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Full Length Research Paper

Compositional and acoustical properties of binary and multi-component oxide glasses

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Correlation between acoustical and compositional properties of TiO_2-Na_2O-CaO-P_2O_5, ZnO-PbO-B_2O_3, Bi_2O_3-V_2O_5-TeO_2, PbO-V_2O_5-TeO_2 and BaO-TeO_2 glass systems has been demonstrated to predict changes in ultrasonic attenuation coefficient and bulk modulus of these glasses. The correlation is based on the recently presented semi-empirical formula of Abd El-Moneim, which correlate the ultrasonic attenuation coefficient at room temperature with packing density, dissociation energy per unit volume and first-order stretching force constant of the glass. In addition, the theoretical values of bulk modulus were evaluated, using Makishima and Mackenzie theory, and compared with the observed values. It has been found that Abd El-Moneim semi-empirical formula as well as Makishima and Mackenzie theory can be applied successfully to predict changes in both ultrasonic attenuation coefficient and bulk modulus if the effect of the basic structural units that are present in the glass network is taken into account.

Key words: Glass, mechanical property, elastic modulus, microstructure, ultrasonic attenuation.

INTRODUCTION

In recent years, the ultrasonic non-destructive pulse-echo technique has been found to be one of the best tools used for evaluating the acoustical parameters of glasses, such as elastic moduli, micro-hardness, Poisson's ratio, ultrasonic attenuation coefficient and Debye temperature. These parameters are very informative about the microstructure as well as the behavior of the network formers and modifiers in the glass. In literature, there are a considerable number of studies dealing with the compositional dependence of acoustical properties in glasses using ultrasonic non-destructive pulse-echo technique (Abd El-Moneim and Alenezy, 2013; Sidkey et al., 2008, 1999; Abd El-Moneim et al., 2006, 2003, 1998; El-Mallawany et al., 2006, 2000, 1998). The agreement between experimentally measured and theoretically calculated values of elastic moduli and Poisson's ratio was studied by many authors on the basis of Makishima and Mackenzie (1973, 1975) theory as well as Rocherulle et al. (1989) model (Abd El-Moneim and Alenezy, 2013; Abd El-Moneim et al., 2003; Abd El -Moneim 2009, 1996; Rocherulle et al., 1989; Makishima and Mackenzie, 1973, 1975).

Correlation between ultrasonic attenuation coefficient and structural parameters of TiO_2-doped CaO-Al_2O_3-B_2O_3 glasses has been studied by Abd El-Moneim (2006). A model consisting of two semi-empirical formulas, which relate the measured ultrasonic attenuation coefficient at room temperature to oxygen density, first-order stretching force constant, atomic ring size, driving ultrasonic frequency and experimental bulk modulus was presented. The presented model appears to be valid for Ag_2O-V_2O_5-TeO_2 and RO-Al_2O_3-B_2O_3 (R = Mg, Ca or Sr) glass systems (Abd El-Moneim, 2007, 2010). More recently, Abd El-Moneim (2012) reported a new semi-empirical formula, which correlate the ultrasonic attenuation coefficient with packing density, dissociation energy per unit volume and first-order stretching force constant of the glass. The validity of this formula has been demonstrated for TiO_2- doped CaO-Al_2O_3-B_2O_3 and Ag_2O-V_2O_5-TeO_2 glass systems (Abd El-Moneim,

2012).

It is very important to generalize the above mentioned formulas (Abd El-Moneim 2006, 2012) by testifying their applicability for all types of the famous glasses, that is, phosphate, borate, gemanate, tellurite and silicate glasses. To the best of this study's knowledge, validity of these formulas has not been testified for phosphate and silicate glasses so far. Therefore, the goal of the present work is to complete the previous work on these formulas (Abd El-Moneim, 2007, 2006, 2011, 2012) by testifying their validity for TiO_2-Na_2O-CaO-P_2O_5,BaO-TeO_2, Bi_2O_3-V_2O_5-TeO_2, PbO-V_2O_5-TeO_2 and ZnO-PbO-B_2O_3 glass systems. Also, the theoretical values of bulk modulus will be evaluated on the basis of Makishima and Mackenzie (1973, 1975) theory and compared with the observed values.

