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Effect of Temperature on Viscosity B- Coefficients of Some Transition Metal Chlorides and Magnesium Chloride in Water and In Water + Methanol Mixtures.

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ABSTRACT

Relative viscosities of some transition metal chloridesviz., manganese chloride, cobalt chloride, nickel chloride, copper chloride, cadmium chloride and magnesium chloride have been determined in water and water + methanol solvents respectively at different compositions (5%,10%,15%,20% and 35% w/w of methanol) at 303.15K . Effect of temperature was also analysed in 5% of methanol at five different temperatures (298.15 , 303.15 ,308.15 ,313.15 and 318.15 K).The measured viscosity data has been analysed by Jones – Dole equation and viscosity B-coefficients are determined. Results have also been interpreted in terms of ion –ion and ion – solvent interactions. Activation parameters are also determined which are helpful in depicting the mechanism of viscous flow. In the present study, all these transition metal chlorides and magnesium chloride act as structure promoters in water and in water + methanol solvent systems.

Keywords: Relative Viscosity, B - Coefficient, methanol + water, structure promoters.

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INTRODUCTION

The viscosity of a liquid is a measure of its resistance to flow. Since the flow takes place by displacement of the equilibrium position of the molecules, so studies of viscosity can lead to information about these displacements. It has been found by a number of workers that addition of electrolytes either breaks or makes the structure of the liquid. The viscosity of electrolyte solutions is usually studied to obtain information of ion – solvent interactions [1-4].In the present study, viscosity of some transition metal chlorides is determined in water and aqueous mixtures of methanol + water and very less attention has been paid for these electrolytes in the solvents taken[5- 14]. Also, in the present study, the measured viscosity data of transition metal chlorides are also used to obtain the B- coefficients. This data is expected to highlight the role of transition metal chlorides in the presence of aqueous solution of methanol and its influence with temperature.

These studies are helpful in geology, oceanography, boiler engineering and in oil recovery systems. These fields require aqueous data over wide range of temperature, pressure and composition. Due to the importance in geology, industrial and biological systems, a need has developed for transport data for transition metal salts in aqueous solutions. In biological systems metals like copper, iron and manganese are required for various purposes, while other metals like nickel and cadmium can poison enzymes by substituting for their required metals. Transition metals are also important in geology because they are components of various minerals, while engineers are interested in their role in corrosion processes.

Alcohols have wide applications in industry and science as reagents or solvents. In organic synthesis, alcohols serve as versatile intermediates.

MATERIALS AND METHODS

Manganese chloride, cobalt chloride, nickel chloride, copper chloride, cadmium chloride and magnesium chloride all were of Analytical grade and used as it is after drying them over calcium chloride desiccators. Fresh double distilled water was used for the preparation of binary mixtures of methanol with water and as a standard solvent. The binary aqueous mixtures of methanol as well as the solutions of transition metal chlorides of concentration 0.005m to 0.1m has been prepared by weight and the conversion of molal solutions (m) into molar solution (C) were made by using the following expression

$$C = \frac{1000\, d\, m}{1000 + m M_2} \qquad (1)$$

Where d and M_2 is the density and molecular weight of the each transition metal chloride. The densities of solutions of transition metal chlorides and magnesium chloride were measured with the help of an apparatus as described by the Ward and Millero [15].The accuracy in the measurement of solutions densities was found to be 1 x 10^{-4} gcm^{-3}.The relative viscosities of the solutions of transition metal chlorides and magnesium chloride were determined with the help of Ostwald viscometer with the flow time 119 seconds at 298.15 K. Measurements were repeated until three successive determinations were obtained with in ±0.1 seconds. No kinetic energy correction required, as the flow time was greater than 100 seconds. The relative viscosities of the solutions were determined by usual procedure [16-17].The density and viscosity studies were carried out in water thermostat (± 0.01K).

RESULTS AND DISCUSSION

The relative viscosities and densities of the solutions of transition metal chlorides namely manganese chloride, cobalt chloride, nickel chloride, copper chloride, cadmium chloride and magnesium chloride in water and in methanol + water (5%, 10%, 15%, 20% and 35% w/w) were determined at 303.15K.The relative viscosity data has been analysed with Jones Dole equation [18].

