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Full Length Research Paper

Elastic moduli prediction and correlation in soda lime silicate glasses containing ZnO

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Soda lime silicate glass (SLS) containing zinc oxide (ZnO) was prepared and its elastic properties through the principle of Rocherulle's model were investigated. Different density, dissociation energy and elastic moduli were calculated theoretically for each glass samples containing different weight percentage of ZnO and these values were compared with experimental results by using ultrasonic pulse echo technique. Thereafter, the values of elastic moduli (including Young's modulus, bulk modulus, shear modulus and Poisson's ratio), derived from experimental data of the glass were compared with those theoretically results calculated in term of Rocherulle's model. According to Rocherulle model, the value of Poisson's ratio decreased as weight percentage of ZnO in SLS glass increased and the elastic moduli tends differ to the experimental values and it was further discussed.

Key words: Elastic moduli, Poisson's ratio, glass systems.

INTRODUCTION

One of the biggest problems in industrial world today is the disposal of waste materials. Most of the waste in the world is dumped in landfill sites but many countries have limited number of suitable sites and furthermore, this disposal method is non-environmental friendly. A number of methodologies of treatment and recycling have been developed to minimize the harmful effects in the environment caused by the landfill disposal of solid waste. Among the materials being recycled today, glass is one of the most difficult materials to be recycled.

Glass is a solid material which is a mixture of inorganic compounds. It is usually transparent or translucent, hard, brittle and pure, to the elements of glass. Generally, it is made by rapid cooling of molten glass with no visible crystals formed. The basic structural unit of silica glass is the silicon-oxygen tetrahedral and these tetrahedral are linked to other tetrahedral into 3-dimensional network (Huang and Behrman, 1991). Yuan et al. (2001) proposed the structure of such glass in form of short range order and the pure silica glass has a density of 2.200 gcm^{-3}. The melting temperature for preparation of soda-lime silicate (SLS) glass is about 700°C which is lower than melting temperature of pure silica, and it is a soft glass because of its relatively low softening temperature. The common commercial SLS glass has density of 2.500 gcm^{-3} and its Young modulus and Poisson ratio are, 72 and 0.25 GPa respectively (Wei et al., 2005). Generally, silica acts as a glass former. Addition of Na_2O can lower the melting point of glass. However, it will increase the solubility of glass in polar solvent such as water. Therefore, CaO is added to decrease the effect of soda on the solubility of the glass and it also increases the hardness and durability of the glass (Mattencci et al., 2002).

A number of the elastic properties studies of the glass-forming materials have been carried out in order to understand the behavior of glass and glass ceramics (Matori et al., 2010). There are some models to analyze the structure and correlate the elastic moduli of the oxide glass. Previous research carried out by Bridge et al. (1983), Higazy and Bridge (1985), El-Mallawany (1990), El-Mallawany et al. (1994) and Sidkey et al. (1997) had been aimed to explain via qualitatively, the behavior of the structure of the oxide glasses in terms of the number of network bond per unit volume, the average atomic ring size, the average stretching force constant and the mean cross-link density. In this research, the raw material of SLS glass is directly used from milled glass bottles. Pure ZnO powder is homogenously mixed with SLS glass

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powder with different weight ratios of ZnO and then, being melted, moulded and annealed to produce a solid glass. Each glass samples were tested by ultrasonic pulse echo machine to determine the ultrasonic shear velocity and longitudinal velocity at room temperature and both values were used to determine the Young's modulus, bulk modulus, shear modulus, longitudinal modulus and Poisson's ratio (Mak and Gauthier, 1993). Since the strength of materials increases with their elastic moduli, therefore, it is possible to assess strength, indirectly from their elastic property (Hwa et al., 2000). The research of the elastic constants of the glassy materials gives important information about the structure of non-crystalline solids since they are directly related to the inter-atomic forces and potentials (Hwa et al., 2000; Smyth, 1959; Makishima et al., 1978; Wei et al., 2001; Bridge et al., 1983; Elshafie, 1997; Abd El-Moneim et al., 1998; El-Mallawany, 1998).

