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Research Article

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Studies in thermodynamic properties of electrolytes in aqueous solution of amino acid at different temperatures

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ABSTRACT

Ultrasonic velocity (u), density (ρ), and viscosity (η) have been measured for NaCl and MgCl₂ in aqueous alanine solution at 298.15, 303.15, and 308.15K. From these experimental data, thermodynamic parameters i.e. adiabatic compressibility (β a), intermolecular free length (L_f), Vander Waal's constant (b), internal pressure (Π_i) and solvation number (S_n) have been obtained for all the mixtures, with a view to investigate the solute-solute and solute-solvent interactions. Adiabatic compressibility and intermolecular free length decreases with increase in concentration and temperature. Van der Waal's constant (b), internal pressure (Π_i) and solvation number (S_n) increase with increase in temperature and concentration of electrolytes. These derived parameters have been further used to elicit the ion-ion, solute – solvent and ion-amino acids interactions in the mixtures.

Keywords: ultrasonic velocity (u), thermodynamic parameters such as adiabatic compressibility (β a), intermolecular free length (L_f), Van der Waal's constant (b).

INTRODUCTION

There are many approaches and spectroscopic techniques used to determine the structure - function relationship of bimolecules. They are X-ray crystallography, chromatography, NMR, EPR, vibration and Raman spectroscopy, neutron & light scattering, circular dichroism (CD), IR and ultrasonic velocity measurements. Among these techniques, ultrasonic velocity measurements have been found to be most powerful tool in the investigation of structure and molecular interactions occurring in the solutions. Most of the chemical and biological functions of bimolecules take place in aqueous medium. Important bimolecules are proteins, carbohydrates, fats, enzymes, vitamins, hormones and nucleic acids. Among these proteins [24, 26] are found in all parts of the body and they have an enormous variety of functions. As the proteins are large complex molecules, direct study of protein interactions is difficult. One useful approach that can be used to understand these interactions is to study those compounds which mimic some aspects of the protein structure. Recently, there has been considerable interest in the determination of various thermodynamic properties of such model compound e.g. amino acids, small peptides and their derivatives. Amino acids are the building blocks of the molecular structure of the important and very complex class of compounds known as proteins.

Electrolytes are expected to influence water structure and the importance of contribution from structural changes of the solvent to the thermodynamic properties of aqueous solutions of biological moleculels is often stressed. The information on the zwitter – ionic nature of amino acids in water is given in the literature [1-4]. The properties of proteins such as their structure, solubility, denaturation activity of enzymes, etc. are greatly influenced by electrolytes [5]. An electrolyte when dissolved in water perturbs the arrangement of water molecules with strong electric field of its ions. This property of electrolytes is known as structure maker or structure breaker and has been

widely used to understand the effect of electrolytes on the structure and function of both proteins and nucleic acids [6, 7].

How the ion-ion and ion-amino acids interactions together with ion-water and amino acid – water ones are altered in electrolytes and amino acids solutions is the aim of current investigations. To our knowledge, no systematic efforts have been made to study ion-ion and ion- amino acid interactions in aqueous alanine solutions. A systematic programme has been undertaken to investigate the role of inter and intra – molecular interactions in aqueous solutions of amino acids with mono and divalent electrolytes.

In the present investigation, we report the experimental studies of u, ρ and η and the results of derived thermodynamic parameters like adiabatic compressibility (βa), intermolecular free length (L_f), Vander Waal's constant (b), internal pressure (Π_i) and solvation number (Sn) of electrolytes in aqueous solutions of alanine at different temperatures. These derived thermodynamic parameters are expected to highlight the role of electrolytes in aqueous alanine solutions and its influence with concentrations and temperature.