THEORY

The estimation of elastic moduli based on glass composition is very useful for the development of glass materials. The most widely used model is the one proposed by Makishima and Mackenzie (1973, 1975), which expressed bulk modulus, K_{th} , as a function of packing density, V_t , and dissociation energy per unit volume, G_t , of the glass as follows:

$$K_{th} = 10 V_t^2 G_t \quad (1)$$

$$V_t = \frac{\rho}{M} \sum_i x_i V_i \quad (2)$$

$$G_t = \sum_i G_i x_i \quad (3)$$

where x_i , G_i and V_i are the mole fraction, dissociation energy per unit volume and packing factor of the oxide component i , respectively, whereas ρ is the density of the glass and M is its molecular weight.

Recently, Abd El-Moneim (2006) derived the following semi-empirical formula, which correlate the ultrasonic attenuation coefficient, α , with both the first-order stretching force constant, F , and experimental bulk modulus, K_e , of the glass at room temperature:

$$\alpha = \chi \frac{1}{F} \left[\frac{F}{K_e} \right]^{m/n} \quad (4)$$

where χ is a frequency dependent constant, whereas m and n are two positive powers. The average first-order stretching force constant of a multi-component glass can be obtained from an equation reported by Higazy and Bridge (1986) as:

$$F = \frac{\sum_i x_i n_i f_i}{\sum_i x_i n_i} \quad (5)$$

where n_i is the number of network bonds per cation (coordination number of cation) in oxide component i with cation-anion bond length r_i and first-order stretching force constant $f_i = \frac{17}{r_i^3}$. More recently, Abd El-Moneim (2012) eliminated bulk modulus from Equations (1) and (4) and reported the following semi-empirical formula, which correlate the room temperature ultrasonic attenuation coefficient, α , to dissociation energy per unit volume, first-order stretching force constant and packing density of the glass:

$$\alpha = \chi \frac{1}{F} \left[\frac{F^2}{10 G_t V_t} \right]^{m/n} \quad (6)$$

The above assumptions suggest that the first-order stretching force constant, packing density and dissociation energy per unit volume can be used to predict changes of ultrasonic attenuation coefficient in glasses at room temperature.

ANALYSIS AND DISCUSSION

Experimental values of density beside those of bulk modulus and ultrasonic attenuation coefficient, which have been measured at room temperature and at 5 MHz frequency, are summarized in Tables 1 to 5 for the investigated BaO-TeO_2, Bi_2O_3-V_2O_5-TeO_2, PbO-V_2O_5-TeO_2, ZnO-PbO-B_2O_3 and TiO_2-Na_2O-CaO-P_2O_5 glass systems, respectively. All the experimental data were taken from elsewhere (Begum and Rajendran, 2007; Rajendran et al., 2003, 2007; Palanivelu and Rajendran, 2006; Sharma et al., 2005). It is seen from the tables that all the parameters are strongly dependent upon the type and concentration of oxides that constitute the glass network. The variation of density with composition in each glass system is well understandable from the variation in molecular weights of the added modifiers, which are larger in the order: Bi_2O_3>PbO> V_2O_5>TeO_2 >BaO> P_2O_5 >ZnO> TiO_2 > B_2O_3 > Na_2O >CaO.

The values of packing density, dissociation energy per unit volume and theoretical bulk modulus, which have been calculated according to Makishima and Mackenzie theory (Makishima and Mackenzie, 1973, 1975), are listed in Tables 1 to 5 for the same studied glasses. The Applied values of V_i and G_i for Bi_2O_3, PbO, V_2O_5,

Table 1. Physical, acoustical and compositional properties of xBaO-(1-x) TeO_2 (wt%) glass system (x = 9, 12, 15, 18, 20 and 22 wt%).

Composition ($mol\ \%$) BaO – TeO_2	ρ (g/cm^3)	α (dB/cm)	K_e (GPa)	V_t	G_t ($Kcal/cm^3$)	K_{th} (GPa)
9.3 – 90.7	5.646	0.45	32.86	0.51673	12.600	33.64
12.4 – 87.6	5.645	0.46	32.98	0.51552	12.490	33.19
15.5 – 84.5	5.613	0.46	32.85	0.51139	12.400	32.43
18.5 – 81.5	5.587	0.49	32.88	0.50790	12.382	31.94
20.6 – 79.4	5.584	0.54	32.85	0.50690	12.205	31.36
22.7 – 77.3	5.583	0.61	32.68	0.50606	12.132	31.07

Data of ρ, α and K_e are those reported by Begun and Rajendran (2007)

Table 2. Physical, acoustical and compositional properties of xBi_2O_3-(50-x)V_2O_5-$50TeO_2$ glass system (x = 0, 5, 10, 15, 20 and 25 wt%).

