Turkish Journal of Physics

http://journals.tubitak.gov.tr/physics/

Research Article

Turk J Phys
(2014) 38: 136 – 144

doi:10.3906/fiz-1306-13

# Structural, physical, and electrical properties of boro-vanadate-iron glasses doped with $K_2O$ alkali

**Harshvadan R. PANCHAL***
Department of Chemical and Physical Sciences, University of Toronto Mississauga, Mississauga, Ontario, Canada

**Received:** 24.06.2013 • **Accepted:** 14.11.2013 • **Published Online:** 17.01.2014 • **Printed:** 14.02.2014

**Abstract:** The structural, physical, and electrical properties of semiconducting oxide glasses with composition $xK_2O$: (95-X) [$B_2O_3$: $2V_2O_5$]: $5Fe_2O_3$ ($0 \leq x \leq 30$) have been studied. The glasses were prepared by splat-quenching method. Fourier transform infrared spectroscopy (FTIR) studies of these glasses were carried out to study the effect of the modifier and glass former in the glasses. These changes in the molecular structure, bond length, and vibrational group were studied in the spectral range of 400–1350 $cm^{-1}$. The increase in the intensity and shifting of vibrational bands occurred towards lower wave numbers at 1400–1350 $cm^{-1}$, 1200 $cm^{-1}$, and 100–940 $cm^{-1}$. These structural changes were compared with the electrical conductivity data to see a more clear effect of the glass modifier $K_2O$ in the network formed by glass former $V_2O_5$-$B_2O_3$. Electrical conductivity of these glasses was studied for all glasses and results were explained on the basis of Mott's theory of conduction mechanism. A decrease in the conductivity with increase of V-V spacing was observed, which was also confirmed by structural changes observed in the FTIR results of these glasses. The present glasses exhibited the structural changes and confirmed the n-type of semiconducting nature with adiabatic hopping due to polarons.

**Key words:** Electrical conductivity, polaron, activation energy, nonbridging oxygen, adiabatic hopping

## 1. Introduction

Extensive studies have been carried out on semiconducting oxide glasses containing transition metal oxides for their glass structure and electrical conduction mechanisms [1–5]. These semiconducting glasses have been extensively studied due to their various applications in the fields of electrical switching, memory switching, and optical and cathode materials for making solid-state devices and optical fibers [6–11]. The addition of any transition metal oxide makes glasses semiconducting in nature and electrical conduction is observed due to the hopping of electrons [1,3]. Glasses containing $V_2O_5$ are semiconducting in nature and exhibit stable switching, whereas $V_2O_5$-$B_2O_3$ glasses are widely used as soldering and molding materials [11]. Recent studies by many researchers have focused on the electrical behavior of these glasses and structure studies using various techniques.

Fourier transform infrared spectroscopy (FTIR) is one of the best tools for looking into the structure of these glasses. Various bonding energies of solids may be found by infrared absorption. Pure vibrational spectra are observed in the range of 400–160 $cm^{-1}$, whereas separation of the vibrational energy level is larger when observed at high frequencies, i.e. lower wave numbers. Infrared spectroscopies of pure $B_2O_3$-$V_2O_5$ have been

*Correspondence: hr_panchal@yahoo.com

extensively investigated [3,4]. In boro-vanadate glasses, structure, distribution, and interaction between V-O-V and B-O-B polyhedrals play very important roles in electron transport. Effects of nonbridging oxygen during formation of glasses have great impacts on the structure of glasses and electrical conduction process [12]. The electrical conduction in these glasses was also explained on the basis of structure properties and changes in structures that occurred in glasses, which was supported by infrared and physical studies. The present study is focused on the structural, physical, and electrical conductivity of a boro-vanadate-iron glass system containing different amounts of $K_2O$ alkali.

The present study of conduction mechanism was made on a $K_2O$: $V_2O_5$: $B_2O_3$: $Fe_2O_3$ glass system. $Fe_2O_3$ was added as a Mossbauer probe. In the present paper, electrical properties of an $xK_2O$: (95-X) [$B_2O_3$: $2V_2O_5$]: $5Fe_2O_3$ glass system have been examined in the temperature range of 315–435 K and an attempt has been made to elucidate the contribution of $K_2O$ and $V_2O_5$ to the structure and electrical conduction process in the glasses.

