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International Journal For Research in
Applied Science and Engineering Technology



INTERNATIONAL JOURNAL FOR RESEARCH

IN APPLIED SCIENCE & ENGINEERING TECHNOLOGY

Volume: 9 Issue: V Month of publication: May 2021

DOI: <https://doi.org/10.22214/ijraset.2021.34138>

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The Structural, Optical and Mechanical Properties of Sodium Borate Glasses

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Abstract: Binary $[B_2O_3-Na_2O]$ glass is synthesized by conventional melt quenching technique. XRD pattern of all the samples confirms amorphous nature of the glass. Longitudinal and shear ultrasonic velocities are measured at room temperature using Pulse echo overlap method. The density of the glasses is determined using Archimedes principle. Elastic moduli, molar volume, Poisson's ratio, acoustic impedance, microhardness and Debye temperature have been calculated from the density and ultrasonic velocity data. The above parameters are analyzed in terms of structural changes in glasses by varying their composition. The modifying action of divalent oxide is also discussed.

Keywords: XRD, Density and ultrasonic velocity.

I. INTRODUCTION

Ultrasonic characterization of materials is a versatile tool for the inspection of their microstructure and their mechanical properties. This is possible because of the close association of the ultrasound waves with the elastic and inelastic properties of the materials. A solid is differentiated from liquid by its elastic properties. The application of a stretching force to a solid is met with considerable resistance. The magnitude of deformation produced is proportional to the applied force. It is natural for any elastic substance to overcome the instantaneous deformation, when the applied force is removed 'glass' as a solid undergoes the above phenomena such that the ratio of stress to strain is a constant and defined as 'modulus of elasticity'.

Nowadays, there is an ever-increasing interest in the measurement of elastic properties of solids using ultrasonic methods due to their non-destructive nature. Their measurement yields information concerning the forces that operate between the atoms or ions comprising solids. Since the elastic properties describe the mechanical behavior of solids, this information is of fundamental importance in interpreting and understanding the nature of bonding in the solid state.

Density is considered as an important physical property in the field of glass science, especially where the glass structure is concerned. The measurement of density is a very sensitive tool that easily detects changes in geometrical configuration, coordination number, cross-link density and the dimensions of the interstitial space in the glass matrix. Besides the measured data, the evaluated parameters are also viable for characterizing the glasses as a function of their composition and describing the compactness of glass structures (1.2). A variety of glass systems have been investigated by ultrasonic measurements which yield information about the structure and mechanical properties of the glasses (3.4).

Conventional oxides such as SiO_2 , B_2O_3 , V_2O_5 , etc. can form glasses by themselves or when mixed with considerable quantities of non-glass forming oxides (5.6). B_2O_3 is one of the most common glass formers because of its lower cations size, higher bond strength, smaller heat of fusion and valence (=3) of B. The addition of alkali oxide to B_2O_3 network creates $[BO_{4/2}]$ units upto 33% of modifying oxide; further addition of modifying oxide leads to reconversion of $[BO_{4/2}]$ to $[BO_{3/2}]$ units (7). Modification is generally brought about by the reaction of the oxygen supplied by the modifier oxide with pure borate glass which results in structural changes. Alkali borate glasses have been extensively studied over the years to elucidate the nature and relative concentration of various borate units constituting the glass network. These glasses are highly useful materials for vacuum ultraviolet optics and semiconductor lithography due to their presence of stable glass forming range and transparency from the near UV to the middle infrared region (8). It is also used for applications in solid state devices, cathode materials for batteries, gas and chemical sensors. For such technical applications, it is obvious that an understanding of structure and elastic properties of glasses is necessary. In view of the aforementioned perspective, the prime goal of the present investigation is to measure the ultrasonic velocities and density of the binary BN $[(100-x)B_2O_3-xNa_2O, 30 \leq x \leq 50]$ glass use pulse echo technique and Archimedes principle respectively. These values have been used to evaluate the elastic moduli, Poisson's ratio, acoustics impedance, microhardness and Debye temperature which will give insight into the rigidity of the glass network.

II. EXPERIMENTAL

Glass samples belonging to binary glass systems are prepared using the technique described in chapter II in the temperature range 750–1100°C. The amorphous nature, density and ultrasonic velocity in the glass samples are studied using the methods, Xrd, density and ultrasonic velocity. The nomenclature and the composition in mol % of BN glass samples are listed in Table 1.0 and the photographs of the prepared glasses are displayed in Plates 1.0.

