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Optical Properties of Borotellurite Glasses Containing Metal Oxides

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Abstract. Glass samples of the system: 5MₓOᵧ-20B₂O₃-75TeO₂ : MₓOᵧ = WO₃, Nb₂O₅, PbO, Nd₂O₃, Y₂O₃, Eu₂O₃ were prepared by melt quenching and characterized by X-ray diffraction, density, Differential Scanning Calorimetry, UV-visible and FTIR spectroscopy. XRD patterns confirmed the amorphous structure of all samples. Glass transition temperature was maximum in borotellurite glass containing Y²O₃. Refractive index, atomic polarizability and basicity increased in the following order of ions: Y³⁺ < Eu³⁺ < Pb²⁺ < Nd³⁺ < Nb³⁺ < W⁶⁺. FTIR studies showed that PbO is outstanding in enhancing the concentration of tetrahedral borons in the borotellurite network.

Keywords: Borotellurite glasses, XRD, DSC, FTIR and UV-Visible spectroscopy.

PACS: 81.05.Kf, 61.05.cp, 65.60.+a, 63.50.-x and 78.40.-q

INTRODUCTION

Tellurite glasses have a wide range of applications. One of the commercial applications of tellurite glasses is in the field of optical communication due to their outstanding properties like high refractive index, high optical non-linearity and good infrared transmittance [1-5]. Tellurite glasses have been reported to exhibit 30 times higher Raman gain coefficients than silica glass and find application in Raman amplifiers and non-linear optical waveguides [6, 7]. Many studies are reported on the technological importance of tellurite glasses containing transitional metal and/or rare earth ions in memory switching devices and as cathode materials for batteries [8].

In this work, we report the study of short-range structure and optical and thermal properties of borotellurite glasses doped with W⁶⁺, Nb³⁺, Pb²⁺, Nd³⁺, Y³⁺ and Eu³⁺. Glasses were characterized by density, X-ray diffraction (XRD), Differential Scanning Calorimetry (DSC) and UV-visible and FTIR spectroscopy.

EXPERIMENTAL METHODS

Glasses of the system: 5MₓOᵧ-20B₂O₃-75TeO₂ : MₓOᵧ = WO₃, Nb₂O₅, PbO, Nd₂O₃, Y₂O₃, Eu₂O₃ were prepared using H₃BO₃ (Sigma Aldrich Inc., USA, 99.5%), TeO₂ (Sigma Aldrich Inc., USA, 99%), and WO₃, Nb₂O₅, PbO, Nd₂O₃, Y₂O₃ and Eu₂O₃ as starting materials. The chemicals in the above mentioned compositions were weighed and sintered at 300°C for 24 h and then melted in a temperature range of 800°C to 900°C in a platinum crucible. Glass samples were prepared by normal quenching technique in which a small amount of melt was quenched on a brass block and a button shaped sample was obtained and immediately transferred to a furnace kept at a temperature of about 80°C lower than the glass transition temperature. Samples were annealed for 30 min and then slowly cooled to room temperature.

RESULTS AND DISCUSSION

X-ray diffraction measurements were performed on Bruker D8 Focus X-ray diffractometer with Cu Kα radiation (λ =1.54056 Å) in the 2θ range of 10⁰-70⁰. Figure 1 shows the XRD patterns of borotellurite
glasses doped with metal oxides. These patterns do not show any sharp peaks, but the characteristic broad humps due to short-range order in glasses.

FIGURE 1. XRD patterns of borotellurite glasses doped with different metal oxides.

Density of glasses were measured by Archimedes principle using an electronic balance with an accuracy of 10⁻⁴ g. Turpentine oil was used as an immersion liquid. Density and molar volume values are given in Table 1. Density increases on adding metal oxides in the glass network and is maximum for glass containing PbO [Figure 2].

Thermal studies were performed on SETARAM SETSYS Evolution-1750 system in the temperature range of 200-850°C at a heating rate of 10°C/min, in air flow rate of 20 ml/min in Pt pans. From DSC scans, glass transition temperature, T_g was determined [Table 1]. T_g is maximum for the sample having 5-mol% of Y₂O₃ and minimum for glass containing Pb²⁺ ion. All metal ions except Pb²⁺ strengthened the binary borotellurite glass network due to the incorporation of metal oxide bonds with higher bond enthalpies.

Optical absorption spectra of polished disk shaped borotellurite glasses were measured at room temperature on Shimadzu 1601 double beam UV-visible spectrophotometer in wavelength range of 200-1100 nm. The optical absorption coefficient α(λ) was calculated by dividing the absorbance A, with sample thickness and is plotted in figure 3.

