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Research Article

EFFECT OF DIELECTRIC CONSTANT ON THE SOLVATION OF SODIUM CITRATE

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ABSTRACT

The paper reports on the solvation of Sodium citrate in water, water + CH_3CN and water + DMSO mixtures (v/v) under varying dielectric constant at different temperature. The specific conductance data obtained was analyzed by Kraus-Bray and Shedlovsky conductivity models. Limiting molar conductance (λ_m^0), dissociation constant/ association constant (K_C/K_a) were evaluated for all the solvent compositions. The limiting molar conductance, λ_m^0 decreases with the increase in amount of co-solvent in water, due to increased solvent-solvent interaction and decrease in dielectric constant. Due to high viscosity and molecular size of DMSO in water, lower conductance was observed in water + DMSO media. The K_a values increases with the increase in amount co-solvent in water at all the temperatures studied. Energy of activation of the rate process and related thermodynamic parameters such as ΔH_a, ΔG_a and ΔS_a have been evaluated. ΔG_a is found to be negative indicating spontaneity of the process. Walden product ($\lambda^0_m\eta_0$) and corrected Stoke's radius (r_i) have also been evaluated. Fuoss- Accascina equation was used to identify the ion-pairs and ion-triplets in the system. But the slopes of the plot were found to be less than -0.5, indicating the absence of ion-pairs or ion-triplets. Born relation of solvation was verified. These data have been used to study the nature of ion-solvent interaction and solvent-solvent interactions existing in the system.

Keywords: Dielectric constant, Solvation, Limiting molar conductance, Sodium citrate.

INTRODUCTION

Conductance measurement is an important and simplest tool to get information in regard to ion-solvation[1-3]. The study of conductance of an electrolyte system with different solvents of varying viscosity and dielectric constant provides satisfactory information regarding ion-solvation[4-7]. In recent years there has been an increasing interest in the study of behavior of electrolytes in partial and non-aqueous solvents with a view of investigating the role of partial or non-aqueous solvents in the solvation of an ion[8-10].

Tri-sodium citrate (tri-sodium 2-hydroxypropane-1, 2, 3-tricarboxylate) is a sodium salt of citric acid. It is a common ingredient in lemon, lime and citrus soft drinks such as Ting[11], Chinotto and some ocean spray juices. In 1914, the Belgian doctor Albert Hustin and the Argentine physician successfully used sodium citrate as an anticoagulant in blood transfusion[11]. Even today it is used in blood collection tubes and for the preservation of blood in blood banks[11]. The citrate ion chelates calcium ions in the blood by forming calcium citrate complexes, disrupting the blood clotting mechanism[11]. Recently, Oopvik, et.al., showed improvement in running performance of students over 5 km by 30 seconds[12] by the use of this compound. Sodium citrate is used to relieve discomfort in urinary tract infections such as crystitis, to reduce the acidosis seen in distal renal tubular acidosis, and can also be used as an osmotic laxative. A solution of sodium citrate[13] and citric acid at pH 4.0 is effective in dissolving $Ca_3(PO_4)_2$ (Kidney stone) stone in vitro and vivo. The sodium citrate solution containing an acid is effectively reducing the calcium ion concentration. In the literature, no reports available on the solvation of sodium citrate in partial and non-aqueous solvents and hence became the subject of the present study.

MATERIALS AND METHODS

Sodium citrate (Loba chemi, India) was used without further purification [Melting point: 298^{0}C (Observed value), 300^{0}C (Literature value)]. Triply distilled water (specific conductivity of the order of 1×10^{-7} S cm^{-1}) was used throughout the study. The solvents acetonitrile and dimethylsulphoxide were purified as reported[14] (specific conductivity of the order of 2.0 - 3.0 $\times10^{-7}$ Scm^{-1}). The solutions of sodium citrate in water and water + CH_3CN/ DMSO were prepared as and when required.

Conductivity measurements were carried out using a digital direct reading conductivity meter (Model, CM 180, Elico make) and a dip type calibrated[15] conductivity cell of cell constant 0.975 cm^{-1}. All the measurements were made in a thermostat/ cryostat maintained at the desired temperature ± 0.01^{0}C. The instrument was standardized as described earlier[16].

RESULTS AND DISCUSSION

Limiting molar conductance

The solutions of sodium citrate in various compositions of water + CH_3CN and water + DMSO (v/v) were subjected to conductivity measurement at four different temperatures (283, 293, 303 and 313 ± 0.01K). The specific conductance so obtained was used in the determination of molar conductance. The conductance data was initially subjected to analysis by Kraus-Bray equation[17]. Since the chosen electrolyte is of 3:1 type, the Kraus-Bray equation related to that condition was tried but the plot $(\lambda_m C)^3$ Vs $1/\lambda_m$ was found to be non linear (Fig. not shown) and Kraus-Bray equation of 2:1 electrolyte was tried and the plot $(\lambda_m C)^2$ Vs $1/\lambda m$ was found to be non linear (Fig not shown). Hence the Kraus-Bray equation related to 1:1 electrolyte was applied and the equation may be represented as

$$Na_3C_6H_5O_7 \rightleftharpoons C_6H_5O_7^- + 3Na^+$$

$$\frac{1}{\lambda_m} = \frac{1}{\lambda_m^0} + \frac{\lambda_m C}{\lambda_m^{0\,2} K_c} \qquad (1)$$

Where, λ_m^0 is the limiting molar conductance, C is the concentration in molar and K_C is the dissociation constant. On the basis of the above equation a plot was drawn by taking $\lambda_m C$ along X-axis and $1/\lambda_m$ along Y-axis. The plot was found to be linear (Fig.1, as a representative plot). This proved that the sodium citrate behaves like 1:1 electrolyte. From the intercept and slope of the linear plot, the limiting molar conductance (λ_m^0) and dissociation constant (K_C) were obtained and are shown in Tables 1 and 2 respectively. The measurements were done at different temperatures and various compositions of water + CH_3CN and water + DMSO as it gives with a view of getting the effect of dielectric constant on λ_m^0 or K_C/ K_a.

