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Elastic Properties of Clinopyroxene Based Glasses along Diopside ($CaMgSi_2O_6$)-Jadeite ($NaAlSi_2O_6$) Join

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ABSTRACT

The elastic properties of glasses along Diopside ($CaMgSi_2O_6$)-Jadeite ($NaAlSi_2O_6$) join (Di_x - Jd_{1-x} where x=20, 40, 60, 80, 100 mole %), were obtained by the ultrasonic echography technique, at room temperature. The correlation of elastic moduli with the atomic packing density of these glasses was discussed. The derived experimental values of Young's modulus, bulk modulus, shear modulus and Poisson's ratio for investigated glasses were compared with those theoretically calculated values in terms of the Makishima–Mackenzie model and the modified model presented by Rocherulle.

Keywords: Glass, Diopside, Mechanical properties.

1. INTRODUCTION

Glass is characterized as brittle and easily broken, unlike metals and plastics, when subjected to thermal or mechanical stresses. The thermal shock resistance and fracture toughness of glasses are very important properties, because they estimate the resistance to these stresses. These parameters are directly related to Young's modulus, which, in turn, is influenced predominantly by the glass chemical composition thus, the estimation of the Young's modulus based on glass composition is very useful for the development of glass materials [1, 2]. Moreover, the strength of materials increases with their elastic moduli; it is therefore possible to assess strength indirectly from their elastic properties. Studies of the elastic moduli of the glassy materials give considerable information about the structure of non-crystalline solids, since they are directly related to the interatomic forces and potentials [3-11].

It is useful to predict the elastic properties of polycomponent oxide systems solely from knowledge of the system composition, density and well known tabulated physical properties. Makishima and Mackenzie [3, 4] correlated the elastic moduli of oxide glasses to both packing density and the average strength of chemical bonds in the glass. Rocherulle [8] extended the analysis of Makishima and Mackenzie [3, 4] to oxynitride glasses. They introduced a thermodynamic factor, which results from the substitution of oxygen by nitrogen within the vitreous network. Their results showed that the calculated values of elastic moduli are in good agreement with the experimental values.

The purpose of the present work is to calculate theoretically the elastic moduli and Poisson's ratio from the chemical composition and density data on the basis of Makishima and Mackenzie's [3, 4] model, Rocherulle [8] model and to compare with the experimental values of the investigated glasses. Furthermore, a correlation between the predicted and experimental values of elastic moduli and Poisson's ratio is studied to verify the applicability of these models for the studied glass system.

The solid solution between diopside ($CaMgSi_2O_6$; hereafter referred as Di) and jadeite ($NaAlSi_2O_6$; hereafter referred as Jd) is a subject of relevance from petrologic as well as

technological point of view. Abo-Mosallam et al. [12] studied the structure and crystallization behavior of glasses and glass-ceramics by MAS-NMR along $(CaMgSi_2O_6)_{1-x}$ - $(NaAlSi_2O_6)_x$ - $(Ca_5(PO_4)_3F)_y$ where $0 \leq x \leq 30$ mole % and $y = 7$ mole %. He has reported that with increasing Jd content in the glasses, the polymerization in silicate glass network shifts from Q^2 to Q^3 (Q^n: degree of polymerization; n: number of bridging oxygen's) and Al exists predominantly as Al (IV) species. The glass compositions under investigation have been designed along diopside ($CaMgSi_2O_6$)-jadeite ($NaAlSi_2O_6$) join (Di_x - Jd_{1-x} where x =20, 40, 60, 80, 100 mole %) with varying diopside/jadeite molar ratio as shown in the Table 1. The partial substitution of B_2O_3 for SiO_2 has been made in all compositions in accordance with substitution scheme 0.3 $Si^{4+} \leftrightarrow$ 0.4 B^{3+} so as to decrease the melting point of the glass batch.

2. EXPERIMENTAL

High purity chemical powders SiO_2 (Sigma Aldrich, Germany, purity >99.7%), $CaCO_3$ (Merck, Germany >99.8%), Al_2O_3 (Merck, Germany, ≥98%), H_3BO_3 (Merck, Germany, 99.8%), MgO (Merck, Germany >99.7%), Na_2CO_3 (Merck, Germany, 99.9%) were used for glass melting. For each glass composition as shown in Table 1 the batch of 200 grams was taken and thoroughly mixed by using an agate ball mill. The mixed powder was taken in a platinum crucible and heated in the electric furnace to 1500 °C for one hour and one hour dwell time to obtain bubble free and homogeneous glass. Glasses in the bulk form were produced by pouring the melts on the preheated graphite moulds followed by annealing at 550 °C for one hour.