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

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Where, η and η_0 are the viscosities of solution and solvent respectively. A and B are two constants parameters and c is the molar concentration. Parameter A corresponds to ion- ion interactions and parameter B corresponds to ion – solvent interactions. These parameters A and B have been obtained by plotting a graph between $(\eta_r - 1)/\sqrt{C}$ verses √C for all the transitions metal chlorides and magnesium chloride in water as well as in all the prepared compositions of methanol + water at 303.15 K.All these obtained values of A and B parameters were recorded in Table 1 and a sample plot of $(\eta_r - 1)/\sqrt{C}$ verses √C shown in fig1 for $NiCl_2$ in different compositions of methanol + water at 303.15K.

Table 1: Values of A and B parameters of Jones – Dole equation for some transition metal chlorides and magnesium chloride in water and in different compositions of methanol + water at 303.15 K

Methanol + Water (5% w/w)	A ($dm^{3/2}$ $mol^{-1/2}$)	B (dm^3 mol^{-1})
Manganese chloride		
0	4.41	0.030
5	15.34	0.090
10	15.71	0.080
15	15.94	0.081
20	16.18	0.079
35	16.43	0.075
Cobalt chloride		
0 (Water)	3.08	0.029
5	17.64	0.076
10	17.73	0.087
15	17.86	0.085
20	18.16	0.075
35	18.46	0.085
Nickel Chloride		
0 (Water)	2.81	0.026
5	17.85	0.067
10	18.14	0.064
15	18.25	0.062
20	18.39	0.061
35	18.55	0.057
Copper chloride		
0 (Water)	4.40	0.036
5	12.82	0.061
10	13.15	0.053
15	13.58	0.044
20	14.03	0.032
35	14.29	0.026
Cadmium chloride		
0 (Water)	3.73	0.027
5	20.39	0.053
10	20.72	0.044
15	20.87	0.041
20	21.19	0.035
35	21.39	0.030
Magnesium chloride		
0 (Water)	3.75	0.020
5	17.78	0.053
10	18.01	0.050
15	18.29	0.045
20	18.72	0.033
35	19.10	0.024

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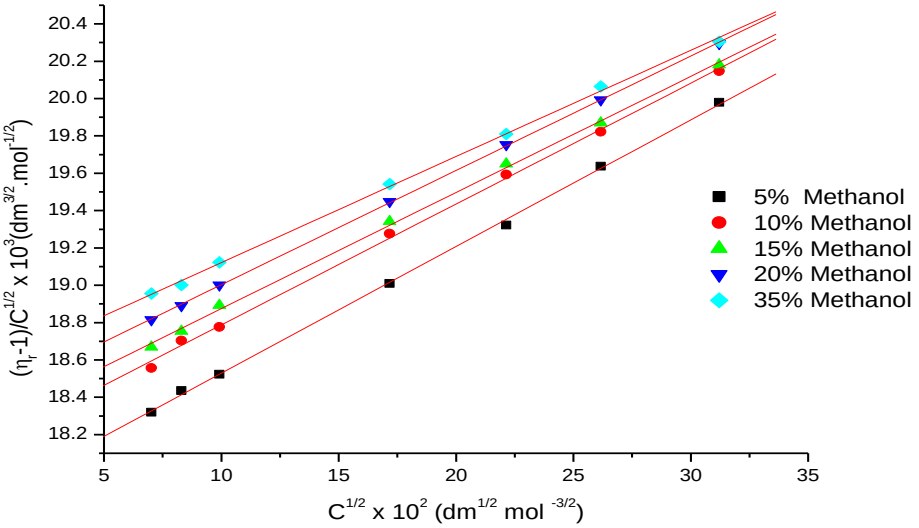

Figure 1: Plot of $(\eta_r-1)/\sqrt{C}$ verses $\sqrt{C}$ for $NiCl_2$ in 5%, 10%, 15%, 20% and 35% w/w compositions of methanol + water.

