

## STUDY ON ULTRASONIC VELOCITY AND ELASTIC PROPERTIES OF $\gamma$ -RADIATED BORATE GLASSES

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**Abstract.** Longitudinal and shear ultrasonic velocities were measured in different compositions of sodium borate glass samples before and after irradiation with  $\gamma$ -rays. Measurements were carried out at room temperature and at 4 MHz frequency using pulse echo technique. Densities of the glass samples were measured using displacement method while toluene was used as immersion liquid. Elastic constants were calculated as well as Debye temperature, softening temperatures and microhardness for all glass samples. Results showed that the effect of radiating sodium borate glass with  $\gamma$ -radiation had no noticeable change on ultrasonic velocity. However, the effect of increasing sodium oxide content on ultrasonic velocity was very clear. The increase in velocity was attributed to the gradual increase in the rigidity of the glass and hence strengthening of the network due to the gradual change of boron atoms from the three-fold to the four-fold coordination of oxygen atoms.

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### 1. Introduction

The largest most important group of glasses are the inorganic oxides and their mixtures. They are commonly used because of their diverse applications in the fields of electronics, nuclear and solar energy technologies, and acousto-optic devices.

The propagation of ultrasonic waves in solids provides important information regarding the solid state motion in materials. The velocity of ultrasonic waves and hence the elastic properties are particularly suitable parameters for characterizing glasses as a function of their composition, because they give some

information about both the microstructure and the elastic properties through the behaviour of the network and the modifier.

Glassy materials as well as some organic ones, mainly polymers, have been increasingly tested for dosimetry purposes. They also were successfully used as detectors for different kinds of radiation [1].  $\gamma$ -radiation causes changes in the physical properties of materials. The changes are strongly dependent on the internal structure of absorbed substances. As a result a displacement will occur of the orbital electrons and possibly atoms within the structure of substance. Friebel et al [2] have found that when alkali borate, silica, or phosphate glasses are irradiated, at room temperature, the most prominently induced electro-magnetic resonance signal is the one associated with holes trapped on bridging or non bridging oxygen.

Sodium borate glasses are composed of the network former  $B_2O_3$  and the modifier  $Na_2O$ . According to Bisco and Warren [3], pure  $B_2O_3$  exists in planar three fold coordination. The addition of net-work modifier ions up to about 15 % produces fourfold coordinated borons by cross linking the planar triangle and will be expected to tighten and strengthen the network. However, the results of the ultrasonic velocity measurements of Gladkov and Tarasov [4] have indicated that the strengthening of the network continues up to at least 35 mol %  $Na_2O$ .

Krause and Kurkjian [5] have used the ultrasonic pulse superposition method to study the velocity of sound waves in sodium borate glasses. On the other hand, Kodama [6] studied the relationship between the elasticity and the structure of sodium borate glasses. He showed the way in which the network former and the modifier contributed to the elasticity of these glasses. Ultrasonic velocities were measured as a function of composition, from which elastic resistances of the network former and the modifier were obtained on the basis of the theory of elastic internal energy [7].

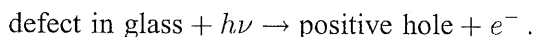
Sidkey et al [8] determined the ultrasonic velocity in samples of sodium borate glasses containing  $Na_2O$  between 5.6 and 27.2 mol. Boron anomaly was studied and results showed that this anomaly should appear at compositions above 28 mol % sodium oxide.

As indicated before,  $\gamma$ -ray radiation causes changes in the physical and chemical properties of the solid materials after passing through them. These changes are strongly dependent on the interaction mechanism of  $\gamma$ -rays with materials. The interaction mechanism of  $\gamma$ -rays with glass samples mainly occurs by means of electronic excitation, electronic ionization, and by primarily atomic displacement of orbital electrons.

