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Research Paper

Viscometric and Thermodynamics Study of uni-bivalent Mixed Electrolytes in Aqueous Solutions at 293.15, 303.15 and 313.15K

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Abstract: *Viscosities (η) and apparent molar volumes (V_ϕ) of the solutions of uni-bivalent mixed electrolytes in aqueous solutions have been determined at 293.15, 303.15and 313.15K. It is seen that the plots of ή$_{rel}$-1/√y verses √y are linear, from which the values of coefficients A and B have been obtained by the method of least squares. The values of coefficient A are positive for the mixed electrolytes (NH$_4$)$_2$C$_2$O$_4$ + K$_2$C$_2$O$_4$, Na$_2$SO$_4$ + K$_2$SO$_4$ and (NH$_4$)$_2$SO$_4$ + K$_2$SO$_4$. The negative values of B in the case of Na$_2$SO$_4$ + K$_2$SO$_4$ and (NH$_4$)$_2$ SO$_4$ + K$_2$SO$_4$ show the presence of strong ion-ion interactions in the aqueous solution of these mixed electrolytes. In each case the values of A tend to decrease with the rise of temperatures which suggests further weakening of ion-ion interactions at elevated temperatures. The values of B are positive which tend to increase more positive with the rise of temperature there by suggesting the presence of strong ion-solvent interactions, which becomes stronger at elevated temperatures.* The variation of ϕ_v with. y, fraction of ionic strength due to the first electrolyte in the mixture of two electrolytes at different temperatures is linear. *From   ή and V_Φ data the values of coefficients A and B of Jones -Dole equation and that of $V_\square^\square$ and S_v of Masson's equation have been obtained. Besides the activation thermodynamic quantities Δμ$_2$$^{0\#}$, ΔH$_2$$^{0\#}$and ΔS$_2$$^{0\#}$ of viscous flow have also been calculated for the mixed electrolytes in aqueous solutions. It is seen that the values of Δμ$_2$$^{0\#}$ are much larger as compared to those of Δμ$_1$$^{0\#}$ for all the uni-bivalent mixed electrolytes. The values of TΔS$_2$$^{0\#}$ are negative while those of ΔH$_2$$^{0\#}$ are positive and that ΔH$_2$$^{0\#}$ > TΔS$_2$$^{0\#}$, thereby suggesting that ion-solvent interaction for the mixed electrolyte system, are nearly complete in the ground state. Further these are the values of TΔS$_2$$^{0\#}$ and ΔH$_2$$^{0\#}$ indicate the transition state is associated with the bond breaking and increase in order.*

Keywords: Viscosities, Apparent molar volumes, mixed electrolyte, aqueous media.

Introduction

The studies of thermodynamic properties of aqueous mixed electrolytic solutions using the Mayer-MeMillian[1,2] theory as developed by Friedman and Anderson have been found to be useful in understanding the specific ion-ion interactions in solution Patil and coworkers[3,4] have determined viscosities of aqueous mixed electrolytic solutions for the systems. KBr- NaBr. KBr-Bu$_4$NBr. NaCI-NaBr and NaCl-Bu$_4$NBr at various constant ionic strength with varying electrolyte mole fractions (y) at 25^{0}C.

The viscosity of electrolyte solutions is usually studied to obtain information on ion-solvent interactions[5-8]. In general the variation of relative viscosity, n_{rel} with the molarity of the solution. C can be represented by Jones- Dole equation[9].

$$\eta_{rel} = \frac{\eta}{\eta_0} = 1 + AC^{1/2} + BC \qquad \ldots\ldots\ldots\ldots (1)$$

where η and η_0 are the viscosity coefficients of the solution and solvent respectively. *A* is a constant depending on the long-range coulombic forces (ion-ion interactions) and *B* is an adjustable parameter which is related to the size of the ions and to the different ion-solvent interactions. The apparent molar volumes at infinite dilution and their variation with concentration of electrolytes also throw light on ion-solvent and ion-ion interactions[10,11].