The DC conductivity of transition metal oxide glass was expressed in [1] with the following equation:

$$\sigma = \left[\frac{\vartheta_0 e^2 c\,(1-c)}{kTR}\right] \exp\left(-2\,\alpha\,R\right) \exp\left(-\frac{W}{kT}\right), \tag{1}$$

where $\nu_o$ is the optical phonon frequency ($\approx 10^{13}$ Hz), e is the electronic charge, c is the mole fraction of the site occupied by the electron, R is the average hopping distance, $\alpha$ is the electron wave function decay constant such that exp (–2 $\alpha$ R) represents an electron overlap integral and hence the probability of tunneling, W is the activation energy, and T is the temperature.

$$\begin{aligned} \text{Note:} W &= W_H + (1/2)W_D \quad \text{for} \quad T > \theta_D/2 \\ &= W_D \qquad\qquad\qquad\quad T < \theta_D/4 \end{aligned} \tag{2}$$

where $W_H$ is the polaron hopping energy, $\theta_D$ is the Debye temperature, and $W_D$ is the disordered energy.

The polaron hopping energy $W_H$ is given by:

$$W_H = \frac{e^2}{4\varepsilon_p}\left[\frac{1}{\nu_p} - \frac{1}{R}\right], \tag{3}$$

where $\varepsilon_p = (1\ /\ \varepsilon_\alpha - 1\ /\ \varepsilon_o)^{-1}$ and $\varepsilon_o$ and $\varepsilon_\alpha$ are static and high-frequency dielectric constant for the glass, respectively. $\nu_p$ is the polaron radius, which is given by [13]:

$$\nu_p = \frac{1}{2}\left(\frac{\pi}{6N}\right)^{1/3}, \tag{4}$$

where N is the number of sites per unit volume.

For a polaron to be small, the polaron radius $\nu_p$ should be smaller than the atomic site spacing (V-V spacing) R [1].

For a nonadiabatic hopping process $J < \hbar\omega_o$, where J is the electron transfer integral, and for adiabatic hopping, $J > \hbar\omega_o$. This approximation is given by Holstein [14].

$$J \begin{array}{c} > \\ < \end{array} \left[\frac{2\ \mathrm{kT}\ \mathrm{W_H}}{\pi}\right]^{1/4} \left[\frac{h\nu_o}{\pi}\right]^{1/2} \begin{array}{l} \text{Adiabatic } (>) \\ \text{Nonadiabatic } (<) \end{array} \tag{5}$$

J can be estimated from the difference of the mean value of hopping energy and experimental activation energy W.

## 2. Experimental

### 2.1. Sample preparation

Analytical grade chemicals $V_2O_5$, $H_3BO_3$, $K_2CO_3$, and $Fe_2O_3$ were taken as raw materials, which were weighed in appropriate amounts to prepare glass samples $xK_2O$:

(95-X) [$B_2O_3$: $2V_2O_5$]: $5Fe_2O_3$ ($0 \leq x \leq 30$) in steps of 5 mol%. Weighed amounts were mixed well and transferred to a porcelain crucible, which was maintained for 1 h at 250 °C to remove the moisture. The temperature of the furnace was then raised to 950 °C and maintained for 4 h. These melts were then quenched onto a clean copper plate at room temperature and subsequently pressed by another plate to provide a faster quenching rate. The X-ray diffraction analysis showed that quenched samples were amorphous.

### 2.2. Density measurement

The density of the glass samples was measured at room temperature by displacement method using methanol as the immersion liquid and a single pan balance of $10^{-5}$ g sensitivity. The density measurements were done to determine the number of metal ions per unit volume (i.e. vanadium ions) in the glasses. The number of transition metal ions was determined by assuming the glass structure as uniform, which helped in calculating the polaron radius [1,2] by knowing the number of atomic sites per unit volume as per Eq. (4).

$$\nu_p = \frac{1}{2}\left(\frac{\pi}{6N}\right)^{1/3}$$

Here, N is the number of sites per unit volume (i.e. $V_{total}$ ions in $V_2O_5$).

### 2.3. FTIR measurement

The glass samples were crushed into a fine powder and small amount of glass powder was mixed and ground with a relatively large amount of KBr, which is transparent to IR radiation. Disks for measurement of IR absorption spectra were prepared by pressing a mixture at a pressure of 10–15 t for few minutes under vacuum. The infrared spectra of quenched samples were taken with a PerkinElmer 577 infrared spectrometer in the range of 4000–400 $cm^{-1}$.