Electrical measurements on telluride phosphate glasses were carried out by Abd El-Kader et al [9] to study the effect of  $\gamma$ -irradiation on these glasses. El Mallowany et al [10] also studied  $\gamma$ -irradiation effect on the ultrasonic

attenuation in telluride glasses. They concluded that in radiating the glass with  $\gamma$ -rays increased both ultrasonic absorption  $\alpha$  and internal friction  $Q^{-1}$ .

The defects produced in glass by irradiation could be represented by the general equation



Hussein et al [1] studied the dc electrical conductivity of the binary  $\gamma$ -irradiated  $\text{MoO}_3\text{P}_2\text{O}_5$  glass to investigate the validity of using such glass samples as  $\gamma$ -ray dosimeters. They found that the dc electrical conductivity has proved to be dose dependent, which showed a decrease in the former with increasing  $\gamma$ -dose. The induced change in dc measurements was found to be essentially related to the energy absorbed in the glass samples due to the exposure to the radiation.

Sanad et al [11] studied the effect of the duration of heat treatment and irradiation on the electrical conductivity and crystallization of barium borate glass containing iron. They found a decrease in conductivity with the increase in time of heat treatment due to a change in the structure of samples. A decrease in conductivity as the time of irradiation increased up to 18 h, and they attributed that to the formation of compact structure. At higher doses of irradiation the increase in conductivity was attributed to the breakage of the B–O bond formed with non-bridging oxygen.

## 2. Experimental

A series of sodium borate glass samples were selected according to the following formula  $X_2\text{Na}_2\text{O}-X_1\text{B}_2\text{O}_3$  where  $X_2 = 2, 5, 10.4, 13.2, 18, 20, 22$  and 27%. The starting materials of sodium carbonate and boric acid were mixed together and calculated to give 30 g. Conversion from mol % to weight fraction was calculated by applying the following relation:

$$W_a = \frac{X_a Z_a}{\sum_{i=1}^n X_i Z_i}$$

where  $W_a$ ,  $X_a$  and  $Z_a$  are the weight fraction, the mol %, the atomic weight of the oxide, respectively, and  $n$  being the number of oxides incorporated in the glass composition.

As for the weight of  $\text{Na}_2\text{CO}_3$  ( $Z_1$ ), this was calculated by applying the relation  $Z = W \frac{M_2}{M_1}$ , where  $M_1$  and  $M_2$  are the molecular weights of sodium oxide and sodium carbonate respectively.

The specified weights of the constituent materials were mixed together and fused at 400 °C in a silica crucible. After complete fusion, the temperature of the muffle furnace was raised to 900 °C and left for two hours in order to improve homogeneity. Occasional stirring of the melt, using a thin silica rod, was carried out before casting. The temperature of the furnace was lowered to 350 °C and left for 30 min. The melt was then poured into a split mold made of mild steel.

The mold which was preheated to 450 °C was held to rest for one hour. Annealing was made at 300 °C and glass samples were allowed to cool to reach room temperature 25 °C. The obtained samples were polished with 0.3  $\mu\text{m}$  alumina powder using precision polishing machine. Two opposite sides of each glass sample were polished to obtain optically flat and mutually parallel faces.

The densities of the glass samples were measured to the third decimal by the displacement method using toluene as immersion liquid.

The ultrasonic velocities were measured at a frequency of 4 MHz, using pulse echo-technique. The elapsed time between the initiation and the receipt of the pulse appearing on the screen of a flaw detector (USM3 Kraut-Kramer) was measured by standard electronic circuit PM 3055 Philips. The ultrasonic velocity was obtained by dividing the round trip distance by the elapsed time. The transducer was coupled to the specimen by an ultrasonic couplant which proved to be very suitable for this purpose. The glass samples were irradiated using  $^{137}\text{Cs}$  gamma ray source at a dose rate of  $24 \times 10^{-2}$  Gy/min at 30 cm from the source at room temperature. The different doses of irradiation were achieved by exposing the sample to the source for different periods of time. The used  $\gamma$ -ray doses are: 0.5, 1, 3, 5, 7 and 10 Gray.

