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Ultrasonic Studies of Physical and Elastic Properties of Sodium Borate Glasses

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Abstract: Borate based glass systems of binary B_2O_3-Na_2O with different compositions are prepared by melt quench technique. The ultrasonic (both longitudinal and shear) velocities are measured at 303K and 10 MHz frequency using the Pulse Echo Overlap method. Density of the glass samples is measured by Archimedes principle using water as immersion liquid. From the measured data, the elastic moduli and other parameters such as molar volume, Poisson's ratio, acoustic impedance, microhardness and Debye temperature have been evaluated and they are used to gain knowledge about the structural and mechanical properties and are correlated to the rigidity and compactness of the glass systems.

I. INTRODUCTION

The ultrasonic non-destructive method has been found to be the excellent technique to study the physical , mechanical and elastic properties of solids. In recent years, great interest in glasses has rapidly been increased due to their diverse applications in electronic, nuclear, solar energy and acousto-optic devices. [1] The main reason is the need for elastic properties of materials like crystals, alloys, plastics, ceramics, glasses and so on in a lot of applications.. Among the various newer techniques, pluse-echo methods are useful where measurements of highest precision are needed

The ultrasonic investigation of solids helps to understand various solid state phenomena such as grain and domain boundary effects in metals, ferromagnetic and ferroelectric materials, the diffusion of atoms, molecules and vacancies through a solid, the motion of imperfection such as dislocation as well as the interaction between the lattice sound vibration and free electrons in metals at low temperatures. The measurements of elastic constants of solids is of considerable interest and significance to both science and technology. This measurement yields valuable information regarding the forces operative between the atoms or ions in a solids. Since the elastic properties describe the mechanical behaviour of materials, this information is of fundamental importance in interpreting and understanding the nature of bonding in the solid state.

In recent years, borate based glasses have attracted the attention of researchers due to their useful physical properties, which make them promising for practical use **[2]**. In borate glasses B_2O_3 is the best and important glass former and has been incorporated in several kinds of glass systems to get physical and chemical properties. Because of its higher field strength, lower cation size, small heat of fusion and trivalent nature of B. In which, B^{3+} ions are triangularly coordinated by oxygen and corner bonded in a random configuration **[3]** The size of B^{3+} ion is very small and it can fit into the trigonal void created by three oxide ions in mutual contact, forming a BO_3 unit. BO_3 units are the primary building blocks in all borate glasses.. B_2O_3 can be considered as having the highest glass formation tendency because molten B_2O_3 does not crystalline by itself even when cooled at a slowest rate.

Alkali borate glasses are highly useful materials for vacuum ultraviolet optics and semiconductor lithography owing to the presence of stable glass forming range and transparency from the near UV to the middle infrared region **[4]** Alkali oxide is a strong modifier for glass systems and it provides transparency and homogeneity. Alkali ion doping into borate glasses introduces interesting structural variation by converting three co-ordinate boron ions into four co-ordinated boron ions and by forming non-bridging oxygen ions **[5]**.

In the BO_3 group, the oxygen is fully bridging and one negative charge each from oxygen satisfies the three positive charges on the boron ion. After the conversion from BO_3 to BO_4, all the oxygen remains bridging and the extra negative charges on the [BO_4] group are satisfied by an alkali ion in the vicinity.

An increase in the BO_4 tetrahedral in the glass structure increases the connectivity of the network, and hence flow relatedproperties decrease and viscosity increases. The addition of alkali oxide to glassy B_2O_3 causes the gradual change in the co-ordination number of boron from three to four. Further addition of alkali oxide causes the production of NBOs and increases the expansion coefficient and decreases the viscosity. This extreme behaviour is termed as the boron anomaly. The structure of alkali borate strongly depends upon the species of alkali ion **[6]**

Borate glasses modified with the addition of alkali oxide Na_2O and observed a non- linear behaviour with the increase of the alkali oxide in binary and mixed alkali borate glasses. Hence, the role of alkali oxide Na_2O in B_2O_3 network is to modify the host structure through the transformation of structural units of borate network from BO_3 to BO_4. **[7]**

II. EXPERIMENTAL

The glass samples having the general chemical formula (1-x) B_2O_3 -xNa_2O mol%) have been prepared by the melt quenching technique. Required quantities of Analar grade B_2O_3, Na_2O were mixed together by grinding the mixture repeatedly to obtain a fine powder. The mixture was melted in a porcelain crucible in an electrically heated furnace under ordinary atmospheric conditions at a temperature of about 900°C 2 hours to homogenize the melt. The obtained glass samples from the melt quenching into preheated brass mould were heat treated at a temperature of about 20 K below their calorimetric glass transition temperature for one hour remove any internal stresses. The obtained glasses were lapped and two opposite sides were polished to be suitable for use in the ultrasonic velocities measurements.