But the present model has certain inherent limitations especially the unaccountability for the effect of ionic mobility and activity co-efficient on conductivity. Hence a better model which could solve the above problems was made use. Shedlovsky model, accounts for the above facts and expected to give absolute limiting molar conductance. The shedlovsky equation[18] may be represented as

$$\frac{1}{S\lambda_m} = \frac{1}{\lambda_m^0} + \frac{C\lambda_m S f_\pm^2 K_a}{\lambda_m^{0\,2}} \qquad (2)$$

Where, λ_m^0 is the limiting molar conductance, C is the concentration, K_a is the association constant, S is the Onsager slope and $f_\pm$ the mean activity co-efficient of the electrolyte. From the intercept and slope of the linear plot $1/S\lambda_m$ Vs $C\lambda_m S f_\pm^2$ (Fig.2, as a representative plot), the limiting molar conductance (λ_m^0) and association constant (K_a) were obtained and are presented respectively in Tables 1 and 2.

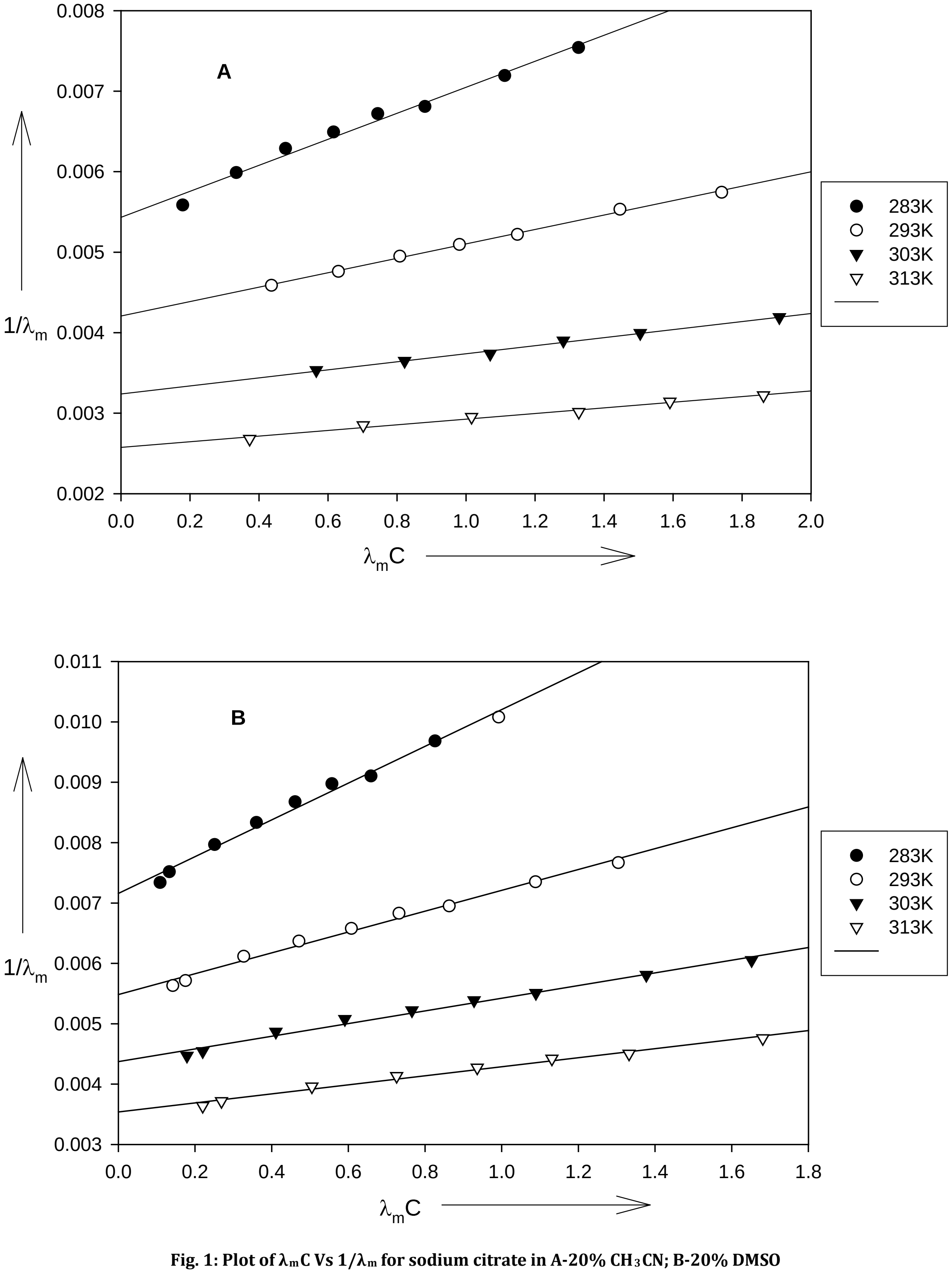


Fig. 1: Plot of $\lambda_m C$ Vs $1/\lambda_m$ for sodium citrate in A-20% CH_3CN; B-20% DMSO

Table 1: Experimental values of molar conductance at infinite dilution (λ^0_m: mho cm^2 mol^{-1}) for sodium citrate from Kraus-Bray (1) and Shedlovsky (2) models in different dielectric constant (ε at 303K) or various compositions (v/v) of water + CH_3CN and water + DMSO

