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Studies of conductivity in mixed alkali vanadotellurite glasses

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A set of novel mixed alkali vanadotellurite glasses $(V_2O_5)_{0.25}(Li_2O)_{0.25}(K_2O)_x(TeO_2)_{1-x}$ $(0.05 \geq x \leq 0.30)$ have been investigated for density and dc conductivity in the temperature range from 380 K to 480 K. The density exhibited a minimum, and molar volume showed a maximum at $x = 0.20$. Conductivity data have been analyzed in the light of Mott's small polaron hopping model and high temperature activation energy values have been determined. The conductivity and activation energy also passed through a minimum and maximum, respectively, at $x = 0.20$. This result has been attributed to the mixed alkali effect. The strength of mixed alkali effect has been estimated. It is for the first time that vanadotellurite glasses are shown to exhibit mixed alkali effect. Various physical and polaron hopping parameters have been determined and discussed.

Key words: *glass; tellurite glass; bulk density; conductivity; mixed alkali effect*

1. Introduction

The electrical conductivity in glasses doped with transition metal ions (TMI) have always been observed to be semiconducting type and that was due to the polaron hopping between multivalent states of TMI [1–3]. Glasses containing Li^+, Ag^+, Cu^+ and Na^+ ions exhibit significant ionic conductivity at ordinary temperatures. An interesting aspect of electrical conduction in ionic glasses is the so called mixed alkali effect (MAE). Mixed alkali doped glasses exhibit lower electrical conductivity compared to that of single alkali composition, for the same total ionic concentrations [4–7]. A general understanding of ion transport in glasses is based on the assumption that cations jump from one position to another similar one while the negatively charged centres remain fixed in position in the glass matrix [7]. The mixed alkali effect has not been completely understood mainly due to the difficulty to determine the conduction pathways for the mobile ions [8]. Ion conducting glasses have technological applications for electrochemical devices such as solid-oxide fuel cells, solid state batteries or

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chemical sensors [9]. MAE has been observed in different physical properties of silicate, borate, germanate and tellurite glasses [10, 11]. MAE has been observed in terms of electrical conductivity and thermal properties in sodium–rubidium–borate glasses [11]. In particular, MAE in terms of glass transition temperature and electrical conductivity has been observed in K_2O-Na_2O-TeO_2 systems [12]. El-Damrawi [13] investigated MAE in Ag and Na doped tellurite and phosphotellurite glasses. Li–Na–Te glasses have been explored for thermal properties and MAE has been reported in terms of specific heat capacity and glass transition temperature [14]. Li–Rb doped phosphate glasses have been investigated for electrical conductivity and pointed out that the strength of MAE depends on the sizes of the alkali ions [15]. Various theoretical models such as Mott's small polaron hopping (SPH), variable range hopping (VRH) [16–19] and Greaves VRH models [20, 21] have been used to explain temperature dependence of conductivity in glasses. There are no reports on physical properties of the glass systems containing both alkali and transition metal ions.

In the present paper, we report the results of investigations of bulk density at room temperature and dc conductivity in function of temperature in the glass systems; $(V_2O_5)_{0.25}(Li_2O)_{0.25}(K_2O)_x(TeO_2)_{1-x}$, $x = 0.05$, 0.10, 0.15, 0.20, 0.25 and 0.30, labelled as MA1, MA2, MA3, MA4, MA5 and MA6. The objective of the present investigation is to study the electrical conductivity in a glass system wherein both TMI and two different alkali ions are present and in terms of the possibility of occurrence of MAE in these systems.

Vanadium oxide (V_2O_5) can also act as glass former. In the present systems, its content is fixed at mole fractions equal to 0.25. In few samples its content is comparable with the main glass former, TiO_2 and therefore V_2O_5 in these systems may play the roles of network former or network modifier or both. No special experiment or analysis has been conducted to ascertain exactly the role of V_2O_5. However, it is often quoted that vanadium exists in two different valence states that is, V^{4+} and V^{5+}. The existence of two different oxidation states paves the way for polaron conduction in the glasses.

2. Experimental

The glass samples were prepared by the melt-quenching technique using the analytical grade V_2O_5, Li_2CO_3, K_2CO_3 and TeO_2 (Sigma-Aldrich). The well ground mixture of chemicals in appropriate weight ratios was taken in porcelain crucible and melted in a muffle furnace at a constant temperature in the range 1200–1300 K for 1 h. The melts were quickly quenched to room temperature and the random pieces of samples were collected. In order to relieve the mechanical stresses, the samples were annealed at 525 K. Glassy nature of the samples was confirmed by X-ray diffraction studies.

