



Evaluation of the elastic properties of monovalent oxides using TeO₂-based glasses

A M EMARA^{1,2}, S F MANSOUR¹, M Y HASSAAN³ and EI SAYED YOUSEF^{2,4,*}

¹Faculty of Science, Department of Physics, Zagazig University, Zagazig, Egypt

²Faculty of Science, Department of Physics, King Khalid University, Abha 9004, Saudi Arabia

³Faculty of Science, Department of Physics, Al-Azhar University, Nasr City 11884, Egypt

⁴Faculty of Science, Department of Physics, Al-Azhar University, Assuit Branch, Assuit, Egypt

*Author for correspondence (omn_yousef2000@yahoo.com)

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Abstract. Quaternary tellurite glasses with composition 75TeO₂–5WO₃–15Nb₂O₅–5M_xO_y in mol%, where M_xO_y = (Na₂O, Ag₂O, ZnO, MgO, CuO, NiO, TiO₂, MnO₂), were prepared by the normal melt-quenching method. The ultrasonic velocities (longitudinal and shear) were measured in these glasses using the pulse-echo technique at room temperature. Their elastic moduli, microhardness and Debye temperature were calculated and discussed in terms of the modifier's ionicity and quantitatively in terms of number of bonds per unit volume and the cross-link density. In this study, the values of ultrasonic velocities, elastic moduli, Debye temperature and microhardness were found to be strongly dependent on three factors, namely: (i) modifier's ionicity; (ii) trigonal pyramid (TeO₃)/trigonal bipyramid (TeO₄) ratio; and (iii) glass transition temperature T_g . We used the Makishima and Mackenzie's model to calculate the theoretical elastic moduli and to indicate that the experimental values were in good agreement with the theoretical values.

Keywords. Oxides; tellurite glass; elastic properties.

1. Introduction

Tellurite-based glasses possess very interesting physical properties such as low melting temperature [1], good mechanical strength and chemical resistance [1,2], high values of linear and non-linear refractive indices [3–7] and good optical transmission in a wide range of infrared (IR) [8,9] and visible wavelengths [3,5,10–12]. Studying the elastic properties is very important for selection of glasses for a particular application, such as optical fibres, because it gives useful information about the structural stability [8,13,14], strength and rigidity of the glass samples [15,16]. Several elastic studies have been reported for the binary and ternary tellurite glasses, such as Nb₂O₅–TeO₂ [17], TeO₂–La₂O₃ [18,19], TeO₂–ZnO [12,20], TeO₂–V₂O₅ [8], TeO₂–WO₃–PbO [21,22], TeO₂–Nb₂O₅–Li₂O [23], TeO₂–V₂O₅–TiO₂ [24] and TeO₂–WO₃–K₂O [25]. From the available literature, it can be understood that ternary tellurite glass systems show excellent thermal resistance against crystallization compared to the binary glasses [26,27]. Addition of transition metal oxides or rare-earth oxides has been suggested to increase the rigidity of the glasses through the formation of bridging oxygen (BO), as reported in many ternary tellurite glass systems [24,28,29]. Similarly, addition of monovalent oxides, such as Li₂O, Na₂O and Ag₂O, has been reported to increase the number of non-bridging oxygen atoms (NBOs) in the ternary tellurite glass systems [30–33].

In this study, we have studied the elastic properties of quaternary tellurite glasses using a variety of different modifiers such as Na₂O, Ag₂O, ZnO, MgO, CuO, NiO, TiO₂ and MnO₂, which can be later on used in various optical applications.

2. Experimental

The glass samples were prepared using 75TeO₂–5WO₃–15Nb₂O₅–5M_xO_y in mol%, where M_xO_y = (Na₂O, Ag₂O, ZnO, MgO, CuO, NiO, TiO₂, MnO₂). The powder mixture of the oxides was taken in a platinum crucible and heated in a melting furnace at 900°C for 30 min. The highly viscous melt was stirred and cast in a cylindrical graphite mould at 750°C. It was then transferred to a furnace maintained at 350°C for 2 h for annealing. The furnace was switched off and the glass samples were allowed to cool. After cooling, the samples were optical polished to produce parallel opposite surfaces for ultrasonic velocity measurements.

