*International Journal of Recent Research and Review, Vol. VII, Issue 1, March 2014*

ISSN 2277 – 8322

# Acoustical and Elastic Properties of $Ni^{2+}$ and $W^{6+}$ Transition Metal Ions Doped with Tellurite Magnesium Borate Glasses Using Pulser-Receiver Technique

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***Abstract- Glasses* of the system, $TeO_2$-MgO-$B_2O_3$ containing different concentrations of NiO and $WO_3$ (ranging from 0 to 1.0 mol % in steps of 0.2) were prepared by conventional rapid melt-quenching method. The amorphous nature of the samples were ascertained using X-ray diffractometry (XRD). Longitudinal, transverse ultrasonic velocities and attenuation of the glasses have been measured using the pulser-receiver method at 5 MHz and at room temperature. The density of the glass samples were measured by relative measurement method. The elastic properties; longitudinal modulus, shear modulus, young's modulus, bulk modulus, and Poisson's ratio together with the microhardness, Debye temperature and thermal expansion coefficient are found to be rather sensitive to the glass composition. 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.**

***Keywords* - Ultrasonic velocities, Elastic moduli, Microhardness, X-ray diffractometry, and Debye temperature.**

## I. INTRODUCTION

Glasses have the unique property of high durability together with transparency at room temperature [1]. Glasses are usually formed by mixing inorganic oxides intimately and then melting them to a homogenous liquid followed by quenching. This gives glass as an isotropic solid material [2].Glasses containing heavy non-transition metal oxides give rise to high refractive indices and non-linear optical properties. Therefore glassesmadewith$TeO_2$, PbOand MgO in addition to other glass formers like $B_2O_3$ have been at the focus of several research investigations.$B_2O_3$ is a good glass former and is a primary component of many large volume industrial glasses including those used in nuclear waste disposal. Boron is generally present in both trigonal and tetrahedral coordination of oxygen. The proportion of trigonal and tetrahedral boron depends on both the chemistry and concentration of the added modifier oxides [3]. Borate glasses are used as electro-optic modulators, electro-optic switches, solid-state laser materials and non-linear optical paramagnetic converters. $TeO_2$ is a conditional glass former, it forms glass only with modifiers like alkali/alkaline earth and transition metal oxide (TMO) or other glass formers. It could not form glass by itself because the Te-O bond is too strongly covalent to permit the requisite amount of distortion. The addition of tellurite to any other glass former or network modifier leads to interesting physical and structural changes [4]. They possess high refractive index, excellent infrared transmittance, high dielectric constant, good chemical durability, and low melting temperature [5]. 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. The octahedrally positioned $Ni^{2+}$ions are of great importance in telecommunications. The concentration of the ions present in tetrahedral or octahedral positions depends on the quantitative properties of modifiers and glass formers, size of the ions in the glass structure, their field strength, and mobility of the modifier cation etc. [6]. Tellurite glasses containing transition metal oxide $WO_3$ has previously attracted some interest, measurements have reported on capacitance and dielectric, molecular refraction, molar volume, glass transition temperature, thermal expansion, infrared spectra, density, optical absorption edge and infrared absorption edge [7].MgO is used as an insulator in industrial cables, as a basic refractory material for crucibles and as a principal fireproofing ingredient in construction materials. As a construction material, magnesium oxide wallboardshave several attractive characteristics: fire resistance, moisture resistance, mould and mildew resistance, and strength. It is also used as a protective coating in plasma displays and also as an oxide barrier in spin tunnelling devices. Pressed MgO is used as an optical material. Crystalline pure MgO has a small use in infrared optics.All these different oxides play a role in determining the ultrasonic velocities as well as the elastic properties of glasses. The measurement of elastic properties of glasses by ultrasonic pulse-echo methods becomes a more interesting subject, due to the non-destructive nature and the high precision of the technique. This measurements yields valuable information regarding the forces operating between the atoms or ions in a solid. Since the elastic properties describe the mechanical behaviour of the materials, so, the study of these properties is of fundamental importance in interpreting and understanding the nature of bonding in the solid state [8].

Since there are no reports so far made on the acoustical analysis of NiO and $WO_3$doped with tellurite magnesium borate glasses, the present investigation has been undertaken. The aim of this present investigation is to investigate the effect of NiO and $WO_3$ in the $15TeO_2$-10MgO -(75-x) $B_2O_3$-xNiO and $15TeO_2$-10MgO -(75-x) $B_2O_3$-x$WO_3$(where x=0 to 1 in steps of 0.2 mol %) glass systems on its elastic and structural properties by ultrasonic velocity measurements.