The amorphous nature of glasses was confirmed by powder X-ray diffraction (XRD) (Philips PW 3710). The density of glass was determined by Archimedes' method in which the sample was weight both in air and immersed in liquid. The liquid used in the present study for density measurement was Ethylene Glycol of known density (1.1132 g/cm^3). The accuracy of the measurement was about ±0.002 g/cm^3.

Table 1 Batch composition of glasses

Glass		MgO	CaO	Na_2O	SiO_2	B_2O_3	Al_2O_3
Di-0	wt.%	-	-	15.65	51.58	7.03	25.74
$NaAlSi_{1.7}B_{0.4}O_6$	mole %	-	-	17.27	58.57	6.89	17.27
Di-20	wt.%	4.01	5.58	12.34	50.84	6.93	20.3
$Ca_{0.2}Mg_{0.2}Na_{0.8}Si_{1.7}Al_{0.8}B_{0.4}O_6$	mole %	6.42	6.42	12.92	54.90	6.42	12.92
Di-40	wt.%	7.91	11.01	9.12	50.12	6.83	15.01
$Ca_{0.4}Mg_{0.4}Na_{0.6}Si_{1.7}Al_{0.6}B_{0.4}O_6$	mole %	12.11	12.11	9.09	51.44	6.06	9.09
Di-60	wt.%	11.7	16.28	6.0	49.42	6.74	9.87
$Ca_{0.6}Mg_{0.6}Na_{0.4}Si_{1.7}Al_{0.4}B_{0.4}O_6$	mole %	17.12	17.12	5.73	48.57	5.73	5.73
Di-80	wt.%	15.39	21.41	2.96	48.74	6.65	4.87
$Ca_{0.8}Mg_{0.8}Na_{0.2}Si_{1.7}Al_{0.2}B_{0.4}O_6$	mole %	21.60	21.60	2.72	45.93	5.43	2.72
Di-100	wt.%	18.97	26.40	-	48.08	6.55	-
$CaMgSi_{1.7}B_{0.4}O_6$	mole %	25.65	25.65	-	43.57	5.13	-

The elastic constants such as Young's modulus (E), shear Modulus (G), bulk modulus (K) and Poison's ratio (σ) of glasses were determined by ultrasonic echography at room temperature. For this purpose, velocities of longitudinal (10 MHz) and transverse (4 MHz) ultrasonic waves in the investigated glass specimens were measured using piezoelectric transducers and associated electronics (ultrasonic flaw detector USD15, Krautkramer GmbH & Co., Huerth, Germany). The overall uncertainty in calculated value is estimated to be ±2% due to several influential effects, such as multiple internal reflections within the transducer, sample thickness and the acoustic impedance mismatch between glass sample and transducer.

3. THEORY

Makishima and Mackenzie [3, 4] presented a theoretical model to calculate the elastic modulus of oxide glasses in terms of chemical composition, packing density and dissociation energy of the oxide constituents. They derived the following relations:

Young's Modulus,

$$E_{cal} = 83.6\, V_t \sum_i G_i\, x_i \qquad (1)$$

Bulk Modulus,

$$K_{cal} = 100\, V_t^2 \sum_i G_i x_i \qquad (2)$$

Shear Modulus,

$$G_{cal} = \left[\frac{300\, V_t^2}{10.2\, V_t - 1}\right] \sum_i G_i x_i \qquad (3)$$

Poisson's Ratio

$$\sigma_{cal} = 0.5 \mid \frac{1}{7.2 V_t} \qquad (4)$$

where V_t is the packing density of the glass sample which is calculated by using the equation:

$$V_t = \frac{\rho}{M} \sum_i V_i\, X_i \qquad (5)$$

where M is effective molecular weight, ρ is the density, X_i is the molar fraction of component i and V_i is a packing factor obtained from the following equation for an oxide A_XO_Y:

$$V_i = 6.023 \times 10^{23}\, \frac{4}{3}\pi\left(XR_A^3 + YR_O^3\right) \qquad (6)$$

where R_A and R_O are the respective ionic radius of metal and oxygen (In the present study, Pauling's ionic radii are used). The dissociation energy per unit volume (G_i), the effective molecular weight (M) and packing density (V_i) of each oxide component present in the Diopside ($CaMgSi_2O_6$)-jadeite ($NaAlSi_2O_6$) are given in Table 2. Rocherulle [8] introduced some

modifications in the expression of the packing factor and expressed it for an oxide A_mO_n as:where ρ_i and M_i are the density and molecular weight of *i*th oxide component respectively. The elastic moduli and Poisson's ratio of a multicomponent glass are given by Rocherulle [8] as:

