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## RESEARCH ARTICLE

## Viscosity, Apparent Molar Volume and Thermodynamic Parameters of Lanthanum Chloride in Absolute and Aqueous Ethanol Mixtures

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## ABSTRACT

Viscosities and apparent molar volumes of different concentrations of lanthanum chloride from  $1 \times 10^{-2}$  to  $9 \times 10^{-2}$  mol dm<sup>-3</sup> have been studied in absolute and aqueous ethanol mixtures at temperatures ranging from 303 to 323 K. Viscosity and density data were analyzed by using two different relations for the prediction of nature of lanthanum chloride in absolute and aqueous ethanol systems. *A* and *B*-coefficients of Jones-Dole equation show ion-ion and ion-solvent interactions respectively. The density data was analyzed in terms of limiting apparent molar volume ( $\phi_v^0$ ) and experimental slopes ( $S_v$ ) obtained from Masson equation has been used to interpret the ion-solvent interaction and ion-ion interaction respectively. On the basis of results evaluated by using the different parameters it was concluded that lanthanum chloride behaves as a structure maker in absolute ethanol and 90 % (v/v) aqueous ethanol, while as the aqueous quantity increases such as 80 to 70 % (v/v) it behaves as structure breaker. Thermodynamic parameters such as energy of activation ( $E_a$ ), free energy change of activation ( $\Delta G^*$ ) and entropy change of activation ( $\Delta S^*$ ) for viscous flow have been evaluated as a function of concentration, solvent and temperature.

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## INTRODUCTION

Methods of physico-chemical analysis have been found to be useful tools in getting sound information about the structure of various liquids and in studying liquid-liquid interactions in binary and ternary systems. These analyses are based on the dependence of physical properties (additive, constitutive and colligative) on the compositions and external conditions of a liquid system. The physico-chemical properties of mixtures of polar and nonpolar liquids such as alcohols and hydrocarbons out of several classes of binary systems have drawn considerable attention from many investigators (Chowdhury *et al.*, 2001). The nature of a solute in a binary solvent system has a fundamental importance in the study of chemical and physical properties of solution regarding to their ionic interactions between solute-solute and solute-solvent. The reason of understanding of the phenomenon attained the curiosity of many researchers (Donald *et al.*, 1995; Khan *et al.*, 2005; Lomesh *et al.*, 2006; Koseli *et al.*, 2006) and to date in spite of colossal amount of research data for pure solvent system (Saeed and Uddin, 2001; Dhanaragu *et al.*, 1997; Choudhury and Roy, 2005), these generate a lot of scanty for mixed solvent system.

Viscosities and Apparent molar volumes of electrolytes provide valuable information about ion-ion, ion-solvent and solvent-solvent interactions (Das and Hazra, 1997; Nikam and Sawant, 1998; Wang *et al.*, 2000; Qadeer and Khalid, 2005; Millero, 1971; Redlick and Meyer, 1964; Saeed *et al.*, 2002; Das and Roy, 2006). In case of water-ethanol binary system, water is a polar solvent while ethanol has a relatively low value of dipole moment and dielectric constant ( $\mu = 5.64 \times 1030$  cm and  $\epsilon_r = 24.3$  at 298.15 K), self associated through hydrogen bonding into chain like association. Thus the behavior of solute is different in pure and aqueous ethanol mixture due to difference in ionic interactions. The electrostatic forces, which tend to maintain a quasilattice of the ions in the solutions, would increase

the viscosity of solutions. The relation of these forces to the concentration in very dilute solutions was expressed by Jones-Dole relation (Jones and Dole, 1929):

$$\eta_{sp} / \sqrt{C} = A + B \sqrt{C} \quad (1)$$

where *C* is the concentration of the electrolyte solution,  $\eta_{sp}$  is the specific viscosity of solution. *A* and *B* are coefficients represent the ion-ion and ion-solvent interactions respectively. The apparent molar volumes ( $\phi_v$ ) were calculated from the density of the solution using equation (Parmar and Guleria, 2005).

$$\phi_v = M / \rho_o - 1000(\rho - \rho_o) / C\rho_o \quad (2)$$

where *M* is the Molecular weight of solute, *C* is the concentration of the electrolyte solution,  $\rho$  is the density of the solution and  $\rho_o$  is the density of the solvent. The purpose of study was to evaluate the structural changes in terms of ion-ion and ion solvent interactions of lanthanum chloride in aqueous ethanol system. An attempt was also made to evaluate the thermodynamic parameters such as energy of activation ( $E_a$ ), free energy change of activation ( $\Delta G^*$ ) and entropy change of activation ( $\Delta S^*$ ) as a function of concentration of lanthanum chloride, ethanol and temperature.

