

Research Article

Elastic Behavior of Borate Glasses Containing Lead and Bismuth Oxides

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PbO and Bi₂O₃ binary borate glasses with different compositions, (MO)_x(B₂O₃)_{1-x} (M = Pb, Bi), have been characterized and ultrasonic velocity as well as density is taken into account. In addition, the results have been compared with those of Ag, K, and Li oxide borate glasses from others. The ultrasonic velocities (both longitudinal and transverse) and density for (PbO)_x(B₂O₃)_{1-x} and (Bi₂O₃)_x(B₂O₃)_{1-x} have been measured accurately and elastic moduli as well as hardness and Poisson's ratio was determined. It has been demonstrated that density and ultrasonic velocities are enhanced by increasing PbO and Bi₂O₃ molar fraction with different values for each borate glass composition. However, the enhancement of ultrasonic velocities did not carry on continuously and after reaching a maximum point, they fell down dramatically. Both PbO and Bi₂O₃ showed almost similar glass improvement in case of density, ultrasonic velocity, and elastic moduli.

1. Introduction

Recently, great interest for electrochemical device applications, such as microbatteries, gas sensors, and electrochromics displays, has increased the attraction to glasses with high ionic conductivity [1, 2]. In comparison with crystalline materials, glasses show many advantages, where these isotropic materials do not have grain boundaries and fabrication is easy with low cost [3]. Generally, boron (B), silicon (Si), and phosphorous (P) oxides are the typical glass forming oxides where borate glasses have high potential for technological applications. Metal oxides such as Bi₂O₃ and Li₂O play an active role in semiconducting glasses [4, 5]. They can be added to glass former like B₂O₃ to enhance their properties [6]. For example, Bi₂O₃ containing glasses are good precursors for the preparation of ceramic high T_c superconductors [7–9]. Glasses containing PbO show high refractive indices with low crystallization tendency, as well as lower melting point and glass transition temperatures [10]. Although they do not form glass on their own, they modify a vitreous network to form glass when they combined with a second glass forming oxide such as B₂O₃ [11]. Several studies

have proved that a preferred host glass for the addition of the above oxides has been B₂O₃ [12, 13]. B₂O₃ is an absolute glass former and is a primary component of many large volume industrial glasses [14–16].

Ultrasonic examinations are considered in understanding the structural characteristics of glass network [17]. The properties of glass such as elastic properties are dependent on the interatomic forces and potential in lattice structure. The ultrasonic wave velocities as well as density of glass can be taken into account to discover the elastic properties of glass network which are strictly related to the glass structure [18]. Therefore, changes in the glass structure due to modifier doping can be directly noted.

Young's, bulk, shear, and longitudinal modulus can be obtained from the density of the solid and longitudinal wave transmission velocity through the solid [19].

Despite so many studies that have been done on borate glasses, the role of different oxides on borate glasses is not understood clearly. The purpose of this work is to compare the effect of different modifier oxides on ultrasonic velocities as well as density of borate glasses containing lead and bismuth oxides with those of other borate glass systems. In

TABLE 1: The room temperature physical and elastic properties of binary borate lead (PB) and bismuth (BB) glasses' density (ρ), molar volume (V_m), atomic volume (V_a), ultrasonic longitudinal (V_l) and transverse (V_s) velocities, longitudinal modulus (L), shear modulus (G), bulk modulus (K), Young's modulus (E), Poisson's ratio (σ), and microhardness (H). PB = $(\text{PbO})_X(\text{B}_2\text{O}_3)_{1-X}$, BB = $(\text{Bi}_2\text{O}_3)_X(\text{B}_2\text{O}_3)_{1-X}$.