The densities (ρ) of the glass samples were measured using a helium pycnometer (AccuPyc 1330 Pycnometer) with an accuracy of $\pm 0.03\%$.

Ultrasonic measurements were carried out using the pulse-echo method in a flaw detector (USM3-Krautkramer) at room temperature. The X-cut and Y-cut transducers were used for longitudinal and transverse modes, respectively. An

Table 1. Sample code, sample composition, density, modifier's ionicity, $\text{TeO}_3/\text{TeO}_4$ ratio.

Sample code	Sample composition in mol%	ρ (g cm ⁻³)	Modifier's ionicity	$\text{TeO}_3/\text{TeO}_4$
Sample 1	75TeO ₂ -5WO ₃ -15Nb ₂ O ₅ -5CuO	5.3107	0.8	0.34
Sample 2	75TeO ₂ -5WO ₃ -15Nb ₂ O ₅ -5MnO ₂	5.2937	0.78	0.22
Sample 3	75TeO ₂ -5WO ₃ -15Nb ₂ O ₅ -5NiO	5.3388	0.83	0.35
Sample 4	75TeO ₂ -5WO ₃ -15Nb ₂ O ₅ -5Ag ₂ O	5.444	0.92	0.63
Sample 5	75TeO ₂ -5WO ₃ -15Nb ₂ O ₅ -5ZnO	5.3235	0.85	0.43
Sample 6	75TeO ₂ -5WO ₃ -15Nb ₂ O ₅ -5MgO	5.285	0.8	0.33
Sample 7	75TeO ₂ -5WO ₃ -15Nb ₂ O ₅ -5TiO ₂	5.244	0.76	0.18
Sample 8	75TeO ₂ -5WO ₃ -15Nb ₂ O ₅ -5Na ₂ O	5.2178	0.89	0.45

Values of $\text{TeO}_3/\text{TeO}_4$ are deduced from Raman spectra as stated in part 1 of Mansour *et al* [35].

oscilloscope (Hewlett-Packard model 54502A oscilloscope) was used to measure the time between the initiation and the receipt of the pulse. The velocity was obtained by dividing the round trip distance by the elapsed time. The accuracy of the method was about $\pm 0.04\%$. All velocity measurements in this study were carried out at a nominal frequency of 5 MHz at room temperature (300 K).

3. Results and discussion

The chemical composition, density and modifier's ionicity results of the glass samples are reported in table 1. From these results, we derived that the ultrasonic velocities and the elastic moduli values of the prepared glasses are dependent on the ionicity values of the network modifiers. Ahrens [34] derived the relation between ionicity and cation radius, r , of the binary oxides as $\text{ionicity} = \exp(-0.1131/r_{\text{cation}}^2)$. The ionicity of the oxide decreases with increasing polarizing power (field strength) of the cation. The values of ionicity calculated for the modifiers in our samples are shown in table 1. The values of $\text{TeO}_3/\text{TeO}_4$ ratio were deduced from the Raman spectra of the samples, where the intensities of the IR major band at 650 cm^{-1} and of the band at 720 cm^{-1} were assigned to TeO_4 and TeO_3 units, respectively [35], and these values are listed in table 1. It can be noticed that the modifier TiO_2 has the lowest value of ionicity ($I = 0.76$), which corresponds to the lowest value of the $\text{TeO}_3/\text{TeO}_4$ ratio (0.18) in sample 7, while the modifier Ag_2O has the highest value of ionicity ($I = 0.92$), which corresponds to the highest value of the $\text{TeO}_3/\text{TeO}_4$ ratio (0.63) in sample 4. This means that the modifier Ag_2O is more ionic than the modifier TiO_2 , making the bonds between the cation Ag^+ and the anion O^{2-} weaker than the bond between the cation Ti^{2+} and the anion O^{2-} and creating more NBOs [5,34]; thus, the value of $\text{TeO}_3/\text{TeO}_4$ ratio increases with increasing ionicity (figure 1a and b). Figure 2a and b shows the variation of longitudinal and shear velocities with different network modifiers. The change in ultrasonic velocities (shear and longitudinal) is strongly dependent on the $\text{TeO}_3/\text{TeO}_4$ ratio. In addition, the change in the glass structure (like packing density, cross-link density, compactness