Young's Modulus,

$$E^*_{cal} = 83.6\, C_t \sum_i G_i\, x_i \qquad (8)$$

Bulk Modulus,

$$K^*_{cal} = 100\, C_t^2 \sum_i G_i x_i \qquad (9)$$

Shear Modulus,

$$S^*_{cal} = \left[\frac{300\, C_t^2}{10.2\, C_t - 1}\right] \sum_i G_i x_i \qquad (10)$$

Poisson's Ratio,

$$\sigma^*_{cal} = 0.5 - \frac{1}{7.2 C_t} \qquad (11)$$

Packing Factor

$$C_t = \sum_i C_i x_i \qquad (12)$$

The values of packing factor (C_i) of various oxides used in Diopside ($CaMgSi_2O_6$)-jadeite ($NaAlSi_2O_6$) system are given in Table 2.

Table 2 Effective molecular weight (*M*), Dissociation energy per unit volume (G_i), Packing density (V_i) and Packing factor (C_i)

	CaO	MgO	Na_2O	SiO_2	B_2O_3	Al_2O_3
M (g/mol)	56.077	40.304	61.979	60.084	69.62	101.961
G_i (Kcal/cm^3)	15.50	20.00	8.90	15.40	18.60	32.00
V_i (cm^3)	9.4	7.6	11.2	14.0	20.8	21.4

C_i	0.5530	0.6750	0.4102	0.6174	0.7619	0.8333

4. RESULTS

The investigated glass compositions were readily castable after 1 h of soaking time at 1500 ºC resulting in homogeneous and transparent glass. The amorphous natures of the quenched glasses were confirmed by XRD analysis as shown in Fig. 1. On the contrary Di-100 crystallized immediately after pouring the melt on the graphite mould as expected (due to direct contact to air and it prone to cracking during annealing so no experiment was done on this sample) but further increase in Jd/Di ratio in glasses led to the formation of stable, transparent and monolithic bulk glasses.

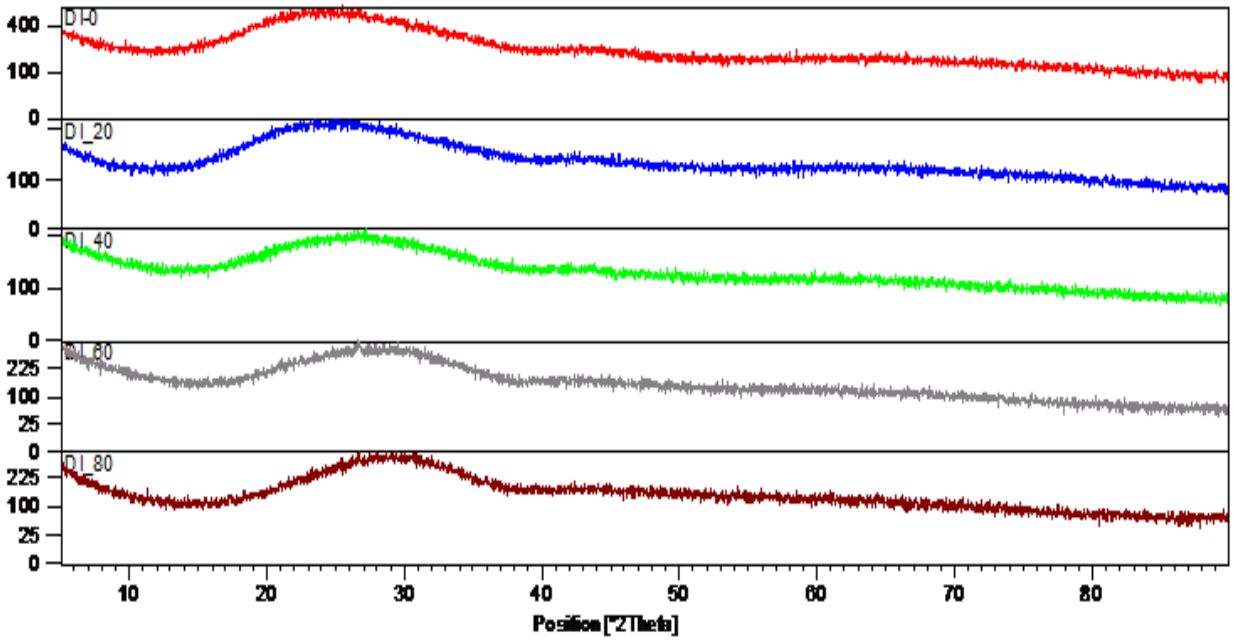


Figure 1:- X-ray diffractograms of glass-powder

In an amorphous solid such as glass, the elastic strain produced by a small stress can be described by two independent elastic constants, C_{11} and C_{44} [13]. The Cauchy relation $2C_{44} = C_{11} - C_{12}$ allows to determine C_{12}, and for pure longitudinal waves $C_{11} = \rho V_l^2$, and for pure transverse waves $C_{44} = \rho V_t^2$ where V_l and V_t are the longitudinal and transverse velocities,