Table 1.0 Nomenclature and composition of binary and ternary glass systems

Plate	Nomenclature	Composition in mol %	Remarks
	B ₂ O ₃ – Na ₂ O		
1.0	BN30	70–30	Mol% of B ₂ O ₃ is decreasing with increasing Na ₂ O content
	BN35	65–35	
	BN40	60–40	
	BN45	55–45	
	BN50	50–50	

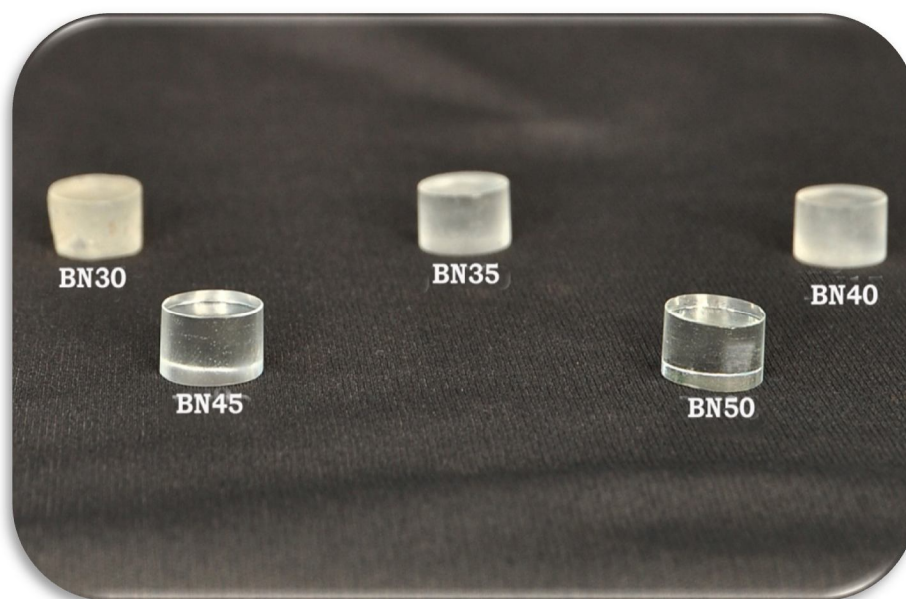


Plate 3.1. Photograph of BN glass samples

A. XRD Analysis

The XRD pattern of BN30, BN50, glasses is shown in Fig. 3.1. The X-ray spectrograms show a broad halo, which reflects the characteristics of amorphous or glassy nature, obtained at around $2\theta \approx 30^\circ$ (8.9). The appearance of broad profiles around $2\theta \approx 30^\circ$ also suggests some short-range order in the preserved BN and BNR glasses (10).

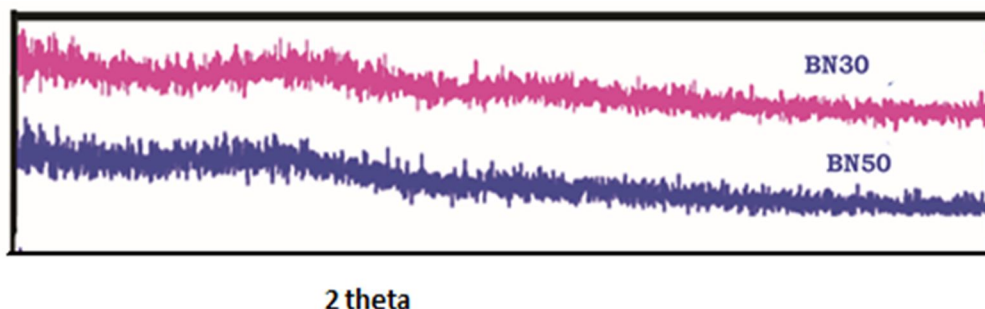
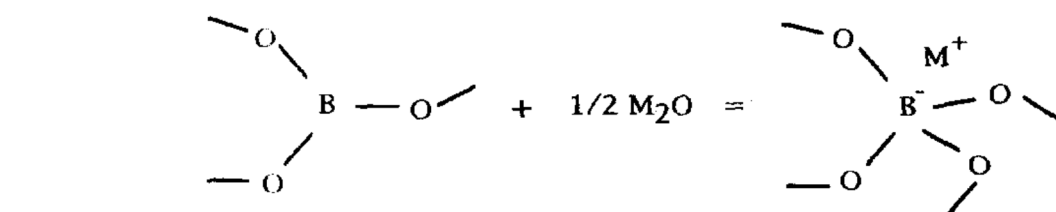
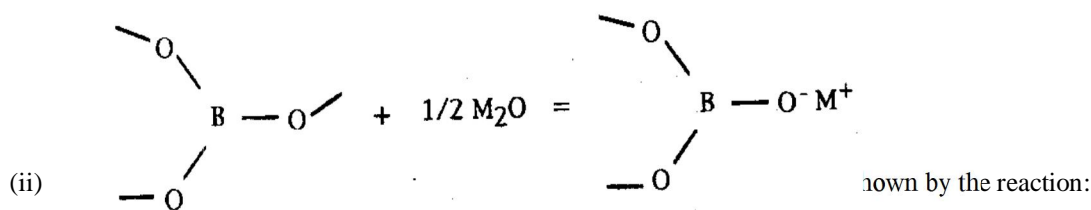


