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Structure and Properties of the Soda-Borate Glasses: Effect of Adding Fe_2O_3 Concentration

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Abstract

Effect of adding iron oxide (Fe_2O_3) in the sodium borate glasses (B_2O_3-Na_2O) in the structural and physical properties was studied. Samples of the studied glass system were prepared with different percent of Fe_2O_3 concentration. This latter was added by substitution of the B_2O_3 and Na_2O at concentration of (05, 10, 15 and 20 by wt %). The structural and physical properties of obtained glass were evaluated. The measurements of physical properties show that, in term of the glass density is relatively high with also high thermal expansion coefficient values with Fe_2O_3 presence. In the studied glass system, addition of transition iron oxide (Fe_2O_3) leads to weakened structure of glass so the transition temperature (Tg) and the softening temperature (Ts) decrease with increasing of Fe_2O_3 addition.

Keywords: Iron oxide; Sodium borate glasses; Thermal expansion coefficient; Transition temperature; Softening temperature

Introduction

In recent years, borate glasses have been the subject of several studies and research works due to their applications in various domains. Indeed, the borosilicate glasses have very important characteristics namely higher resistance to crystallization compared to others system glasses, which confers their use in various scientific and technological applications [1]. Research works of the SiO_2-B_2O_3-Na_2O [2-4] ternary system have shown that alkali cations (Na^+) can played as charge compensators in the four coordinated boron breaking down the original network. In others studies on borate glasses have also shown that the addition of metal oxides (Fe_2O_3) can modified their electrical, optical and magnetic properties furthermore can changes the structure of the glass network [5]. The SiO_2-Na_2O-Al_2O_3-B_2O_3 glass system was used because it is easily doped with certain metals to form nanocrystals and having a transparent window from ultraviolet to near infrared [6]. Dantas et al. have already studied the effect of Fe_2O_3 concentration on the structure of the SiO_2-Na_2O-Al_2O_3-B_2O_3 glass system [7]. Their results prove that the glass transition and crystallization temperatures have changed as a function of Fe_2O_3 concentration and that these alterations were related to structural change in the glass system. The obtained information about of the structure changes in Si-O-Si units of these glasses have been related at the concentration of Fe_2O_3 added to studied system. In another glass system (B_2O_3-PbO-Na_2O), the substitution of Na_2O by Fe_2O_3 led to an increasing the microhardness values and a decreasing the thermal expansion coefficients of the studied glasses [8]. Also, a decreased the conductivity was observed with increasing of replacement Na_2O by Fe_2O_3, this is due to converting of the three coordinated boron atoms [BO_3] to four coordinated boron atoms [BO_4].

For this, the aim of this work is to study the effect of Fe_2O_3 concentration structural, physical properties in B_2O_3-Na_2O glass system. Indeed, the Fe_2O_3 is added by substitution of the B_2O_3 and Na_2O at different concentrations (5, 10, 15 and 20% by wt. %). After, the structural and physical properties of obtained glasses were evaluated, and compared to reference glass (with 0% of Fe_2O_3).

Experimental

Preparation of studied glasses

Iron-borate glasses (BNF) were prepared whose the chemical composition is given in Table 1. For each mixture, the 10% wt. B_2O_3 is substituted by Na_2O and Fe_2O_3 (5%-5%). The finely crushed powders was then placed in a platinum crucible and transferred to an electric furnace at temperature ranging 1000°C with a stage for 1 h. The liquid was then cast in a graphite mold preheated to approximately 250°C to limit the thermal shocks during hardening. The samples were annealing then at 250°C for 1 h.

Test methods

Dilatometric analysis: The expansion curves of glass samples were determined using a dilatometer DIL 402°C at an average speed of heating of 5 $K.min^{-1}$. The sample had a rectangular shape with an 8 mm width and a 20-25 mm length. The glass transition temperature (Tg) was determined from the expansion curve using the interception method, whereas the Ts (softening temperatures) values were determined as the temperatures of the maximum expansions [2].