### 2.4. Electrical conductivity measurement

The samples were cut in the form of rectangular pellets of about 2 mm in thickness. These samples were lapped and polished. Silver paste was painted on the polished rectangular surface of the sample. After polishing, samples were kept at 90 °C for 90 min to remove moisture and stress from the sample. With the painted silver paste, good ohmic contacts were found. These samples were kept between 2 probes of a sample holder and the cell was kept in a furnace, which was controlled by a mercury contact thermometer (JUMO, Germany) and relay. The electrical conductivity of all the glass samples at different temperatures was measured (315–435 K).

## 3. Results and discussion

### 3.1. X-Ray diffraction and density

Among the various characterization techniques, X-ray diffraction plays a very key role in structure characterization. The diffraction patterns were taken from 0° to 70° with a Cu$\alpha$ target. The diffraction pattern showed only big halos with no characterization peak, which illustrates the amorphous nature of all of the prepared glasses (Figure 1).

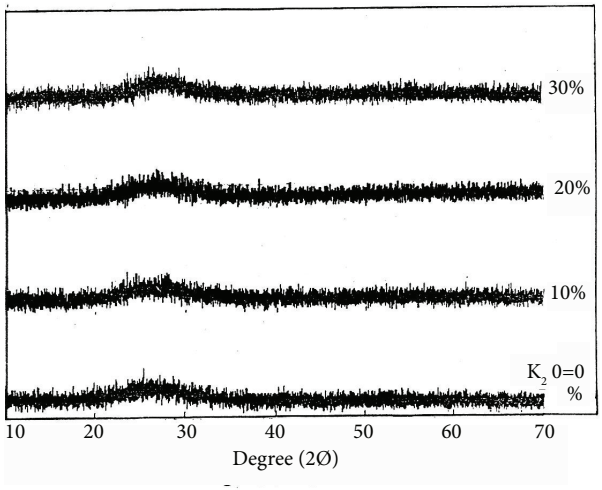


**Figure 1.** X-ray diffraction pattern of the $xK_2O$: (95-X) [$B_2O_3$: $2V_2O_5$]: $5Fe_2O_3$ glass system.

The density of the present glass system was measured using the Archimedes principle. The calculated values of parameters using density measurements are shown in the Table 1, where it can be observed that, as the amount of modifier $K_2O$ is increased, the density of glass samples is also increased. The calculated values of spacing between vanadium atoms (V-V spacing) was found to be increase from 4.16 to 4.52 Å with the content of $K_2O$. These variations in density and hence V-V spacing help in understanding the electrical conductivity behavior of these glasses.

**Table 1.** Density, spacing between vanadium (V-V) and boron (B-B), and polaron radius for $xK_2O$: (95-X) [$B_2O_3$: $2V_2O_5$]: $5Fe_2O_3$ glass system.

| $K_2O$ mol% | Density (g/mL) | $\nu_P$ (Å) | $W_H$ (eV) | B-B spacing (Å) | V-V spacing (Å) | Fe-Fe spacing (Å) |
|---|---|---|---|---|---|---|
| 0 | 2.65 | 1.67 | 0.327 | 5.37 | 4.16 | 11.33 |
| 5 | 2.71 | 1.68 | 0.332 | 5.54 | 4.18 | 11.18 |
| 10 | 2.73 | 1.70 | 0.336 | 5.45 | 4.22 | 11.08 |
| 15 | 2.75 | 1.72 | 0.339 | 5.52 | 4.27 | 11.00 |
| 20 | 2.77 | 1.74 | 0.342 | 5.60 | 4.33 | 10.91 |
| 25 | 2.83 | 1.76 | 0.347 | 5.65 | 4.37 | 10.76 |
| 30 | 2.70 | 1.82 | 0.343 | 5.85 | 4.52 | 10.86 |