respectively. The sound velocities also allow the determination of Young's modulus (E), bulk modulus (K), shear modulus (S) and Poisson's ratio (σ) by the following equations:

$$K = \rho\left(V_l^2 - \frac{4}{3}V_t^2\right) \qquad (14)$$

$$\sigma = \frac{(V_l^2 - V_t^2)}{2(V_l^2 + V_t^2)} \qquad (16)$$

The Density, longitudinal velocity (V_l) and transverse (V_t) sound velocities of diopside ($CaMgSi_2O_6$) – jadeite ($NaAlSi_2O_6$) glasses are given in Table 3.

Table 3 Experimental values of Density (ρ), Longitudinal (V_l) and Transverse velocity (V_t)

Glasses	Density (g/cm^3)	Longitudinal Velocity V_l (m/s)	Transverse Velocity V_t (m/s)
Di-0	2.462	5776.580	3519.403
Di-20	2.510	5933.704	3488.286
Di-40	2.607	6205.345	3599.386
Di-60	2.655	6277.778	3604.466
Di-80	2.766	6560.816	3730.021

Table 4 gives the calculated elastic constants (C_{11}, C_{44} and C_{12}), Young's Modulus (E), Bulk Modulus (K) and Poisson ratio (σ) from experimental sound velocities for diopside ($CaMgSi_2O_6$)-jadeite ($NaAlSi_2O_6$) glasses. The overall uncertainty for above calculated value is estimated to be ±2%.

The expression of $d = 4C_{44}/K$, which was derived by Bergman and Kantor [14] for an inhomogeneous random mixture of fluid and a solid backbone near the percolation limit, gives

an interesting information on effective dimensionality of the materials [15, 16]. Bogue and Sladek [15] called this new parameter ''*d*'' the fractal bond connectivity, where *d* = 3 for three-dimensional tetrahedral coordination polyhedra, *d* = 2 for two-dimensional layer structures, and d = 1 for one dimensional chains, respectively. The calculated d-values for investigated glasses are given in Table 4.

Table 4 Experimental calculated values of elastic constants (C_{11}, C_{44} and C_{12}), Young's Modulus (*E*), Bulk Modulus (*K*), Poisson ratio (σ) and fractal bond connectivity (*d*) of investigated glasses

Glass	C_{11} (GPa)	C_{44} (GPa)	C_{12} (GPa)	*E* (GPa)	*K* (GPa)	σ	$d = 4C_{44}/K$
Di-0	82.118	30.470	21.178	73.423	41.461	0.229	2.940
Di-20	88.223	30.493	27.247	75.376	47.576	0.243	2.564
Di-40	100.309	33.749	32.811	84.135	55.310	0.248	2.441
Di-60	104.595	34.480	35.635	86.486	58.620	0.252	2.353
Di-80	119.941	38.623	42.696	97.422	67.994	0.256	2.272

5. DISCUSSION

The calculated values of longitudinal and transverse elastic constants (C_{11}, C_{44}), Young's modulus (*E*) and bulk modulus (*K*) decreases as the mole % of Jd increases as shown in the Table 4. This implies that glass containing more percentage of Di have a rigid structure in the investigated glasses. The values of Young's modulus are increasing as mole % of Di increases because Di contains Mg^{+2} and Ca^{+2} ions so as we the increase Di, the Mg^{+2} and Ca^{+2} ions increases and the substitution of low-valency ions by high-valency ions enhance elastic moduli because the internal energy is proportional to the effective charge of cations and anions. Thus, the glasses containing an alkaline earth show high elastic modulus [17].

Both interatomic energies (U_0) and atomic packing densities have been taken into account to interpret the elasticity data. Here the substitution of Na^+ and Al^{3+} by Ca^{2+} and Mg^{2+} as the mole % of Di increases and $U_{0\ Mg}$ (7.646), $U_{0\ Ca}$ (6.113) is greater than $U_{0\ Na}$ (5.139), $U_{0\ Al}$ (5.986) so modulus of elasticity increases [18]. Simultaneously the atomic packing density decreases with increase of Jd as shown in Table 5 so the elastic modulus decreases [18].

