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# Volumetric and Viscometric studies of solute –solvent and solute-solute interactions of Glycine in aqueous in aqueous electrolytes at 30°C

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**Abstract:** Densities and viscosities of Glycine in aqueous NaCl and MgCl<sub>2</sub>(0.02& 0.06 M ) solutions have been determined experimentally at 30°C. The results obtained from density and viscosity measurement have been used to calculate A and B coefficients and free energies of activation of viscous flow of solvent  $\Delta \mu_1^{0\#}$  and solute  $\Delta \mu_2^{0\#}$ . The results are discussed in terms of the dehydration effect of the electrolytes upon the amino acids and weak solute- solute and strong solute- solvent interactions. The properties of these amino acids in water and water+ electrolytes solution systems are discussed in terms of the charge , size and hydrogen bonding effect.

**Keywords:** Glycine, Aqueous Electrolytes, Jones- Dole coefficients and Free Energies of Activation

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## 1. Introduction

Amino acids have zwitter-ion and are the constituents of the most important class of biopolymers, i.e. Proteins. Disarrangement water and electrolyte balance in living systems cause a wide variety of health problems. In physiological media such as blood, membranes, cellulose fluids etc., the dipolar character of amino acids (in presence of ions such as Na<sup>+</sup>, K<sup>+</sup>, Mg<sup>+2</sup> and Cl<sup>-</sup> etc., dissolved in body water) has an important bearing on their biological functions. Therefore, a knowledge of water-amino acid interaction the effects on several biological processes occurring in living organism(1). The study of interaction between amino acids and electrolytes in aqueous medium, we present in this paper, the study of interaction between aqueous NaCl and MgCl<sub>2</sub> and Glycine at 30°C. There has been an increased interest in the physicochemical properties of amino acids in aqueous as well as aqueous electrolyte media to understand the role played by the biological molecules in living organism(2-6). In recent years, a number of workers have utilized density and viscosity data to deduce the thermodynamic properties (relative viscosity, Jones -Dole coefficient and free energy of activation of viscous flow) for a number of mixtures solutions (7-9). Structural interactions

of non-ionic solutes with ionic ones in different solvents are important in many fields of chemistry and bio-chemistry. Very recently, we have made systematic effort to investigate the ultrasonic and volumetric properties of amino acids in concentrated electrolytic solution (10-12). It was found that NaCl and MgCl<sub>2</sub> increase the apparent molar volume and decrease the adiabatic compressibility of glycine. This increase could be attributed to the interactions of the ions of the NaCl and MgCl<sub>2</sub> electrolytes and zwitter-ion head group of glycine, causing the transfer of hydrated water molecule to the bulk state.

In the present paper, we report densities,  $\rho$  and viscosities  $\eta$  of Glycine (0.01-0.09 M) in aqueous NaCl and MgCl<sub>2</sub>(0.02& 0.06 M) solutions have been determined experimentally at 30°C. From these experimental data a number of thermodynamic parameters namely, Jones- Dole equation to calculate A and B coefficients and free energies of activation of viscous flow of solute and solvent respectively have been calculated. These parameters were utilized to study various interactions taking place in the solutions of glycine in aqueous NaCl and MgCl<sub>2</sub> at 30°C.

## 2. Experimental

### 2.1. Chemical and Preparation

Glycine (99.5% purity), NaCl and MgCl<sub>2</sub> (99.8% purity) were procured from Merck and S d Fine Ltd. They were used as such without further purification, after drying over calcium chloride in desiccators for more than 48 hours. The viscosities and densities of the amino acids in aqueous electrolytes solution at various concentrations as well as in double distilled de-ionized water were measured experimentally. Aqueous solutions of NaCl and MgCl<sub>2</sub> (0.02 & 0.06 M) were prepared and these were used as a solvents to prepare glycine solutions on mass basis covering the whole composition range. All the solutions were prepared by mass in dry box and were stored in special air-tight bottles and kept in dark to avoid photo chemical degradation. The weighing was done on an Afcoset ER-120A electronic balance with an accuracy ± 0.1 mg.