### 3. Results and Discussion

Table 1 compiles the basic data obtained with the present series of experiments. It contains values for: density  $\rho$ , shear velocity  $V_s$ , longitudinal elastic modulus  $L$ , shear elastic modulus  $G$ , Debye temperature  $\theta_D$  and softening temperature  $T_s$ , for five sodium borate glass samples having  $\text{Na}_2\text{O}$  content of 2.0, 5.0, 10.4, 20.0, and 27.0 mol% at different  $\gamma$ -radiation doses. It is quite clear from this table that the effect of irradiating sodium borate glasses with  $\gamma$ -radiation on the physical properties of these glasses is very weak. Very slight changes were observed. However, for glass sample with 10.4 mol%  $\text{Na}_2\text{O}$ , the remarkable increase in ultrasonic velocity and other physical properties is significant.

Figure 1 shows the relation between longitudinal ultrasonic velocity  $V_{||}$  and irradiation dose, in the range from 0 to 10 Gray. It is observed that, the rate of change of ultrasonic velocity with respect to irradiation dose is more noticeable in the glass sample with 10.4 mol%  $\text{Na}_2\text{O}$  than the other glass samples. The

slope of the straight line representing this relation is 65.111 while for the other glass samples with 2.0, 5.0, 20.0 and 27.0 mol %  $\text{Na}_2\text{O}$  the slopes of the straight lines are 33.036, 26.471, 29.848, and 17.821 respectively.

**Table 1**

Dose (Gray)	$\text{Na}_2\text{O}$ (mol %)	Density $\rho$ ( $\text{kg/m}^3$ )	Shear velocity $V_s$ (m/s)	Long. elastic modulus (GPa)	Shear elastic modulus (GPa)	Debye temp. $\theta_D$ (K)	Softening temp. $T_s$ (K)
0	2	1887	2222.85	27.718	9.323	318.14	268.83
	5	1950	2358.86	32.261	10.854	340.36	305.425
	10.4	2060	2610	41.711	14.032	381.41	380.026
	20	2217	2999.8	59.307	19.954	444.79	517.463
	27	2293	3268.9	72.852	24.561	486.38	628.731
0.5	2		2325.8	30.337	10.207	332.88	294.319
	5		2369.9	32.56	10.95	341.308	308.245
	10.4		2641.32	42.72	14.37	384.928	388.714
	20		3049.06	61.272	20.611	452.06	534.505
	27		3282.2	73.422	24.696	488.31	633.742
1	2		2373.4	31.59	10.624	339.61	306.32
	5		2376.55	32.754	11.02	342.95	310.104
	10.4		2668	43.59	14.66	389.717	397.093
	20		3073.16	62.325	20.964	455.91	543.66
	27		3304.84	74.411	25.043	491.72	642.655
3	2		2397.7	32.242	10.849	343.18	312.822
	5		2431.36	34.274	11.526	350.75	324.353
	10.4		2717.3	45.21	15.21	397.09	411.9035
	20		3105.9	63.58	21.39	460.495	554.63
	27		3316.4	74.961	25.21	493.37	646.94
5	2		2418.6	32.806	11.04	346.18	318.324
	5		2465	35.23	11.85	355.656	333.48
	10.4		2818.22	48.632	16.357	411.81	443.01
	20		3123.9	64.316	21.638	463.17	561.124
	27		3328.04	75.486	25.393	495.15	651.631
7	2		2447.6	33.597	11.306	350.34	326.003
	5		2485.9	35.83	12.05	358.6718	361.278
	10.4		2866.4	50.31	16.92	419.05	457.78
	20		3139.54	64.962	21.86	465.54	566.887
	27		3348.9	76.436	25.714	498.28	659.881
10	2		2471.14	34.253	11.519	353.64	332.157
	5		2502.7	36.315	12.219	361.13	343.851
	10.4		3002.66	55.205	18.575	438.83	507.086
	20		3210.3	67.924	22.845	475.93	592.444
	27		3384.88	78.087	26.255	503.5	673.745

