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Structural and elastic properties of Ni^{2+} and W^{6+} transition metal ions doped with tellurite barium borate glasses using pulser – receiver technique

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ABSTRACT

Longitudinal, shear ultrasonic velocities and attenuation were measured in different compositions of the glass systems of $10TeO_2.15BaO.(75\text{-}x)B_2O_3.xNiO$ and $10TeO_2.15BaO.(75\text{-}x)B_2O_3.xWO_3$ (where x= 0 to 1 mol% in steps of 0.2) at room temperature by using pulser - receiver technique at 5 MHz. The glass samples were prepared by conventional melt-quenching method. The amorphous nature of the samples were ascertained using X-ray diffractometry (XRD). The density of the glass samples were measured by relative measurement method. The measured experimental values are utilized to evaluate elastic moduli, Poisson's ratio, acoustic impedance, internalfriction, microhardness, Debye temperature, and thermal expansion coefficient. Trends of the co-ordination number, cross-link density, mechanical and thermal stability for the systems are discussed in terms of the structural changes taking place due to variations in composition.

Introduction

The values of ultrasonic waves in the glasses as well as the elastic properties are practically very useful for describing glasses as a function of composition since they give information about the microstructure and all the dynamics of glass. Moreover, the elastic properties are related to microscopic properties through the behavior of the network and the modifier[1]. Ultrasonic pulse-echo technique is one of the most useful experimental tool for structural studies of glasses. The ultrasonic attenuation and elastic moduli as a function of composition and temperature are directly related to the glass structure, thus, their measurements allow evaluating the influence of modifiers on glass nature[2,3]. Studies on the elastic moduli of the glasses have given considerable information about their structure, since the elastic moduli are directly related to interatomic forces and potentials, i.e. they will be affected by the rigidity of the structure. The elastic moduli of glasses are influenced by many physical parameters, which is turn can be studied by measuring the ultrasonic velocities[4]. Glasses being isotropic and have only two independent elastic constant: longitudinal and shear elastic moduli. These two parameters are obtained from the longitudinal and shear velocities and density of the glasses. The elastic constants could be deduced[5]. Tellurite based glasses have received much attention in recent years and are considered as potential candidates for non-linear optical devices such as ultra-fast optical switches, power limiters, optical fibres and storage devicesdue to their superior properties viz., high temperature, high refractive index, high chemical durability, high electrical conductivity, low melting temperature, moisture resistant and non-linear optical properties such as second harmonic generation (SGH)[6]. B_2O_3 is one of the most important glass formers incorporated into various kinds of glass systems as a flux material, in order to attain materials with specific physical and chemical properties suitable for high-technological applications[7]. In borate glasses, B_2O_3 is a basic glass former because of its higher bond strength, lower cation size and smaller heat of fusion. Therefore, the structural investigations of boron in these glasses are one of the most attractive points of borate glass formation and related doped systems. In the borate glasses, B^{3+} ions are triangularly coordinated by oxygen atoms and triangle units are corner bonded in a random configuration[8]. Glasses containing barium are also proved to be good radiation shielders[9]. Barium is a good candidate for development of Ba - based radiation shielding glass owing to strong absorption of x-rays, gamma-rays and non-toxicity[10]. In recent years transition metal oxides represent a large family of materials possessing various interesting properties, such as superconductivity, magneto resistance, piezoelectricity solid-state layers luminescent solar energy concentrators (LSCS) and fiber optic communication devices. Among them, tungsten oxide (WO_3) is of intense interest and has been investigated extensively for its distinctive properties. With outstanding electrochromic, photo chromic, gas chromic, gas sensors, photo – catalyst and photoluminescence properties, tungsten oxide has been used to construct "smart window", anti-glare rare view mirrors for automobiles, non-emissive displays, optical recording devices, solid state gas sensors, humidity and temperature sensors, biosensors, photonic crystals and so forth[11]. The divalent nickel ion is an interesting paramagnetic ion to probe in the glass systems. Nickel ions are reported to occupy both tetrahedral and octahedral positions in the glass matrices[12]. The main objectives of the present work are to study elastic property of the tellurite barium borate glasses based on experimental measurements and also to investigate the structural modification of the tellurite barium borate glass network induced by the introduction of NiO and WO_3.