## II. EXPERIMENTAL TECHNIQUE

The glass samples of the formula $15TeO_2$-10MgO -(75-x) $B_2O_3$ - xNiO (TMBN)and$15TeO_2$-10MgO -(75-x) $B_2O_3$-x$WO_3$(TMBW) where x = 0 to 1.0 in steps of 0.2mol % have been prepared by the conventional melt- quenching method. Required quantities of analytical grade of tellurium oxide, magnesium carbonate, boric acid, nickel oxide and tungsten trioxide were obtained from E-Merck, Germany, Hi-media, and Sd-fine chemicals India. The proper compositions were mixed together continuously using an agate mortar to attain good homogeneity. The mixture was melted in alumina crucible at about 1213 K using muffle furnace (TECHNICO) 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 573 K 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 to produce parallel opposite surfaces for ultrasonic velocity measurements. Thickness of the glass samples are measured using digital vernier calliper (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 . These transducers were brought into contact with glass samples by means of a couplant, in order to ensure that there was no air void between the transducers and the glass specimen. Couplant ultrasonic sound gel was used for longitudinal waves while honey was used for shearwaves. The ultrasound gel acts as both a lubricant and an energy conductor. The attenuation coefficient α of the sample in neper per unit length is obtained from the relation

$$I = I_o e^{-\alpha t}$$

where, t is the thickness of the sample, $I_o$ and I are the ratios of amplitude of the two successive echoes.

## III. 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)

$$L = \rho U_l^2 \qquad (1)$$

(ii) Shear modulus (G)

$$G = \rho U_S^2 \qquad (2)$$

(iii) Bulk modules (K)

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

(iv) Poisson's ratio (σ)

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

(v) Young's modulus (E)

$$E = (1+\sigma)\, 2G \qquad (5)$$

(vi) Acoustic impedance (Z)

$$Z = U_l \rho \qquad (6)$$

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

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

where, α - attenuation coefficient and f - frequency of the quartz crystal.

(viii) Microhardness ($H_v$)

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

(ix) Debye temperature ($\theta_D$)

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

where, h, k, N,$V_m$ and$U_m$are the Planck's constant ($6.626 \times 10^{-34}$JS), the Boltzmann's constant($1.38 \times 10^{-23}$JK$^{-1}$), the Avogadro's number ($6.023 \times 10^{23}$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 ($\alpha_p$)

$$\alpha_p = 23.2\,(U_l - 0.57457) \qquad (10)$$

## IV. RESULTS AND DISCUSSION

The experimental values of density (ρ), longitudinal ultrasonic velocity ($U_l$), shear ultrasonic velocity ($U_s$) and attenuation (α) of the tellurite magnesium 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), shear modulus (G), bulk modulus (K), Young's modulus (E), Poisson's ratio(σ), acoustic impedance(Z), internal friction $(Q^{-1})$, microhardness ($H_v$), Debye temperature ($\theta_D$),and thermal expansion coefficient ($\alpha_p$) are presented in the Tables 2-3. X-ray diffraction patterns (Fig. 1) of the studied glass systems reveal the absence of any discrete or continuous sharp crystalline peaks, but show homogenous glassy characters.

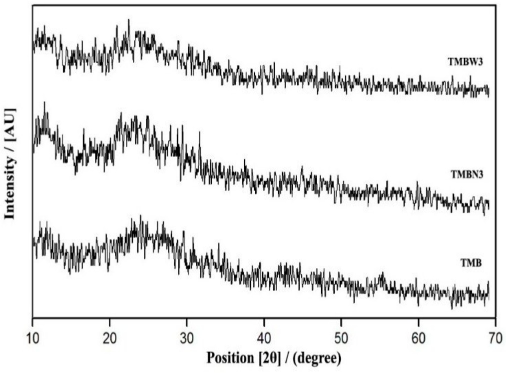

Fig. 1: The powder XRD pattern of glass samples of TMB, TMBN3, and TMBW3 at room temperature

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 theglass dependsupon 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 [9-10]. From the (Table I) it is observed that the values of density decreases with increase in mol% of NiO and $WO_3$ glass systems.

The density values of TMBW glass system are much higher than that of TMBN glasses. The structure of glass depends on the nature of the ions entering in the network and hence the density of glass. Further, it is observed from the above table, thevariation of longitudinalvelocity ($U_l$) and shear velocity ($U_s$) decreases 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 [11]. 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 I 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 [12].

The decrease in the ultrasonic attenuation $\alpha$ may be attributed to the increase of the Ni-O and W-O ionic bonds. This is probably attributed to the increase in the cross-link density in glasses due to 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 [13].

