

## Viscometric and Thermodynamic Studies on Strong Electrolytes-Metal Chlorides and Metal Sulphates in Aqueous Medium at Different Temperatures

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### Abstract

The viscosity values of strong electrolytes like Magnesium chloride, Zinc chloride, Strontium chloride, Cadmium chloride, Barium chloride, Magnesium sulphate and Zinc sulphate have been measured in aqueous solutions at different temperatures ranging from 298.15K to 313.15K at 5K intervals. The viscosity data have been analyzed on the basis of Jones-Dole equation and transition state theory to obtain Falken-Hagen co-efficient ( $A_F$ ), Jones-Dole Co-efficient ( $B_J$ ), temperature co-efficient ( $dB_J/dT$ ), free energy of activation per mole of solvent ( $\Delta\mu_1^{0*}$ ), and solute ( $\Delta\mu_2^{0*}$ ), enthalpy ( $\Delta H^*$ ) and entropy of activation ( $\Delta S^*$ ) of viscous flow. The variation of these parameters has been discussed in the light of solute-solute and solute-solvent interactions. The structural effect is inferred from the sign of temperature co-efficient.

**Keywords:** Strong electrolytes, temperature co-efficient, Jones-Dole co-efficient, enthalpy

### Introduction

Hydrodynamic behaviour of electrolytic solutions has been the focus of intense research activities for the past few years [1-2]. The viscosity is one of the important hydrodynamic properties of liquid and the viscous nature of liquid is due to the shearing effect which arises by the movement of liquid layers over each other. The viscosity and its derived parameters help study the structural change and intermolecular forces of the electrolytic solutions at different concentrations and temperatures. Viscometric studies of electrolytic solution provide important information regarding solute-solute, solute-solvent and solvent-solvent interactions and help in characterizing the structural properties of solution. Various types of interactions help for better understanding about the nature of solute (electrolyte) and solvent, whether the solute modifies or distorts the structure of the solvent. Also it can be used for the development of molecular models for describing the thermodynamic properties of the electrolytic solutions. So the investigation of thermodynamic properties in aqueous electrolytic solutions has been the area of interest of many researchers [3-5].

The present work aims at determining the values of viscosity of metal chlorides ( $MgCl_2$ ,  $ZnCl_2$ ,  $SrCl_2$ ,  $CdCl_2$  and  $BaCl_2$ ) and metal sulphates ( $MgSO_4$  and  $ZnSO_4$ ) in aqueous medium at different temperatures ranging from 298.15K to 313.15K at an interval of 5K and related parameters such as viscosity co-efficient ( $A_F$  and  $B_J$ ), temperature co-efficient ( $dB_J/dT$ ), the free energy of activation for viscous flow of solvent ( $\Delta\mu_1^{0*}$ ), and solute ( $\Delta\mu_2^{0*}$ ), enthalpy ( $\Delta H^*$ ) and entropy of activation ( $\Delta S^*$ ) have

been calculated to study the structural change and possible ion-ion and ion-solvent interactions. Viscosity measurements of these aqueous electrolytic solutions play the vital role in pharmaceutical, medicinal, industrial, agricultural, environmental and technological applications.

## Materials and Methods

All electrolytes like metal chlorides (MgCl<sub>2</sub>, ZnCl<sub>2</sub>, SrCl<sub>2</sub>, CdCl<sub>2</sub> and BaCl<sub>2</sub>) and metal sulphates (MgSO<sub>4</sub> and ZnSO<sub>4</sub>) used were of GR or AR grades and dried over anhydrous CaCl<sub>2</sub> in desiccator before use. All solutions were prepared in conductivity water (Sp. cond. ~10<sup>-6</sup> S.cm<sup>-1</sup>). The solutions were prepared on the molal basis and conversion of molality to molarity was done by using standard expression [6] using the density data at the corresponding temperatures. The electrolyte content in the solutions varied over a concentration range of 6.0x10<sup>-3</sup>M to 8.0x10<sup>-2</sup>M for viscosity measurements. Viscosity measurements were made using an Ostwald's viscometer in a water thermostat whose temperature was controlled to ±0.05K and efflux time was determined using a digital stop clock with an accuracy of ±0.01s. An average of three sets of flow of times for each solution was taken for the calculation of viscosity. The values of viscosity so obtained were accurate to within ±0.3x10<sup>-3</sup>cP.