It is possible that corrosive melts at high temperatures can attack the porcelain made crucibles. However, there are numerous reports wherein the glasses were prepared at the temperature ranges operated here [22, 23], and no chemical analysis of the samples was carried out and the set compositions were assumed. Due to unavailability of the facilities and expertise no chemical analysis has been performed of the present glasses and the set compositions are taken for granted. Moreover, the integrity of our porcelain crucibles after quenching the melt was observed to be intact.

Densities at room temperature were measured by following the Archimedes principle using a sensitive single pan balance (Sartorius). Carbon tetrachloride, CCl_4 (density = 1.595 g/cm^3) was used as an immersion liquid. The uncertainty on density was estimated to be ±0.026 g/cm^3.

Samples (4±0.04) mm thick of cross sectional areas ranging from (30±0.30) mm^2 to (50±0.50) mm^2 were cut for dc conductivity measurements and the silver electrodes were painted on two major surfaces of the samples. Conductivities in the temperature range 300–525 K were measured by the two probe method. The currents and voltages were measured using a digital nanoammeter and a multimeter to the accuracy of ±0.1nA and ±10mV, respectively. The temperature of the sample was measured to the accuracy of ±1 K using a chromel-alumel thermocouple and a digital microvoltmeter. To verify the reproducibility and minimize errors, the experiment was repeated and data collected over several experiments has been averaged. The conductivity was determined from the equation: $\sigma = t/RA$, where t is the thickness, R is the resistance and A is the cross sectional area of the sample.

The errors on conductivity, $\Delta\sigma$, were estimated taking into account errors on the measured thickness, Δt, areas, ΔA, voltages, ΔV, and currents, ΔI. The estimated errors on conductivity were found to lie in the range 4–5%.

The ionic conductivity is often measured by the ac impedance spectroscopy to avoid the polarization effects on the sample–contact interfaces. However, there are numerous reports in which the ionic conductivity has been measured using the dc method [2, 3, 24–26]. Since ions Li^+ and K^+ incorporated into the glass network are lighter ones and in many instances the ionic conductivity has been detected by the dc method, we have adopted the dc technique to measure ionic conductivity of the present glasses. To avoid initial polarizations, if any, the current measurements were actually started after about 30 s of keeping constant voltage across the sample. The resistance measurements were repeated several times to check the reproducibility of the data.

3. Results

3.1. Density and molar volume

The density of the present glasses was found to be in the range of 2.24 to 3.22 g·cm^{-3} (Table 1).

Table 1. Physical properties of Li_2O and K_2O doped vanadotellurite glasses

Glass compositions in mole fractions				$\rho \pm 0.026$ [$g\cdot cm^{-3}$]	$V_m \pm 0.450$ [$cm^3\cdot mol^{-1}$]	$N \pm 0.04$ [$10^{21} cm^{-3}$]	$R \pm 0.01$ [nm]	$\sigma \times 10^{-6}$ [$ohm^{-1}\cdot m^{-1}$] at 450 K
V_2O_5	Li_2O	K_2O	TeO_2					
0.25	0.25	0.05	0.45	3.220	40.891	5.331	0.572	6.23±0.09
0.25	0.25	0.10	0.40	2.895	45.112	4.793	0.593	4.31±0.06
0.25	0.25	0.15	0.35	2.462	52.612	4.076	0.626	2.45±0.10
0.25	0.25	0.20	0.30	2.236	57.451	3.702	0.646	1.16±0.09
0.25	0.25	0.25	0.25	2.368	53.796	3.920	0.634	2.25±0.12
0.25	0.25	0.30	0.20	2.748	45.968	4.549	0.604	8.68±0.06