T(K)/ ε	76.7 (0%)		72.6(10%)		68.6(20%)		60.7(40%)		53.0(60%)	
	1	2	1	2	1	2	1	2	1	2
					Water + CH_3CN					
283	218	218	192	196	181	183	165	172	149	156
293	281	279	250	253	235	238	210	212	185	190
303	349	347	322	327	307	307	263	270	232	238
313	423	425	408	408	384	384	357	357	285	289
					Water + DMSO					
T(K)/ ε	76.7 (0%)		76.5(10%)		75.2(20%)		74.0(40%)		70.5 (50%)	
283	218	218	170	174	139	138	77	77	57	59
293	281	279	219	223	178	180	107	109	77	80
303	349	347	274	279	222	227	133	138	102	104
313	423	425	340	342	273	274	169	172	125	130

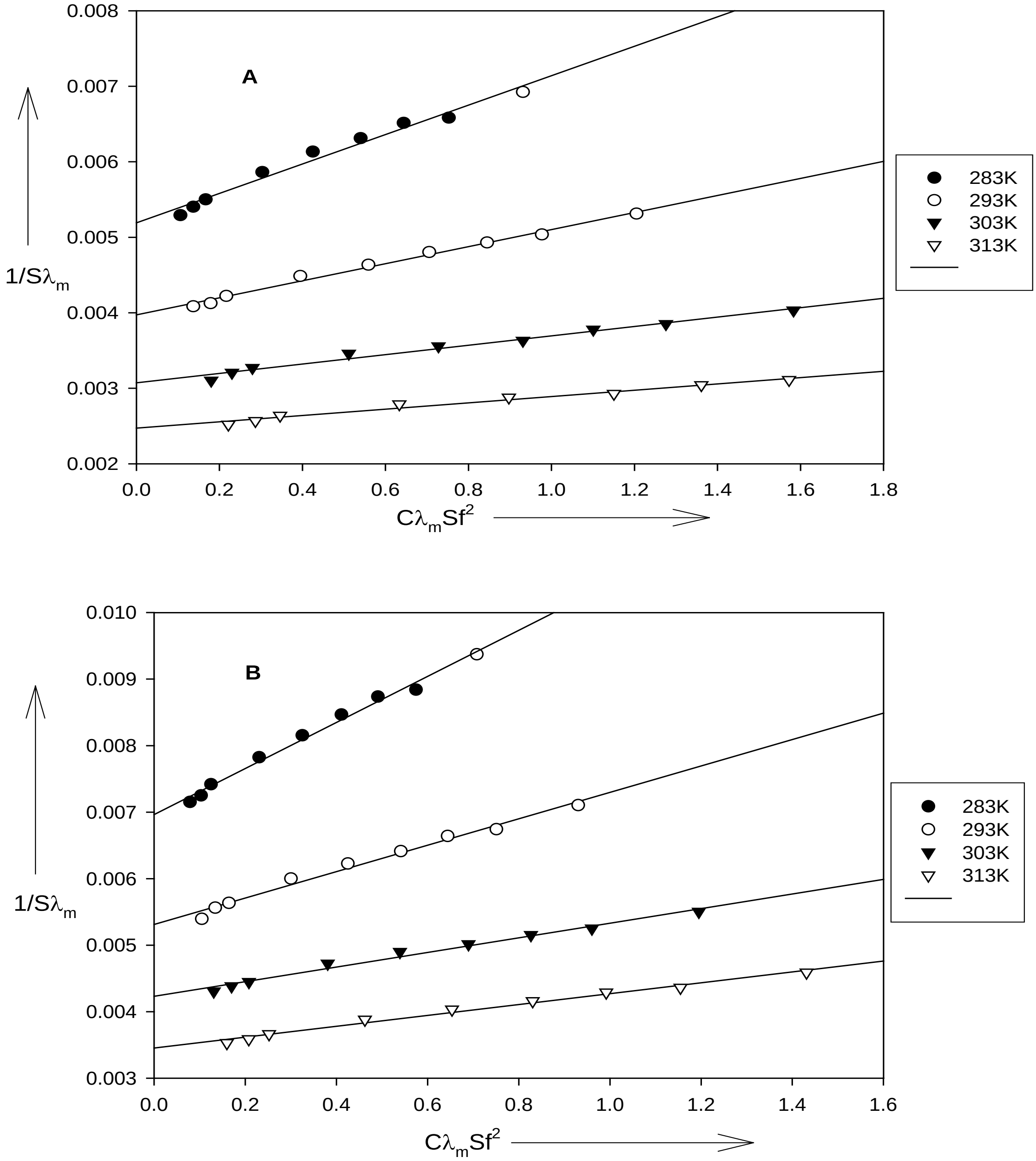


Fig. 2: Plot of $1/S\lambda_m$ Vs $C\lambda_m Sf_\pm^2$ for sodium citrate in A-20% CH_3CN

From the Table 1, it is clear that the temperature enhanced the conductivity value for all compositions of water + CH_3CN and water + DMSO. This is expected due to the fact that the increase in temperature increases the thermal energy and mobility of ions leading to increase in conductivity.

Limiting molar conductance decreases with increase in the amount of co-solvent (CH_3CN or DMSO) due to the increased solvent-solvent interaction and decreased dielectric constant value from 76.7 to 53.0 in case of water + CH_3CN and 76.7 to 70.5 in case of water + DMSO at 303K. The limiting molar conductance varies with the solvent as well as with temperature as expected. The minimum conductance value was observed at 50% DMSO (ε = 70.5), whereas maximum was observed at 60% acetonitrile (ε = 53.0) at all studied temperatures. Even though both the solvents are dipolar, DMSO is structure former and acetonitrile is structure breaker[19]. Relatively high viscosity and molecular size of DMSO over CH_3CN have lead to lower conductivity of sodium citrate in water + DMSO media. Since the viscosity and conductivity are inversely related naturally the conductance should be high in the presence of acetonitrile in water. The present result is in conformity with this trend. DMSO is expected to undergo self association[19] leading to varied molecular size and formation of some interstitial cavity in water. This cavity may trap some of cations released from sodium citrate bringing down the conductance and also show increased ion-solvent interaction.