Density and molar volume measurements: The densities were determined out using Archimedes' method with xylene as an immersion fluid. The relative error in these measurements was about ± 0.03 $g{\bullet}cm^{-3}$ and the molar volume V_m was calculated from the molecular weight M and the density ρ according to the relation: $V_m=M/\rho$. Molar volume samples is determined by the following formula: $V_m=P_m/MV$ (molecular weight of glass/Density) in cm^3/mol.

Theoretical calculus of properties of glasses: In a glass structure, the various components have an important role determining certain properties of the glass material. It is possible to estimate the properties of studied glasses by calculating by means of additive formulas these properties from the mentioned glass composition [2].

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Results and Discussion

Fe2O3 effect on glass coloration

Aspect of the samples: Pictures were taken on studied glass samples to see influence of Fe_2O_3 on the macroscopic aspect particularly the glass coloration. Coloration aspect is shown in Figure 1. All glass variants are colored (dark blue) except the BNF1 sample. This coloration is due to the iron oxide (brick red) taking place by transmission in the visible.

Visible transmission: Glasses containing conventional network modifiers are usually completely colorless in the visible region of the spectrum. It changes if the glasses contain at the same time the second family elements of the periodic Table such as: Cu, Ti, V, Cr, Mn, Fe, Co, and Ni are the most important [9]. It occurs to these electron jumps, that is to say, removals, even under the effect of a low-energy light, so it appears colorations in the visible range. The color observed depends primarily on the electronic configuration and therefore the nature of the element and the environment (oxygen ions). Among the theories that explain the coloration phenomenon is that of the Ligand-field theory of Hartmann. This theory predicts that the glasses coloration by the transition metals "3d" which is due to electronic transitions between energy levels of the electron-degenerate d. For free ions (any field), electronic levels "3d" are identical in energy. However, when a metal ion transition is surrounded by some anion, referred to as "ligands", the electrical interaction causes some disruption of the energy levels fields whose amplitude is function of the field, number and geometrical arrangement of anions in the neighborhood.

The Number of formed different levels is depending to the electronic configuration and the coordination number of the cation. The photon absorption by electronic transitions between levels "3d" causes discoloration in the visible range [10].

Most metal ions transitions "3d" are in tetrahedral coordination or octahedral in oxide glasses from which the change in the coordination number will be obtained with the energy differences, depending on the number of electrons present 3d. The color is also affected by the concentration of the cation dye, the effect of the concentration of the dye ion is clear: the species resulting dyes with more absorption; the effects of changes in the network trainers ions and ions present modifiers are due to alteration in the distance of the strips and the force between the coloring ions and surrounded ligands.

Thermal properties

It can be seen that glass transition (Tg) and softening (Ts) temperatures decreased with increasing concentrations of Fe_2O_3 by reducing of B_2O_3 (Figures 2 and 3). This is due to the decreased bond strength intensity of the various glass matrix components. Previous studies were carried on some glasses have been proven this finding [6]. The addition of iron oxide contributed to creation of non-bridging oxygen so the coefficients of expansion increased.

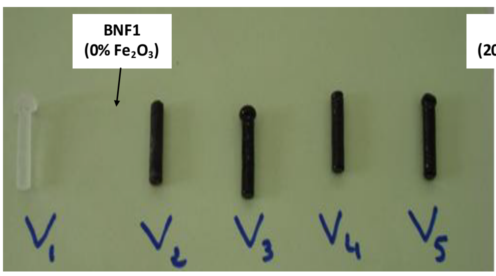

Figure 1: Aspect of glass samples..

Physical properties

Table 2 gives respectively the density and the molar volume of all studied glasses. It is noted that the density of glass samples increased with the addition of iron oxide and sodium oxide. This is explained by filling the voids between the structural units (silica tetrahedra, boron, boron triangles), thus decrease the molar volume and density increase (Figure 4). Given that such concentrations, the reference glass (BNF1) which is a sodoborate glass has a lower density compared to other glasses.