It is clear from, Table 1 that both parameters A and B have positive values for the selected transition metal chlorides and magnesium chloride in water and in all five compositions (5% - 35%) of methanol + water at 303.15K.The values of A are positive and magnitude of A increases with the increase in the content of methanol in water. This indicates the presence of significant ion- ion interactions and these interactions becomes more and more with the increase in the content of methanol in water. This may be trait to decrease in the solvation of ions of transition metals and indicates that methanol have more affinity for water than those of transition metal chlorides. Also from table 1 it is clear that values of B-Coefficients are positive for all the transition metal chlorides taken here for study in water and in binary aqueous mixtures of methanol. Also these values are decreasing with the addition of methanol content in water resulting in decrease in ion-solvent interactions indicating that water has more affinity for solvent than that for electrolyte. The viscosity data has also been analysed on the basis of transition state treatment of relative viscosity as proposed by Feakins [19].The parameter B in terms of transition state theory is given by the following relation:

$$B = \frac{\overline{V_1^0} - \overline{V_2^0}}{1000} + \frac{\overline{V_1^0}}{1000}\left[\frac{\Delta\mu_2^0 - \Delta\mu_1^0}{RT}\right] \qquad (3)$$

Where,$\overline{V_1^0}$is mean volume of the solvent$\overline{V_2^0}$ is the partial molar volume of the transition metal chlorides, $\Delta\mu_1^0$ and $\Delta\mu_2^0$ are the free energies of activations per mole of pure solvent and transition metal chlorides respectively and given by the expressions:

$$\Delta\mu_1^0 = RTIn\left(\eta_0\overline{V_1^0}/hN\right) \qquad (4)$$

$$\Delta\mu_2^0 = \Delta\mu_1^0 + RT/\overline{V_1^0}\left[1000B - \left(\overline{V_1^0} - \overline{V_2^0}\right)\right] \qquad (5)$$

Where R is the gas constant is the absolute temperatureη_0 is the viscosity of the solvent, h is the Planck's constant and N is the Avogadro number.

The values of $\Delta\mu_1^0$are calculated with the help of relation 4 and are given in Table 2.

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Table 2: Values of $\overline{V_1^0}$, $\overline{V_2^0}$, $\Delta\mu_1^0$ and $\Delta\mu_2^0$ for some transition metal chlorides and magnesium chloride in different compositions of methanol + water at 303.15K

Methanol+ water % compositions(w/w)	5%	10%	15%	20%	35%
$\overline{V_1^0}$ ($dm^3\ mol^{-1}$)	18.68	19.18	19.79	20.33	22.40
$\Delta\mu_1^0$ (kJ mol^{-1})	61.36	61.54	61.81	61.94	62.52
Manganese chloride					
$\overline{V_2^0}$ ($dm^3\ mol^{-1}$)	61.50	59.91	56.22	49.07	45.79
$\Delta\mu_2^0$ (kJ mol^{-1})	79.99	78.40	76.76	75.29	73.62
Cobalt chloride					
$\overline{V_2^0}$ ($dm^3\ mol^{-1}$)	127.37	123.76	118.85	116.29	112.87
$\Delta\mu_2^0$ (kJ mol^{-1})	86.39	86.78	85.32	83.16	82.32
Nickel chloride					
$\overline{V_2^0}$ ($dm^3\ mol^{-1}$)	98.79	96.05	91.75	89.19	85.49
$\Delta\mu_2^0$ (kJ mol^{-1})	81.31	80.15	78.94	78.12	76.13
Copper chloride					
$\overline{V_2^0}$ ($dm^3\ mol^{-1}$)	41.01	35.98	34.72	29.49	29.16
$\Delta\mu_2^0$ (kJ mol^{-1})	72.69	70.72	69.37	67.15	66.21
Cadmium chloride					
$\overline{V_2^0}$ ($dm^3\ mol^{-1}$)	68.02	62.03	57.72	53.05	52.06
$\Delta\mu_2^0$ (kJ mol^{-1})	75.28	72.96	71.86	70.42	69.33
Magnesium chloride					
$\overline{V_2^0}$ ($dm^3\ mol^{-1}$)	144.21	142.81	139.03	135.92	133.15
$\Delta\mu_2^0$ (kJ mol^{-1})	85.48	84.37	82.77	80.40	77.72

For mixed solvents, each solvent mixture was treated as pure and the molar volume taken as a mean volume given as:

$$\overline{V_1^0} = (x_1M_1 + x_2M_2)/d_1 \qquad (6)$$

Where x_1, M_1 and x_2M_2 are the mole fractions and molecular weights of the water and methanol respectively and d_1 is the density of methanol + water as solvent. The values of $\overline{V_2^0}$ for the selected transition metal chlorides and magnesium chlorides calculated with the help of the viscosity data and are recorded in Table 2.The values of $\Delta\mu_2^0$ and $\overline{V_1^0}$ calculated with the help of relations 5 and 6 are also recorded in Table 2.The $\Delta\mu_1^0$ and $\overline{V_1^0}$ values are constant in all solvent compositions.This indicates that $\Delta\mu_1^0$ is dependent on B-coefficient and $(\overline{V_1^0} - \overline{V_2^0})$ factors.

