Temperature dependence of the mechanical behavior of ZnO-Na2O-P2O5 glasses

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Abstract. In this research, we investigate the mechanical behaviour of zinc sodium phosphate glass system with the various concentration of chromium oxide doped. The glass system was synthesized using the melt quenching technique. The concentration of chromium oxide varying from 0.5% to 2.5%. The ultrasonic wave velocities have been determined as functions of temperature from 273 K and 333 K. The transit time data and density are used to determine all of the mechanical behavior for these glasses, we found that the mechanical behaviour of the glass system decrease as the Cr2O3 content increase, which indicates a weakening of the binding energy in the network.

1. Introduction

Glasses is used in many forms and plays varied roles, both passive and active. These include transmitting optical components such as lenses, windows, light guides and substrates for thin film coatings, gain media in form of rods, disks, and fibers, magneto-optic materials exhibiting, diamagnetic or paramagnetic Faraday rotation for optical isolators, acousto-optic material for photonic switching in integrated optics [1,2]. Incorporation of Cr2O3 in modifiers into glass networks produces the energy level structure necessary for the manufacture of non linear magneto-optical devices, such as magnetically tunable lasers and amplifiers for the telecommunication industry [3]. Even though there have been limited studies of zinc sodium phosphate glasses system in term of the mechanical properties and the glass structure.

Basically many types of biomaterials can be applied to dental restorative; metals, polymers, glasses, ceramics, and composites. However, the materials are selected according to the specific properties which are general guidelines for the study of dental restorative materials. These properties should be considered the biological conditions of the tooth itself, especially the presence of microorganisms. The proper dental restorative materials should build the biological bonding in tissue. Glasses systems structure have a great impact on biological mechanism and processability, especially antimicrobial properties. Some biomolecules can be replaced by glasses system [4]. On another glasses system polymer combined, the structure of the polymer was transformed to change the bonding and mechanical properties by containing bioactive glass in the glass ionomer cement [5].

The mechanical behaviour of common glasses has been analysed for along time and is now well known. To obtain more complete and accurate information about the mechanical behaviour of ZnO-
Na$_2$O-P$_2$O$_5$ glass, we measured, and report herein, the results of measurements of transit times of both longitudinal and shear ultrasonic waves at various temperatures. From the results we deduce the elastic stiffness moduli of each type of wave and use then to calculate the bulk modulus. The behavior of the ultrasonic velocity as a function of temperature includes an excess contribution; over that expected from the vibrational anharmonicity and thermally activated relaxation. This work provides detailed understanding of mechanical behaviour of ZnO-Na$_2$O-P$_2$O$_5$ phosphate glasses.

This research synthesized zinc sodium phosphate glass system with different chromium oxide concentration using melt quenching technique. Among the various glasses oxides system, phosphate glass can accommodate active ions without losing its distinctive properties [6,7]. The phosphate glass has other attractive properties, which has a high thermal expansion coefficient, high refractive index, low dispersion, low melting point, high electrical conductivity, and various structures to receive some cation or anion exchange [8]. However, phosphate glass has a weak resistance to chemicals, has a high hygroscopic, and easily vapour [9]. In relation to the dental restorative materials, this research also study the physical properties of Cr$_2$O$_3$ doped in ZnO-Na$_2$O-P$_2$O$_5$ glass system Incorporation of Cr$_2$O$_3$ in modifiers into glass networks produces the energy level structure necessary for the manufacture of biomaterial devices, such as dental restorative.

2. Experimental techniques
The glass system of ZnO-Na$_2$O-P$_2$O$_5$ were prepared, by melting dry mixtures of Cr$_2$O$_3$ content varying from 0.5% to 2.5%. Mixed batches of 20 g each were placed in platinum crucibles and heated at 300°C to remove the water for an hour, and 1000°C to remove the residual of H$_2$O for an hour and then the mixtures were melted at 1100 °C for an hour in an electric furnace. To ensure proper mixing and homogeneity, the molten liquid was shaken frequently and vigorously. After being checked, the melt was cast by pouring as fast as possible into a hot steel split mould to quench it to form a glass. The glass was immediately transferred to an annealing furnace at 300°C where it was kept at that temperature for 3 hours to relieve any residual stress which could cause embrittlement. At the end of annealing process the furnace was switched off and the glasses left to cool down to room temperature, at cooling rate 0.5 °C/min. The samples containing up to 2.5 mol % Cr$_2$O$_3$ were transparent and free from crack and bubbles, purple, good optical quality glass. After preparation, sample were stored in plastic containers in a dessicator until the measurement of ultrasonic wave velocities was performed. To do measurement of ultrasonics the glasses were cut and polished first to produce flat faces parallel about 5-7 mm apart. X-cut or Y-cut quartz transducers of 5 MHz frequency were used for the generation and detection of longitudinal and shear ultrasonic wave respectively. The grease was used to bond the transducer to the sample. Longitudinal and shear waves velocity ($V_L$ and $V_S$, respectively) were determined from the transit times of ultrasonic waves using the pulse-echo-overlap technique. However, the absolute values of the transit time and the velocity of the ultrasonic wave pulse were much less accurate due to variation in the thickness of the material bonding the transducer to the sample.

