expected since $K_{bc}$ depends on the number of network bonds per unit volume ($N_b$) and the first order stretching force constant F (Eq. 9).

Moreover, the number of network bonds per unit volume increases from $9.19\times10^{28}$ to $14.17\times10^{28}$ $m^{-3}$, the average stretching force constant increases from 216.43 to 217.50 N $m^{-1}$ with the increase of ZnO content. Furthermore, values of the ratio ($K_{bc}/K_{exp}$) range between 3.09 and 4.43 which indicate a relatively open three dimensional network.

Debye temperature ($\theta_D$) is a characteristic property of a solid lattice related to its acoustic phonon spectrum (Varshneya, 1994). It represents the temperature at which nearly all modes of vibration in a solid are excited (Sidkey *et al.*, 1999). Also increase in the rigidity of the glass is associated with an increase in the lattice vibrations.

The Debye temperature (Fig. 3) decreases from 259 to 251 K as ZnO content increases. It decreases when the ultrasonic velocity decreases. The observed decrease in $\theta_D$ indicates a monotonic decrease in the total vibrational energy of the system. This is because any of the conceivable vibrational units resulting from the substitution will be of lower energy.

Micro-hardness expresses the stress required to eliminate the free volume (deformation of the network) of the glass. The free volume in the glass is the openness of the glasses over that of the corresponding crystal (Saddeek, 2004). Therefore, application of high hydrostatic pressure will reduce this free volume, i.e., the glasses will be compacted. Figure 4 shows that micro-hardness of the glass system decrease with the addition

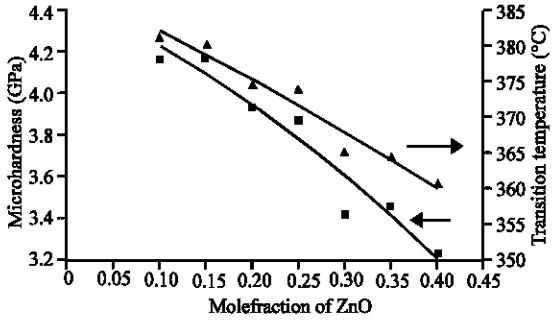


Fig. 4: Micro-hardness and glass transition temperature of $(TeO_2)_{1-x}(ZnO)_x$

of ZnO. It is presumably because of the connectivity of the glass structure decreases (Varshneya, 1994).

DTA curves for the studied glass samples with different ZnO contents have been obtained to determine the glass transition temperature ($T_g$) values. The composition dependence of $T_g$ in the present glass system reveals that $T_g$ (Fig. 4) decreases from 381.17 to 360.52°C with an increase in ZnO content. The decrease in the glass transition temperature values implies that number of bridging oxygen group decreases.

The observed decrease in micro-hardness and $T_g$ with the increase of ZnO content indicate a weakening in the glass structure which may be due to the introduction of weaker ionic bonds in the glass network. Tellurite glass with higher percentage of ZnO has a lower value of $T_g$ that is ZnO oxides creates a weaker tellurite glass (Mallawany, 1999).

The fractal bond connectivity, (d) of these glasses ranges between 2.47 and 2.97 (Table 2). These suggest an intermediate connectivity between a two and a three-dimensional disordered network, which implies a marked degree of cross-links between $TeO_2$ chains. As the ZnO content increases, the value of fractal bond connectivity was found to decrease, indicating the weakening of structure in the glass.

## CONCLUSIONS

The present study on the binary $TeO_2$-ZnO glass system revealed the following conclusions:

- The density increases as the ZnO content increases while the molar volume decreases.
- There is an observed decrease in the ultrasonic velocity, the elastic moduli, the Debye temperature and the glass transition temperature with an increase in ZnO content which was attributed to the decrease in the network of the glass structure and the creation of non-bridging oxygens in a direct result of the transition of $TeO_4$ to $TeO_3$.

- The observed decrease in elastic modulli is also mainly due to the constant number of cations per glass formula unit and the increase in the average crosslink density and this indicates the breaking of network linkage and the weakness of the network structure.
- The decrease in the glass transition temperature values implies that number of bridging oxygen group decreases. It is also due to the increase of both the average crosslink density and the stretching force constant.
- The fractal bond connectivity suggests an intermediate structure between a two and a three-dimensional disordered network, indicating cross-links between $TeO_2$ chains.

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