# Electrical conductance of a mixture of sodium and potassium nitrates in aqueous medium

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Electrical conductance and density measurements of $[x\text{NaNO}_3 + (1-x)\text{KNO}_3] + R\text{H}_2\text{O}$ system were taken as functions of $x$, $R$, and temperature. The mixed alkali effect on molar conductance ($\Lambda$) was found to be negligible upto $R = 25$ and becomes significant in the region where $R < 25$. It has been shown that for mixed electrolytic system in aqueous medium the concentration range at which the mixed alkali effect on $\Lambda$ starts becoming significant can be predetermined by plotting the difference in $\Lambda$ of the two pure electrolytic solutions versus $R$. The concentration dependence of $\Lambda$ has been described satisfactorily by the expression $\Lambda = \Lambda_{\text{FLK}} \exp(Bc + Cc^2)$ where $\Lambda_{\text{FLK}}$ is the Falkenhagen–Leist–Kelbg equation for $\Lambda$, $B$ and $C$ are empirical constants, and $c$ is the molar concentration. The observed values of the ion-size parameter have indicated more ionic association in $\text{KNO}_3$ solution than in $\text{NaNO}_3$ solution.

*Key words*: electrical conductance, sodium nitrate, potassium nitrate, mixed alkali effect.

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On a mesuré la conductivité électrique et la densité du système $[x\text{NaNO}_3 + (1-x)\text{KNO}_3]R\text{H}_2\text{O}$ en fonction de $x$, de $R$ et de la température. On a trouvé que l'effet des alcalins mixtes sur le conductivité ($\Lambda$) est faible jusqu'à une valeur de $R = 25$ et qu'il devient important lorsque $R < 25$. Pour le système électrolytique mixte en milieu aqueux, on a démontré que l'intervalle de concentration dans lequel l'effet des alcalins mixtes commence à se faire sentir peut être prédéterminé en faisant un graphique de la différence entre les $\Lambda$ des deux solutions électrolytiques pures en fonction de $R$. L'équation $\Lambda = \Lambda_{\text{FLK}} \exp(Bc + Cc^2)$, dans laquelle $\Lambda_{\text{FLK}}$ est l'équation de Falkenhagen–Leist–Kelbg pour $\Lambda$, $B$ et $C$ sont des constantes empiriques et $c$ est la concentration molaire, permet de décrire correctement la relation qui existe entre la concentration et la conductivité $\Lambda$. Les valeurs observées pour le paramètre de la grosseur de l'ion indiquent qu'il y a plus d'association en solution dans le $\text{KNO}_3$ qu'en solution dans le $\text{NaNO}_3$.

*Mots clés* : conductivité électrique, nitrate de soude, nitrate de potasse, effet des alcalins mixtes.

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## Introduction

Properties of ternary systems containing two alkali metal ions in a fixed glassy medium are interesting due to a phenomenon known as mixed alkali effect (MAE) which is of both practical and theoretical importances (1, 2). This phenomenon refers to non-additivity of various physical properties caused when the relative amounts of the alkali metal ions are varied while the concentration of the glassy medium is a constant. Normally MAE is most pronounced for properties related to ionic transport.