### 2.2. Measurement of Density and Viscosity

The densities were measured with a single capillary pycnometer (made of Borosil glass) of bulb capacity of 8 × 10<sup>-6</sup> m<sup>3</sup>. The marks of the stems were calibrated using double distilled water at 30°C. The pycnometer was kept for about 30 minutes in a thermostatic water bath so that the thermal fluctuation in density was minimized. The viscosity measurements were carried out by Ubbelohde type suspended level viscometer which was first calibrated with doubled distilled water. The viscometer was allowed to stand in an electronically controlled thermostatic water bath for 30 minutes to minimize the thermal fluctuation. The time of fall was recorded with a stop –watch of least count 0.1 s. At least three time recorded were obtained, and the average value was used as the experimental flow time. Poiseuille's equation was employed to calculate the viscosity of the amino acid + electrolyte + water solutions.

$$\eta = \frac{\pi r h g r 4 t}{8 V} = \rho \beta t \quad (1)$$

Here  $\rho$  is the density of the amino acids solutions,  $h$  the height of the column in the viscometer,  $g$  is the acceleration due to gravity,  $r$  is the radius of the capillary,  $l$  the length capillary and  $t$  is the time of fall of the solution of volume  $V$ . The term  $h, g, r, l$  and  $V$  are constant for a given viscometer therefore these have been replaced by single term  $\beta$ . The temperature of the water bath was maintained at 30°C. The viscosity and density data were found to be accurate within ± 0.1 % and ± 0.01% respectively.

## 3. Results and Discussion

The densities and viscosities of Glycine (0.02 & 0.06 M) in aqueous NaCl and MgCl<sub>2</sub> (0.02 & 0.06 M) determined at 30°C and are presented in Table 1. It is observed from Table 1 that densities  $\rho$  and viscosities  $\eta$  for all the ternary systems increase with increase in molarities of glycine. The values of  $\rho$  and  $\eta$  increase with increase in concentration of amino

acids in all the ternary systems under investigation, which appear to be due to hydrophobic properties of solutes i.e. H-bond forming. This may be attributed to the formation of clusters by the amino acids and strong intermolecular forces in the solute. The changes in structure of solvent or solution as a result of H-bond formation lead to decrease in intermolecular free length (13). Solute may occupy the interstitial spaces in solvent or get solvated forming new weaker bonds. It was suggested (14-16) that what is experimentally observed for any system, reflects the compromise between the tendency for the ion and the peptide to interact with each other and inclination of the solutes to associate with the solvent.

**Table 1.** Densities ( $\rho$ ) and viscosities ( $\eta$ ) of glycine + aqueous NaCl and glycine + aqueous MgCl<sub>2</sub> at 30°C

C (mol.l <sup>-1</sup> )	$\rho$ (kg m <sup>-3</sup> )	$\eta$ (×10 <sup>-3</sup> Nm <sup>-2</sup> s)	$\rho$ (kg m <sup>-3</sup> )	$\eta$ (×10 <sup>-3</sup> Nm <sup>-2</sup> s)
Glycine + aqueous + NaCl				
	0.02 M		0.06 M	
0.00	1020.6	0.843	1003.8	0.862
0.10	1021.3	0.847	1004.6	0.866
0.30	1028.9	0.912	1007.3	0.890
0.50	1031.2	0.918	1011.8	0.913
0.70	1035.7	0.931	1020.6	0.942
0.90	1041.3	0.959	1024.7	0.967
Glycine + aqueous MgCl <sub>2</sub>				
	0.02M		0.06 M	
0.00	998.7	0.825	1006.2	0.903
0.10	1004.0	0.876	1006.4	0.916
0.30	1013.4	0.914	1015.7	0.960
0.50	1024.9	0.991	1017.5	0.989
0.70	1034.2	1.027	1020.9	1.014
0.90	1038.5	1.180	1025.3	1.062

The viscosity data were used to calculate the relative viscosity using Jones- Dole equation [17]

$$\eta_{rel} = \eta / \eta_o = [1 + AC^{1/2} + BC] \quad (2)$$

Where,  $\eta$  and  $\eta_o$  viscosities of the solutions and solvent respectively.  $B$ , is the Jones- Dole coefficient [17], an empirical constant, and is measure of ion-solvent interaction. Its values depend on the size and shape of the solute particles. The  $A$  is the Falkenhagen coefficient [18] which indicates ion-pair electrostatic interactions. They were obtained by a least square treatment as the intercepts and slopes of the linear plots of  $\eta / \eta_o - 1/C^{1/2}$  versus  $C^{1/2}$  and their values are given in Table 2.