### 3.2. FTIR

Figure 2 shows FTIR spectra of pure $B_2O_3$, $V_2O_5$, and $Fe_2O_3$, which are considered for the purpose of comparison. It is seen from the spectrum (line a) in Figure 2 that $B_2O_3$ has characteristic absorption bands at 1471 $cm^{-1}$, 1200 $cm^{-1}$, 780 $cm^{-1}$, and 653 $cm^{-1}$. The peaks at 1471 $cm^{-1}$ and 1200 $cm^{-1}$ are assigned to $BO_3$ triangles [12,13,15]. The peaks at 780 $cm^{-1}$ and 653 $cm^{-1}$ are attributed to bending vibrations of the B-O-B band [15,16]. Figure 2 also shows the FTIR spectrum (line b) of pure $V_2O_5$, which has characteristic features at 1020 $cm^{-1}$, 850 $cm^{-1}$, and 613 $cm^{-1}$. The sharp band at 1020 $cm^{-1}$ is assigned to the vanadyl group of the V = O bond, whereas 850 $cm^{-1}$ is related to symmetric stretching vibrations along the V-O-V chain involved in corner sharing of the $VO_5$ polyhedra [16]. A very weak absorption peak at 613 $cm^{-1}$

corresponds to the V-O-V bond of the symmetrical or bending mode of vibrations. Figure 2 for $Fe_2O_3$ has the characteristic feature at 563 $cm^{-1}$ attributed to the vibrations of $FeO_4$ groups of $Fe_2O_3$.

The effect of modifier $K_2O$ were studied by keeping $V_2O_5$ and $B_2O_3$ at a ratio of 2:1 for fixed amounts of $Fe_2O_3$ at 5 mol%. The FTIR spectra of all glass samples for $K_2O$ = 0 mol% to 30 mol% are given in Figure 3. The main features of these spectra are the appearances of resonance peaks at 1430 $cm^{-1}$, 1353 $cm^{-1}$, 1192 $cm^{-1}$, 1253 $cm^{-1}$, 1104 $cm^{-1}$, and 1000–940 $cm^{-1}$ and very weak broad bands at 804–602 $cm^{-1}$ and 860–651 $cm^{-1}$. A close examination of the spectra reveals a very small shift of the vibrational band at 1430 $cm^{-1}$ up to 20 mol% $K_2O$. When the amount of $K_2O$ is further increased, the vibrational band at 1430 $cm^{-1}$ splits into 2 vibrations, 1353 $cm^{-1}$ and 1435 $cm^{-1}$ , which are assigned to characteristics of the B-O-B linkage in which both borons are triangularly coordinated or – B – B < stretching vibrations [15]. The peaks at 1435 $cm^{-1}$ and 1353 $cm^{-1}$ are slightly shifted to 1437 $cm^{-1}$ and 1350 $cm^{-1}$ wavenumbers at 30 mol% $K_2O$. In the pure borate glasses, the main structural element is a boroxol ring of plane trigonal configuration with B-O bond length of 1.36 ± 0.005 Å, whereas the B-O bond length for the $BO_4$ tetrahedra was 1.47 ± 0.01 Å. In the spectra, the increase in bond length is seen as the shifting of an absorption band toward the lower wavenumber. In binary borate glasses, up to 14 mol% $K_2O$ does not break bridging in the $BO_3$ groups to form $BO_4$ tetrahedra [15,16]. One $K_2O$ causes the formation of 2 $BO_4$ tetrahedra, which participate in a 3-dimensional network, thus