From the above survey of literature it appears that studies on the determination of densities and viscosities of aqueous solutions of uni-bivalent mixed electrolytes vis-à-vis ion-ion and ion-solvent interactions are still lacking. With this aim in view the total study has been undertaken. The following uni-bivalent mixed electrolyte systems in aqueous media keeping ionic strength constant, Interactions are still lacking .With this aim in view the title study have been undertaken. The following uni- bivalent mixed electrolytes have been used :

i. $Li_2SO_4 + (NH_4)_2SO_4$,
ii. $Na_2SO_4 + K_2SO_4$,
iii. $Na_2SO_4 + Li_2SO_4$,
iv. $Na_2SO_4 + (NH_4)_2 C_2O_4$,
v. $Na_2SO_4 + (NH_4)_2 C_2O_4$,
vi. $K_2SO_4 + Li_2SO_4$,
vii. $K_2SO_4 + (NH_4)_2C_2O_4$,
viii. $(NH_4)_2SO_4 + K_2SO_4$, and
ix. $NH_4)_2C_2O_4 + K_2C_2O_4$

The studies have been undertaken in the light of the following aspects:-

I.   Determination of densities and viscosities of aqueous solutions of above mentioned uni-bivalent mixed electrolytes at different temperatures (293.15, 303.15 and 313.15 + 0.0 1K) as a function of fraction of the ionic strength due to the first electrolyte in the mixture of two electrolytes (here in after denoted by *y)* keeping the ionic strength constant at a particular value depending upon the solubility of the electrolytes,

II.   Analysis viscosity data in the light of Jones-Dole equation ,

III.   (iii) Determination of apparent molar volume (ϕ_v) from the density data as a function of *y* at different temperatures and calculation of the values of constants Φ_v and S_v of Masson's equation,

IV.   Determination of free energies of activation for viscous flow $\Delta\mu_1^{0\#}$ and $\Delta\mu_2^{o\#}$ per mole of solvent and solute respectively, and also the calculation   of entropy of activation $\Delta S_2^{0\#}$ and enthalpy of activation $\Delta H_2^{0\#}$ for the viscous flow process of the uni-bivalent mixed electrolytes solutions at different temperatures with a view to interpret the solution behavior of mixed electrolytes , and

V.   Ascertaining structure-making or structure-breaking capacities of mixed electrolyte systems in aqueous solution.

Material and Methods
All the uni-bivalent electrolytes were of analytical reagent grade and were used after drying in a vacuum oven at 110^0 C for 10 to 12 hours. The standard stock

solutions of these electrolytes were prepared in doubly distilled water (specific conductivity- 10^{-6} ohm^4 cm ^{4}) The solutions of mixed electrolytes of different composition with *y* varying in the range of 0.0 of 1.0, were prepared by mixing requisite volume of the solutions of individual electrolytes, in a measuring flask keeping. The ionic strength constant.

The densities and viscosities of aqueous solutions of mixed electrolytes were determined at different temperatures (293.15, 303.15 and 313.15 + 0.01). The viscosity data were analyzed in the light of the modified form of Jones-Dole equation[9] as given blow:
Jones-Dole equation is

$$\eta_{rel} = \frac{\eta}{\eta_0} = 1 + A\sqrt{C} + BC \qquad \dots\dots\dots\dots(2)$$

Or

$$\frac{n_{rel} - 1}{\sqrt{C}} = A + B\sqrt{C} \qquad \dots\dots\dots\dots (3)$$

$$\frac{(n_{rel} - 1)}{\sqrt{y}} = A + B\sqrt{y} \qquad \dots\dots\dots\dots (4)$$

Thus from the linear plots of $\acute{\eta}_{rel}$-1/√y verses √y   the value of *A* and *B* can be obtained.
The apparent molar volume (ϕ_v) of aqueous solutions of uni- bivalent mixed electrolyte systems was determined as a function of y i.e. fraction of ionic strength due to the first electrolyte in the mixtures of two electrolytes, at different temperatures using the following equation

$$\phi_v = \frac{1ooo(\rho_0 - \rho)}{C\rho_0} + \frac{\overline{M}}{\rho_0} \qquad \dots\dots\dots\dots\dots \quad (5)$$

Where ρ and ρ are the densities of solvent and solution respectively and C is the molarity of the solution, $\overline{M}$ is the effective molecular weight of the mixed electrolyte system given by [12]

$$\overline{M} = \frac{n_1 M_1 + n_2 M_2}{n_1 + n_2} \qquad \dots\dots\dots\dots\dots \quad (6)$$

Where n_1 and n_2 are the number of moles M_1 and M_2 are the molecular weights of first and second electrolytes respectively.

