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Viscosity B- coefficient and apparent molal volume studies of aqueous electrolytes solutions at 298.15 K

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ABSTRACT

KEYWORDS

Electrolytes; Dnsity; Viscosity; Ion interactions.

Viscosities and densities of solution of aqueous electrolytes at different molalities have been measured at 298.15 K. By using the data obtained and the appropriate thermodynamic equations, the parameters like apparent molal volume ϕ_v , partial molal volume ϕ_v^0 , relative viscosity η_{rel} , Jones-Dole B and Falkenhagen coefficient A, free energy of activation per mole of solvent $\Delta \mu_1^{0#}$ and free energy of activation per mole of solute $\Delta \mu_2^{0#}$ of the aqueous electrolytes solution have been computed. Results show that there are weak ion-ion interaction and ion-solvent interaction are very strong and greater structure making ability in case of MgCl₂, KNO₃ and Na₂SO₄. The order of structure making ability as Mg²⁺ > Na⁺ > K⁺ > Ba²⁺. © 2008 Trade Science Inc. - INDIA

1. INTRODUCTION

The work on viscosity coefficient of electrolytic solutions has been carried out by many workers^[1-3]. It has been found by number of workers that addition of electrolytes either breaks or makes the structure of liquid. Viscosity and its derived parameters provide valuable information regarding the shape and size of these molecules. The hydrophobic and hydrophilic properties of fluid has considered as a measure of solute-solvent and solute-solute interactions. Motin^[4] reported the apparent molal volume and viscosities of NaCl, NH₄Cl, CuCl₂, CuSO₄ and MgSO₄ in water and water + urea solutions at 308.15, 313.15 and 323.15 K. Therefore a deeper knowledge of the temperature influence of the pure and binary systems at a molecular is essential for the understanding of many chemical and biological processes in these media. In recent year some workers^[5-8] have utilized density and viscosity data deduced the thermodynamic properties of metal electrolytes solution. The measurement of viscosities of solutions from the basis of the studies of solute-solvent interactions. The metal ions play a vital role in life systems. In these work, the volumetric and viscometric studies can provide a meaningful information regarding solute-solute and solutesolvent interactions in aqueous electrolyte solutions. For these purpose, viscosities and densities of the aqueous electrolytes viz. NaCl, KCl, MgCl, BaCl, NaNO, KNO₃ CH₃COONa, CH₃COOK and Na₂SO₄ at different molalities (0.10, 0.15, 0.20, 0.25, 0.30, 0.35, 0.40, 0.45 and 0.50) m have been studied at 298.15 K. Several thermodynamic parameters such as relative viscosity, apparent molal volume ϕ_{y} , partial molal volume ϕ_{v}^{0} at infinite dilution, Jones –Dole coefficient^[9] B,

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Falkenhagen coefficient^[10] A, free energy of activation per mole of solvent $\Delta \mu_1^{0\#}$ and free energy of activation per mole of solute $\Delta \mu_2^{0\#}$ were computed from experimental data. Such data are expected to highlight the role of the cation and anion of an electrolyte is influencing its partial molal volume at infinite dilution in the solvent. These considerations prompted us to undertake the present study.

2. MATERIALS AND METHODS

NaCl and KCl (>99%), MgCl₂ and BaCl₂(99%), NaNO₃ KNO₃ CH₃COONa and CH₃COOK (99%) and Na_2SO_4 (>98 %) were procured from B. D. H. and A. C. S. grade. The chemicals with quoted purities were dried first in an oven at 110°C and then at room temperature in a vacuum over phosphorous pentaoxide for at least 48 hours and were then used without further purifications. The weighing were done on an Afcoset ER-120 A electronic balance with a precision of ± 0.1 mg. Viscosities measurements were carried out using a calibrated Ostwald viscometer at 298.15 K. Runs were repeated until three successive determinations were obtained within \pm 1s. Because all the flow times were greater than 100s. The estimated errors in viscosity measurements were of the order of ± 0.1 % Kg m⁻¹ s⁻¹. Densities of water and aqueous electrolytes solutions were measured using double capillary pycnometer made of Borosil glass. The capillary, with graduated marks, has uniform bore and could be closed by well fitting glass cap. The marks were calibrated with deionized and double distilled water at 298.15 K. The densities of water were determined from the mass of the solution in the pycnometer after reaching thermal equilibrium with a electronically thermostatic water bath at 298.15 K \pm 0.02 K. The accuracy of the density measurement was ± 0.01 Kg m⁻³.