Makishima and Mackenzie (1975) have worked out a theoretical model to calculate the elastic moduli of oxide glasses in terms of the packing density of chemical compositions and the dissociation energy of oxide constituents per unit volume. They also used the electrostatic attraction energy of ions within a crystal and Gruneisen's first rule to explain the dissociation energy as a function of Young's modulus, bulk modulus, shear modulus and Poisson's ratio (Hemono et al., 2006). Their theory was applied to several non-crystal and multi-component oxide glasses. The theoretical elastic modulus determination using Rocherulle's model suggests that, the elastic modulus of glass is composition and dissociation energy dependence (Makishima and Mackenzie, 1973). Rocherulle et al. (1989) expended the research of Makishima and Mackenzie to oxynitride glasses and acquain a thermodynamic factor to account for the substitution of oxygen by the nitrogen within the glass network. The elastic modulus obtained by this model is independent of sample glass density but depend on density of each oxide in the glass. The prime goal of the present work is to compare the experimental values of elastic modulus obtained by pulse echo technique method with the theoretical values obtained from Rocherulle model.

EXPERIMENTAL PROCEDURE

SLS glass was obtained from waste material and crushed until it became powder, in size of few microns. The SLS glass powder and ZnO powder were weighed by using electronic digital weighing machine to prepare the $(ZnO)_x(SLS)_{1-x}$ glasses (x = 0.05, 0.10, 0.20, 0.30 and 0.40 wt. %). After mixing, the batch of mixture was dry milled for 18 h to increase their homogeneity. 30 g of batch sample was placed in alumina crucible and preheat at 400±1°C in electric furnace for 1 h. Then, the batch was melted at 1400±1°C for 2 h in another furnace. After that, the molten glass was poured onto a preheated stainless-steel mold. The molten glass solidified rapidly in room temperature and was then placed into another furnace at 400°C for an hour for annealing process. Finally, the glass samples were cut into a dimension of (1.0 cm × 2.0 cm × 0.3 cm) by using diamond blades before being grinded, using silicon carbide paper from 120 to 600 meshes in order to get a smooth surface and to remove the cutting deformation marks. The surface of the glass samples must be parallel to get good results for the ultrasonic measurement. Finally, the samples were polished, using polishing machine with few drops of diamond polisher with size of 6 and 1 µ to obtain a perfect surface.

The density of samples was determined, using Archimedes' principle. The sample was weighed in air and in acetone, using electronic digital weighing machine of uncertainty ±0.0001 g at room temperature. According to Archimedes' principle, the buoyant force is of equal magnitude to the weight of acetone displaced by the volume of the glass sample. It is an upwards force. Hence, the density of glass, ρ_{glass} can be expressed as:

$$\rho_{glass} = \frac{\text{weight of glass in air}}{\text{weight of glass in acetone}} \times \rho_{acetone}$$

where $\rho_{acetone} = 0.789\ \text{gcm}^{-3}$.

Acetone was chosen because it is inert to glass samples and has low surface tension and thus, discourages air bubbles from adhering to the sample when it is immersed into it. The MBS8000 ultrasonic data acquisition model was used to determine the ultrasonic shear velocity and longitudinal velocity of all glass samples at room temperature with frequency of 5MHz. Two quartz transducers in this machine, which are *X*-cut transducer and *Y*-cut transducer, were required to generate longitudinal wave and transverse wave respectively (Uozumi et al., 1972). Burnt honey was used to ensure a good contact between sample and transducers (Sidek et al., 2004). Therefore, it can reduce refraction of wave in air gap as well as reduce the loss of acoustic energy due to unwanted reflection (Sidek et al., 2005).

RESULTS AND DISCUSSION

Five series of glasses were successfully prepared and they were transparent, bubble-free and homogeneous. After melting and quenching, each glass samples was chemically analyzed by ICP and the chemical compositions were given in Table 1.