**Table 2.** Falkenhagen coefficient, (A), Jones Dole coefficient, (B), Free energy of activation per mole of solvent, ( $\Delta \mu_1^{0\#}$ ) and solute ( $\Delta \mu_2^{0\#}$ ) of Glycine + aqueous NaCl and Glycine + aqueous MgCl<sub>2</sub> at 30°C

C (mol l <sup>-1</sup> )	A (10 <sup>-2</sup> dm <sup>3/2</sup> mol <sup>-1/2</sup> )	B (10 <sup>-1</sup> dm <sup>3</sup> mol <sup>-1</sup> )	$\Delta \mu_1^{0\#}$ (KJ mol <sup>-1</sup> )	$\Delta \mu_2^{0\#}$ (KJ mol <sup>-1</sup> )
Glycine + water				
NaCl				
0.02	5.926	0.958	9.12	28.39
0.06	-4.726	1.858	9.22	41.86
Glycine + water				
MgCl <sub>2</sub>				
0.02	-6.290	4.302	9.22	37.85
0.06	-0.320	1.950	9.13	72.34

The viscosity,  $\eta$ , of dilute solution of non-electrolytes is represented by

$$\eta = \eta_0(1+BC) \quad (3)$$

For a dilute solution of unsolvated spherical colloidal suspension, has derived by Einstein relation

$$\eta_{rel} = 1 + 2.5\phi \quad (4)$$

Where  $\phi$  is the volume fraction of the solute. If this equation is valid or amino acids, Eq. (3) becomes

$$\eta_{rel} = 1 + 0.0025 V_h C \quad (5)$$

Where  $V_h$  is the hydrodynamic volume, since  $AC^{1/2}$  term in Eq. 2 can be assumed to be negligible in a dilute solution, the following relation holds

$$B = 0.0025V_h \quad (6)$$

Hakinet.al[19] may be assumed that the partial molar volume at infinite dilution of the unsolvated solute particle in a continuum solvent. The more B values in the mixed solvent might mean a more hydrodynamic volume in the mixed solvent.

A perusal of Table 2 shows that the values of A coefficients are negative and those of B coefficients are large positive for all the ternary systems under investigation, thereby suggesting the ion-ion interactions are weak and ion-solvent interactions are strong. The positive value of B with electrolytes concentration of water molecules as a result of shielding of polar terminal groups of glycine molecules is due to increased interaction between these polar ends and ions of the electrolytes. They have estimated the contraction of water around the appositively charged group is caused by electrostatic ion- solvent interaction and is called as electrostriction. A mutual comparison of the NaCl and MgCl<sub>2</sub> shows that values of A and B are larger in case of MgCl<sub>2</sub> than NaCl. Mg<sup>+2</sup> ions being smaller in size, has an intense force field and hence a strong hydration co sphere around it. Therefore hydration of MgCl<sub>2</sub>( ion-solvent ) will be much more than that of NaCl. Further to this effect is superimposed the effect of interaction of cations (Na<sup>+</sup> and Mg<sup>+2</sup>) and anion ( Cl<sup>-</sup>) with negative and positive charge centers of glycine respectively (ion-ion interaction). The double charge on Mg<sup>+2</sup> ion results in intense electric field and thus, the possibility of

interactions with glycine is larger in case of glycine with aqueous MgCl<sub>2</sub> ternary system. The hydration behavior of amino acids considered the following interactions (a) Akhtar[4] as the terminal groups of zwitter-ions of amino nature which may hydrophobic, hydrophilic or amphiphilic, acids, -NH<sub>3</sub><sup>+</sup> and COO<sup>-</sup> are hydrated in electrostatic manner whereas hydration of intervening backbone depends on its, (b) the overlap of hydration groups co spheres of terminal (-NH<sub>3</sub><sup>+</sup> and COO<sup>-</sup>) and the adjacent groups result in volume change . (c) Electrostriction of NH<sub>3</sub><sup>+</sup> group is greater than the COO<sup>-</sup> by a factor of 10. Table 2 shows that the values of B are positive which indicates strong solute-solvent interactions.