Dissociation and Association constants

The values of dissociation constant (K_C) calculated from the Kraus-Bray plot and association constant (K_a) obtained from the Shedlovsky plot are shown in Table 2. K_C increased with increase in temperature in both the case of solvent mixtures.

In this case the dissociation constant did not found reciprocal to the association constant even though one expects the reciprocal relation. This might be due to the non consideration of electrophoretic effect, relaxation effect, inter ionic activity co-efficient in the case of Kraus-Bray equation. In general, the variation of these values either with temperature or with the solvent composition is mainly due to the change in dielectric constant and the reduction in thickness of ionic atmosphere surrounding the polar species. It is true that ion-solvent and solvent-solvent interactions affects limiting molar conductance and association constant which is proved by the data given in Tables 1 and 2. It is also known[1,2] that they are governed by various factors like dielectric constant, viscosity, chemical effect like short range interactions (ion-dipole, ion-ion induced dipole dispersion hydrophobic and hydrophilic interactions) as well as cavity and related solvent structural effects. To prove the formation of ion pairs and ion triplets, a plot of log λ_m Vs log C was drawn (Fig. not shown) on the basis of Fuoss equilibrium equation[20]. But in the present case, the slope values are found to be less than -0.5 (values are not shown), as against the expected -0.5 as per Fuoss for a system with ion-pair and hence there may not be any notable amount of ion association.

Walden Product

Walden product[21] relates molar conductance at infinite dilution (λ_m^0) to viscosity (η_0) of the solvent as

$$\lambda_m^0 \eta_0 = \frac{Ze_0 F}{6\pi r T} \quad (3)$$

where r is the Stoke's molecular radius. The product was calculated at all temperatures and percentage compositions of both the solvent mixtures and the values are shown in Table 3. Walden product decreased with the increase in the amount of co-solvent in water at all temperatures. The variation of Walden product from one composition to other in water + CH_3CN or water + DMSO mixtures clearly indicates the preferential solvation of cation by CH_3CN or DMSO molecules. It depends on the viscosity of the medium, the exchange of ions with neighbouring solvent molecules and movement of ions through the holes in hydrogen bonded solvent mixtures. From the eq. 3, it is clear that Walden product is directly related to Stoke's molecular radius. But in the present case corrected Stoke's radius (r_i) is calculated and shown in Table 4. Corrected Stoke's radius slightly decreased with the increase in either temperature or percentage composition, indicating that it is not fully responsible for the variation of λ_m^0 (Table.1). So other solvent structural properties are responsible for the variation of λ_m^0.

Table 2: Experimental values of dissociation constant (K_C) and association constant (Ka) for sodium citrate at different dielectric constant (ε at 303K) of water + CH_3CN and water + DMSO mixture

	K_C					**K_a**				
T(K)/ε	**76.7**	**72.6**	**68.6**	**60.7**	**53.0**	**76.7**	**72.6**	**68.6**	**60.7**	**53.0**
				Water + CH_3CN						
283	0.020	0.022	0.019	0.012	0.004	45.6	49.5	52.2	108.5	289.5
293	0.019	0.023	0.020	0.013	0.005	47.4	43.5	55.5	89.8	236.8
303	0.018	0.022	0.021	0.014	0.006	51.7	41.7	44.2	42.2	217.5
313	0.016	0.021	0.021	0.011	0.006	59.6	49.9	53.0	43.3	192.0

	K_C					**K_a**				
T(K)/ε	**76.7**	**76.5**	**75.2**	**74.0**	**70.5**	**76.7**	**76.5**	**75.2**	**74.0**	**70.5**
				Water + DMSO						
283	0.020	0.026	0.017	0.013	0.009	45.6	46.3	54.6	77.6	135.0
293	0.019	0.025	0.019	0.012	0.010	47.4	43.2	54.7	96.7	124.0
303	0.018	0.025	0.020	0.014	0.010	51.7	44.3	47.9	90.4	116.0
313	0.016	0.022	0.019	0.011	0.011	59.6	45.6	49.5	85.2	110.0

Table 3: Computed values of Walden product ($\lambda^0_m\eta_0$: mho cm^2 mol^{-1}poise) for sodium citrate at different dielectric constant (ε at 303K) of water + CH_3CN and water + DMSO mixture

T(K)/ε	**76.7**	**72.6**	**68.6**	**60.7**	**53.0**	**76.7**	**76.5**	**75.2**	**74.0**	**70.5**
	Water + CH_3CN					**Water + DMSO**				
283	2.85	2.77	2.54	2.08	1.52	2.85	2.75	2.82	2.61	2.57
293	2.83	2.67	2.43	2.03	1.47	2.83	2.69	2.69	2.70	2.46
303	2.79	2.71	2.64	2.12	1.56	2.79	2.54	2.55	2.35	2.35
313	2.77	2.80	2.66	2.24	1.62	2.77	2.55	2.47	2.32	2.13

Table 4: Computed values of corrected Stoke's radius (r_i in A^0) for sodium citrate at different dielectric constant (ε at 303K) of water + CH_3CN and water + DMSO mixture

T(K)/ε	76.7	72.6	68.6	60.7	53.0	76.7	76.5	75.2	74.0	70.5
	Water + CH_3CN					Water + DMSO				
283	2.29	2.25	2.23	2.20	2.25	2.29	2.00	1.98	2.00	1.97
293	2.25	2.22	2.21	2.18	2.25	2.25	1.98	1.97	1.95	1.96
303	2.21	2.18	2.15	2.14	2.20	2.21	1.96	1.95	1.95	1.93
313	2.18	2.13	2.11	2.09	2.16	2.18	1.92	1.93	1.93	1.94

Thermodynamic Parameters

Since conductance can be related to the rate process[22], the variation of λ_m^0 with temperature can be given by the equation

