



Impact of SrO ions on dielectric properties and ac conductivity of Vanadoborate glasses

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Abstract

A series of borate glasses containing V_2O_5 and SrO were prepared by melt quenching technique and investigated for dielectric properties in the frequency range 100Hz to 5.5MHz and ac electrical conductivity in the temperature range 313 K to 473 K. The high-temperature electrical conduction data were analysed following Mott's small polaron hopping theory. Both ϵ' and ϵ'' decreased with increase in SrO concentration and increased with increase in temperature and decreased with increase in frequency. The ac conductivity decreased and activation energy increased with increase of SrO concentration at all frequencies. This may hint that strontium ions may not at all contribute the total conductivity and the observed conductivity may be due to hopping of polarons alone.

Keywords: Borate Glasses; Transition metal ions; Strontium ions; AC conductivity

1. Introduction

Over the last few decades, the researchers have shown much interest on oxide glasses such as B_2O_3 , V_2O_5 , SrO, P_2O_5 and TeO_2 due its exceptional properties and potential use for technological applications viz. Solid-state batteries, sensors, fuel cells and electrochromic display devices [1,2]. Among the oxide glasses, B_2O_3 based glasses have drawn much attention on the technological interest because of greater bond strength, small cationic dimension and low heat fusion [3]. These glasses are well-thought-out to be structurally more promising and establish them as good candidates in doping them with varied amounts of alkali, alkaline and transition metal oxides are doped, the boroxol rings broken and borate units BO_3 and BO_4 are formed [4].

Borate glasses doped with high concentration of transition metal ions (TMI) viz. V_2O_5 , CuO , NiO , Fe_2O_3 , CoO and leads to electronic conduction which usually arises by the transfer of electron from ions in the lower valence state to higher valence states respectively [5,6]. This categorizes them as a form of highly disordered amorphous semiconductors. The electronic conduction mechanism in borate glasses containing different TMIs in single and mixed proportions has been reported by many researchers [7-9]. Electronic conduction in such glasses show mixed electronic and ionic conductivity and moreover they also exhibit changeover of conduction mechanism from electronic to ionic or vice versa at a particular composition [10]. Transport mechanism of electrons in these glasses containing TMI was explained by the hopping of electrons between low valence states to high valence states of TMI which is termed as small polaron hopping [11]. However, some oxide glasses incorporated with TMI have been shown interesting properties in terms of resistivity, frequency dependent AC conductivity and dielectric properties at some temperatures [12].

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Numerous studies in terms of DC electrical conduction, structure of Strontium and other vanadate glasses have been made with vanadium pentoxide greater than 0.5 mole fraction [13-15]. Very few studies have been reported on glasses containing lower molar fraction of V_2O_5 in presence of B_2O_3 and SrO glass matrix [14-16]. These mentioned glasses have technological applications such as optical amplifiers in telecommunications, optical fibers, electrochemical batteries, electrical memory switching and cathode materials for making solid-state devices [17-19].

In the present communication, we report the information on effect of incorporation of SrO in vanadium borate glasses with the help of impedance spectroscopy technique and investigated the ac electrical conductivity and dielectric properties over a wide range of temperature and frequency scale in the following mentioned compositions, $0.5(B_2O_3) - (0.5-x) (V_2O_5) - x(SrO)$ where $x=0, 0.1, 0.2, 0.3, 0.4$. These glasses are coded as BVS1, BVS2, BVS3, BVS4 and BVS5. The main objective of the present work is to understand the impact on ac electrical conductivity and dielectric properties by the addition of SrO at the cost of V_2O_5 content.

2. Experimental Method

The melt-quenching method has been employed to synthesize the present glass samples. For this, the required analytical grade reagents, namely, V_2O_5 (99.9) SrO (99.5) and H_3BO_3 (99.99%) were procured from Himedia Chemicals. The chemicals with suitable composition were mixed thoroughly in order to obtain homogeneity. The mixture was then taken in a silica crucible and melted between the temperatures ranging from 1423 K to 1473 K to get different glass compositions. After confirming the formation of homogenous transparent melt, the melt was then transferred on to a thick steel plate and quickly quenched by pressing with another similar steel plate. Different sized sample pieces were collected. The samples were annealed at 473 K to remove thermal strains, if any, in them.