Composition ($mol\ \%$) Bi_2O-V_2O_5-TeO_2	ρ (g/cm^3)	α (dB/cm)	K_e (GPa)	V_t	G_t ($Kcal/cm^3$)	K_{th} (GPa)
0 - 47 - 53	3.9956	0.973	30.03	0.5761	14.664	48.67
2 - 43 - 55	4.3764	1.187	31.83	0.5970	14.408	51.35
4 - 40 - 56	4.7969	1.032	33.61	0.6240	14.190	55.25
6 - 36 - 58	5.1884	0.929	34.65	0.6390	13.934	56.90
8 - 32 - 60	5.6240	0.800	35.87	0.6559	13.678	58.84
11 - 27 - 62	6.0314	1.030	36.75	0.6537	13.332	56.97

Data of ρ, α and Ke are those reported by Rajendran et al. (2003)

Table 3. Physical, acoustical and compositional properties of xPbO-(50-x)V_2O_5-$50TeO_2$ glass system (x = 0, 5, 10, 15, 20 and 25 wt%).

Composition ($mol\ \%$) PbO-V_2O_5-TeO_2	ρ (g/cm^3)	α (dB/cm)	K_e (GPa)	V_t	G_t ($Kcal/cm^3$)	K_{th} (GPa)
0 – 47– 53	3.996	0.97	30.30	0.57620	14.664	48.69
4 – 42 – 54	4.240	0.99	31.30	0.57745	14.203	47.36
8 – 38 – 54	4.510	1.02	32.40	0.58354	13.780	46.92
12 – 34 – 54	4.807	1.08	33.80	0.58982	13.357	46.47
16 – 29 – 55	5.133	1.14	34.20	0.59077	12.897	45.01
20 – 24 – 56	5.383	0.72	33.70	0.57910	12.436	41.70

Data of ρ, α and Ke are those reported by Palanivelu and Rajendran (2006)

TeO_2, BaO, P_2O_5, ZnO, TiO_2, B_2O_3, Na_2O and CaO are those reported by Inaba et al. (1999). In the case of ZnO-PbO-B_2O_3 glasses, all B atoms are considered to be three-coordinated with oxygen.

Agreement between theoretical and experimental bulk modulus

Figure 1 shows the relation between theoretical and experimental values of bulk modulus for all the studied glass systems. The straight line in the figure is the 1:1 correlation line (slope equal one). It can be seen clearly from this figure that the agreement between theoretical and experimental bulk modulus of BaO-TeO_2 and TiO_2-Na_2O-CaO-P_2O_5 glasses was excellent. The correlation ratio ranges between 95 and 99.4% in BaO-TeO_2 glasses and between 76 and 95% in TiO_2-Na_2O-CaO-P_2O_5 glasses. In the case of PbO-V_2O_5-TeO_2, Bi_2O_3-V_2O_5-TeO_2.

Table 4. Physical, acoustical and compositional properties of xZnO-2xPbO-(1-3x)B_2O_3 (mol%) glass system.

x (mol %)	ρ (g/cm^3)	α (dB/cm)	V_t	G_t (BO3) (Kcal/cm^3)	K_{th} (GPa)	K_e (GPa)	C_t	G_t (BO4) Kcal/cm^3)	K^*_{th} (GPa)
10	3.745	1.13	0.65262	5.015	21.36	51.59	0.66886	15.424	69.00
13.3	4.185	1.31	0.62245	5.439	21.07	57.62	0.64704	14.376	60.19
20	5.286	1.38	0.57772	6.300	21.027	54.76	0.60274	12.248	44.50
22	5.487	1.06	0.54791	6.557	19.86	56.51	0.58952	11.613	40.36
23	5.743	0.96	0.54818	6.686	20.09	49.87	0.58290	11.295	38.38
25	5.869	1.21	0.51185	6.943	18.19	56.07	0.56968	10.66	34.60
26	5.985	1.28	0.49884	7.071	17.60	47.45	0.56307	10.342	32.79

Data of ρ, α and Ke are those reported by Sharma et al. (2005)

Table 5. Physical, acoustical and compositional properties of xTiO_2-(31-x)Na_2O-24CaO-45P_2O_5 glass system (x = 0, 0.5, 1.0, 1.5, 2.0 and 2.5 wt%).

