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Correlation between acoustical and structural properties of glasses: Extension of Abd El-Moneim model for bioactive silica based glasses

Amin Abd El-Moneim

Department of Basic Science, College of Education, University of Dammam, P.O. Box: 2375, Dammam 31451, Saudi Arabia

HIGHLIGHTS

- Abd El-Moneim model was extended for bioactive glasses.
- Ultrasonic attenuation was correlated with structural parameters.
- Correlation was carried out in Si–Na–Ca–P glasses.
- The model is valid for all investigated glass samples.
- Agreement between theoretical and experimental elastic moduli was studied.

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ABSTRACT

Correlation between room temperature ultrasonic attenuation coefficient and the most significant structural parameters has been studied in the bioactive silica based glasses, for the first time. The correlation has been carried out in the quaternary SiO_2–Na_2O–CaO–P_2O_5 glass system using the two semi-empirical formulas, which have been presented recently by the author. Changes in the elastic properties, related to the substitution of SiO_2 by alkali Na_2O and alkaline earth CaO oxides, have also been deduced by evaluating the mean atomic volume, packing density, fractal bond connectivity and density of the analogous crystalline structure. Furthermore, values of the theoretical elastic moduli have been calculated on the basis of Makishima-Mackenzie theory and compared with the corresponding observed values. Results show that the correlation between ultrasonic attenuation coefficient and the oxygen density, average atomic ring size, first-order stretching force constant and experimental bulk modulus was achieved at 5 MHz frequency. Values of the theoretically calculated shear modulus are in excellent correlation (C. R. >95%) with the corresponding experimental ones. The divergence between the theoretical and experimental values of bulk modulus has been discussed.

1. Introduction

In recent years, the biomaterials such as bioactive glasses and glass-ceramics have been developed for different newer applications in the field of medicine [1]. Different recent studies reveal a numerous successful implementations of bioactive glasses such as replacement of damaged or deceased body parts [2–7]. The silica content is one of the major factors which determine the properties of the bioactive glasses and it forms a firm chemical bond to bone [2]. Peitl et al. [8] suggested that, the bioactive glassy materials should have SiO_2 content smaller than 60 mol%, high concentrations of Na_2O and CaO as well as high CaO/P_2O_5 ratio.

Mechanical/acoustical properties, i. e. elastic moduli, Poisson's ratio, acoustic Debye temperature, hardness and ultrasonic attenuation coefficient, of bioactive glasses are very important parameters for practical applications and also for understanding the structural characteristics and bonding strength states of glass network. In the recent years, the study of acoustical properties of bioactive glasses as a function of composition using an ultrasonic pulse-echo technique becomes an interesting subject [1,9–11]. This is due to the fact that, the ultrasonic waves are closely related to the elastic and inelastic properties of the glass. Marikani et al. [9] have prepared the bioactive $47P_2O_5$-30.5CaO-(22.5-x)Na_2O-xK_2O glasses and investigated their acoustical properties using ultrasonic pulse-echo technique. The influence of SiO_2 content on the structure, glass transition temperature, ultrasonic attenuation, Vickers hardness, fracture toughness, fracture surface energy and elastic moduli

E-mail address: aminabdelmoneim@hotmail.com.

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of the bioactive (43-x)SiO_2-(25.5-x)Na_2O-(25.5-x)CaO-6P_2O_5 (wt%) glass system has been investigated by Rajendran and his co-workers [10]. The effect of heat treatment on the structure, stability, mechanical properties, and elastic stiffness of the bioactive SiO_2–Na_2O–CaO–P_2O_5 and SiO_2–Na_2O–CaO–K_2O–MgO–P_2O_5–B_2O_3 glasses has also been studied [1,11]. The temperature dependence of mechanical properties showed the existing of softening in the glass network structure as temperature increases.

In the few last years, the correlation between ultrasonic attenuation coefficient and the critical structural parameters of the glass has been attracting considerable attention by the present author and his co-workers [12–16]. In 2006, Abd El-Moneim [12] presented a model of two semi-empirical formulas, which correlate the experimentally measured ultrasonic attenuation coefficient at room temperature with the oxygen density, average atomic ring size, first-order stretching force constant and experimental bulk modulus of the glass. These semi-empirical formulas have been verified for variety of borate-, tellurite- and phosphate-based glasses, such as TiO_2-doped CaO–Al_2O_3–B_2O_3, RO-Al_2O_3–B_2O_3 (R = Mg, Ca or Sr), ZnO–PbO–B_2O_3, TiO_2–Na_2O–CaO–P_2O_5, Ag_2O–V_2O_5–TeO_2, BaO–TeO_2, PbO–P_2O_5, Li_2O–TeO_2–B_2O_3–$P_2O_5$and Cr_2O_3–Na_2O–ZnO–P_2O_5 glasses [12–16].