Code	X (mol%)	ρ (kgm ⁻³)	V_m (cm ³ mol ⁻¹)	Velocity (m/s)			Elastic moduli (GPa)				H	σ
				V_a	V_l	V_s	L	G	K	E		
Pure Borate	0	1838	37.88	7.576	3469	1901	22.12	6.64	13.26	17.08	0.95	0.29
PB1	25	3781	28.57	6.722	4528	2597	77.52	25.50	43.52	64.00	4.17	0.25
PB2	30	4153	27.86	6.795	4235	2390	74.48	23.72	42.86	60.08	3.70	0.27
PB3	35	4439	27.79	7.036	4142	2305	76.16	23.58	44.71	60.17	3.53	0.28
PB4	40	4852	27.01	7.108	3960	2277	76.09	25.16	42.55	63.04	4.14	0.25
PB5	45	5044	27.50	7.535	3747	2197	70.82	24.35	38.36	60.28	4.25	0.24
PB6	50	5252	27.88	7.965	3707	2106	72.17	23.29	41.11	58.78	3.70	0.26
PB7	55	5528	27.87	8.321	3564	2034	70.22	22.87	39.72	57.56	3.68	0.26
PB8	60	5892	27.46	8.580	3279	1836	63.35	19.86	36.87	50.51	3.02	0.27
BB1	35	4907	42.46	8.50	3825	2385	71.79	27.91	34.58	65.98	5.92	0.18
BB2	40	5262	43.36	8.67	3767	2294	74.67	27.69	37.75	66.75	5.44	0.21
BB3	45	5613	44.18	8.84	3692	2196	76.51	27.07	40.42	66.39	4.94	0.23
BB4	50	6035	44.37	8.88	3556	2134	76.31	27.48	39.67	66.98	5.16	0.22
BB5	55	6122	46.98	9.40	3524	2098	76.03	26.95	40.10	66.05	4.93	0.23
BB6	60	6249	49.20	9.84	3479	2054	75.63	26.36	40.48	64.98	4.70	0.23
BB7	65	6502	50.33	10.07	3440	2015	76.94	26.40	41.74	65.41	4.60	0.24
BB8	70	6730	51.57	10.31	3356	1988	75.80	26.60	40.33	65.41	4.79	0.23

addition, elastic moduli, Poisson's ratio, and microhardness are obtained for different borate glass compositions by analyzing those mentioned properties. In present study, B_2O_3 plays as a glass former and Li_2O , PbO , Bi_2O_3 , Ag_2O , and K_2O serve as borate glass modifiers.

2. Materials and Methods

2.1. Sample Preparation. Bismuth and lead borate glasses were successfully synthesized by conventional melt quenching. Precursors with purity of bismuth oxide powder (purity 99.975%), lead oxide (purity 99.8%), and boron oxide (purity 99.8%) are used in the preparation of the glass samples. After mixing of B_2O_3 with oxides and drying the mixture in 400°C , the mixture was melted for 1 hour in furnace at 1000°C . After that, the melt was quenched into the preheated metal mould to obtain transparent glass cylinder with 12 mm diameter. In order to relieve the residual stress, the glass samples were annealed in 350°C for 1h. Finally, the cylindrical samples were cut and polished with 1-2 cm height for ultrasonic measurements.

2.2. Measurements. Archimedes method was taken into account to obtain density of the glasses as described elsewhere [16]. For ultrasonic velocity measurement in glass sample MATEC MBS 8000 was used. All measurements were taken at 5 MHz frequency and at room temperature. Elastic modulus (longitudinal, shear, bulk, and Young's) as well as Poisson's ratio and microhardness of bismuth and lead borate glasses with different contents has been determined from the measured ultrasonic velocities and density using the standard relations [20] (density (ρ), molar volume (V_m), ultrasonic longitudinal (V_l) and transverse (V_s) velocities, longitudinal

modulus (L), shear modulus (G), bulk modulus (K), Young's modulus (E), Poisson's ratio (σ), and microhardness (H)):

$$\begin{aligned}
 \text{Longitudinal modulus: } L &= \rho V_l^2, \\
 \text{Shear modulus: } G &= \rho V_s^2, \\
 \text{Bulk modulus: } K &= L - \frac{4}{3}G, \\
 \text{Young's modulus: } E &= (1 + \sigma) 2G, \\
 \text{Poisson's ratio: } \sigma &= \frac{L - 2G}{2(L - G)}, \\
 \text{Microhardness: } H &= \frac{(1 - 2\sigma)(1 + \sigma) 2G}{6(1 + \sigma)}.
 \end{aligned} \tag{1}$$

