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Determination of Ultrasonic Velocities Theoretically for Tellurite Glasses Using Makishima and Mackenzie Model

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Abstract

Makishima and Mackenzie model was used to determine both ultrasonic wave velocities (Longitudinal and Shear) theoretically for many tellurite glasses. The model is mainly depends on the values of the experimentally measured density. Then, the problems of determining the measured densities of amorphous glasses (as the density depends on the geometry of the network structure of these glasses) and the slope of linear regression between the experimentally determined bulk modulus and the product of packing density and experimental Young's modulus, were solved in this search work. The results showed good agreement between the experimentally measured values of densities and both ultrasonic wave velocities, and those theoretically determined.

Keywords: Glasses; Ultrasonic wave velocities; elastic moduli; Makishima & Mackenzie model
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1. Introduction

TeO_2 – based glasses have recently become of great interest for use in optical communication systems, Kosuge et al., 1998. Tellurite glasses are well known to have the highest refractive indices among oxide glasses in the visible and near IR region, Afifi and Marzouk, 2003. The systems TeO_2 – WO_3 – K_2O, TeO_2 – WO_3 – Bi_2O_3 and TeO_2 – WO_3 – PbO are the most promising in this respect because they contain oxides of the heaviest metals, such as tungsten, lead and bismuth, Kosuge et al., 1998, Safonov et al., 1992 and Safonov and Semonova, 1989. Previous studies by Gaman et al., 1972 and Rhee et al., 1974 on vanadium tellurite glasses showed that they are semiconducting glasses and they switch when a high electrical field is applied.

Also, Pure B_2O_3 glass has a random three-dimensional network of BO_3 triangles with a large fraction of almost planar B_3O_6 boroxol rings. It is known that borate glasses show the anomalous composition dependences of physical properties, such as density, Kodama et al., 1994, sound velocity, Matsuda et al., 2009 and Kawashima et al. 2009 and thermal expansion Kathrine K. Smyth and Matthew B. Magida, 1983, by the addition of alkali oxide to pure B_2O_3 glass.

Longitudinal and shear ultrasonic wave velocities were measured by Gaafar et al., 2009 in binary Li_2O-$2B_2O_3$ glasses doped with different transition metal oxides (TMOs) (where TMO = V_2O_5, Fe_2O_3, Cr_2O_3, NiO, TiO_2, MnO_2 and CuO) using pulse echo technique. Measurements were carried out at 4 MHz frequency and at room temperature. Elastic moduli and some other physical parameters such as acoustic impedance, Debye temperature, thermal expansion coefficient, and latent heat of melting were calculated. Results indicated that these parameters depend upon the TMO modifier i.e., the ionic radius of the transition metal cation. Quantitative analysis has been carried out, in order to obtain more information about the structure of these glasses, based on bond compression model, and the Makishima and Mackenzie model, i.e., the cation-anion bond of each TMO.

The ternary xV_2O_5–$(40- x)Li_2O$–$60B_2O_3$ glass system, where x = 1, 2, 3, 4 and 6 mol%, was prepared by Sidkey et al., 2008 using normal quenching. The composition dependence of these parameters, in addition to the glass-transition temperature, suggested that vanadium ions were incorporated into these glasses as a network modifier, resulting in the reconversion of BO_4 tetrahedra to BO_3 triangles by the breaking of B–O–B linkages and the formation of nonbridging oxygens (NBOs). The

results were explained quantitatively in terms of fractal bond connectivity, average atomic volume, network dimensionality, packing density, number of network bonds per unit volume, cross-link density and atomic ring size. The Makishima and Mackenzie model appeared to be valid for the studied glasses when the fate of BO_4 tetrahedra and creation of NBOs are taken into account.