3. RESULTS AND DISCUSSION

Viscosities and densities of the aqueous solutions of NaCl, KCl, MgCl₂, BaCl₂, NaNO₃, KNO₃, CH₃COONa, CH₃COOK and Na₂SO₄ at different molalities (0.10, 0.15, 0.20, 0.25, 0.30, 0.35, 0.40, 0.45 and 0.50) m have been measured at 298.15 K and reported in TABLE 1. The values of η and ρ decrease and increase the concentration of electrolytes in

Physical CHEMISTRY An Indian Journal

FABLE 1: Viscosities, η , densities, ρ , relative viscosities,
η_{rel} and apparent molal volume ϕ_{v} of electrolytes in water at
298.15 K

$m \pmod{1^{-1}}$	-η 10 ⁻³	ρ 10 ⁻³	mucl 10 ⁻³	φ _v 10 ⁻⁶
m (mor r)	(Nm^2s)	(kg m^{-3})	- ηrei 10	$(m^{3} mol^{-1})$
	So	dium chlo	ride	
0.10	0.5256	1000.9	0.6175	19.49
0.15	0.5422	1002.0	0.6369	25.84
0.20	0.5499	1004.0	0.6460	24.01
0.25	0.5665	1008.0	0.6655	14.88
0.30	0.5734	1009.1	0.6737	18.22
0.35	0.5780	1010.2	0.6791	21.64
0.40	0.5821	1011.0	0.6838	16.23
0.45	0.5847	1014.9	0.6869	19.80
0.50	0.6118	1026.1	0.7188	12.67
	Pota	assium ch	loride	
0.10	0.6531	1001.1	0.7673	35.66
0.15	0.6706	1002.3	0.7879	42.01
0.20	0.6740	1004.0	0.7918	40.17
0.25	0.6811	1005.1	0.8002	43.08
0.30	0.6896	1008.9	0.8102	34.99
0.35	0.7169	1012.0	0.8423	32.08
0.40	0.7353	1014.0	0.8639	32.40
0.45	0.7455	1019.1	0.8758	25.96
0.50	0.7934	1020.2	0.9321	28.84
	Magr	nesium ch	loride	
0.10	0.4617	1007.0	0.5424	104.81
0.15	0.4232	1008.2	0.4972	131.15
0.20	0.4115	1009.1	0.4835	144.32
0.25	0.3950	1013.2	0.4641	140.19
0.30	0.3945	1017.9	0.4635	134.09
0.35	0.3617	1023.8	0.4249	126.87
0.40	0.2975	1025.1	0.3495	133.99
0.45	0.2542	1026.0	0.2986	139.53
0.50	0.2095	1030.0	0.3046	137.94
	Ba	rium chlo	ride	
0.10	0.6188	1012.1	0.7270	85.52
0.15	0.6079	1020.2	0.7140	91.87
0.20	0.5954	1027.0	0.6995	90.04
0.25	0.5865	1040.1	0.6890	72.89
0.30	0.5677	1049.3	0.6670	71.48
0.35	0.5557	1055.0	0.6528	93.40
0.40	0.5417	1062.0	0.6364	87.28
0.45	0.5293	1077.1	0.6218	66.91
0.50	0.5102	1086.2	0.5994	66.71
Sodium nitrate				
0.10	0.6063	1001.1	0.7123	57.35
0.15	0.6101	1002.0	0.7168	52.61
0.20	0.6197	1003.4	0.7281	55.75
0.25	0.6292	1006.1	0.7392	49.61
0.30	0.6570	1010.9	0.7719	58.83
0.35	0.6863	1014.1	0.8062	36.86 25.20
0.40	0.7219	1017.2	0.0401	33.39