The frequency related parameters such as, capacitance, C , and dissipation factor, $\tan \delta$, were measured by means of LCR meter (ZM2376, NF Corporation, Japan) for various frequencies ranging from 100 Hz to 5.5 MHz and temperature starting from 313 K to 473 K. The dielectric constant (ϵ'), dielectric loss factor (ϵ'') and ac conduction (σ_{ac}) were determined using the equations [20],

$$\epsilon' = \frac{Cd}{\epsilon_0 A} \quad (1)$$

$$\epsilon'' = \epsilon' \tan \delta \quad (2)$$

$$\sigma_{ac} = \omega \epsilon_0 \epsilon'' \quad (3)$$

where, ϵ_0 is the permittivity of free space, d is thickness of the glass sample and A is cross-sectional area of the sample.

3. Results and Discussion

3.1. Dielectric constant and loss

The plots of variation of $\ln(\epsilon')$ with respect to $\ln(F)$ for all BVS glass systems with respect to composition at 463 K is shown in Fig. 1. The measured ϵ' and ϵ'' values were found to lie in the range of 10^2 to 10^3 and 10^3 to 10^4 respectively. These measured values of ϵ' and ϵ'' are comparable to those in the literature [21,22]. It can be observed from Figs. 1 and 2 that both ϵ' and ϵ'' decreases with increase in frequency and increases with increase in temperature [23]. The decrease in ϵ' and ϵ'' as a function of frequency indicates that at higher frequencies the alkali ions don't follow the field variation and also because of the interfacial effects i.e., occurrence of space charge polarization [20].

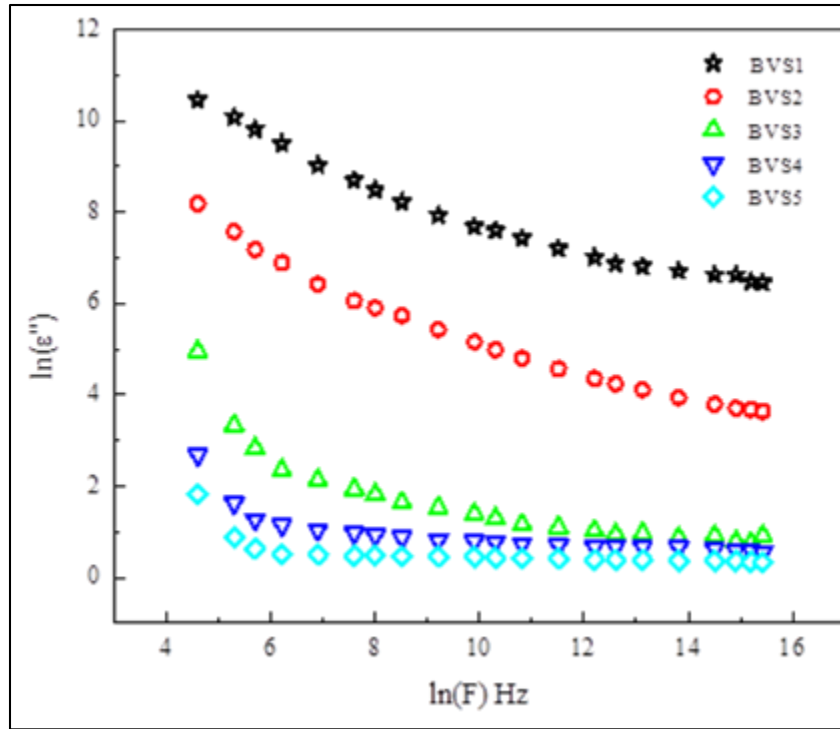


Figure 1 Plot of $\ln(\epsilon'')$ versus $\ln(F)$ at 463 K temperature for all BVS series glasses

