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Temperature Dependence of Dielectric Conductivity of Borosilicate Glasses Modified by Iron Oxide

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Abstract

The temperature dependent conductivity of xFe_2O_3. (43-x) B_2O_3. $25SiO_2$. $30Na_2O$. $2Al_2O_3$ glass system in the composition range of 1 - 12 mol% Fe_2O_3 was measured over temperature range from 40-400°C. The observed dependence follows simple Arrhenius relation as evidenced by the exponential variation of conductivity with temperature. The mechanism responsible for DC conductivity is primarily governed by the transport of sodium ions and electronic hopping of iron charge careers through the localized glass network. The electrical conductivity was found to be increased with raising temperature due to the thermal activation of the charge carriers. Both DC conductivity and activation energy values were significant effected by Fe_2O_3 addition- especially at higher content- which may be correlated with the variation in the two distinct conduction mechanisms present in the studied glass system.

Keywords: Borosilicate glasses, Iron oxide, Electrical properties, Structural units, DC conductivity, Arrhenius formula

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اعتماد الموصلية الكهربائية على درجة الحرارة لزجاج البورسليكات المعدل بأكسيد الحديد

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المخلص

تم قياس التوصلية الكهربية المعتمدة على درجة الحرارة لزجاج $xFe_2O_3.(43-x)B_2O_3.25SiO_2.30Na_2O.2Al_2O_3$ في نطاق درجات حرارة يتراوح بين 40 و400 درجة مئوية. يتبع هذا الاعتماد علي معادلة أرينيوس , كما يتضح من السلوك الأسّي للتوصيلية مع درجة الحرارة. يمكن ربط الآلية المسؤولة عن التوصيلية الكهربية عبر شبكة الزجاج المحلية بحركة أيونات الصوديوم والقفز الإلكتروني لأيونات الحديد. من خلال النتائج، وُجد أن التوصيل الكهربائي يزداد مع ارتفاع درجة الحرارة، نظرًا لتنشيط حاملات الشحنة حراريًا. بالإضافة، تأثرت كلا من التوصيلية الكهربية وطاقة التنشيط بشكل كبير بإضافة أكسيد الحديد خاصة عند التركيزات العالية والتي قد تكون مرتبطة بآليتي التوصيل المختلفتين اللتين تعتمدان بشكل مباشر على تركيب الشبكة الهيكلية للزجاج قيد الدراسة.

الكلمات المفتاحية: زجاج البوروسيليكات، أكسيد الحديد، الخصائص الكهربية، الوحدات التركيبية، التوصيلية الكهربية، معادلة أرينيوس

Introduction

Borosilicate glasses have attracted high scientific significance (LIMA et al., 2012;EL-DAMRAWI et al.,2016; CIŻMAN et al,2020; Gharghar & El-Jadal,2022) due to their desirable chemical and physical features including a low thermal expansion coefficient and softening temperature, and their high hardness and chemical corrosion resistance. The presence of both B_2O_3 and SiO_2 within borosilicate network creates a mixed glass former impact on the structure and properties. Adding chemical modifiers to the borosilicate glasses plays a significant role in further modifying its structural and physical properties. In particular, glasses formulated with transition metal oxides, have attracted a significant interest due to their optical and electrical properties, making them suitable for

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various technological applications. Among these, Fe_2O_3 is regarded as one of the most potential candidates for enhancing the glass properties, particularly in the area of electrical, optical, and magnetic characteristics. (EL-DAMRAWI et al.,2016; LIMA et al., 2012; Gharghar& El-Jadal,2022; CIŻMAN et al,2020, CIŻMAN et al,2017). Several researchers have investigated glasses of various compositions containing Fe_2O_3 and demonstrated that borosilicate glasses containing Fe_2O_3 exhibit an enhancement of the structural, optical and magnetic properties as well as electric behavior. El-Damrawi G.et al (EL-DAMRAWI et al.,2016) revealed that the presence of low Fe_2O_3 content is consumed to modify borosilicate network based on the creation of non-bridging oxygens (NBOs) in silicate network and the transformation of BO3 to BO4 species. Moreover, changes in the network connectivity accompanied by increasing amount of Fe_2O_3 are attributed to the formation of iron former units at the expense of BO4 groups and NBOs. CIŻMAN, A., et al (CIŻMAN et al,2017) reported that, increasing Fe content at a constant Na concentration in the studied glass system causes a stronger formation of [FeO4/2]-Na^+ linkage rather than those in sodium-borate [BO4]-Na^+ species.