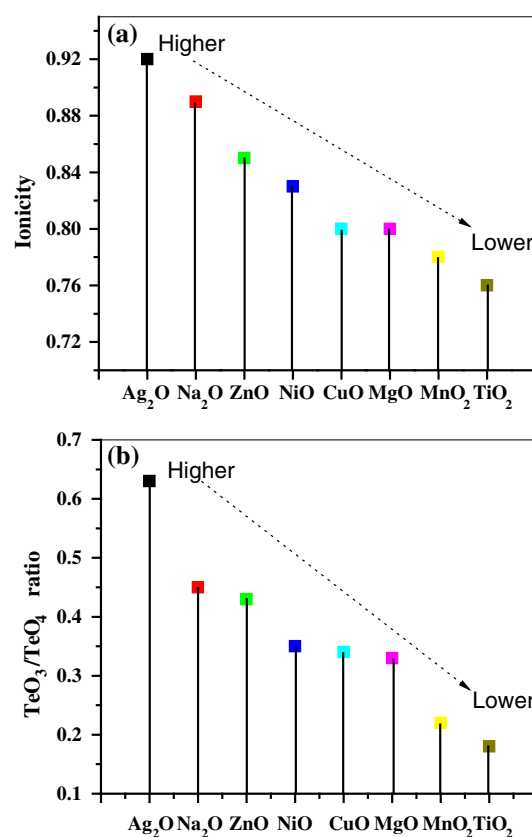


Figure 1. (a) The variation of ionicity value with different network modifiers. (b) $\text{TeO}_3/\text{TeO}_4$ ratio with different network modifiers.

and dimension of interatomic spacing) also reflected on the change in ultrasonic velocities, as reported in pervious literature [36–38]. Thus, sample 4 having the modifier Ag_2O has the lowest value of ultrasonic velocity due to its highest ratio of NBO ($\text{TeO}_3/\text{TeO}_4 = 0.63$) in the glassy network, where these NBOs can absorb the ultrasonic waves more than the BO atoms [39]. In contrast, sample 7 with the modifier TiO_2 has the highest value of ultrasonic velocity. The data of longitudinal (L), shear (S), bulk (Ke) and Young's (E) elastic moduli for quaternary glasses $75\text{TeO}_2-5\text{WO}_3-15\text{Nb}_2\text{O}_5-5\text{M}_x\text{O}_y$ in mol%, where $\text{M}_x\text{O}_y = (\text{Na}_2\text{O}, \text{Ag}_2\text{O}, \text{ZnO}, \text{MgO}, \text{CuO}, \text{NiO},$