## MATERIALS AND METHODS

Reagent grade lanthanum chloride of E. Merck was used without further purification. The ethanol of Analar grade was used. Aqueous ethanol mixtures ranges from 70 to 90 % (v/v) were prepared in double distilled water having specific conductance  $0.06 \mu\text{S cm}^{-1}$ . All the glassware used were of Pyrex A grade quality. The densities of solvent and solutions were measured with relative density bottle having the capacity of 10 cm<sup>3</sup>. Ostwald viscometer type Techniconominal constant 0.1 Cs/S capillary ASTM D 445, was used to measure the viscosity at different temperatures. In order to keep

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the temperature constant throughout the course of experiment, a thermostatic water bath (type Haake-13, Karlsruhe, Germany) was used. Lanthanum chloride solutions in the concentration ranging from  $1.0 \times 10^{-2}$  to  $9.0 \times 10^{-2}$  mol dm<sup>-3</sup> were prepared in absolute and aqueous ethanol mixtures. A known volume of solution was taken in viscometer, which is vertically placed in a glass tube attached with thermostatic water bath having a constant circulation of water to maintain constant temperature during the experimental work. Time flow for solvent and solutions were taken by stopwatch having a least count of  $\pm 0.02$  seconds. Reproducibility of the results was checked by taking each measurement three times.

## RESULTS AND DISCUSSION

The values of viscosities and densities of different concentrations of lanthanum chloride ranging from  $1 \times 10^{-2}$  to  $9 \times 10^{-2}$  mol dm<sup>-3</sup> in ethanol and aqueous ethanol mixtures at different temperatures ranging from 303 to 323 K are tabulated in Table 1 and 2 respectively. The results show an increase in viscosity with increase in concentration of lanthanum chloride at fixed temperature. This are due to the fact that with increasing concentration of salt, the number of collision between the molecules also increased resulting a loss in kinetic energy, therefore the molecules tend to stick together which increases the viscosity. The viscosities were found to decrease with increasing temperature and increase in concentration of ethanol. Results show that in absolute ethanol the values of viscosities were lower as compare to that in aqueous ethanol mixtures because of increase in hydration of ethanol molecules with increasing water contents which decreases the viscosity of lanthanum chloride. From density data it was shown that with the increase in concentration of lanthanum chloride density increases because of increase in number of molecules per unit volume while reverse behaviour was observed with the increase in temperature due to increase in volume of solution. The values of density decreased with the increase in percent composition of ethanol (70 - 100 %) because of increase in hydration of ethanol molecules as shown in Table 2. The values of ion-ion interactions and ion-solvent interactions in terms of *A* and *B*-coefficients of Jones-Dole parameters are shown in Table 3. *A* and *B*-coefficients were evaluated from the intercept and slope of the linear plot of  $\eta_{sp}/\sqrt{C}$  versus  $\sqrt{C}$  respectively. Representative plot of  $\eta_{sp}/\sqrt{C}$  versus  $\sqrt{C}$  is shown in Fig 1. The negative values of *A*-coefficient show less ion-ion interaction. The *B*-coefficient of the Jones-Dole empirical relation expressed of the relative viscosities of electrolyte solutions as a function of their concentration provide information concerning the solvation of the ions and their effect on the structure

**Table 1. Viscosities ( $\eta$ ) of lanthanum chloride in absolute and aqueous ethanol mixtures at different temperatures**