3. Results and Discussions

The codes, composition, density, molar volume, and elastic modulus are given in Table 1 for PbO and Bi_2O_3 borate glasses (PB and BB). To explore the changes in the structure of glasses, the density is a powerful tool where the structural softening/compactness, change in geometrical configuration, coordination number, cross-link density, and dimension of interstitial spaces of all borate glasses affected chemical composition [21]. It can be seen from Figure 1 that the density of glasses increases linearly with decreasing of B_2O_3 content. These changes in density by the increase of Bi_2O_3 and PbO are compared with Ag_2O , K_2O , and Li_2O borate glasses that were studied elsewhere [22, 23].

In general, density and molar volume illustrate opposite behavior; nevertheless in the case of Bi_2O_3 both density and

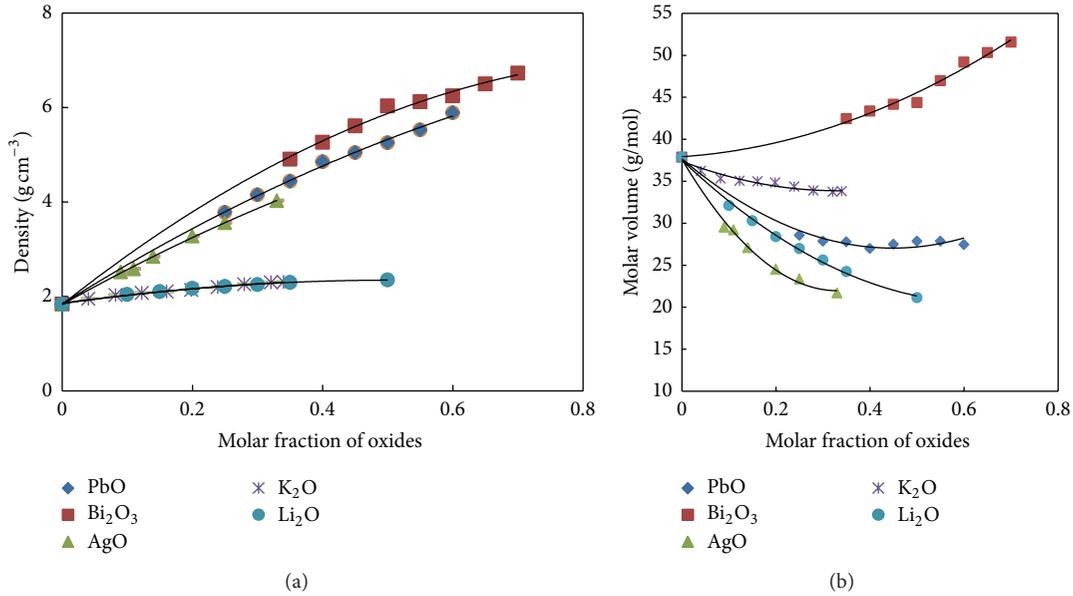


FIGURE 1: (a) Density (ρ) and (b) molar volume (V_m) of binary borate glasses as a function of modifier oxide concentrations of Pb and Bi oxides with those of Ag, K, and Li [22, 23].

molar volume increase with increasing of Bi_2O_3 content due to expansion of the glass network by bismuth (Table 2). Bi ions go into the glass network interstitially; therefore, some network bonds of B_2O_3 are ruptured and ionic bonds between Bi ions and individually bonded oxygen atoms replace them. Hence, if one assumed that only consequence of Bi cations addition was to break down the network bonds, then an increase in the molar volume with Bi_2O_3 content would be expected for the total vitreous range of the studied glass system.

The atomic masses of Pb, Bi, Ag, K, Li, and B are 207.20, 208.98, 107.87, 39.10, 6.94, and 10.81 and their atomic radii are 1.81, 1.63, 1.75, 2.77, 2.05, and 1.17 Å, respectively. This explains the increase in density. Figure 2 completely proves that density is linearly dependent on molar mass.