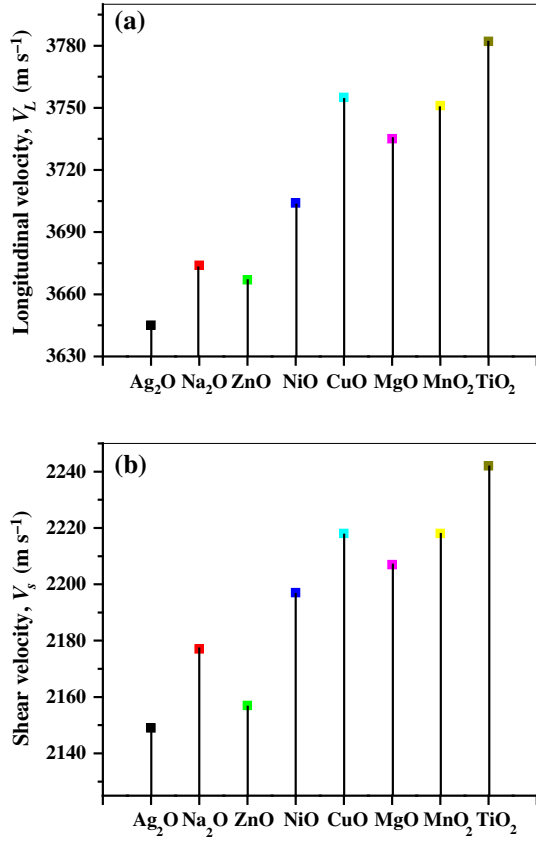


Figure 2. (a) Variation of longitudinal velocity value with different network modifiers. (b) Variation of shear velocity value with different network modifiers.

TiO₂, MnO₂), are summarized in table 2. These values were calculated using the formulae given below [21].

$$L = \rho V_L^2, \quad (1)$$

$$S = \rho V_S^2, \quad (2)$$

$$K = L - \frac{4}{3}S, \quad (3)$$

$$E = 2(1 + \sigma) \cdot S, \quad (4)$$

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

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

where ρ is the density, V_L and V_S are the measured longitudinal and shear ultrasonic velocities, respectively, and σ and H are Poisson's ratio and microhardness, respectively. Longitudinal modulus, L , ranged from 70.43 to 75 GPa; shear modulus, S , from 24.7 to 26.35 GPa; Young's modulus, E , from 60.78 to 64.79 GPa and bulk modulus, K , from 37.45 to 40 GPa (table 2). The large difference in the values between L and S was due to the volume effect, where the change in

volume due to compressions and expansions involved in longitudinal strains was pronounced while there was no change in the volume due to shear strains.

The values of the elastic moduli for the quaternary tellurite glasses used in this study are higher than that of pure TeO₂ glass, binary TeO₂-WO₃ [12], and are in good agreement with the values calculated for the ternary system TeO₂-Nb₂O₅-ZnO [15].

The quantitative interpretation of the elastic moduli may be explained according to the number of bonds per unit volume (n_b) of the glass, as given in the equation [12] $n_b = \frac{N_A}{V_a} \sum_i (n_f x)_i$, where x is the mole fraction of component oxide, N_A is the Avogadro's number, V_a is the molar volume of the glass, n_f is the coordination number of the cation and i denotes the component oxide. It is observed that sample 7 has the highest values of elastic moduli $L = 75$, $S = 26.35$, $E = 64.79$ and $K = 40$ GPa, which corresponds to the highest value of number of bonds per unit volume, i.e., $n_b = 8.12 \times 10^{28}$ (m⁻³), as these network bonds generate strong covalent forces resisting deformation [5].

Microhardness (H), in GPa, is the quantity that determines the stress required to eliminate the free volume. It is calculated using equation (6). The value of microhardness increases from 4.368 to 4.761 GPa. The increase of microhardness is expected from the increase in elastic moduli. The highest value of microhardness is 4.761 GPa, for sample 7, with the modifier TiO₂ having the lowest value of ionicity (0.76), while the lowest two values of microhardness are 4.368 and 4.465 GPa, for the samples 5 and 4, with the modifiers ZnO and Ag₂O having the highest values of ionicity (0.85 and 0.92), respectively.

Our results on the effect of reducing TeO₂ can be discussed in the light of results from previous works on ternary tellurite glasses, where several elastic studies on ternary tellurite glasses with unaltered TeO₂ content have been reported.