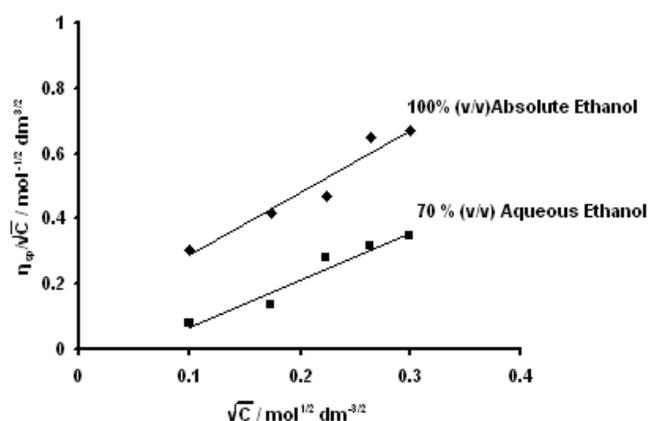
[LaCl <sub>3</sub> ].10 <sup>2</sup> (mol dm <sup>-3</sup> )	$\eta$ /(m.P) at temperatures (K)				
	303	308	313	318	323
<b>Absolute Ethanol</b>					
1.0	1.083	0.957	0.861	0.789	0.732
3.0	1.128	0.996	0.907	0.803	0.749
5.0	1.162	1.047	0.927	0.835	0.764
7.0	1.231	1.072	0.967	0.846	0.776
9.0	1.263	1.090	0.982	0.865	0.788
<b>90% (v/v) Aqueous Ethanol</b>					
1.0	1.210	1.129	0.996	0.903	0.831
3.0	1.304	1.146	1.031	0.933	0.844
5.0	1.367	1.223	1.061	0.960	0.857
7.0	1.433	1.249	1.082	1.004	0.884
9.0	1.509	1.302	1.106	1.025	0.920
<b>80% (v/v) Aqueous Ethanol</b>					
1.0	1.310	1.298	1.139	1.038	0.919
3.0	1.541	1.350	1.197	1.071	0.962
5.0	1.680	1.424	1.249	1.168	1.009
7.0	1.512	1.461	1.307	1.218	1.036
9.0	1.643	1.496	1.362	1.245	1.099
<b>70% (v/v) Aqueous Ethanol</b>					
1.0	1.640	1.403	1.173	1.066	0.949
3.0	1.667	1.450	1.230	1.109	0.990
5.0	1.729	1.490	1.260	1.142	1.034
7.0	1.760	1.520	1.300	1.196	1.069
9.0	1.798	1.550	1.340	1.236	1.144

**Table 2. Densities ( $\rho$ ) of lanthanum chloride in absolute and aqueous ethanol mixtures at different temperatures.**

[LaCl <sub>3</sub> ].10 <sup>2</sup> (mol dm <sup>-3</sup> )	$\rho$ /(g.cm <sup>-3</sup> ) at temperatures (K)				
	303	308	313	318	323
<b>Absolute Ethanol</b>					
1.0	0.8177	0.8157	0.8118	0.8067	0.8031
3.0	0.8230	0.8219	0.8204	0.8144	0.8060
5.0	0.8276	0.8257	0.8249	0.8174	0.8099
7.0	0.8336	0.8277	0.8284	0.8211	0.8181
9.0	0.8416	0.8351	0.8310	0.8292	0.8262
<b>90% (v/v) Aqueous Ethanol</b>					
1.0	0.8524	0.8468	0.8459	0.8413	0.8330
3.0	0.8581	0.8512	0.8518	0.8483	0.8428
5.0	0.8595	0.8545	0.8570	0.8494	0.8439
7.0	0.8656	0.8579	0.8594	0.8540	0.8461
9.0	0.8659	0.8637	0.8601	0.8548	0.8535
<b>80% (v/v) Aqueous Ethanol</b>					
1.0	0.8874	0.8802	0.8769	0.8756	0.8719
3.0	0.8927	0.8887	0.8852	0.8820	0.8774
5.0	0.8984	0.8921	0.8873	0.8837	0.8820
7.0	0.9033	0.8964	0.8951	0.8919	0.8876
9.0	0.9094	0.9005	0.8981	0.8945	0.8919
<b>70% (v/v) Aqueous Ethanol</b>					
1.0	0.9161	0.9105	0.9071	0.9028	0.8998
3.0	0.9211	0.9142	0.9124	0.9080	0.9036
5.0	0.9257	0.9187	0.9148	0.9144	0.9088
7.0	0.9303	0.9259	0.9238	0.9197	0.9142
9.0	0.9354	0.9282	0.9264	0.9213	0.9192