Our velocity results were used with our density data to calculate the longitudinal and shear elastic constant using the formulas,

\[ C_{11} = \rho V_L^2 \] \hspace{1cm} \text{(1)}
\[ C_{44} = \rho V_S^2 \] \hspace{1cm} \text{(2)}

where $\rho$ is the density.

The sample was placed in temperature controlled cabinet. Density was determined by Archimedes principle using toluene as the immersion.

3. Results and discussion
The velocities of longitudinal $V_L$ and shear $V_S$ ultrasonic waves, the elastic stiffness $C_{11}$ and $C_{44}$, and the bulk modulus $B^0$, of zinc sodium phosphate glass at various temperature and mol % of Cr$_2$O$_3$ are shown in Figure 1-4. In general, the measured properties correlate well with those of other transition metal glass determined previous researcher [10] as might be expected from their similar structural
configuration. Figure 1 and 2, showed that an abrupt change in velocity in the range of temperature between 270 K and 303 K. Longitudinal velocity showed more significant changes compared to shear velocity. The effects of increase of Cr$_2$O$_3$ content on the elastic stiffness bear an interesting relationship. As the Cr$_2$O$_3$ content is increase from 0.5 mol % to 2.5 mol % the elastic constant C$_{11}$ and C$_{44}$ decrease with increases in temperature. The value of elastic constant C$_{44}$ is smaller than value of elastic constant C$_{11}$ as shown in Figure 3 and Figure 4. Apparently the character of the glass are easy to bend but difficult to compress.

From Figure 5, the bulk modulus for x Cr$_2$O$_3$(1-x)P$_2$O$_5$ glasses tend to decreases with the increase in mol % of Cr$_2$O$_3$ substantially as the structure changes from the three dimensional network to the chain structure typical of the metaphosphate composition. This shows that the stiffness of the glass system at room temperature also decreases with the increase of content of Cr$_2$O$_3$ which indicates a weakening of the binding energy in the network. However the fact remained that for x Cr$_2$O$_3$ (1-x)P$_2$O$_5$ glasses, the mechanical behaviour is lower than the binary rare-earth previous reported [11-12].

![Figure 1](image1.png)

**Figure 1.** The longitudinal velocity of various mol % Cr$_2$O$_3$ vs temperature.

![Figure 2](image2.png)

**Figure 2.** The shear wave velocities of various mol % Cr$_2$O$_3$ vs temperature.
Figure 3. The temperature dependences of the elastic stiffnesses $C_{11}$ of various mol\% Cr$_2$O$_3$.

Figure 4. The elastic stiffnesses $C_{44}$ of various mol\% Cr$_2$O$_3$ vs temperature.

Figure 5. The bulk modulus of various mol \% Cr$_2$O$_3$ vs temperature.
4. Conclusions
The mechanical behaviour of zinc sodium phosphate glass system with the various concentration of chromium oxide doped from 0.5% to 2.5% mole % has been measured using ultrasonic technique as functions of temperature from 273 K to 333 K. The change of the measurement induced by application of temperature on the natural wave velocity and it was obvious found to be linearly dependent upon temperature. In general, the mechanical behaviour at this composition of the glass system is a characteristic feature of the Cr$_2$O$_3$ phosphate glasses. Acoustic softening is greatest in those glasses having the long chain methaphosphate structure. It can be concluded that this mode softening is related to valency instability.

References
[1] J H Campbell and T I Suratwala 2000 J. Non-Cryst. Solids 318 263-264
[2] R K Brow, C A Click and T M Alam 2000 J. Non Cryst. Solids 274 9-16
[3] R Harani, C A Hogarth, M M Ahmed and D F C Morris 1984 J. Materials Science letters 3 843-844
[4] B Sylvain, B Lionel, Cassagnetes and E B Jean 2003 Photonics Tech. Lett. 15(4) 516-518
[5] S Wolf, J Soures, O Lewis, J Bunkenburg, D Brown, S Jacobs, G Mourou and J Zimmermann 1980 Appl. Opt. 19(3) 409-19
[6] M Emanuele, L Joris, M Daniel, S Abate and Z Vincenzo 2013 J. Non-Cryst. Solids 362 147–51
[7] G H Dieke and R Sarup 1996 J. Chem.Phys. 31 371
[8] M Vincenzo, E Sglavo, D Mura and J L Milanese 2014 Int. J. Appl. Glass Sci. 5 57-64
[9] G A Saunders, T Brennan, M Acet and M Cankurtaran 2001 J. Non Cryst. Solids 282 291-3.05
[10] R Bogue and R J Sladek 1990 Physical Review B 42(8) 5280-5288
[11] H B Senin, G A Saunders and P J Ford 1994 Physics and Chemistry of Glasses 35 109-114
[12] A A Kutub, A E M Osman and C A Hogarth 1986 J. Materials Science letters 21 3517-3520