It is clear from table 2, that the values of $\Delta\mu_2^0$ are positive and more in magnitude than $\Delta\mu_1^0$ values. This informs that the transition state formation is less favoured in the presence of the selected transition metal chlorides and magnesium chloride. This indicates that the formation of transition state is followed by the breaking and distortions of the intermolecular bonds between methanol and water.

Many workers [20] intensified that instead of parameter B, $\frac{dB}{dT}$ is a better parameter for determine the structure making/breaking nature of any solute. In order to make use of $\frac{dB}{dT}$ parameter, the effect of temperature must be studied.

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Effect Of Temperature

Since the behaviour of the selected transition metal chlorides in different compositions 5%,10%,15%,20% and 35% w/w of methanol + water are found to be linear and identical at 303.15 K so, only 5% w/w methanol + water system has been selected for studying the effect of temperatures 298.15 ,303.15,308.15,313.15 and 318.15 K. The relative viscosity data has been again analysed with Jones Dole equation [18] and the values of A and B coefficients are recorded in table 3.The plot of $(\eta_r - 1)/\sqrt{C}$ verses $\sqrt{C}$ has been found to be linear for all the selected transition metal chlorides at five temperatures (298.15K to 318.15K).A sample plot for nickel chloride in 5% w/w methanol + water system is shown in fig.2 at different temperatures. This is in accordance to the Jones – Doles equation. The plot of $(\eta_r - 1)/\sqrt{C}$ verses $\sqrt{C}$ has been fitted to least square fit method to obtain the data the values of A and B parameters.

Table 3: Values of A and B parameters of Jones-Dole equations for some transition metal chlorides in water and in 5% w/w of methanol + water at 298.15, 303.15, 308.15, 313.15 and 318.15 K.

Temperature (K)	298.15	303.15	308.15	313.15	318.15
Manganese chloride + water					
A ($dm^{3/2}$ $mol^{-1/2}$)	4.34	4.41	4.51	4.60	4.78
B (dm^3 mol^{-1})	0.0348	0.0344	0.0341	0.0334	0.0313
Cobalt chloride + water					
A ($dm^{3/2}$ $mol^{-1/2}$)	2.87	3.08	3.12	3.24	3.33
B (dm^3 mol^{-1})	0.0299	0.0293	0.0277	0.0264	0.0253
Nickel chloride + water					
A ($dm^{3/2}$ $mol^{-1/2}$)	2.68	2.81	2.99	3.05	3.26
B (dm^3 mol^{-1})	0.0271	0.0262	0.0259	0.0251	0.0246
Copper chloride + water					
A ($dm^{3/2}$ $mol^{-1/2}$)	4.33	4.40	4.51	4.67	4.85
B (dm^3 mol^{-1})	0.0379	0.0365	0.0344	0.0322	0.0270
Cadmium chloride + water					
A ($dm^{3/2}$ $mol^{-1/2}$)	3.44	3.73	3.81	2.97	2.84
B (dm^3 mol^{-1})	0.0320	0.0272	0.0248	0.0418	0.0445
Magnesium chloride + water					
A ($dm^{3/2}$ $mol^{-1/2}$)	3.41	3.75	3.80	4.08	4.19
B (dm^3 mol^{-1})	0.0264	0.0204	0.0198	0.0163	0.0156
Manganese chloride + 5% w/w methanol + water					
A ($dm^{3/2}$ $mol^{-1/2}$)	9.23	9.42	9.71	10.02	10.60
B (dm^3 mol^{-1})	0.125	0.121	0.113	0.111	0.103
Cobalt chloride + 5% w/w methanol + water					
A ($dm^{3/2}$ $mol^{-1/2}$)	0.76	1.09	2.42	2.84	3.07
B (dm^3 mol^{-1})	0.233	0.226	0.197	0.196	0.189
Nickel chloride + 5% w/w methanol + water					
A ($dm^{3/2}$ $mol^{-1/2}$)	3.81	4.11	5.11	5.57	5.76
B (dm^3 mol^{-1})	0.143	0.138	0.133	0.126	0.122
Copper chloride + 5% w/w methanol + water					
A ($dm^{3/2}$ $mol^{-1/2}$)	5.34	6.26	6.95	7.20	7.37
B (dm^3 mol^{-1})	0.132	0.123	0.122	0.120	0.119
Cadmium chloride + 5% w/w methanol + water					
A ($dm^{3/2}$ $mol^{-1/2}$)	7.52	8.05	8.97	9.50	10.06
B (dm^3 mol^{-1})	0.112	0.104	0.103	0.094	0.085
Magnesium chloride + 5% w/w methanol + water					
A ($dm^{3/2}$ $mol^{-1/2}$)	5.25	5.59	5.90	6.27	6.88
B (dm^3 mol^{-1})	0.182	0.181	0.180	0.177	0.175