Fig. 1.0. XRD diffractogram of BN30, BN50, glasses

III. BN GLASS SYSTEM

In general, the introduction of oxygen from a modifier oxide ($M_2O = Li_2O, Na_2O$ and K_2O) into boron oxide glass brings about one of the two possibilities:

- (i) Creates non-bridging oxygen, and oxygen co-ordination around the boron remains three as shown by the following reaction



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-related properties 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 decrease in viscosity. This extreme behaviour is termed as the boron anomaly. The structure of alkali borate strongly depends upon the species of alkali ion (11).

IV. RESULTS AND DISCUSSION

The experimental values of density, molar volume, longitudinal velocity and shear velocity of the BN glasses with respect to change in mol% of Na_2O are listed in Table 3.2. The calculated elastic moduli and Poisson's ratio are reported in Table 3.3. The remaining parameters such as acoustic impedance, microhardness and Debye temperature are presented in Table 3.4.

Figs. 3.2 - 3.5 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 - 3.13. On analyzing the experimental and derived parameters, interesting observation has been obtained on the addition of Na_2O with B_2O_3 .

The variation of density and molar volume of BN glasses with Na_2O mol% are depicted in Table 3.2. It can be seen that the density increases from 2.344 to 2.639 kgm^{-3} with increase in Na_2O content and correspondingly molar volume of the glasses decreases. The reason for density increase in this case is related to the type of structural units that form when Na_2O is incorporated into the glass structure. Same variations were observed by Doweidar 1990. They have shows that when Li_2O is substituted for B_2O_3 , there is a continuous increase in density and it can be explained that both BO_4 tetrahedra and asymmetric BO_3 triangle are considerably denser than the symmetric BO_3 triangles. Similar results have been deduced by Kodama (1991), from ultrasonic velocity and density measurements in sodium borate glasses.

Ultrasonic velocity is a tool in revealing the degree of change in structure with the composition of glasses (12). In general decrease in ultrasonic velocities is related to increase in the number of non-bridging oxygen (NBO) and consequently the decrease in connectivity of the glass network (13). In the present investigation, the progressive addition of Na_2O in B_2O_3 glass matrix makes a fall in both longitudinal and shear velocities. The decrease in ultrasonic velocities is noted which results in the structural rearrangement of the glasses such that B-O-B bonds are broken and the bridging oxygens are transferred into NBOs. The formation of NBO will decrease the connectivity of the glass and the broken bonds are replaced by ionic bonds between the interstitial modifying ions (Na^+) and non-bridging oxygen of the network.

Table 1.2 Values of density, molar volume, longitudinal velocity and shear velocity of BN glass system

Name of the sample	Density ρ (10^3 kgm^{-3})	Molar volume V_m ($10^{-6} \text{ m}^3/\text{mol}$)	Ultrasonic velocity (ms^{-1})	
			Longitudinal (U_l)	Shear (U_s)
BN30	2.344	29.4	5698.2	3028.2
BN35	2.441	24.4	5442.8	2819.3
BN40	2.513	20.8	5245.1	2689.5
BN45	2.588	18.0	5034.5	2531.0
BN50	2.639	15.8	4817.1	2415.2

Table 1.3 Values of longitudinal, shear, bulk and Young's moduli and Poisson's ratio of 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)
BN30	76.11	21.49	47.45	56.02	0.3032
BN35	72.31	19.40	46.44	51.09	0.3166
BN40	69.14	18.18	44.90	48.05	0.3216
BN45	65.60	16.58	43.49	44.13	0.3309
BN50	61.24	15.39	40.71	41.01	0.3321

Many reports (13.14) show that in pure B_2O_3 , the structural unit forming the network is a planer BO_3 triangle. As sodium oxide is incorporated to the network, two competing processes that utilize the sodium cation take place. One process is the formation of BO_4 tetrahedra and the sodium cation is located adjacent to the electrically negative BO_4 unit to provide local charge neutrality. The other process is the formation of electrically negative non-bridging oxygen atoms on BO_3 unit coupled with the adjacent sodium cation. Above 33.3 mol% of sodium oxide, the formation of non-bridging oxygen atoms is significant and the fraction of boron atoms in BO_3 configuration increases quantitatively with increasing Na_2O content in the glass. These reports support the present investigation.

