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Study of Molecular Interionic interactions of Ammonium Magnessium Sulphate in 50 % DMSO –water System at different temperature.

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Abstract

Thermodynamic or conductance studies are one of the most important tools to study the solute-solvent interactions and the effects of solute on the structure of solvent. The best understanding of these effects can be attained from the knowledge of transport number, dissociation constants, limiting equivalent conductance, degree of association or dissociation, Walden products and activity coefficients of the electrolyte solutions. Density, viscosity were measured for Ammonium Magnesium Sulphate in 50 % DMSO-water medium at temperature 308.15 and 313.15 K. The densities (ρ) of the solvent and solutions were measured by using a bicapillary pycnometer having a bulb capacity of 10 ml at various temperatures. The viscosities of the solution were evaluated by measuring flow time of solution using an Ubbelohde suspended-level viscometer, calibrated with double distilled demineralised water.[1]

Molar volume and apparent molar volume ($\phi^o v$) have been calculated from density measurement and their concentration dependence are discussed. The related thermodynamic parameters such as apparent molar volume, limiting apparent molar volume, Jones dole A and B coefficient of Ammonium Magnesium Sulphate in 50 % DMSO have been applied as a way to study different interactions. By using density and viscosity data structural interactions like solute-solute, solute-solvent have received vital importance in physical chemistry. The apparent and partial molar volume provides usefull information about various types of interactions occurring in solution. These studies are of great help in characterizing the structure and properties of solution Knowledge of ion-solvent interactions in aqueous and non aqueous media is considerable fundamental and technological importance.[2] The parameters obtained by Masson equation and Jones-Dole equation are limiting apparent molar volume ($\phi^o v$) and their associated constant (Sv), viscosity A and B coefficient of Jones – Dole equation. The results show strong solute-solvent interactions at higher temperature 313.15 k.

Key words

Density, viscosity, limiting apparent molar volume, Jones-Dole equation, Masson equation, Viscosity coefficient,

Introduction

Thermodynamics has great importance in physical chemistry as well as in solution chemistry. Accurate knowledge of the physico-chemical properties of solutions has great relevance in theoretical and applied areas of research. Such parameter is functionally dependent on temperature. When only small quantities of liquid are available or where greater accuracy is required, the density of liquid is best determined by means of vessels of accurately defined volume called pycnometer. The pycnometer invented by spengel and modified by Ostwald is very popular for accurate density measurement [3]. A bicapillary pycnometer was chosen for the present work. The pycnometer was washed with chromic acid to remove obstructions in the capillary. This enabled a smooth flow of liquid without drops sticking behind. This was further washed by with distilled water and acetone and then dried with stream of warm air from hot blower. It was further weighed accurately on Dhona balance having sensitivity of 0.1 mg. The pycnometer was filled with air free double distilled water by dipping the end of A limb with a water taken in a beaker. Water filled up to mark D in limb B by capillary action. The pycnometer was weighed again to obtain the mass of water taken in it. It was mounted accurately vertical in a glass sided thermostat.

The temperature of the thermostat was controlled with in 0.01^{o}C. The heights of water in limbs A and B say h_1 and h_2 respectively were noted at various temperatures. The heights of water in limbs A and B say h_1 and h_2 respectively were noted at various temperatures. From the known densities of water at various temperatures and the weight of the water taken by a pycnometer, from the weight of pycnometer corresponding volumes of water were calculated.

The weight of pycnometer is on dhona balance it is very sensitive. These volumes were plotted against the total height (h_1+ h_2) of water levels in pycnometer yielding straight line graph with slope, intercept 0.005, 10.92 respectively. This graph is called calibration curve. This slope and intercept values are used as constant in calculation of density.[4]

Pycnometer was removed from the thermostat and cleaned as above. It was filled with experimental liquid and mounted in the thermostat. The procedure was repeated to find the total heights (h_1+h_2) for experimental liquids at various temperatures. From these total heights, the corresponding volumes of liquids under investigation were obtained from the calibration curve and the corresponding densities of solutions are determined.