M. S. Gaafar et al., 2011 have introduced the Artificial Neural Network (ANN) technique to simulate and predict important parameters such as density, longitudinal and shear ultrasonic velocities and elastic moduli (Longitudinal and shear moduli) for more than 30 glass compositions. The authors showed that the ANN results were found to be in successful good agreement with those experimentally measured parameters. Then the authors have used the ANN model to predict the acoustic properties of some new tellurite glasses. For this purpose, four glass systems x Nb_2O_5 - (1-x) TeO_2, 0.1 PbO - x Nb_2O_5 - (0.9-x) TeO_2, 0.2 PbO - x Nb_2O_5 - (0.8-x) TeO_2 and 0.05 Bi_2O_3 - x Nb_2O_5 - (0.95-x) TeO_2 were prepared by the authors using melt quenching technique. The results of ultrasonic velocities and elastic moduli showed that the addition of Nb_2O_5 as a network modifier provides oxygen ions to change [TeO_4] tbps into [TeO_3] bps.

In the present search work, many different tellurite and borate glass compositions were used to solve the problems of Makishima and Mackenzie model (as it depends on the experimentally measured density values) to make theoretical determination of the densities of these glasses and consequently obtain both the ultrasonic (Longitudinal and Shear) wave velocities theoretically.

2. Theoretical Considerations

Makishima and Mackenzie, 1973 and 1975 proposed a theoretical model for the direct calculation of Young's modulus of oxide glasses in terms of their chemical compositions taking into consideration the two parameters; dissociation energy of the oxide constituents per unit volume (G_i) and packing density of glasses (V_t). In a simple one component glass such as fused silica, Young's modulus was given as:

$$E_{(M-M)} = 2V_t G \tag{1}$$

For poly-component glasses, Young's modulus was given as,

$$E_{(M-M)} = 2V_t \sum_i G_i x_i \tag{2}$$

where $E_{(M-M)}$ is Young's modulus, x_i is the molecular fraction of component i in the glass system.

The packing density V_t, is defined as:

$$V_t = \left(\frac{\rho_m}{M}\right)\sum_i V_i x_i \tag{3}$$

where M is the effective molecular weight, ρm is the density of the poly-components glass system and V_i is the packing factor of the oxide and can be determined from the equation for an oxide AxOy having ions A and O with Pauling ionic radii given as:

$$V_i = 6.023\times10^{23}\left(\frac{4\pi}{3}\right)\left[xR_A^3 + yR_O^3\right] \tag{4}$$

where R_A and R_O are the respective ionic radii of the cation and anion.

Makishima and Mackenzie, 1973 and 1975 studied the agreement between the experimental values of Young's modulus (E_{M-M}), of many oxide glasses and those calculated from their theoretical model. The agreement was good for the majority of glasses, especially for silica glass, proving that Eq. (1) is satisfactory for estimating elastic modulus of a simple one component glass system. A problem appeared for poly-component glasses with Young's modulus values greater than 90 GPa where the theoretically calculated values were found to be less than the measured values. Furthermore, Makishima - Mackenzie model also requires the measured density of glass for which the glass-melting needs to be done before estimating their elastic moduli. Therefore, Makishima and Mackenzie refined their model in order to estimate Young's modulus values directly from the designed compositions without melting.

Makishima and Mackenzie, 1975 extended their theoretical study and combined Gruneisen's equation with Young's modulus equation of glass to derive new formula for the theoretical calculation of bulk modulus, shear modulus, and Poisson's ratio of glass. According to Gruneisen first rule, the bulk modulus, K, is given as:

$$KV_o = \frac{1}{9}mn|U_o| \tag{5}$$

where V_o is the equilibrium volume found where the net force vanishes, m and n are constants, and U_o is the equilibrium energy for volume V_o. Then, they obtained the following equation:

$$K_{(M-M)} = \left[\frac{m|m-n|}{24\pi}\right]\beta V_t E_{(M-M)} \quad (6)$$

Makishima and Mackenzie pointed out that it was very difficult to calculate m and n which are important factors. They therefore, examined the correlation between bulk modulus and the product of packing density and Young's modulus and found good linearity between the experimentally obtained bulk modulus (using ultrasonic technique) and the product of the packing density multiplied by the experimentally obtained Young's modulus of many glasses. Therefore, the slope β of the linear regression was determined, and the bulk modulus of the glass was expressed as:

$$K_{(M-M)} = \beta V_t E_{(M-M)} \quad (7)$$

Therefore, Young's modulus $E_{M\text{-}M}$, shear modulus $S_{(M-M)}$, and Poisson's ratio σ_{M-M}, are given as:

$$E_{(M-M)} = 2V_t G \quad (8)$$

$$S_{M-M} = \frac{3E_{(M-M)}K_{(M-M)}}{9K_{(M-M)} - E_{(M-M)}} \quad (9)$$

$$\sigma_{M-M} = \frac{E_{(M-M)}}{2S_{(M-M)}} - 1 \quad (10)$$

Now there are two problems, which are (ρ_m) is the experimentally obtained density of the poly-components glass system and the slope of the linear regression β.