Figure 3 shows the longitudinal and transverse velocities (V_l) and (V_s). Both V_l and V_s increase with addition of modifier oxides that alkali borate glasses (LB, KB) have higher improvement than others. The highest belongs to LBs in which LB4 has value of 6702 and 3990 m/s for V_l and V_s , respectively. According to relations 1 to 4 elastic moduli are directly related to ultrasonic velocities (Figure 4).

Therefore, LBs have the highest elastic moduli as well, with 86.27 GPa, 35.20 GPa, 99.31 GPa, and 52.38 GPa for LB4's E, G, L, and K, respectively.

The variations of ultrasonic velocities and elastic moduli can be explained on the basis of structural consideration of borate glassy networks which depend on bond strength, packing density, coordination number, and cross-linking of units. In all present glass systems shown in Figure 3, their ultrasonic velocities are enhanced by increasing of modifier oxide contents. However, in higher content of oxides they are decreasing after reaching peak points where it is known as boron anomaly. These points could be described as optimum

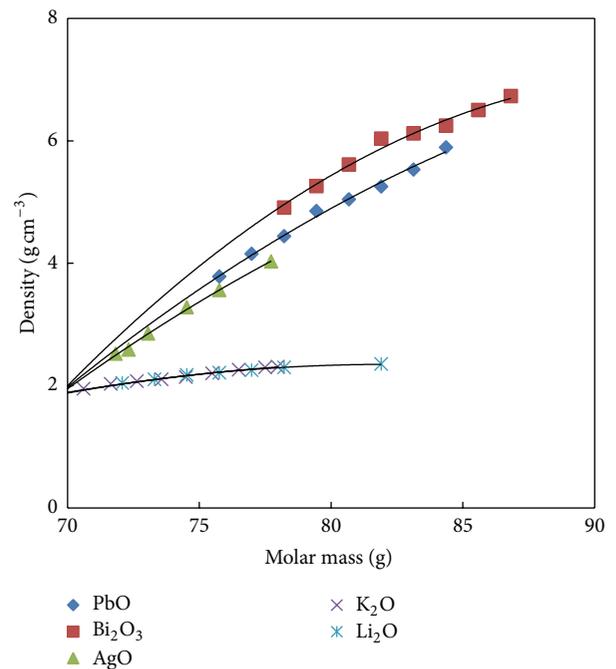


FIGURE 2: Changes in density (ρ) of binary borate glasses as a function of modifier oxide concentrations of Pb and Bi oxides with those of Ag, K, and Li molar masses (M) [22, 23].

compositions to obtain the best network structural modifications. The ultrasonic velocities and elastic moduli increase up to ~ 0.3 molar fractions in alkali borate oxides K_2O and Li_2O and then decrease. These optimum compositions are shown in Table 3, in addition to nonlinear functions ($y = ax^2 + bx + c$) of ultrasonic velocities with respect to molar fraction.

TABLE 2: Nonlinear regression analysis of the variables ($y = ax^2 + bx + c$) for density and molar volume in respect to molar fraction of Pb, Bi, K, Ag, and Li oxides [22, 23].

Oxides	Molar volume versus molar fraction				Density versus molar fraction			
	a	b	c	R^2	a	b	c	R^2
PbO	32.287	-21.278	37.384	0.935	-3.072	8.648	1.842	0.999
Bi ₂ O ₃	23.045	3.743	37.930	0.985	-5.702	10.941	1.831	0.998
K ₂ O	52.798	-47.373	37.672	0.979	-1.443	1.791	1.863	0.988
Ag ₂ O	142.660	-94.498	37.623	0.994	-3.072	7.695	1.831	0.999
Li ₂ O	41.412	-53.041	37.517	0.997	1.921	1.957	1.849	0.996