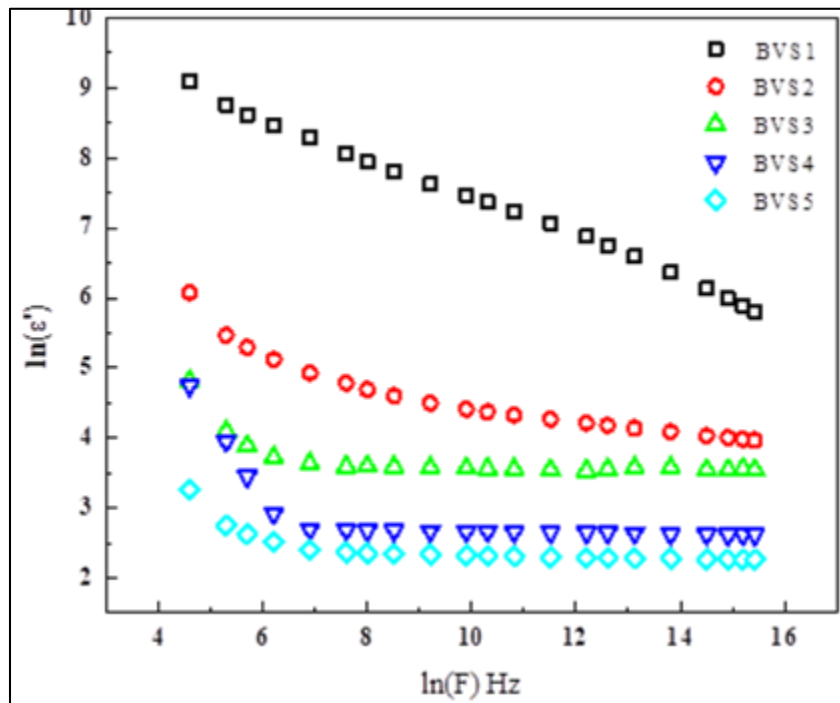


Figure 2 Plot of $\ln(\epsilon')$ versus $\ln(F)$ at 463 K temperature for all BVS series glasses

Figs. 3 and 4 represents the plot of ϵ' vs frequency and ϵ'' vs frequency at different temperatures for BVS4 glass series. From these curves, it is observed that both ϵ' and ϵ'' decreases with increase in frequency and increase with increase in temperature. This kind of behavior may be due to the fact that the interfacial dipoles don't have adequate time to orient themselves in the electric field directions [24]. The nature of ϵ' and ϵ'' with frequencies at a particular temperature for all the other glasses of the BVS series exhibited similar behavior.

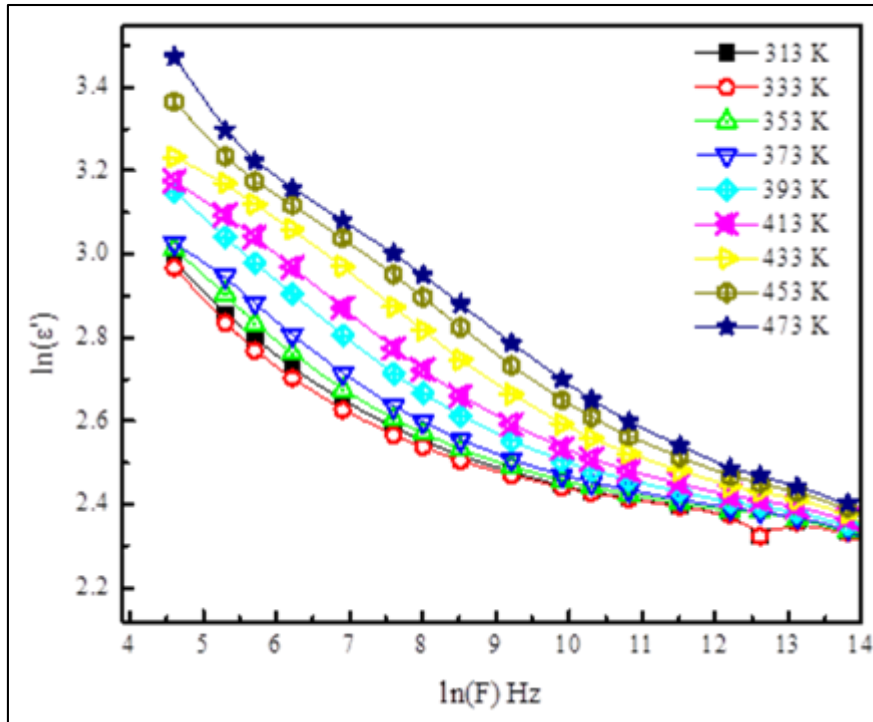


Figure 3 Plot of $\ln(\epsilon')$ vs $\ln(F)$ for BVS4 glass sample at different temperatures