Poisson's Ratio decreases as Jd increases because as we substitutes the Na^+ and Al^{3+} by Ca^{2+} and Mg^{2+} as the mole % of Di increases so the amount of alkali content decreases and packing density is also increasing as shown in Table 5 so Poisson's ratio increases [18]. In that glasses as the amount of aluminum increases Al coordination changes from 6 (small Al quantities) to 4 (Al is network forming). As the network former Al decreases the atomic packing density, whereas in comparison packing density is enhanced by sixfold network modifying Al atoms. Consequently, Poisson's ratio exhibits a slight increase at low Al contents, However when we increases the mole % of Jd the network connectivity is increased [12] so the poison ratio decrease, because the higher connectivity of network decrease the Poisson's ratio.

The fractal bond connectivity data, which shows the *d*-value of these, glasses around 2.1 to 2.7 as shown in Table 3 implies that as we increase the Di the connectivity of the structure decreases. Similar results have also been reported on diopside-jadeite-fluorapatite glasses by Abo-Mosallam et al. [12].

Table 5 Theoretical calculated Packing density (V_t), Young's modulus (E_{cal}), Shear Modulus (S_{cal}), Bulk Modulus (K_{cal}) and Poisson's ratio (σ_{cal}) of investigated glasses from the model of Makishima and Mackenzie [3, 4]

Glass	V_t	E_{cal} (GPa)	S_{cal} (GPa)	K_{cal} (GPa)	σ_{cal}
Di-0	0.523	75.925	32.874	47.498	0.427
Di-20	0.525	75.543	32.680	47.441	0.427
Di-40	0.539	76.900	33.070	49.580	0.425

Di-60	0.547	77.589	33.258	50.767	0.424
Di-80	0.569	80.210	34.093	54.593	0.411

For the studied glasses, Table 5 gives the theoretically calculated packing density (V_t), the elastic moduli (E_{cal}, S_{cal} and B_{cal}) and the Poisson's ratio based on the model of Makishima and Mackenzie. The theoretical values for the packing density (C_t), Young's modulus, shear modulus, bulk modulus, and Poisson ratio based on the Rocherulle model for the studied glass samples are given in Table 6.

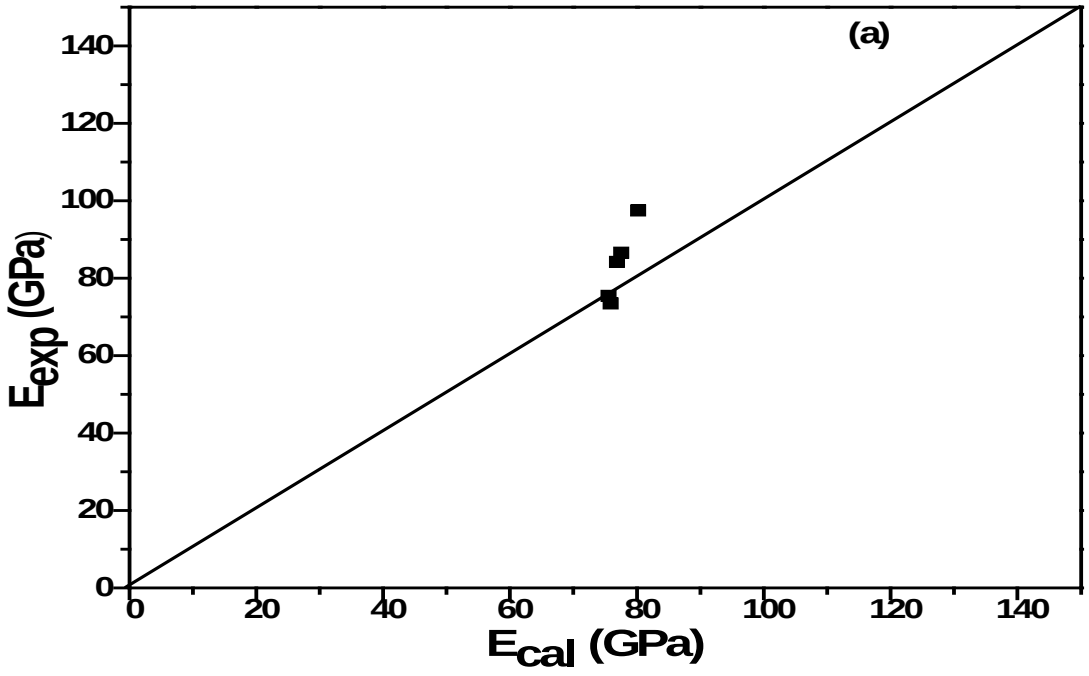
Table 6 Theoretical calculated Packing factor (C_t), Young's modulus (E_{cal}*), Shear Modulus (S_{cal}*), Bulk Modulus (K_{cal}*) and Poisson's ratio (σ_{cal}*) of investigated glasses from the model of Rocherulle [8]