**Table 3. Values of *A* and *B*-coefficient of Jones-Dole parameters for lanthanum chloride in absolute and aqueous ethanol mixtures at different temperatures**

Ethanol (% v/v)	<i>A</i> and <i>B</i> -coefficients at different temperatures (K)				
	303	308	313	318	323
<b><i>A</i>-coefficient /(dm<sup>3</sup>mol)<sup>-1/2</sup></b>					
100	0.093	0.007	0.540	-0.120	-0.092
90	-0.422	-0.334	0.217	0.137	-0.059
80	-0.014	-0.105	-0.090	-0.078	-0.138
70	-0.081	-0.264	-0.337	-0.376	0.523
<b><i>B</i>-coefficient /(dm<sup>3</sup>mol<sup>-1</sup>)</b>					
100	1.92	1.85	1.00	0.86	0.50
90	2.13	2.77	1.25	1.62	1.35
80	1.65	2.33	2.72	3.00	3.30
70	1.46	2.07	2.56	2.80	3.27



**Fig 1. Plot of  $\eta_{sp}/\sqrt{C}$  versus  $\sqrt{C}$  for lanthanum chloride in absolute and 70% (v/v) aqueous ethanol mixture at 303 K.**

of the solvent in the near environment of the solute particles (Donald *et al.*, 1995). It is a measure of effective solvodynamic volume of solvated ions, the size and shape of solute as well as structural effects induced by the solute-solvent interactions make a major contribution to relative viscosity. When a solute dissolve in a solvent, a hole is made in the liquid with rupture of intermolecular bonds, and the solute is inserted. Some of the solvent molecules are attached to the ions because ion-solvent interactions and this causes an increase in the viscosity of solution show a positive contribution to the viscosity *B*-coefficient. On the other hand, these solvent molecules must be

wrenched out of the bulk solvent and this breaking of the solvent structure causes a decrease in viscosity of solution results a negative contribution to viscosity  $B$ -coefficient. Thus, the  $B$ -coefficient is the resultant of these two opposite factors (Janardhan and Sivasankar, 1978). The lower value of  $B$ -coefficient in absolute ethanol indicate structure making nature of lanthanum chloride, while as the content of alcohol decreased from absolute to 70 % (v/v) aqueous ethanol the value of  $B$ -coefficient increased show structure breaking nature of lanthanum chloride. The results show that the values of  $B$ -coefficient decreased with rise in temperature in absolute ethanol and 90 % (v/v) aqueous ethanol represent structure promoting effect due to the ordering and a sort of enforcement due to the solvent around the solute show structure maker. While in 70 and 80 % (v/v) aqueous ethanol mixtures increased in values of  $B$ -coefficient with rise in temperature show structure breaking of regular arrangement of solvent molecules due to dipole interaction with solute. The densities measured for the solution of lanthanum chloride in absolute ethanol and aqueous ethanol at different temperatures is shown in Table 4 have been used to calculate the apparent molar volumes. The limiting

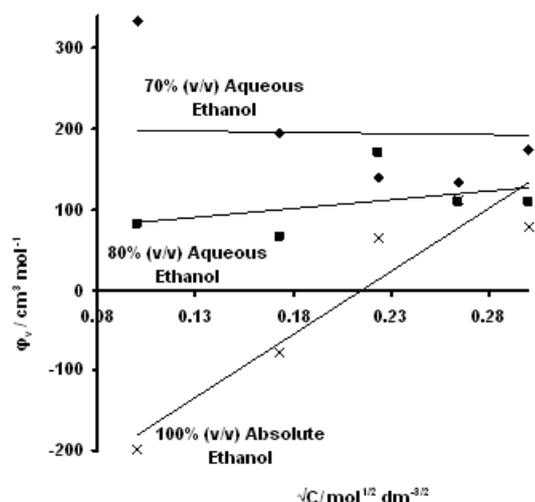
**Table 4. Values of  $\phi_v^0$  and  $S_v$  for lanthanum chloride in absolute and aqueous ethanol mixtures at different temperatures.**