TABLE 1 countinue next page

Full Paper

				TA
m (mol l ⁻¹)	-η 10 ⁻³ (N m ⁻² s)	ρ 10 ⁻³ (kg m ⁻³)	- ηrel 10 ⁻³	$\phi_v 10^{-6}$ (m ³ mol ⁻¹)
	So	dium chlo	oride	
0.10	0.5256	1000.9	0.6175	19.49
0.15	0.5422	1002.0	0.6369	25.84
0.20	0.5499	1004.0	0.6460	24.01
0.25	0.5665	1008.0	0.6655	14.88
0.30	0.5734	1009.1	0.6737	18.22
0.35	0.5780	1010.2	0.6791	21.64
0.40	0.5821	1011.0	0.6838	16.23
0.45	0.5847	1014.9	0.6869	19.80
0.50	0.6118	1026.1	0.7188	12.67
	Pot	assium ch	loride	
0.10	0.6531	1001.1	0.7673	35.66
0.15	0.6706	1002.3	0.7879	42.01
0.20	0.6740	1004.0	0.7918	40.17
0.25	0.6811	1005.1	0.8002	43.08
0.30	0.6896	1008.9	0.8102	34.99
0.35	0.7169	1012.0	0.8423	32.08
0.40	0.7353	1014.0	0.8639	32.40
0.45	0.7455	1019.1	0.8758	25.96
0.50	0.7934	1020.2	0.9321	28.84
	Mag	nesium ch	loride	
0.10	0.4617	1007.0	0.5424	104.81
0.15	0.4232	1008.2	0.4972	131.15
0.20	0.4115	1009.1	0.4835	144.32
0.25	0.3950	1013.2	0.4641	140.19
0.30	0.3945	1017.9	0.4635	134.09
0.35	0.3617	1023.8	0.4249	126.87
0.40	0.2975	1025.1	0.3495	133.99
0.45	0.2542	1026.0	0.2986	139.53
0.50	0.2095	1030.0	0.3046	137.94
	Ba	arium chlo	oride	
0.10	0.6188	1012.1	0.7270	85.52
0.15	0.6079	1020.2	0.7140	91.87
0.20	0.5954	1027.0	0.6995	90.04
0.25	0.5865	1040.1	0.6890	72.89
0.30	0.5677	1049.3	0.6670	71.48
0.35	0.5557	1055.0	0.6528	93.40
0.40	0.5417	1062.0	0.6364	87.28
0.45	0.5293	1077.1	0.6218	66.91
0.50	0.5102	1086.2	0.5994	66.71
	S	odium nit	rate	
0.10	0.6063	1001.1	0.7123	57.35
0.15	0.6101	1002.0	0.7168	52.61
0.20	0.6197	1003.4	0.7281	55.75
0.25	0.6292	1006.1	0.7392	49.61
0.30	0.6570	1010.9	0.7719	38.83
0.35	0.6863	1014.1	0.8062	36.86
0.40	0.7219	1017.2	0.8481	35.39
0.45	0.9040	1019.0	1.062	36.47
0.50	0.9105	1022.0	1.069	35.33