Elastic properties of the system of glass (90- x)TeO₂-10Nb₂O₅-(x)ZnO ($x = 0-15$ mol%) have been studied by Mohamed *et al* [15]. The initial drop in ultrasonic velocity and related elastic moduli observed at $x = 5$ mol% indicates weakening of network rigidity of the glass system due to structural modification as a direct effect of TeO₂ reduction and existence of NBO. However, further replacement of TeO₂ by ZnO at $x > 5$ mol% contributed to the increase in BO, causing the rigidity of the glass network to improve.

The ultrasonic velocity measurements of the tricomponent tellurite glasses TeO₂-V₂O₅-TiO₂ showed a linear increase in the density, ultrasonic velocities, elastic moduli, Debye temperature and microhardness with the addition of TiO₂ mol%, while Poisson's ratio and softening temperature decreased linearly with the addition of TiO₂. This decrease in Poisson's ratio was due to the increase in the average cross-link density as the TiO₂ content increased [24].

The ternary glass system TeO₂-V₂O₅-Ag₂O studied by El-Mallawany *et al* [39] showed that ultrasonic velocity decreased rapidly when Ag₂O content was increased from 5 to 25 wt% because of an increase in NBO which caused

splitting of the glassy network. The NBO atoms can absorb ultrasonic waves more than the BO atoms. $\text{TeO}_2\text{-WO}_3\text{-K}_2\text{O}$ [25] and $\text{TeO}_2\text{-B}_2\text{O}_3\text{-Ag}_2\text{O}$ glasses showed a monotonous decrease in the ultrasonic velocity and elastic moduli.

In the glass system, $\text{Na}_2\text{O-V}_2\text{O}_5\text{-TeO}_2$ [38], both shear and longitudinal velocities showed a small steady decrease with the addition of Na_2O from $x = 5$ mol% to $x = 15$ mol%, followed by a large decrease at $x > 15$ mol%. Longitudinal modulus (L), shear modulus (S), Young's modulus (E), hardness (H) and Debye temperature (θ_D) also showed similar behaviour to the ultrasonic velocities. The decrease in elastic moduli was suggested to be due to the weakening of the network rigidity of the glass system with the increase in the formation of NBO, as revealed by the Raman spectroscopy.

Poisson's ratio σ is defined as the ratio between lateral and longitudinal strains produced when the tensile force is applied. The value of Poisson's ratio ranged from 0.2286 to 0.2355. The behaviour of the variation of Poisson's ratio was nearly opposite to that observed for the variation in elastic moduli. It was noticed that the highest value of Poisson's ratio was 0.2355 for sample 5, which corresponded to the lowest value of microhardness (4.368 GPa).

We know that two factors affect the change in Poisson's ratio value, namely (i) the average cross-link density, n_c , where Poisson's ratio decreases with increasing cross-link density (for constant ratio of bond bending to stretching force constant) and (ii) the ratio of bond bending to stretching force constant, where Poisson's ratio decreases with increasing ratio of bond bending to stretching force constant (at constant cross-link density). The average cross-link density, n_c [5], was calculated using the relation

$$\bar{n}_c = \frac{\sum_i x_i (n_c)_i (N_c)_i}{\sum_i x_i (N_c)_i}, \quad (7)$$

where x is the mole fraction of component oxide, n_c is the cross-link density per cation, N_c is the number of cations per glass formula unit and i denotes the component oxide. The values of n_c are listed in table 1. It can be noted that Poisson's ratio increased with decreasing average cross-link density, n_c . Another possible variable affecting the Poisson's ratio of glass is the relation of Poisson's ratio with E/S ratio applied to the three chain network [40,41]. From table 2, it is clear that the behaviour of the Poisson's ratio is nearly like the behaviour of E/S ratio, where σ relates to the modulus of elasticity and modulus of rigidity, as shown in the equation $\sigma = \left(\frac{E}{2S_{ij}} - 1\right)$. This relationship is applicable only to an isotropic body in which there is one value for the S_{ij} (constant of elasticity) and that value is independent of direction. Generally, this is not the case for single crystals; however, the relationship represented by these equations is a good approximation for glasses and for most polycrystalline ceramic materials.