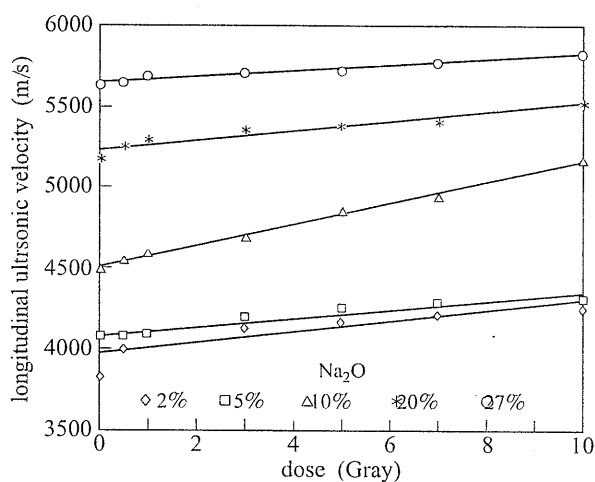


Fig. 1. Relation between longitudinal ultrasonic velocity and irradiation dose

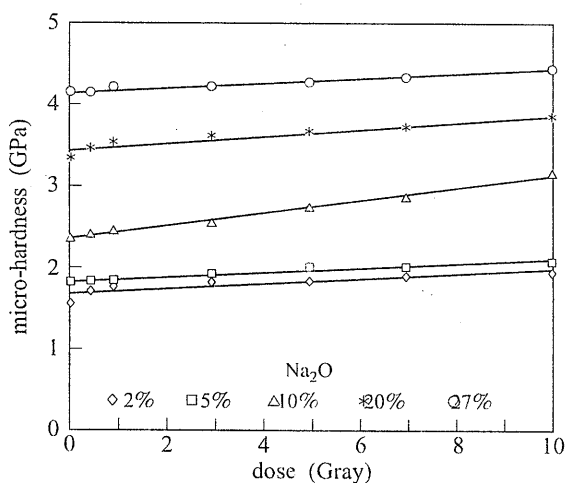


Fig. 2. Relation between microhardness  $H$  and gamma irradiation dose

Figure 2 shows the relation between microhardness  $H$  and  $\gamma$ -irradiation dose for five glass samples with Na<sub>2</sub>O contents of 2.0, 5.0, 10.4, 20.0, and 27.0 mol%. Microhardness was calculated according to the equation given by Kodama [7] in the form

$$H = (1 - 2\sigma)Y/6(1 + \sigma)$$

where  $\sigma$  is the Poisson's ratio, and  $Y$  is the Young's modulus. The figure shows the same futures as those observed in Fig. 1. The relations between

Young's and bulk modulus of elasticity and  $\gamma$ -irradiation dose are shown in Figs 3 and 4.