Materials and Methods

The glass samples of the formula$10TeO_2.15BaO.(75\text{-}x)B_2O_3.xNiO$ and $10TeO_2.15BaO.(75\text{-}x)B_2O_3.xWO_3$ where x= 0 to 1 in steps 0.2 mol % have been prepared by the conventional melt-quenching technique. Required quantities of analytical grade of TeO_2, $BaCo_3$, H_3Bo_3, NiO and WO_3 were obtained from E-merck, Germany and Sd-Fine chemicals, India. The proper compositions were mixed together by grinding the

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mixture repeatedly to obtain a fine powder. The mixture is melted in alumina crucible at about 1113 K for about 45 minutes to homogenize the melt. The melt was quickly quenched by pouring on to a copper plate and covering with another plate and the random pieces of samples thus formed were collected. Then the glass samples were annealed at 573K for two hours to avoid the mechanical strains developed during the quenching process. The samples prepared were chemically stable and non-hygroscopic. The prepared glass samples were polished and the surfaces are made perfectly plane and smoothened by diamond disc and diamond powder. Thickness of the glass samples are measured using digital vernier caliper (MITUTOYO DIGIMATIC CALIPER) with an accuracy of 0.0001 mm. The amorphous nature of glass samples was confirmed by X-ray diffraction technique using an x-ray diffractometer (Model : X' PERT POWDER XRD SYSTEM FROM PANALYTICAL). Density (ρ), at room temperature was measured by following Archimedes principle using a sensitive single pan digital balance (Model : SHIMADZU AX 200). The xylene was used as an immersion liquid. The ultrasonic wave (longitudinal and shear) velocities and attenuation of the glass specimens were measured using ultrasonic high energy pulser receiver (PANAMETRICS – 5800 PR) technique at room temperature by making use of X-cut and Y-cut transducers at 5 MHz.The attenuation coefficient α of the sample in neper per unit length is obtained from the relation

$$I = Ioe^{-\alpha t}$$

Where, t is the thickness of the sample, I_O and I are the ratios of amplitude of the two successive echoes.

Theory and Calculation

The elastic and thermal properties of the glass specimens were investigated at room temperature by using the measured values of density (ρ), longitudinal velocity (U_l), shear velocity (U_S) and attenuation (α).

(i) Longitudinal modulus (L)

$$L = \rho U_l^2 \tag{1}$$

(ii) Shear modulus (G)

$$G = \rho U_S^2 \tag{2}$$

(iii) Bulk modules (K)

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

(iv) Poisson's ratio (σ)

$$\sigma = \left(\frac{L-2G}{2(L-G)}\right) \tag{4}$$

(v) Young's modulus (E)

$$E = (1+\sigma)\, 2G \tag{5}$$

(vi) Acoustic impedance (z)

$$Z = U_l\rho \tag{6}$$

(vii) Internal friction (Q^{-1})

$$Q^{-1} = \frac{\alpha}{8.66\pi f U_l} \tag{7}$$

where, α - attenuation coefficient andf - frequency of the quartz crystal

(viii) Microhardness (H)

$$H = (1-2\sigma)\frac{E}{6(1+\sigma)} \tag{8}$$

(ix) Debye temperature (θ_D)

$$\theta_D = \frac{h}{k}\left(\frac{9N}{4\pi V_m}\right)^{1/3} U_m \tag{9}$$

where, h, k, N, V_m and U_m are the Planck's constant ($6.626\text{x}10^{-34}$JS), the Boltzmann's constant ($1.38\text{x}10^{-23}\text{JK}^{-1}$), the Avogadro's number ($6.023\text{x}10^{23}\text{mol}^{-1}$), the molar volume and mean sound velocity of the sample respectively where

$$U_m = \left[\frac{1}{3}\left(\frac{2}{U_S^3} + \frac{1}{U_l^3}\right)\right]^{-1/3}$$