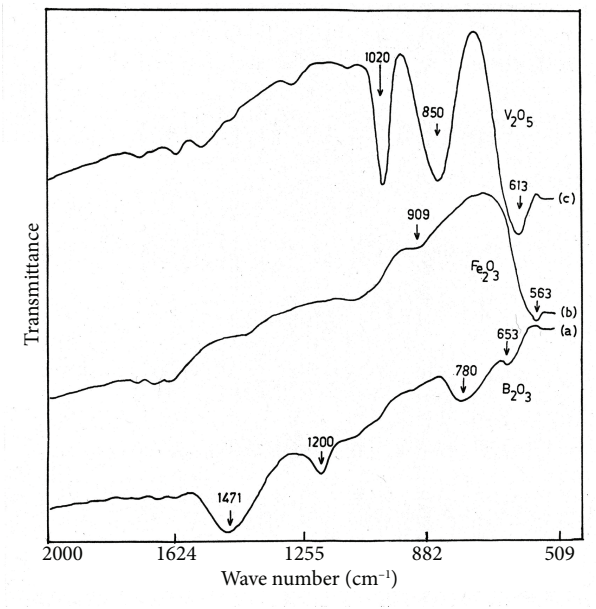


**Figure 2.** FTIR spectra of pure (a) $B_2O_3$, (b) $V_2O_5$, and (c) $Fe_2O_3$.

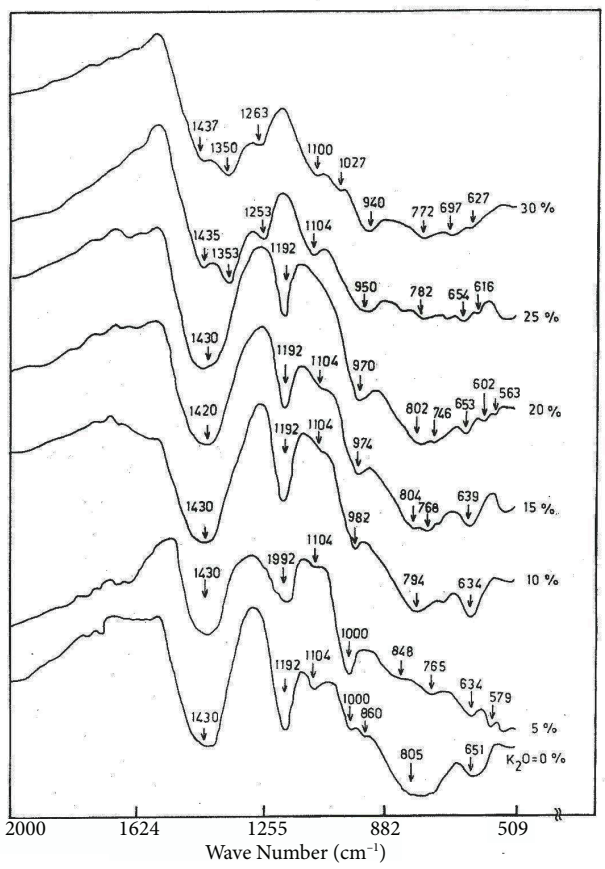


**Figure 3.** FTIR spectra of $xK_2O$: (95-X) [$B_2O_3$: $2V_2O_5$]: $5Fe_2O_3$ glass system for $K_2O$ = 0 to 30 mol%.

strengthening the structure [17]. The B-O-B separation has been observed to increase, which was calculated from density measurements [15], as shown in Table 1.

Another feature of the IR spectra is the presence of a highly intense peak at 1192 $cm^{-1}$, which is assigned to $BO_3$ polyhedra. This peak that disappeared after 20 mol% $K_2O$ shows a presence of a new peak at 1253 $cm^{-1}$, which is assigned to the = B-O-B $\equiv$ linkage in which boron is tetrahedrally coordinated [15,16]. For all glass samples from $K_2O$ = 0 to 30 mol%, a vibrational peak at 1104 $cm^{-1}$ is observed, which is assigned to the vibrations of nonbridging oxygen in the form of = B-$O^-$ or B – O = B < $O^-$ [15]. The vibrational bands at 1000–940 $cm^{-1}$ in the spectra are assigned to the isolated V = O double bond of the $VO_5$ polyhedra with the increase of $K_2O$. This vibrational band shifting towards the lower wavenumber suggests an increase in bond length of the V = O bond. This is due to the fact that the added $K_2O$ goes into the structure at an interstitial position, leaving its oxygen. Now K+ ions directly interact with the oxygen of the V = O bond, thereby weakening the frequency of vibration shifts toward lower wavenumbers. Added $K_2O$ gives rise to the formation of a nonbridging oxygen atom, thereby weakening the V = O bond. The presence of a weak and wide absorption band at 860 $cm^{-1}$ and 848–634 $cm^{-1}$ suggests that it is the sum of the number of absorption bands corresponding to the V-O-V band of slightly varying bond lengths and angles [18–20]. Thus, the formation of nonbridging oxygen results in the increase of distance between V-V atoms, which is also supported by the density measurement data shown in Table 1. This variation of V-V spacing is also supported by the electrical conductivity data, which are explained in Section 3.3.