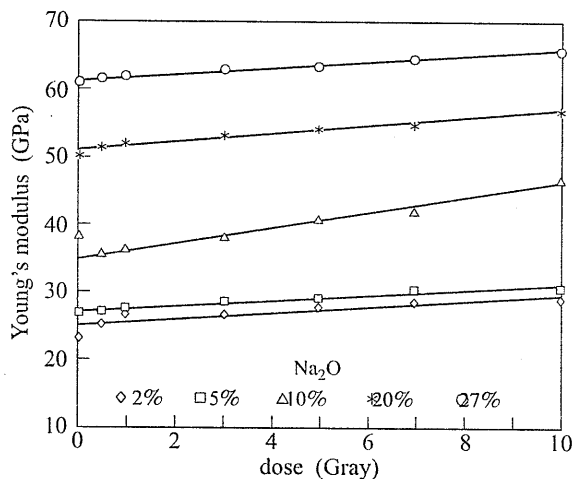


Fig. 3. Relation between Young's modulus and gamma irradiation dose

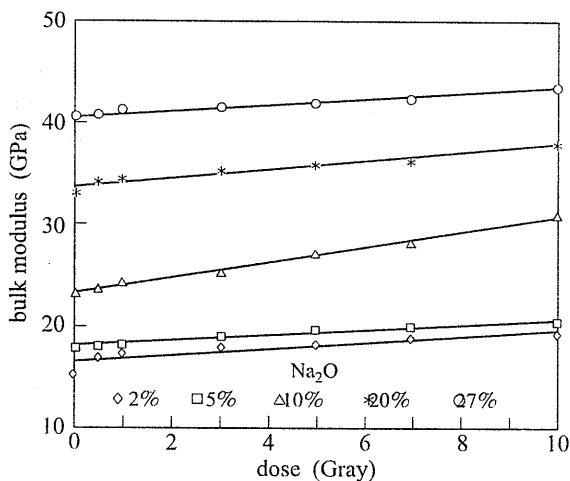


Fig. 4. Relation between bulks modulus and gamma irradiation dose

Again, the rate of change of Young's or bulk modulus with respect to irradiation dose is more noticeable in the glass sample with 10.4 mol % Na<sub>2</sub>O content. Figures 3 and 4 show the same characteristics like Figs 1 and 2.

Values of elastic moduli given in Table 1 were calculated according to the

well-known equations:

$$\begin{array}{ll}
 \text{longitudinal modulus} & L = \rho V_{\parallel}^2 \\
 \text{shear modulus} & G = \rho V_s^2 \\
 \text{bulk modulus} & K = L - \frac{4}{3}G \\
 \text{Young's modulus} & Y = \frac{G(3L - 4G)}{L - G}
 \end{array}$$

Debye temperature  $\theta_D$  was calculated according to the equation given in [8]

$$\theta_D = 251.2 V_m \frac{(nM)^{1/3}}{\rho}$$

where  $V_m$  is the mean ultrasonic velocity given by

$$\frac{3}{V_m^3} = \frac{1}{V_{\parallel}^2} + \frac{2}{V_s^2}$$

Here  $n$  is the number of atoms in the chemical formula,  $M$  is the effective molecular weight, and  $\rho$  is the density of the glass. Debye temperature represents the temperature at which all modes of vibrations in solid are excited and its increase implies an increase in rigidity of the network and hence an increase in velocity. The variation of Debye temperature with irradiation dose (Table 1) shows an increase in  $\theta_D$  for 10.4 mol % Na<sub>2</sub>O, which is relatively more than in the other glass samples. This increase is possibly due to structural change in the network.

The observed increase in ultrasonic velocity and all other physical properties of 10.4 mol % Na<sub>2</sub>O content glass sample is not due to the  $\gamma$ -irradiation effect, but merely related to structure changes of the network. According to Bisco and Warren [3], pure B<sub>2</sub>O<sub>3</sub> exists in planar three-fold coordination. The addition of network modifier (Na<sub>2</sub>O ions) produces four-fold coordinated boron by cross linking the planar triangles and will be expected to tighten and strengthen the network. Moreover, below 10.4 mol % Na<sub>2</sub>O content, the boron atoms with coordination number three predominate and make B<sub>2</sub>O<sub>3</sub> soft and easily deformed by stress. Above 10.4 mol % Na<sub>2</sub>O content, the boron atoms begin to change from three-fold to four-fold with coordination number of 4 [12]. This tetrahedral structure gives B<sub>2</sub>O<sub>3</sub> maximum rigidity and increases in velocity indicating that the network former becomes rigid and resists deformation.

Moreover, it has been reported that [13], with increasing Na<sub>2</sub>O content, the first incorporation of alkali oxide leads to the coordination shift [BO<sub>3</sub>] → [BO<sub>4</sub>] with alkali ions adjoining the [BO<sub>4</sub>] tetrahedron. The structure is thus strengthened and consequently the velocity increases. This is because the number of

points of linkage of polyhedrons rises from three to four. This is often located around 15 mol%  $\text{Na}_2\text{O}$  content (10.4 in our study). Therefore, at this concentration "critical composition" alkali borate glasses often exhibit borate anomaly.