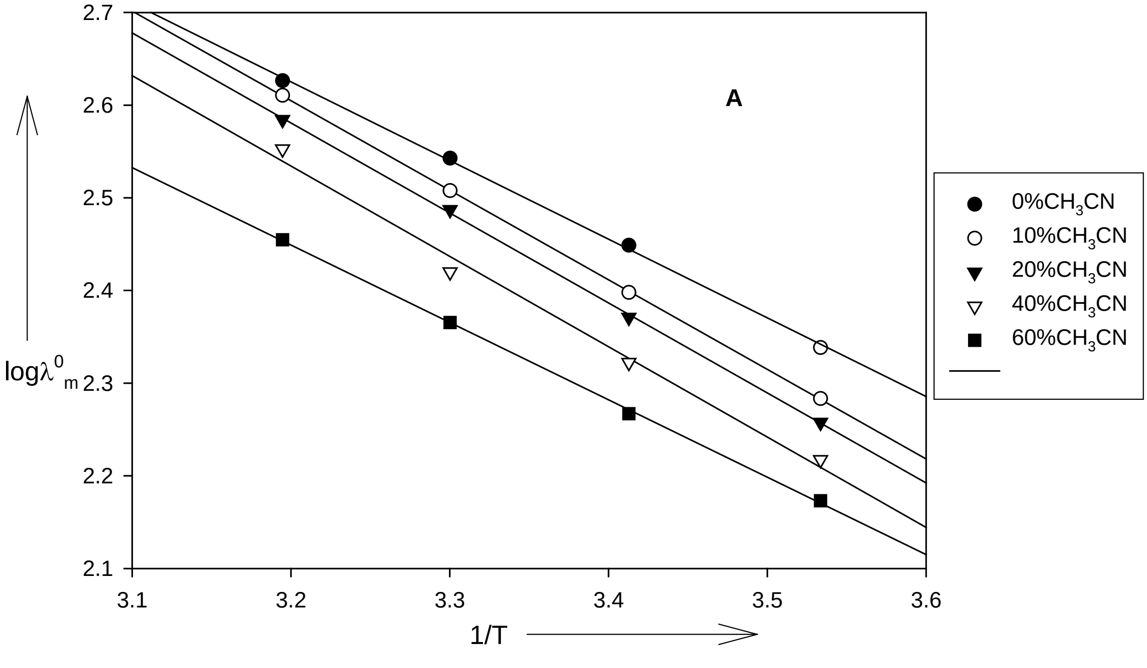
$$\lambda_m^0 = A.e^{\frac{-E_a}{RT}} \quad (4)$$

Where, E_a is the energy of activation of the conducting process. E_a was calculated[22] from the slope of the linear plot of log λ_m^0 Vs 1/T (Fig.3) and the values are shown in Table.5.

The energy of activation of the rate process increases till 20% of CH_3CN and later decreases. In the case of water + DMSO, E_a increases with the increase in percentage composition except at 10% (ε = 76.5) and 20% (ε = 75.2) of DMSO. ΔH_a the change in enthalpy of association was calculated from the slope of the linear plot of log K_a Vs 1/T (Fig. not shown). The computed values are shown in Table 5. The value of ΔH_a is negative except at 0% and 20% of CH_3CN and 0% and 10% of DMSO. Change in free energy and change in entropy were calculated from the equation $\Delta G_a = -RT \ln K_a$ and ΔS_a from $(\Delta H_a - \Delta G_a)/T$ respectively and the values are presented in the Table 5. ΔG_a is found to be negative, indicating spontaneity of the process. ΔS_a, the change in entropy is found to be positive in all the cases except at 40% CH_3CN indicating steric hinderence in the system.

Table 5: Computed thermodynamic parameters for sodium citrate at different dielectric constant (ε at 303K) of water + CH_3CN and water + DMSO mixture

Thermo dynamic parameters	Water + CH_3CN				
	ε = 76.7	ε = 72.6	ε = 68.6	ε = 60.7	ε = 53.0
E_a ($kJmol^{-1}$)	18.5	18.6	18.7	17.8	16.2
ΔH_a ($kJmol^{-1}$)	4.4	-5.7	3.4	-16.6	-7.8
ΔG_a ($kJmol^{-1}$)	-9.7	-9.5	-9.7	-10.3	-13.4
ΔS_a ($kJ\ K^{-1}mol^{-1}$)	0.048	0.012	0.044	-0.021	0.018
	Water + DMSO				
	ε = 76.7	ε = 76.5	ε = 75.2	ε = 74.0	ε = 70.5
E_a ($kJmol^{-1}$)	18.5	17.0	16.3	19.5	21.0
ΔH_a ($kJmol^{-1}$)	4.4	1.8	-2.4	-4.0	-5.5
ΔG_a ($kJmol^{-1}$)	-9.7	-9.4	-9.8	-11.1	-11.9
ΔS_a ($kJ\ K^{-1}mol^{-1}$)	0.048	0.037	0.025	0.024	0.021