Concerning the (ρ_m), Zou and Toratani, 2001 derived modified equations on the basis of Makishima - Mackenzie model for direct determination of the elastic modulus of glasses from their compositions. According to the traditional crystalline model of glass structure, the authors assumed that a polycomponent glass is a mixture of microcrystals with the same local structure of corresponding oxide (A_xO_y) component i, so the Young's modulus coefficient Ei, of the component i can be given as:

$$E_i = 2V_i G_i \quad (11)$$

where Vi is the packing density factor of component i which is equivalent to (V_t) only when the glass is simple one component system. If the A – O bond energy of oxide A_xO_y is similar, in the crystal or in the glass, the packing factor (V_i) can be evaluated from the basic properties of oxide A_xO_y as:

$$V_i = \left(\frac{\rho_i}{M_i}\right)6.023\times10^{23}\left(\frac{4\pi}{3}\right)\left[xR_A^3 + yR_O^3\right] \quad (12)$$

Based on the foundational definition of specific modulus Messier and Patel, 1995, the specific modulus coefficient (S_i), of component i in a glass can be expressed as:

$$S_i = \frac{E_i}{\rho_i} = 2G_i\frac{V_i}{\rho_i} \quad (13)$$

Substituting Eq. (12) for (13), the modulus coefficient, Si, can be rewritten as:

$$S_i = 2\left(\frac{G_i}{M_i}\right)6.023\times10^{23}\left(\frac{4\pi}{3}\right)\left[xR_A^3 + yR_O^3\right] \quad (14)$$

Then the specific modulus of a poly-component glass can be expressed, based on the modified additive rule, as follows:

$$S = \gamma\sum_i S_i x_i \quad (15)$$

where (γ) is the modified coefficient introduced by the authors. Eq. (12) was established under assumption that oxide A_xO_y has the same co-ordination number regardless of the oxide being in the crystal or in the glass. However, Eq. (14) was established under assumption that oxide A_xO_y has the same co-ordination number regardless of the oxide being in the crystal or in the glass. However, the local structure of each oxide component in glass is, in general, somewhat different to that in its crystal state. Experiments carried out by the authors showed that the difference in results is mainly due to variation of the packing density of oxide component in glass. If the local structure of oxides does not change when added them into glass, the density of the glass based on the additive rule (ρ_i) can be simply evaluated by integrating the densities of the oxide components weighted by their molecular fractions. However, the measured densities are, in fact, different to the calculated values due to the variation of their co-ordination structures. Such a difference between the measured and calculated densities of glass just reflects the variation of local structures of oxide components in the glass. The modified coefficient (γ) is therefore defined as:

$$\gamma = \frac{\sum_i \rho_i x_i}{\rho_m} \quad (16)$$

where ρm is the measured density of the glass. Therefore, the problem of (ρ_m) is the density of the poly-components glass system can be solved by linear regression between (ρ_m) is the density of the poly-components glass system and the density of the glass composition based on the additive rule (ρ_i) of different glass compositions and the slope of regression will be equals to the coefficient (γ). Therefore the density can be obtained theoretically using equation (34) which takes the form:

$$\rho_{cal} = \gamma \sum_i \rho_i x_i \quad (17)$$

where (ρ_{cal}) is the calculated density of amorphous glass composition. Moreover, the problem of the slope (β) of the linear regression between the experimental bulk modulus K_e and the product of V_t and experimental Young's modulus (E_e) was solved.