## 4. Electrical conductivity

The glass modifier plays a very significant role in the formation of glasses and hence changes the properties of glasses like electrical conductivity, thermal switching, and density. The temperature dependence electrical conductivity of the present glass system has been explained with a theory based on polaron hopping conduction [1]. Dependence of electrical conductivity on the amount of glass modifier has also been studied by many researchers [11,21,22]. In the present glass system, the amount of glass modifier is increased from 0 mol% to 30 mol%, whereas the amount of $V_2O_5$ is decreasing with a simultaneous decrease of $B_2O_3$. The amount of $Fe_2O_3$ has been kept constant in all glass system. Figure 4 shows the dependence of conductivity on the amount of modifier $K_2O$. The added $K_2O$ goes into the glass structure by occupying an interstitial position in the $V_2O_5$-$B_2O_3$ network. Because of the breaking of the $V_2O_5$ structure, atomic site spacing between V-V increases. The V-V spacing calculated from the density measurements shows a change from 4.16 to 4.52 Å. The conductivity of the present glass system is also found to increase from 1.59 $\times$ $10^{-5}$ to 2.04 $\times$ $10^{-7}\Omega^{-1}$ $cm^{-1}$ (at 100 °C) with the increase of activation energy from 0.407 to 0.536 eV. A similar type of behavior was also observed and discussed by Devidas et al. [21] and Sanchez et al. [23]. Thus, with the increase of $K_2O$ amount, the atomic site spacing between V-V is found to increase, which directly affects the mobility of a charge carrier. Additionally, $V^{+4}/V_{total}$ is observed to be dependent on $K_2O$ amount. Figure 5 shows the plot of $\log\sigma$ (at 100 °C) and activation energy (W) versus V-V spacing. The observed activation energies for all glass samples were found to be slightly higher than the value obtained for other $V_2O_5$-based glasses [21,22]. The variations of conductivity are observed in all glasses from $10^{-6}$ to $10^{-8}\Omega^{-1}$ $cm^{-1}$ and $\log\sigma$ versus $10^3/T$ is linear (Figure 6), which shows the semiconducting behavior of the present glass system. The Seebeck measurements were also studied for the present glass system [24,25]. Mori et al. reported $V_2O_5$-$Sb_2O_3$-$TeO_2$ and $V_2O_5$-$Bi_2O_3$-$TeO_2$ glasses as n-types of semiconductors [26,27].

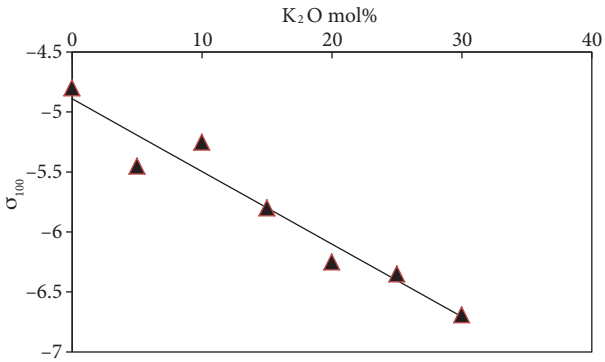


**Figure 4.** The dependence of conductivity $\sigma_{100}$ on the amount of modifier $K_2O$ mol%.

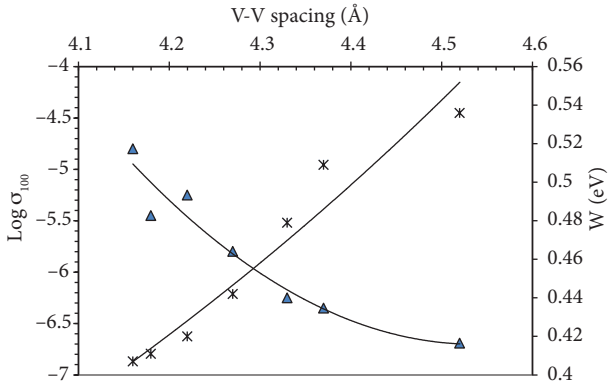


**Figure 5.** Plot of $\log\sigma$ (at 100 °C) and W (eV) versus V-V spacing.

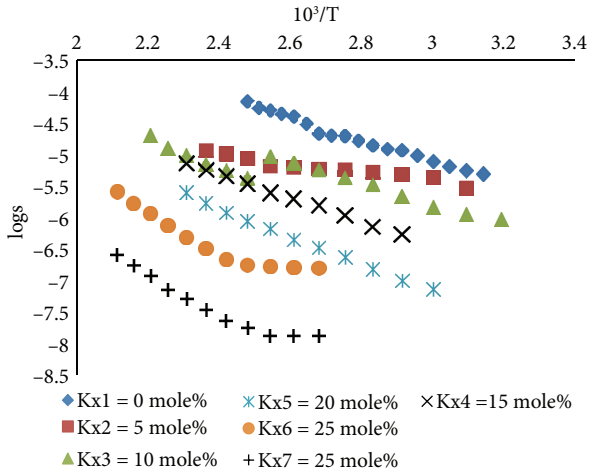


**Figure 6.** The dependence of conductivity $\log\sigma$ versus 1000/T for the $xK_2O$: (95-X) $[B_2O_3: 2V_2O_5]$: $5Fe_2O_3$ glass system.