However, to the best of this study's knowledge, the validity of Abd El-Moneim semi-empirical formulas [12] has not been testified for the silicate glasses so far. In addition, the silica based bioactive glasses have recently shown a great success in many clinical applications in both dental and orthopedic field. Therefore, the first goal of the present work is to demonstrate the validity of Abd El-Moneim semi-empirical formulas [12] for the bioactive silica based (97.4-x_1-x_2) SiO_2-x_1Na_2O-x_2CaO-2.6 P_2O_5 (mol%) glasses. Secondly, the effect of SiO_2 substitution by Na_2O and CaO on the structure and elastic properties of these glasses has been explored by evaluating many critical compositional parameters, such as the mean atomic volume, fractal bond connectivity, packing density, mean atomic ring size, density of the analogous crystalline structure, average first-order stretching force constant and dissociation energy per unit volume of the glass. Finally, the theoretical values of bulk modulus and shear modulus have been calculated on the basis of Makishima-Mackenzie theory [17,18] and compared with the corresponding experimental values.

2. Theory

2.1. Makishima-Mackenzie model [17,18] for packing density, dissociation energy per unit volume and elastic moduli

Generally, the elastic properties of glasses are closely related to interatomic forces and potentials in the lattice structure. Makishima and Mackenzie [17,18] presented a theoretical model for predicting the elastic moduli and Poisson's ratio of multi-component oxide glasses in terms of chemical composition, packing factors and dissociation energies of the oxide constituents. According to this model, the packing density (defined as the ratio between the minimum theoretical volume occupied by the ions and the corresponding ionic volume of the glass), V_t, dissociation energy per unit volume (volume density of energy), G_t, bulk modulus, K_{th}, and shear modulus, S_{th}, of a multi-component glass can be calculated from the following relations:

$$V_t = \frac{\rho}{M}\sum_i x_i V_i = \frac{\rho}{\sum x_i M_i}\sum x_i V_i \quad (1)$$

$$G_t = \sum_i G_i x_i \quad (2)$$

$$K_{th} = 10 G_t V_t^2 \quad (3)$$

$$S_{th} = 3K_{th}/(10.2V_t - 1) \quad (4)$$

where x_i, M_i, G_i and V_i are the respective molar fraction, molecular weight, dissociation energy per unit volume and packing factor of the oxide component i, whereas ρ is the density of the glass and$M = \sum x_i M_i$ is its molecular weight. The packing factor and dissociation energy per unit volume can be obtained from equations given for the oxide A_bO_c as:

$$V_i = \frac{4}{3}\pi N_a\left(bR_A^3 + cR_O^3\right) \quad (5)$$

$$G_i = \frac{\rho_i}{M_i}U_i \quad (6)$$

where R_A and R_O are the respective ionic radius of metal A and oxygen O, whereas N_a is Avogadro's number and U_i is the dissociation energy per mole (molar dissociation energy) of oxide component i with density ρ_i.

2.2. Abd El-Moneim model [12] for room temperature ultrasonic attenuation coefficient

According to the ring deformation model [19], the network of oxide glasses can be regarded as two- or three-dimensional structure of A–O covalent bonds (A = cation and O = anion) and atomic rings of different sizes as shown in Fig. 1.

Ultrasonic attenuation coefficient describes the total reduction in ultrasonic intensity due to absorption of energy by the medium and deflection of energy from the path of the beam by reflection, refraction and scattering [14]. Quite recently, Abd El-Moneim [12] derived the following two semi-empirical formulas, which correlate the experimentally measured ultrasonic attenuation coefficient at room temperature, α, with the mean atomic ring size, ℓ, oxygen density, $[O]$, average first-order stretching force constant, F, and experimental bulk modulus, K, of the glass:

$$\alpha = \chi_1 \frac{[O]}{F}\ell^m \quad (7)$$

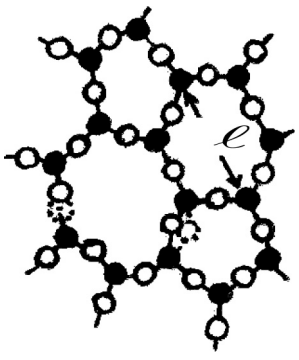

Fig. 1. Schematic two-dimensional representation of a glassy material with average atomic ring size ℓ. ● represents cations and o represents anions.