Attempt are being made to look for the MAE in media other than glass. In hydrate melt medium MAE also is found to be present but to a lesser extent (3–6). Another suitable medium for investigating MAE is the aqueous medium. Investigation of MAE on transport properties in aqueous medium has a two-fold interest. (*i*) Data on the properties of mixed aqueous electrolytes are necessary because many systems of practical, biological, geological or chemical interest involve mixed aqueous electrolytes. Studies on the transport properties of mixed aqueous electrolytes have, however, been relatively few compared to those on their thermodynamic properties. (*ii*) One of the characteristics of MAE is that it shows a complicated dependence on the total alkali metal ion concentration. To examine this aspect, aqueous medium is more convenient. Any mixed alkali metal ion system in aqueous medium can be represented as $[xM_1A + (1-x)M_2A] + R\text{H}_2\text{O}$, where $M_1$ and $M_2$ are the two alkali metal ions and $R$ is the number of moles of $\text{H}_2\text{O}$. Our previous attempt (7) to look for MAE in aqueous medium by measuring electrical conductances of a mixture of sodium and potassium thiocynates in water at 25°C has shown that significant MAE on molar conductance exists only in the region where $R < R_0 \simeq 10$. In view of the fact that NaSCN solution exhibits specific conductance maximum at $R \simeq 10$ and for KSCN solution it occurs at $R \simeq 6$, the value of $R_0$ is considered to have a correlation with the concentration (lower one) at which the specific conductance maximum of one of the constituent electrolytes of the mixed system occurs (7). Furthermore, it has been suggested that in the region where MAE exists the mixed electrolytic system has a quasi-crystalline-type structure. To further examine this type of dependence of MAE on R or on the total alkali metal ion concentration, electrical conductances of another mixed electrolyte, viz., $[x\text{NaNO}_3 + (1-x)\text{KNO}_3] + R\text{H}_2\text{O}$, are reported here as functions of $x$, $R$, and temperature.

## Experimental section

$\text{NaNO}_3$ (BDH, Laboratory Reagent) and $\text{KNO}_3$ (BDH, Laboratory Reagent) were recrystallized from their solutions in doubly distilled water and were then dried for several days over anhydrous $\text{CaCl}_2$ in a vacuum desiccator. Using these salts molal solutions were prepared. To convert molal concentrations into molar concentrations densities of the solutions were measured using a calibrated glass pycnometer of about 7 mL capacity. Conductivity measurements were made at 1 kHz using the CDM 83 conductivity meter (Radiometer, Copenhagen) and a dip type CDC 304 conductivity cell. This cell has three electrodes in the form of pure platinum bands on a glass tube. All measurements were made in a thermostated (±0.02°C) water bath.

## Results and discussion

The experimental values of molar conductance of $[x\text{NaNO}_3 + (1-x)\text{KNO}_3] + R\text{H}_2\text{O}$ system measured as functions of $x$, $R$, and temperature are presented in Table 1.[3] The density data are

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[3] Complete set of tables may be purchased from the Depository of Unpublished Data, Document Delivery CISTI, National Research Council of Canada, Ottawa, Canada K1A 0S2.

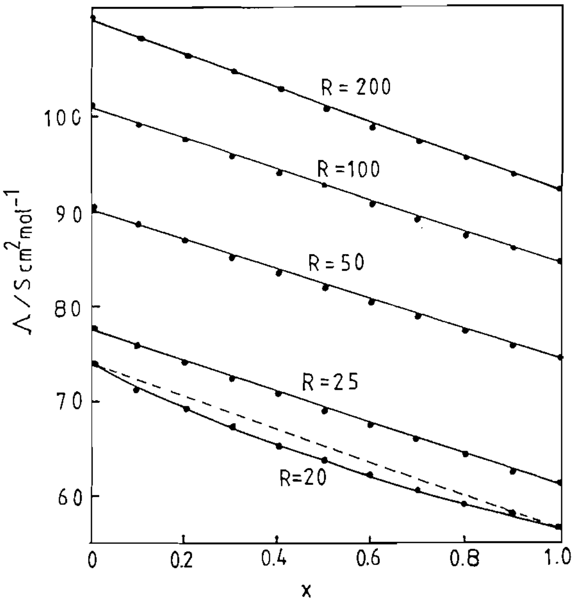

FIG. 1. Variation of Λ of [$x$NaNO$_3$ + (1 − $x$)KNO$_3$] + $R$H$_2$O system with $x$ at 298 K for different $R$ values.

presented as a function of temperature in Table 2.[3] The measured values of conductance were found to be reproducible to within ±0.75% accuracy.