Composition (mol %) TiO_2-Na_2O-CaO-P_2O_5	ρ (g/cm^3)	α (dB/cm)	K_e (GPa)	V_t	G_t (Kcal/cm^3)	K_{th} (GPa)
0.0-40.2-34.4-25.4	2.608	0.57	38.36	0.5387	10.06	29.19
0.5-40.0-34.2-25.3	2.606	0.58	38.28	0.5379	10.12	29.28
1.0-39.0-34.5-25.5	2.614	0.56	38.39	0.5400	10.23	29.83
13.3-33.8-30.4-22.5	2.617	0.45	38.30	0.5333	11.98	34.07
17.1-32.0-29.2-21.7	2.624	0.53	38.43	0.5328	12.522	35.55
20.6-30.3-28.2-20.9	2.631	0.51	38.81	0.5321	13.03	36.89

Data of ρ, α and Ke are those reported by Rajendran et al., (2007)

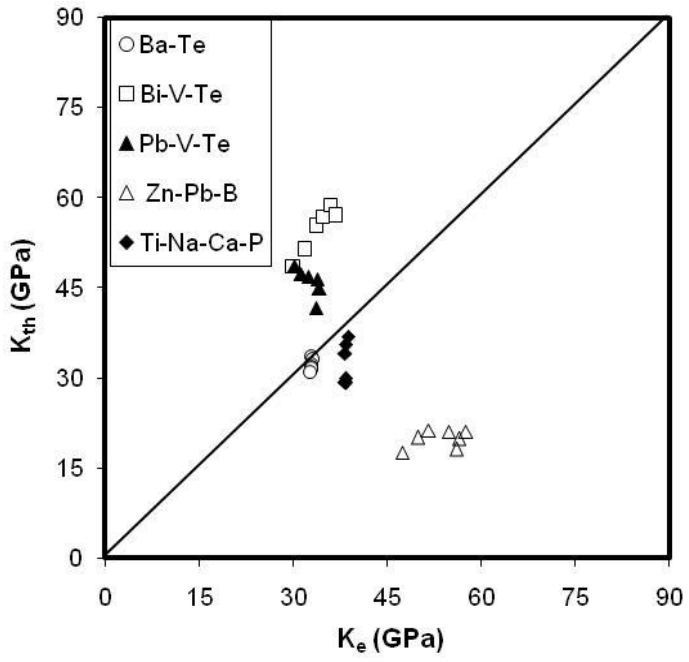

Figure 1. Relationship between theoretically calculated and experimentally measured bulk modulus. The solid line is the line of 1:1 correlation.

and ZnO-PbO-B_2O_3 glass systems, the agreement between theoretical and experimental values of bulk modulus was not satisfactory. The correlation ratio ranged between 62.2 and 81% in PbO-V_2O_5-TeO_2 glasses, between 61 and 65% in Bi_2O_3-V_2O_5-TeO_2 glasses and between 34 and 44.4% in ZnO-PbO-B_2O_3 glasses.

It is well known that the basic structural units of pure P_2O_5, B_2O_3 and TeO_2 glasses are PO_4 tetrahedra with P=O bonds, BO_3 triangles and TeO_4 trigonal bipyramids (tbps), respectively (Bridge et al., 1983). The basic structural units are connected to each other through P-O-P linkages in pure P_2O_5 glass, B-O-B linkages in pure B_2O_3 glass and Te-O-Te linkages in pure TeO_2 glass. The addition of BaO to pure TeO_2 glass changes the Te coordination polyhedron from TeO_4 trigonal bipyramids to TeO_3 trigonal pyramid (tp) by breaking the Te-O-Te linkages and creation of non-bridging oxygens (NBOs) according to the following reaction:

$$BaO + Te - \underset{ax}{O} \underset{eq}{} - Te \rightarrow Te - \underset{ax}{O^-}\ Ba^{2+}\ {}^-\underset{eq}{O} - Te$$