## Theoretical Aspects

### Viscosity coefficients (A<sub>F</sub> and B<sub>J</sub>):

The empirical equation of Jones-Dole [7] relates the relative viscosity of an electrolytic solution to the concentration (c) of the electrolyte as follows:

$$\eta_r = \frac{\eta}{\eta_0} = 1 + A_F c^{\frac{1}{2}} + B_J c \quad \dots (1)$$

$$\Rightarrow \left( \frac{\eta_r - 1}{c^{\frac{1}{2}}} \right) = A_F + B_J c^{\frac{1}{2}}$$

where  $\eta_r$  is the relative viscosity,  $\eta$  and  $\eta_0$  are the viscosity of the solution and solvent respectively. A<sub>F</sub> is the Falken-Hagen co-efficient characterizes the ionic interaction and B<sub>J</sub> is the Jones-Dole co-efficient depends on the size of the solute and the nature of solute-solvent interactions [8]. The values of A<sub>F</sub> and B<sub>J</sub> have been determined from the intercept and slope of linear plots of  $(\eta_r - 1)/c^{1/2}$  versus  $c^{1/2}$ , respectively.

The temperature coefficient (dB<sub>J</sub>/dT) values were calculated from the slope of the curve obtained with B<sub>J</sub>-coefficient values against temperatures[9]. The sign of (dB<sub>J</sub>/dT) value provides information regarding the structural change ability of solute in solvent system.

### Transition State theory:

The viscosity data have also been analyzed on the basis of transition state theory for the relative viscosity of aqueous electrolytic solutions as suggested by Feakins et al [10] using the relations

$$\Delta\mu_1^{0*} = 2.303RT \log(\eta_0 \bar{V}_1^0 / hN) \quad \dots (2)$$

$$\Delta\mu_2^{0*} = \Delta\mu_1^{0*} + \left( RT/\bar{V}_1^0 \right) \left[ 1000B_J - (\bar{V}_1^0 - \bar{V}_2^0) \right] \quad \dots (3)$$

where  $\Delta\mu_1^{0*}$  is the free energy of activation per mole for viscous flow of solvent and  $\Delta\mu_2^{0*}$  is the free energy of activation per mole for viscous flow of solute. R, h and N are universal gas constant, Plank's constant and Avogadro's number, respectively. T is the temperature in Kelvin.

$\bar{V}_1^0$  is the partial molar volume of the solvent and was calculated by the relation

$$\bar{V}_1 = \frac{M_{\text{solvent}}}{d} \quad \dots (4)$$

$\bar{V}_1^0$  is the partial molar volume of the solute which is equal to the limiting apparent molar volume at infinite dilution

i.e.,  $\bar{V}_1^0 = \bar{V}_1^0$  .... (5)

The free energy of activation of viscous flow of solutions ( $\Delta\mu^{0*}$ ) was calculated by using the following equation [11]

$$\Delta\mu^{0*} = n_1\Delta\mu_1^{0*} + n_2\Delta\mu_2^{0*} \quad \dots (6)$$

where  $n_1$  and  $n_2$  are the number of moles of solvent and solute, respectively.

The enthalpy ( $\Delta H^*$ ) and entropy ( $\Delta S^*$ ) of activation of viscous flow were computed using the equation

$$\Delta\mu^{0*} = \Delta H^* - T\Delta S^* \quad \dots (7)$$

The values of  $\Delta H^*$  and  $\Delta S^*$  were obtained from the intercepts and slopes, respectively, of the plots of  $\Delta\mu^{0*} \sim T$ .

The above parameters contribute to the structural information regarding solute and solute-solvent interactions.

## Results and discussion

The experimental values of viscosity ( $\eta$ ) for strong electrolytes like metal chlorides ( $\text{MgCl}_2, \text{ZnCl}_2, \text{SrCl}_2, \text{CdCl}_2$ , and  $\text{BaCl}_2$ ) and metal sulphates ( $\text{MgSO}_4$  and  $\text{ZnSO}_4$ ) in aqueous solutions have been determined at 298.15, 303.15, 308.15 and 313.15K and values of viscosity and relative viscosity ( $\eta_r$ ) of above electrolytes at different temperatures are presented in Table 1.