Properties of studied glasses

According to the calculation methods, there is an increase in the coefficients of thermal expansion as and is added sodium and iron oxide, and by reducing the boron oxide as the structure becomes less rigid and network modifiers contribute to the creation of non-bridging oxygen which increases the expansion of the network (Table 3).

The refractive index is an optical property related to the polarizability of oxygen atoms, we also notice an increase in the refractive index as we add sodium and iron oxide which promotes the creation of non-oxygen bridging and therefore their polarizability increases. Increases were observed in the values of modulus of elasticity, and this behavior is similar to the same coefficient of thermal expansion (non-bridging oxygen's of creation by adding modifiers network) and the opposite fish louse coefficient (decrease). Decreases observed in the values of mechanical strength and this is due to the creation of non-bridging oxygen that caused voids in the glass structure so open and less rigid structures.

As the thermal conductivity is a property related to transport phenomena in matter, we note in the case of our glasses, an increase in value with the increase in modifier ions (Na^+) and iron oxide that facilitating the transfer of thermal conductivity through the vitreous mass.

The electrical permittivity is also related to the polarizability of oxygen ions, which notes the increase in its value with the increase in non-bridging oxygen's created with the Fe_2O_3 addition.

Conclusion

In the glass system B_2O_3-Na_2O, the addition of iron oxide (Fe_2O_3) leads to the change of coloration and properties of studied glass. Fe_2O_3 has contributed to creation of non-bridging oxygen. So, the glass structure is weakened that transition and softening temperatures decrease, densities and coefficients of dilatation have significantly increased.

The creation of non-bridging oxygen led to an increase of the elastic modulus and consequently an improvement the mechanical properties as the traction and the compression resistance.

The increase of the polarizability of the ions oxygen has to lead to the increase of the refractive index and the as well as the electric permittivity. On the other hand, it can be noted that the properties of transfer have increased such as the thermal conductivity.

Nomenclature

α: Thermal dilation coefficient, 10^{-6}, k^{-1}

nd: Index of refraction

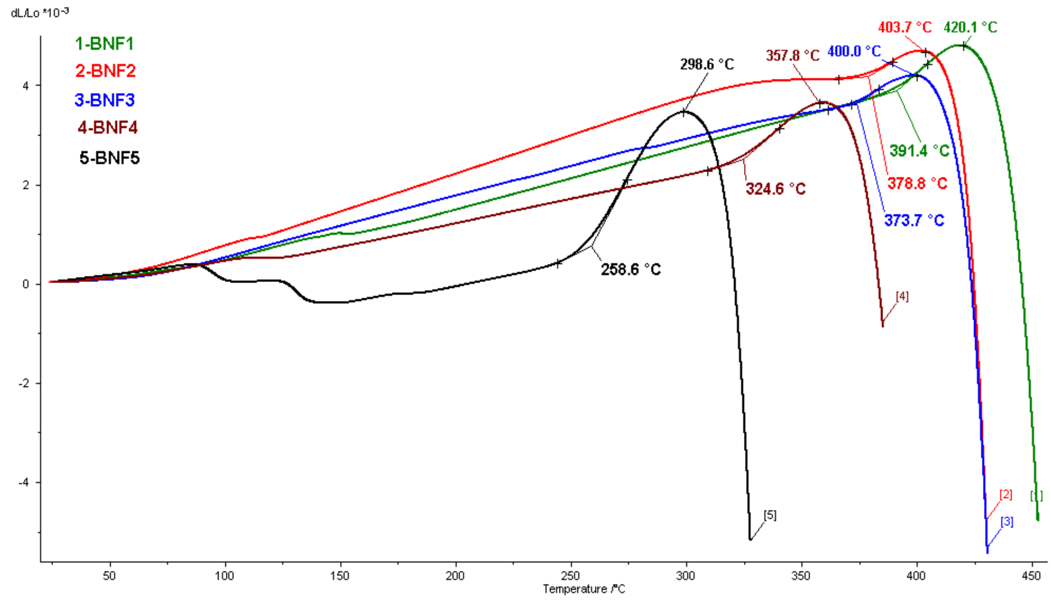

Figure 2: Curves of dilatation thermal of studied glasses.