Figure 1 show that the density of glass samples increases with increasing weight percentage of ZnO in the SLS glass due to several factors. SLS glasses contain alot of non-bridging oxygen with highly open glass network. The addition of ZnO will increase their mass and volume. However, the increase in mass is larger than the increase in volume. An increase of density of the glasses probably results in changes of crosslink density by adding of ZnO. The ZnO added into a glass, fills up the interstitial sites of the glass structure and expense of all Na_2O, CaO and SiO_2 will cause the increase in density values, up to, 3.313 gcm^{-3}. This is related to the larger atomic mass of ZnO (81.389 gmol^{-1}). It is expected that the density and the molar volume should show opposite behaviour to each other. Insertion of ZnO cause the decrease in the value of molar volume and this can be attributed to the introduction of Zn^{2+} ions as network modifiers or formers into the glass network.

The sound velocities (both longitudinal and transverse) of $(ZnO)_x(SLS)_{1-x}$ and pure SLS glasses were given in

Table 1. Chemical composition of all glasses in weight (%).

Designation	Composition (wt. %)									
	SLS	**ZnO**	**SiO_2**	**CaO**	**Na_2O**	**Al_2O_3**	**K_2O**	**MgO**	**ZnO**	**Others**
S1	95	5	65.7	10.7	11.6	2.7	1.4	2.1	5.2	0.6
S2	90	10	63.1	10.2	11.0	2.4	1.3	1.9	9.6	0.5
S3	80	20	55.4	9.1	10.2	2.1	1.1	1.7	19.7	0.7
S4	70	30	45.9	7.9	8.4	1.8	0.9	1.4	29.6	4.1
S5	60	40	39.2	7.6	7.4	1.4	0.6	1.2	38.9	3.7

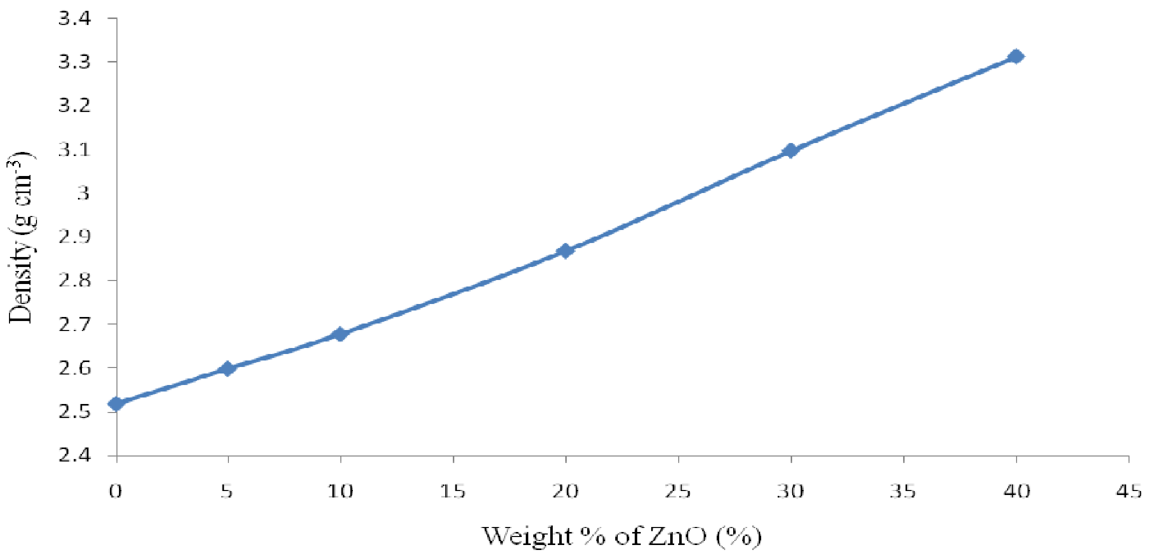

Figure 1. Graph of glass density, ρ versus weight percentage of ZnO in SLS glass.