(Begum and Rajendran, 2007). In the case of xZnO-xPbO- (1-3x)B_2O_3 glasses, PbO enters the glass network as a modifier by the transformation of BO_3 units into BO_4 groups (two BO_4 groups for each added oxygen atom)

(Sharma et al., 2005). The extra negative charges on the [BO_4] groups are satisfied by the heavy Pb^{2+} ions in the vicinity. The dissociation energy per unit volume of BO_4 groups is much greater than that of BO_3 units (Makishima and Mackenzie, 1973, 1975). It can be seen clearly from Figure 2 that all ZnO-PbO-B_2O_3 glass samples have values of theoretical bulk modulus much smaller than the experimentally measured ones. Thus, divergence between the theoretical and experimental values of bulk modulus can be attributed to ignoring the concentration of BO_4 groups in the network of these glasses. Formation of BO_4 groups instead of BO_3 units is expected to increase the dissociation energy per unit volume and packing density of the glass, which consequently increases the theoretical bulk modulus.

Rocherulle et al. (1989) studied the agreement

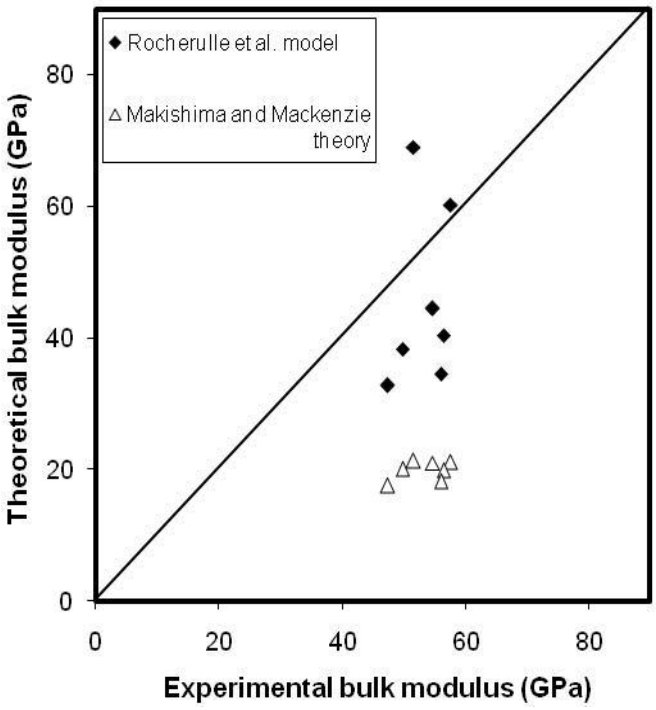

Figure 2. Relationship between theoretical and experimental bulk modulus of ZnO-PbO-B_2O_3 glasses. The solid line is the line of 1:1 correlation.

between the theoretically calculated and experimentally measured values of elastic moduli and Poisson's ratio of glasses. The authors (Rocherulle et al., 1989) made modification on the equation of packing density in order to improve the agreement between the theoretical and experimental values. They reported the following relations for calculating both the packing density and bulk modulus of multi-component glasses (Rocherulle et al., 1989):

$$C_t = \sum_i \frac{\rho_i}{M_i} x_i V_i \quad (7)$$

$$K_{th}^{*} = 10 C_t G_t \quad (8)$$

where ρ_i and M_i are the density and molecular weight of the oxide component i, respectively. In the light of the above discussion and considering all Batoms are four-coordinated with oxygen, the new values of C_t, G_t and K_{th}^{*} for the present ZnO-PbO-B_2O_3 glass samples can be calculated on the basis of Rocherulle et al. (1989) model. These values are given also in Table 4. The applied G_i values are those reported previously by Makishima and Mackenzie, 1973, 1975. Figure 2 shows that the values of K_{th}^{*} agree much better with the observed values when compared with those calculated on the basis of Makishima and Mackenzie (1973, 1975) theory. The correlation ratio between the observed and K_{th}^{*} values ranges between 96% in 25ZnO-50PbO-25B_2O_3 glass sample and 62% in 13.3 ZnO-26.6PbO-60.1B_2O_3 glass sample.