Nowadays, ionic and electronic conduction mechanism (LENARCIAK et al.2019; CIŻMAN et al,2017) in the disordered glass materials have garnered large theoretical and experimental attention. The disordered structure of these materials significantly affects their overall electrical conductivity, which is primarily governed by the modifier ion concentration as well as the mobility of charge carriers. Previous studies (LENARCIAKet al.2019; CIŻMAN et al,2017; RYSIAKIEWICZ-PASEK et al. 2020) have proposed that the migration of alkali ions in glass occurs through a thermally activated hopping mechanism, which strongly affected by both the concentration of defects and the mobility of these defects within the glass network. Besides, the electrical conductivity of oxide glasses containing Fe_2O_3 is characterized by the presence of iron in two different Fe^{2+} and Fe^{3+} ions.

The main objective of this study is to analyze the impact of Fe_2O_3 replacing B_2O_3 on the electrical conductivity of sodium borosilicate glasses based on the relevant properties that have been extensively examined and reported in previous studies (EL-DAMRAWI et al.,2016; Gharghar & El-Jadal,2022).

Experimental procedure

Material Preparation

The studied series of borosilicate glasses has the formula of $xFe_2O_3.(43-x)\ B_2O_3.25SiO_2..30Na_2O.2Al_2O_3$ (with x= 1 - 12 mol% Fe_2O_3) as shown in table.1. Reagent grade of high purity SiO_2, H_3BO_3, Na_2CO_3, Al_2O_3 and Fe_2O_3 was melted under normal atmospheric conditions in alumina crucibles at a temperature range from 1250˚C-1520˚C. The homogeneous melt was then poured into a ceramic plate or steel plate and pressed by another plate to obtain glass discs at room temperature.

Table.1. The prepared samples compositions (mol%) of the formula $xFe_2O_3.(43-x)B_2O_3.\ 25SiO_2.30Na_2O.2Al_2O_3$.

Fe_2O_3	0	1	2	4	6	10	12
B_2O_3	43	42	41	39	37	33	31
SiO_2	25	25	25	25	25	25	25
Na_2O	30	30	30	30	30	30	30
Al_2O_3	2	2	2	2	2	2	2

Electrical Conductivity characterization

Samples for DC conductivity measurements were prepared in the form of discs with radius 0.8 cm and thickness varied between 0.2 and 0.5 mm. The conductivity measurements were conducted over a temperature range from room temperature to 400°C.

Figure 1 indicates the conductance cell used for the electrical conductivity measurements (Abdelghany,2009).

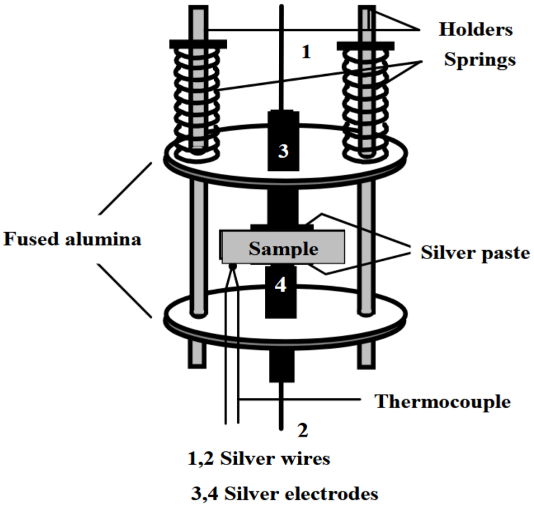

Figure 1: The electrical conductivity cell

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The conductivity (σ) of the prepared samples is defined as a function of the thickness of the sample l, the measured resistance R and the coated area a as follow:

$$\sigma = \frac{l}{Ra} \quad (1)$$

Result and discussions

The temperature dependence of DC conductivity was measured at a temperature in 40˚C to 400˚C range for all investigated samples. **Figure 2** depicts the dependence of the logarithm of DC conductivity on the reciprocal of temperature (1/T). As can be observed, log σ decreases linearly with increasing 1/T which reveals the ionic nature of the electric conduction. In such a case, Arrhenius formula has been used to describe the DC conductivity dependence on its temperature:

$$\sigma = \sigma_o \exp(-E/kT) \quad (2)$$

Where: σ_o, E, K, and T denote the frequency factor, the activation energy, the Boltzmann constant and the absolute temperature respectively.