**Table 1. Values of  $\eta$  (Pa.s) and  $\eta_r$  for aqueous electrolyte solutions at different temperatures.**

Electrolytes	cx10 <sup>-3</sup> (mol.m <sup>-3</sup> )	$\eta \times 10^3$				$\eta_r$			
		Temperature(K)							
		298.15	303.15	308.15	313.15	298.15	303.15	308.15	313.15
MgCl <sub>2</sub>	0.006	0.889	0.775	0.705	0.631	0.997	0.971	0.979	0.966
	0.008	0.890	0.796	0.717	0.641	0.998	0.997	0.996	0.982
	0.009	0.892	0.808	0.721	0.647	1.001	1.013	1.001	0.991
	0.010	0.900	0.812	0.731	0.652	1.010	1.018	1.015	0.998
	0.030	0.917	0.818	0.747	0.665	1.029	1.025	1.038	1.018
	0.050	0.927	0.832	0.757	0.674	1.040	1.043	1.051	1.032
	0.080	0.938	0.841	0.762	0.679	1.053	1.054	1.058	1.040
ZnCl <sub>2</sub>	0.006	0.894	0.801	0.723	0.657	1.003	1.004	1.004	1.006
	0.008	0.897	0.802	0.726	0.658	1.007	1.005	1.008	1.008
	0.009	0.898	0.804	0.728	0.658	1.008	1.008	1.011	1.008
	0.010	0.900	0.808	0.730	0.659	1.010	1.013	1.014	1.009
	0.030	0.905	0.812	0.733	0.662	1.016	1.018	1.018	1.014
	0.050	0.921	0.827	0.747	0.671	1.034	1.036	1.038	1.028
	0.080	0.933	0.841	0.753	0.677	1.047	1.054	1.046	1.037

SrCl <sub>2</sub>	0.006	0.919	0.812	0.739	0.661	1.031	1.018	1.026	1.012
	0.008	0.921	0.814	0.741	0.662	1.034	1.020	1.029	1.014
	0.009	0.922	0.815	0.742	0.663	1.035	1.021	1.031	1.015
	0.010	0.923	0.816	0.743	0.664	1.036	1.023	1.032	1.017
	0.030	0.938	0.836	0.749	0.675	1.053	1.048	1.040	1.034
	0.050	0.947	0.841	0.758	0.680	1.063	1.054	1.053	1.041
	0.080	0.959	0.852	0.772	0.694	1.076	1.068	1.072	1.063
CdCl <sub>2</sub>	0.006	0.894	0.805	0.722	0.649	1.003	1.009	1.003	0.994
	0.008	0.896	0.805	0.724	0.651	1.005	1.009	1.006	0.996
	0.009	0.896	0.807	0.724	0.651	1.006	1.011	1.006	0.997
	0.010	0.897	0.809	0.726	0.653	1.007	1.014	1.008	1.000
	0.030	0.904	0.817	0.728	0.655	1.015	1.024	1.011	1.003
	0.050	0.907	0.819	0.732	0.657	1.018	1.026	1.017	1.006
	0.080	0.913	0.824	0.740	0.667	1.025	1.033	1.028	1.021
BaCl <sub>2</sub>	0.006	0.893	0.794	0.721	0.641	1.002	0.995	1.001	0.982
	0.008	0.894	0.796	0.722	0.642	1.003	0.997	1.003	0.983
	0.009	0.896	0.797	0.723	0.644	1.006	0.999	1.004	0.986
	0.010	0.897	0.798	0.725	0.646	1.007	1.000	1.007	0.989
	0.030	0.902	0.802	0.731	0.654	1.012	1.005	1.015	1.002
	0.050	0.912	0.815	0.738	0.658	1.024	1.021	1.025	1.008
	0.080	0.921	0.826	0.746	0.669	1.034	1.035	1.036	1.025
MgSO <sub>4</sub>	0.006	0.913	0.802	0.735	0.651	1.025	1.028	1.021	0.997
	0.008	0.916	0.822	0.740	0.652	1.028	1.030	1.028	0.998
	0.009	0.923	0.826	0.743	0.664	1.036	1.035	1.032	1.017
	0.010	0.925	0.831	0.746	0.666	1.038	1.041	1.036	1.020
	0.030	0.928	0.834	0.750	0.669	1.042	1.045	1.042	1.025
	0.050	0.934	0.838	0.755	0.673	1.048	1.050	1.049	1.031
	0.080	0.948	0.847	0.758	0.68	1.064	1.061	1.053	1.041
ZnSO <sub>4</sub>	0.006	0.919	0.822	0.736	0.657	1.031	1.030	1.022	1.006
	0.008	0.923	0.824	0.743	0.660	1.036	1.033	1.032	1.011
	0.009	0.926	0.829	0.745	0.669	1.039	1.039	1.035	1.025
	0.010	0.928	0.835	0.749	0.672	1.042	1.046	1.040	1.029
	0.030	0.947	0.843	0.755	0.677	1.063	1.056	1.049	1.037
	0.050	0.951	0.847	0.767	0.684	1.067	1.061	1.065	1.047
	0.080	0.963	0.863	0.771	0.693	1.081	1.081	1.071	1.061