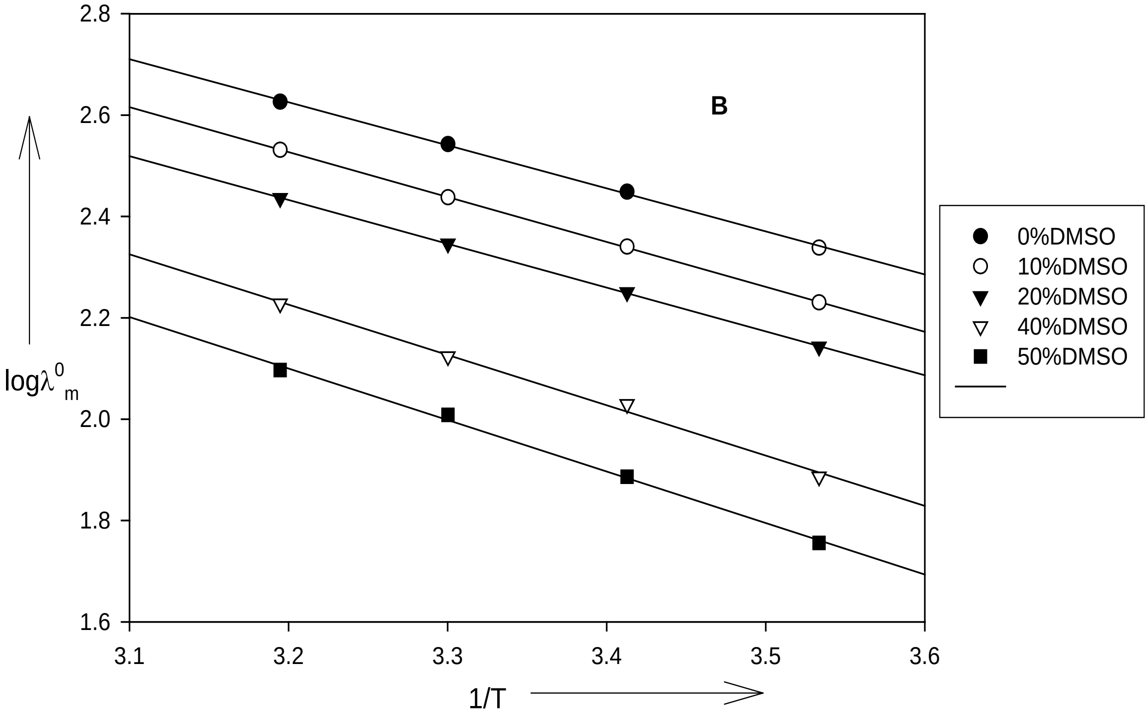


Fig. 3: Plot of log λ^0_m Vs 1/T for sodium citrate in A-water+CH_3CN

B-water + DMSO

Thermodynamics of solvation

Solvation is a process where an ion gets covered by the solvent molecules and form a sheath on an ion and the number of solvent molecules present in this sheath is called solvation number. Born proposed[23] a relation between thermodynamics of solvation $\Delta G_{s\text{-}s}$, $\Delta H_{s\text{-}s}$ and $\Delta S_{s\text{-}s}$ and the radius of the solvated ion and the dielectric constant of the solvent as

$$\Delta G_{s\text{-}s} = -\frac{N_A(Z_i e_0)^2}{2r_i}\left(1-\frac{1}{\varepsilon}\right) \quad (5)$$

$$\Delta S_{s\text{-}s} = \frac{N_A(Z_i e_0)^2}{2r_i}\frac{1}{\varepsilon^2}\frac{\partial\varepsilon}{\partial T} \quad (6)$$

$\Delta H_{s\text{-}s} = \Delta G_{s\text{-}s} + T\,\Delta S_{s\text{-}s}$ (7)

Values of thermodynamics of solvation computed on the basis of the above equations and are shown in Table 6. The values are different from those of thermodynamics of association due to the involvement of different mechanism in these two cases. The stability of the species in solution depends on the free energy change arising from solute-solvent interaction. Larger the negative value higher will be the stability of the species in that solution. Observed large negative value of $\Delta G_{s\text{-}s}$ indicate the stability of the species in solvent media than in water which is supported by the exothermic character of the reaction indicated by the negative $\Delta H_{s\text{-}s}$. The $\Delta S_{s\text{-}s}$ values are very small, suggest the requirement of steric restriction for the formation of ion pairs or ion association. To verify the Born relation a plot of -$\Delta G_{s\text{-}s}$ Vs $1/r_i$ was drawn and was found to be linear (Fig. 4) proposing that the Born model of solvation is applicable in this case.

Solvation number (S_n)

Solvation number (S_n) may be defined as the number of solvent molecules around an ion that have better interaction with the central ion and is greater than to that with the other solvent molecules either in the secondary region or in the bulk of the solution. They do not have any translational degree of movements. Solvation number reflects the magnitude of ion-solvent interaction in the system. Solvation number can be calculated using the relation[24]

$$S_n = \frac{d_{AB} - r_i}{r_{sol/sol\,mix}} \quad (8)$$

Where d_{AB} is the inter nuclear distance and r_i is the corrected Stoke's radius of an ion. Difference between d_{AB} and r_i gives the surplus number of solvent molecules present around an ion. When it is divided by the actual radius of the solvent /solvent mixture, the resulting value represents the solvation number. The inter nuclear distance or the closest distance of approach was calculated on the basis of the following equation[24]

$$\log\lambda_m{}^0 = \log\lambda - \frac{Z_A Z_B e^2}{\varepsilon d_{AB} kT} \quad (9)$$

Here Z_A, Z_B are the ionic charge on the cation and anion , e is the electronic charge, ε is the dielectric constant, d_{AB} is the inter nuclear distance, k is the Boltzman constant and T is the absolute temperature.

The plot of log λ^0_m versus 1/ε (was obtained by varying % composition of water + CH_3CN and water + DMSO) was drawn and found to be linear at all temperatures (Fig. 5). It gave a slope of $Z_A Z_B e^2/d_{AB}kT$ from which the value of inter nuclear distance d_{AB} was determined. S_n appear to be high indicating the existence of solvent separated ion pair. Calculated S_n values are shown in Table 7.