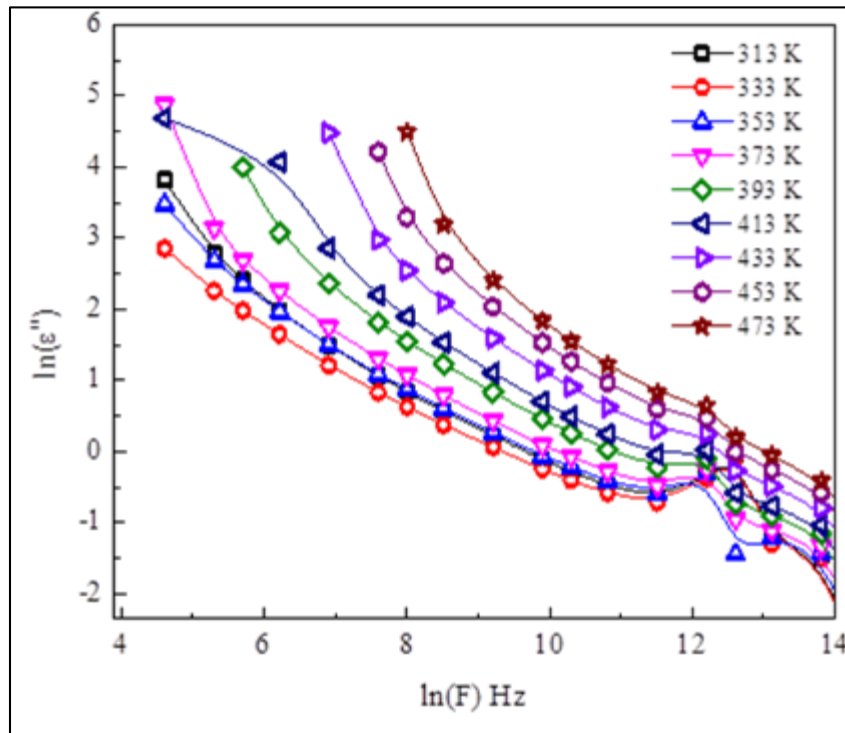


Figure 4 Plot of $\ln(\epsilon'')$ vs $\ln(F)$ for BVS4 glass sample at different temperatures

In order to study the compositional effect of SrO concentration on ϵ' and ϵ'' of the BVS glasses, the plot of variation of ϵ' and ϵ'' (Inset) with molar fraction of SrO content measured at 500 kHz and temperature at 453 K is depicted in Fig. 5. From this figure, it can be noted that ϵ' and ϵ'' decreases with increase in SrO concentration. This may be due to the fact that the decrease in electronic contribution to the total polarizability [25].