Viscosity is one of the most important physical properties of liquid which throws light for better understanding of the molecular interaction as well as the structural change occurred in the solutions containing electrolyte [12]. It is observed from table 1 that the values of viscosity ( $\eta$ ) increases with increase in concentration and decreases with rise of temperature for all the electrolyte solutions of metal chlorides and metal sulphates. This increasing trend of  $\eta$  indicates the existence of molecular interaction occurring in the electrolyte solutions [13] but the decreasing values of  $\eta$  with temperature may be due to more thermal agitation and reduction of attractive forces between the ions [14] which might be attributed

to the decrease in solvation of ions by water. The typical plots of  $(\eta_r - 1)/c^{1/2}$  versus  $c^{1/2}$  of metal chlorides and metal sulphates at 298.15K are shown in Figure 1 (a)-(b).

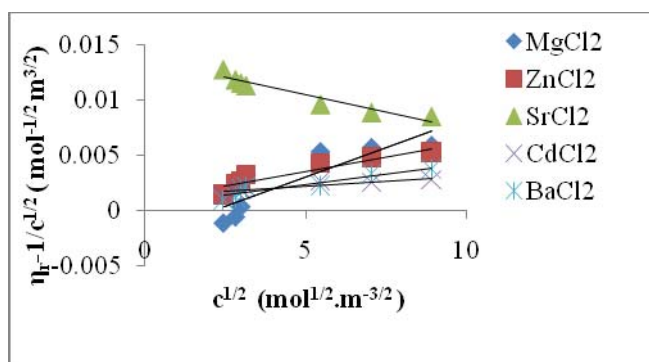


Figure 1(a).Plot of  $(\eta_r - 1)/c^{1/2}$  vs  $c^{1/2}$  in aqueous metal chlorides at 298.15K.

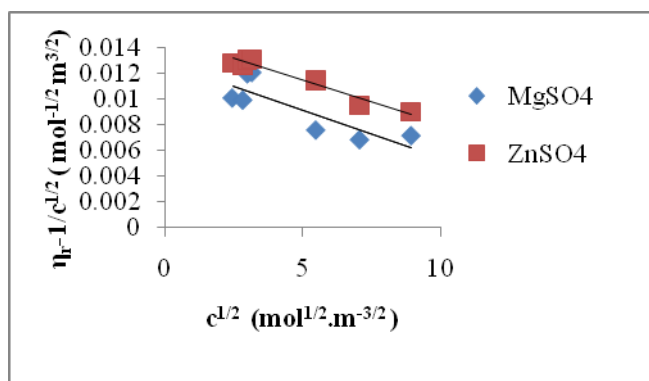


Figure 1(b).Plot of  $(\eta_r - 1)/c^{1/2}$  vs  $c^{1/2}$  in aqueous metal sulphates at 298.15K.