Table 6: Computed thermodynamics of solvation for Sodium citrate at different dielectric constant (ε at 303K) of water + CH_3CN and water + DMSO mixture

T(K)/ ε	76.7 (0%)			72.6(10%)			68.6(20%)			60.7(40%)			53.0(60%)		
	1	2	3	1	2	3	1	2	3	1	2	3	1	2	3
							Water+CH_3CN								
283	309.7	0.018	304.6	315.0	0.020	309.5	317.6	0.020	311.8	321.9	0.021	315.3	313.4	0.019	308.0
293	315.0	0.020	309.1	319.0	0.022	312.6	320.2	0.022	313.9	324.0	0.023	317.2	313.1	0.020	307.3
303	320.5	0.020	314.3	324.7	0.021	318.2	328.9	0.023	322.1	329.8	0.023	322.9	320.1	0.022	313.4
313	324.7	0.023	317.5	332.1	0.025	324.1	334.9	0.026	326.7	337.4	0.031	327.7	325.7	0.032	315.5
T(K)/ ε	**76.7(0%)**			**76.5(10%)**			**75.2(20%)**			**74.0(40%)**			**70.5(50%)**		
							Water+DMSO								
283	309.7	0.018	304.6	354.5	0.017	349.8	358.0	0.015	353.9	354.4	0.019	349.1	359.6	0.018	354.5
293	315.0	0.020	309.1	357.9	0.018	352.6	359.7	0.016	355.0	363.1	0.021	357.2	361.3	0.020	355.6
303	320.5	0.020	314.3	361.4	0.020	355.3	363.2	0.020	357.1	363.0	0.036	352.1	366.6	0.033	356.4
313	324.7	0.023	317.5	368.7	0.025	360.9	366.7	0.024	359.1	366.6	0.012	362.9	364.5	0.020	358.2

1. $-\Delta G_{s\text{-}s}$ (kJ mol^{-1}) 2. $\Delta S_{s\text{-}s}$ (kJ $K^{-1}mol^{-1}$) 3. $-\Delta H_{s\text{-}s}$ ($kJmol^{-1}$)

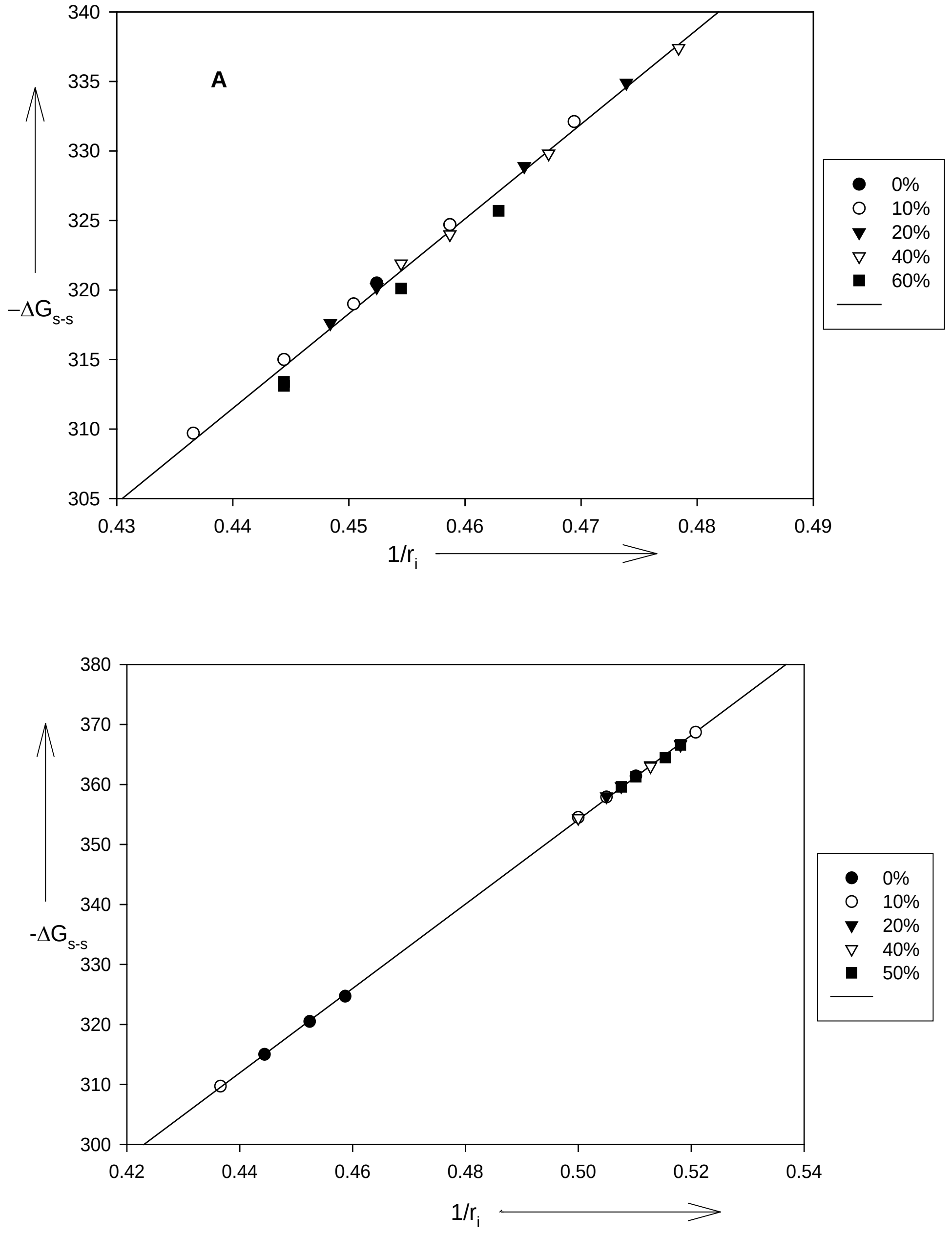
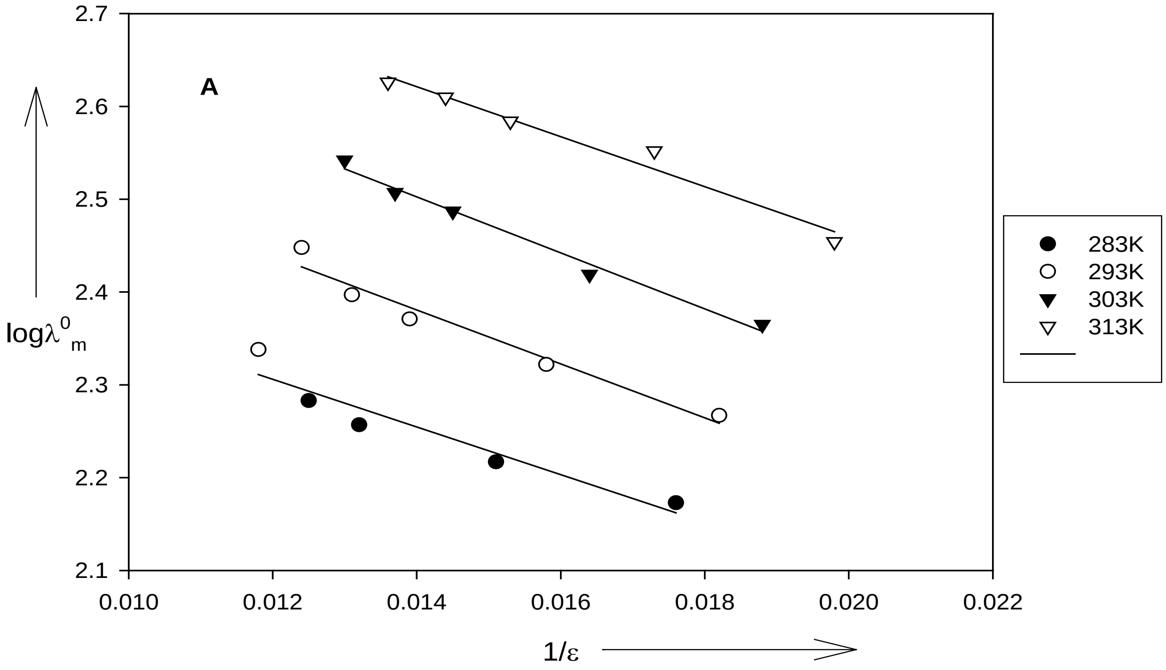