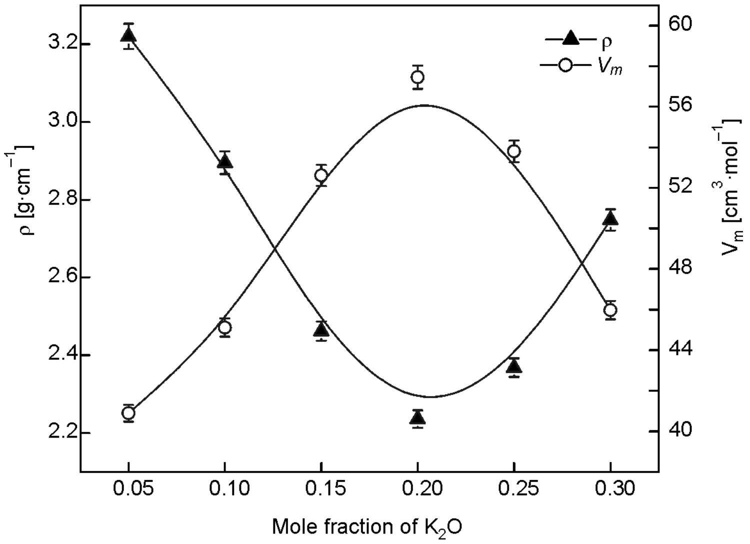

Fig. 1. Compositional dependence of bulk density and molar volume.
The solid lines are guides to the eye

The density decreased with K_2O content up to 0.20 mole fraction and increased for further doping of K_2O (Fig. 1). The molar volume, V_m, was calculated from the density data using the relation $V_m=M/\rho$ [27], where M is the molecular weight of the glass which is given as

$$M = 0.25 M_{V_2O_5} + 0.25 M_{Li_2O} + x M_{K_2O} + (0.5 - x) M_{TeO_2}$$

The values of V_m were found to be in the range 40.89–57.45 $cm^3\cdot mol^{-1}$ and increased continuously with increase in K_2O up to 0.20 mole fraction and decreased for further increase in K_2O (Fig. 1).

The vanadium ion density, N [27] was estimated using the relation

$$N = 2\left(\left(\frac{\rho m_{\mathrm{V_2O_5}}}{M_{\mathrm{V_2O_5}}}\right)N_A\right)$$

where ρ is the density of the glass, $m_{\mathrm{V_2O_5}}$ is the mole fraction of V_2O_5, $M_{\mathrm{V_2O_5}}$ is molecular weight of V_2O_5 and N_A is the Avogadro number. The calculated values of N are presented in Table 1. The error on N values was estimated using the relation

$$\Delta N = \left(\frac{2N_A m_{\mathrm{V_2O_5}}}{M_{\mathrm{V_2O_5}}}\right)\Delta\rho$$

and was found to be $\pm 0.01\times10^{21}$.

3.2. dc conductivity

In the studied temperature range, the measured conductivities were found in the range from 10^{-5} to 10^{-8} $\mathrm{ohm^{-1}\cdot m^{-1}}$ being in agreement with the reported dc conductivities in alkali ions doped similar and other glass systems [2, 9–15]. The temperature dependence of dc conductivity was observed to be of a semiconductor type. The conductivity decreased monotonously with increase in concentration of alkali ions up to the 0.20 mol fraction of K_2O and then it increased for further increase in total alkali concentration. The initial decrease and then increase in conductivity with K_2O content is believed to be due to mixed alkali effect and this is discussed elaborately in the following section.

4. Discussion

4.1. Density and molar volume

The variation of density and molar volume with alkali concentration in the present glasses indicate that the glass network becomes continuously loose packed up to 0.20 mol fraction of K_2O and shows a sign of tight packing of network for further doping of the second alkali [28–30]. The monotonous increase in V_m with the addition of second alkali up to 0.20 mol fraction of K_2O reveals no significant change in the topology of glass network in the studied systems [30]. However, the observed increase in V_m with further addition of K_2O above 0.20 mol fraction may be a sign for change in the structure and the topology of the glass network. The composition dependence of bulk density, in the present glasses, suggests the occurrence of mixed alkali effect.

No structural investigations have been conducted on the present glasses and no structural information on mixed alkali vanadotellurite glasses is available in the literature, either. However, structural aspect of single and mixed alkali tellurite glasses has been reported [12, 31], where it was concluded that in these systems non-bridging oxygen ions (NBOs) exist. The NBOs number increases with the increase of second alkali and their concentration becomes maximum at the composition corresponding to conductivity minimum. This may be the case in the present glass systems also.