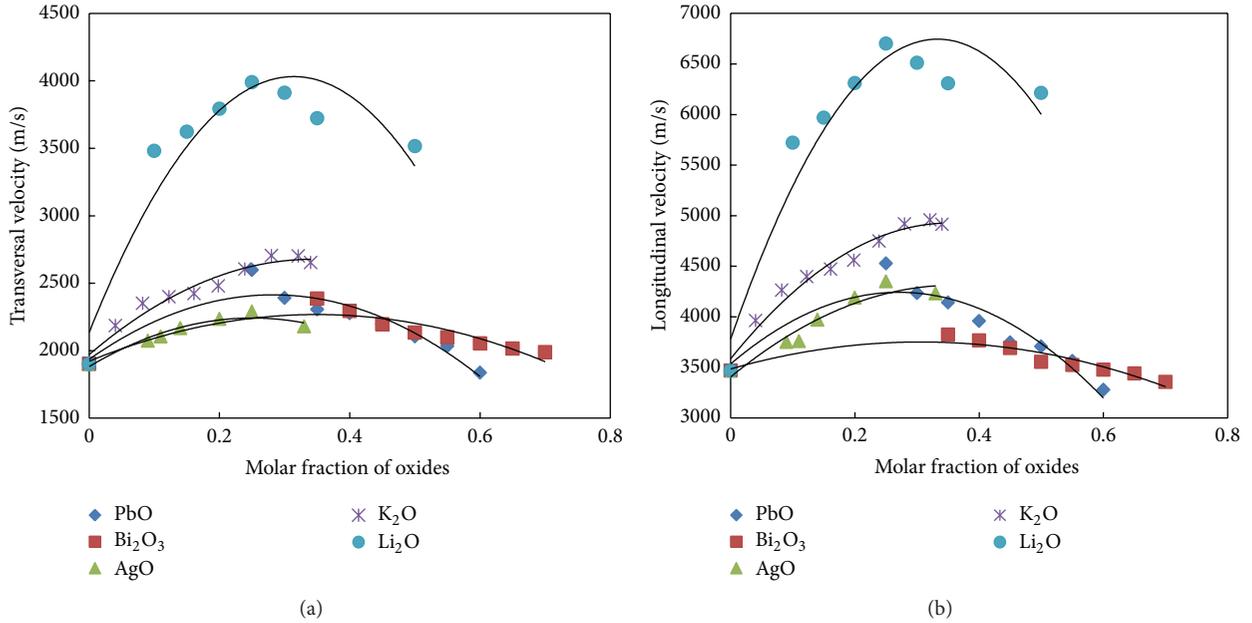


FIGURE 3: (a) Transverse (V_s) and (b) longitudinal (V_l) sound velocities of binary borate glasses as a function of modifier oxide contents of Pb and Bi oxides with those of Ag, K, and Li [22, 23].