(x) Thermal expansion coefficient (α_p)

$$\alpha_p = 23.2\,(U_l - 0.57457) \tag{10}$$

Result and discussion

The experimental values of density (ρ), longitudinal ultrasonic velocity (U_l), shear ultrasonic velocity (U_s), and attenuation (α) of the tellurite barium borate glasses with respect to the change in mol percentage of NiO and WO_3 used as network modifier (NWM) are listed in the Table 1. The calculated values of longitudinal modulus (L), shearmodulus (G), bulk modulus (K), Young's modulus (E), Poisson's ratio (σ), acoustic impedance (Z), internal friction (Q^{-1}), microhardness (H), Debye temperature (θ_D) and thermal expansion coefficient (α_p)are presented in the Tables 2-3. The X-ray diffraction patterns (Fig. 1) of the studied glass systems reveals the absence of any discrete or continuous sharp crystalline peaks, but show homogenous glassy characters. The density is an important measure of the glass; its value stands on its own as an intrinsic property capable of casting the light on the short-range structure. The changes in the composition of the glass depend upon the structural compactness, modification of the geometrical configurations, etc., in the glass network. Thus, the density seems to be clearly reflecting the underlying atomic arrangements in a quantitative manner and lends support to the ideas of Krogh-Moe[13,14].From Table 1, it is observed that the values of density decreases with increase in mol% of NiO and WO_3 glass systems. The density values of TBBW glass system are much higher than that of TBBN glasses. The structure of glass depends on the nature of the ions entering in the network and hence, the density of glass. Further, the decreasing nature of density with the addition of Ni^{2+} and $W^{6+}$was most likely due to the lower effective ionic radius of Ni^{2+} ions (69 pm) compared to W^{6+} ion (60 pm) leading to a decrease in the interatomic distance and hence a more compact network was formed[15]. Further, it is observed from the above table, the values of longitudinal velocity (U_l) and shear velocity (U_s) are decreased with increasing of NiO and WO_3 contents, but the rate of increase of U_l is greater than that of U_s. The large difference between U_l and U_s arises from volume effects. The change in volume due to compressions and expansions involved in longitudinal strain is pronounced while no change in volume is involved in shear strains. The decrease in ultrasonic velocity is because of the formation of non-bridging oxygen (NBO) which makes the glass soft[16]. The attenuation coefficient describes the total reduction in the intensity due to absorption of energy by the medium and the deflection of energy from the path of the beam by reflection, refraction and scattering.

Table 1. Composition, measured values of density (ρ), longitudinal velocity (U_l), shear velocity (U_s) and attenuation (α) of TBB, TBBN and TBBW glasses at room temperature

Sample Label	Composition (mol%)	Density ρ/(kg.m^{-3})	Ultrasonic Velocity U/(m.s^{-1})		Attenuation α/(nepers.unit length^{-1})
			Longitudinal (U_l)	Shear (U_s)	
TeO_2 – BaO–B_2O_3(TBE)					
TBE	15-10-75	1557.5	4605.38	2614.38	64.2
System 1 : TeO_2 – BaO–B_2O_3 –NiO(TBBN)					
TBBN 1	15-10-74.8-0.2	3369.7	4897.96	2620.09	24.28
TBBN2	15-10-74.6-0.4	2656.6	4705.88	2590.09	15.33
TBBN3	15-10-74.4-0.6	2195.9	4615.38	2542.37	15.23
TBBN4	15-10-74.2-0.8	2002.1	4597.70	2537.00	14.64
TBBN5	15-10-74.0-1.0	1268.4	4515.68	2531.65	8.48
System 2 : TeO_2 – BaO–B_2O_3 – WO_3(TBBW)					
TBBW 1	15-10-74.8-0.2	5282.5	4743.08	2616.38	58.88
TBBW 2	15-10-74.6-0.4	3794.7	4616.32	2580.65	44.73
TBBW 3	15-10-74.4-0.6	3640.2	4596.74	2537.65	32.89
TBBW 4	15-10-74.2-0.8	2487.5	4562.74	2520.65	29.83
TBBW 5	15-10-74.0-1.0	1939.5	4511.28	2510.46	25.93

Table 2.Values of elastic moduli and Poisson's ratio of TBB, TBBN and TBBW glasses at room temperature