The nomenclature and the composition in mol % of BN glass samples are listed in Table .1

Table 1 Nomenclature and composition of sodium borate glasses

Nomenclature B_2O_3-Na_2O	Composition in mol %	Remarks
BN1	75-25	Mol% of B_2O_3 is decreasing with increasing Na_2O content
BN2	70-30	
BN3	65-35	
BN4	60-40	
BN5	55-45	

III. RESULTS AND DISCUSSION

The experimental values of density, longitudinal velocity and shear velocity of the different glass specimen with respect to change in mol% of Na_2O are listed in Table 1. The calculated values of molar volume, elastic moduli and Poisson's ratio are reported in Table 2. The remaining parameters such as acoustic impedance, microhardness and Debye temperature are presented in Table 3.

Figs.1,2 show the variation of density, molar volume, longitudinal velocity and shear velocity with different composition of Na_2O. The elastic moduli and the remaining parameters for BN glasses are shown in Figs.3-6. On analyzing the experimental and derived parameters, interesting observation has been obtained on the addition of Na_2O with B_2O_3.

Density measurement is an important tool to identify the structural changes of the glass network. In other words, the density is related to how tightly the ions and ionic groups are packed together in the structure. The increase in density of the BN system reveals the change in the structure of the glass network with increasing Na_2O content. That is, the increase in density may be attributed to the more close packed nature of the glass structure. The decrease in molar volume can be attributed to closing up of structure of glasses.

Table 2 Values of density, molar volume, longitudinal velocity and shear velocity of BN glass system

Name of the sample	Density $\rho(10^3\ kgm^{-3})$	Molar volume $V_m\ (10^{-6}m^3/mol)$	Ultrasonic velocity (ms^{-1})	
			Longitudinal (U_l)	Shear (U_s)
BN1	2.546	26.59	4002.2	2401.2
BN2	2.5387	25.90	4036.8	2512.3
BN3	2.6479	25.28	4098.1	2593.5
BN4	2.7594	24.12	4143.5	2608.0
BN5	2.8123	23.53	4206.1	2684.2

Table 3 Values of longitudinal, shear, bulk and Young's modulusand Poisson's ratioof BN glass system

Name of the sample	Longitudinal modulus L (GPa)	Shear modulus G (GPa)	Bulk modulus K (GPa)	Young's modulus E (GPa)	Poisson's ratio (σ)
BN1	40.78	14.67	21.20	3.5782	0.2187
BN2	42.34	16.40	20.47	3.8837	0.1839
BN3	44.46	17.81	20.72	4.1532	0.1659
BN4	47.37	18.76	22.35	4.3991	0.1719
	49.75	20.26	22.73	4.6865	0.1564
BN5					

Table 4 Values of acousticimpedance, microhardness and Debye temperature of BN glass system

Name of the sample	Acoustic impedance Z ($10^7 kgm^{-2} s^{-1}$)	Microhardness H (GPa)	Debyetemperature θ_D(K)
BN1	1.0189	2.7520	320.3
BN2	1.0490	3.4563	336.8
BN3	1.085	3.9661	349.9
BN4	1.1433	4.1046	357.6
BN5	1.1828	4.6406	370.5

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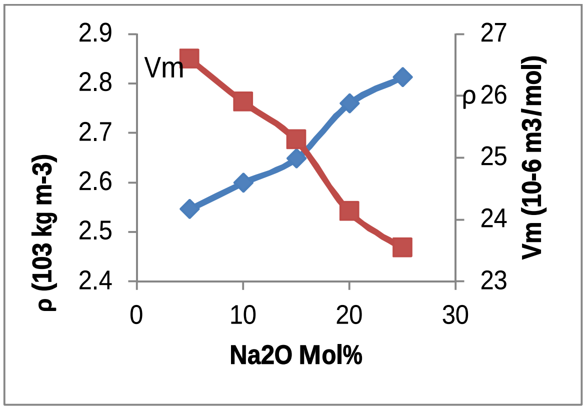