Debye temperature, θ_D , represents the temperature at which nearly all modes of vibrations in a solid are excited. It was

Table 2. Longitudinal and shear velocities, V_L and V_S (m s^{-1}); longitudinal modulus, L (GPa); shear modulus, S (GPa); bulk modulus, K (GPa); Young's modulus, E (GPa); microhardness, H ; Poisson's ratio, σ ; average cross-link density, n_c ; number of bonds per unit volume, n_b ; E/S ratio, elastic moduli calculated by Makishima and Mackenzie's model, E_m , K_m and S_m (GPa); and Poisson's ratio, σ_m .

Sample no.	Ultrasonic velocity (m s^{-1})				Elastic moduli (GPa)					Elastic moduli (GPa)					
	V_L	V_S	L	S	E	K	H	σ	n_c	E/S	$n_b \times 10^{28} \text{ m}^{-3}$	E_m	K_m	S_m	σ_m
Sample 1	3755	2218	74.8	26.1	64.37	40	4.667	0.2321	2.609	2.4663	8.03	68.6	47.35	27.25	0.2585
Sample 2	3751	2218	74.5	26	64.13	39.7	4.667	0.2312	2.609	2.4665	8	70.57	49.4	27.96	0.2619
Sample 3	3704	2197	73.25	25.77	63.32	38.89	4.662	0.2286	2.609	2.4571	8.09	67.6	46.9	26.8	0.25977
Sample 4	3645	2149	72.3	25.1	62.03	38.8	4.465	0.234	2.58	2.4713	7.71	66.69	45.9	26.51	0.2578
Sample 5	3667	2157	71.5	24.7	61.20	38.5	4.368	0.2355	2.609	2.4777	8.05	68.3	47.27	27.12	0.2592
Sample 6	3735	2207	73.7	25.7	63.42	39.4	4.603	0.2317	2.609	2.4677	8.08	70.8	49.2	28.09	0.2602
Sample 7	3782	2242	75	26.35	64.79	40	4.761	0.2291	2.695	2.4588	8.12	71.3	49.46	28.29	0.25974
Sample 8	3674	2177	70.43	24.73	60.78	37.45	4.467	0.229	2.58	2.4577	7.93	67	45.9	26.65	0.2567

Table 3. Molar volume (V_a), mean velocity (V_m), Debye temperature (θ_D), glass transition temperature (T_g), thermal stability ΔT and IR major band position.

Sample no.	V_a ($\text{m}^3 \text{ mol}^{-1}$) ($\times 10^{-6}$)	V_m (m s^{-1})	θ_D (K)	T_g ($^{\circ}\text{C}$)	ΔT	IR major band position
Sample 1	32.98	3005.2	361.1	397	118	676
Sample 2	33.15	3004.7	362.1	411	109	674
Sample 3	32.76	2974.6	358.2	423	111	672
Sample 4	33.57	2912.6	349.5	379	92	657
Sample 5	32.92	2924.6	354.4	406	107	669
Sample 6	32.77	2990.1	360.1	421	102	675
Sample 7	33.41	3035.8	364.9	428	122	685
Sample 8	33.4	2948	351.6	395	96	666

T_g , ΔT and IR major band position are taken from part 1 of Mansour *et al* [35].

