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Acoustic Properties of Borate Glasses Doped with Ag_2O

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Abstract: Na_2O-CaO-P_2O_5-B_2O_3-Ag_2O glasses of different compositions were prepared. The ultrasonic velocities (both longitudinal and shear) were measured at 10 MHz at room temperature using the pulse echo overlap method. Various parameters, viz., longitudinal modulus, shear modulus; bulk modulus, Young's modulus, acoustic impedance, Poisson's ratio, micro hardness, thermal expansion coefficient and Debye temperature were evaluated. The observed results have been explained in terms of the change in the structure and physical properties of the glass network with change in Ag_2O content.

Key words: Acoustic properties, Borate glass, Mechanical properties, Ag_2O, X-ray diffraction

INTRODUCTION

Glass structure is a basic issue to understand the behaviour of the materials. The velocity of ultrasonic waves and hence the elastic moduli are particularly suitable for characterizing glasses as a function of composition [1]. Ultrasonic investigation on solids is gaining much importance nowadays and interest in glasses has rapidly increased because of improving information technology. Elastic properties are very informative about the structure of solids and they are directly related to inter-atomic potentials.

In recent years, attention has been focussed more on glassy materials in view of their larger optical non-linearity and high optical quality with fast response time [2]. Ultrasonic tools are very important for characterizing materials because they have many applications in physics, chemistry, engineering, biology, food industry, medicine, oceanography, seismology, etc. Nearly all of these applications are based on two unique features of ultrasonic waves [3]:

- Ultrasonic waves travel slowly, about 100,000 times slower than electromagnetic waves. This provides a way to display information in time, create variable delays, etc.
- Ultrasonic waves can easily penetrate opaque materials, whereas many other types of radiation such as visible light cannot. Since ultrasonic wave sources are inexpensive, sensitive and reliable, this provides a highly desirable way to probe and image the interior of opaque objects.

Network formers B_2O_3 and P_2O_5 glasses have been of great importance for a variety of applications. Boron oxide is one of the best known glass formers, as well as a very good host matrix for heavy metal oxide (HMO). This is because it vitrifies up to very high additions of the heavy metal oxides.

Borate glass is one of the most characteristic glasses having unique super-structures (SS) of intermediate range order (IRO) such as boroxol ring, tetraborate, etc. The conversion between threefold coordinated boron and fourfold coordinated boron with the addition of modifier oxides is found in short-range order (SRO) and becomes one of the main factors bringing about the variety of SSs. The glass properties are also known to show unique dependencies on the types of the added modifier oxide and their concentration [4]. The ability of boron to exist in three and four oxygen coordinated environments and the high strength of covalent B-O bonds enables borate to form stable glasses. Coordination number and connectivity (number of bridging bonds) usually determine the melting point and Poisson's ratio. Glasses having higher coordination number tend to lower the bond strength. A detailed investigation of coordination number is therefore needed to understand the structural properties of glasses. The thermal stability and the lattice vibrations within the glass systems have been related to the measurement of softening and Debye temperature.

The present work aims to measure the ultrasonic velocity (both longitudinal and shear) and density of glasses namely

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Na_2O-CaO-P_2O_5-B_2O_3 (NCPB) and Na_2O-CaO-P_2O_5-B_2O_3-Ag_2O (NCPBA) doped with Ag O_2 glasses. These values have been used to evaluate longitudinal, Young's, bulk and shear moduli, Poisson's ratio, acoustic impedance, micro hardness, Debye temperature and thermal expansion coefficient, which will give further insight into the rigidity and structure of glasses.

MATERIALS AND METHODS

Preparation of Glasses: The samples of Na_2O-CaO-P_2O_5- B_2O_3 (NCPB) and Ag_2O doped (NCPBA) glass systems having different compositions were prepared by the rapid quenching method. Batches of each glass composition are listed in Table 1. Analytical grade materials of purity more than 99.9% of Na_2O- CaO- P_2O_5- B_2O_3 and Ag_2O chemicals were used to prepare the glass samples.