Ethanol (% v/v)	$\phi_v^0$ and $S_v$ at different temperatures (K)				
	303	308	313	318	323
	$\phi_v^0 \times 10^{-2} / (\text{cm}^3 \text{mol}^{-1})$				
100	-6.049	-13.43	-9.619	-3.367	-3.727
90	1.400	2.278	-4.391	-2.686	3.254
80	-2.122	5.630	2.943	0.610	4.804
70	2.366	2.885	4.564	3.770	2.536
	$S_v \times 10^{-2} / (\text{cm}^3 \text{mol}^{-3/2})$				
100	24.21	51.25	36.50	15.66	17.88
90	2.390	-0.830	21.06	16.12	-6.388
80	11.01	-14.64	-6.551	2.192	-11.75
70	-3.142	-4.421	-10.72	-8.578	-3.477

apparent molar volumes ( $\phi_v^0$ ) and experimental slopes ( $S_v$ ) were determined by applying the least square method to the plots of  $\phi_v$  versus  $\sqrt{C}$  using the Masson's equation.

$$\phi_v = \phi_v^0 + S_v \sqrt{C} \quad (3)$$

The values of  $\phi_v^0$  is also related to ion-solvent interaction and  $S_v$  is a constant dependent on charge and salt type and can be related to ion-ion interactions. The plot of  $\phi_v$  and  $\sqrt{C}$  were linear in all cases and from the intercept and slope, the values of  $\phi_v^0$  and  $S_v$  respectively can be obtained as shown in Fig 2.



**Fig 2. Plot of  $\phi_v$  versus  $\sqrt{C}$  for lanthanum chloride in absolute and 80% (v/v) and 70% (v/v) aqueous ethanol mixture at 318 K.**

Table 4 reveals that the values of  $\phi_v^0$  were negative for absolute and 90 % ethanol which indicates presence of weak ion-solvent interaction while in case of 80 % and 70 % ethanol positive  $\phi_v^0$

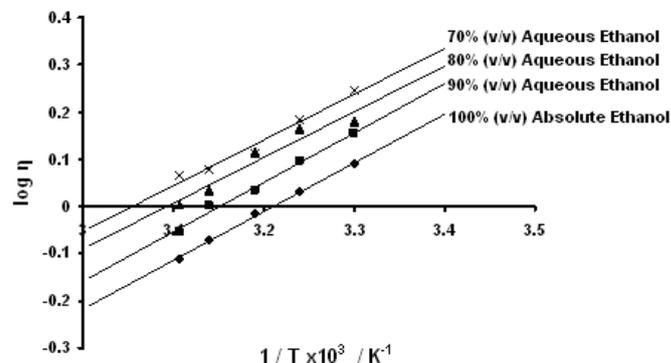
values show strong ion-solvent interaction due to increase in water content. It is also clear from Table 4 that values of  $S_v$  are negative for absolute and 90 % ethanol which indicate weak ion-ion interaction. The positive and large value of  $S_v$  for 80 % and 70 % ethanol indicates strong ion-ion interaction (Parmar and Guleria, 2005). Because of the nature of  $B$ -coefficients and their ion additive properties potentially important correlations exist between the coefficients and other ion additive properties. In this sense the  $B$ -coefficients may provide the key for the validation of a host of thermochemical data (Donald *et al.*, 1995). The results for the thermodynamic parameters such as energy of activation ( $E_\eta$ ), free energy change of activation ( $\Delta G^*$ ) and entropy change of activation ( $\Delta S^*$ ) in absolute and aqueous ethanol mixtures are tabulated in Table 5. The effect of temperature on viscosity is given by (Roy and Jha, 2001):