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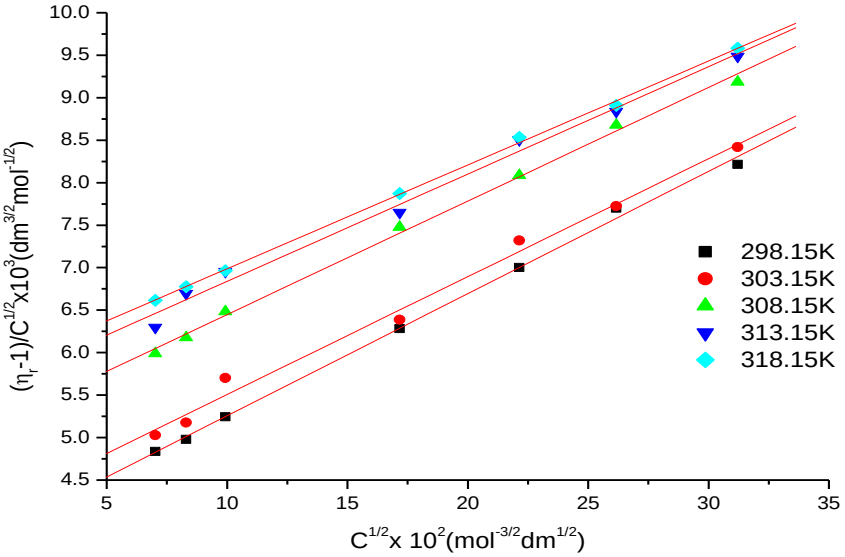

Figure 2: Plot of $(\eta_r-1)/\sqrt{C}$ verses $\sqrt{C}$ for $NiCl_2$ in 5% methanol + water at 298.15K, 303.15K, 308.15K, 313.15K and 318.15K.

It is clear from Table 3,that the values of A parameter are positive in water as well as in 5% w/w methanol + water for all the transition metal chlorides and this parameter A values increases with the rise in temperature. So, it suggests that the presence of ion- ion interactions and these interactions are more strengthen with the rise of temperature. In order words, the presence of strong ion – ion interactions may be attributes to decrease in the salvation of transition metal chlorides as well as of magnesium chloride. From Table 3, the values of parameter B are positive for the selected transition metal chlorides and magnesium chloride in both water as well as in 5% w/w methanol + water at all temperatures. These positive values indicates the presence of strong ion – solvent interactions but these interactions are further weakened with the rise in temperature for the transition metal chlorides and magnesium chloride taken in the present study, as the values of parameter B decreases with the rise of temperature. The value of $\frac{dB}{dT}$ is negative for the transition metal chlorides as well as for the magnesium chloride both in water and in 5% w/w methanol + water. This indicates the structure making nature of transition metal chlorides in water as well in binary aqueous mixtures of methanol + water systems.

The data of viscosity coefficients at 298.15, 303.15, 308.15, 313.15 and 318.15 K have also been interpreted by applying the transition state theory. The values of $\Delta\mu_1^0$ and $\Delta\mu_2^0$ have been recorded in Table 4.

According to Feakins model [20] $\Delta\mu_2^0 > \Delta\mu_1^0$ for the solutes having positive B values, greater is the value of $\Delta\mu_2^0$, more is the structure making ability of the solute.Thereby, it is determined that the Co^{2+} ion is an efficient structure maker and Cd^{2+} ion is least. In the present study, the order of structure making is $Co^{2+} > Mg^{2+} > Ni^{2+} > Mg^{2+} > Cu^{2+} > Cd^{2+}$.