Appropriate amounts in mol% of chemicals in powder form were weighed using single pan balance having a sensitivity of ±0.0001 g. The homogenisation of the appropriate mixtures of the component of chemicals was achieved by repeated grinding using an agate mortar. The homogeneous mixture was put in a platinum crucible and placed in a muffle furnace. Melting was carried out under controlled conditions at temperatures 1100-1180K for system. The molten sample was cast into a copper mold having the dimension of 10 mm diameter and 6 mm height. Then the glass samples were annealed at 473K for two hours to avoid the mechanical strain developed during the quenching process. The two opposite faces of glass were highly polished to ensure a good parallelism. All glasses were cleaned with acetone to remove the presence of any foreign particles. The samples prepared were chemically stable and non-hygroscopic. The amorphous nature in all glass samples was confirmed using X-ray diffraction (XRD).

Density Measurement: The density (ρ) of all glass samples was determined by employing the Archimedes principles using benzene as buoyant and applying the relation,

$$\rho = \frac{W_1}{(W_1 - W_2)}\rho_B \quad (1)$$

Where W_1 and W_2 are the weights of the glass samples in air and in the buoyant respectively, ρ_B is the density of the buoyant at 303K.

Ultrasonic Velocity Measurement: The ultrasonic longitudinal and shear velocities of the specimen were determined by using the Pulse-Echo method. The pulser section generates electrical pulses, which are converted into ultrasonic signals using X-cut and Y-cut quartz transducers having the fundamental frequency of 10 MHz. These transducers act as both transmitter and receiver of the ultrasonic pulse. Precise transit time measurements were carried out by cross-correlation technique described elsewhere [4]. The precise ultrasonic velocities (U_ℓ and U_s) in each glasses were obtained by measuring the precise transit time (t) using the relation [5].

$$U = \frac{2d}{t} \quad (2)$$

Where U, the velocity of the specimen (ms^{-1}), d, thickness of the specimen (mm) and t, transit time in micro seconds.

Mechanical Properties: The elastic constants of the glass specimen were calculated at room temperature from the measured density (ρ), longitudinal velocity (U_ℓ) and shear velocity (U_s) by using the following standard expressions,

Longitudinal Modulus $L = \rho U_l^2$ (3)

Shear Modulus $G = \rho U_s^2$ (4)

Bulk Modulus $K = L - \left(\frac{4}{3}\right)G$ (5)

Young's Modulus $E = (1+\sigma)2G$ (6)

Poisson's ratio $\sigma = \left[\frac{L-2G}{2(L-G)}\right]$ (7)

Acoustic Impedance $Z = U_\ell \rho$ (8)

Micro Hardness $H = (1-2\sigma)\frac{E}{6(1+\sigma)}$ (9)

Debye Temperature $\theta_D = \frac{h}{K}\left[\frac{9N}{4\pi V_m}\right]^{1/3} Um$ (10)

Mean Sound Velocity $U_m = \left[\frac{1}{3}\left[\frac{2}{U_s^2} + \frac{1}{U_\ell^3}\right]\right]^{-\frac{1}{3}}$ (11)

Thermal Expansion Coefficient $\alpha_p = 23.2(U_\ell - 0.57457)$ (12)

Table 1: The Nominal composition, values of density (ρ), longitudinal velocity (U_l), shear velocity (U_s) of NCPB and Ag_2O doped Glass systems

	Nominal glass composition (mol %)						Ultrasonic velocity U ms^{-1}	
Sample name	Na_2O	CaO	P_2O_5	B_2O_3	Ag_2O	Density r x 10^{-3} kgm^{-3}	Longitudinal (U_l)	Shear (U_s)
NCPB0	24.4	26.9	2.6	46.1	0	5.6712	5072.9	2912.9
NCPBA05	24.4	26.9	2.6	46.1	0.5	5.6065	4924.0	2767.1
NCPBA10	24.4	26.9	2.6	46.1	1.0	5.5523	5220.9	3064.0
NCPBA15	24.4	26.9	2.6	46.1	1.5	5.5079	5618.6	3461.7
NCPBA20	24.4	26.9	2.6	46.1	2.0	5.4408	6028.6	3871.7

Table 2: Values of elastic moduli of NCPB and Ag_2O doped Glass systems

Sample name	Longitudinal modulus L x 10^{9} Nm^{-2}	Shear modulus G x 10^{9} Nm^{-2}	Bulk modulus K x 10^{9} Nm^{-2}	Young's modulus E x 10^{9} Nm^{-2}	Poisson's ratio s
NCPB0	146.0	48.12	81.84	120.70	0.2542
NCPBA05	136.0	42.92	78.77	108.96	0.2694
NCPBA10	151.3	52.12	81.80	108.25	0.2372
NCPBA15	173.8	66.00	85.80	107.75	0.1939
NCPBA20	197.7	81.50	89.03	95.87	0.1493