The variation of apparent molar volume (ϕ_v) with the molar concentration of the electrolyte solution is governed by Masson's equation[13]

$$\phi_v = \phi_v^0 + S_v \sqrt{C}$$

In the present study, the modified form of this equation in terms of *y* is to be determined on the basis of the linearity or otherwise of Φ_v versus √y plots Φ_v versus y plots.

Free energies of activities of viscous flow $\Delta\mu_1^{0\#}$ and $\Delta\mu_2^{0\#}$ per mole of solute and solvent respectively were determined at different temperatures. The values of entropy and enthalpy of activation of viscous flow were calculated from the following equations;

$$d\frac{(\Delta\mu_2^{0\#})}{dT} = -\Delta S_2^{0\#} \qquad \ldots\ldots\ldots\ldots\ldots(8)$$

$$\Delta H_2^{0\#} = \Delta\mu_2^{0\#} + T\Delta S_2^{0\#} \qquad \ldots\ldots\ldots\ldots\ldots(9)$$

The experimental data in regard to different aspects of the present study have been presented in the tables.

The densities and viscosities of aqueous solutions of uni-bivalent mixed electrolytes have been determined at different temperatures viz. 239.15, 303.15 ± 0.01 K, as a function of y, the fraction of ionic strength due to first electrolyte in the mixture of two electrolytes

keeping the ionic strength of the solution constant at an appraise value.

The viscosity data of the solutions have been analyzed in the light of modified form of Jones- Dole equation as under:

$$\frac{(n_{rel}-1)}{\sqrt{y}} = A + B\sqrt{y} \qquad \ldots\ldots\ldots\ldots\ldots(10)$$

where y is the fraction of ionic strength due to the first electrolyte in the mixture of two electrolytes. It is seen that the plots of $\acute{\eta}_{rel}-1/\sqrt{y}$ verses $\sqrt{y}$ are linear, from which the values of coefficients A and B have been obtained by the method of least squares. The results have been presented in Table 1.

Table 1: Values of coefficients A and B of Jones-Dole equation for uni-bivalent mixed electrolyte systems in aqueous medium at different temperatures

Mixed electrolyte system	Ionic Strength (I)	$A(dm^{3/2}mol^{-1/2})$			$B(dm^3 mol^{-1})$		
		293.15K	303.15K	313.15K	293.15K	303.15K	313.15K
$Li_2SO_4 + (NH_4)_2SO_4$	3.0	-0.0227	-0.0642	-0.1536	0.4070	0.4250	0.4384
$Na_2SO_4 + K_2SO_4$	1.5	0.8074	07406	-0.4901	-0.6145	-0.5735	-0.4503
$Na_2SO_4 + Li_2SO_4$	3.0	-0.0124	-0.1130	-0.2891	0.9448	0.9609	1.0091
$Na_2SO_4 + (NH_4)_2SO_4$	3.0	-0.0249	-0.0369	-0.1209	0.4127	0.8016	0.9948
$Na_2SO_4 + (NH_4)_2C_2O_4$	0.6	-0.4386	-0.7995	-0.09138	0.4480	0.8001	0.9733
$K_2SO_4 + Li_2SO_4$	1.5	-0.0437	-0.0592	-0.0948	0.3497	0.3882	0.3892
$K_2SO_4 + (NH_4)_2C_2O_4$	0.6	-0.6357	-0.9582	-1.0449	0.7973	1.0773	1.1966
$(NH_4)_2SO_4 + K_2SO_4$	1.5	-06399	-0.5336	-0.3072	-0.5666	-0.3429	-0.2081
$(NH_4)_2C_2O_4 + K_2C_2O_4$	1.5	-0.0783	-0.0498	-0.0325	0.2337	0.2564	0.4066