*Variation of* Λ *with x*

The variation of Λ with $x$ at different $R$ values is shown in Fig. 1. At all experimental tempertures less than 1% deviation of Λ from additivity has been observed upto $R = 25$. Rysselberghe and Nutting (8) also reported less than 1% deviation from additivity in Λ of [$x$NaNO$_3$ + (1 − $x$)KNO$_3$] + 55.56H$_2$O system at 298 K. The MAE is, however, considered to be not very significant when the non-additivity in Λ is <1%. At $R = 20$, on the other hand, 2% deviation of Λ from additivity (Fig. 1) has been observed for $x = 0.5$ at 298 K thereby envisaging the occurrence of the MAE on Λ. Earlier it was suggested (7), as mentioned in the Introduction, that the MAE on electrical conductance starts becoming significant when the value of $R$ falls in the concentration range where conductivity maximum of at least one of the components of the mixed electrolytic solution appears. The existence of MAE at $R = 20$ in the [$x$NaNO$_3$ + (1 − $x$)KNO$_3$] + $R$H$_2$O system is, however, contrary to our above suggestion since conductivity maximum has not been observed or reported for KNO$_3$ solution upto saturation point and for NaNO$_3$ solution it is found at $R \simeq 6.9$. What is then the concentration range at which the MAE on Λ becomes significant in an aqueous medium? To seek a probable answer to this question we have plotted in Fig. 2 the difference (ΔΛ) in the Λ values of (*i*) NaSCN and KSCN solutions (7) and (*ii*) NaNO$_3$ and KNO$_3$ solutions as a function of $R$ at 298 K. It is evident from Fig. 2 that at high $R$ values ΔΛ depends linearly on $R$ and at some concentration range deviation from the linear dependence starts to take place. It is very interesting to observe that the concentration range at which the MAE on Λ starts becoming significant coincides with the concentration range where dependence of ΔΛ on $R$ deviates from linearity. Incidentally, in the case of NaSCN and KSCN solutions the concentration range around which ΔΛ deviates from linearity coincides with the range at which NaSCN solution exhibits specific conductance maximum. In Fig. 2 we also checked the dependence of ΔΛ on $R$ for a few other pair of electrolytes, viz., RbCl and CsCl (at 296 K), NaCl and KCl (at 298 K), and LiCl and RbCl (at 296 K), with the help of their reported data (9–11). In all cases, the dependence of ΔΛ on $R$ follows a general trend, viz., (*i*) linear dependence at large $R$ values, (*ii*) at some concentration range, which depends on the pair of electrolytes considered, deviation from linearity is observed and (*iii*) the plot of ΔΛ vs. $R$ passes through a maximum at some lower $R$ value. Therefore, for mixed electrolyte systems in aqueous medium the concentration range at which the MAE on Λ begins to become significant can be predetermined by plotting the difference in Λ of the two pure electrolytic solutions versus $R$.