RESULTS

The experimental values of density (ρ), longitudinal velocity (U_l) and shear velocity (U_s) of the different glass specimen with respect to change in mol % of doped Ag_2O are listed in Table 1. The calculated longitudinal modulus (L), Shear modulus (G), bulk modulus (K), Young's modulus (E) and Poisson's ratio (σ) are reported in Table 2. Figs. 1 - 4 shows the variation of acoustic impedance (Z), micro hardness (H), thermal expansion coefficient (α_p) and debye temperature (θ_D) in five NCPBA glasses with composition of doped Ag_2O (0.0, 0.5, 1.0, 1.5 and 2.0).

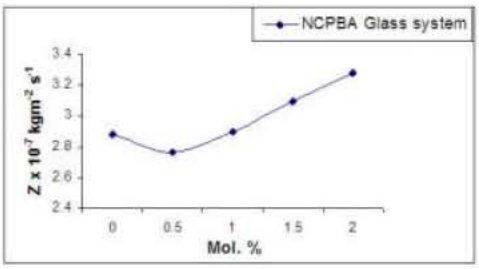

Fig. 1: Variation of acoustic impedance (Z) with pure and Ag_2O doped in mol%

On analyzing the experimental and derived parameters, interesting observation has been obtained on the addition of Ag_2O doped with Na_2O-CaO-P_2O_5 and B_2O_3.

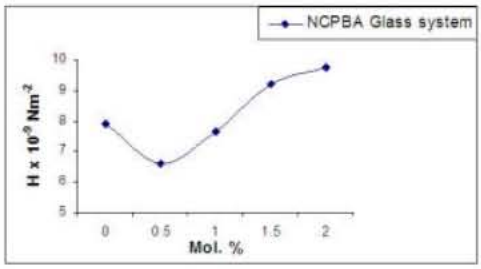

Fig. 2: Variation of micro hardness (H) with pure and Ag_2O doped in mol%

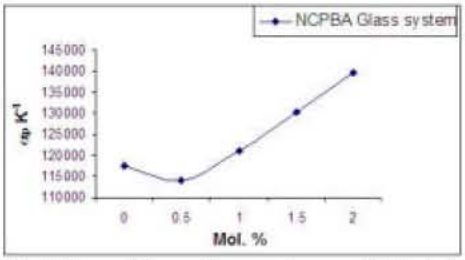

Fig. 3: Variation of thermal expension coefficient (α_p) with pure and Ag_2O doped in mol%

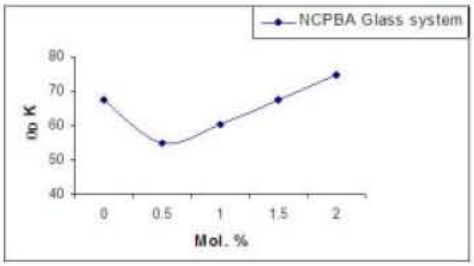

Fig. 4: Variation of Debye temperature (θ_D) with pure and Ag_2O doped in mol%

DISCUSSION

In NCPB and NCPBA glasses, the density(ρ) shows a continuous decrease (Table 1) with increase in mol% of Ag_2O doped content respectively, which is due to the structural change takes place in glass network. The structure of glass depends on the nature of ions entering in the network and hence the density. Similar trend was observed [5,6].

It is observed from the Table 1 that both longitudinal (U_l) and shear velocity (U_s) increase linearly with increase in mol% of doped Ag_2O in NCPBA glass system, but the rate of increase of U_l is greater than that of U_s. The increase in ultrasonic velocity (both longitudinal and shear) is attributed to an increase in packing density because of the transformation of coordination. Due to this increase in packing density, the rigidity of the glass system increases and hence ultrasonic velocities and elastic constants.

The longitudinal, shear, bulk and Young's moduli (Table 2) increase over the entire range of composition of doped Ag_2O in NCPBA glass systems. The increase in the values of the elastic constants has been attributed to an increase in the packing density and rigidity and hence the formation of stronger structural building units in the glass network [7].