Potassium nitrate					
0.10	0.6700	1001.1	0.5703	62.48	
0.15	0.7598	1005.3	0.6467	48.71	
0.20	0.8095	1006.1	0.6890	56.87	
0.25	0.8920	1009.1	0.7592	53.74	
0.30	0.9027	1012.2	0.7683	51.65	
0.35	0.9223	1015.0	0.7850	50.65	
0.40	0.9269	1017.1	0.7889	51.55	
0.45	1.0231	1022.0	0.8709	45.95	
0.50	1.0304	1024.0	0.8770	47.48	
	S	odium aceta	ate		
0.10	0.6465	1001.3	0.7595	120.62	
0.15	0.3723	1001.8	0.4373	115.21	
0.20	0.2026	1002.0	0.2381	117.01	
0.25	0.1827	1003.0	0.2147	112.88	
0.30	0.1664	1005.2	0.1955	110.13	
0.35	0.1478	1008.8	0.1736	102.43	
0.40	0.1224	1011.2	0.1438	101.67	
0.45	0.1084	1015.0	0.0987	96.62	
0.50	0.1059	1016.1	0.0702	98.60	
		Potassium	acetate		
0.10	0.5930	1000.2	0.6967	75.55	
0.15	0.5609	1001.0	0.6590	79.16	
0.20	0.5543	1001.0	0.6512	78.70	
0.25	05251	1003.3	0.6169	70.82	
0.30	0.5142	1004.2	0.6041	78.76	
0.35	0.5095	1006.0	0.5986	72.97	
0.40	0.4492	1009.2	0.5278	68.63	
0.45	0.4379	1012.1	0.5144	65.26	
0.50	0.4360	1014.2	0.5122	64.56	
Sodium sulphate					
0.10	0.6229	1011.0	0.7318	23.44	
0.15	0.5599	1015.0	0.6578	22.90	
0.20	0.4651	1016.1	0.5464	47.77	
0.25	0.4504	1017.1	0.5291	62.70	
0.30	0.4408	1019.1	0.5178	69.30	
0.35	0.3502	1021.3	0.4115	74.02	
0.40	0.2620	1023.2	0.3079	77.51	
0.45	0.1322	1026.1	0.1553	78.08	
0.50	0.082	1028.4	0.0967	80.51	

water, under investigation, which appear to be hydrophobic properties of solutes i.e. H- bond forming. The change in structure of solvent or solution as a result of H-bond formation leads to decrease in intermolecular free length.

The apparent molal volume, ϕ_v were calculated from measured density data using the following equation : $\phi_v = [1000 (\rho^0 - \rho) / m \rho^0] + M / \rho^0$ (1) where M is the molecular weight of the solute, m is the molality of the solutions, ρ and ρ^0 are the densities of solution and solvent respectively. The values of apparent molal volume, ϕ_v of these systems are recorded in TABLE 1.



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Positive values of ϕ_v (TABLE 1) of these systems indicating the presence of strong solute-solvent interactions. The contraction of water in vicinity of charged group is caused by electrostatic solute-solvent interactions. The strong solute-solvent interactions are indicating the presence of large electrostriction effect in these systems. The partial molal volume, ϕ_v^{0} at infinite dilution, was evaluated by taking an average of all the data points, standard deviations pertain to the mean value^[11]. In all the other cases the standard partial molal volume were obtained by least square treatment to the plot of ϕ_v versus m^{1/2}, using Masson's equation^[12] $\phi_v = \phi_v^{0} + S_v m^{1/2}$ (2)

Here, S_v is the experimental slope, which is considered to be volumetric pair wise interaction coefficient^[13,14]. The derived values of ϕ_v^0 along with coefficient S_v are summarized in TABLE 2. The values of ϕ_v^0 for NaCl, KCl, MgCl₂, BaCl₂, NaNO₃, KNO₃, CH₃COONa, CH₃COOK and Na₂SO₄ are positive, the presence of strong interaction between ions and water molecules i.e. increase the electrostriction effect.

The salt of NaCl, KCl and $MgCl_2$ contain monovalent and bivalent cations but the same anion. The cations are of different nature and are hydrated by water molecules. According to Vaslows^[15], concept of hydration, small cations normally enhance hydrogen bonded structural grouping in liquid. As radius of Mg^{2+} , is the smallest it may occupy the smallest space in the hydration sheath. For the larger radius of K^+ , it occupies a larger space in the hydration sheath than Na⁺. That is the interaction of Mg^{2+} with water is greater than Na⁺ and K⁺.

