Investigation of Acoustical Parameters in Ternary B₂O₃ – MnO₂ –Al₂O₃ GLASSES

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Abstract: Glass samples of composition 60B₂O₃ - (40-x)MnO₂-xAl₂O₃ (where x=0, 5, 10, 15 and 20 mol %) were synthesized by melt quench technique. Longitudinal and shear velocities have been measured at 5 MHz frequency by Pulse - Echo Overlap method. The density of the glasses has also been determined. The density increases while their molar volume values decreases with the increase of Al₂O₃ concentration. The amorphous nature of the glass samples were checked by XRD technique. Elastic moduli such as longitudinal, bulk, shear and Young’s moduli, Poisson’s ratio, acoustic impedance, microhardness and Debye temperature were calculated from the measured data. The variations of the above parameters with change in composition have been discussed in terms of structural changes in the glass network.

Keywords: XRD, Ultrasonic velocity, elastic moduli, rigidity.

1. Introduction

Among the various non-destructive evaluation techniques, ultrasonic technique is a versatile tool for investigating the changes in microstructure, deformation process and mechanical properties of materials. The ultrasonic waves are closely related with the elastic and inelastic properties of the materials (1). The study of elastic properties of glasses has inspired many researchers (2, 3), because their measurement yields information concerning the forces that are operative between the atoms or ions comprising solid. This is basically important in interpreting and understanding the nature of bonding of the solid state.

B₂O₃ is one of the best glass formers (4) and it exhibits unique structural features. It is well know that the main structural units of the borate network are (BO₃) triangles and (BO₄) tetrahedral, may form different super structural units; boroxol and metaborate rings, metaborate chains, pentaborate, triborate, diborate and pyroborate. (5). Borate glasses containing various transition metal ions have been under extensive investigation of the technological applications especially in microelectronics, optical glasses and solid state laser (6). Transition metal doped borate glasses have been studied by several authors (7-9). Among all transition metal ions, manganese (Mn) ion is particularly interesting because it exists in all transition metal ions, manganese (Mn) ion is particularly

2. Experimental

2.1. Sample preparation

The glass samples under investigations were prepared by the conventional melt quench technique. The required amount in mol% of different chemicals in powder form was weighed using single pan balance having an accuracy of ± 0.0001g. The nominal compositions of BMA glass system are listed in Table-1.

<table>
<thead>
<tr>
<th>S. No</th>
<th>Nomenclature</th>
<th>Compositions in mol%</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>BMA 1</td>
<td>60-35-5</td>
</tr>
<tr>
<td>2</td>
<td>BMA 2</td>
<td>60-30-10</td>
</tr>
<tr>
<td>3</td>
<td>BMA 3</td>
<td>60-25-15</td>
</tr>
<tr>
<td>4</td>
<td>BMA 4</td>
<td>60-20-20</td>
</tr>
</tbody>
</table>

The chemicals were first thoroughly mixed together by grinding the mixture repeatedly to obtain a fine powder. The powder was melted in silica crucible at about 900°C in muffle furnace for few minutes to homogenize the melts. The molten sample is cast into a copper mould having dimensions 10mm diameter and 6mm length. All these glasses were annealed for three hours at 250°C to avoid mechanical strain developed during the quenching process. The prepared samples are chemically stable and non-hygroscopic. The glass samples are polished and the surfaces are made perfectly plane and smoothened by diamond disc and diamond powder. Thickness of the samples has been measured using vernier calipers with an accuracy of 0.0001mm.


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2.2. Measurements

2.2.1 XRD
The amorphous nature of the samples is confirmed by X-ray diffraction technique using GE-Inspection technology 3003TT model made in Germany copper target operating voltage 40 kV 300 mA current rate.

2.2.2. Velocity
The longitudinal and shear velocities of the glass specimens were measured using the Pulse – Echo Overlapping method at 303K by making use of 5MHz X-cut and Y-cut transducers. These transducers were brought into contact with each of the samples by means of a couplant, in order to ensure that there was no air void between the transducers and the specimen. By applying constant pressure on the probe, the echo waveforms were obtained on the display unit and stored in the memory.