Also, It is clear from Table 4, that the quantity $(\Delta\mu_2^0 - \Delta\mu_1^0)$, change in activation energy per mole of solute on replacing one mole of solvent by one mole of solute at infinite dilution is positive and more for transition metal chlorides and magnesium chloride in 5% w/w methanol + water at different temperatures (298.15 K to 318.15 K) ,thereby it is concluded that the formation of the transition state is less favoured in the presence of these salts in the selected temperature range.

The entropy of activation ΔS_2^0 for transition metal chlorides and magnesium chloride has been calculated from the following equations [19].

$$\frac{d(\Delta\mu_2^0)}{dT} = -\Delta S_2^0 \qquad (7)$$

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Table 4: Values of $\overline{V_1^0}$, $\overline{V_2^0}$, $\Delta\mu_1^0$ and $\Delta\mu_2^0$ for some transition metal chlorides and magnesium chloride in 5% w/w methanol in water at 298.15, 303.15,308.15,313.15 and 318.15 K.

Temperature (K)	298.15	303.15	308.15	313.15	318.15
$\overline{V_1^0}$ ($dm^3 mol^{-1}$)	18.67	18.68	18.75	18.78	18.88
$\Delta\mu_1^0$ ($kJmol^{-1}$)	60.71	61.36	62.03	62.70	63.41
Manganese chloride					
$\overline{V_2^0}$ ($dm^3 mol^{-1}$)	64.89	61.50	57.86	55.80	52.86
$\Delta\mu_2^0$ ($kJ mol^{-1}$)	83.52	83.49	82.91	83.23	82.72
Cobalt chloride					
$\overline{V_2^0}$ ($dm^3 mol^{-1}$)	131.81	127.68	125.55	123.95	120.75
$\Delta\mu_2^0$ ($kJ mol^{-1}$)	106.67	106.63	103.64	104.50	104.18
Nickel chloride					
$\overline{V_2^0}$ ($dm^3 mol^{-1}$)	104.61	96.83	93.87	87.21	84.09
$\Delta\mu_2^0$ ($kJ mol^{-1}$)	91.20	90.61	90.59	89.73	89.76
Copper chloride					
$\overline{V_2^0}$ ($dm^3 mol^{-1}$)	43.16	40.89	37.27	35.53	33.29
$\Delta\mu_2^0$ ($kJ mol^{-1}$)	81.50	81.05	81.26	81.78	82.10
Cadmium chloride					
$\overline{V_2^0}$ ($dm^3 mol^{-1}$)	73.56	68.13	63.37	58.38	53.66
$\Delta\mu_2^0$ ($kJ mol^{-1}$)	82.95	82.15	82.29	81.32	80.21
Magnesium chloride					
$\overline{V_2^0}$ ($dm^3 mol^{-1}$)	151.56	142.52	138.67	135.74	132.50
$\Delta\mu_2^0$ ($kJ mol^{-1}$)	102.56	102.53	103.04	103.46	103.96

Table 5: Entropy $T\Delta S^0$ and enthalpy ΔH_2^0 of activation for viscous flow of some transition metal chlorides and magnesium chloride in 5% w/w of methanol in water at 298.15, 303.15, 308.15, 313.15 and 318.15 K

Temperature (K)	298.15	303.15	308.15	313.15	318.15
Manganese chloride					
$(\Delta\mu_2^{o*} - \Delta\mu_1^{o*})$ ($kJ mol^{-1}$)	22.81	22.13	21.87	20.53	19.31
$T\Delta S^0$ ($kJ mol^{-1}$)	51.31	52.17	53.03	53.89	54.75
ΔH_2^0 ($kJ mol^{-1}$)	134.84	135.67	135.94	137.12	137.47
Cobalt chloride					
$(\Delta\mu_2^{o*} - \Delta\mu_1^{o*})$ ($kJ mol^{-1}$)	45.96	45.27	41.60	41.79	40.77
$T\Delta S^0$ ($kJ mol^{-1}$)	82.61	84.00	85.38	86.77	88.15
ΔH_2^0 ($kJ mol^{-1}$)	189.29	190.64	189.03	191.27	192.34
Nickel chloride					
$(\Delta\mu_2^{o*} - \Delta\mu_1^{o*})$ ($kJ mol^{-1}$)	30.49	29.25	28.55	27.03	26.35
$T\Delta S^0$ ($kJ mol^{-1}$)	62.52	63.57	64.61	65.66	66.71
ΔH_2^0 ($kJ mol^{-1}$)	153.72	154.18	155.20	155.40	156.47
Copper chloride					
$(\Delta\mu_2^{o*} - \Delta\mu_1^{o*})$ ($kJ mol^{-1}$)	20.79	19.69	19.22	19.08	18.69
$T\Delta S^0$ ($kJ mol^{-1}$)	28.74	29.22	29.70	30.18	30.66
ΔH_2^0 ($kJ mol^{-1}$)	110.24	110.27	110.96	111.97	112.76
Cadmium chloride					
$(\Delta\mu_2^{o*} - \Delta\mu_1^{o*})$ ($kJ mol^{-1}$)	22.24	20.79	20.26	18.61	16.81
$T\Delta S^0$ ($kJ mol^{-1}$)	77.75	79.06	80.36	81.66	82.97
ΔH_2^0 ($kJ mol^{-1}$)	160.71	161.21	162.66	162.99	163.19
Magnesium chloride					
$(\Delta\mu_2^{o*} - \Delta\mu_1^{o*})$ ($kJ mol^{-1}$)	41.86	41.17	41.01	40.75	40.56
$T\Delta S^0$ ($kJ mol^{-1}$)	17.91	18.21	18.51	18.82	19.12
ΔH_2^0 ($kJ mol^{-1}$)	120.48	120.75	121.56	122.28	123.08