Sample Label	Longitudinal Modulus L/(×10^{10} N.m^{-2})	Shear Modulus G/(× 10^{10}N.m^{-2})	Bulk Modulus K/(×10^{10} N.m^{-2})	Young's Modulus E/ (×10^{10} N.m^{-2})	Poisson's Ratio (σ)
TeO_2 – BaO–B_2O_3(TBE)					
TBE	3.3034	1.0645	1.8841	2.6874	0.2623
System 1 : TeO_2 – BaO–B_2O_3 – NiO(TBBN)					
TBBN 1	8.0839	2.3133	4.9995	6.0127	0.2996
TBBN2	5.8831	1.7822	3.5068	4.5721	0.2827
TBBN3	4.6776	1.4194	2.7851	3.6399	0.2822
TBBN4	4.2322	1.2886	2.5141	3.3017	0.2811
TBBN5	2.5864	0.8129	1.5025	2.0661	0.2708
System 2 : TeO_2 – BaO–B_2O_3 – WO_3(TBBW)					
TBBW 1	11.8839	3.6161	7.0624	9.2666	0.2813
TBBW 2	8.0867	2.5272	4.7171	6.4327	0.2727
TBBW 3	7.6918	2.3442	4.5662	6.0049	0.2808
TBBW 4	5.1786	1.5805	3.0713	4.0473	0.2804
TBBW 5	3.9472	1.2224	2.3173	3.1188	0.2757

Table 3.Values of acoustic impedance (Z), internal friction (Q^{-1}), microhardness (H_v),Debye temperature (θ_D) and thermal expansion coefficient (α_p) of TBB, TBBN and TBBW glasses at room temperature.

Sample Label	Z/(×10^7kg. $m^{-2}s^{-1}$)	(Q^{-1}) /(×10^{-11} dB.s^2m^{-1})	H_v/(×10^9 N.m^{-2})	θ_D/(K)	α_p/(×10^2 m.s^{-1})
TeO_2 – BaO–B_2O_3(TBE)					
TBE	0.7172	5.1239	1.6869	267.14	1068.31
System 1 : TeO_2 – BaO–B_2O_3 – NiO(TBBN)					
TBBN 1	1.6505	1.8221	3.0214	351.27	1136.19
TBBN2	1.2501	1.9738	2.5818	321.16	1091.63
TBBN3	0.9965	1.2129	2.0610	295.83	1070.63
TBBN4	0.9205	1.1704	1.8805	286.21	1066.53
TBBN5	0.5728	0.6905	1.2421	244.90	1047.50
System 2 : TeO_2 – BaO–B_2O_3 – WO_3(TBBW)					
TBBW 1	2.4783	4.5629	5.2723	407.43	1100.26
TBBW 2	1.7517	3.5615	3.8295	359.16	1070.85
TBBW 3	1.6737	2.6299	3.4257	348.14	1066.31
TBBW 4	1.1350	2.3587	2.3138	304.25	1058.42
TBBW 5	0.8750	2.1127	1.8279	278.44	1046.48