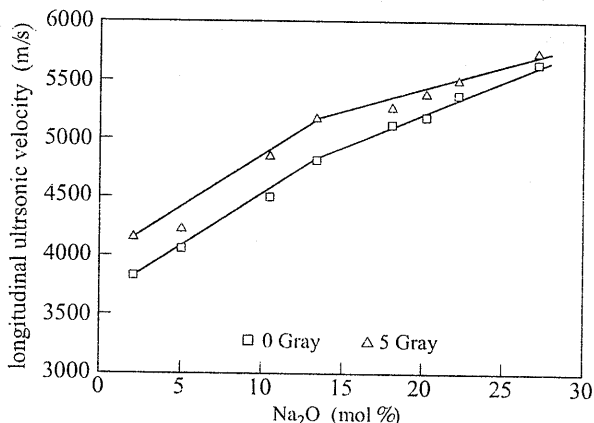


Fig. 5. Relation between longitudinal ultrasonic velocity and sodium oxide content

Figure 5 depicts the relation between longitudinal ultrasonic velocity and sodium oxide content, for irradiated and nonirradiated two glass samples. It can be seen from the figure that the ultrasonic velocity increases rapidly with increasing  $\text{Na}_2\text{O}$  mol % content up to about 10.4 mol %  $\text{Na}_2\text{O}$ . Above 10.4 mol %  $\text{Na}_2\text{O}$  to 27.0 mol %  $\text{Na}_2\text{O}$ , a gradual increase in velocity is observed. The plot can be divided into two segments having different slopes. However, the gradual increase in ultrasonic velocity indicates the gradual increase in rigidity and strengthening of the network continues up to 27.0 mol %  $\text{Na}_2\text{O}$ . The results obtained in this study are in good agreement with the measurements of Gladkov and Tarasov [4], Kraus and Kurkjian [5], and Lorosch et al [14]. The increase in rigidity of glass with the addition of  $\text{Na}_2\text{O}$  is related to the change of the structural unit forming the network. In the composition range 2.0 and 10.4 mol %  $\text{Na}_2\text{O}$  (first segment in Fig. 5), boron atoms change from the three-fold to the four-fold coordination of oxygen atoms by addition of modifier to form a three-dimensional connection of the network.

This means that the glasses in this composition range are composed of soft  $\text{B}_2\text{O}_3$  and hard  $\text{Na}_2\text{O}$ . This indicates that the modifier is enclosed within the network in such a way that the modifier prevents the deformation of the surrounding soft network.

At the composition range  $\text{Na}_2\text{O} = 10.4$  mol % (Critical composition), alkali borate glasses often exhibit borate anomaly near this composition range [15].

The fact that the elastic modulus of  $B_2O_3$  is maximal at this composition indicates that the borate network becomes rigid to high degree. Around 10.4 mol %  $Na_2O$ , the three different species of boroxol ring, tetraborate group and diborate group exist in large quantities at the same time [16]. It seems therefore that a combination of these borate groups forms the rigid network. Consequently, the degree to which the modifier prevents the deformation of the surrounding rigid network decreases.