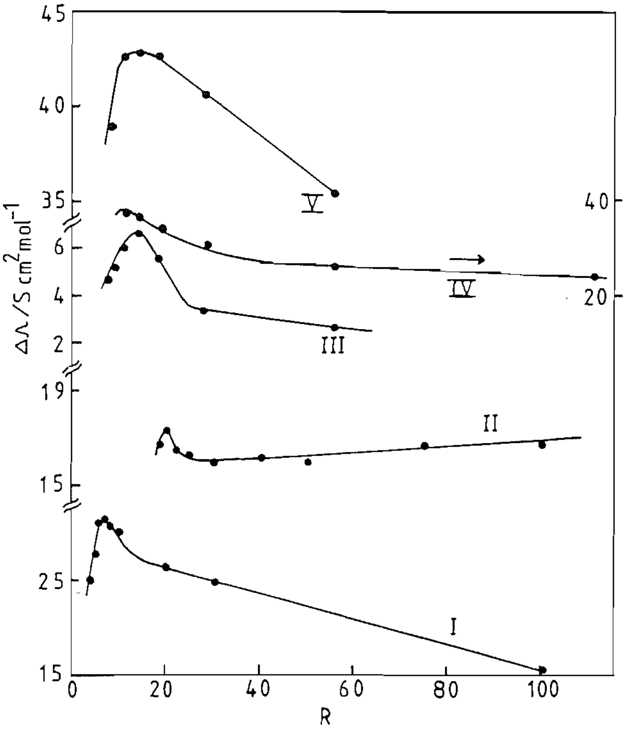

FIG. 2. Plot of difference in Λ values of aqueous electrolytic solutions at 25°C vs. $R$ (*I*, NaSCN and KSCN; *II*, NaNO$_3$ and KNO$_3$; *III*, RbCl and CsCl (at 23°C); *IV*, NaCl and KCl; and *V*, LiCl and RbCl (at 23°C).

*Variation of* Λ *with* R

We recently reported (7) a new isothermal equation of the form

$$[1] \quad \Lambda = \Lambda_{\mathrm{FLK}} \exp\,(Bc + Cc^2)$$

to describe the concentration dependence of Λ of electrolytic solutions, both single and mixed electrolytes, from dilute region upto high concentration. In eq. [1], $B$ and $C$ are empirical constants and $c$ is the molar concentration. Concentration in the exponential part of eq. [1] can also be expressed in other units. $\Lambda_{\mathrm{FLK}}$ refers to the Falkenhagen–Leist–Kelbg (FLK) equation (12) for Λ and is of the form

$$[2] \quad \Lambda_{\mathrm{FLK}} = \left[\Lambda_0 - \frac{B_1c^{1/2}}{1 + B_0a_0}\right]\left[1 - \frac{B_2c^{1/2}F}{1 + B_0a_0}\right]$$

For 1:1 electrolytes, $B_1 = 82.501/\eta_0(DT)^{1/2}$, $B_2 = 82.04 \times 10^4/(DT)^{3/2}$, $B_0 = 50.29 \times 10^8c^{1/2}/(DT)^{1/2}$, and $F = [\exp(0.2929B_0a_0) - 1]/0.2929B_0a_0$. $\Lambda_0$ is the molar conductance of the solution at infinite dilution, $\eta_0$ is the viscosity of water, $D$ is the dielectric constant of water at temperature $T$, and $a_0$ is known as the ion-size parameter. In eq. [1] the parameters whose values need to be determined are $\Lambda_0$, $a_0$, $B$, and $C$. While fitting data to eq. [1] it was reduced to a three-parameter equa-

TABLE 3. Best-fit parameters of eq. [1] for molar conductance of $[x\text{NaNO}_3 + (1-x)\text{KNO}_3] + R\text{H}_2\text{O}$ system