The viscosity data were analyzed by the help of Jones-Dole Eq. (1) and the values of viscosity coefficients are presented in Table 2. A perusal of Table 2 shows that the values of Falken-Hagen coefficient ( $A_F$ ) are positive for almost all aqueous electrolyte solutions (except in few cases) at different temperatures. Since ( $A_F$ ) represents the contribution from inter-ionic electrostatic forces or solute-solute interaction [15], it shows that there is a strong ion-ion interaction present in the electrolyte solutions.

In order to focus more light on these electrolytes the role of Jones-Dole co-efficient ( $B_j$ ) has also been presented. The co-efficient ( $B_j$ ) is a measure of the effective solvodynamic volume of solvated ions which accounts for the ion-solvent interactions that is the structural effect of the solvent in the solutions [16-17] and it also measures the order or disorder introduced by the solute in the solvent and directly depends on the size, shape and charge of the solute molecule.

It is a fact that, when an electrolyte dissolves in a solvent, some of the solvent molecules are surround by the ions (generated from the electrolytes) because of ion-solvent interactions and this causes an increase in viscosity of the electrolyte solutions (a positive contributions to  $B_j$ ).On the other hand, these solvent molecules have to be wrenched out of the bulk solvent and this breaking of the solvent structure causes a decrease in viscosity of the electrolyte solutions (a negative contribution to  $B_j$ ).Therefore, the values of  $B_j$  in the electrolyte solutions is the resultant of these two opposite factors .

As observed, the values of  $B_J$  co-efficient for all the electrolytes are positive at all temperatures excepting few cases indicating the existence of ion-solvent interaction.

It is observed that, in most of the electrolytes the values of  $B_J$  are smaller as compared to  $A_F$  values supporting the behaviour of  $V_\phi^0$  and  $S_V$ , respectively studied earlier[18]. The temperature co-efficient ( $dB_J/dT$ ) values give important information regarding the structural effects (structure making or structure breaking) and solute-solvent interactions of the electrolyte in solvent media is a better criteria [19-24] than Jones-Dole co-efficient( $B_J$ ). The viscosity study of different electrolytes has shown that the positive and negative sign of  $dB_J/dT$  indicates structure breaker and structure maker, respectively reported by number of workers [25]. The values of  $dB_J/dT$  is found to be less positive in almost all electrolytes except in few cases in aqueous solution, the less positive values of temperature co-efficient indicates less stronger ion-solvent interactions as well as less structure breaking tendency of electrolytes in aqueous solutions.

Table 2. Values of parameters  $A_F$  ( $m^{3/2}.mol^{-1/2}$ ),  $B_J$  ( $m^3.mol^{-1}$ ),  $\bar{V}_1^0$  ( $m^3.mol^{-1}$ ),  $\bar{V}_2^0$  ( $m^3.mol^{-1}$ ),  $\Delta\mu_1^{0*}$  ( $kJ mol^{-1}$ ) and  $\Delta\mu_2^{0*}$  ( $kJ mol^{-1}$ ) for aqueous electrolyte solutions at different temperatures.

Electrolytes	Temp./K	$A_F \times 10^3$	$B_J \times 10^3$	$(dB_J/dT) \times 10^5$	$\bar{V}_1^0 \times 10^5$	$\bar{V}_2^0 \times 10^3$	$\Delta\mu_1^{0*}$	$\Delta\mu_2^{0*} \times 10^{-3}$
MgCl <sub>2</sub>	298.15	-2.332	1.068	6.850	1.807	-245.28	9.167	112.9
	303.15	-4.947	1.467		1.809	-336.63	9.046	157.5
	308.15	-5.612	1.677		1.812	-266.15	8.936	199.5
	313.15	-11.70	2.139		1.816	-281.03	8.831	266.5
ZnCl <sub>2</sub>	298.15	0.944	0.524	-2.032	1.807	-313.89	9.167	28.86
	303.15	0.681	0.637		1.809	-331.85	9.046	42.50
	308.15	2.090	0.414		1.812	-492.02	8.936	-11.01
	313.15	1.962	0.260		1.816	-405.74	8.831	-20.92
SrCl <sub>2</sub>	298.15	13.63	-0.628	4.281	1.807	-369.91	9.167	-136.9
	303.15	6.954	0.112		1.809	-341.90	9.046	-32.00
	308.15	11.53	-0.499		1.812	-362.45	8.936	-121.8
	313.15	4.257	0.289		1.816	-306.86	8.831	-2.51
CdCl <sub>2</sub>	298.15	1.355	0.175	2.782	1.807	-450.48	9.167	-37.85
	303.15	3.739	0.013		1.809	-364.74	9.046	-49.05