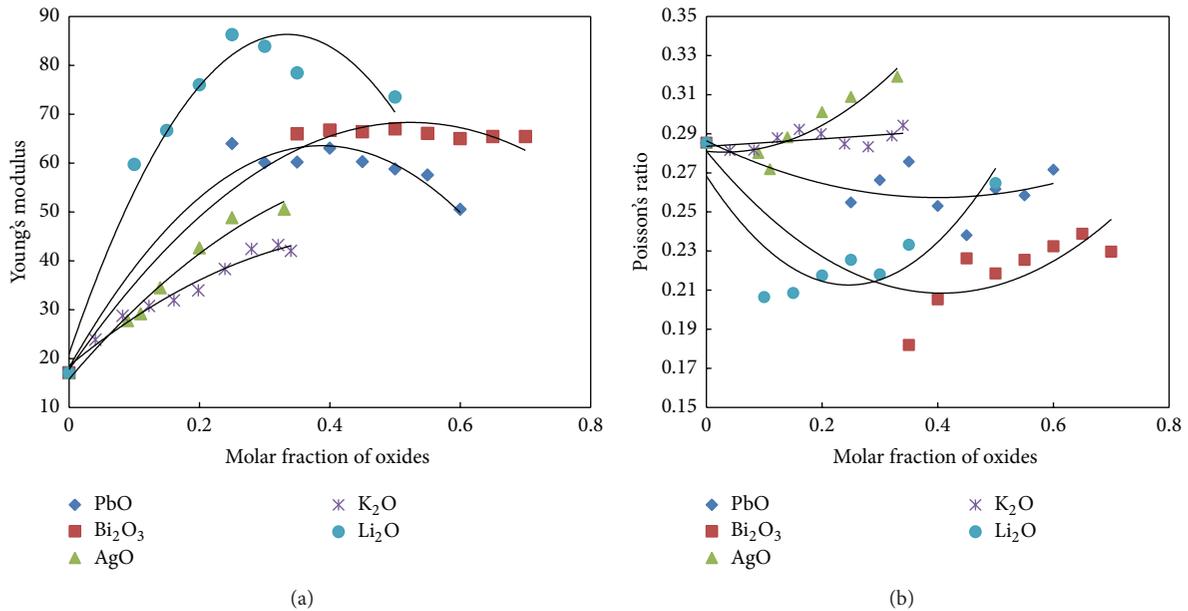


FIGURE 4: (a) Young's modulus and (b) Poisson's ratio of binary borate glasses as a function of modifier oxides concentration of Pb and Bi oxides with those of Ag, K, and Li [22, 23].

TABLE 3: Nonlinear regression analysis of the variables ($y = ax^2 + bx + c$) for longitudinal and transverse ultrasonic velocities as well as optimum contents of modifier oxides (op = optimum points) [22, 23].

Oxides	V_l versus molar fraction					V_s versus molar fraction					V_l versus V_s		
	a	b	c	R^2	OP	a	b	c	R^2	OP	b	c	R^2
Pb	-9650	5228	3535	0.88	0.25	-5993	3384	1936	0.89	0.25	1.64	270.84	0.91
Bi	-2845	1740	3486	0.89	0.35	-2839	1982	1922	0.83	0.35	1.00	1.51	0.87
K	-9796	7200	3623	0.97	0.30	-5186	3817	1994	0.95	0.30	1.86	-78.51	0.99
Ag	-7024	5047	3406	0.93	0.25	-5743	2877	1882	0.96	0.25	2.40	-1.14	0.97
Li	-26720	17800	3781	0.92	0.25	-19218	12074	2135	0.91	0.25	1.53	566.66	0.98

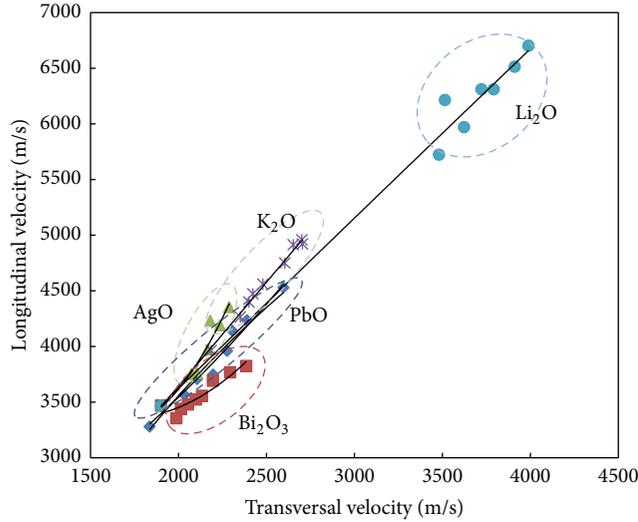


FIGURE 5: Correlation between transverse (V_s) and longitudinal (V_l) ultrasonic velocities in binary borate glasses containing Pb and Bi oxides, as well as oxides of Ag, K, and Li. [22, 23].

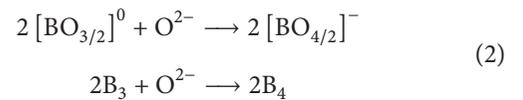
It also shows that longitudinal (V_l) and transverse (V_s) ultrasonic velocities increase and decrease in the same pattern, however with different rates (Figure 5). V_l and V_s in borate glasses with bismuth oxides increase with the same rate. However, in ABs and KBs borate glasses rising of V_l is almost twice V_s .

Modulus of elasticity can be a characteristic of glass since glass is also considered as an elastic substance [24, 25]. This modulus increases as the expansion at a certain applied stress reduces. That would have happened if the glass structure was rigid and therefore contained the fewest possible nonbridging oxygen. When an oxide is introduced to borate, the field strength of the cation determines the strength of the structure. By increasing the oxides content in the borate glass, the structure becomes more rigid, and then density increases as well; hence, the elastic moduli increase [17, 26]. It may also be noted that the rate of change in elastic moduli is more evident in longitudinal modulus (L) than in case of transverse or shear modulus (G). This designates resistance to deformation which is probably due to presence of large number of covalent bonds.