4.2. dc conductivity

According to Mott's small polaron hopping model (SPH) [16, 17], the electrical conductivity in non-adiabatic regime is expressed as,

$$\sigma = \frac{\sigma_0}{T}\exp\left(-W/k_B T\right)$$

where W is the activation energy and σ_0 is the pre-exponential factor given as

$$\sigma_0 = \nu_0 N e^2 R^2 C(1-C)\exp(-2\alpha R)/k_B$$

where, $\nu_o=\theta_D k_B/h$ is the optical phonon frequency [32], $\theta_D=2T_D$, is the Debye temperature, N is the concentration of TMI, R is mean spacing between the TMI given as $R=N^{-1/3}$, α is the tunnelling factor and C is the fraction of reduced TMI concentration to that of total TMI concentration. The estimated values of N, R, θ_D and ν_0 for the present glasses are presented in Table 1. The errors on R were calculated using the formula $\Delta R = (\Delta N/3N^{4/3})$.

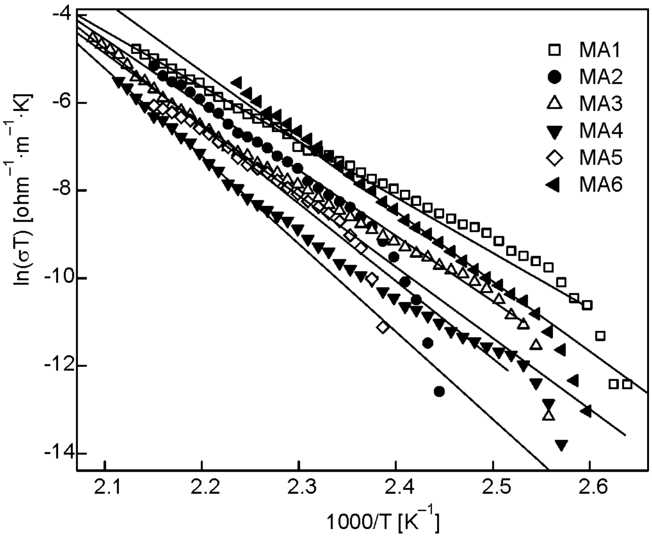

Fig. 2. Temperature dependences of dc conductivity in MA glasses. The solid lines are least square linear fits to the data

According to Eq. (1), the plots of ln(σT) vs. ($1/T$) were made for the present glasses and they are shown in Fig. 2. The dependences were found to be linear at high temperatures and non-linear at low temperatures. The precise values $\theta_D/2$ were not determined due to larger uncertainties in the measured conductivities at lower temperatures. The general behaviour of the curves is observed to be similar to that reported for alkali doped vanadophosphate [2], vanadotellurite and phosphotellurite glasses [10–13]. The least square lines were fit to the data corresponding to high temperatures. The best fit of $\ln\sigma$ vs. $1/T$ gave r^2 from 0.9992 to 0.9996 (r is the correlation coefficient). From the slopes of the lines, the activation energies, W, were determined (Table 2). The W values for the present glasses were found to lie between 1.091 eV and 1.638 eV. These values are in close agreement with those reported in literature [2, 9–13]. The computational errors on activation energies were estimated as per $\Delta W = (\Delta\text{slope})k_B$ and they are found to lie within ±0.002 eV. The activation energies of the present glasses are in near agreement with mixed alkali borate glasses [4, 5], phosphate glasses [2, 33] and tellurite glasses [10–13].

Table 2. Polaron hopping parameters for MA glasses

Glass	$W \pm 0.002$ [eV]	$WH \pm 0.01$ [eV]	$\varepsilon_p \pm 0.002$	$r_p \pm 0.004$ [nm]	J ± 0.001 [eV]	$N(E_f)$ [10^{21} eV$^{-1}\cdot$m^{-3}]	$\gamma_p \pm 0.01$
MA1	1.091	0.651	0.99	0.231	0.122	1.17	21.00
MA2	1.289	0.770	0.81	0.239	0.121	0.89	24.81
MA3	1.522	0.909	0.65	0.252	0.119	0.64	29.29
MA4	1.638	0.978	0.59	0.261	0.118	0.54	31.52
MA5	1.487	0.888	0.66	0.256	0.119	0.63	28.61
MA6	1.377	0.822	0.75	0.243	0.120	0.73	26.50

The variation of activation energy W, and conductivity σ at 450 K in function of mole fraction of second alkali content K_2O are plotted in Fig. 3. From Figure 3 it can be observed that the conductivity decreases with the increase of K_2O content and reaches minimum at 0.2 mole fraction and increases for further increase of K_2O content. On the other hand, the activation energy W increases with the increase of K_2O content and reaches maximum at 0.2 mole fraction and decreases for further increase of K_2O content. The conductivity going through minimum and activation energy passing through maximum at 0.2 mole fraction of the second alkali content is due to mixed alkali effect taking place in MA glasses.