	308.15	1.373	0.172		1.812	-368.32	8.936	-27.80
	313.15	-2.881	0.585		1.816	-400.59	8.831	26.48
BaCl <sub>2</sub>	298.15	0.377	0.389	5.752	1.807	-282.43	9.167	14.66
	303.15	-3.245	0.827		1.809	-347.43	9.046	66.75
	308.15	-0.105	0.493		1.812	-458.58	8.936	4.86
	313.15	-9.343	1.459		1.816	-418.51	8.831	149.2
MgSO <sub>4</sub>	298.15	12.84	-0.737	7.601	1.807	-297.75	9.167	-141.9
	303.15	13.94	-0.872		1.809	-330.79	9.046	-167.6
	308.15	11.92	-0.689		1.812	-319.67	8.936	-142.5
	313.15	1.156	0.469		1.816	-328.19	8.831	20.23
ZnSO <sub>4</sub>	298.15	14.85	-0.673	5.924	1.807	-400.10	9.167	-147.28
	303.15	14.68	-0.706		1.809	-419.28	9.046	-156.79
	308.15	12.42	-0.493		1.812	-334.78	8.936	-116.96
	313.15	5.134	0.243		1.816	-337.22	8.831	-13.54

The viscosity data were also analysed on the basis of the transition state theory for relative viscosity of all the electrolyte solutions at all range of temperatures using Eq. (3). The calculated values of  $\Delta\mu_1^{0*}$ ,  $\Delta\mu_2^{0*}$ ,  $\bar{V}_1^\ddagger$  and  $\bar{V}_2^\ddagger$  are also presented in Table 2. The values of  $\Delta\mu_2^{0*}$  indicate that its behaviour is quite similar to B<sub>j</sub> in almost all electrolytes and the formation of the transition state is accompanied by the rupture and distortion of the intermolecular forces in solvent structure.

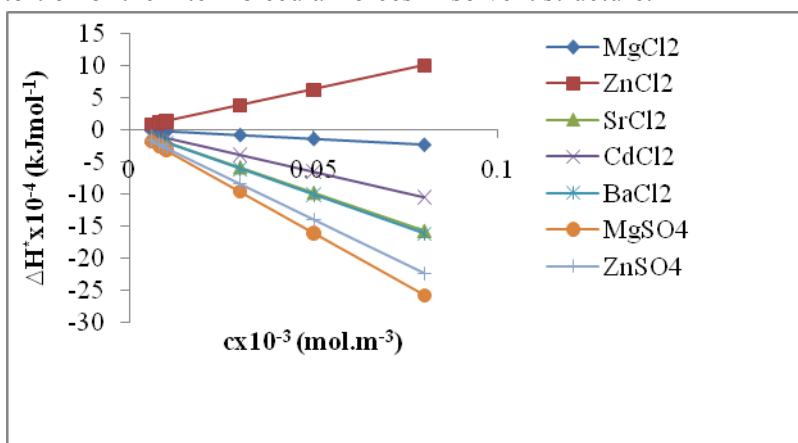


Figure 2. Plot of enthalpy ( $\Delta H^*$ ) vs  $c$  in aqueous metal chlorides and metal sulphates at different temperatures.



Table 3. Values of parameters enthalpy,  $\Delta H^*$  ( $\text{kJmol}^{-1}$ ) and entropy,  $\Delta S^*$  ( $\text{Jmol}^{-1}\text{K}^{-1}$ ) for aqueous electrolyte solutions at different temperatures.