The values of longitudinal, shear, bulk and Young's moduli (Table II) decreases with increase in concentration of NiO and $WO_3$ contents. 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 by longer-range forces. The decreasing elastic moduli indicating a reduction in network rigidity. The ring formation of NiO and $WO_3$ with boron is reduced magnesium and tellurium ions possibly try to modify the ring-like structure into smaller ring formation, causing a decrease in elastic moduli. Boron atoms form in borate glasses structural units $BO_3$ with three-coordinated boron atom and $BO_4$ with four-coordinated boron atom [14]. 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. MgO is 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 ($Mg^{2+}$ ions) take the interstitial positions [15].

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 [16]., 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 has Poisson's ratio in the order of 0.1 - 0.2, while structures with low cross-link density has Poisson's ratio in the order of 0.3 - 0.5. In the studied glass system, the values of Poisson's ratio (Table II) increase and it is vary from 0.2 to 0.28. The increase in Poisson's ratio is due to breaking of network linkages and formation of smaller structural units in the glass samples [17]. Further, a high cross-link density leads to an increase in Poisson's ratio. The decrease in acoustic impedance and internal friction (Table III) 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 friction indicate the slower atomic or molecular movements. The continuous decrease in microhardness, Debye temperature and thermal expansion coefficient (Table III) reveals the presence of non-bridging oxygen ion (NBO) and this causes the formation of soft glassy network ([13], [18]). A linear decrease in the Debye temperature observed in both glass systems suggesting ring formation does not occur in borate glasses. Further the magnitude of micro hardness of the TMBW glasses is much higher than TMBN glasses which confirm TMBW glasses possess higher rigidity than TMBN glass system.

## V. CONCLUSION

The elastic moduli of the $15TeO_2$-10MgO-(75-x)$B_2O_3$-xNiO and $15TeO_2$-10MgO-(75-x) $B_2O_3$-x$WO_3$ glass systems show many enhancements with the progressive addition of NiO and $WO_3$. The enhancements were 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 shows that it depends on the atomic weight of the metal atom in the network modifier (NWM). The decreasing elastic moduli found in both glasses indicates a reduction in network rigidity. The estimated acoustical, elastic and mechanical properties of the nickel and tungsten doped tellurite magnesium borate glasses throw light on the rigidity and compactness in structural network. However the TMBW series of glass possess higher rigidity, strength and compactness of the glass network over the TMBN glasses.

## VI.ACKNOWLEDGEMENTS

The author wish to express their gratitude to Dr. Krishnan Balasubramaniyan, Prof. and Head, Centre of NDT evaluation, IIT., Chennai for having provided the necessary experimental setup, for velocity measurements in glass samples.

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Table I

Composition, measured values of density (ρ), longitudinal velocity ($U_l$), shear velocity ($U_s$)and attenuation (α) of TMB, TMBN and TMBW glasses at room temperature.

| **Sample Label** | **Composition (mol %)** | **Density ρ / ($kg.m^{-3}$)** | **Ultrasonic Velocity U / ($m.s^{-1}$)** | | **Attenuationα / (nepers. unit length$^{-1}$)** |
|---|---|---|---|---|---|
| | | | **Longitudinal ($U_l$)** | **Shear ($U_s$)** | |
| $TeO_2 - MgO - B_2O_3$ (TMB) | | | | | |
| TMB | 15-10-75 | 2445.0 | 5660.38 | 3196.88 | 27.15 |
| System 1 : $TeO_2 - MgO - B_2O_3 - NiO$(TMBN) | | | | | |
| TMBN 1 | 15-10-74.8-0.2 | 1878.3 | 5657.20 | 3191.49 | 32.13 |
| TMBN 2 | 15-10-74.6-0.4 | 1717.3 | 5633.80 | 3133.16 | 31.65 |
| TMBN 3 | 15-10-74.4-0.6 | 1573.0 | 5504.59 | 3092.78 | 29.70 |
| TMBN 4 | 15-10-74.2-0.8 | 1490.3 | 5464.59 | 3061.23 | 28.99 |
| TMBN 5 | 15-10-74.0-1.0 | 1329.1 | 5405.41 | 3045.69 | 28.75 |
| System 2 : $TeO_2 - MgO - B_2O_3 - WO_3$ (TMBW) | | | | | |
| TMBW 1 | 15-10-74.8-0.2 | 2425.6 | 5640.38 | 3141.36 | 44.92 |
| TMBW 2 | 15-10-74.6-0.4 | 2403.2 | 5479.45 | 3084.83 | 37.70 |
| TMBW 3 | 15-10-74.4-0.6 | 2198.1 | 5429.86 | 3007.52 | 37.29 |
| TMBW 4 | 15-10-74.2-0.8 | 2165.0 | 5357.14 | 2919.71 | 34.43 |
| TMBW 5 | 15-10-74.0-1.0 | 1827.9 | 5329.86 | 2905.57 | 27.27 |