The BaCl₂, NaNO₃, KNO₃, CH₃COONa, CH₃COOK and Na₂SO₄ contain cations Ba⁺², Na⁺, K⁺ and their anions are Cl⁻, CH₃COO⁻, NO₃⁻ and SO₄⁻²⁻ respectively. The behavior of cations and anions differ essentially in the presence of each other, which may influence the water structure differently. The larger size of Cl⁻, CH₃COO⁻, NO₃⁻ and SO₄⁻²⁻ anion may occupy a larger space in the hydration sheath. The SO₄⁻²⁻ should be more highly solvated than Cl⁻, CH₃COO⁻ and NO₃⁻ due to its higher number electronegative groups. Sulphate ion, containing four oxygen atoms, may enhance, the formation of hydrogen bonding in water solutions^[16].

Among the electrolytes studied (NaCl, KCl, MgCl₂, BaCl₂, NaNO₃, KNO₃, CH₃COONa, CH₃COOK and Na₂SO₄), the ϕ_v^0 values obtained for MgCl₂ and Na₂SO₄ are seen to be abnormally higher than those of the other electrolytes (shown in TABLE

Physical CHEMISTRY Au Judian Journal

TABLE 2:	Partial mola	r volume, ø º	, at infinite
dilution,exp	erimental slope, S	5 Falkenhager	coefficient, A,
Jones-Dole	coefficient, B, fre	e energy of act	ivation for the
solvent, Δµ ⁰	$_{1}^{*}$ and solute, $\Delta \mu$	^{0#} , of electroly	es in water at
298.15 K	1 .	2	

Electrolytes $\phi_v^0 10^{-6}$ $(m^3 mol^{-1})$	$\begin{array}{c} S_v 10^{-6} \\ (m^3 mol^{-3/2} \\ l^{-1/2}) \end{array}$	A (dm ^{3/2} mol ^{-1/2})	B (dm ³ mol ⁻¹)	$\frac{\Delta \mu^{0\#}_{1} = 9.051}{0^{-4}}$ $\frac{\Delta \mu^{0\#}_{2} 10^{-4}}{(\text{kJ mol}^{-1})}$
NaCl 20.08	-24.89	-5.96	5.27	72.23
KCl 20.82	-20.11	-6.13	5.17	71.11
MgCl ₂ 246.13	178.00	-5.93	5.93	84.57
BaCl ₂ 39.45	-38.93	-10.47	15.10	20.72
NaNO ₃ 38.77	27.00	-6.91	6.68	92.02
KNO ₃ 62.56	18.50	-8.13	8.20	30.82
CH ₃ COONa 17.08	-126.25	-5.77	4.88	67.00
CH ₃ COOK 64.49	-10.00	-5.51	3.33	46.37
Na ₂ SO ₄ 218.53	331.66	-4.64	7.40	10.45

2). This abnormally may be accounted for by the fact that Mg^{+2} and Na^+ can form an octahedral complex with water^[4]. TABLE 2 shows that the value of S_v are positive in case of the systems $MgCl_2$, $NaNO_3$, KNO_3 and Na_2SO_4 indicating, thereby strong ion-pair interactions. On the basis of studies conducted on a variety of hydrophilic, hydrophobic and amphiphilic solutes, it has been shown that positive S_v values are generally associated with solutes showing an hydrophilic character^[17] as in the $MgCl_2$, $NaNO_3$, KNO_3 and Na_2SO_4 . The negative values of S_v obtained in the case of NaCl, KCl, $BaCl_2$, CH_3COONa and CH_3COOK which indicates ion- ion interactions are weak and less complex ion formation.