| $x$ | $\Lambda_0/S\ \text{cm}^2\ \text{mol}^{-1}$ | $a_0 \times 10^8/\text{cm}$ | $-B$ | $-C$ | Std. dev. in $\Lambda$ |
|---|---|---|---|---|---|
| 283 K | | | | | |
| 0.0 | 104.72 | 1.45 | −0.0720 | 0.2423 | 0.0764 |
| 0.1 | 102.91 | 1.46 | −0.0740 | 0.2544 | 0.0634 |
| 0.2 | 101.10 | 2.02 | −0.0116 | 0.1649 | 0.0861 |
| 0.3 | 99.29 | 2.43 | 0.0251 | 0.1094 | 0.0486 |
| 0.4 | 97.48 | 2.41 | 0.0134 | 0.1680 | 0.0804 |
| 0.5 | 95.67 | 2.92 | 0.0595 | 0.0642 | 0.1561 |
| 0.6 | 93.86 | 3.28 | 0.0798 | 0.0385 | 0.1180 |
| 0.7 | 92.05 | 3.56 | 0.0915 | 0.0312 | 0.1136 |
| 0.8 | 90.24 | 3.81 | 0.0999 | 0.0288 | 0.1135 |
| 0.9 | 88.43 | 4.11 | 0.1100 | 0.0237 | 0.1227 |
| 1.0 | 86.62 | 4.34 | 0.1154 | 0.0241 | 0.9044 |
| 288 K | | | | | |
| 0.0 | 117.68 | 1.88 | −0.0206 | 0.1594 | 0.0965 |
| 0.1 | 115.69 | 1.78 | −0.0384 | 0.2152 | 0.0594 |
| 0.2 | 113.70 | 2.34 | 0.0191 | 0.1113 | 0.1046 |
| 0.3 | 111.72 | 2.57 | 0.0375 | 0.0875 | 0.0630 |
| 0.4 | 109.73 | 2.66 | 0.0366 | 0.1223 | 0.0655 |
| 0.5 | 107.74 | 2.96 | 0.0624 | 0.0609 | 0.1313 |
| 0.6 | 105.75 | 3.34 | 0.0842 | 0.0329 | 0.1285 |
| 0.7 | 103.76 | 3.61 | 0.0956 | 0.0259 | 0.1219 |
| 0.8 | 101.78 | 3.86 | 0.1038 | 0.0231 | 0.1059 |
| 0.9 | 99.79 | 4.14 | 0.1130 | 0.0181 | 0.1041 |
| 1.0 | 97.80 | 4.36 | 0.1182 | 0.0184 | 0.1073 |
| 293 K | | | | | |
| 0.0 | 131.16 | 1.96 | −0.0156 | 0.1666 | 0.0961 |
| 0.1 | 128.99 | 2.09 | −0.0031 | 0.1487 | 0.0861 |
| 0.2 | 126.83 | 2.40 | 0.0233 | 0.1084 | 0.1074 |
| 0.3 | 124.66 | 2.63 | 0.0414 | 0.0843 | 0.0587 |
| 0.4 | 122.49 | 2.72 | 0.0405 | 0.1187 | 0.1507 |
| 0.5 | 120.33 | 3.06 | 0.0692 | 0.0537 | 0.1516 |
| 0.6 | 118.16 | 3.36 | 0.0858 | 0.0316 | 0.1177 |
| 0.7 | 115.99 | 3.64 | 0.0974 | 0.0243 | 0.1112 |
| 0.8 | 113.82 | 3.86 | 0.1049 | 0.0211 | 0.1134 |
| 0.9 | 111.66 | 4.12 | 0.1135 | 0.0165 | 0.1189 |
| 1.0 | 109.49 | 4.37 | 0.1204 | 0.0137 | 0.1206 |
| 298 K | | | | | |
| 0.0 | 114.60 | 1.99 | −0.0204 | 0.1690 | 0.3874 |
| 0.1 | 142.29 | 2.55 | 0.0369 | 0.0790 | 0.0595 |
| 0.2 | 139.98 | 2.65 | 0.0392 | 0.0931 | 0.1416 |
| 0.3 | 137.67 | 2.88 | 0.0562 | 0.0690 | 0.0896 |
| 0.4 | 135.36 | 3.24 | 0.0820 | 0.0312 | 0.2118 |
| 0.5 | 133.05 | 3.27 | 0.0817 | 0.0332 | 0.1689 |
| 0.6 | 130.74 | 3.46 | 0.0899 | 0.0277 | 0.1197 |
| 0.7 | 128.43 | 3.67 | 0.0976 | 0.0257 | 0.1310 |
| 0.8 | 126.12 | 3.94 | 0.1081 | 0.0176 | 0.0781 |
| 0.9 | 123.81 | 4.15 | 0.1157 | 0.0146 | 0.1060 |
| 1.0 | 121.50 | 4.36 | 0.1209 | 0.0123 | 0.0342 |
| 303 K | | | | | |
| 0.0 | 159.46 | 2.04 | −0.0095 | 0.0170 | 0.0685 |
| 0.1 | 156.94 | 2.36 | 0.0241 | 0.1021 | 0.0976 |
| 0.2 | 154.42 | 2.41 | 0.0216 | 0.1256 | 0.1605 |
| 0.3 | 151.90 | 2.70 | 0.0475 | 0.0805 | 0.0748 |
| 0.4 | 149.38 | 3.10 | 0.0766 | 0.0407 | 0.3011 |
| 0.5 | 146.86 | 3.13 | 0.0765 | 0.0408 | 0.2083 |
| 0.6 | 144.33 | 3.33 | 0.0861 | 0.0326 | 0.1650 |
| 0.7 | 141.81 | 3.58 | 0.0965 | 0.0262 | 0.1539 |
| 0.8 | 139.29 | 3.83 | 0.1070 | 0.0174 | 0.1008 |
| 0.9 | 136.77 | 4.04 | 0.1138 | 0.0141 | 0.1426 |
| 1.0 | 134.25 | 4.23 | 0.1187 | 0.0129 | 0.1556 |