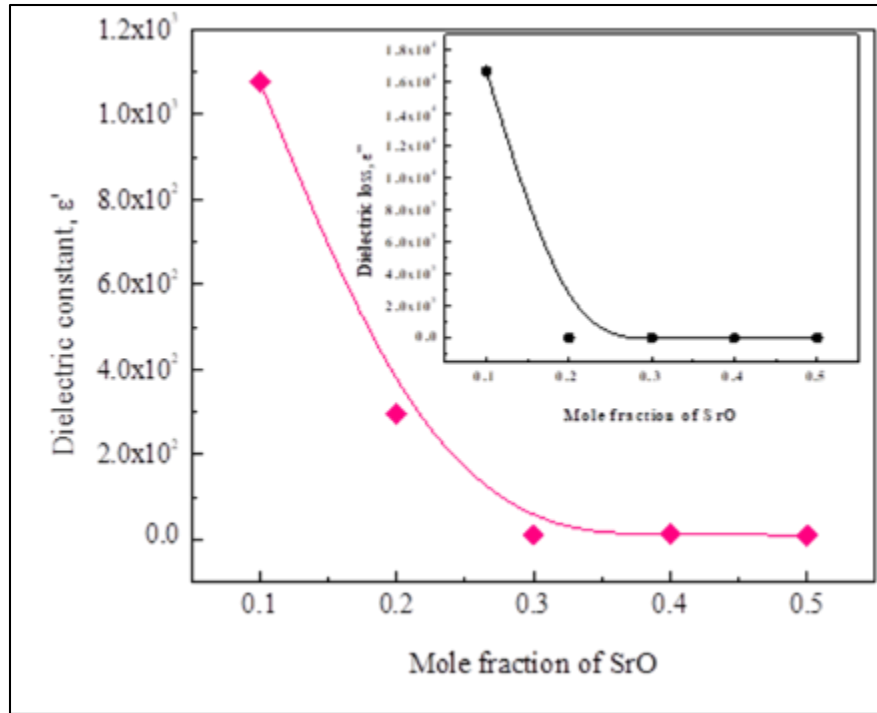


Figure 5 Compositional variation of dielectric constant, ϵ' at 500 kHz and temperature, 453 K for BVS glass series. Inset plot represents the compositional variation of dielectric loss, ϵ'' at 500 kHz and temperature, 453 K for BVS glass series. Solid lines are guides to the eye

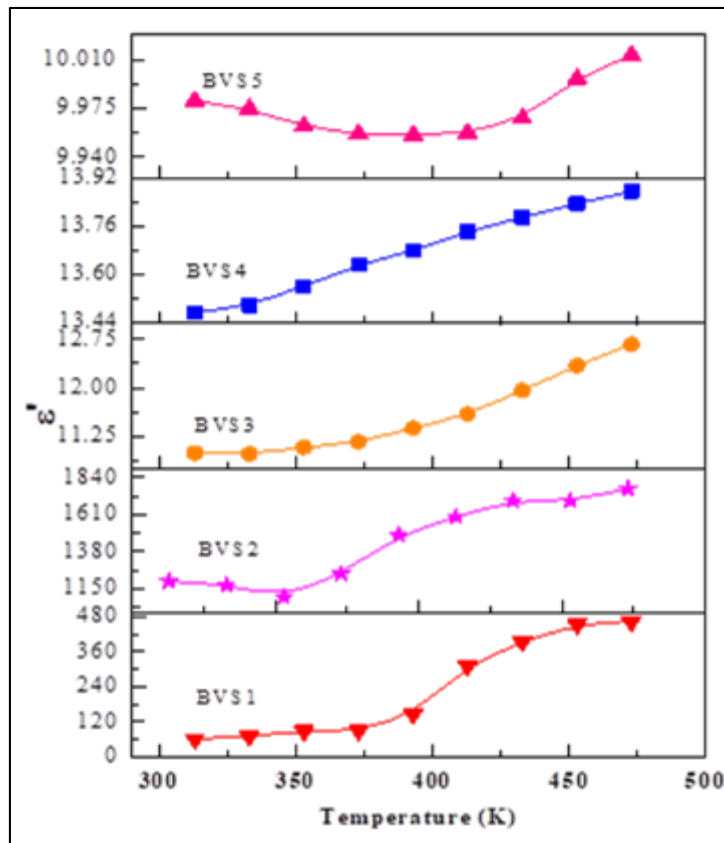


Figure 6 Variation of dielectric constant (ϵ') versus temperature at 1 kHz for different molar fractions of BVS glass series

Fig. 6 represents the variation of ϵ' measured at 1 kHz and at different temperatures for the BVS glass series doped with different concentrations of SrO. From this figure, it can be seen that as temperature increases the increase in ϵ' was observed for all the BVS series glasses. This is due to the fact that the orientational polarization is subjected to the thermal movement of molecules, meanwhile at low temperatures the dipoles cannot orient themselves. Consequently, as the increase in temperature, the orientation of the dipoles turns out to be easier and therefore an increase in the orientational polarization is observed, which subsequently increases the ϵ' [26]. The nature of variation of ϵ'' with temperature is found to be similar for the BVS glass series.