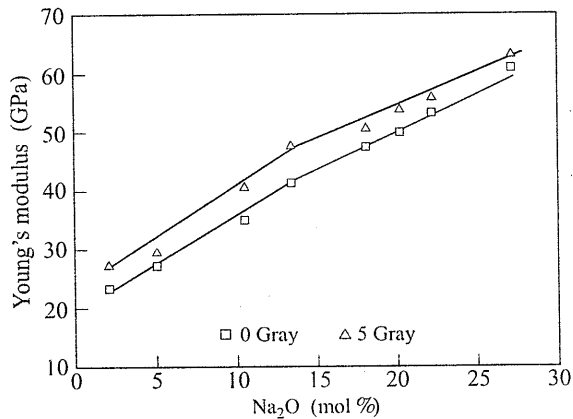


Fig. 6. Relation between Young's modulus and sodium oxide content

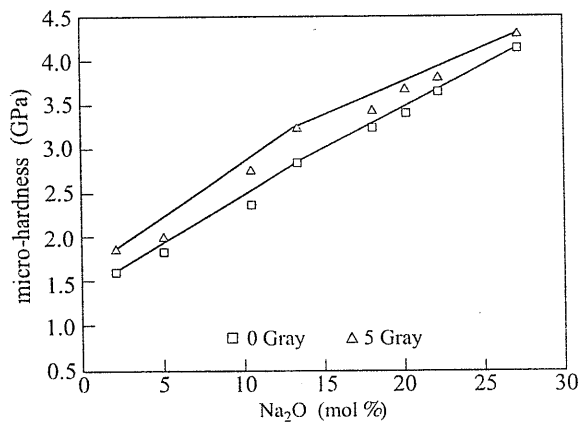


Fig. 7. Relation between micro-hardness and sodium oxide content

In the composition range 10.4 and 27.0 mol %  $Na_2O$ , the boron atoms continue changing from the three-fold to the four-fold coordination of oxygen atoms. Replacing  $B_2O_3$  (coordination number  $n_f = 3$ , and cross-link density  $n_c = 1$ ) by  $Na_2O$ , (coordination number  $n_f = 6$  and cross-link density  $n_c = 4$ ),

the structure will be more linked and the resistance to the deformation by stress will be more strong. This leads to the increase in glass rigidity as evidenced by the increase in glass velocity.

Glasses are considered as elastic substances and thus can be characterized through a modulus of elasticity. This modulus increases as the lengthening at a certain applied stress diminishes. That will be the case if the glass structure is rigid and therefore contains the fewest possible nonbridging oxygen. When an alkali is introduced to  $B_2O_3$ , the strengthen of the structure depends on the field strength of the cation.

The relatively open structure of  $B_2O_3$  glass makes its modulus of elasticity low. With increasing  $Na_2O$  content of the binary borate glass, the structure becomes more rigid and the modulus of elasticity increases. This increase is very pronounced and according to Takahashi [17] reaches the maximum value when the number of B atoms is the coordination number 4 has its maximum.

The relations between Young's modulus and microhardness and  $Na_2O$  mol % content are shown in Figs 6 and 7 respectively. One can notice that the rate of change of Young's modulus or microhardness with respect to  $Na_2O$  content is not constant, but two different slopes can be observed in these plots. The first in the range from 2-10.4 mol %  $Na_2O$  and the second is in the range from 10.4 to 27.0 mol %  $Na_2O$ .

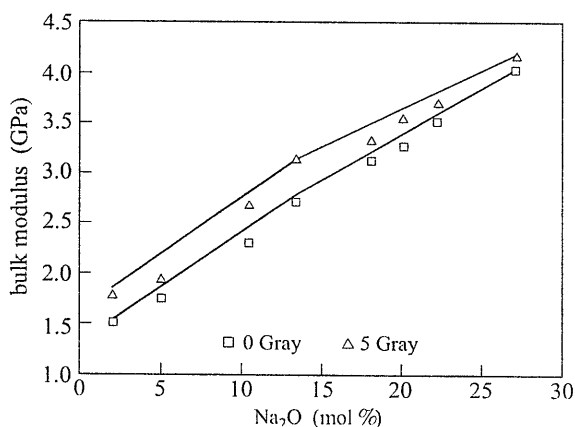


Fig. 8. Relation between bulk modulus and sodium oxide content

Bulk modulus is the elastic property of glasses which can be derived most easily from the glass structure.  $B_2O_3$  possesses an open structure with many open spaces. The addition of  $Na_2O$  will occupy spaces by these cations, which should lead to an increase in bulk modulus. With low  $Na_2O$  contents, the bulk modulus is small since many open spaces are present which will be filled most quickly by the large  $Na^+$  ions (ion radius of Na is 0.098 nm) so that the bulk

modulus increases. With higher Na<sub>2</sub>O contents, the deformability of the cations becomes decisive. This is clear from Fig. 8 which shows the relation between bulk modulus and Na<sub>2</sub>O mol %.