TABLE 3 (*concluded*)

| $x$ | $\Lambda_0/S\ \text{cm}^2\ \text{mol}^{-1}$ | $a_0 \times 10^8/\text{cm}$ | $-B$ | $-C$ | Std. dev. in $\Lambda$ |
|---|---|---|---|---|---|
| 308 K | | | | | |
| 0.0 | 174.18 | 2.01 | −0.0162 | 0.1968 | 0.1370 |
| 0.1 | 171.49 | 2.39 | 0.0261 | 0.1049 | 0.1190 |
| 0.2 | 168.79 | 2.44 | 0.0234 | 0.1288 | 0.1727 |
| 0.3 | 166.10 | 2.73 | 0.0481 | 0.0890 | 0.0992 |
| 0.4 | 163.40 | 3.13 | 0.0795 | 0.0386 | 0.3183 |
| 0.5 | 160.71 | 3.18 | 0.0809 | 0.0351 | 0.2392 |
| 0.6 | 158.01 | 3.35 | 0.0885 | 0.0301 | 0.2045 |
| 0.7 | 155.32 | 3.61 | 0.1003 | 0.0210 | 0.1585 |
| 0.8 | 152.62 | 3.85 | 0.1098 | 0.0134 | 0.1097 |
| 0.9 | 149.93 | 4.04 | 0.1154 | 0.0114 | 0.1671 |
| 1.0 | 147.23 | 4.20 | 0.1192 | 0.0107 | 0.1952 |
| 313 K | | | | | |
| 0.0 | 189.21 | 2.11 | −0.0071 | 0.1879 | 0.1636 |
| 0.1 | 186.35 | 2.45 | 0.0312 | 0.1008 | 0.1613 |
| 0.2 | 183.48 | 2.48 | 0.0261 | 0.1313 | 0.1698 |
| 0.3 | 180.62 | 2.74 | 0.0485 | 0.0963 | 0.1081 |
| 0.4 | 177.75 | 3.13 | 0.0801 | 0.0413 | 0.4065 |
| 0.5 | 174.89 | 3.23 | 0.0858 | 0.0291 | 0.3041 |
| 0.6 | 172.03 | 3.37 | 0.0914 | 0.0267 | 0.2326 |
| 0.7 | 169.16 | 3.62 | 0.1023 | 0.0180 | 0.1876 |
| 0.8 | 166.30 | 3.84 | 0.1119 | 0.0100 | 0.1193 |
| 0.9 | 163.43 | 4.02 | 0.1165 | 0.0089 | 0.2060 |
| 1.0 | 160.57 | 4.16 | 0.1198 | 0.0085 | 0.2158 |