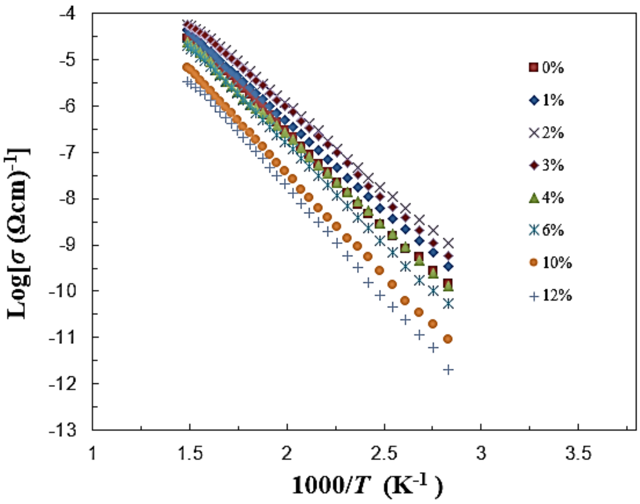

Figure 2:Temperature dependence of the electric conductivity of the studied glass samples

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Figure 3 represents the electric conductivity (at T=200,250,300, and 350°C) as a function of Fe_2O_3 content. There can also be a noticeable trend of increasing DC conductivity with increasing temperature due to increment of the thermally activated drift mobility of charge carriers during hopping process. Thus, the temperature increment enhances the mobility of hopping ions which in turn leads to an increase in the DC conductivity.

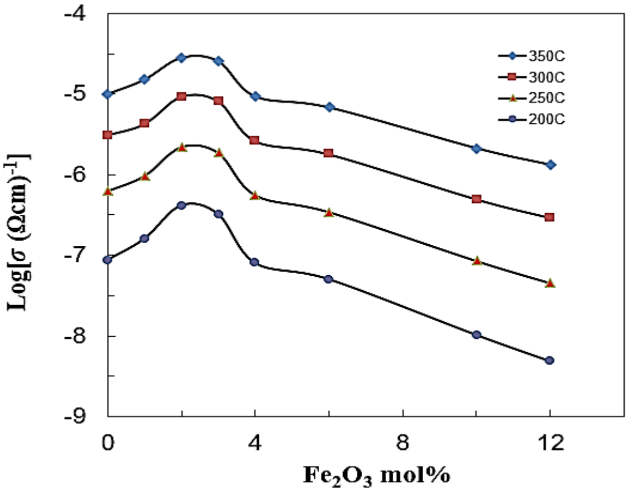

Figure 3: Temperature dependence of the natural logarithm of the electric conductivity (at T=200,250,300,350°C) as a function of Fe_2O_3 content (mol%).

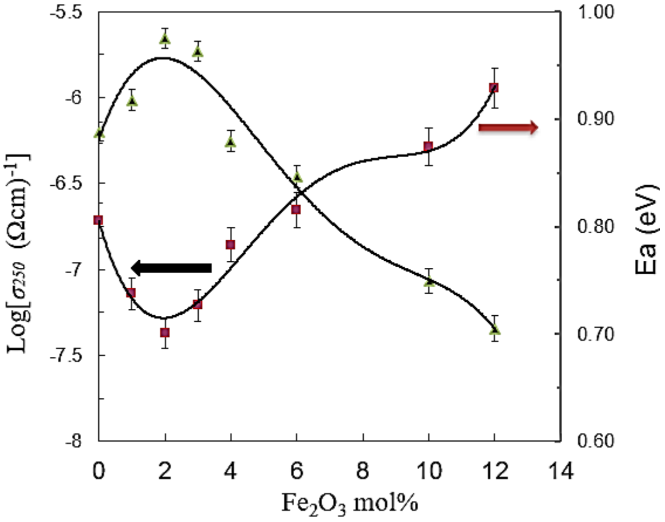

Figure 4: The natural logarithm of the electric conductivity at 250°C and the activation energy E as a function of Fe_2O_3 content (mol%).

