Journal of Non-Crystalline Solids 405 (2014) 141–147

Contents lists available at ScienceDirect

Journal of Non-Crystalline Solids

journal homepage: www.elsevier.com/ locate/ jnoncrysol

Correlation between acoustical and structural parameters in some oxide glasses

Amin Abd El-Moneim

Physics Department, Faculty of Science, Zagazig University, Zagazig, Egypt

ARTICLE INFO

Article history:
Received 12 June 2014
Received in revised form 1 September 2014
Accepted 10 September 2014
Available online 19 September 2014

Keywords:
Glass;
Acoustical property;
Elastic modulus;
Attenuation coefficient;
Microstructure

ABSTRACT

Correlation between acoustical and structural parameters has been carried out in TiO_2–Na_2O–CaO–P_2O_5, ZnO–PbO–B_2O_3 and BaO–TeO_2 glass systems. The correlation is based on the recently presented Abd El-Moneim semi-empirical formulas, which relate the room temperature ultrasonic attenuation coefficient with oxygen density, average atomic ring size, first-order stretching force constant and experimental bulk modulus of the glass. Fractal bond connectivity and mean atomic volume have also been evaluated and interpreted in terms of composition to deduce changes in the structure of the glass. Furthermore, values of the theoretical bulk modulus have been calculated on the basis of Rocherulle et al. model and compared with the corresponding observed values. It has been found that, majority of the studied glass samples have two-dimensional structure and the network strength changes in the order BaO–TeO_2 glasses ≺ TiO_2–Na_2O–CaO–P_2O_5 glasses ≺ ZnO–PbO–B_2O_3 glasses. Abd El-Moneim semi-empirical formulas can be applied successfully to predict changes of ultrasonic attenuation coefficient in these glasses at room temperature. Also, Rocherulle et al. model is valid for majority of the investigated glass samples.

1. Introduction

Acoustical properties play a significant role in understanding the structural characteristics of glasses, since they are directly related to the microscopic structure through the behavior of the network formers and modifiers [1–13]. Recently, the non-destructive ultrasonic 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. Extensive studies on the acoustical and structural properties of borate, phosphate, silicate and tellurite glasses have been reported in the recent years in view of their technological applications [1–13]. In addition, attempts have been made to estimate elastic moduli and Poisson's ratio of glasses from packing density and dissociation energy per unit volume of chemical oxides that constituent the glass network [4,6,14–17]. The most widely used model is that proposed by Makishima and Mackenzie [14,15] and extended thereafter by Rocherulle et al. [17] to oxynitride glasses.

Recently, Abd El-Moneim [18–22] presented number of studies on the correlation between ultrasonic attenuation coefficient and structural parameters of oxide glasses. A model of two semi-empirical formulas, which correlate the experimentally measured ultrasonic attenuation coefficient at room temperature with oxygen density, average atomic ring size, first-order stretching force constant and experimental bulk modulus, was reported [19]. These semi-empirical formulas have been verified for TiO_2-doped CaO–Al_2O_3–B_2O_3, Ag_2O–V_2O_5–TeO_2 and RO–Al_2O_3–B_2O_3 (R = Mg, Ca or Sr) glass systems [20,21]. More recently, Abd El-Moneim [22] derived a new semi-empirical formula, which correlates the room temperature ultrasonic attenuation coefficient with packing density, dissociation energy per unit volume and first-order stretching force constant of the glass. The same author verified his semi-empirical formula for TiO_2-doped CaO–Al_2O_3–B_2O_3 and Ag_2O–V_2O_5–TeO_2 glass systems [22].

The applicability of Abd El-Moneim model [19,22] for a wide variety of phosphate, borate, gemanate, tellurite and silicate glasses is worth demonstrating. Recently, the acoustical properties of TiO_2–Na_2O–CaO–P_2O_5, ZnO–PbO–B_2O_3 and BaO–TeO_2 glass systems have been studied using ultrasonic pulse-echo technique [7,10,11]. In the present work, the validity of Abd El-Moneim semi-empirical formulas [19,22], which correlate the room temperature ultrasonic attenuation coefficient with oxygen density, average atomic ring size, first-order stretching force constant and experimental bulk modulus, has been demonstrated for these glass systems. Special attention is also given on the structure of the glass by evaluating and discussing the compositional dependence of fractal bond connectivity and mean atomic volume. Finally, the

E-mail address: aminabdelmoneim@hotmail.com.

http://dx.doi.org/10.1016/j.jnoncrysol.2014.09.014
0022-3093/

agreement between the theoretically calculated and experimentally measured values of bulk modulus is studied on the basis of Rocherulle et al. model [17].

2. Theory

2.1. Abd El-Moneim model [18,19] for room temperature ultrasonic attenuation coefficient

Multi-component oxide glasses can be regarded as solution of oxides, in which no definite combinations occur. Each oxide exerts its own influence so that any physical or structural property of the glass as a whole may reasonably be considered as the sum of the influences of all individual oxides. 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.