From α vs λ plots, absorption edge, λ_o was determined as the wavelength at which α = 9 cm⁻¹ [Table 2]. The absorption edge shifted towards longer wavelengths on adding metal oxides. Optical band gap, E_g of glasses was determined from λ_o. E_g was used to calculate the optical electronegativity χ, refractive index n, and the molar volume variation in borotellurite glasses containing metal oxides.

FIGURE 2. Density and molar volume variation in borotellurite glasses containing metal oxides.

FIGURE 3. Absorption spectra (α vs λ) of borotellurite glasses.

From α vs λ plots, absorption edge, λ_o was determined as the wavelength at which α = 9 cm⁻¹ [Table 2]. The absorption edge shifted towards longer wavelengths on adding metal oxides. Optical band gap, E_g of glasses was determined from λ_o. E_g was used to calculate the optical electronegativity χ, refractive index n,
electronic polarizability $\alpha_o$ and optical basicity $\Lambda$, of glasses using the formulae reported in the literature [9]. The calculated values are given in table 2.

Optical energy gap of glasses containing metal ions was lower than undoped borotellurite glass (i.e. sample 20BTe) except for the sample containing Y$^{3+}$. The optical electronegativity was lowest for glass having W$^{6+}$. Values of refractive indices follow the trend: 5Y-20BTe < 20BTe < 5Eu-20BTe < 5Pb-20BTe < 5Nd-20BTe < 5Nb-20BTe < 5W-20BTe.

CONCLUSIONS

Borotellurite glasses were prepared with 5-mol% of W$^{6+}$, Nb$^{3+}$, Pb$^{2+}$, Nd$^{3+}$, Eu$^{3+}$, Y$^{3+}$ and their properties were compared with undoped borotellurite glass. Glass transition temperature is maximum for 5Y-20BTe sample. Refractive index, atomic polarizability and basicity increases in the following order of dopant ions: Y$^{3+}$ < 20BTe < Eu$^{3+}$ < Pb$^{2+}$ < Nd$^{3+}$ < Nb$^{3+}$ < W$^{6+}$. Conversely optical energy gap is lowest for glass containing WO$_3$ and is highest for the glass with Y$_2$O$_3$. Also, PbO produces a maximum increase in $N_4$ while WO$_3$ suppresses it.

REFERENCES


### TABLE 2. Optical energy gap and $N_4$ in borotellurite glasses containing metal ions.

<table>
<thead>
<tr>
<th>Sample Code</th>
<th>Absorption edge $\lambda_o$ [nm]</th>
<th>Energy gap $E_o$ [eV]</th>
<th>$\alpha_o$</th>
<th>$\Lambda$</th>
<th>$N_4$</th>
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<td>3.4</td>
<td>0.913</td>
<td>2.28</td>
<td>2.68</td>
</tr>
</tbody>
</table>

Optical energy gap of glasses containing metal ions was lower than undoped borotellurite glass (i.e. sample 20BTe) except for the sample containing Y$^{3+}$. The optical electronegativity was lowest for glass having W$^{6+}$. Values of refractive indices follow the trend: 5Y-20BTe < 20BTe < 5Eu-20BTe < 5Pb-20BTe < 5Nd-20BTe < 5Nb-20BTe < 5W-20BTe.

The FTIR absorption spectra show three bands in the wavenumber ranges of 500-800 cm$^{-1}$, 800-1150 cm$^{-1}$ and 1150-1550 cm$^{-1}$ [Figure 4]. The first band in the wavenumber range of 500-800 cm$^{-1}$ is due to Te-O vibrations in different Te-O units. The bands in the wavenumber ranges of 820-1140 cm$^{-1}$ and 1150-1550 cm$^{-1}$ are due to B-O stretching vibrations in BO$_4$ and BO$_3$ units respectively [4]. Areas under the second and third bands i.e. $A_4$ and $A_3$ are calculated and the ratios of these areas $A_4/(A_4+A_3)$ is considered proportional to the fraction of tetrahedral borons ($N_4$) in the glass network. It is clear from the data presented in table 2 that $N_4$ is nearly the same for the undoped borotellurite sample: 20BTe and the glasses doped with Nd$^{3+}$ and Nb$^{3+}$. Maximum $N_4$ is in the sample containing Pb$^{2+}$ and the minimum is observed in the glass with W$^{6+}$.

FIGURE 4. FTIR absorption spectra of borotellurite glasses doped with metal ions.