The increase in Young's modulus values with addition of modifier oxides has contributed to increase in rigidity of

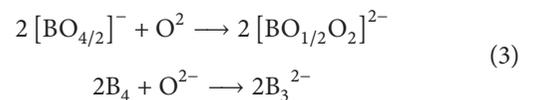
borate glassy network. This may be due to the increasing of the modifiers [27] which can increase Coulomb contribution to the lattice energy. This mechanism was discussed for the role of Bi and Pb [28, 29]. Shear and Young's moduli have a straight connection with bond bending (F_b) and bond stretching (F) force constant, respectively [30]. So, the increase in E values with modifier oxides can be attributed to the increase in F , and the increase of G with oxide concentrations can indicate the increase in F_b bulk and Young's moduli. Compositional variation of experimental values of Poisson's ratio is demonstrated in Figure 4. Poisson's ratio decreases in full range but faster in Bi-rich. The cross-link density of two, one, and zero is related to Poisson's ratio of 0.15, 0.3, and 0.4, respectively [31]. Poisson's ratio starts from 0.27 in Pb-rich and decreases to 0.21 in Bi-rich limit which means that the cross-link density increases from nearly one to two.

As mentioned the introduction of alkali oxides (Li_2O and K_2O) enhanced ultrasonic velocities and elastic moduli of borate glasses very well. It can be explained that, in alkali borate glasses, conversion of the trigonal borons ($BO_{3/2}$ units) to tetrahedral borons ($BO_{4/2}$ units) is by the coordination of O^{2-} to two trigonal borons:



Modified borate glass structures by adding alkali oxides are shown in Figure 6. As long as maximum of 50% of B_3 changes to B_4 , the creation of B_4 proceeds [12]. Then, by further increasing of the modifier concentration, the concentration of B_4 decreases quickly. In diborate composition B_3 and B_4 concentrations are equivalent, and the corresponding mole fraction of the alkali oxide is 0.33.

In a case of lithium borate glasses, the diborate structure does not seem to favor B_4 - B_4 connections [32]. Structure in borate glasses with less than 0.33 alkali oxide molar fractions takes rise to open by the connectivity of the tetrahedral and the diffuse spread of the negative charge together. Nevertheless, for alkali oxide mole fractions more than 0.33, B_4 units collapse and form