EXPERIMENTAL SECTION

All the chemicals used were of AR grade and dried over anhydrous $CaCl_2$ in desiccator's before use. All solutions were prepared in de-ionized and distilled water (degassed by boiling), having specific conductivity 10⁻⁶ Scm⁻¹. The stock solutions [8] of 1M concentration of alanine were prepared by weighing the alanine on a digital balance with an accuracy of $\pm 1 \times 10^{-4}$ g. Solutions of NaCl & MgCl₂ were prepared by mass on the molality concentration scale. Uncertainties in solution concentrations were estimated at $\pm 1 \times 10^{-5}$ mol. kg⁻¹ in calculations. The solutions were kept in the special air tight bottles and were used within 12 hours after preparation to minimize decomposition due to bacterial contamination.

Ultrasonic velocity was measured with a single crystal interferometer (F- 81, Mittal Enterprises, New Delhi) at 2 MHz. The interferometer was calibrated against the ultrasonic velocity of water used at T = 298.15K. The present experimental value is 1497.08 ms⁻¹ which is in good agreement with literature values [12] 1496.69ms⁻¹. Accuracy in the velocity measurement was ± 1.0 ms⁻¹.

The density measurements were performed with precalibrated specific gravity bottle with an accuracy of $\pm 2 \times 10^{-2}$ kg m⁻³. An average of triple measurements was taken into account. Sufficient care was taken to avoid any air bubble entrapment.

Viscosity was measured with Ostwald type viscometer. The flow of time was measured with a digital stop watch capable of registering time accurate to ± 0.1 s. An average of three or four sets of flow of times for each solution was taken for the purpose of calculation of viscosity. The accuracy of the viscosity measurements was $\pm 0.5\%$. Accuracy in experimental temperature was maintained at ± 0.1 K by means of thermostatic water bath.

RESULTS AND DISCUSSION

Ultrasonic velocity (u), density (ρ) and viscosity (η) of the solutions were obtained using the relations;

Ultrasonic velocity	$u = n x \lambda$		(1)
Density	ho = m / V		
Viscosity	$\eta_2 = [t_2 / t_1] x [$	$\rho_2 / \rho_1] \ge \eta_1$	(3)

Where, n & λ are frequency and wavelength; V is the volume of the solution; η_1 & η_2 are the viscosities of the water and solutions; t_1 , t_2 are time of flow water and solution and ρ_1 & ρ_2 are the densities of the water and solution.

From the experimental data of ultrasonic velocity (u), density (ρ) and viscosity (η), the various thermodynamic parameters such as the adiabatic compressibility (β a); intermolecular free length (L_f); Vander Waal's constant (b); internal pressure ($_{\Pi i}$) and solvation number (S_n) for six different concentrations (m) NaCl and MgCl₂ at temperatures 298.15, 303.15 and 308.15K and at 2MHz frequency have been computed using the following relations [9 – 19]

Adiabatic compressibility $\beta a = 1 / u^2$. ρ ------ (4)

Intermolecular free length $L_f = K / u. \rho^{1/2}$ ------ (5)

Vader Waal's constant b = M / ρ [1- (RT / Mu²)] [$\sqrt{1 - Mu^2} / 3RT - 1$] --- (6) Internal pressure $\Pi i = bRT (K\eta / u_{\infty})^{1/2}$. $\rho^{2/3} / M^{7/6}$ ---- (7)

Solvation number $S_n = [n_1 / n_2] \times [1 - (\beta s / \beta s^0)]$ ----- (8)

Where, K is the temperature dependant Jacobson constant, T is the absolute temperature. ρ_0 , ρ and u_o , u are the density and ultrasonic velocity of solvent and solution respectively.

For the amino acid systems (water + alanine + NaCl) and (water + alanine + MgCl₂) ultrasonic velocity (u), density (ρ) and the coefficient of viscosity (η) for different concentrations have been measured at constant frequency of 2 MHz and at different temperatures. The data obtained are used to evaluate βa , L_f, b, Π_i and Sn. The experimental values of u, ρ , η and the calculated values of βa , L_f, b, Π_i and Sn are shown in Table – 1 and 2 for both the systems. The graph plotted for u, βa , L_f, b, Π_i and Sn at different temperatures and various concentrations at 2 MHz frequency are shown in Fig. – 1 to 12.