Viscosity study is another fundamental transport property with the help of which the ion-solvent interactions can be investigated. The viscosity data involving low concentration of electrolyte has generally been analysed by the Jones-Dole equation.[5] Various types of interactions exist between the solute in solutions and these solute-solute and solute-solvent interactions are of current interest in all branches of chemistry. These interactions provide better understanding of solute modifiers or distort the structure of the solvent.[6] Molar volume provides various types of interactions exist between solute and solution and these solute-solute and solute-solvent interactions are of current interest in all branches of chemistry. These studies help in elucidating the structure of molecule[7].

Thermodynamics is a fundamental subject of great importance in physical chemistry and chemical engineering. The properties of liquid mixtures basically depend on its local structure, expressed in terms of packing density and volume. It changes with composition and temperature. This change in composition changes properties like density and viscosity of mixtures. The nature and type of interactions in binary organic liquid mixtures have been studied in terms of mixing parameters such as excess molar volume.[8]

Viscosity measures the fluid quality. The important information regarding solute-solute, solute-solvent and solvent-solvent interactions in an aqueous and in nonaqueous solution study by viscometric study. The experimental measurement of density, viscosity and derived parameters such as apparent molar volume, this data provide some significant information regarding the state of interactions in solution. Viscometric measurement of electrolyte solution have been widely used in order to obtain information regarding solute-solvent interactions. The dependence of concentration of viscosity in concentrated electrolyte solution was studied by V. Vand.[9] Interactions of electrolytes in binary mixtures of two liquids have been studied in terms of B coefficient of viscosity. Viscometric method used to study the behavior of certain electrolyte dissolved in binary mixtures. Density was analyzed in terms of Masson equation and viscosity was analyzed in terms of Jones-Dole equation.

Material and Methods

The chemicals NN, Dimethyl Formamide and Dimethyl sulphoxide employed were of analytical grade and purchased from E.Merck, company, used without further purification.

Demineralised water were double distilled and used. The component Ammonium Magnesium Sulphate was purified by crystalline and dried in oven at 50 ^{O}C and then used to prepare solutions of different proportion. Commercially available AR grade chemicals dimethylsulphoxide (DMSO) obtained from E-Merck chemical company and double distilled water were used for preparing the electrolyte solutions. DMSO is aprotic solvent and is strongly associated due to highly polar sulphoxide group. Water was distilled in quick fit apparatus over alkaline $KMnO_4$ followed by further distillation.

The aqueous solution of Ammonium Magnesium Sulphate was obtained by accurately weight amount of Ammonium Magnesium Sulphate were dissolved in particular solvent to give 1 M concentration. The Ammonium Magnesium Sulphate is used as electrolyte was supragrade quality 99.5 % pure. (E-Merck Chem.) After formation of 1M stock solution of Ammonium Magnesium Sulphate, it is used for preparation of different concentrations 0.2M to 1.4M. The concentrations were obtained by using dilution technique. The solutions were stored in dark colour amber bottles which are kept in dry box.

Bicapillary pycnometer was used for measurement of density at temperature 308.15 and 313.15 K. The mass were measured by using Dhona electronic balance. The Ubbelohde viscometer is a U-shaped piece of glassware with a reservoir on one side and a measuring bulb with a capillary on the other side. The time that the liquid level takes to flow between two ring marks indicates the viscosity of test fluid.89. A viscometer was selected having a flow time of about 181 seconds for water at 298.15 K Viscosity measurements are carried out by using Ubbelohde viscometer. The experimental measurements of flow time of the solution between two points on the viscometer were performed at three times for each solution and the average results were noted. Time was monitored by using stopwatch. The calibration of glassware's like pycnometer and viscometer[10] was carried out by using double distilled water. An average of three readings was taken. The density and viscosity measurements were carried out in a glass side thermostatic water-bath.