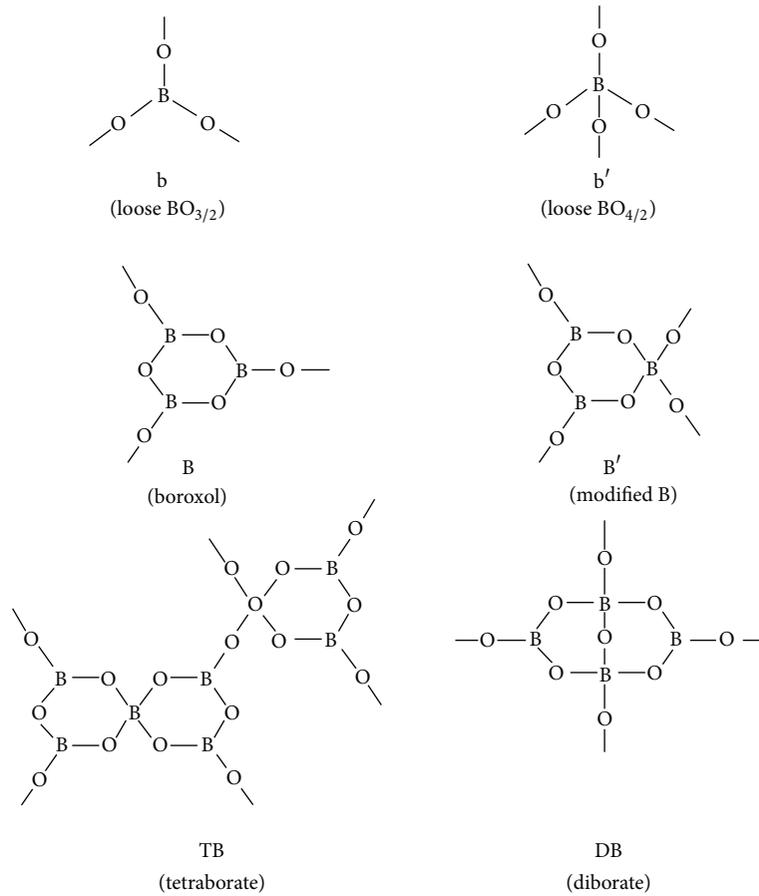


FIGURE 6: Different structural units in alkali borate glasses [12].

Consequently, network collapses and volume is utilized. Therefore, expansion of network occurs which contributes to network rigidity reduction. By decreasing the atomic bond rigidity ultrasonic velocities and elastic moduli reduce. In addition, rapid reversals occur in the variation of refractive indices, T_g , thermal expansivity, and so forth, as a function of alkali composition [12]. All of these variations are directly or indirectly related to energy density and the observed variations are often referred to as borate anomaly.

Other borate oxides, PB, BB, and AB, are showing almost similar behavior in terms of ultrasonic velocities and elastic moduli. Modification of borate glasses by PbO can be explained by two approaches. The first one is the assumptions that cation ions of oxides, such as Pb^{2+} in PBs, enter interstitially as a result of the addition of oxides, such as PbO, into the borate glass networks. Therefore, some types of modification of B–O–B linkages, which already exists in the glass, into B–O–Pb bonds will occur. The conversion of these linkages results in an increase in the molar volume and a decrease in the packing density which expand the glass network. Therefore, the reduction in the rigidity due to the formation of PBOs will contribute to the decrease in the ultrasonic velocity.

The second approach is the suggestion by Higazy and Birba [28], where the longitudinal strain in a bond is directly

dependent on the bond stretching force constant. In the studied glasses, the longitudinal strain in the main chains (B–O–B linkages) is affected by the modifying role of PbO, as these oxides decrease the overall stretching force constant ($F_{\text{B-O}}$) and consequently increase the bond length of these linkages, so the longitudinal wave velocity will decrease. On the other hand, the shear strain changes with the bond bending force constant (F_b). Thus, the decrease of the shear wave velocity indicates that the addition of the modifiers has no bending effect on the behavior of bond bending force constant; that is, the increase in the modifiers cations did not contribute to filling the network interstices.

4. Conclusion

New binary borate base glasses added with bismuth and lead oxides were successfully prepared and their structures were analyzed by ultrasonic waves. Three other borate glasses including lithium, potassium, and silver oxides from others were compared. It has been demonstrated that density and ultrasonic velocities are enhanced by increasing PbO and Bi_2O_3 molar fraction with different values for each borate glass composition. However, the enhancement of ultrasonic velocities did not carry on continuously and after reaching a maximum point, they fell down dramatically.

Density of borate glasses is enhanced uniformly by increasing of modifier oxides content where bismuth, lead, and silver oxides glasses demonstrated larger value than potassium and lithium oxides due to their larger molar masses.

The ultrasonic velocities of borate glasses added with modifier oxides increased as a result of higher glass network rigidity and vibration. Additionally, while the contents of all modifier oxides enhanced, the elastic moduli and micro-hardness were improved. However, Poisson's ratio illustrates a slight change.

Improving in ultrasound velocities as well as elastic moduli is much higher for Li_2O borate glasses due to their higher compactness as a result of lower molar volume.

Oxide borate glasses demonstrated declining in ultrasonic velocities and elastic moduli in higher contents of modifier oxides which occurred due to reduction of glass network rigidity. In alkali borate glasses such as Li_2O and K_2O borate glasses, B_4 networks collapse into B–O–B linkages. However, in other borate glasses B_4 turns into B–O–M in which both address the decreasing of glass network rigidity.

In general comparison, both PbO and Bi_2O_3 showed almost similar glass improvement in case of density and ultrasonic velocity; nevertheless Li_2O in the other study pointed up the best improvement behavior due to lower density, higher ultrasonic velocity, and elastic moduli enhancements.

Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

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