Figs. 3.13.4 show the variations of elastic moduli with the Na_2O content. It is observed from the figures that the elastic moduli decrease with increasing concentration of Na_2O . This is due to the development of weaker B-O-Na linkages in the place of strong B-O-B linkages.

Normally, the rigidity decrease in the glasses contributes to the decrease in velocity and hence elastic moduli, but on the other hand causes an increase in Poisson's ratio. The values of Poisson's ratio (Table 1.3) are found to increase from 0.3032 to 0.3321. The addition of Na_2O in BN glass matrix increases the Poisson's ratio, and this reveals the presence of non-bridging oxygen and hence becomes more loosely packed (14.15).

Table 1.4 Values of acoustic impedance, microhardness and Debye temperature of BN glass system

Name of the sample	Acoustic impedance Z ($10^7 \text{ kgm}^{-2} \text{ s}^{-1}$)	Microhardness H (GPa)	Debye temperature θ_b (K)
BN30	1.3357	2.8199	394.6
BN35	1.3286	2.3766	393.7
BN40	1.3181	2.1614	392.3
BN45	1.3029	1.8691	390.9
BN50	1.2712	1.7231	388.5

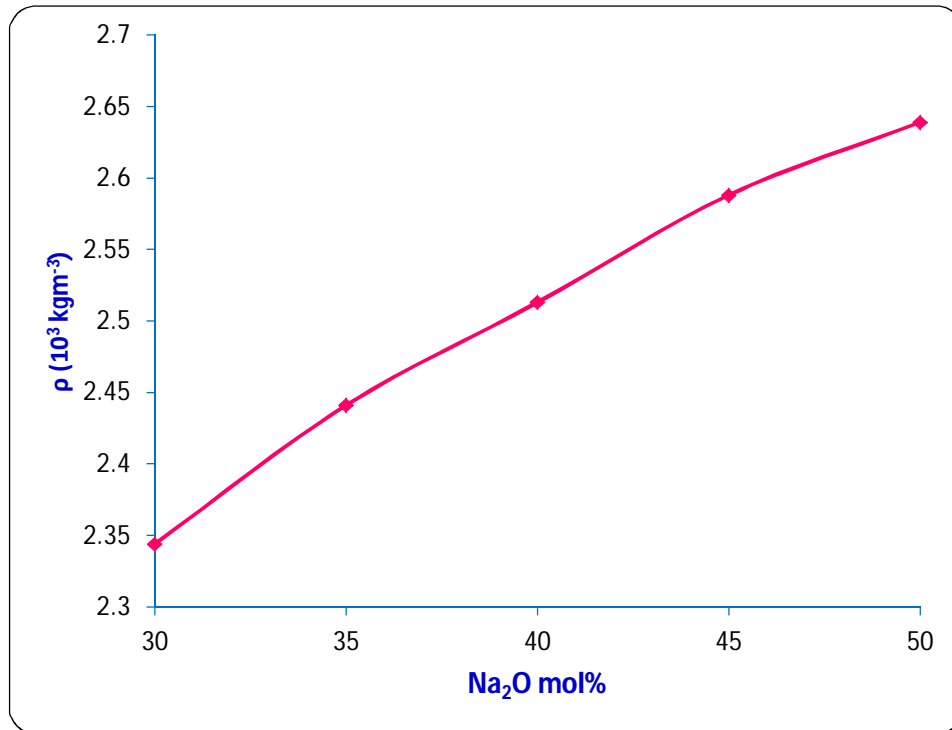


Fig. 1.2. Variation of density with Na₂O mol%

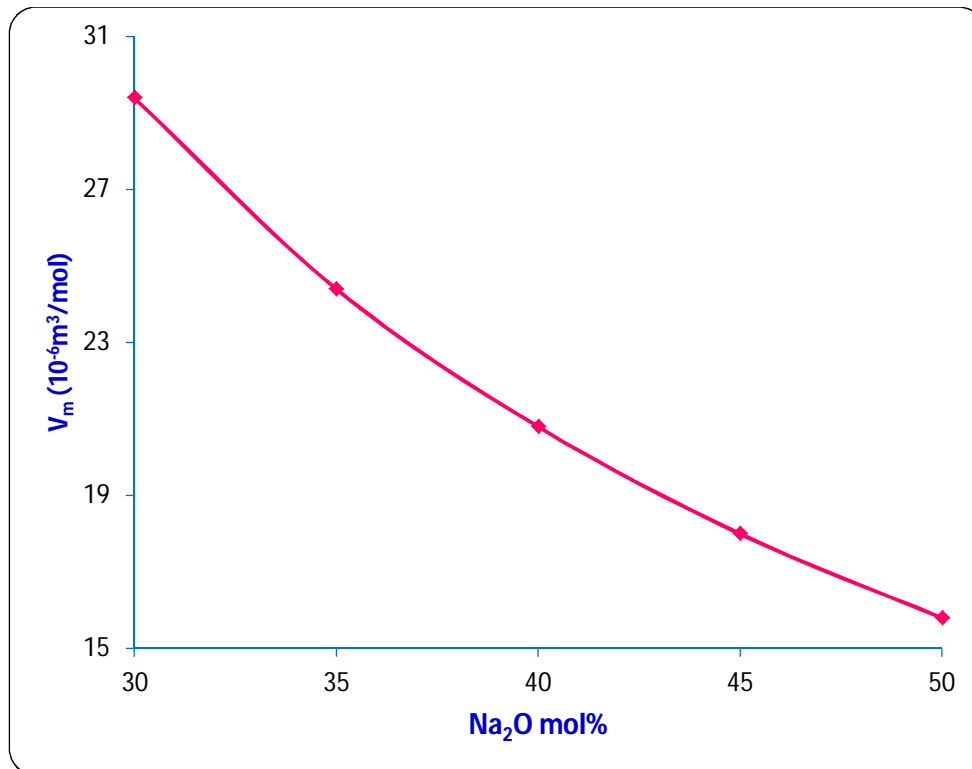


Fig. 1.3. Variation of molar volume with Na₂O mol%

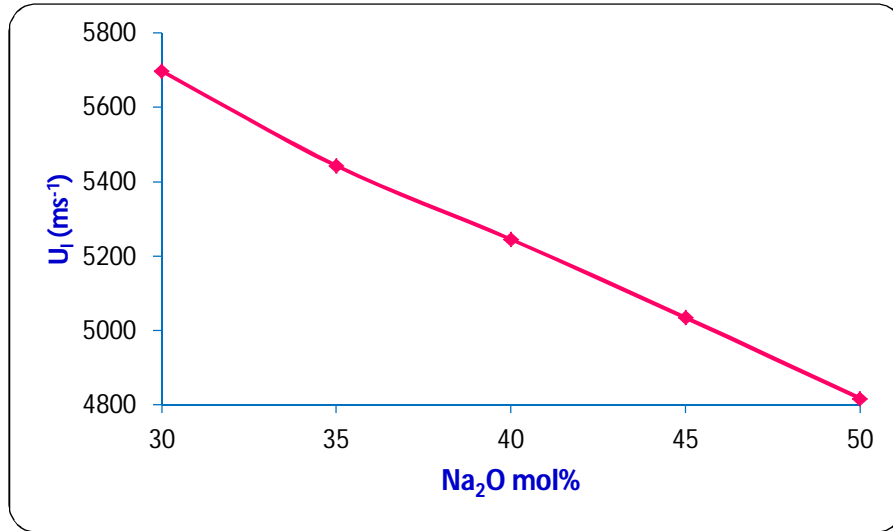


Fig. 1.4. Variation of longitudinal velocity with Na₂O mol%

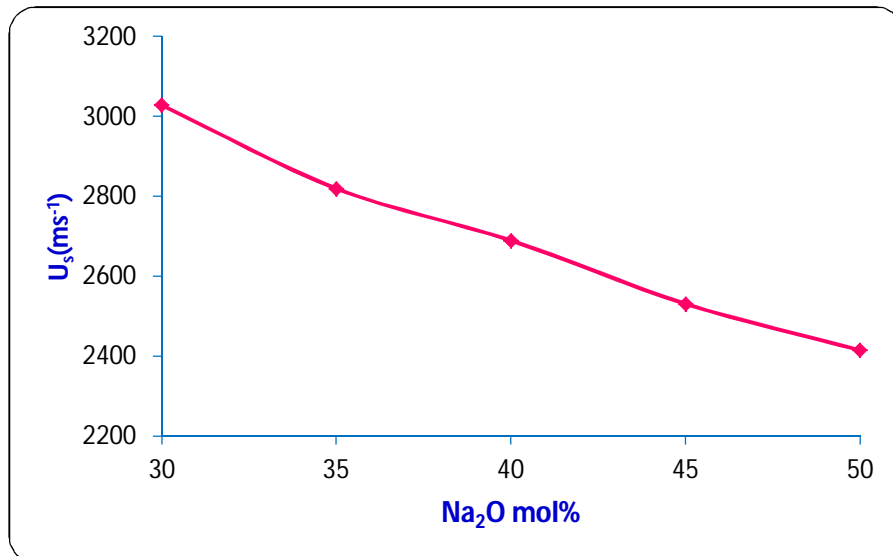


Fig. 1.5. Variation of shear velocity with Na₂O mol%

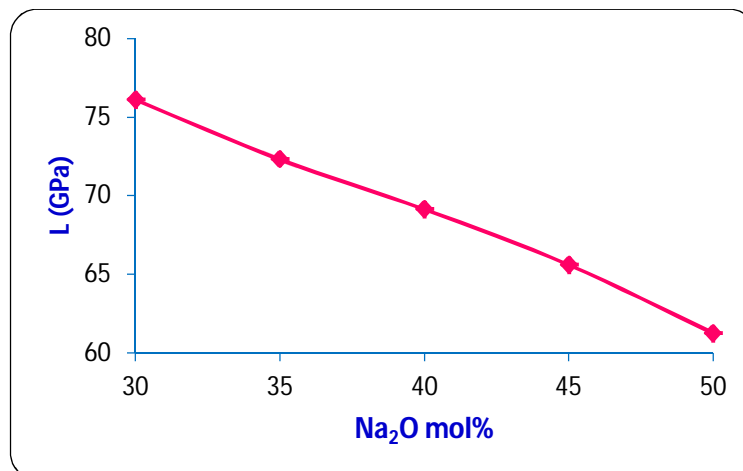


Fig. 1.6. Variation of longitudinal modulus with Na₂O mol%