Results and Discussion

The densities and viscosities of Ammonium Magnesium Sulphate of different concentrations like 0.2 to 1.4 M in 50 % aqueous DMSO were determined at temperature 308.15 and 313.15 K. It is observed from table- 1 that densities ρ and viscosities η increase with increase in concentration of Ammonium Magnesium Sulphate.

1. **Apparent molar volume of Ammonium Magnesium Sulphate-**

The apparent molar volume of solution is a quantity defined with the purpose of isolating the contribution of each component to the non-ideality of the mixture. It shows the change in the corresponding solution property like volume when that entire component is added to the solution. It is described as apparent because it appears to represent the molar property of component in solution. The study of structural interactions by means of molar volume has received vital importance in physical chemistry. The apparent molar volume of the salts depends on their concentration due to the ion-ion interactions. The extrapolation of the experimental ϕv value to infinite dilution to obtain $\phi^0 v$ by redlich equation.

The apparent and partial molar volumes have proven to be a very useful tool in elucidating the interactions occurring in aqueous and non-aqueous solutions. The apparent molar volume ϕv were calculated by using equation [11],

$$\phi v = M2 /d - (d-d0) /(M\ d\ d0)$$

The apparent, limiting apparent molar volume and partial molar volumes provide useful information about various types of interactions occurring in solution i.e. solute-solute, solute-solvent and solvent-solvent occurring in solution. The apparent molar volume (ϕv) was determined from density of solution using following equation

$$\phi v = M/\rho_o - 1000(\rho-\rho_o)/\rho_o C$$

Where ρ_o and ρ are the densities of solvent and solution respectively, C is the molar concentration and M is molecular weight of solute. Apparent molar volume of solute changes with the square root of the molar concentration and obeys Masson equation.[12]

$$\phi v = \phi^o v + Sv\ (C)^{1/2}$$

Where $\phi^o v$ is limiting apparent molar volume of the solute and Sv is the experimental slope.

An apparent molar volume property of solution component is quantity with the purpose of isolating the contribution of each component to the non ideality of the mixture. The ϕv value has been positive and increases with increasing concentration of electrolyte Ammonium Magnesium Sulphate. Apparent molar volume of the solution of Ammonium Magnesium Sulphate in DMSO-water mixture at 308.15 and 313.15 k are reported.

Limiting apparent molar volume ($\phi^o v$) of ammonium magnessium sulphate in DMSO – water solution is depending on temperature which can be represented by following equation.

$$\phi^o v = a_O + a_1 T + a_2 T^2$$

The value of coefficient a_0, a_1 and a_2 are calculated by differentiating above equation with respect to temperature T, where T is temperature.

$$\phi^o v = 50.64 + 0.1899\ T + 0.000052\ T^2$$

Density data of Ammonium Magnesium Sulphate was analyzed with the help of Masson equation. Slope Sv and intercept $\phi^o v$ constants are obtained by linear plots of $C^{1/2}$ Vs ϕv. The possible explanations for positive slope that the ionic association as the concentration of the electrolyte Ammonium Magnesium Sulphate is increased, there is weakening the solute-solvent interaction. The increase of Sv with increase of temperature suggest that more and more solute is accommodated in the void space left in associated solvent molecules and then enhance the structure of solvent molecule. Apparent molar volume at different temperature is given in table 1 and table 2.

Table 1: Density ρ and viscosity η and apparent molar volume of Ammonium Magnesium Sulphate in 50 % DMSO-water at temperatures 308.15 k.

Conc.	**P**	**η**	**ϕ_v**
C	**g cm^{-3}**	**m Pa s**	**$Cm^3\ mol^{-1}$**
0.2	1.03523	1.12976	-2008.5
0.4	1.04259	1.14337	-999.83
0.6	1.04493	1.14595	-890.96
0.8	1.04683	1.14735	-695.16
1.0	1.04894	1.14885	-540.74
1.2	1.05090	1.14900	-311.28
1.4	1.05281	1.15200	-112.55

Table 2: Density ρ and viscosity η and apparent molar volume of Ammonium Magnesium Sulphate in 50 % DMSO-water at temperatures 313.15 k.