In the case of V_2O_5-containing tellurite glasses (Bi_2O_3-V_2O_5-TeO_2 and PbO-V_2O_5-TeO_2 glasses), the theoretical values of bulk modulus are much greater than the corresponding experimental ones. IR and ultrasonic studies indicated that the substitution of V_2O_5 by Bi_2O_3 in Bi_2O_3-V_2O_5-TeO_2 glasses or PbO in PbO-V_2O_5-TeO_2 glasses results in the conversion of TeO_4 trigonal bipyramid into TeO_3 trigonal pyramid by breaking of V-O-Te linkages and creation of non-bridging oxygens (NBOs) (Rajendran et al., 2003; Palanivelu and Rajendran, 2006). Thus, weakening of the glass structure and reduction in the rigidity of the network takes place. The disagreement between the theoretical and experimental bulk modulus of these glasses may be due to the following reasons:

1. Lack of information about the concentration of NBOs. The creation of NBOs in the network is expected to reduce the packing density and dissociation energy, which consequently decreases the theoretical bulk modulus.

2. In the calculations, we have ignored the presence of some structural units, such as TeO_3 tps and VO_5 groups with V=O bonds, which exist as a part of a building block of the glass structure. This is because the concentration, packing density and dissociation energy of these groups are unknown so far. For TeO_2 oxide, packing density of 14.7 cm^3/ mol and dissociation energy of 54 kJ/ cm^3 was used for TeO_4 trigonal bipyramids, which has been determined empirically from Young's modulus of a single-component TeO_2 glass (Inaba et al., 1999). Also, the applied values of packing factor and dissociation energy

of V_2O_5 oxide are taken as that of four-fold coordinated VO_4 groups.

Correlation between ultrasonic attenuation coefficient and compositional parameters

It is an important to demonstrate the applicability of the semi-empirical formula (6) for the present BaO-TeO_2, Bi_2O_3-V_2O_5-TeO_2, PbO-V_2O_5-TeO_2 ZnO-PbO-B_2O_3 and TiO_2-Na_2O-CaO-P_2O_5 glass systems. This formula suggests that the ultrasonic attenuation coefficient of a glassy material depend on three major parameters, which are the packing density, dissociation energy per unit volume and first order stretching force constant of the glass. A plot of $F\alpha$ versus $F/10G_tV_t^{2}$ yields a proportional relation. Figure 3 illustrates the relationship of $F\alpha$ versus $F/10G_tV_t^{2}$ for all the studied glass

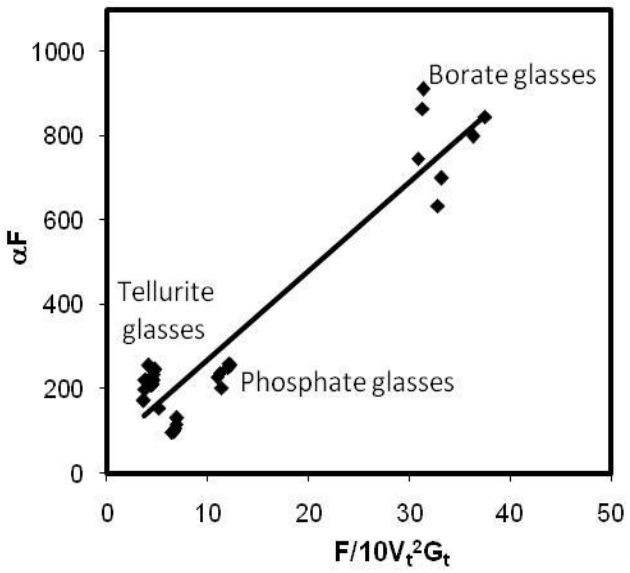

Figure 3. Relationship between $F\alpha$ and $F/10G_tV_t^{2}$ in BaO-TeO_2, Bi_2O_3-V_2O_5-TeO_2, PbO-V_2O_5-TeO_2, ZnO-PbO-B_2O_3 and TiO_2-Na_2O-CaO-P_2O_5 glass systems. The solid line is drwawn as a guide to the eye.