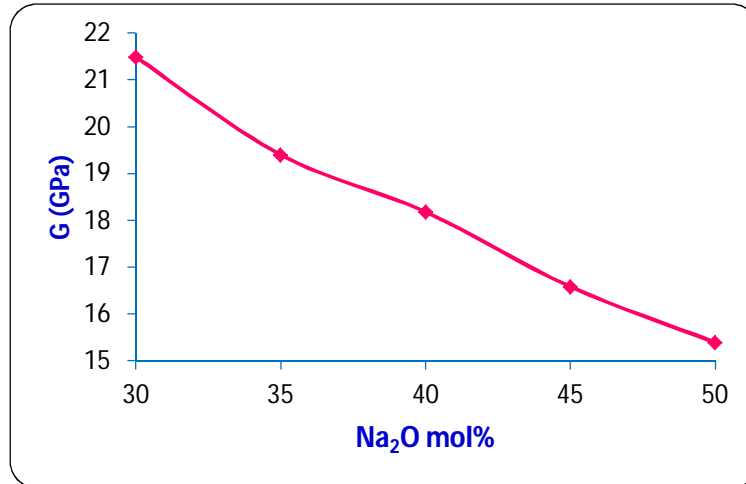


Fig. 1.7. Variation of shear modulus with Na₂O mol%

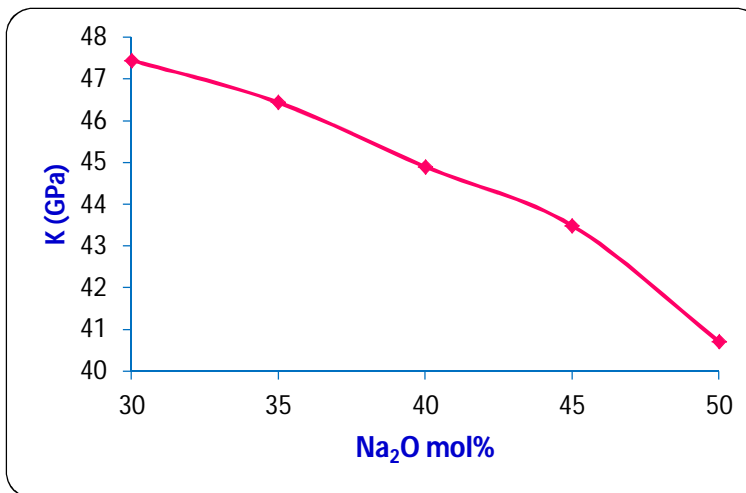


Fig. 1.8. Variation of bulk modulus with Na₂O mol%

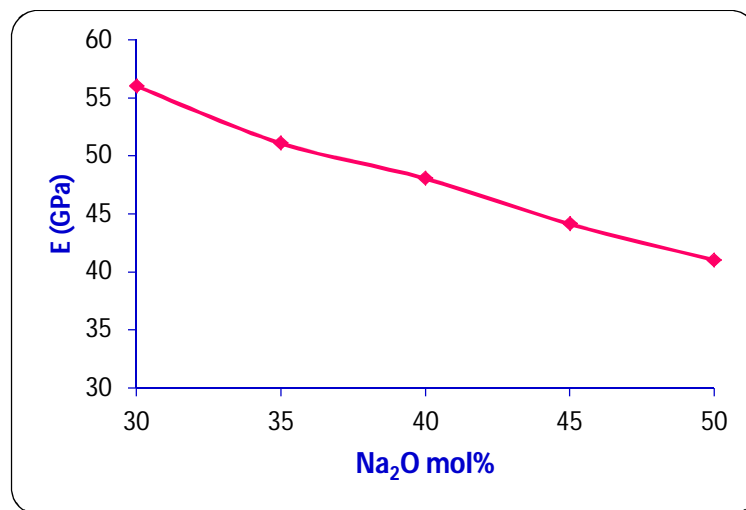


Fig. 1.9. Variation of Young's modulus with Na₂O mol%

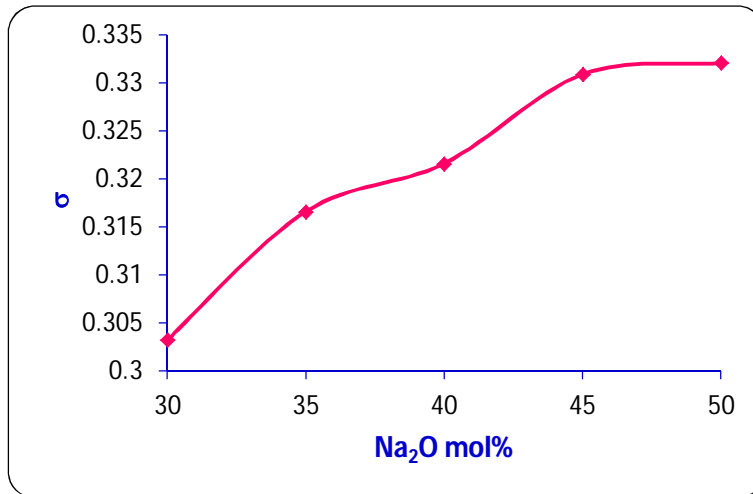


Fig. 1.10. Variation of Poisson's ratio with Na₂O mol%

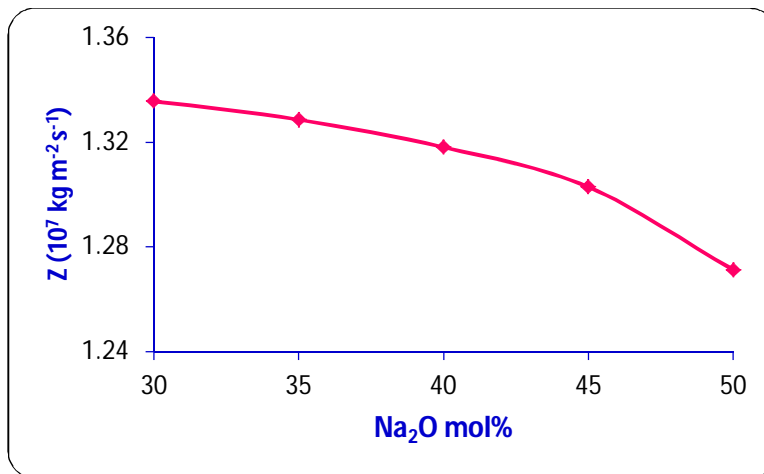


Fig. 3.11. Variation of acoustical impedance with Na₂O mol%

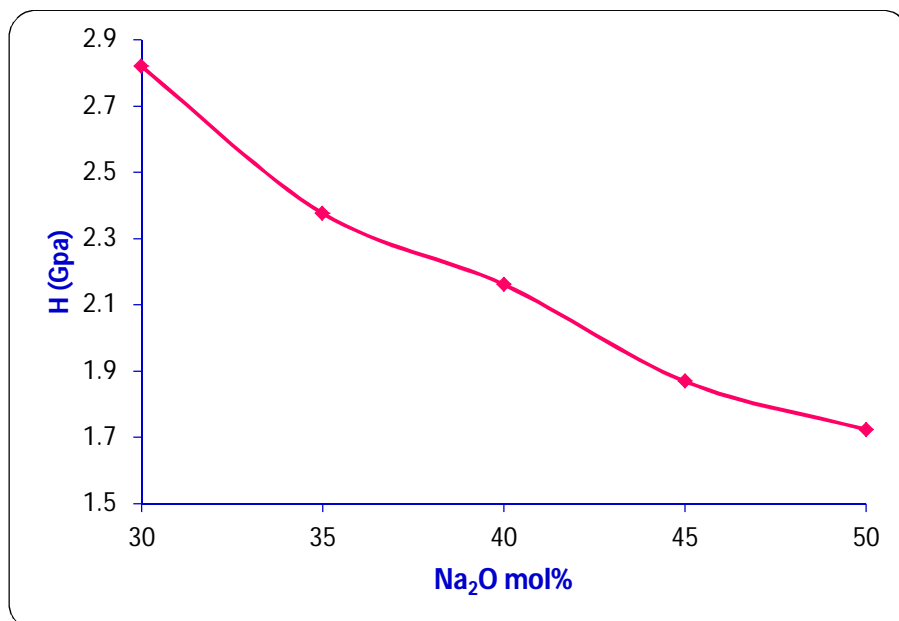


Fig. 1.12. Variation of microhardness with Na₂O mol%

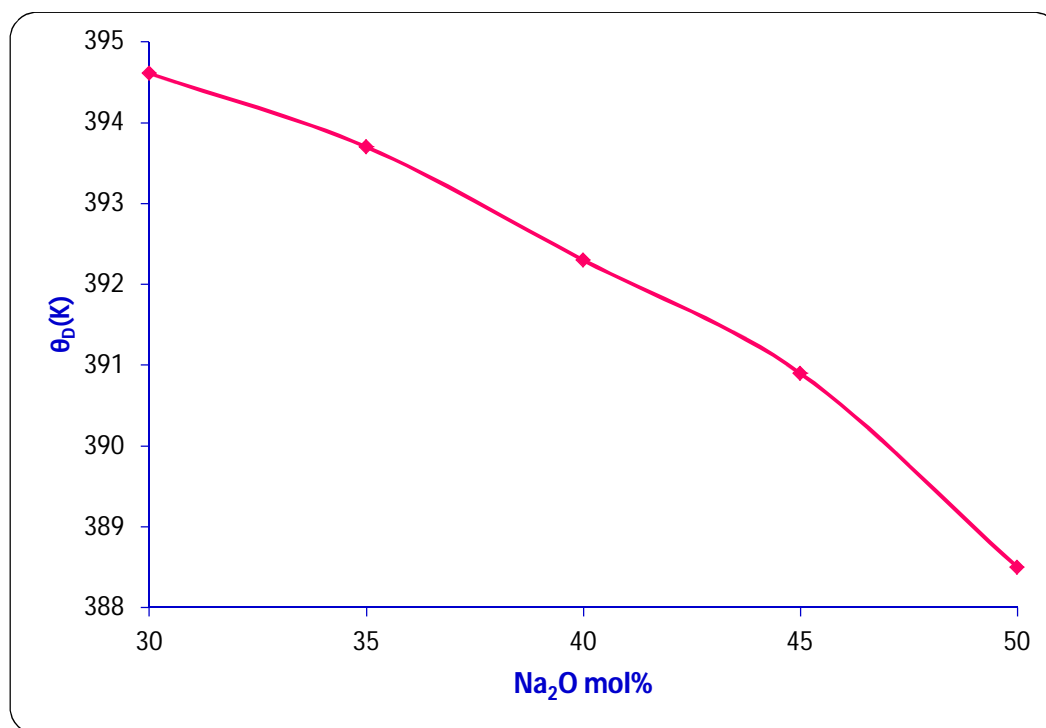


Fig. 1.13. Variation of Debye temperature with Na₂O mol%