The variation of Poisson's ratio in NCPBA glass system is shown in Table 2. The decrease in Possion's ratio indicates that the atoms experience lesser transverse contracting strain acting on them and hence become more loosely packed [8].

The amorphous nature of glass samples was confirmed by X-ray diffraction technique using an X-ray diffractometer. XRD shows an evidence of unmelted or crystalline particles in a quenched glass. The XRD pattern of NCPB and NCPBA glasses are shown in Figs 5 and 6 respectively.

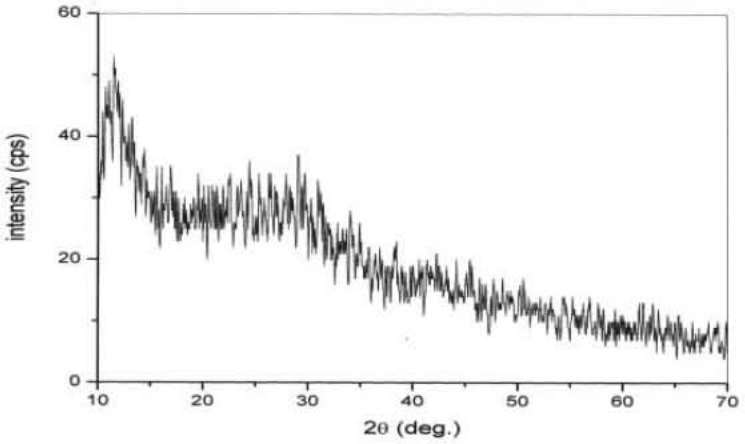

Fig. 5: X- Ray Diffractogram of Ncpb Glass

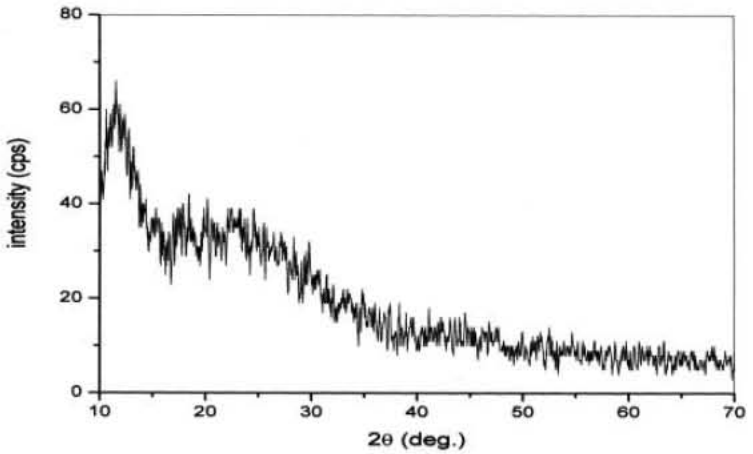

Fig. 6: X- Ray Diffractogram of Ncpba Glass

The acoustic impedance increases (Fig. 1) with increase in mol% of doped Ag_2O content in NCPBA glass system confirming the increase in rigidity of the structure of the glass. Similar trend is obtained [9]. Further, the increase in micro hardness (H) strengthens the softening points, as the network modifier (NCPBA) content is increased. The continuous increase in micro hardness as well as Poisson's ratio reveals the absence of non-bridging oxygen ion (NBO) and this causes the formation of glassy network.

The observed results are further confirmed by considering another parameter, Debye temperature, obtained directly from the measured velocity. It increases with increase in mol% of doped Ag_2O content in NCPBA system as shown in Fig. 4. The increase in Debye temperature is possibly due to the charge center coming closer than the distance required statistically to achieve a more effective coulombian interaction. Such interaction can give rise to high energy vibrational modes, thereby increasing the Debye temperature [10,11]. The thermal expansion coefficient increases with increase in mol% of Ag_2O and hence the rigidity of the structure of the glass.

CONCLUSION

From the this study, it is concluded that

- The gradual decrease in density of the glass specimen indicates the dependence of weight of the metal atom in the network modifier (NWM). The magnitude of the density is in the order NCPB > NCPBA.
- The ultrasonic velocities (U_l and U_s) of NCPB and NCPBA glasses vary linearly and the magnitude of velocity is in order NCPB < NCPBA.
- The estimated acoustical, elastic and mechanical properties of NCPB and NCPBA glasses throw light on the rigidity and compactness in structural network.

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