Ultrasonic velocity is calculated using the relation \( U = \frac{2d}{t} \) ……… (1)
where d and t are the thickness of the specimen (mm) and transit time in microsecond

2.2.3. Density
The density of the glass samples was measured using relative measurement technique. Ionized water was used as a buoyant liquid. The glass samples were weighed both in air and after immersing in water at 303K. The weight of the glass samples was measured in a single pan with an accuracy of 0.0001g. The density was calculated using the formula \( \rho = \frac{W_A - w_w}{W_A - w_w} \) (2) \( W_A \) and \( w_w \) are the weight of the glass samples in air and in water respectively and \( \rho_w \) is the density of water at 303K.

3. Theory and Calculation
The elastic moduli and other parameters of the glass specimen are calculated using the measured density, longitudinal velocity and shear velocity as given below:

\[ L = \rho U^2 \] (3)
\[ G = \rho U^2 \] (4)
\[ K = L \left( \frac{3}{2} \right) G \] (5)
\[ (1 + \sigma) = \frac{L - 2G}{G} \] (6)
\[ \rho Z = \rho U \] (7)
\[ \rho U \] (8)
\[ \rho V_m \] (9)
\[ \rho U_m \] (10)
where \( \rho \), \( U \), \( V_m \), \( h \), \( k \), \( N \) and \( V_m \) are the density, longitudinal velocity, shear velocity, Planck’s constant, Boltzmann’s constant, Avogadro’s number and molar volume respectively.

Mean sound velocity \( U_m = \left[ \frac{1}{3} \left( \frac{2}{U_l^2} + \frac{1}{U_s^2} \right) \right]^{1/3} \)

4. Results and Discussion

XRD Analysis
X-ray diffraction is a useful method to detect readily the presence of crystals in a samples. From the result of X-ray diffraction, the prepared glass system ([Fig1] was found to be in the form of broad halo, which is characteristic of amorphous structure (14). This indicates the absence of long range atomic order and lack of periodicity of the three dimensional network.

The density, molar volume, longitudinal velocity and shear velocity, of the different glass specimen with respect to change in the mol% of the \( \text{Al}_4\text{O}_5 \) are reported in Table 2. Elastic moduli and Poisson’s ratio are given in Table 3. The, acoustic impedance, microhardness and Debye temperature are presented in Table 4.

**Table 2: Values of density, molar volume, longitudinal velocity and shear velocity of BMA glass system**

<table>
<thead>
<tr>
<th>Name of the sample</th>
<th>Density ( (\rho) \times 10^3 \text{kgm}^{-3} )</th>
<th>Molar volume ( (V_m) \text{cm}^3/\text{mol} )</th>
<th>Ultrasonic velocity ms(^{-1} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>BMA1</td>
<td>4.2467</td>
<td>18.287</td>
<td>4898.4</td>
</tr>
<tr>
<td>BMA2</td>
<td>4.3327</td>
<td>17.922</td>
<td>5010.6</td>
</tr>
<tr>
<td>BMA3</td>
<td>4.4101</td>
<td>17.86</td>
<td>5239.9</td>
</tr>
<tr>
<td>BMA4</td>
<td>4.5492</td>
<td>17.4686</td>
<td>5358.5</td>
</tr>
</tbody>
</table>

**Table 3: Values of longitudinal, shear, bulk and Young’s moduli and Poisson’s ratio of BMA glass systems**

<table>
<thead>
<tr>
<th>Name of the sample</th>
<th>Longitudinal modulus ( L ) 10^9 \text{Nm}^{-2}</th>
<th>Shear modulus ( G ) 10^9 \text{Nm}^{-2}</th>
<th>Bulk modulus ( K ) 10^9 \text{Nm}^{-2}</th>
<th>Young’s modulus ( E ) 10^9 \text{Nm}^{-2}</th>
<th>Poisson’s ratio ( \sigma )</th>
</tr>
</thead>
<tbody>
<tr>
<td>BMA1</td>
<td>101.896</td>
<td>29.766</td>
<td>62.208</td>
<td>77.014</td>
<td>0.2936</td>
</tr>
<tr>
<td>BMA2</td>
<td>108.777</td>
<td>31.824</td>
<td>66.344</td>
<td>82.312</td>
<td>0.2932</td>
</tr>
<tr>
<td>BMA3</td>
<td>121.086</td>
<td>36.876</td>
<td>71.916</td>
<td>94.481</td>
<td>0.281</td>
</tr>
<tr>
<td>BMA4</td>
<td>130.623</td>
<td>40.469</td>
<td>76.664</td>
<td>103.241</td>
<td>0.2755</td>
</tr>
</tbody>
</table>