Glass	C_t	E_{cal}* (GPa)	S_{cal}* (GPa)	K_{cal}* (GPa)	σ_{cal}*
Di-0	0.62886	91.292	38.050	68.672	0.41266
Di-20	0.62608	90.088	37.579	67.476	0.41304
Di-40	0.62308	88.896	37.115	66.255	0.41346
Di-60	0.62159	88.169	37.256	65.556	0.41367
Di-80	0.61962	87.346	36.506	64.738	0.41394

The correlation between the experimental values of Young's modulus and those calculated from the theory of Makishima and Mackenzie [3, 4] is shown in Fig. 2 (a).

This figure shows that the calculated values are less than the observed values and the correlation is not satisfactory and Fig. 2 (b) shows the correlation between the observed and theoretically calculated values of Young's modulus on the basis of the Rocherulle [8] model. This figure clearly shows that this model is also not good in predicting most of the observed values of Young's modulus. The calculated values are more than that of experimental values while same

behavior is observed with the bulk modulus, experimental values are not in satisfactory agreement with the calculated values from both of the model as shown in Fig. 3 (a) and (b).



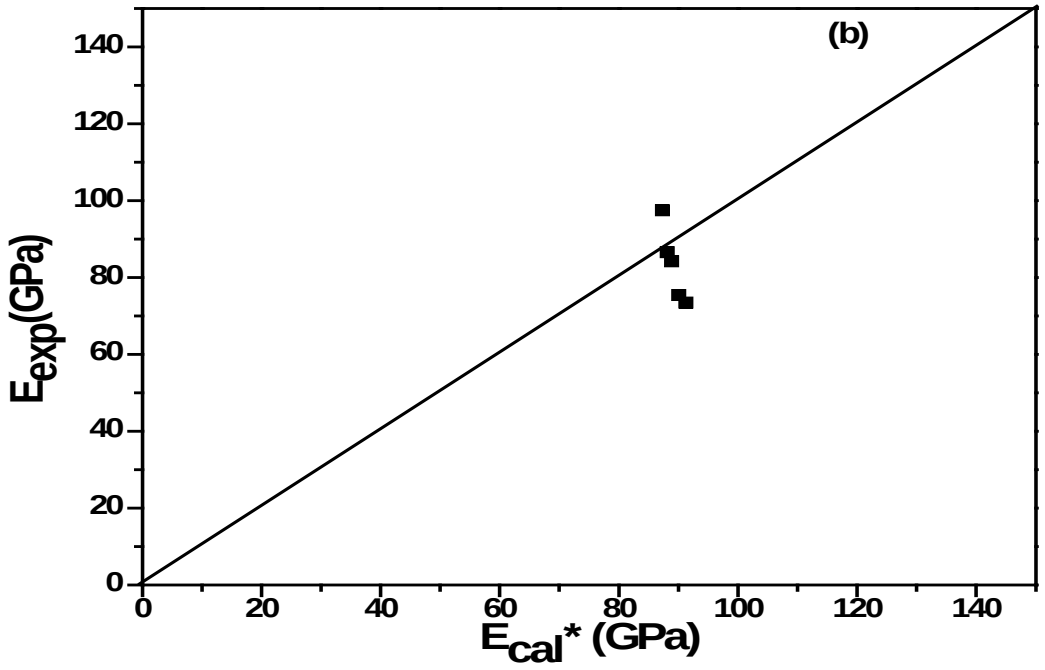


Figure 2:- Agreement between observed and theoretical calculated values of Young's modulus in the present study according to (a) Makishima and Mackenzie's [3, 4] model; (b) Rocherulle [8] model