Table 2. Longitudinal velocities are almost twice the transverse velocities at the same × weight percentage of ZnO. For example, at × = 0.30, the longitudinal velocity is 4084 ms^{-1} whereas, transverse velocity is 2013 ms^{-1}. At ×= 0.40 the longitudinal velocity decreases to 3541 ms^{-1} whereas, transverse velocity decreases to 1704 ms^{-1}. As ZnO act as network modifiers, the Zn^{2+} will cause the breaking of the oxygen bonds which connect two former cations and leads to the creation of non-bridging oxygen bond (single bonded oxygen ion) and these make the glass network become weak (Sidek et al., 2005). The results indicate that, the ultrasonic velocities of these glasses which decrease with increasing ZnO content, caused by the glass network, with less ZnO composition, should contain less non-bridging oxygens (NBOs), and consequently give higher elastic strength.

The elastic strain in an amorphous solid (such as glass) generated by a minor stress, can be described by two independent elastic constants, C_{11} and C_{44} (Chen et al., 2000). 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, respectively, are the longitudinal and transverse velocities (Cheng et al., 2003). The Young's modulus (E), bulk modulus (K), shear modulus (S) and Poisson's ratio (σ), can be determined from the sound velocities by the following equations (Higazy and Bridge, 1985):

$$\text{Young's modulus, } E = \rho V_L^2 \, \frac{3V_L^2 - 4V_T^2}{V_L^2 - V_T^2} \tag{1}$$

$$\text{Bulk modulus, } K = \frac{1}{3} \rho \, (3V_L^2 - 4V_T^2) \tag{2}$$

$$\text{Shear modulus, } S = \rho V_T^2 = C_{44} \tag{3}$$

$$\text{Poisson's ratio, } \sigma = \frac{V_L^2 - 2V_T^2}{2(V_L^2 - V_T^2)} \tag{4}$$

The elastic constants (C_{11} and C_{44}), Young's modulus (E), bulk modulus (K) and Poisson's ratio (σ) from experimental sound velocities for glass samples were given in Table 3. The overall uncertainty for the above

Table 2. The density and sound velocities (both longitudinal and transverse) for $(ZnO)_x(SLS)_{1-x}$ and pure SLS glasses.

Designation	Composition (wt. %)		ρ (g cm^{-3})	V_L (m s^{-1})	V_T (m s^{-1})
	SLS	ZnO			
SLS	100	0	2.520	4956	3202
S1	95	5	2.600	4836	3115
S2	90	10	2.679	4667	3048
S3	80	20	2.869	4640	2667
S4	70	30	3.098	4084	2013
S5	60	40	3.313	3541	1704

Table 3. The calculated elastic constants (C_{11} and C_{44}), Young's modulus (E), bulk modulus (K), Poisson's ratio (σ), and fractal bond connectivity (d) from experimental data for $(ZnO)_x(SLS)_{1-x}$ and pure SLS glasses.

Designation	C_{11} (GPa)	C_{44} (GPa)	E (GPa)	K (GPa)	σ	$d=4C_{44}/B$
SLS	62.54	26.77	58.63	29.24	0.1645	3.78
S1	60.81	25.23	57.80	27.17	0.1455	3.71
S2	58.35	24.89	56.15	25.17	0.1281	3.96
S3	61.79	20.41	51.17	34.57	0.2533	2.36
S4	51.69	12.58	33.64	34.95	0.3395	1.44
S5	41.54	9.62	25.96	28.71	0.3493	1.29

calculated quantities is estimated to be about ±1%. Bergman and Kantor (1984) derived an expression of $d = 4C_{44}/K$, (given information on effective dimensionality of the materials) for an inhomogeneous random mixture of fluid and a solid backbone near the percolation limit. This new parameter, d, called as fractal bond connectivity here, d = 3, for 3D tetrahedral coordination polyhedral, d = 2 for 2D layer structures, and d = 1 for 1D chains, respectively (Bogue and Sladek, 1990; Saunders et al., 2001). From Table 3, the calculated longitudinal elastic constant (C_{11}) of the studied glasses are lower than the pure SLS glass.