For a polaron to be small, the polaron radius should be greater than the radius of the iron on which the electron is localized but less than the distance R separating 2 sites. As is evident for the present glass system (Table 2), the value of the polaron radius lies between 1.67 and 1.82 Å, which is less than R, the atomic site separation 4.16–4.52 Å. This satisfies the condition for small polaron hopping. Similar results were also reported for PbO-$P_2O_5$-$V_2O_5$ glasses [27].

Holstein [14] suggested that the hopping process is controlled by the activation energy and is given by Eq. (5). Calculated values of $W_H$ lie in the range of 0.327–0.343 eV (from Table 2). As per Eq. (5), the values of the right-hand side were calculated for an optical phonon frequency of approximately $10^{13}$ Hz. The values lie in the range of 0.0124–0.0126 eV. These values are less than the value of J, which lies in the range of 0.0689–0.01973 eV, satisfying the condition for adiabatic hopping in different types of glasses. Hirashima et al. also reported adiabatic hopping in PbO-$P_2O_5$-$V_2O_5$ glasses [27], as did El-Desoky in $V_2O_5$-$B_2O_3$-BaO glasses [28].

Thus, in the present glass system, the effect of the glass modifier plays an important role in the conductivity behavior, which is supported by FTIR and density measurement data. The increase of $K_2O$

amounts in the glass network modified the structure and led to an increase of V-V spacing, thereby decreasing the conductivity of these glasses. The n-type of the semiconducting nature of the glasses was also confirmed by Seebeck measurements of the present glass system [24,25].

**Table 2.** Electrical conductivity parameters and other related parameters for $xK_2O$: (95-X) [$B_2O_3$: $2V_2O_5$]: $5Fe_2O_3$ glass system.

| $K_2O$ mol% | W (eV) | J | $\left(\frac{2kTW_H}{\pi}\right)^{1/2}\left(\frac{h\vartheta_o}{\pi}\right)^{1/2}$ (eV) | $\sigma_{100}$ ($\Omega^{-1}$ cm$^{-1}$) | V-V spacing (Å) | Mobility ($\mu$) |
|---|---|---|---|---|---|---|
| 0 | 0.407 | 0.0689 | 0.0124 | $1.59 \times 10^{-5}$ | 4.16 | $4.70 \times 10^{-7}$ |
| 5 | 0.411 | 0.0729 | 0.0125 | $3.55 \times 10^{-6}$ | 4.18 | $7.50 \times 10^{-8}$ |
| 10 | 0.420 | 0.0819 | 0.0125 | $5.62 \times 10^{-6}$ | 4.22 | $1.11 \times 10^{-7}$ |
| 15 | 0.442 | 0.1039 | 0.0126 | $1.59 \times 10^{-6}$ | 4.27 | $3.10 \times 10^{-8}$ |
| 20 | 0.479 | 0.1409 | 0.0126 | $5.62 \times 10^{-7}$ | 4.33 | $9.00 \times 10^{-9}$ |
| 25 | 0.509 | 0.1709 | 0.0126 | $4.47 \times 10^{-7}$ | 4.37 | $7.00 \times 10^{-9}$ |
| 30 | 0.536 | 0.1979 | 0.0126 | $2.04 \times 10^{-7}$ | 4.52 | $3.10 \times 10^{-9}$ |

## 5. Conclusion

The structural and electrical properties of $xK_2O$: (95-X) [$B_2O_3$: $2V_2O_5$]: $5Fe_2O_3$ ($0 \leq x \leq 30$) mol% have been studied. The structural changes in these glasses were observed due to the formation of nonbridging oxygen, which was due to formation of $BO_4$ from the $BO_3$ group. Spacing between V-V was found to increase as a result of the increase of $K_2O$ content. These results also support the electrical conductivity data. Furthermore, the increase in V-V spacing inhibits the mobility of small polarons as well as the hopping of energy. This results in the decrease of conductivity from $1.59 \times 10^{-5}$ to $2.04 \times 10^{-7} \Omega^{-1}$ cm$^{-1}$ (at 100 °C) and the increase of activation energy from 0.407 to 0.536 eV. This change is also confirmed from the IR spectroscopy measurement of the present glass system. The present glass system confirmed that the n-type of semiconducting nature and conduction is due to polaron hopping, which is adiabatic in nature.