Viscosities and relative viscosities of the aqueous electrolytes solution NaCl, KCl, MgCl₂ BaCl₂, NaNO₃, KNO₃, CH₃COONa, CH₃COOK and Na₂SO₄ at different molalities are recorded in TABLE 2. The relative viscosities of these systems were determined at 298.15 K from the viscosity data. These relative viscosities were analyzed by the Jones-Dole^[9] semi empirical equation :

$$\eta_{m1} = \eta / \eta_0 = [1 + Am^{1/2} + Bm]$$
(3)

where η and η_0 are the viscosities of solution and solvent respectively. A is the Falkenhagen coefficient which indicates ion-pair electrostatic interaction and B is the Jones – Dole coefficient, an empirical constant, and is measure of ion-solvent interaction. Its values of B depend on the size and shape of the solute particles. They were obtained by a least square treatment as intercept and slope of the linear plots of $\eta / \eta_0 - 1$

$/ m^{1/2} versus m^{1/2}$.

A per usual of TABLE 2 shows that the values of B is positive and large for the NaCl, KCl, MgCl₂, BaCl₂, NaNO₃, KNO₃, CH₃COONa, CH₃COOK and Na₂SO₄ solutions. They shows that the presence of very strong ion-solvent interaction Mg⁺² ion being smaller in size than Na⁺, K⁺ and Ba⁺² has an intense force field and hence strong hydration around it. Therefore hydration of MgCl₂ and Na₂SO₄. The SO₄⁻² ions should be more highly solvated due to its higher number of electronegative groups. Sulphate ion containing four oxygen atoms, may enhance the formation of hydrogen bonding in water solution. The values of A coefficient are negative in these systems thereby showing the presence of weak ion-ion interactions. Yasmin and coworkers^[8] have observed such anomalous behavior of A and B.

According to the transition state theory of the relative viscosity of the electrolyte solutions proposed by Feakins et al.^[18]the B- coefficient is given as,

$\mathbf{B} = (\overline{\mathbf{V}}_1^0 - \overline{\mathbf{V}}_2^0) / 1000 + \overline{\mathbf{V}}_1^{0} \, {}^{0}[(\Delta \mu^{0\#}_2 - \Delta \mu^{0\#}_1) / \, \mathbf{RT}] / 1000 \qquad (4)$

where $\Box V_1^0$ and $\Box V_2^0$ are the partial molal volumes of the solvent and solute at infinite dilution respectively, $\Delta \mu_1^{0\#}$ is the free energy of activation per mole of the solvent and $\Delta \mu_2^{0\#}$ is the free energy of activation per mole of the solute. The values of $\Delta \mu_1^{0\#}$ and $\Delta \mu_2^{0\#}$ were calculated using the following equations: $\Delta \mu_1^{0\#} = \mathbf{RT} \ln (\eta_0 V_1^0 / hN)$ (5) and

$$\Delta \mu_{2}^{0\#} = \Delta \mu_{1}^{0\#} + (RT / \overline{V}_{1}^{0}) [1000B - (\overline{V}_{1}^{0} - \overline{V}_{2}^{0})]$$
(6)

where N is the Avogadro's number. R and h are the gas and Planck's constant. T is the absolute temperature. The values of $\Delta \mu^{0\#}_{1}$ is constant and $\Delta \mu^{0\#}_{2}$ is dependent mainly on B-coefficient and $\Box V_{1}^{0-} \Box V_{2}^{0}$ terms. It is also clear from TABLE 2 that the values of $\Delta \mu^{0\#}_{2}$ are larger than $\Delta \mu^{0\#}_{1}$ suggesting that the formation of transition state is accompanied by the breaking and distortion of the intermolecular bonds. The values of $\Delta \mu^{0\#}_{2}$ are large of NaCl, KCl, MgCl₂, BaCl₂, NaNO₃, KNO₃, CH₃COONa, CH₃COOK and Na₂SO₄ solutions reveals the order of structure making ability as Mg⁺² > K⁺ > Na⁺ > Ba⁺². T

he hydration of Mg^{+2} is stronger than that of K⁺, Na⁺ and Ba⁺² ion since the electric charge of Mg^{+2} is twice that of K⁺ and Na⁺ and the radius of the former is much shorter than that in later. This suggests maximum structure making ability and extent of interaction is greater in case of MgCl₂ and KNO₃. The studies suggest that ion-solvent interaction in these systems.

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