Then, the ultrasonic velocities (U_l, the longitudinal velocity and U_s, the shear velocity) can be computed using the following equations;

$$\mathbf{L} = \boldsymbol{\rho} \mathbf{U_l^2}$$

$$\mathbf{S} = \boldsymbol{\rho} \mathbf{U_s^2}$$

$$\mathrm{E} = 2(1+\sigma)\mathrm{S} \quad (18)$$

$$\mathbf{K} = \mathbf{L} - \left(\left(\frac{\mathbf{4}}{\mathbf{3}}\right)\mathbf{S}\right)$$

3. Analysis and discussions

Fig. 1 shows the relations between the measured densities (ρ_m) for many different tellurite glasses, which were taken from references Sidkey et al., 2008, Sidkey and Gaafar, 2004, El-Mallawany, 1990, Paul et al., 2000, Lambson et al., 1985, El-Mallawany and Saunders, 1988, El-Mallawany and Saunders, 1987, Hart, 1983, Rajendran, 2003, Saddeek, 2007, Abd El-Aal and Afifi, 2009, Gaafar et al., 2009, Gaafar, et al., 2009, Gaafar et al., 2009, Singh et al., 1989, Hager, 2002, Saddeek, 2009, and the calculated densities of the glass compositions based on the additive rule (ρ_i). The slope of the relation which is the modified coefficient ($\gamma_{Tellurite}$) for tellurite glasses. The modified coefficient value as found to be 0.948 for tellurite. Then Eq. 17 will take the following forms:

$$\rho_{cal} = 0.948 \sum_i \rho_i x_i \quad (19)$$

for tellurite glasses with correlation factor 99.8 %. Results of the measured densities and calculated densities are listed in Table 1(a) and 1(b) for tellurite glasses.

Fig. 2 shows the relations between the experimentally determined bulk moduli (K_e) and the product of packing density and Young's modulus ($V_t.E_{M\text{-}M}$) for tellurite glass compositions. The slope ($\beta_{Tellurite}$) was found to be as 1.319 for tellurite glasses. Then Eq. 7 will take the following forms:

$$K_{cal} = 1.319 . V_t E_{(M-M)} \quad (20)$$

for tellurite glasses with correlation factor 99.1 %.

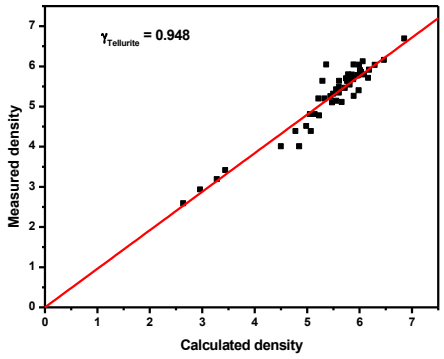

Fig. 1. The relation between the measured densities and the calculated densities of the glass compositions based on the additive rule (ρi) for tellurite glasses.

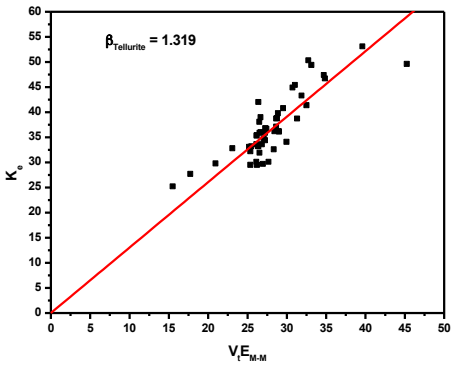

Fig. 2. The relation between the measured bulk moduli and the product of the packing density and calculated Young's modulus for tellurite glasses.

Table 1: (a) Glass Composition (mol. %)

Glass compositions (mol. %)													
TeO_2	WO_3	K_2O	CeO_2	CuO	B_2O_3	Li_2O	Bi_2O_3	V_2O_5	La_2O_3	Nb_2O_5	PbO	Sm_2O_3	BaO
0.8	0.2	0											
0.8	0.15	0.05											
0.8	0.1	0.1											
0.8	0.05	0.15											
0.8	0	0.2											
0.9			0.1										
0.84				0.16									
0.821				0.179									
0.81				0.19									
0.797				0.203									
0.1					0.6	0.3							
0.2					0.5	0.3							
0.3					0.4	0.3							
0.35					0.35	0.3							
0.5							0	0.5					
0.5							0.05	0.45					
0.5							0.1	0.4					
0.5							0.15	0.35					
0.5							0.2	0.3					
0.5							0.25	0.25					
0.65				0				0.35					
0.65				0.075				0.275					
0.65				0.1				0.25					
0.65				0.125				0.225					
0.65				0.15				0.2					
0.65				0.175				0.175					
0.9									0.1				
0.669										0.124	0.207		
0.75									0.04		0.21		
0.9												0.1	
1													
0.85	0.15												
0.8	0.2												
0.79	0.21												
0.67	0.33												
0.77	0.14												0.09
0.74	0.21		0.05										
0.77	0.2								0.03				
0.5	0.3										0.2		
0.75	0.2											0.05	
0.95										0.05			
0.9										0.1			
0.85										0.15			
0.8										0.2			
0.9											0.1		
0.7										0.2	0.1		
0.7										0.1	0.2		
0.6										0.2	0.2		
0.75							0.05			0.2			
0.7							0.05			0.25			