The ultrasonic velocity (u), for (water + Alanine + NaCl) and (water + Alanine + MgCl₂) amino acid – electrolytes solutions at 2 MHz frequency at different temperatures and various concentrations (m) have been determined using equation (1) and presented in Table – 1 and 2. The variation in ultrasonic velocity of solutions depends on the intermolecular free length (L_f), Erying and Kincaid [12, 16, 17] proposed the increase in ultrasonic velocity with the decrease of intermolecular free length (L_f) and vice versa [12, 16]. Intermolecular free length is a predominant factor, as it determines the ultrasonic velocity in fluid state. Presence of an ion alters the intermolecular free length. Hence ultrasonic velocity of a solution will be different from that of solvent.

Ultrasonic velocity (u) is related to intermolecular free length. As the free length decreases due to the increase in concentration, the ultrasonic velocity has to increase and vice versa. The experimental results of the author support the above statement in both the cases. The variation of ultrasound speeds of systems with molal concentrations of NaCl, MgCl₂ in 1M alanine is given in Tables 1 & 2. From Tables -1 and 2, it is observed that ultrasonic velocity in aqueous alanine solution increases with increase in concentration of solutes NaCl and MgCl₂. consequently ultrasonic velocity of system increases depending on the structural properties of solutes. The solute that increases the ultrasonic velocity is of structure maker type (SM). The value of ultrasonic velocity of NaCl in aqueous alanine is less as compared to the value of MgCl₂ as shown in Fig. – 1 and 2. When NaCl or MgCl₂ is dissolved in solution, the sodium ion (Na⁺) or (Mg⁺⁺) has a structure breaking effect, would disrupt the water structure [16]. This could be followed by structural reorganization leaving the molecules in closely fitting helical cavities. This would cause an increase in closed packed structure as the cohesion between water molecules [18] increases. This makes the liquid medium less compressible and hence the ultrasonic velocity increases above that of pure value.

Density (ρ) of the solution in both the systems increases with concentration of NaCl and MgCl₂. However, density of the solutions is less in aqueous alanine for NaCl as compared to MgCl₂. Viscosity of solutions shows a non-linear behavior in both the systems. Density is a measure of solvent – solvent and ion - solvent interactions [20]. Increase in density with concentration indicates the increase in solvent – solvent and solute - solvent interactions. Whereas decrease in density indicates the decrease in magnitude of solute – solvent and solvent – solvent interactions. Increase in density with concentration is due to the shrinkage in the volume which in turn is due to the presence of solute molecules. That means the increase in density with concentration indicates the decrease in density with concentration indicates the solvent. It is clear that solvent – solvent interactions bring about a bonding, probably hydrogen bonding between them. So, size of the resultant molecule increases and there will be decrease in density. In fact the values of density and viscosity of any system vary with increase in concentration of solutions. The change in structure of solvent or solutions as a result of hydrogen bond formation or dissociation or hydrophobic (structure – breaking) or hydrophilic (structure – forming) nature of solute. Thus hydrogen bond forming or dissociating properties can be correlated with change in density and viscosity.

When an ion is added to a solvent, it attracts certain solvent molecules toward itself by wrenching the molecules from bulk of the solvent due to the forces of electrostriction. Due to this, the available solvent molecule for the next incoming ion gets decreased. This process is known as compression. Every solvent is having a limit for compression called the limiting compressibility value. The compressibility of a solvent is higher than that of a solution and it decreases with increase in concentration. The adiabatic compressibility is calculated using equation (4). The calculated values of (β_a) are presented in Tables – 1 and 2. The graph for (β_a) versus molality (m) of NaCl and MgCl₂ are plotted and shown in Fig. – 3 and 4 respectively. In the present case adiabatic compressibility (β_a) decreases with increase in concentrations of NaCl and MgCl₂. This is because as the concentration of solute increases, a larger portion of water molecules are electrostatic and the amount of bulk water decreases causing the compressibility to decrease. In the present study d β / dc is negative which indicates the electrostriction of water