systems. The applied values of F are those of B-O-B linkage (660 N/m) in ZnO-PbO-B_2O_3 glasses, Te-O-Te linkage (216 N/m) in BaO-TeO_2, Bi_2O_3-V_2O_5-TeO $_2$ and PbO-V_2O_5-TeO_2 glasses and Ti-O-P linkage (323.5 N/m) in TiO_2-Na_2O-CaO-P_2O $_5$ glasses. The figure shows clearly that the value of $F\alpha$ increases linearly with the increase of the ratio of $F/10G_tV_t^{2}$, which suggests that the semi-empirical formula (6) satisfies the ultrasonic attenuation data of these glasses very well. Figure 4 shows the relationship between $\log(F\alpha)$ and

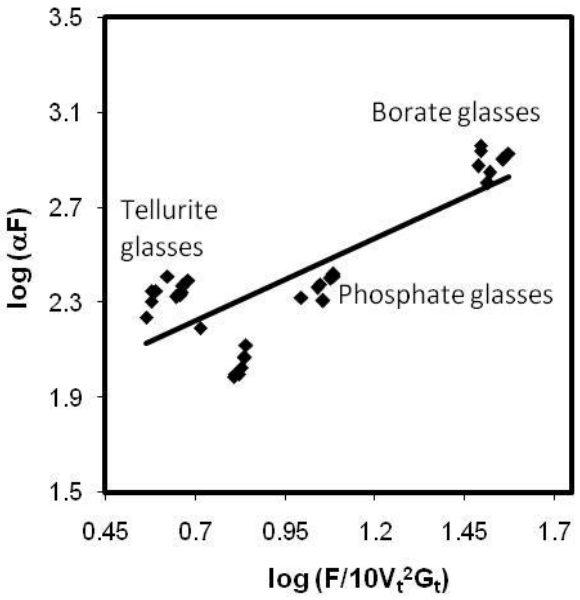

Figure 4. Relationship between $\log(F\alpha)$ and $\log F/10G_tV_t^{2}$ in BaO-TeO_2, Bi_2O_3-V_2O_5-TeO_2, PbO-V_2O_5-TeO_2, ZnO-PbO-B_2O_3 and TiO_2-Na_2O-CaO-P_2O_5 glass systems. The solid line represents the filting of the data.

$\log(F/10G_tV_t^{2})$ in for the same glass systems under investigation. The equation of the fitted curve can be represented by the following relation:

$$\alpha = 54.7\frac{1}{F}\left[\frac{F}{10G_tV_t^{2}}\right]^{0.691} \quad (9)$$

with correlation ratio of 64.6%. This equation suggests that the best fitting of equation (6) to the attenuation data of BaO-TeO_2, Bi_2O_3-V_2O_5-TeO_2, PbO-V_2O_5-TeO_2 ZnO-PbO-B_2O_3 and TiO_2-Na_2O-CaO-P_2O_5 glasses was achievedwhen $(m/n)=0.691$and $\chi_5=54.7$.

Considering the effect of all the structural units, it is first necessary to identify all the types of bonds present in the prepared glass and then to ascribe appropriate values of G_i, V_i, G_t, F and V_t for each type of bonds by using neutron diffraction, Raman EXAFS and FTIR data. Thus, taking into account the effect of the basic structural units that are present in the glass structure as well as the uncertainty inherent in experimental measurements, it is believed that the semi-empirical formula (6) (Abd El-Moneim 2012) as well as Makishima and Mackenzie theory (Makishima and Mackenzie, 1973, 1975) can be applied successfully to predict changes in the acoustical properties of the investigated BaO-TeO_2, Bi_2O_3-V_2O_5-

TeO_2, PbO-V_2O_5-TeO_2, ZnO-PbO-B_2O_3 and TiO_2-Na_2O-CaO-P_2O_5 glasses.

Conclusions

Correlation between acoustical and compositional properties was demonstrated in TiO_2-Na_2O-CaO-P_2O_5, ZnO-PbO-B_2O_3, Bi_2O_3-V_2O_5-TeO_2, PbO-V_2O_5-TeO_2 and BaO-TeO_2 glass systems. The following conclusions were reached:

(i) Packing density and dissociation energy per unit volume of the constituent oxides are powerful tools capable of exploring changes in acoustic properties of these glasses.
(ii) The model of Rocherulle et al. (1989) is valid for the majority of ZnO-PbO-B_2O_3 glasses.
(iii) Abd El-Moneim semi-empirical formulas can be applied successfully to predict changes in the ultrasonic attenuation data if the effect of the basic structural units that are present in the glass structure is taken into account.

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