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Both changes in the natural logarithm of the DC conductivity at 250°C and the activation energy E with iron oxide are shown in Figure 4.
Overall, it was found that (MEIKHAIL & ABDELGHANY,2017; LENARCIAK et al.2019; CIŻMAN et al,2017), the electrical properties of iron-containing sodium borosilicate glasses can be attributed to the migration of sodium ions and the electron hopping between the Fe^{2+} and Fe^{3+}valence states. As it can be a notable. there are two regions of change, in the first region, a significant increase in the electrical conductivity can be characterized by a relatively low iron oxide. In such a case, iron oxide modifies the borosilicate network by breaking part of the Si–O bonds and creating bridging bonds in borate network. The presence of NBOs in the glass network has been shown to enhance the migration of Na^+ ions (EL-DAMRAWI et al.,2016; Eremyashev et al,2014; CIŻMAN et al ,2020). As a result, the DC conductivity is expected to increase and sodium ions could be incorporated into the borosilicate network and became the dominant charge carriers. The second region where the highest rate of variation in conductivity is observed by adding a higher content of iron oxide. The substitution of B_2O_3 by Fe_2O_3 (EL-DAMRAWI et al.,2016; Eremyashev et al,2014) involved the formation of Fe-O-Si and Fe-O-B linkages at the expense of non-bridging bonds in both the borate and the silicate Networks. As a result, creation of more bridging bonds into the glass network would affect the electrical conductivity since both number of charge carriers and their mobility would be affected. Hence, decreasing the mobility of the charge species with increasing Fe_2O_3 content may be considered due to increasing more bridging oxygen atoms in both the borate and the silicate sites.
Figure 5 shows a linear reduction of log σ with rising the activation energy. This indicated that the conductivity is dominated by the activation energy which depends mainly upon the type and concentration of interconnected bonds in the glass network. As it is discussed in previous work, more bridging bonds will increase with increasing Fe_2O_3 contents thereby the mobility of the charge carriers would decrease and they need higher energy to migrate.

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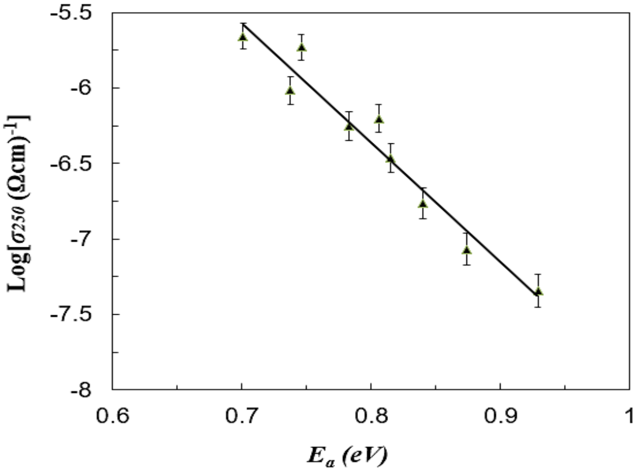

Figure 5: Correlation between the natural logarithm of electric conductivity at 250°C and the activation energy E of the studied glasses

Conclusion

The findings of the investigated work of iron-doped sodium borosilicate glasses can be summarized as follows:

- The electrical conductivity of sodium borosilicate glasses containing iron oxide can be described by two mechanisms: The migration of sodium ions and the hopping of electrons between iron oxidation states.
- The mobility of charge carriers increases through the thermal activation with a possible enhancement of the hopping rate, thereby promoting an increase in the electric conductivity.
- The electric conductivity, as well as activation energy values show a strong dependence on the type of the iron oxide role in the glass network.
- It is expected that there is a strong correlation between the variation of the structure and the electrical conduction mechanism of the modified borosilicate network.
- The production of NBO's plays the essential role of increasing ion mobility in the borosilicate network. The high ability of sodium ions to migrate within the glass structure is responsible for the conductivity processes in glasses with low concentration of iron.
- On the other hand, increasing more bridging bonds upon replacing B_2O_3 by Fe_2O_3 would affect the electrical conduction of the glass, since the movement of charge carriers would be affected. Thus, for samples with higher iron content, the

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electrical conductivity is controlled by the structural rearrangement caused mainly by the competition between FeO4 and BO4 units.

- The variation of activation energy monotonically with the charge carrier's mobility leads to conclusion that the conductivity in this case is mobility dependence.

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