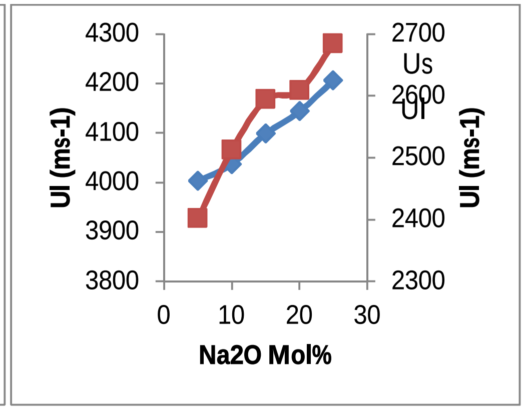
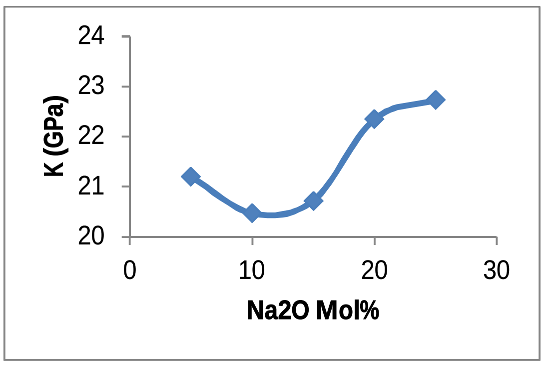


Fig 1 Variation of density and molar Fig 2 Variation of longitudinal (U_l) volume with Na_2O mol%

shear(Us) velocities of Na2Omol%



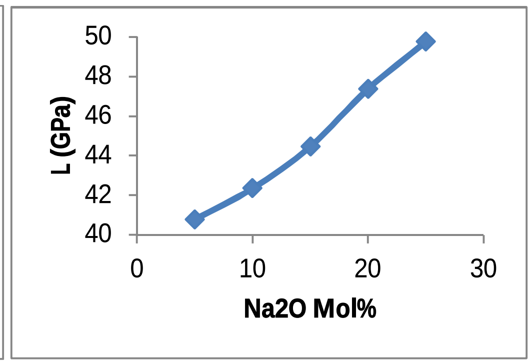
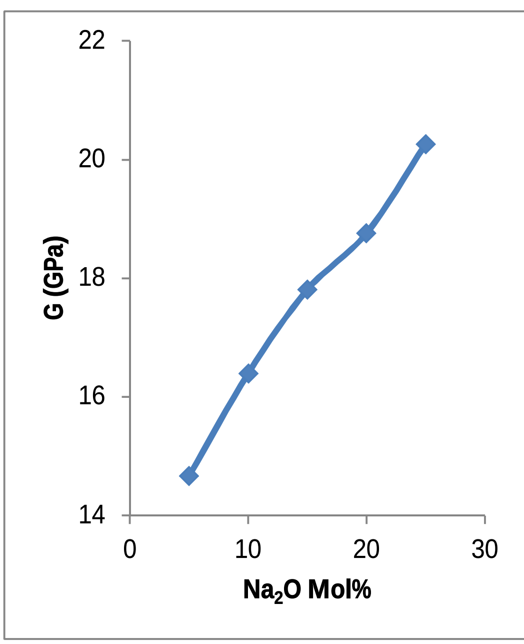


Fig 3 Variation of bulk modulus Fig 4 Variation of longitudinal with Na_2O

modulus with Na_2O



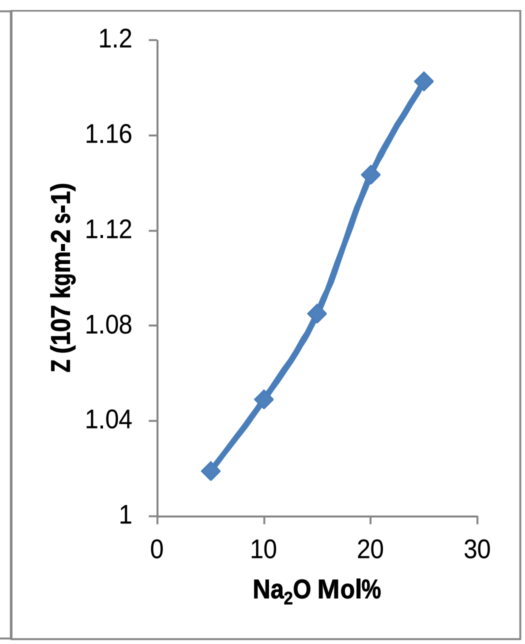


Fig 5 Variation of shear modulus with Na_2O mol%

Fig 6 Variation of acoustic impedance with Na_2O

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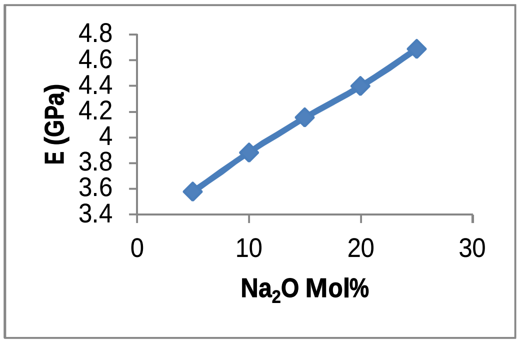