**Table 2**

Na <sub>2</sub> O (mol %)	Density (kg/m <sup>3</sup> )	Shear ultrasonic velocity (m/s)	Longitudinal elastic modulus (GPa)	Shear elastic modulus (GPa)	Debye temp. $\theta_D$ (K)	Softening temp. $T_s$ (K)
$\gamma$ -irradiation dose 0 Gy						
2	1887	2222.85	27.718	9.323	318.14	268.83
5	1950	2358.86	32.261	10.854	340.36	305.425
10.4	2060	2610	41.711	14.032	381.41	380.026
13.2	2127	2795.6	49.422	16.63	411.81	439.905
18	2178	2961.48	56.788	19.11	437.5	501.211
20	2217	2999.76	59.307	19.954	444.79	517.463
22	2218	3108.22	63.698	21.425	459.87	559.006
27	2293	3268.8	72.852	24.501	486.38	628.731
$\gamma$ -irradiation dose 5 Gy						
2	1887	2418.6	32.806	11.04	346.18	318.324
5	1950	2465	35.23	11.85	355.656	333.48
10.4	2060	2818.22	48.632	16.357	411.81	443.01
13.2	2127	2999.53	56.904	19.146	441.86	506.439
18	2178	3054.3	60.403	20.316	451.1	532.83
20	2217	3123.9	64.316	21.638	463.17	561.124
22	2218	3185.4	66.899	22.5	471.27	587.047
27	2293	3328.04	75.486	25.393	495.15	651.631
$\gamma$ -irradiation dose 10 Gy						
2	1887	2471.14	34.253	11.519	353.64	332.157
5	1950	2502.7	36.315	12.219	361.13	343.851
10.4	2060	3002.66	55.205	18.575	438.83	503.086
13.2	2127	3051.96	58.902	19.815	449.52	524.148
18	2178	3103.26	63.446	21.34	462.33	559.679
20	2217	3210.3	67.924	22.845	475.93	592.444
22	2218	3290.92	71.407	24.022	486.95	626.773
27	2293	3384.9	78.087	26.255	503.5	673.745

Table 2 gives values of density  $\rho$  of the glass samples, shear velocity  $V_s$ , shear modulus  $G$ , longitudinal modulus  $L$ , Debye temperature  $\theta_D$ . Softening temperature  $T_s$  was calculated from the expression given by Anderson [18] as

$$T_s = \frac{V_s^2 M}{C^2 n}$$

Here  $V_s$  is the shear velocity,  $M$  is the effective molecular weight,  $C$  is a con-

stant of proportionality and equals to  $0.5074 \times 10^5 \text{ cm s}^{-1} \text{ K}^{1/2}$ . The softening temperature is the temperature at which viscous flow changes to plastic flow and is related to microhardness.

The greater microhardness implies great softening temperature. It can be seen from Table 2 that as the  $\text{Na}_2\text{O}$  increases, the physical parameters increase.

#### 4. Conclusion

The results obtained from the study under report lead to the following conclusions.

1. Sodium borate glasses irradiated with  $\gamma$ -radiation showed no noticeable changes in ultrasonic velocity.
2. The effect of increasing sodium oxide content on ultrasonic velocity is very clear. From 2.0 mol %  $\text{Na}_2\text{O}$  to 10.4 mol %  $\text{Na}_2\text{O}$ , the increase in ultrasonic velocity is gradual due to the gradual increase in the rigidity of the glass and hence the strengthening of the network due to the gradual change of boron atoms from the three fold to the four fold coordination of oxygen atoms.
3. At the composition range  $\text{Na}_2\text{O} = 10.4 \text{ mol } \%$  (critical composition) three different species of boroxol ring, tetraborate group, and diborate group exist in large quantities at the same time.
4. In the composition range 10.4 and 27.0 mol %  $\text{Na}_2\text{O}$  the boron atoms continue changing from the three fold to the four fold coordination of oxygen atoms and ultrasonic velocity increases as the rigidity of the glass is increased.

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