obtained from the measured ultrasonic velocities, V_L and V_S , using the expression [15]

$$\theta_D = \left(\frac{h}{k_B}\right) \left(\frac{3PN_A}{4\pi V_a}\right)^{1/3} v_m, \quad (8)$$

where h is the Planck's constant ($6.626 \times 10^{-34} \text{ J s}^{-1}$), k_B is the Boltzmann's constant ($1.3806 \times 10^{-23} \text{ J K}^{-1}$), N_A the Avogadro's number ($6.02214 \times 10^{23} \text{ mol}^{-1}$), V_a is the molar atomic volume calculated from the effective molecular mass and the density (i.e., M/ρ), P is the number of atoms in the chemical formula and v_m is the mean sound velocity defined by the relation: $v_m = (3v_L^3 v_S^3 / v_L^3 + v_S^3)^{1/3}$. The values of Debye temperature are listed in table 3. They increased from 349.5 to 364.9. Its increase implies an increase in the rigidity of the glass. Debye temperature potentially depends on changes in V_a , P and V_m as a result of modifier change, as shown in equation (11); however, it is observed that the behaviour of V_m is similar to that of θ_D and therefore, it can be concluded that the change in θ_D is controlled by the value of V_m . It is observed that the Debye temperature is inversely proportional to the ionicity of the modifiers, while it is directly proportional to T_g and ΔT . The highest value of θ_D (364.9 K) corresponds to the lowest value of the ionicity of the modifier TiO_2 in sample 7, the lowest value of $\text{TeO}_3/\text{TeO}_4$ (0.18), the highest value of T_g (428°C), the highest value of thermal stability ΔT (122°C) and the highest value of IR major band position (685 cm^{-1}), while the lowest value of θ_D (349.5 K) corresponds to the highest value of the ionicity of the modifier Ag_2O in sample 4, the highest value of $\text{TeO}_3/\text{TeO}_4$ (0.18), the lowest value of T_g (379°C), the lowest value of thermal stability ΔT (92°C) and the lowest value of IR major band position (657 cm^{-1}).

Makishima and Mackenzie presented a theoretical calculation model [42,43], in terms of chemical composition of oxide glasses, by only taking into consideration the dissociation energy of the oxide constituents per unit volume (G_i) and the packing density of ions (V_i). The elastic moduli and

Poisson's ratio were given as $E = 2V_i G_i$, for a polycrystalline glass ($E = 2V_t \sum_i G_i X_i$).

The V_i was defined as $V_i = (\rho/M_w) \sum_i V_i X_i$, where M_w is the effective molecular weight (kg mol^{-1}), ρ is the density (kg m^{-3}), X_i is the mole fraction of component i (mol%) and V_i is the packing factor obtained ($\text{m}^3 \text{ mol}^{-1}$); for example, in the following equation for oxide A_xO_y , $V_i = 6.023 \times 10^{23} [(4\pi/3)(xR_A^3 + yR_O^3)]$, R_A and R_O are the respective ionic radius of metal and oxygen. A semiempirical relation was found between the packing density of ions (V_i), dissociation energy per unit volume (G_i) and bulk modulus [43] $K = 2.4V_i^2 G_i$. The shear modulus and Poisson's ratio were given as $S = 3EK/9K - E$; $\sigma = E/2S - 1$. The results were compared with the experimental values given in table 2. The results show a good agreement between the experimental and the calculated values of Young's, bulk and shear moduli and Poisson's ratio, where sample 7 has the highest value of E , K , S in both experimental and theoretical values while sample 8 has the lowest values.

4. Conclusion

The incorporation of network modifiers Na_2O , Ag_2O , ZnO , MgO , CuO , NiO , TiO_2 and MnO_2 oxides into the ternary glass system $\text{TeO}_2\text{-WO}_3\text{-Nb}_2\text{O}_5$ showed an increase in the elastic properties. The addition of weak ionic modifiers, such as TiO_2 , to the ternary glass led to the preservation of the number of BOs in the glass network, resulting in higher values of elastic moduli, microhardness and Debye temperature; whereas, the addition of strong ionic modifiers, such as Ag_2O , led to the creation of more NBOs in the glass network, resulting in a decrease in these values. Finally, the ionicity of the modifiers was identified as a very important factor that affected the elastic properties of the tellurite glass systems.

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