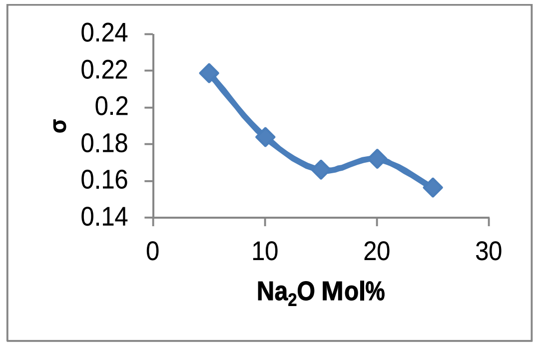
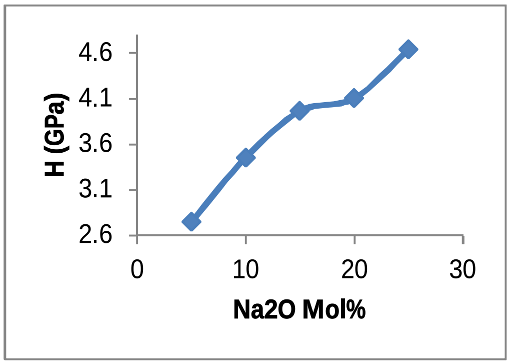


Fig 7 Variation of Young's modulus with Na_2O mol%

Fig 8 Variation of Poisson ratio with Na_2O mol%



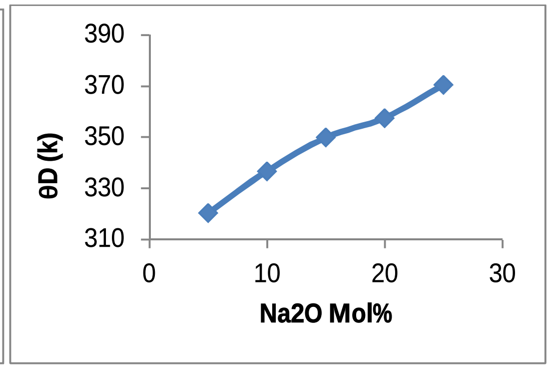


Fig 9 Variation of micro harness with Na_2O mol%

Fig10 Variation of Debye temperature with Na_2O mol%

It has been reported that the change in geometrical configuration, co-ordination number, cross link density and dimensions of inertial space of glass decides the ultrasonic velocity and hence, ultrasonic velocity is an important parameter revealing the degree of changes with composition of glasses.**[8]**

It is observed from the Figs. 2 that both longitudinal (U_l) and shear velocity (U_s) increase linearly with increase in mol% of Na_2O content. The increase in ultrasonic velocity has been attributed to an increase in packing density arises, because of the transformation of coordinated boron BO_3 into BO_4 units. This increase indicates the rigidity of the glass system increases and hence the ultrasonic velocities and elastic constants..

It is observed from the figures 3-7 that the elastic moduli increase with increasing concentration of Na_2O. This increase in elastic moduli is related to the changes occurring in compactness and cross – link density of the glass network.

The poisson's ratio is useful in exploring the degree of cross link density of the glass network and it is the ratio of transverse and linear strains for a linear stress. In the BN glasses, the drop in Poisson's ratio with the addition of Na_2O mol% indicates the increasing cross – link density and dimensionality of the glass network..

Debye's temperature is an important parameter of a solid and is expressed by equations describing the properties arising for atomic vibrations. The gradual increase of Debye's temperature in BN glass systems indicate the increase in the rigidity of the glass systems. **[9]**

The increasing of acoustic impedance and thermal expansion co-efficient are due to increase in rigidity of the glass structure. The acoustic impedance of sodium borate glasses increases, which results in the introduction of new network forming groups and large impedance to the propagation of ultrasonic waves. This also indicates that the strength of the glass network increases. **[10]**

IV. CONCLUSION

The ultrasonic velocity, density, evaluated parameters and results are in good agreement with each other. The conclusion drawn from these studies for B_2O_3 – Na_2O glasses are summarized below:

The density and molar volume is observed to concur with the general behavior which indicates the increase in connectivity of the network structure.

All the modulus, Acoustic impedance, microhardness and Debye's temperature values found to increase and Poisson's ratio decreases with the alkali ion concentration thus increasing the dimensionality and cross-link density of the as prepared glass samples.

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