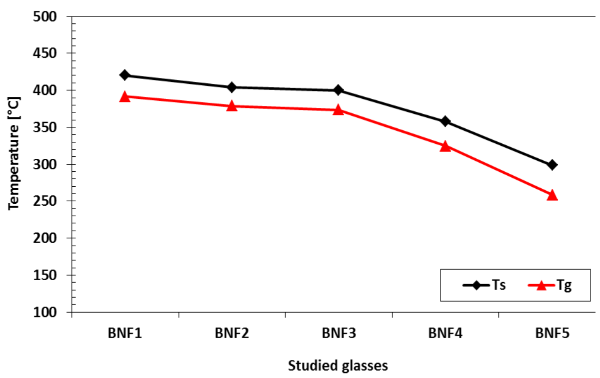

Figure 3: Variation of the glass transition temperatures (Tg) and dilatometric deformation (Ts) of studied glasses.

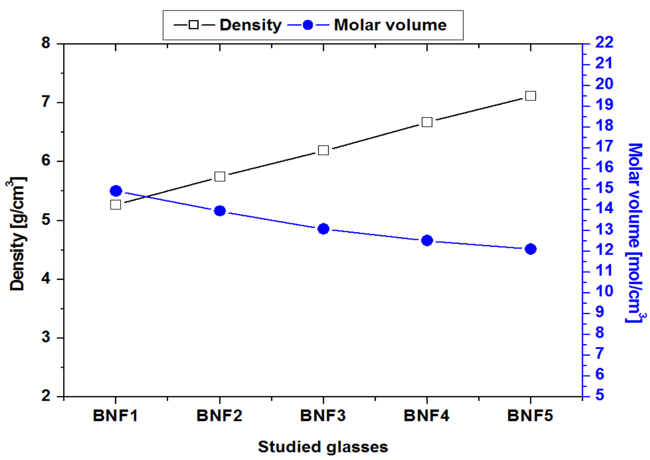

Figure 4: Density and molar volume of glasses studied.

	B_2O_3 [wt.%]	Na_2O [wt.%]	Fe_2O_3 [wt.%]
BNF1	**95**	**5**	**0**
BNF2	**85**	**10**	**5**
BNF3	**75**	**15**	**10**
BNF4	**65**	**20**	**15**
BNF5	**55**	**25**	**20**

Table 1: Chemical compositions of studied glasses (BNF).

Glass	D_{glass} (g/cm³)	Mv (cm³/mol)
BNF1	5,26	14,91
BNF2	5,75	13,95
BNF3	6,19	13,09
BNF4	6,67	12,52
BNF5	7,11	12,11

Table 2: Density and Molar volume of glasses studied.

Glasses	α	n_d	E	σ	δt	δc	λc	ε
BNF1	1 .977	1.47	260	97.4	67.30	863	0.048	6.42
BNF2	4.280	1.49	312	147	65.80	843	0.053	7.84
BNF3	6.580	1.53	350	173.8	64.50	793	0.058	7.96
BNF4	8.882	1.54	381	206	64.35	757	0.061	8.12
BNF5	11.179	1.57	420	249.7	63.00	747	0.066	8.57

Table 3: Calculated properties of studied glasses.

E: Longitudinal modulus of elasticity, kbar

σ: Surface tension 10^{-3}, N/m

σc: Compressive stress (σc) or with traction (σt), MN/m²

Cp: Heat capacity, J/gk

λc: Thermal conductivity, W/mk

ε: Electric permittivity

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