The values of coefficient *A* are positive for the mixed electrolytes (NH$_4$)$_2$C$_2$O$_4$ + K$_2$C$_2$O$_4$, Na$_2$SO$_4$ + K$_2$SO$_4$ and (NH$_4$)$_2$SO$_4$ + K$_2$SO$_4$. The negative values of *B* in the case of Na$_2$SO$_4$ + K$_2$SO$_4$ and (NH$_4$)$_2$ SO$_4$ + K$_2$SO$_4$. and which show the presence of strong ion-ion interactions in the aqueous solution of these mixed electrolytes. On the other hand, the values *A* are negative for the mixed electrolytes which indicate the presence of weak ion-ion interaction in aqueous solutions of these mixed electrolytes: Li$_2$SO$_4$ + (NH$_4$)$_2$SO$_4$,Na$_2$SO$_4$ + K$_2$SO$_4$,Na$_2$SO$_4$ + Li$_2$SO$_4$,Na$_2$SO$_4$ + (NH$_4$)$_2$ C$_2$O$_4$,Na$_2$SO$_4$ + (NH$_4$)$_2$ C$_2$O$_4$,K$_2$SO$_4$ + Li$_2$SO$_4$,K$_2$SO$_4$ + (NH$_4$)$_2$C$_2$O$_4$,(NH$_4$)$_2$SO$_4$ + K$_2$SO$_4$; and NH$_4$)$_2$C$_2$O$_4$ + K$_2$C$_2$O$_4$. In each case the values of *A* tend to decrease with the rise of temperatures which suggests further weakening of ion-ion interactions at elevated temperatures.

All the mixed electrolyte systems except that of Na$_2$SO$_4$ + K$_2$SO$_4$ and (NH$_4$)$_2$SO$_4$ + K$_2$SO$_4$. The negative values of *B* are positive which tend to increase more positive with the rise of temperature there by suggesting the presence of strong ion-solvent interactions, which becomes stronger at elevated temperature. The negative value of *B* in the case of Na$_2$SO$_4$ + K$_2$SO$_4$ and (NH$_4$)$_2$ SO$_4$ + K$_2$SO$_4$ indicate the presence of weak ńon-solvent interactions.

The apparent molar volumes (Φ_v) of aqueous solutions of uni-bivalent mixed electrolyte systems have been determined as a function of y. keeping the ionic strength constant. The results have been presented in the table 2.

Table 2: Values of limiting apparent molar volume (ϕ_v^0) and experimental slope (S_v) of uni-bivalent mixed electrolyte systems in aqueous medium at different temperatures

Mixed electrolyte system	Ionic Strength (I)	$V_\phi^0 (cm^3 mol^{-1})$			$S_v (cm.^3 dm^{3/2}.mole^{-3/2})$		
		293.15K	303.15K	313.15K	293.15K	303.15K	313.15K
$Li_2SO_4 + (NH_4)_2SO_4$	3.0	101.06	103.06	104.45	-15.23	-16.18	-18.52
$Na_2SO_4 + K_2SO_4$	1.5	-229.22	-225.48	-220.66	215.63	214.99	214.29
$Na_2SO_4 + Li_2SO_4$	3.0	96.51	99.22	101.10	-28.02	-29.37	-31.17
$Na_2SO_4 + (NH_4)_2SO_4$	3.0	96.07	98.31	99.55	-25.26	-26.33	-28.86
$Na_2SO_4 + (NH_4)_2C_2O_4$	0.6	258.11	261.91	263.74	-473.63	-478.01	-479.70
$K_2SO_4 + Li_2SO_4$	1.5	67.76	77.52	82.48	-278.54	-279.30	-293.88
$K_2SO_4 + (NH_4)_2C_2O_4$	0.6	292.88	303.00	314.08	-1045.10	-1049.50	-1103.90
$(NH_4)_2SO_4 + K_2SO_4$	1.5	-198.43	-194.83	-190.85	270.43	269.35	255.59
$(NH_4)_2C_2O_4 + K_2C_2O_4$	1.5	157.86	160.76	161.56	14.94	14.30	10.51

For a single electrolyte the concentration dependence of $\Phi_v^{0\#}$ is given by Masson's equation[13].

$$\phi_v = \phi_v^0 + S_v \sqrt{C} \qquad \ldots\ldots\ldots\ldots\ldots..(11)$$

where $\Phi_v^{0\#}$ is called the limiting apparent molar volume and is equal to the partial molar volumes ($\overline{V}_2^0$) of the electrolyte at infinite dilution and S_v is the experimental slope.