Conc.	**P**	**η**	**ϕv**
C	**g cm^{-3}**	**m Pa s**	**$Cm^3\ mol^{-1}$**
0.2	1.03422	1.12888	-906.63

0.4	1.04152	1.14225	-839.93
0.6	1.04393	1.14485	-679.99
0.8	1.04598	1.14658	-448.98
1.0	1.04790	1.14778	-170.99
1.2	1.05095	1.14567	-97.663
1.4	1.05315	1.15112	-94.154

The negative sign of Sv suggests that the electrolytes behave as structure breakers in that particular solvent. The Sv values (solute-solute interaction) decreases as the size of the cation increases. $\phi^o v$ values are increased as the temperature is increases. The large $\phi^o v$ values of Ammonium Magnesium Sulphate reveal the strong solute-solvent interactions and preferential solvation of ions. The apparent molar volume ϕv has good agreement with reported value of M. Parmar and A. Khanna[13] . Temperature is increases then Sv value is decreases i.e solute-solute interaction decreases which are shown in table-1.

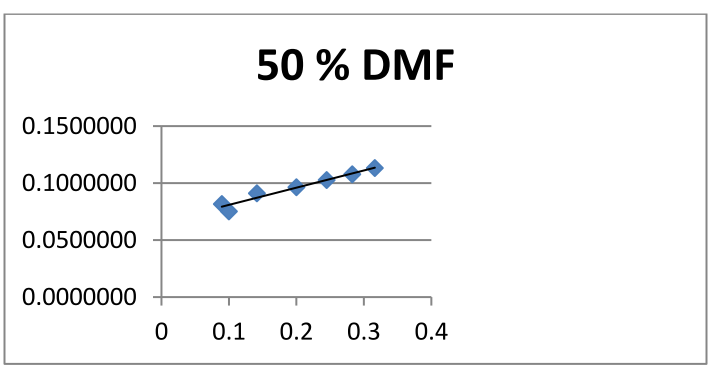

Fig-1 Plot of $C^{1/2}$ vs ϕv of Ammonium Magnesium Sulphate at 308.15 K

The results indicate as temperature is increases then $\phi^{o}v$ value increases. Solute-solvent interactions are increases.

2. **Viscometric Study of Ammonium Magnesium Sulphate in solution**

In 1929 Jones and Dole gave a quantitative formulation of the effect with their empirical equation viscosity data was analyzed according to Jones-Dole equation. The determination of the viscosity parameter can be used to interpret the structural property and solute-solvent interaction. It has been founded by number of workers , that the addition of an electrolyte will either break or make the structure of the fluid. The making or breaking of the structure of a fluid has been considered as a measure of the solute – solvent interaction. The viscosity data involving low concentration of electrolyte has generally been analysed by the Jones-Dole equation Time of flow of Ammonium Magnesium Sulphate of different concentrations like 0.2 to 1.4 M was measured at temperature 308.15 and 313.15 k. The viscosity of solution were represented by equation,[14]

$$\eta/\eta_o = t\,\rho / t_o\,\rho_o$$

Where η, t, ρ are the absolute viscosity , time of flow and density of solution while η_o, t_o, ρ_o are the same quantities for the solvent water. Viscosity of solution was calculated by using Jones- Dole equation. Viscometer constant is falkenhagen coefficient and Jones-Dole coefficient. These parameters was used to interpret the solute-solute, solute-solvent interactions. Viscosity data has been analyzed with the help of Jones-Dole equation from the linear plot of C1/2 versus $(\eta / \eta_o - 1)$ C1/2 . The values of coefficient A and B of Jones-Dole equation have been determined by computerized least square method.[15] Interionic attraction theory which as developed by Deb. A is the Falkenhagen coefficient which is the measure of solute-solute interactions[i] and B is the Jones- Dole coefficient which is the measure of solute - solvent interaction.