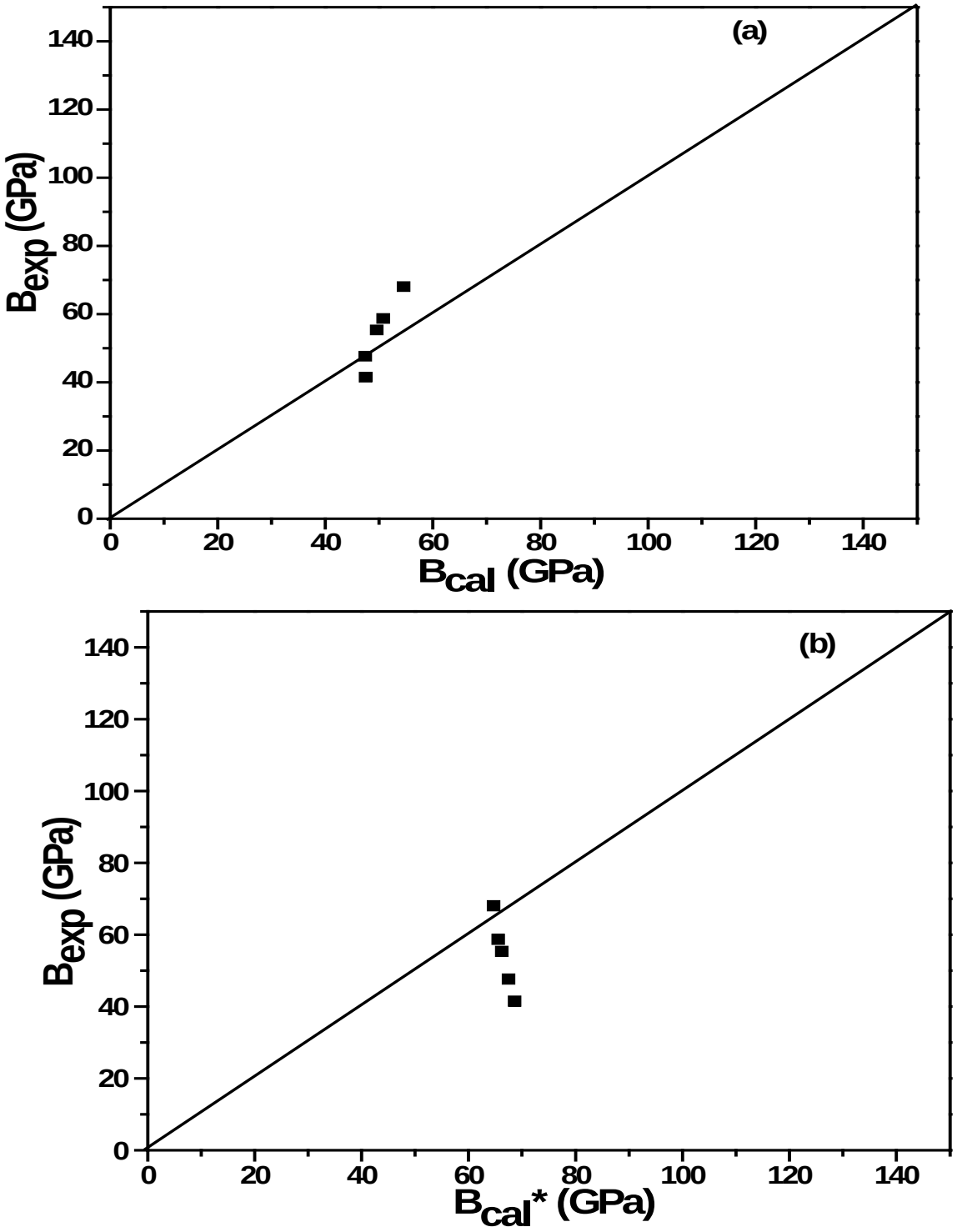


Figure 3:- Agreement between observed and theoretical calculated values of Bulk modulus in the present study according to (a) Makishima and Mackenzie's [3, 4] model; (b) Rocherulle [8] model