Table 1: (b) Measured density (ρ (M)), theoretical density (ρ (cal)), measured longitudinal velocity (U_l (M)), theoretical longitudinal velocity (U_l (cal)), measured shear velocity (U_s (M)), theoretical shear velocity (U_s (cal)), experimental bulk modulus (Ke (M)), theoretical bulk modulus (K (cal)) and $V_t.E$, the product of packing density and theoretical Young's modulus for tellurite glasses.

TeO_2	ρ (M)	ρ (cal)	U_l (M)	U_l (cal)	U_s (M)	U_s (cal)	K_e (M)	K_{th} (cal)	$V_t.E_{(M-M)}$
0.8									
0.8	5.766	5.664	3366	3381	1951	1932	36.1	36.6	29
0.8	5.453	5.432	3288	3310	1888	1897	33	33.4	25.3
0.8	5.091	5.199	3190	3229	1805	1857	29.7	30.3	21
0.8	4.766	4.967	3130	3139	1734	1811	27.6	27.2	17.8
0.9	4.5	4.735	3058	3035	1681	1757	25.1	24.1	15.6
0.84	5.706	5.561	3429	3351	2102	1939	33.5	34.6	26.2
0.821	5.622	5.471	3390	3375	1981	1949	35.2	34.6	26.2
0.81	5.707	5.482	3476	3383	1887	1952	41.9	34.9	26.5
0.797	5.785	5.489	3477	3388	2034	1954	38	35.1	26.6
0.1	5.608	5.497	3684	3394	2232	1957	38.9	35.3	26.7
0.2	2.58	2.509	5869	6242	3381	3369	49.5	59.8	45.3
0.3	2.919	2.813	5556	5580	3089	3065	53	52.3	39.7
0.35	3.177	3.117	5090	5021	2875	2806	47.3	45.9	34.8
0.5	3.404	3.269	4714	4772	2752	2689	41.3	42.9	32.6
0.5	3.996	4.274	3655	3617	2096	1936	30	34.6	26.2
0.5	4.376	4.537	3591	3569	2056	1937	31.8	35.1	26.6
0.5	4.797	4.8	3507	3520	1995	1935	33.5	35.5	26.9
0.5	5.188	5.063	3416	3470	1937	1932	34.6	35.8	27.1
0.5	5.624	5.326	3330	3421	1882	1926	35.8	36	27.3
0.65	6.031	5.589	3250	3371	1833	1920	36.7	36.1	27.3
0.65	3.996	4.604	3992	3763	2362	2042	34	39.6	30
0.65	4.376	4.815	3694	3587	2159	1953	32.5	37.5	28.4
0.65	4.797	4.885	3364	3524	1950	1922	30	36.6	27.8
0.65	5.188	4.955	3210	3459	1859	1891	29.6	35.7	27
0.65	5.624	5.025	3066	3392	1769	1859	29.4	34.6	26.3
0.9	6.031	5.095	2939	3322	1682	1827	29.4	33.5	25.4
0.669	5.685	5.452	3415	3351	2093	1945	33.1	33.7	25.6
0.75	5.888	5.705	3294	3287	1906	1884	35.3	34.6	26.3
0.9	6.145	6.138	3038	2994	1711	1730	32.7	30.5	23.2
1	5.782	5.557	3447	3386	2149	1975	33.1	34.8	26.4
0.85	5.101	5.374	3404	3438	2010	1999	33.1	34.9	26.5
0.8	5.25	5.591	3532	3398	2031	1950	36.6	36.2	27.5
0.79	5.766	5.664	3366	3429	1951	1959	36.1	37.6	28.5
0.67	5.39	5.678	3561	3435	2080	1961	37	37.9	28.7
0.77	5.7	5.852	3555	3509	2098	1983	38.6	41.4	31.4
0.74	5.669	5.58	3378	3360	1952	1930	35.9	35.3	26.8
0.77	5.781	5.772	3408	3416	2011	1939	36	38.4	29.1
0.5	6.027	5.687	3480	3445	2035	1967	39.7	38.2	28.9
0.75	6.68	6.506	3169	3182	1786	1793	38.7	38	28.8
0.95	6.11	5.755	3515	3470	2067	1986	40.7	39	29.6
0.9	5.475	5.323	3352	3439	1876	1981	35.8	35.1	26.6
0.85	5.414	5.272	3464	3568	1911	2039	38.6	37.9	28.7
0.8	5.302	5.222	3677	3692	1949	2096	44.8	40.6	30.8
0.9	5.242	5.171	3922	3812	2087	2150	50.2	43.3	32.8
0.7	5.845	5.722	3091	3259	1746	1890	32.1	33.5	25.4
0.7	5.529	5.52	3767	3789	2079	2124	46.6	46	34.9
0.6	6.025	5.97	3151	3249	1784	1842	34.3	36	27.3
0.75	5.904	5.868	3692	3503	2177	1954	43.2	42.1	32
0.7	5.33	5.324	3633	3680	1879	2092	45.3	41	31.1
0.85	5.13	5.273	3990	3803	2175	2149	49.3	43.8	33.2