The increasing of ZnO content in the glass samples may play important roles in the explanation of the decreasing value of elastic constant. Young's modulus illustrates the changes of the chemical bond and the bond strength in the glass structure. Young's modulus is defined as $(F/A)(\Delta L/L)$, ratio of stress over strain. The value of E indicates how much strain a glass sample can withstand when a certain amount of stress is applied on it. The Young's modulus of glass samples is less than 100 GPa. Thus, the decreased value of Young's modulus indicated that the glass samples do not tolerate well to the strain.

Poisson's ratio is in the range of 0.14 to 0.34, where pure SLS glass shows this ratio to be 0.16 and indicates the higher Poisson's ratio obtained since most of the bonds are ionic, in comparison with pure SLS glass, which is predominantly covalent (Smyth, 1959; Abd El-Moneim et al., 1998). Bridge et al. (1983) suggested a close correlation between the value of Poisson's ratio and the cross-link density (defined as the number of bridging bonds per cation) of the glass structure. The cross-link density of 2, 1 and 0 are related to the value of Poisson's ratio of 0.15, 0.30 and 0.40, respectively. Consequently, Poisson's ratio for the glass samples is about 0.24, which implies a 2D layer structure. This finding agrees well with the prediction from the fractal bond connectivity model, which the d-value of the glass samples is about 2.

It is always interesting to compare the experimental results with the prediction from the existing theories. Makishima and Mackenzie (1973, 1975) proposed a model to calculate the elastic moduli of oxide glasses by using the dissociation energy of oxide constituents per unit volume and the packing density of chemical compositions by the following equations:

$$E_{cal} = 83.6\, C_t \textstyle\sum_i G_i X_i \qquad (5)$$

$$K_{cal} = 100\, C_t^2 \textstyle\sum_i G_i X_i \qquad (6)$$

$$S_{cal} = \left[\frac{300\, C_t^2}{10.2\, C_t - 1}\right] \textstyle\sum_i G_i X_i \qquad (7)$$

$$\sigma_{cal} = 0.5 - \frac{1}{7.2\, C_t} \qquad (8)$$

where G_i and X_i are the dissociation energy per unit volume and the mole fraction of the oxide component i, respectively.

Table 4. Experimental elastic moduli and Poisson's ratio values for $(ZnO)_x(SLS)_{1-x}$ and pure SLS glasses and those calculated from Rocherulle model.

Sample (wt. %)	C_t	G_t (kcal cm^{-3})	Elastic moduli and Poisson's ratio							
			Experimental				Calculated			
			E	K	S	σ	E'_{cal}	K'_{cal}	S'_{cal}	σ'_{cal}
SLS	0.594	14.942	58.63	29.24	27.3	0.164	74.22	52.75	31.37	0.266
5	0.592	14.753	57.80	27.17	25.2	0.145	73.07	51.78	30.80	0.265
10	0.590	14.558	56.15	25.17	24.9	0.128	71.88	50.79	30.32	0.264
20	0.586	14.154	51.17	34.57	20.4	0.253	69.44	48.75	29.33	0.263
30	0.582	13.727	33.64	34.95	12.6	0.339	66.89	46.64	28.29	0.261
40	0.578	13.275	25.96	28.71	9.6	0.349	64.23	44.46	27.21	0.260

In order to calculate the packing density (V_t) of oxides glasses in form of A_mO_n, the packing factor (V_i) of i^{th} oxide component should be calculated first from the following equation:

$$V_i = (6.023 \times 10^{23}) \frac{4}{3} \pi \, (mR^3_A + nR^3_O) \qquad (9)$$

where R_A and R_O are the respective Pauling's ionic radius of cation A and anion O.

Since the value of packing factor (V_i) is independent to the ith oxide component on the sample, (V_i) of an oxide is constant for all samples. Therefore, the packing density for the glasses is given as:

$$V_t = \left(\frac{\rho}{M}\right) \sum_i V_i X_i \qquad (10)$$

where M is the effective molecular weight and X_i is the mole fraction of component i.