$$\eta/\eta_o = \eta_r = 1 + AC^{1/2} + BC$$

Where η_r is relative viscosity, C is molar concentration, A is the falkenhagen coefficient which shows solute-solute interactions and B is the Jones-dole coefficient which shows solute-solvent interactions. Viscosity of aqueous solution of Ammonium Magnesium Sulphate in 50 % DMSO at different concentrations like 0.2 and 1.2 M were measured. The plots of $C^{1/2}$ Vs $[\eta/\eta_o - 1]\, C^{1/2}$ are linear in all cases indicating Jones-Dole equation. The A coefficient represents the contribution from the interionic electrostatic forces.The B coefficient represents the solute – solvent.

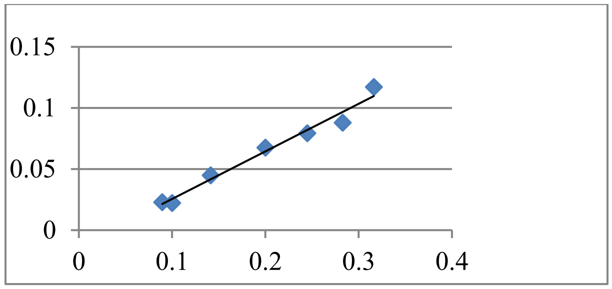

Fig-2 Plot of $C^{1/2}$ Vs [η / η_o -1] $C^{1/2}$of Ammonium Magnesium Sulphate at 308.15 K

Values of viscosity coefficient like A and B were determined by Jones-Dole equation[15] by using computerized least squares method which is shown in table-2

Table.3 Limiting apparent molar volume ($\phi^o v$), experimental slope (Sv) and viscosity Coefficient A and B at 308.15 and 313.15 k.

T	**$\phi^o v$**	**Sv**	**B**	**A**
K	**$cm^3\ mol^{-1}$**	**$cm^3\ L^{1/2}\ mol^{-3/2}$**	**$dm^3\ mol^{-1}$**	**$dm^{3/2}\ mol^{-1/2}$**
308.15	-1478.9	5112.3	0.4850	-0.0266
313.15	-1569.0	5678.4	0.6285	-0.0402

From the viscosity and density of Ammonium Magnesium Sulphate solutions in binary mixture of aqueous DMSO are at 308.15, 313.15 K temperatures. All the values of ϕ_v^0 at all temperatures are negative.[16] Falkenhagen and Jones-Dole coefficient are positive which indicate solute-solvent interactions. As the temperature is increases then B Coefficient is increases which show higher solute-solvent interactions.[17] As the temperature increases the viscosity A coefficient is decreases i.e solute-solute interaction is low at high temperature.

CONCLUSION

The apparent molar volumes (ϕ_v) and viscosity B coefficients (Jones Dole) for Ammonium Magnesium Sulphate in aqueous Dimethyl sulphoxide (DMSO) were evaluated from density (ρ) and viscosity (η) at various temperatures as 308.15, 313.15 K with the help of bicapillary pycnometer and Ubbelohde viscometer instruments respectively. The density data for all the solutions were analysed in limiting apparent molar volume (ϕ_v^o) and experimental slope (S_v) obtained from Masson equation. The viscosity data of solution were analysed in term of A and B coefficient obtained from Jone-Dole equation.Density, viscosity and apparent molar volume is increases as the concentration of Ammonium Magnesium Sulphate is increases in 50 % DMSO. As temperature is increases density and viscosity of Ammonium Magnesium Sulphate in 50 % DMSO is decreases. Falkenhagen coefficient and Jones-Dole coefficient at different temperature shows solute-solvent interactions are stronger at higher temperatures. At lower temperature solute-solute interaction are present.

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