The above relation has been applied to the present study of uni-bivalent mixed electrolyte systems where the plots of Φ_v versus $\sqrt{y}$ have been found to be non-linear, whereas those of Φ_v versus y are found to be linear. From this it follows that in aqueous solutions of uni-bivalent mixed electrolytes. The variation of Φ_v with fraction of ionic strength due to the first electrolyte in the mixture of two electrolytes i.e. y, at different temperatures is linear and is governed by the following modified form of Masson's equation[13].

$$\phi_v = \phi_v^0 + S_v y \qquad \ldots\ldots\ldots\ldots\ldots.. (12)$$

The values of $\Phi_v^{0\#}$ and experimental slope, S_v for uni-bivalent mixed electrolyte systems the aqueous solutions of have been obtained by computerized least square fit of the above equation. The values of $\Phi_v^{0\#}$ and S_v have been presented in Table 2.

It is seen that in aqueous medium, the values of $\Phi_v^{0\#}$ for all the uni-bivalent mixed electrolytes except for $Na_2SO_4 + K_2SO_4$ and $(NH_4)_2SO_4 + K_2SO_4$ mixed systems are positive and become larger with the rise of

temperature. This shows the presence of strong ion-solvent interactions. Which become increasingly stronger with the rise of temperature. The negative values of $\Phi_v^{0\#}$ in the case of aqueous solutions of for $Na_2SO_4 + K_2SO_4$ and $(NH_4)_2SO_4$ indicate the presence of weak ion solvent interactions.

The values of S_v are negative for all the uni-bivalent mixed electrolytes except that of $Na_2SO_4 + K_2SO_4$ $(NH_4)_2SO_4 + K_2SO_4$ and $(NH_4)_2C_2O_4 + K_2C_2O_4$ which show the presence of weak ion-ion interactions in these systems. This conclusion is agreement with these derived on the basic of values of coefficient *A* of Jones-Dole equation, However, for $Na_2SO_4 + K_2SO_4$ $(NH_4)_2SO_4 +K_2SO_4$and $(NH_4)_2C_2O_4 + K_2C_2O_4$ mixed electrolyte systems in aqueous medium the values of S_v are largely positive which suggest the presence of strong ion-ion interactions in these mixed electrolyte systems.

Free energies of activation for viscous flow
The free energies of activation of viscous flow, $\Delta\mu_1^{0\#}$ and $\Delta\mu_2^{0\#}$ per mole of solvent (water) and solute (mixed electrolyte systems) respectively, have been determined and presented in table 3 and 4.

It is seen that the values of $\Delta\mu_1^{0\#}$ are much larger as compared to those of $\Delta\mu_1^{0\#}$ for all the uni-bivalent mixed electrolytes except for $Na_2SO_4 + K_2SO_4$ and $(NH_4)_2SO_4 + K_2SO_4$ mixed systems where $\Delta\mu_2^{0\#}$ is negative and thus is smaller than $\Delta\mu_1^{0\#}$.

From this it follow that $(\Delta\mu_2^{0\#} > \Delta\mu_1^{0\#}) > 0$ for the following mixed electrolytes which show that in aqueous media, these mixed electrolytes behave as structure makers[14]:

Li_2SO_4 + $(NH_4)_2SO_4$, Na_2SO_4 + Li_2SO_4 Na_2SO_4 + $(NH_4)_2SO_4$, Na_2SO_4 + $(NH_4)_2C_2O_4$, K_2SO_4 + Li_2SO_4, K_2SO_4, +$(NH_4)_2C_2O_4$ and $(NH_4)_2C_2O_4$ + $K_2C_2O_4$.

On the other hand, ($\Delta\mu_2^{0\#} > \Delta\mu_1^{0\#}$) < 0 for the mixed electrolytes: Na_2SO_4 + K_2SO_4 and $(NH_4)_2SO_4$ + K_2SO_4, which shows that these mixed electrolyte systems behave as structure breakers.