Bulk modulus is defined as the ratio between isotropic pressure and relative volume change. According to the ring deformation model [23], the bulk modulus of a glassy material is combined with both the average first-order stretching force constant, F and mean atomic ring diameter, ℓ, according to the following relation:

$$K = \chi_1 F \ell^{-n} \tag{1}$$

where χ_1 is a constant and n is a positive power, with a value of ~4 for oxide glasses. The average first-order stretching force constant of multi-component glasses could be obtained from an equation reported by Bridge and Higazy in the form [24]

$$F = \frac{\sum_i x_i n_i f_i}{\sum_i x_i n_i} \tag{2}$$

where x_i is the molar fraction of the oxide component i, whereas n_i is the coordination number of cation and f_i is the first-order stretching force constant between cation and anion.

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. Bridge and Patel [25] expressed quantitatively the total number of loss centers per unit volume of the glass, N, in terms of both the oxygen density, $[O]$, and mean atomic ring size as following:

$$N = \chi_2 [O] \ell^{m} \tag{3}$$

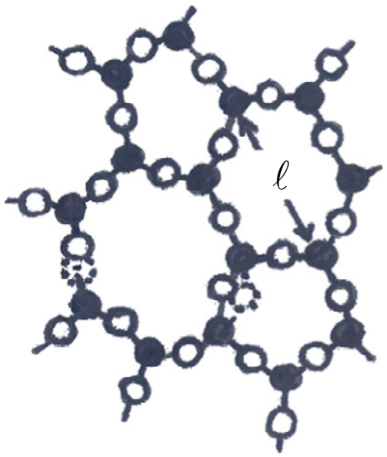

Fig. 1. Schematic two-dimensional representation of a glassy material with average atomic ring size ℓ. Full circles represent cations and open circles represent anions.

where χ_2 is another constant, m is a positive power and $[O]$ is given by [26]:

$$[O] = N_a N_O \frac{\rho}{M} \tag{4}$$

where ρ is the glass density, N_O is the number of oxygen atoms in the glass formula, N_a is Avogadro's number and M is the molecular weight of the glass, which is calculated by using the relation $M = \sum_i x_i M_i$, where M_i is the molecular weight of the oxide component i.

Abd El-Moneim [18] suggested that, the experimentally measured ultrasonic attenuation coefficient at room temperature is related to the total number of loss centers per unit volume and average first-order stretching force constant of the glass according to the following relation:

$$\alpha = \chi_3 \frac{N}{F} \tag{5}$$

where χ_3 is a frequency dependent constant. Recently, Abd El-Moneim [19] derived quantitatively the following two semi-empirical formulas, which correlate the room temperature ultrasonic attenuation coefficient with oxygen density, average atomic ring size, first-order stretching force constant and experimental bulk modulus of the glass:

$$\alpha = \chi_4 \frac{[O]}{F} \ell^{m} \tag{6}$$

$$\alpha = \chi_5 \frac{1}{F} \left[\frac{F}{K}\right]^{m/n} \tag{7}$$

where $\chi_4 = \chi_2 \chi_3$ and χ_5 are two frequency dependent constants. Eqs. (6) and (7) suggest that the average first-order stretching force constant, experimental bulk modulus, oxygen density and mean atomic ring size can be used to predict the compositional dependence of ultrasonic attenuation coefficient in glasses at room temperature.

2.2. Rocherulle et al. model [17] for bulk modulus

The estimation of elastic moduli based on glass composition is very useful for the development of glassy materials. Rocherulle et al. [17] have made an attempt to improve the agreement between the experimentally measured values of elastic moduli and those theoretically calculated on the basis of Makishima–Mackenzie theory [14,15]. They introduced some modifications in the expression of packing factor, which has been proposed by Makishima–Mackenzie [14,15] for oxides, and reported the following relations for calculating both the packing density, C_t and bulk modulus, K_{th}^{*} of multi-component glasses:

$$C_t = \sum_i \frac{\rho_i}{M_i} x_i V_i \tag{8}$$

$$K_{th}^{*} = 10 C_t^2 G_t \tag{9}$$

where V_i and G_i are the respective packing factor and dissociation energy per unit volume (volume density of energy) of the oxide component i, whereas $G_t = \sum_i G_i x_i$ is the dissociation energy per unit volume of the glass.

3. Analysis and discussion

Tables 1 to 3 summarize the experimental values of density, bulk modulus and ultrasonic attenuation coefficient of the investigated $BaO–TeO_2$, $ZnO–PbO–B_2O_3$ and $TiO_2–Na_2O–CaO–P_2O_5$ glass systems, respectively. All data of bulk modulus and ultrasonic attenuation coefficient were measured at room temperature and 5 MHz frequency using ultrasonic pulse-echo technique [7,10,11]. It is seen from these tables