Electrolytes	$\text{cx}10^{-3}$ ( $\text{mol.m}^{-3}$ )	$\Delta H^* \times 10^{-4}$	$\Delta S^* \times 10^{-4}$
MgCl <sub>2</sub>	0.006	-1.641	-5.891
	0.008	-2.219	-7.903
	0.009	-2.508	-8.910
	0.010	-2.797	-9.915
	0.030	-8.575	-30.02
	0.050	-14.35	-50.13
	0.080	-23.02	-80.29
ZnCl <sub>2</sub>	0.006	0.842	2.572
	0.008	1.092	3.383
	0.009	1.217	3.789
	0.010	1.342	4.195
	0.030	3.842	12.31
	0.050	6.343	20.43
	0.080	10.09	32.60
SrCl <sub>2</sub>	0.006	-1.101	-3.621
	0.008	-1.499	-4.874
	0.009	-1.698	-5.501
	0.010	-1.896	-6.127
	0.030	-5.874	-18.66
	0.050	-9.854	-31.20
	0.080	-15.82	-50.01
CdCl <sub>2</sub>	0.006	-0.706	-2.430
	0.008	-0.973	-3.287
	0.009	-1.106	-3.715
	0.010	-1.239	-4.144
	0.030	-3.902	-12.71
	0.050	-6.566	-21.28
	0.080	-10.56	-34.14
BaCl <sub>2</sub>	0.006	-1.127	-3.965
	0.008	-1.533	-5.332
	0.009	-1.736	-6.015
	0.010	-1.939	-6.699
	0.030	-6.000	-20.37
	0.050	-10.06	-34.04
	0.080	-16.15	-54.55
MgSO <sub>4</sub>	0.006	-1.848	-5.998
	0.008	-2.495	-8.043
	0.009	-2.819	-9.067
	0.010	-3.142	-10.09
	0.030	-9.612	-30.55
	0.050	-16.08	-51.00
	0.080	-25.78	-81.69
ZnSO <sub>4</sub>	0.006	-1.590	-5.152
	0.008	-2.151	-6.915
	0.009	-2.431	-7.797
	0.010	-2.712	-8.681
	0.030	-8.322	-26.32
	0.050	-13.93	-43.96
	0.080	-22.35	-70.43

It is evident from Table 3 and Figure 2 that, the  $\Delta H^*$  values are negative and decrease with increase in concentration of electrolytes which might be due to the formation of the activated species necessary for viscous flow and energy releasing where as the positive values for other electrolytes suggest the energy consuming process[26]. Figure 3 shows the values of  $\Delta S^*$  are negative and decreases with increase in concentration of electrolyte in the solutions. It is a fact that the formation of the activated species required for viscous flow appears easily but the positive values of  $\Delta S^*$  indicate the net order of the system decreasing, i.e., randomness of solute molecules in solvent increases [27]. This indicates the existence of solute-solvent interactions in electrolyte solutions.



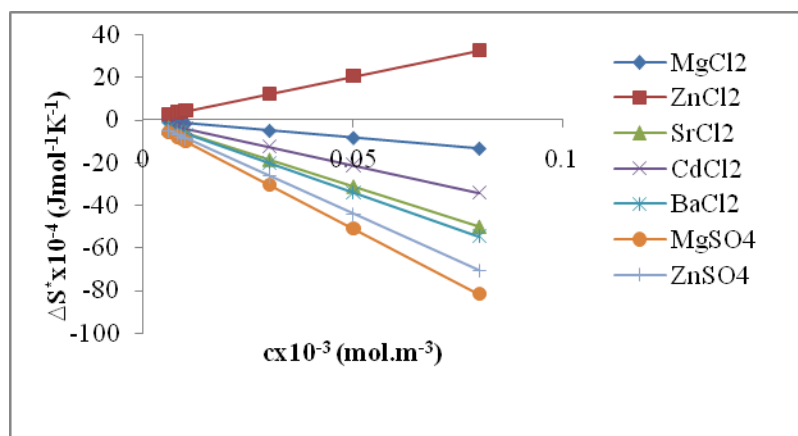


Figure 3. Plot of entropy ( $\Delta S^*$ ) vs  $c$  in aqueous metal chlorides and metal sulphates at different temperatures.

## Conclusion

The results of the present study on strong electrolytes reveal that there exists a molecular interaction and structural change occurred in the solutions in varying degree depending on the nature of the electrolytes and structure of the solvent system. The values of temperature co-efficient concluded the existence of ion-solvent interaction leads to structure making/breaking capacities of the electrolytes. The formation of transition state is accompanied by rupture of the intermolecular interaction.

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