The dynamic structure model (DSM) and matrix mediated coupling (MMC) have been mainly used to discuss the observed results. In addition to DSM and MMC, there exists a number of structural models or views which have been proposed to explain the dynamics of diffusion of alkali ions in glasses [8, 12, 34]. There is no single model which can explain MAE in all types of glasses and our presently studied systems are special and complicated ones as they contain both TMI and mixed alkali ions. In these systems, the second alkali ions were introduced at the cost of TeO_2 and therefore no models have been invoked to analyze the observed MAE in the present systems. In the TMI and alkali doped glasses, the conductivity is due to both polarons and alkali ions. Polaron hopping between TMI sites of different oxidation or valency gives rise to electronic conductivity and ionic movement interstitially leads to ionic conductivity.

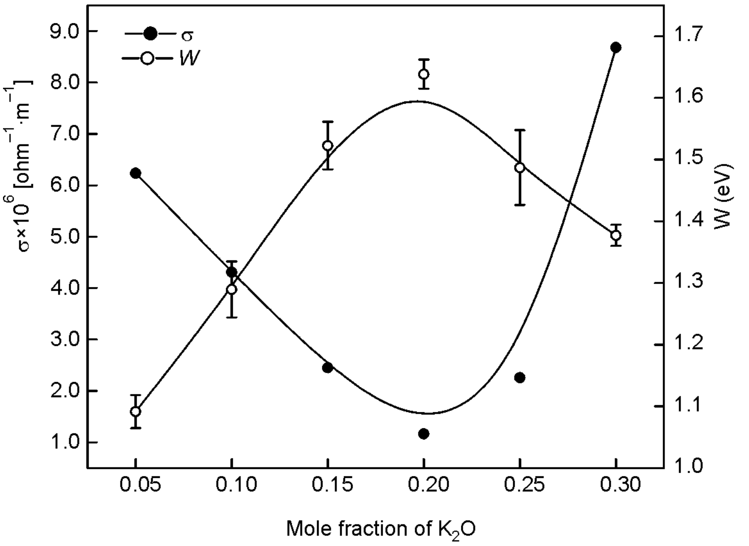

Fig. 3. Compositional dependence of the high temperature activation energy and conductivity at 450 K. The solid lines are guides to eye

Bazian et al. [35, 36] proposed an explanation for the phenomenon of mixed electronic-ionic conduction in oxide glasses. In the glasses where both ions and polarons coexist there must be some interaction between them. The argument goes as follows: mobile electrons or polarons formed by the capture of the moving electrons by TMI of low valency (V^{4+} in the case of vanadium) are attracted by the oppositely charged cations (Li^+ and K^+). This so formed cation–polaron pair tends to move together as a neutral entity. Then the migration of these pairs does not involve any net displacement of charge so this process does not contribute to the electrical conductivity. These authors compared this phenomenon with that of interaction assumed or explanation of the so called mixed alkali effect. So, in the present systems the number of polaron-ion pairs may increase upon increasing K_2O content and that in turn decreases the total conductivity. The conductivity reaches minimum when polaron–ion number reaches a possible maximum. There is another explanation due to Jayasinghe et al. [37], where the continuous blocking of electronic paths with the addition of ionic content is proposed.

The strength of MAE, $\Delta\ln\sigma_{dc}$, has been estimated [6] from

$$\Delta \ln \sigma_{dc} = \ln \sigma_{dc\,\text{lin}} - \ln \sigma_{dc\,\text{min}}$$

where $\ln\sigma_{dc\,\mathrm{lin}}$ represents logarithm of conductivity on the linear interpolated line between two end points in $\ln\sigma$ vs. x plot, which correspond to x for the minimum conductivity (x stands for mole fraction of K_2O). The term $\ln\sigma_{\mathrm{dc\,min}}$ represents the logarithm of the minimum experimental value of conductivity. The estimated value of $\Delta\ln\sigma_{dc}$ for the present glasses is 2.02 and this is in the range of values reported for other binary alkali glass systems [6].