3.2. Conductivity

The temperature dependent variation of ac conductivity, σ_{ac} in the present glasses is understood to be due to the thermal stimulation process and is also due to the hopping of electrons among the different valency states of vanadium⁺ charge carriers in the glass network [27]. Mott's SPH model has been used for explaining the high temperature dependent ac conductivity. The graph of $\ln(\sigma_{ac}T)$ versus $(1000/T)$ for different frequencies for BVS4 glasses are presented in Fig. 7. The σ_{ac} values were found to be in the order of $10^{-5} \Omega^{-1}m^{-1}$ to $10^{-7} \Omega^{-1}m^{-1}$. These values are nearly comparable with the σ_{ac} values reported in our previous work [28] and other glass systems [6,29].

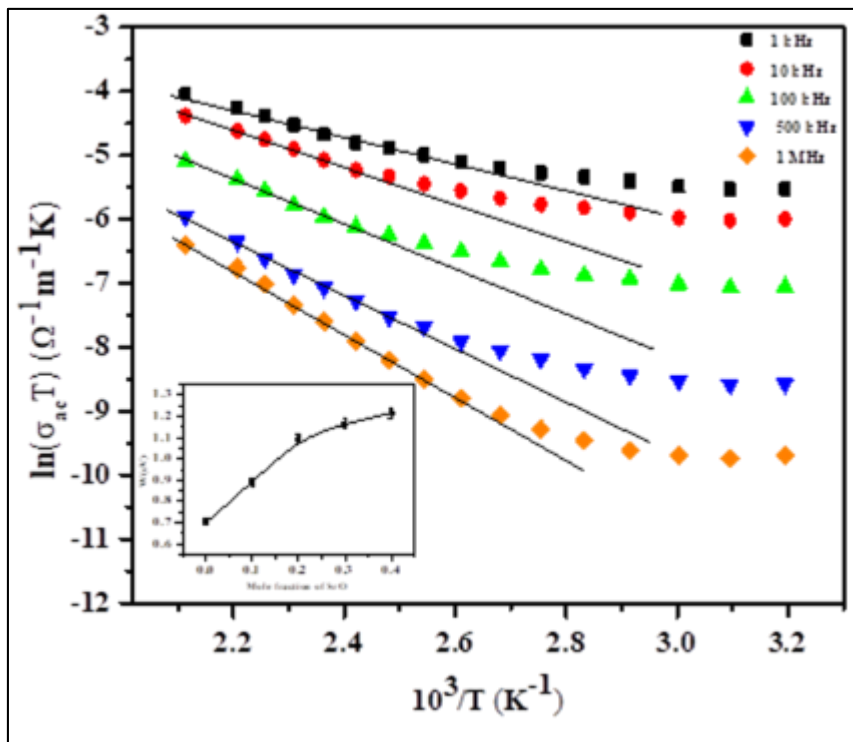


Figure 7 Plot of ac conductivity at different frequencies for the glass sample BVS4. The lines are the linear fits to the data. Inset plot represents the variation of activation energy with composition

As per the Small Polaron Hopping theory, the electrical conduction in non-adiabatic system is given as [30],

$$\sigma = \frac{\sigma_o}{T} \exp\left(-\frac{E_{ac}}{k_B T}\right) \quad (4)$$

where, E_{ac} is the activation energy and σ_o is the pre-exponential factor.

The profiles of $\ln(\sigma_{ac}T)$ against $(1000/T)$ at a different frequency for BVS4 glasses are made as per Eq. (4) and shown in Fig. 7. The least square lines were fit to the data in the high temperature (above T_D) region. The slopes of linear fits were determined and activation energies, E_{ac} , were determined. The E_{ac} values lies between 0.70 eV to 1.21 eV.