$$\eta = A \exp(E_\eta / RT) \quad (4)$$

**Table 5. Thermodynamic parameters for lanthanum chloride in absolute and aqueous ethanol mixtures at 303 K.**

$[LaCl_3] \cdot 10^2$ (mol dm <sup>-3</sup> )	Thermodynamic Parameters at 303 (K)		
	Energy of Activation ( $E_\eta$ ) (kJ mol <sup>-1</sup> )	Free Energy change of Activation ( $\Delta G^*$ ) (kJ mol <sup>-1</sup> )	Entropy change of Activation ( $\Delta S^*$ ) (kJ mol <sup>-1</sup> K <sup>-1</sup> )
<b>Absolute Ethanol</b>			
1.0	15.75	64.93	-0.161
3.0	16.83	65.02	-0.159
5.0	17.36	65.10	-0.158
7.0	18.85	65.26	-0.165
9.0	19.08	65.34	-0.167
<b>90% (v/v) Aqueous Ethanol</b>			
1.0	15.94	64.46	-0.159
3.0	17.50	65.18	-0.157
5.0	19.17	65.29	-0.152
7.0	19.36	65.42	-0.151
9.0	20.06	65.56	-0.150
<b>80% (v/v) Aqueous Ethanol</b>			
1.0	19.67	65.22	-0.153
3.0	19.22	65.32	-0.152
5.0	18.12	65.40	-0.156
7.0	18.06	65.50	-0.157
9.0	16.81	65.56	-0.164
<b>70% (v/v) Aqueous Ethanol</b>			
1.0	22.36	65.19	-0.140
3.0	21.38	65.26	-0.145
5.0	21.14	65.35	-0.146
7.0	20.20	65.41	-0.149
9.0	18.50	65.48	-0.155

where  $A$  is constant,  $E_\eta$  is the activation energy for the viscous flow. The energy of activation ( $E_\eta$ ) for the viscous flow was evaluated from the plot of  $\log \eta$  versus  $1/T$ , which is a linear and the slope gives the value of energy of activation ( $E_\eta$ ). The representative plot of  $\log \eta$  versus  $1/T$  is shown in Fig 3.



**Fig 3. Plot of  $\log \eta$  versus  $1/T$  for lanthanum chloride at concentration  $7.0 \times 10^{-2}$  (mol dm<sup>-3</sup>) in absolute, 90% (v/v), 80% (v/v) and 70% (v/v) aqueous ethanol mixture.**

The results for the energy of activation ( $E_\eta$ ) show that it increases with increase in concentration of lanthanum chloride in absolute

ethanol and going from higher to lower concentration of ethanol the values the energy of activation decreased with increase in concentration of lanthanum chloride. This is due to the fact that configuration changes by means of shearing, interatomic and intermolecular forces resulting lanthanum chloride as structure maker in high ethanol content and structure breaker in less ethanol content due to the thermodynamic hydration of ions in more aqueous content. Free energy change of activation ( $\Delta G^*$ ) is given by the expression:

$$\Delta G^* = RT \ln (\eta V / h N_A) \quad (5)$$

where  $h$  is Planck's constant,  $N_A$  is Avogadro's number,  $V$  is the volume of one mole of solution particles. The entropy change of activation ( $\Delta S^*$ ) is expressed as:

$$\Delta S^* = E_h - \Delta G^* / T \quad (6)$$

Results tabulated in Table 5, indicate that the values of free energy change of activation ( $\Delta G^*$ ) and entropy change of activation ( $\Delta S^*$ ) increase with increase in temperature and concentration of lanthanum chloride. The increased values of entropy change of activation ( $\Delta S^*$ ) with the decrease in concentration of ethanol show an increase in disorderness of binary solvent system. It also proves lanthanum chloride behaves as a weak complex in aqueous ethanol while high value of entropy change of activation ( $\Delta S^*$ ) absolute ethanol indicate structure making phenomenon.

## Conclusion

The ionic interaction of lanthanum chloride in absolute and aqueous ethanol system was analyzed by viscosity and apparent molar volume data. It was concluded that lanthanum chloride behaves as structure maker in absolute ethanol and 90 % (v/v) aqueous ethanol while in 80 % and 70 % (v/v) aqueous ethanol it behaves as structure breaker. In aqueous ethanol as the concentration of ethanol decreases the ions are readily hydrolyzed in water.



The tendency of hydrolysis increases with decrease in the ionic size. In non-aqueous media such as absolute ethanol, salt of weak complex  $MX_6^{-3}$  was prepared (Janardhan and Sivasankar, 1978).

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