The measurement of attenuation of ultrasonic waves in the glass system is the loss of energy during the propagation in the medium[17]. Table 1 shows the values of attenuation decreases with increasing of NiO and WO_3 concentrations which confirms the strengthening nature of these glasses as suggested from the composition dependence ultrasonic velocities[18].The decrease in the ultrasonic attenuation α may be attributed to the decrease of the Ni-O and W-O ionic bonds. This is probably attributed to the increases in the cross-link density in glasses due to the introduction of Ni and W ions with coordination number two and six. The changes in the nature of the chemical bond and the bond strength in the glass structure are normally incorporated in young's modulus which has the ability to determine the fracture behaviour involved in the glasses. On the other hand, the bulk modulus is more sensitive in exploring the changes in the cross-link density and bond stretching force constant [14].The variation of longitudinal, shear, bulk and Young's moduli (Table 2) decreases with an increase in concentration of NiO and WO_3 contents. The obvious decreases in elastic moduli are due to the continuous reduction in the rigidity of glass samples[19].The elastic properties of covalent networks are very sensitive to average coordination number; i.e. high-coordination-bond networks form relatively hard glasses, and their elastic moduli are determined by covalent forces, whereas low-coordination bond networks form relatively soft glasses, and their elastic moduli are determined bylonger-range forces. The ring formation of BaO with boron is reduced tellurite ions possibly try to modify the ring-like structure into smaller ring formation, causing a decrease in elastic moduli[20]. Boron atoms form in borate glasses structural units BO_3 with three-coordinated boron atom and BO_4 with four-coordinated boron atom[21]. It is well known that the effect of introduction of alkali metal oxides in B_2O_3 glass is the conversion of Sp^2 planer BO_3 units into more stable SP^3 tetrahedral BO_4 units. Each BO_4 unit is linked to two such other units and one oxygen from each unit with a metal ion and the structure leads to the formation of long tetrahedron chains. BaO is the modifier oxide and enters the glass network by breaking up the random network. Normally the oxygens of these oxides break the local symmetry while the cations (Ba^{2+} ions) take the interstitial positions[22]. The monotonic decrease in elastic moduli infers the absence of structural phase changes[23,24] and more creations of NBO. Poisson's ratio can be explained on the basis of the effect of tensile stress on an oriented chain of atoms or ions. If strain is lateral to the chain, its effect is maximum for lowest cross-links. Rajendran et al.,[24] reported that Poisson's ratio is affected by the changes in the cross-link density of the glass network, and the structure with high cross-link density have Poisson's ratio in the order of 0.1-0.2, while structures with low cross-link density have Poisson's ratio in the order of 0.3-0.5. In the studied glass system, the values of Poisson's ratio (Table 2) decrease and it varies from 0.2 to 0.3. The decrease in Poisson's ratio is due to making of network linkages and formation of smaller structural units in the glass samples[20]. Further, an increase in cross-link density leads to a decrease in Poisson's ratio. Further, the decrease in acoustic impedance and internal friction (Table 3) is due to the decrease in compactness and rigidity of the structure of the glass. The behaviour of internal friction is a measure of heat produced with in a material by conversion of mechanical strain energy, when it is subjected to fluctuating stress. The smaller values of internal frictionindicate the slower atomic or molecular movements. The continuous decrease in microhardness, Debye temperature and thermal expansion coefficient (Table 3) reveal the presence of Non-bridging oxygen ion (NBO) and this causes the formation of soft glassy network[25,26]. A linear decrease in the Debye temperature observed in both glass systems suggesting that ring formation does not occur in borate glasses. Further, the magnitude of microhardness of the TBBW glasses is much higher than TBBN glasses which confirm TBBW glasses possess higher rigidity than TBBN glass system.

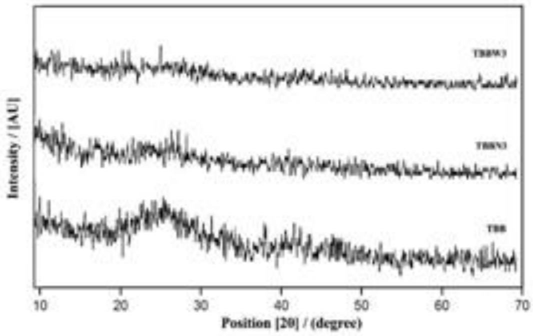

Fig 1. The powder XRD pattern of glass samples of TBB, TBBN3, TBBW3 at room temperature

Conclusion

The elastic moduli of the $10TeO_2.15BaO.(75\text{-}x)B_2O_3.xNiO$ and $10TeO_2.15BaO.(75\text{-}x)B_2O_3.xWO_3$ glass systems show many enhancements with the progressive addition of NiO and WO_3. The enhancement was attributed to the increase in the cross link density, the packing density, and the rigidity of the glass network. The decrease in density of the glass specimens show that it depends on the atomic weight of the metal atom in the network modifier (NWM). The decreasing elastic moduli indicate a reduction in network rigidity of the glass samples. It is generally accepted that nickel and tungsten ion enter the glass structure originally in one valance state viz., Ni^{2+} and W^{6+}. Both Ni^{2+} and W^{6+} ions are octahedrally coordinated with oxygen, but W^{6+} ions are more predominant over the Ni^{2+} ions according to the glass composition. The estimated acoustical, elastic and mechanical properties of the nickel and tungsten doped with tellurite barium borate glasses throw light on the rigidity and compactness in structural network. However the TBBW series of glass possess higher rigidity, strength and compactness of the glass network over the TBBN glasses.

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