## References


[1] Mott, N. F. *J. Non-cryst. Solids* **1968**, *1*, 1–17.

[2] Al-Hajry, A.; Al-Shahrani, A..; El-Desly, M. M. *Mater. Chem. Phys*, **2006**, *95*, 300–306.

[3] Sindhu, S.; Sanghi, S.; Agarwal, S. A.; Seth, V. P.; Kishore, N. *Physica B* **2005**, *365*, 65–75.

[4] Sen, S.; Ghosh, A. *J. Non-cryst. Solids* **1999**, *258*, 29–33.

[5] Austin, I. G.; Mott, N. F. *Adv. Phys,* **1969**, *18*, 71, 41–102.

[6] Darke, C. F.; Scanlan, L. F.; Enegel, A. *Phys. Status Solidi* **1969**, *32*, 193. Mansingh, A.; Singh, R.; Krupanidhi, S. B. *Solid State Electron.* **1980**, *23,* 649–654.

[7] Montani, R. A.; Levy, M. J.; Souquet, L. *J. Non-cryst. Solids* **1992**, *149*, 249–156.

[8] Panchal, H. R.; Kanchan, D. K. *Turk. J. Phys.* **1999**, *23*, 969–976.

[9] Mekki, A.; Khattak, G. D; Siddiqui, M. N. *Int. J. Mod. Phys. B* **2010**, *24*, 1471–1488.

[10] Wozniak, I.; James, P. F. *Glass Technol.* **1984**, *25*, 98–104.

[11] Sanad, A. M.; Kashif, I.; El-Sharkawy, A. A.; El-Saghir, A. A.; Farouk, H. J. *Mater. Sci.* **1986**, *21*, 3490.

[12] Kashif, I.; Farouk, H.; Aly, A. S.; Sanad, A. M. *Phys. Chem. Glasses* **1991**, *32*, 77–78.

[13] Holstein, T. *Ann. Phys.* **1959**, *8*, 34.

[14] Li, H.; Lin, H.; Chen, W.; Luo, L. *J. Non-cryst. Solids* **2006**, *352*, 3069–3073.

[15] Kanchan, K.; Panchal, H. *Turk. J. Phys.* **1998**, *22*, 989–996.

[16] Bratu, I.; Oana, P.; Culea, M. *Phys. Status Solidi A* **1987**, *100*, K195–K198.

[17] Bachmann, H. G.; Ahmed, F. R.; Barnes, W. H. *Z. Kristallogr.* **1961**, *115*, 110–131.

[18] Assem, E.; Elmehasseb, I. J. Mater. Sci. **2011**, *46*, 7, 2071–2076.

[19] Kim, Y. Y.; Kim, K. H.; Choi, J. S. *J. Phys. Chem. Solid* **1989**, *50*, 903–908.

[20] Devidas, G. B.; Sankarappa, T.; Chougule, B. K.; Prasad, G. *J. Non-cryst. Solids* **2007**, *353*, 426–434.

[21] Dixit, V. G. ; Singh, K. *J. Phys. Chem. Solids* **1983**, *44*, 859–864.

[22] Sanchez, C.; Linage, J. *J. Non-cryst. Solids* **1984**, *65*, 285–300.

[23] Panchal, H. R.; Kanchan, D. K. In *Proceedings of the DAE Solid State Physics Symposium*, University of Kurukshetra, Kurukshetra, India, 27–31 December 1998; Mukopadhyay, R. K.; Saikh, A. M.; Godwal, B. K., Eds.; Hyderguda, Hyderabad, India: Universities Press (India) Limited, **1999**, p. 256–257.

[24] Panchal, H. R. PhD, Department of Physics, The Maharaja Sayajirao University of Baroda, Gujarat, India, 2000.

[25] Mori, H.; Sakata, H. *J. Mater. Sci. (UK)* **1996**, *31*, 1621.

[26] Hirashima, H.; Nishi, K.; Yoshida, T. *J. Am. Ceram. Soc.* **1985**, *68*, 486–489.

[27] El-Desoky, M. M. *Phys. Status Solidi A* **2003**, *195*, 422–428.