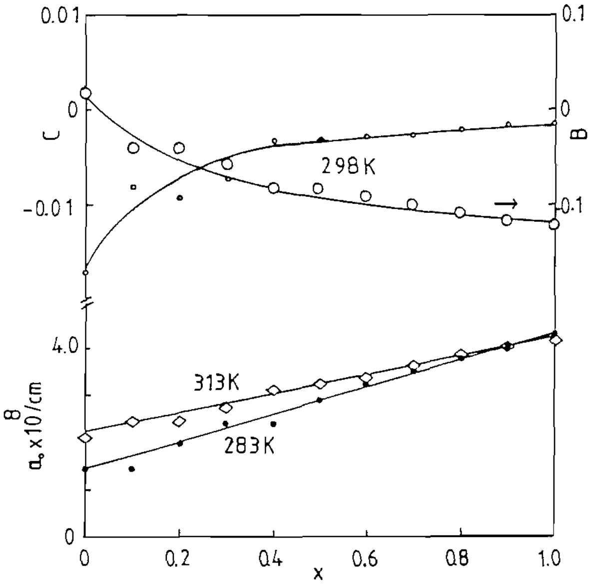

FIG. 3. Variation of $a_0$, $B$, and $C$ of $[x\text{NaNO}_3 + (1-x)\text{KNO}_3] + R\text{H}_2\text{O}$ system with $x$.

tion, by substituting the reported (13) values of $\Lambda_0$ for $x = 0$ and $x = 1$ and for other values of $x$ the corresponding $\Lambda_0$ values were estimated using the additivity principle (14). The best-fit values of $a_0$, $B$, and $C$ obtained by using an iterative least-squares fitting program are listed in Table 3.

It is evident from Table 3 and Fig. 3 that $a_0$ increases linearly with $x$. At 298 K $a_0$ = 1.99 Å for $\text{KNO}_3$ solution ($x = 0$) and for $\text{NaNO}_3$ solution ($x = 1$) $a_0$ = 4.36 Å. Association into ionic pairs is reported (15–17) in aqueous solutions of both $\text{NaNO}_3$ and $\text{KNO}_3$. The considerably low value of $a_0$ for $\text{KNO}_3$ solution, also observed by others (15), envisages more ionic association in $KNO_3$ solution than in $NaNO_3$ solution. Increase in the degree of association of alkali salts in aqueous medium with increasing size of the alkali metal ion was also reported by Kay (15). From Fig. 3 it can further be seen that the $a_0$ parameter for $NaNO_3$ solution is almost temperature independent whereas the $a_0$ of $KNO_3$ solution increases with increase in temperature. This is analogous to the observation made by Goldsack *et al.* (10) in the cases of NaCl and KCl solutions. The degree of ionic association in $KNO_3$ solution therefore seems to decrease with increasing temperature.

The dependence of $B$ and $C$ parameters on $x$ is also shown in Fig. 3 at one particular temperature. $C$, which is always a negative quantity, has the lowest value for pure $KNO_3$ solution and its value increases rapidly as $K^+$ ions are replaced by $Na^+$ ions. In the range from $x = 0.5$ to $x = 1.0$, $C$ appears to remain almost independent of the $x$ value. In the case of $B$, it has a highest positive value for $KNO_3$ solution and its value decreases with increasing $x$ value, becoming negative above a particular value of $x$. The value of $x$ at which B becomes zero is dependent on the temperature. For example, at 298 K $B = 0$ at $x = 0.04$ whereas at 288 K $B = 0$ at $x = 0.16$. Similar behaviour was observed in the case of $[x\text{NaSCN} + (1 - x)\text{KSCN}] + R\text{H}_2\text{O}$ system also (7). This type of dependence of $B$ on $x$ may be explained by correlating, in the light of Wishaw-Stokes equation (18), the exponential part of eq. [1] to the reciprocal of viscosity of the system. Consequently, solutions whose $B$ is positive and $C$ is negative may be expected to show negative viscosity (relative viscosity $< 1$). In fact, aqueous $KNO_3$ solution is known (9) to exhibit negative viscosity nearly upto 318 K. At 298 K the viscosity of $KNO_3$ solution is therefore expected to decrease with concentration upto $c = B/C = 1.2$ mol $dm^{-3}$, which is in good agreement with the experimentally observed value (9). Furthermore, the $B$ parameter appears to have a relation with the $\Delta E$ parameter of the Samoilov model (19) for hydration of ions.

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