4.3. Parameters associated with polaron hopping

Considering a strong electron-phonon interaction, Austin and Mott [17] proposed that

$$W = W_H + \frac{W_D}{2} \quad \text{for} \quad T > \frac{\theta_D}{2}$$

$$W \approx W_D \quad \text{for} \quad T < \frac{\theta_D}{4}$$

where W_H is the polaron hopping energy and W_D is the disorder energy arising from the energy difference of the neighbours between two hopping sites.

Polaron hopping energy W_H was calculated using the formula [38]:

$$W_H = \frac{W_p}{2} = \frac{e^2}{4\varepsilon_p}\left(r_p^{-1} - R^{-1}\right)$$

Where W_p is the polaron binding energy, ε_p is the effective electric permittivity, which can be determined from the relation, $\varepsilon_p = e^2/4Wr_p$ [16] and the small polaron radius, $r_p = (1/2)(\pi/6N)^{1/3}$ [39]. The estimated values of W_H, ε_p and r_p, are listed in Table 2 and they are comparable with reported values for other similar glasses [1–3, 38]. The errors on W_H and r_p were calculated to be ±0.01 eV and ±0.004 nm, respectively.

In the SPH model, the polaron bandwidth J, which is a measure of electron wavefunction overlap on adjacent sites, is given by [32, 40]

$$J > \left(\frac{2kTW_H}{\pi}\right)^{1/4}\left(\frac{h\nu_0}{\pi}\right)^{1/2} \quad \text{for adiabatic SPH}$$

$$J < \left(\frac{2kTW_H}{\pi}\right)^{1/4}\left(\frac{h\nu_0}{\pi}\right)^{1/2} \quad \text{for non-adiabatic SPH}$$

The polaron bandwidths were calculated from the relation $J = J_0\exp(-\alpha R)$, where $J_0 = W_{H(\mathrm{min})}/4$ [39]. The calculated values of J are listed in Table 2. The value of α was taken from the literature quoted for TMI doped glasses ($\alpha = 20$ nm^{-1}) [32]. From Table 2, it can be noted that J for all the glasses satisfies the condition for non-adiabatic conduction given in Eq. (6) and Holstein's condition, i.e., $J < W_H/3$ [41]. The dependences of J, ε_p and $N(E_F)$ on K_2O content are similar, that is they decrease up to 0.20 mole fraction and increase for further increase of K_2O content.

The densities of states, $N(E_F)$, near the Fermi level were calculated [38] from:

$$J \approx \frac{e^3 N(E_F)^{1/2}}{\varepsilon_p^{3/2}}$$

The estimated values of $N(E_F)$ are shown in Table 2 and they are of the order of 10^{21} $eV^{-1}\cdot m^{-3}$. These $N(E_F)$ values are in agreement with the reported values for many TMI doped glasses [38]. The values of the small polaron coupling constant γ_p, which is a measure of electron-phonon interaction, were calculated using the relation $\gamma_p=2W_H/h\nu_o$ [16,38] and shown in Table 2. The nature of variation of W_H, r_p and γ_p with K_2O content is same as W that is, they increase up to 0.20 mole fraction and decrease for further increase in K_2O content. The γ_p behaviour reveals that the electron-phonon interaction becomes stronger with increase in K_2O concentration, which in turn decreases the electronic contribution to the total electrical conductivity. For higher concentrations of K_2O, γ_p decreases and hence the increase in electrical conductivity was observed.

5. Conclusion

A set of mixed alkali vanadotellurite glasses prepared by melt quench technique was subjected to room temperature density and dc conductivity studies. The density decreased up to 0.20 mole fraction of K_2O and increased for further increase in K_2O content. This is attributed to the structural changes taking place in the glasses at 0.20 mole fraction of K_2O.

The electrical conductivity and activation energy passed through minimum and maximum, respectively, at 0.20 mole fraction of second alkali (K_2O) content. This must be due to mixed alkali effect. The estimated strength of mixed alkali effect agrees with the results reported for other glasses. In view of the absence of any universal theory to explain mixed alkali effect, no theoretical model has been invoked to describe mixed alkali effect in the present glasses. It is concluded that the observed effect is due to interaction of alkali ions among themselves and their interaction with the remaining glass network offered by V_2O_5–TeO_2. It is for the first time that vanadotellurite glasses are shown to be exhibiting mixed alkali effect.

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