**Table 4: Values of acoustic impedance, microhardness and Debye temperature of BMA glass systems**

<table>
<thead>
<tr>
<th>Name of the sample</th>
<th>Acoustic impedance ( Z \times 10^3 \text{kgm}^{-1}\text{s}^{-1} )</th>
<th>Microhardness ( H \times 10^6 \text{Nm}^{-2} )</th>
<th>Debye temperature ( \theta_D \text{K} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>BMA1</td>
<td>2.08</td>
<td>4.094</td>
<td>398.81</td>
</tr>
<tr>
<td>BMA2</td>
<td>2.17</td>
<td>4.383</td>
<td>409.67</td>
</tr>
<tr>
<td>BMA3</td>
<td>2.31</td>
<td>5.395</td>
<td>437.66</td>
</tr>
<tr>
<td>BMA4</td>
<td>2.43</td>
<td>6.055</td>
<td>503.12</td>
</tr>
</tbody>
</table>

**Table 4**: Values of acoustic impedance, microhardness and Debye temperature of BMA glass systems
tetrahedral coordination. The presence of AlO

...from the modifier oxides in

...the modifier oxides in the glass Abd El-moneim etal. (17) reported that CaO

...the formation of Al-O-B linkage and they concluded that there is an increase in the connectivity of the glass compared with the parent pure B2O3 glasses.

...increase in the packing density, rigidity and hence the density of the glasses.

...Al2O3 indicating the conversion of some BO3 units into BO4 units. Further the increase of Al2O3 content at the expense of MnO2 cause the increase in ultrasonic velocity and a simultaneous increase in the number of BO4 units that increase the stability of the glasses. Al3+ incorporated in the glasses as a network former and to form AlO4 tetrahedral and B-O-Al linkages.

...in longitudinal (U L) and shear (U S) velocities increase almost linearly with the increase in concentration of Al2O3, but the rate of increase is greater than that of U S. The velocity of B2O3-MnO2 glass is increased by the introduction of Al2O3 indicating the conversion of some BO3 units into BO4 units. Further the increase of Al2O3 content at the expense of MnO2 cause the increase in ultrasonic velocity and a simultaneous increase in the number of BO4 units that increase the stability of the glasses. Al3+ incorporated in the glasses as a network former and to form AlO4 tetrahedral and B-O-Al linkages.

Table 3 shows the values of longitudinal, shear, bulk and Young’s moduli as a function of Al2O3 concentration which varies in a similar fashion as ultrasonic velocities. The increase in the values of elastic moduli has been attributed to an increase in the packing density, rigidity and hence the formation of stronger structural building units in the glass network. The large difference between L and G arises from volume effect. The change in volume due to compressions and expansions involved in longitudinal strain is pronounced while no change in volume is involved in shear strain (18).

In general, Poisson’s ratio of the order of 0.1 to 0.2 reveals high cross link density while low cross link density has the Poisson’s ratio between 0.3 and 0.5. Ultrasonic velocities can be utilized in the calculation of the Poisson’s ratio. The value of the Poisson’s ratio decreases from 0.2936 to 0.2755 with the increase of Al2O3 content as shown in table 3. This decrease can be explained in terms of the introduction of covalent bond that formed glass network as B-O-Al.

The acoustic impedance increases with increase in mol% of Al2O3 content in the glass system confirming the increase in rigidity of the structure of the glass. Further, the increase in microhardness expresses the stress required to eliminate the free volume of the glass in the present study, the increasing microhardness indicated the increase in structural connectivity of the glasses.

The results are further confirmed by another parameter, the Debye’s temperature which represents the temperature at which all modes of vibration in solid are excited and its increasing trend implies an increase in the rigidity of glass. From table 4. it can be observed that the increase in Debye temperature values confirms the occurrence of strong ring formation in glasses. Such an enhancement of Debye’s temperature is attributed to the increase in the number of atom in the chemical formula of the glass and increase in ultrasonic velocity (19). The continuous increase of Debye temperature also suggests that the compactness and structure leading to increase in mean sound velocity.

5. Conclusion

The density, ultrasonic velocities and other evaluated parameters of studied B2O3 – MnO2 – Al2O3 glasses have revealed that;

(i) the density of the studied glass system increases whereas the molar volume decreases with increase in mol% of Al2O3 which indicates there is an increase in connectivity of the network structure.

(ii) The elastic moduli and remaining parameters increase while Poisson’s ratio decreases indicating the increase in rigidity of the network structure.

References