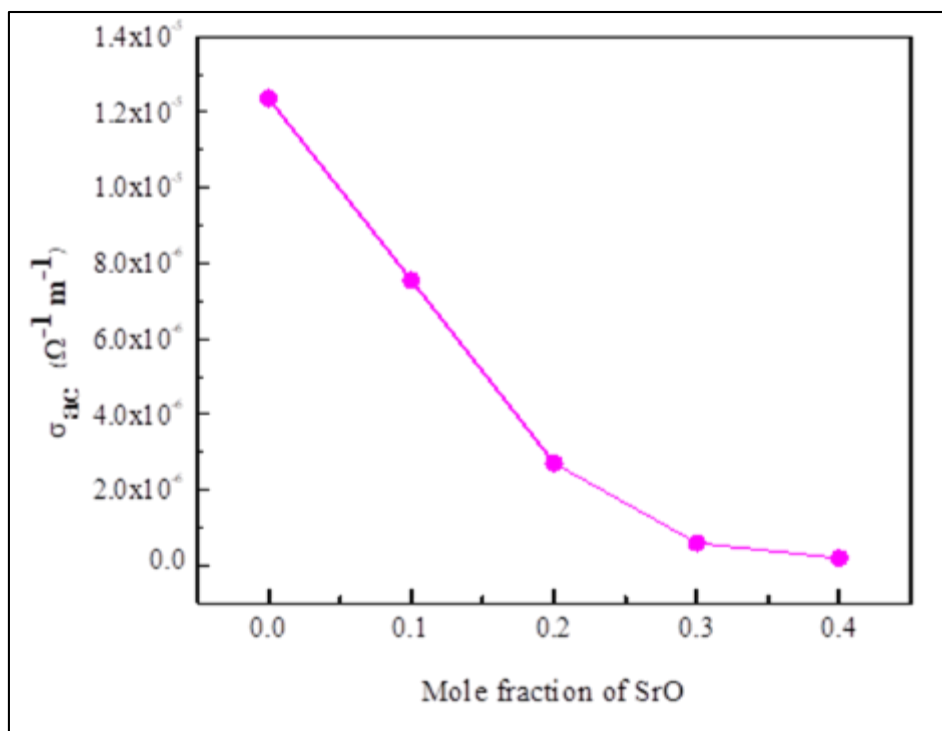


Figure 8 Variation of σ_{ac} versus molar fraction of SrO at 463 K for BVS glasses

Fig. 8 illustrates the variation of ac conductivity at 463 K as a function of the glass composition. It can be noticed from the figure that the conductivity decreases with increase in the SrO concentration. In the present glass system, the increase in activation energy from 0.70 eV to 1.21 eV was revealed that the electrical conduction was hindered by SrO that the conductivity abruptly falls as soon as the SrO is brought into the glass network. Similarly, the activation energy is found to increase rapidly for SrO doped glass samples (Fig. 7 inset). This type of variation in conductivity and activation energy indicates that strontium ions are not at all contributing to the total conductivity. Hence, the observed conductivity may be electronic alone which is due to the hopping of electrons between multivalent states of vanadium. Similar variation has been observed in terms of the dc conductivity studies as reported in our work [28, 31].

4. Conclusion

A set of borate glasses of the composition $0.5(B_2O_3) - (0.5-x) (V_2O_5) - x(SrO)$ were synthesized using the melt-quenching method. For the present series of glasses were investigated for compositional, frequency and temperature dependence of dielectric and ac conductivity over a wide range. Dielectric properties and ac conductivity have been measured in the temperature range 313 K to 473 K and frequency from 100 Hz to 5.5 MHz. Both ϵ' and ϵ'' decreased with increase in SrO concentration. This is attributed to the fact that decrease in electronic contribution to the total polarizability. As temperature increases the increase in both ϵ' and ϵ'' and decreased with increase in SrO concentration were observed.

The Mott's SPH model has been employed to analyze the high temperature ac conductivity data. The ac conductivity decreased and activation energy increased with increase of SrO concentration at all frequencies. Based on these results it is concluded that the strontium ions may not at all contribute the total conductivity and the observed conductivity may be due to hopping of polarons alone due to the presence of V_2O_5 .

Compliance with ethical standards

Disclosure of conflict of interest

No conflict of interest to be disclosed.

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