Rocherulle et al. (1989) introduced some modification for the packing factor and expressed it for an oxide A_mO_n as:

$$C_i = (6.023 \times 10^{23}) \frac{4}{3} \pi \left(\frac{\rho}{M}\right) (mR^3_A + nR^3_O) \qquad (11)$$

Rocherulle model gave the expression of the elastic moduli and Poisson's ratio for the present glasses as:

$$E'_{cal} = 83.6 \, C_t \sum_i G_i X_i \qquad (12)$$

$$K'_{cal} = 100 \, C_t^2 \sum_i G_i X_i \qquad (13)$$

$$S'_{cal} = \left[\frac{300 \, C_t^2}{10.2 \, C_t - 1}\right] \sum_i G_i X_i \qquad (14)$$

$$\sigma'_{cal} = 0.5 - \frac{1}{7.2 \, C_t} \qquad (15)$$

and

$$C_t = \sum_i C_i X_i \qquad (16)$$

From Table 4, the theoretically calculated packing density (C_t), Young's modulus (E), bulk modulus (K), shear modulus (S), and Poisson's ratio (σ), based on the Rocherulle's model for the studied glass are given. It can be seen that all the calculated parameters show a small change (range between 73.07 and 64.23 GPa for Young's modulus, between 51.78 and 44.46 GPa for bulk modulus and between 30.80 and 27.21 GPa for shear modulus) with the increasing of ZnO content. The increasing in ZnO content will break down some of the bridging bonds in the network and hence, create non-bridging oxygen atoms. The creation of non-bridging oxygens in the network is expected to reduce the connectivity and decrease the elastic moduli of the $(ZnO)_x(SLS)_{1-x}$ glass.

The result of experimental elastic moduli (Young's modulus, shear modulus and bulk modulus) of $(ZnO)_x(SLS)_{1-x}$ glass, are compared with the theoretically calculated value from Rocherulle's model as shown in Figures 2, 3 and 4. An excellent agreement of the results on the Young's modulus, shear modulus and bulk modulus are obtained from Rocherulle models for the studied glasses and all the value are of similar shape compared to the theoretically calculated values.

Conclusions

The density of glass samples increases, from 2.600 to 3.313 gcm^{-3} as the weight percentage of ZnO in the SLS glass increases from, 5 to 40%. This is due to the Zn ion which is relatively heavier than Si^{4+} ion, and the Zn^{2+} tends to occupy the interstitial sites in glass structure. The effect of mass increase is larger than the effect of packing density decrease upon the value of glass density. The packing factor, C_t of $(ZnO)_x(SLS)_{1-x}$ glass decreases gradually from 0.592 to 0.578 as the weight percentage of ZnO in the SLS glass increases. It happens because, the C_t of glass sample was composition dependent and the value C_t of ZnO is relatively