molecules. Hydrophilic [23, 25] solutes often show negative compressibility due to the ordering that is induced by them in water structure [12]. The electrolytes like NaCl, KCl, MgCl₂, etc. interact with charged centre of alanine. In alanine, containing methyl group, the methyl group tightens the water molecules around itself and hence the values of adiabatic compressibility for alanine are higher than those of glycine. It is found that the adiabatic compressibility adecreases with increasing the concentration of the solute as well as temperature. The increase in velocity and decrease in compressibility were attributed to the formation of hydrogen bonds between solute and solvent molecules [21]. Decrease in adiabatic compressibility with concentration supported that the solvent molecules are fully compressed due to electrical forces of solute molecules [22].

The values of intermolecular free length for (water + alanine + NaCl) and (water + alanine + MgCl₂) systems have been calculated using equation (5). Increase in concentrations leads to decrease in gap between two species which is referred by intermolecular free length (L_f). With the increase in concentration the intermolecular free length L_f decreases & this trend is clearly observed which shown in Figs- 5 and 6. Intermolecular free length (L_f) is a predominant factor in determining the variations of ultrasonic velocity in fluids and in their solutions. From Tables – 1 and 2, it has been observed that, in the present investigation, intermolecular free length decrease linearly on increasing concentrations of solutes. The decreased in L_f with increase of solutes concentrations in solution indicates that there are significant interactions between solute - solute and solvent suggesting the structure promoting behavior of solutes. In (water + alanine + NaCl) & (water + alanine + MgCl₂) systems, ultrasonic velocity increase with concentrations indicates stronger the intermolecular forces in the solution. This gives increase in closed packed structure of aqueous alanine i.e. enhancement of the closed structure. This provides the cohesion between alanine and water molecules increases. The reduction in degree of dissociation among the liquid molecules of the mixtures. Thus the inter-molecular distance decreases with concentrations. The decrease in free length may due to the gain of dipolar association, making up of hydrogen bonds in the molecules of the liquid mixtures.

Using the experimental data of ultrasonic velocities, densities and viscosities for (water + alanine + NaCl) and (water + alanine + MgCl₂) systems, the values of Vander Waal's constant (b) have been calculated using equation (6) and it has been presented in the Tables – 1 and 2. It is observed that the Vander Waal's constant (b) shows a continuous increasing behavior with concentrations of NaCl and MgCl₂ in (water + alanine + NaCl) and (water + alanine + MgCl₂) systems as per Fig. – 7 and 8. This is because the dissociation of a closed packing of the molecules inside the shell [22]. The values of Vander Waal's constant (b) for NaCl are slightly less than the values for MgCl₂ because of divalent nature of MgCl₂.

Internal pressure (Π i) with different concentrations and temperatures for (water + alanine + NaCl) and (water + alanine + MgCl₂) systems has been obtained using experimental and calculated data by standard relation (7). From Tables -1 and 2, it is observed that internal pressure (Π i) shows increase and decrease non-linear nature as per Fig. - 9 and 10. This shows the orientation of the solvent molecules around the ions, which may be due to the influence of electrostatic field of ions. This means the solution becomes less compressible. It indicates the associating tendency of the molecules in the amino acid solutions. Conversely, the reduction in internal pressure shows dissociating tendency of molecules in the solution.

Solvation number (S_n) depends on the number of moles of the solutes and solvent molecules. For systems (water + alanine + NaCl) and (water + alanine + MgCl₂), the values of solvation number have been calculated using Relation (8) and are given in the tables 1 and 2. It is clearly observed that solvation number (S_n) increases with increase in concentrations of NaCl and MgCl₂ whereas, decrease in the values of solvation number as temperature increases. This suggests significantly strong interaction in both the systems. The increase in (S_n) supports structure maker (SM) tendency of electrolytes [20, 21]. But decrease in the value of (S_n) at high temperature favors presence of solute – solvent interactions. Interaction of MgCl₂ with aqueous alanine is stronger as compared to interaction of NaCl with aqueous alanine due to divalent nature of MaCl₂ as depicted in the Fig. – 11 and 12.