Figs.1.11 and 1.12 show the variation of acoustic impedance and microhardness of the glasses. The values of acoustic impedance indicate that the addition of Na₂O to B₂O₃ causes the splitting of the glass network, thereby increasing the formation of NBO atoms resulting in lower impedance to the propagation of ultrasonic waves in the specimen (16). This is further supported by the evaluated microhardness.

The Debye temperature is an important parameter of solid materials in the determination of elastic moduli and atomic vibration. θ_D represents the temperature at which all the high frequency lattice vibrations modes are excited. It is known that Debye temperature depends directly on the mean ultrasonic wave velocity (17). The observed decrease in Debye temperature is mainly attributed to the change in the atoms per unit volume and also to the existence of non-bridging oxygen as a direct effect of the insertion of Na₂O into the borate network structure.

V. CONCLUSION

The density, ultrasonic velocities and elastic properties of binary BN glasses have revealed the following conclusions:

- A. The density values of the glass system studied were found to have increased, while the molar volume decreased.
- B. The velocities, elastic moduli and remaining parameters decrease and the Poisson's ratio increases as the concentration of Na₂O content is increased. This indicates that the glass network becomes loosely packed and the degree of disorder increases with the increase in the concentration of Na₂O content.
- C. X-Ray diffraction pattern shows the amorphous behavior of the prepared samples.

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