Table II

Values of elastic moduli and Poisson's ratio of TMB, TMBN andTMBWglasses at room temperature.

| **Sample Label** | **Longitudinal Modulus L/ (×$10^{10}$ N. $m^{-2}$)** | **Shear Modulus G/ (× $10^{10}$ N. $m^{-2}$)** | **Bulk Modulus K/(x$10^{10}$ N.$m^{-2}$)** | **Young's Modulus E/ (×$10^{10}$ N.$m^{-2}$)** | **Poisson's Ratio (σ)** |
|---|---|---|---|---|---|
| $TeO_2 - MgO - B_2O_3$ (TMB) | | | | | |
| TMB | 7.8338 | 2.4988 | 4.5022 | 6.3260 | 0.2658 |
| System 1 : $TeO_2 - MgO - B_2O_3 - NiO$(TMBN) | | | | | |
| TMBN 1 | 6.0113 | 1.9132 | 3.4604 | 4.8465 | 0.2666 |
| TMBN 2 | 5.4507 | 1.6858 | 3.2030 | 4.3025 | 0.2761 |
| TMBN 3 | 4.7663 | 1.5046 | 2.7602 | 3.8199 | 0.2694 |
| TMBN 4 | 4.4494 | 1.3963 | 2.5877 | 3.5502 | 0.2713 |
| TMBN 5 | 3.8831 | 1.2329 | 2.2393 | 3.1252 | 0.2674 |
| System 2 : $TeO_2 - MgO - B_2O_3 - WO_3$ (TMBW) | | | | | |
| TMBW 1 | 7.7168 | 2.3936 | 4.5254 | 6.1046 | 0.2752 |
| TMBW 2 | 7.2155 | 2.2869 | 4.1664 | 5.7996 | 0.2680 |
| TMBW 3 | 6.4805 | 1.9882 | 3.8296 | 5.0846 | 0.2787 |
| TMBW 4 | 6.2133 | 1.8456 | 3.7526 | 4.7569 | 0.2887 |
| TMBW 5 | 5.1926 | 1.5432 | 3.1351 | 3.9771 | 0.2886 |

Table III

Values of acoustic impedance (Z), internal friction($Q^{-1}$), microhardness ($H_v$), Debyetemperature ($\theta_D$) and thermal expansion coefficient ($\alpha_p$) of TMB, TMBN and TMBW glasses at room temperature

| **Sample Label** | **Z /($\times 10^7$ kg.m$^{-2}$ s$^{-1}$)** | **($Q^{-1}$)/ ($\times 10^{-11}$ dB.s$^2$m$^{-2}$)** | **$H_v$/($\times 10^9$ N.m$^{-2}$)** | **$\theta_D$ / (K)** | **$\alpha_p$/($\times 10^2$ m.s$^{-1}$)** |
|---|---|---|---|---|---|
| $TeO_2$ – MgO -$B_2O_3$ (TMB) | | | | | |
| TMB | 1.3840 | 1.7639 | 3.9015 | 381.52 | 1313.08 |
| System 1 : $TeO_2$ – MgO -$B_2O_3$ – NiO(TMBN) | | | | | |
| TMBN 1 | 1.0626 | 2.0886 | 2.9769 | 348.83 | 1312.34 |
| TMBN 2 | 0.9675 | 2.0660 | 2.5163 | 332.73 | 1306.91 |
| TMBN 3 | 0.8659 | 1.9842 | 2.3131 | 318.69 | 1276.93 |
| TMBN 4 | 0.8142 | 1.9509 | 2.1289 | 309.85 | 1267.65 |
| TMBN 5 | 0.7184 | 1.9560 | 1.9118 | 296.57 | 1253.92 |
| System 2 : $TeO_2$ – MgO -$B_2O_3$ – $WO_3$ (TMBW) | | | | | |
| TMBW 1 | 1.3681 | 2.9288 | 3.5872 | 373.90 | 1308.44 |
| TMBW 2 | 1.3168 | 2.5302 | 3.5371 | 365.21 | 1271.10 |
| TMBW 3 | 1.1935 | 2.5256 | 2.9332 | 345.70 | 1260.00 |
| TMBW 4 | 1.1598 | 2.3635 | 2.5999 | 333.98 | 1242.72 |
| TMBW 5 | 0.9743 | 1.8816 | 2.1749 | 313.69 | 1236.39 |