CONCLUSION

Ultrasonic velocity, density viscosity have been measured for NaCl and MgCl₂ in aqueous alanine solution at 298.15, 303.15, and 308.15K. The variation in ultrasonic velocity, density and viscosity and other related thermodynamic parameters such as adiabatic compressibility, intermolecular free length, Vander Waal's constant (b); internal pressure (π) and solvation number (S_n) of NaCl & MgCl₂ at various concentrations and temperatures in aqueous alanine shows the variation to be increase and decrease non-linear. Consequently ultrasonic velocity of system increases depending on the structural properties of solutes. It is clear that solutes causing electrostriction lead to decreases in the compressibility of the solution. Hydrophilic solutes often show negative compressibility, due to ordering that is induced by them in water structure. The solute that increases the ultrasonic velocity is of structure maker. The non linearity confirms the presence of solute-solvent, ion-ion, ion-amino acid interactions. The observed

molecular interaction, complex formation, hydrogen bond formation are responsible for the hetero-molecular interaction in the liquid system. This provides useful information about inter and intra molecular interactions of the mixture as existing in the liquid system.



Fig. 1 – Plots of ultrasonic velocity (u) versus concentration (m) of NaCl in aqueous alanine solution at 298.15, 303.15, and 308.15K





Fig. 3 – Plots of adiabatic compressibility (β_a) versus concentration (m) of NaCl in aqueous alanine solution at 298.15, 303.15, and 308.15K

 $\label{eq:generalized} \begin{array}{l} Fig.~4-Plots~of~adiabatic~compressibility~(\beta_a)~versus~concentration~(m)~of~MgCl_2~in~aqueous~alanine~solution\\ at~298.15,~303.15,~and~308.15K \end{array}$



 $\label{eq:Fig.5-Plots} Fig. 5-Plots of intermolecular free length (L_f) versus concentration (m) of NaCl in aqueous alanine solution at 298.15, 303.15, and 308.15K$



Fig. 7 – Plots of Vander Waal's constant (b) versus concentration (m) of NaCl in aqueous alanine solution at 298.15, 303.15, and 308.15K





Fig. 9 – Plots of internal pressure (π_i) versus concentration (m) of NaCl in aqueous alanine solution at 298.15, 303.15, and 308.15K

Fig. 10 – Plots of internal pressure (π_i) versus concentration of MgCl₂ in aqueous alanine solution at 298.15, 303.15, and 308.15K



Fig. 11 – Plots of solvation number (S_n) versus concentration (m) of NaCl in aqueous alanine solution at 298.15, 303.15, and 308.15K

 $\label{eq:Fig.12-Plots} Fig. 12-Plots of solvation number (S_n) versus concentration of MgCl_2 in aqueous alanine solution at 298.15, 303.15, and 308.15K$