According to the transition state theory of the relative viscosities of electrolytic solutions proposed by Feakins et al (20), the B- coefficient given as

$$B = (\bar{V}_1^0 - \bar{V}_2^0) / 1000 + \bar{V}_1^0 [ (\Delta \mu_2^{0\#} - \Delta \mu_1^{0\#}) / RT] \quad (7)$$

Where  $\bar{V}_1^0$ -and  $\bar{V}_2^0$  are the partial molar volumes of the solvent and solute at infinite dilution , respectively ,  $\Delta \mu_1^{0\#}$  the free energy of activation per mole of the solvent and  $\Delta \mu_2^{0\#}$  is the free energy of activation per mole of the solute . The  $\Delta \mu_1^{0\#}$  and  $\Delta \mu_2^{0\#}$  were calculated from the equation

$$\Delta \mu_1^{0\#} = RT \ln ( \eta^0 \bar{V}_1^0 / h N_A ) \quad (8)$$

and

$$\Delta \mu_2^{0\#} = \Delta \mu_1^{0\#} + RT / \bar{V}_1^0 [ 1000 B - (\bar{V}_1^0 - \bar{V}_2^0) ] \quad (9)$$

Where R, h and N are the gas constant, Planck's constant and constant respectively and T are the absolute temperature. The values of  $\Delta \mu_1^{0\#}$  and  $\Delta \mu_2^{0\#}$  for different compositions of glycine in aqueous NaCl/ MgCl<sub>2</sub> are given in Table 2. Table 2 shows that  $\Delta \mu_2^{0\#}$  are larger than  $\Delta \mu_1^{0\#}$  suggesting that the formation of transition state is accompanied by the breaking and distortion of the intermolecular bonds. Moreover, the greater values of  $\Delta \mu_2^{0\#}$  than  $\Delta \mu_1^{0\#}$  suggest that the metal chlorides under study, behave as structure makers/promoters in different concentration ranges of glycine. Greater values of  $\Delta \mu_1^{0\#}$  have also been reported in mixtures of sucrose solution(21).

A comparison of  $\Delta \mu_1^{0\#}$  and  $\Delta \mu_2^{0\#}$  values of the two solutes result the structure making ability of Mg<sup>+2</sup> is greater than Na<sup>+</sup> which may be due to stronger solute-solvents interaction in MgCl<sub>2</sub> solutions. Therefore the hydration of MgCl<sub>2</sub> will is much more than that of NaCl. The double charge on Mg<sup>+2</sup> results in intense electric field and the radius of Mg<sup>+2</sup> is much smaller than the Na<sup>+</sup> thus the possibility of interactions with glycine is larger in case of glycine + aq MgCl<sub>2</sub> ternary systems. The greater values of  $\Delta \mu_2^{0\#}$  at 0.06M concentration for both the electrolytes which indicates the maximum structure making ability. Increase in concentrations of electrolytes from 0.02 & 0.06 M probably causes disruption of the intermolecular bonds of the solvent, thereby decreasing the values of  $\Delta \mu_2^{0\#}$ .

Thus the trends and magnitude of the various parameters obtained from viscosity measurement reported in this paper. The studies suggest that ion- solvent interactions are stronger

and ion-ion interactions are weak. The extent of interactions and structure making ability is greater in case of  $MgCl_2$ . The  $\delta B/\delta T$  is a better criterion for determining the structure making/ breaking nature of any electrolyte rather than simply the B- coefficient.

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