Table 3 and 4: Values of $\Delta\mu_2^{0\#}$ for uni-bivalent mixed electrolyte systems in aqueous medium at different temperatures

Mixed electrolyte system	Ionic Strength (I)	$\Delta\mu_2^{0\#} (kJmol^{-1})$		
		293.15K	303.15K	313.15K
$Li_2SO_4 + (NH_4)_2SO_4$	3.0	66.28	71.12	75.32
$Na_2SO_4 + K_2SO_4$	1.5	-116.45	-113.89	-98.66
$Na_2SO_4 + Li_2SO_4$	3.0	138.28	145.29	156.75
$Na_2SO_4 + (NH_4)_2SO_4$	3.0	66.35	122.96	154.47
$Na_2SO_4 + (NH_4)_2C_2O_4$	0.6	90.43	144.89	174.95
$K_2SO_4 + Li_2SO_4$	1.5	50.45	62.80	64.95
$K_2SO_4 + (NH_4)_2C_2O_4$	0.6	143.87	189.93	213.24
$(NH_4)_2SO_4 + K_2SO_4$	1.5	-105.82	-77.47	-59.84
$(NH_4)_2C_2O_4 + K_2C_2O_4$	1.5	49.56	55.66	78.95

Free energy of activation, $\Delta\mu_1^{0\#} (kJmol^{-1})$ for water at different temperature

293.15K	303.15K	313.15K
20.58	20.71	20.85

Table 5: Values of Entropy of activation $(T\Delta S_2^{0\#})$ and Enthalpy of activation $(\Delta H_2^{0\#})$ for the aqueous solutions of uni-bivalent mixed electrolyte systems at different temperatures

Mixed electrolyte system	Ionic Strength (I)	$(T\Delta S_2^{0\#})$ (kJmol⁻¹)			$\Delta H_2^{0\#}$ (kJmol⁻¹)		
		298.15K	303.15K	298.15K	298.15K	303.15K	313.15K
$Li_2SO_4 + (NH_4)_2SO_4$	3.0	-132.63	-137-16	-141-68	198.91	208.27	217.00
$Na_2SO_4 + K_2SO_4$	1,5	-260.75	-269.64	-278.54	144.30	155.76	179.87
$Na_2SO_4 + Li_2SO_4$	3.0	-270.71	-279.94	-289.18	408.99	425.24	445.92
$Na_2SO_4 + (NH_4)_2SO_4$	3.0	-1291.69	-1335.75	-1379.82	1358.04	1458.72	1534.29
$Na_2SO_4 + (NH_4)_2C_2O_4$	0.6	-1238.89	-1281.15	-1323.41	1329.32	1426.04	1498.36
$K_2SO_4 + Li_2SO_4$	1.5	-212.48	-219.73	-226.98	262.93	282.53	291.93
$K_2SO_4 + (NH_4)_2C_2O_4$	0.6	-1016.75	-1051.43	-1086.12	1160.62	1241.37	1299.36
$(NH_4)_2SO_4 + K_2SO_4$	1.5	-673.96	-696.95	-719.94	568.14	619.48	660.10
$(NH_4)_2C_2O_4 + K_2C_2O_4$	1.5	-430.77	-445.47	-460.16	480.33	501.12	539.11

The values of entropy and enthalpy of activation for viscous flow for the aqueous solutions of uni-bivalent mixed electrolyte systems have been obtained at different temperatures (vide Eqns. 8&9) and the results have been presented in Table 3. It is seen that in each case the values of $T\Delta S_2^{0\#}$ are negative, while those of $\Delta H_2^{0\#}$ are positive and that $\Delta H_2^{0\#} t > T\Delta S_2^{0\#}$ thereeby suggesting that ion-solvent interactions for the mixed electrolyte systems, under discussion are nearly complete in the ground state. Further these values of $T\Delta S_2^{0\#}$ and

$\Delta H_2^{0\#}$ indicate that the transition state is associated with the bond breaking and increase in order[15-17].

Conclusion

From the volumetric and viscometric studies reported in this paper, it has been concluded the identical results are obtained .From both these studies with regard to ion-solvent and ion-ion interaction in aqueous solution of mixed uni-bivalent electrolyte.

Acknowledgement

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