Table - 1 : Variation of thermodynamic parameters for (Water + Alanine + NaCl) system at								
different concentration (m) and temperatures.								
m	u	ρ	η	β x 10 ⁻¹⁰	L _f	b x 10 ⁻⁵	л _i х 10 ⁻⁵	Sn
mol.Kg ⁻¹	m/s	Kg/m ³	Nsm-2	$m^2 N^{-1}$	^{0}A	$m^3 mol^{-1}$	Nm ⁻²	
				298 .15 K				
0	1543.6	1050	1.23235	3.99706	0.41121	18.78955	398.5	0
0.034	1578.64	1056.4	1.24818	3.79844	0.40086	19.1537	401.8	16.8
0.071	1585.8	1063.2	1.24963	3.74014	0.39777	19.16964	402.9	43
0.106	1604.9	1072	1.26167	3.62167	0.39142	19.29597	406.2	94.1
0.14	1607.46	1080.8	1.29934	3.58075	0.3892	19.22114	413.6	139
0.175	1618.12	1085.2	1.2923	3.5194	0.38585	19.32106	412.8	199
				303.15 K				
0	1570.32	1048	1.1377	3.86957	0.40828	18.99361	388.8	0
0.034	1593.04	1054.4	1.1388	3.73716	0.40124	19.20479	389.7	11.4
0.071	1594.1	1062	1.13932	3.70548	0.39953	19.13167	390.9	28.2
0.106	1608.4	1071.2	1.17069	3.60862	0.39428	19.19161	397.7	67.2
0.14	1610.48	1078.8	1.19471	3.57395	0.39238	19.13254	402.8	102
0.175	1622.93	1084.4	1.19861	3.50115	0.38836	19.23166	404	158
				308.15 K				
0	1584	1046.8	1.03922	3.80738	0.40865	19.02483	377.4	0
0.034	1595.36	1052.8	1.04697	3.73196	0.40458	19.10439	379.5	6.18
0.071	1603.4	1060	1.05456	3.66953	0.40118	19.1223	381.8	24.4
0.106	1611.6	1067.6	1.08303	3.60643	0.39772	19.13683	387.9	53.2
0.14	1618.8	1076	1.09493	3.54651	0.3944	19.12406	391.3	91.8
0.175	1628.52	1082.4	1.10405	3.48358	0.39088	19.17559	393.7	142

Table - 2 : Variation of thermodynamic parameters for (Water + Alanine + MgCl ₂) system at								
different concentration (m) and temperatures.								
m	u	ρ	η	β x 10 ⁻¹⁰	L _f	b x 10 ⁻⁵	л _i х 10 ⁻⁵	Sn
mol.Kg ⁻¹	m/s	Kg/m ³	Nsm-2	$m^2 N^{-1}$	^{0}A	m ³ mol ⁻¹	Nm ⁻²	
				298.15 K				
0	1543.6	1050	1.03069	3.99706	0.41121	18.78955	398.5	0
0.034	1566.24	1065.6	1.25542	3.82551	0.40229	18.9644	401.8	14.5
0.071	1612.84	1070	1.55957	3.59521	0.38999	19.62676	415.2	67.1
0.106	1604.9	1086.3	1.60581	3.52134	0.38559	19.5724	429.1	119
0.14	1618.64	1100	1.75742	3.46982	0.38313	19.52942	448.2	176
0.175	1636.22	1109.2	1.87766	3.36749	0.37744	19.75322	464.6	262
				303.15 K				
0	1570.32	1048	0.79285	3.86957	0.40828	18.99361	388.7	0
0.034	1583.5	1062.4	1.22933	3.75384	0.40213	19.07216	389.7	9.95
0.071	1584.16	1068.4	1.45347	3.72965	0.40083	19.15054	409.3	24
0.106	1618.4	1086	1.55373	3.51559	0.38916	19.43403	421.1	91.1
0.14	1624.8	1098	2.13041	3.44983	0.3855	19.47636	438	144
0.175	1628.78	1108.4	2.12168	3.40078	0.38275	19.51335	458.2	201
				308.15 K				
0	1584	1046.8	0.91275	3.80738	0.40865	19.02483	380.2	0
0.034	1602.7	1058.4	1.04792	3.67829	0.40166	19.21892	378.4	11.5
0.071	1604.8	1065.6	1.0519	3.64388	0.39978	19.29307	399.1	28.9
0.106	1623.6	1084	1.2718	3.49955	0.39178	19.37283	411.3	81
0.14	1631	1097.2	1.16509	3.42615	0.38765	19.43512	428.8	134
0.175	1645.7	1105.6	1.93509	3.33965	0.38272	19.60519	446.2	205

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