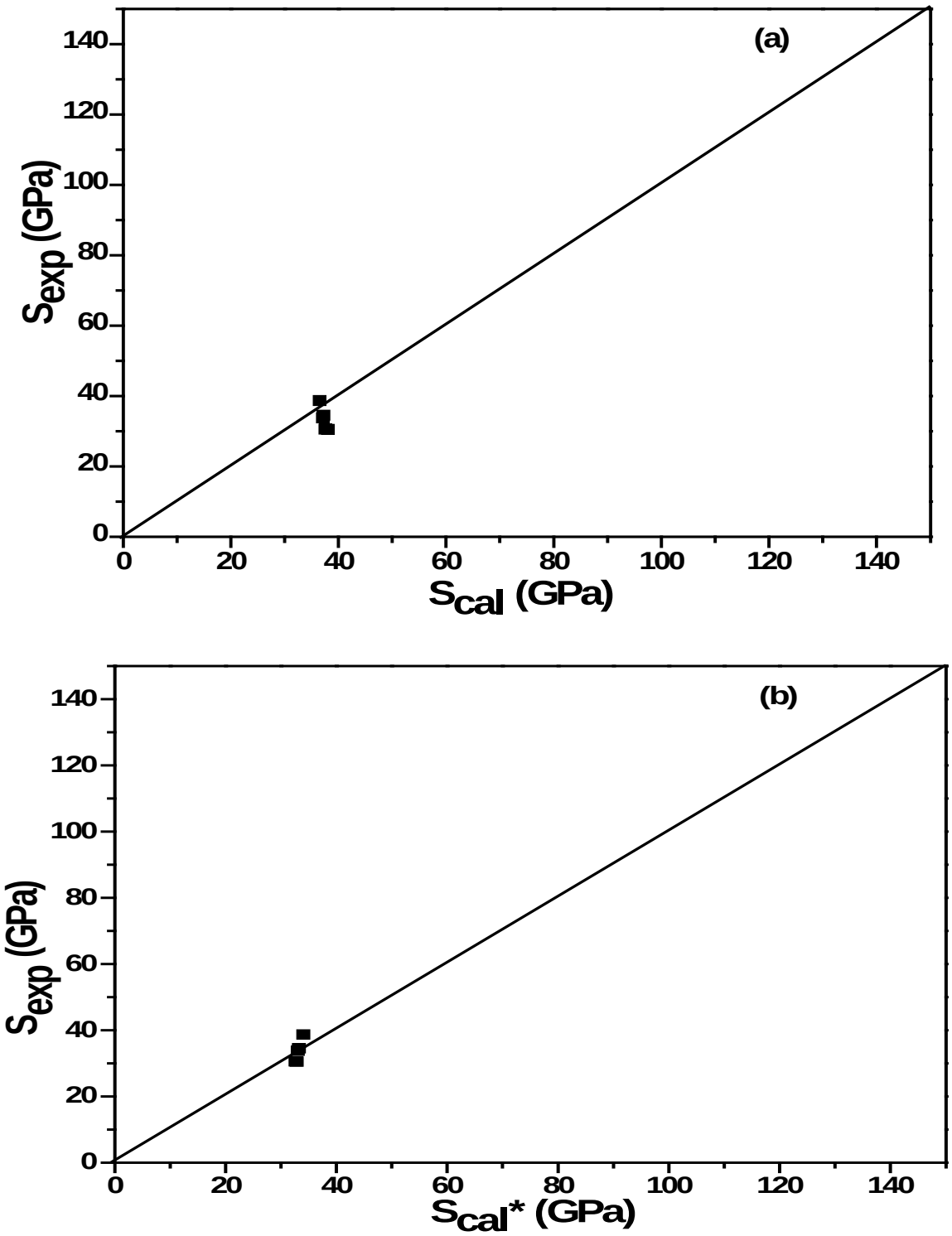


Figure 4:- Agreement between observed and theoretical calculated values of Shear modulus in the present study according to (a) Makishima and Mackenzie's [3, 4] model; (b) Rocherulle [8] model

The disagreement between the experimental and the theoretically calculated results are in the range of 0.2–18% and 0.3–20% for Young's modulus and for bulk modulus, respectively by Makishima and Mackenzie's theory and this disagreement between the experimental and the theoretically calculated results are in the range of 2-20% and 5-40% for Young's modulus and for bulk modulus, respectively by Rocherulle model. The calculated values of Poisson's ratio from the theory of Makishima and Mackenzie [3, 4] and Rocherulle [8] are not satisfactory with the experimental values.

Fig. 4 (a) and (b) shows the agreement between the observed and theoretically calculated values of shear modulus from the Makishima and Mackenzie model [3, 4] and the Rocherulle [8] model, respectively. Fig. 4 (a) shows that the calculated values from the Makishima and Mackenzie model [3, 4] is in satisfactory agreement (between 88 and 98% in Fig. 4 (a)) with the experimental values, So this model is valid for the studied glass system taking into account the uncertainty of experimental data. Fig. 4 (b) shows that the calculated values from the Rocherulle [8] model are not in the satisfactory agreement (between 80 and 95% in Fig. 4 (b)) with the experimental values.

6. CONCLUSION

Elastic moduli and Poisson's ratio decreases with the increase of Jd content in glasses along diopside ($CaMgSi_2O_6$)-jadeite ($NaAlSi_2O_6$) join. Comparison of theoretical and experimental values of elastic moduli and Poisson's ratio of the diopside ($CaMgSi_2O_6$)-jadeite ($NaAlSi_2O_6$) glass system leads to the conclusions that:

1. The correlation between the observed and calculated values of Young's modulus from Makishima and Mackenzie [3, 4] model as well as Rocherulle [8] model is not satisfactory.
2. The satisfactory agreement between the observed and theoretically calculated values of shear modulus is valid only for Makishima and Mackenzie [3, 4] model and values from Rocherulle [8] model are not satisfactory.
3. The calculated values of bulk modulus and Poisson's ratio from the theory of Makishima and Mackenzie [3, 4] and Rocherulle [8] are not satisfactory.

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