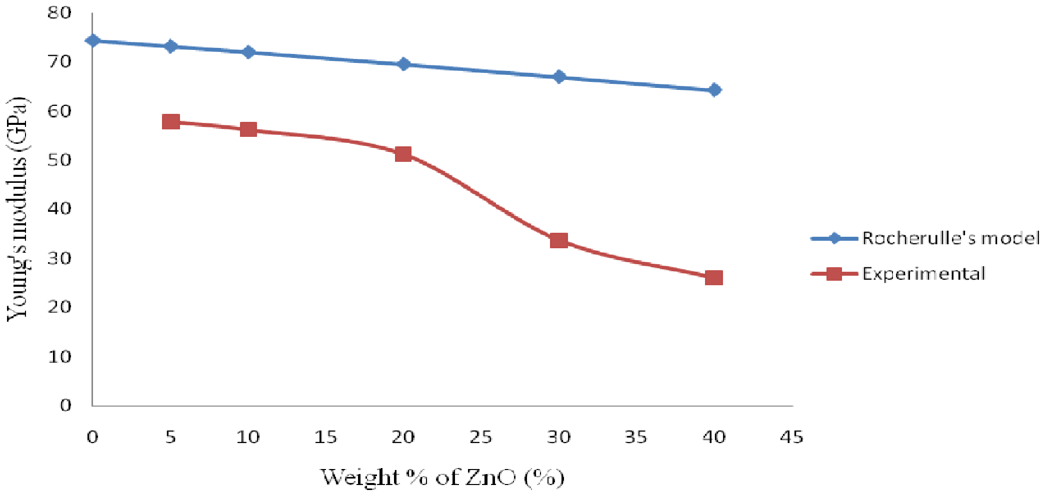

Figure 2. Graph of Young's modulus, *E* versus weight percentage of ZnO in SLS glass.

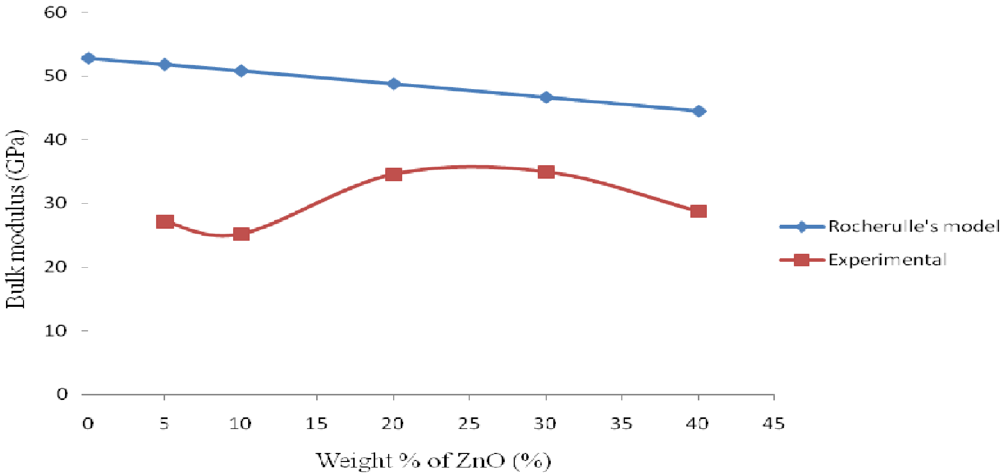

Figure 3. Graph of bulk modulus, K versus weight percentage of ZnO in SLS glass.

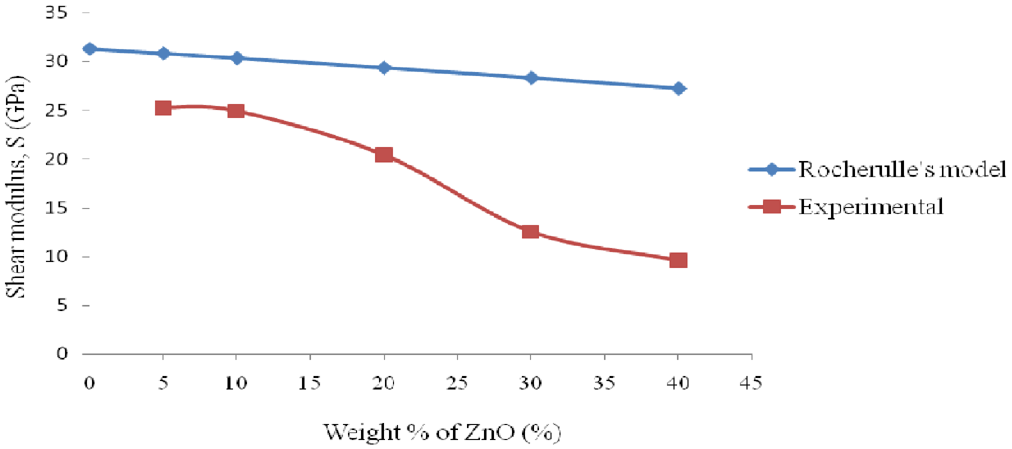

Figure 4. Graph of shear modulus, *S* versus weight percentage of ZnO in SLS glass.

relatively less than SLS glass.

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