Fig. 4: Plot of $-\Delta G_{s\text{-}s}$ Vs $1/r_i$ for sodium citrate in A-water + CH_3CN; B-water + DMSO



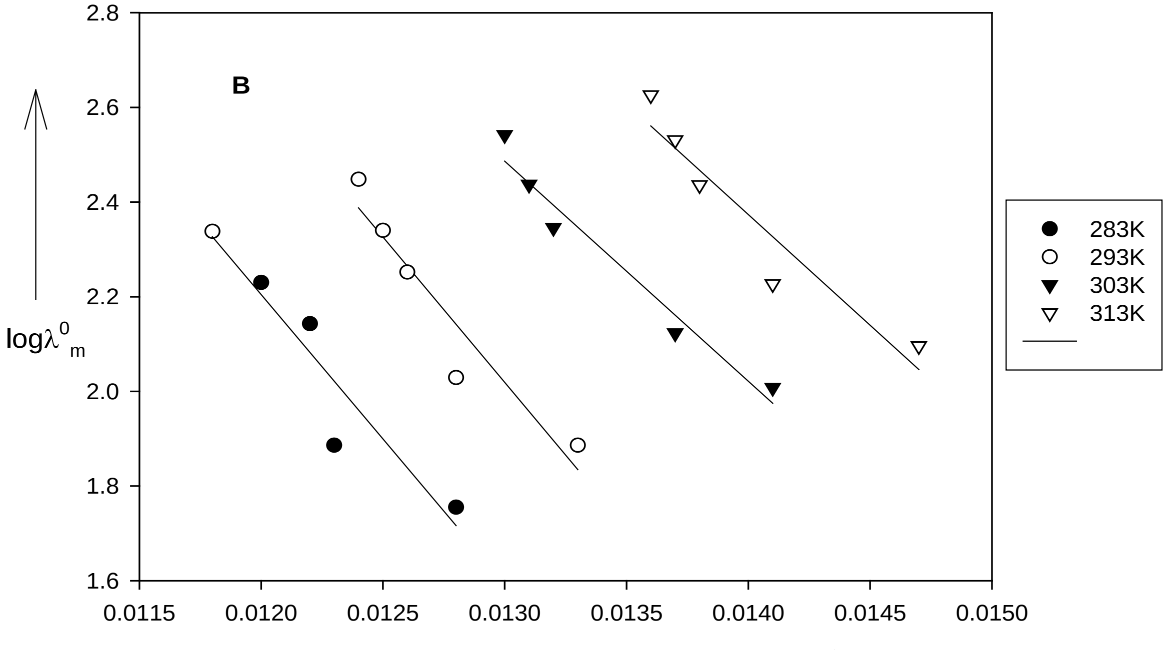


Fig. 5: Plot of log λ^0_m Vs 1/ε for sodium citrate in A- water + CH_3CN; B- water + DMSO

Table 7: Computed values of solvation number for the species of sodium citrate in water + CH_3CN and water + DMSO mixture at

T(K)	Water + CH_3CN	Water + DMSO
283	25.4	8.3
293	21.1	8.2
303	17.4	8.9
313	19.7	5.5

CONCLUSION

The limiting molar conductance (λ_m^0) varies with the solvent as well as with temperature as expected. Lower conductance value was observed in the water + DMSO compared to water + CH_3CN mixture at all studied temperatures. This is due to structure forming nature of DMSO and structure breaking nature of acetonitrile. It is further proved by viscosity and molecular size of these two solvents. The viscosity and molecular size of DMSO is high compare to CH_3CN and hence lower conductance was observed in the case DMSO medium.

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