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The values of ΔS_2^0 have been calculated from the slopes of linear plot of $\Delta\mu_2^0$ verses T. The enthalpy of activation ΔH_2^0 has been calculated with the help of relationship:

$$\Delta H_2^0 = \Delta\mu_2^0 + T\Delta S_2^0 \qquad (8)$$

A sample plot has been shown for nickel chloride is shown in fig 3. The $T\Delta S_2^0$ and ΔH_2^0 values at different temperatures are recorded in Table 5.

It is clear from Table 5 that both parameters enthalpy and entropy of activation are positive for all the transition metal chlorides and magnesium chloride taken in the present study. These positive values of ΔH_2^0 and $T\Delta S_2^0$ parameters indicates that the transition state theory is associated with bond breaking and decrease in order.

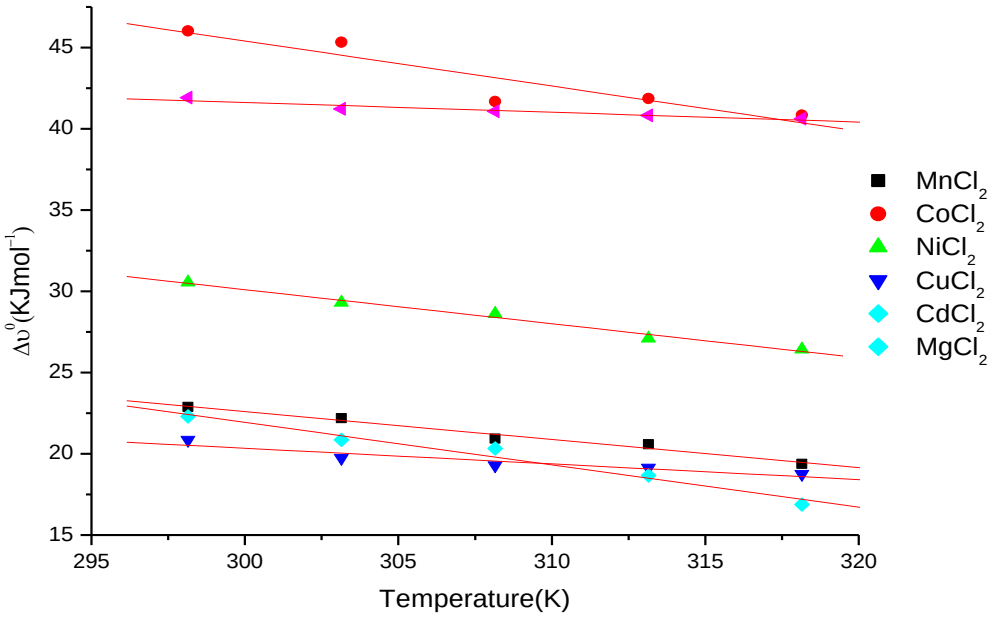

Figure 3: Plot of variation of $\Delta\mu_2^0$ with temperature for $MnCl_2$, $CoCl_2$,$NiCl_2$,$CuCl_2$,$CdCl_2$ and $MgCl_2$ in 5%w/w of methanol+ water.

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