The results of the experimentally determined bulk moduli (K_e), theoretically obtained bulk moduli (K_{cal}) and the product of packing density and Young's moduli ($V_t.E_{M-M}$) for the glass compositions under investigation were listed in Table 1(a) and 1(b).

Then both the ultrasonic wave velocities (U_l & U_s) were obtained theoretically using equations (18) and compared with those experimentally determined as shown in Fig. 3 and 4 and Table 1(a) and 1(b). The results showed good agreement between those obtained experimentally and theoretically obtained, and the deviations were found to be about 3% on average. Finally, those results can led one to conclude that this method is good for theoretical prediction or simulation of both ultrasonic wave velocities for any tellurite glasses using the data of packing densities and dissociation energies of the constituent oxides.

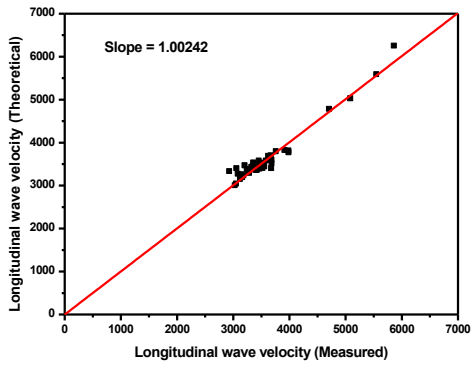

Fig. 3. The relation between the experimentally determined longitudinal ultrasonic wave velocities and the theoretically determined longitudinal ultrasonic wave velocities for Tellurite glasses.

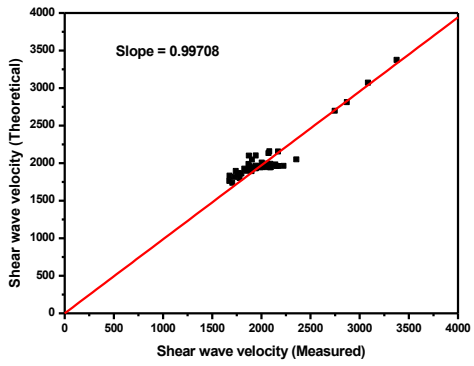

Fig. 4. The relation between the experimentally determined shear ultrasonic wave velocities and the theoretically determined shear ultrasonic wave velocities for tellurite glasses.

4. Conclusions

The author's solution was found to be applicable for characterizing amorphous glass materials using the complete theoretical model, which now give a good chance to simulate the structures of glass materials before experimental processing.

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