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Volumetric, viscometric and ultrasonic studies of some amino acids in aqueous 0.02 M $LiClO_4.3H_2O$ solutions at 298.15K

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ABSTRACT

The Density viscosity and ultrasonic velocity measurement were carried out in aqueous solution of glycine, L-alanine, L-valine, L-Leucine and L-phenyl alanine in 0.02 M $LiClO_4.3H_2O$ at 298.15 K. The values of apparent molar volume, limiting apparent molar volume, adiabatic compressibility, apparent molar adiabatic compressibility have been evaluated from density and sound speed data. These values are used for calculatating the number of water molecule hydrated(n_H) to the amino acids. The viscosity data have been analyzed by using Jones-Dole equation. Transfer volumes at infinite dilution from water to aqueous lithium per chlorate have been also calculated . Transfer parameter have been interpreted in terms of solute-cosolute interaction on the basis of co-sphere overlap model .All the results were interpreted in the light of ion -ion and ion – solvent interactions and of structural effect of solutes in solutions.

Key words: Density, Jones-Dole equation, transfer function, co-sphere overlap model, interaction coefficients.

INTRODUCTION

Amino acids are the fundamental substances for building proteins. Ultrasonic studies of some amino acids in 0.02 M $LiClO_4.3H_2O$ are of great important to obtain about various types of interactions in solutions and have been used to understand the factors responsible for the thermodynamic stability of proteins and their unfolding behavior. Knowledge of various solute-solvent and solute-solute interactions this interaction is very important to understand various fundamental phenomenons like stability of proteins, folding/ unfolding processes, denaturation of proteins aggregation, several biochemical processes such as protein dehydration in aqueous solutions[1-4]. The interaction of amino acids with electrolyte in the aqueous solutions and there temperature dependence of these interactions also play a important role in understanding nature of action of bioactive molecule, the thermodynamic behavior of biochemical process in the body and the stability of the organism found in submarine hot springs[5-7]. Extensive ultrasonic data have been reported at 298.15K .We understand that much more relevance and significance can be achieved by studied compounds of biological importance at temperature close to physiological temperature 308.15K being close to the optimum temperature of several living species offers a better choice for such experimentation.

Lithium (Beryllium and aluminum) are not essential element but they can interfere with biochemical process in vivo by interacting with phosphate containing molecule. One interesting possibility is the involvement of lithium with phosphate compounds, this can occur at two levels.

1. Lithium could interact with the phosphate compounds of Na^+/K^+ or Mg^{2+}/Ca^{2+} pumps.
2. It could interact with the inositol phosphate.

Lithium is of course a major psychotherapeutic drug and it's action may in part to be based on these possibilities[8]. Definitive evidence as to mode of action of lithium is currently unavailable[8].

Therefore thermodynamic is study of amino acids in presence of Lithium salt is undertaken. The results obtained may be useful in order to understand mode of acting of Lithium.

MATERIAL

Five amino acids namely glycine, L-alanine, L-valine, L-Leucine and L-phenyl alanine of highest purity were obtained from Sigma chemicals Co. Amino acids were dried in vacuum oven for 24 hrs on kept over P_2O_5 in vacuum desiccators. Lithum per chloate (LOBA Chemie) were used without any purification. All the solutions were prepared on the molarity basis. The samples were weighted on a mettler balance having accuracy of 0.01mg. Water used to prepare solutions was obtained by distilling deionised water over alkaline $KMnO_4$ and it was thoroughly degassed prior to its use. The specific conductance of the water used was less than 0.055×10^{-6} s cm^{-1}

METHODS

The densities of the solutions were measured using a single capillary pycnometer made up of borosil glass with a bulb of total volume of 8 cm^3 and capillary with internal diameter of 0.1cm was chosen[9] for the present work. The details pertaining to calibration experimental set up and operational procedure have been previously described[9]. An average of triplicate measurement was taken in to account. The reproducibility of density measurement was + - 3 X 10^{-5} g cm^{-3}. Viscosity was measured by means of suspended-level Ubbelhood viscometer with flow time approximately 300s for distilled water at 298.15 K. The time of reflux was measured with a stop watch capable of recording + 0.1 s .An average of three or four sets of flow times for each fluids was taken for the purpose of calculation of viscosity .The reproducibility of viscosity results was found to be within $+0.003 \times 10^{-2}$ Nm^{-2} Ultrasonic velocities for the solution were measured using M-81 model, 2 MHz. Ultrasonic interferometer (Mittal Enterprises, New Delhi) with a reproducibility of +- 0.4 ms^{-1} at 298.15K The temperature was constantly maintain by controlled temperature water bath (Gemini scientific instruments, Madras) having accuracy of ± 0.01 ^{0}C.

RESULTS AND DISCUSSION

In the present study, densities of these solutions are increasing with increase in concentration of amino acids, but decreases with increase in temperature.

The viscosities of the solution were determined by using the following relation.

$$\frac{\eta}{\rho} = \left(at = \frac{b}{t} \right) \qquad ...(1)$$

Where t is the efflux time, a and b are viscometer constants. The measured values of viscosities are accurate to within 0.001mPas. Viscosities of these solutions are also increasing with increase in concentration of amino acids but decrease with increase in temperature.

It is observed from Table 1 that the ultrasonic velocity increases with the increase in concentration of amino aids in 0.02 M $LiClO_4.3H_2O$. The increase in ultrasonic velocity in any solution indicates the maximum association among the molecules present in solution. In the present study was observed with steady increase in ultrasonic velocity as concentration, this is due to maximum possible association in aqueous $LiClO_4$ solutions. Same behavior is observed in the acoustical properties of fructose and maltose in water, aqueous NH_4Cl solution[10], ultrasonic studies of some amino acids in binary aqueous $MgCl_2$ $2H_2O$[9] and in aqueous 1,4 dioxane[26] solutions.

The ultrasonic velocity of amino acids in 0.02 M $LiClO_4.3H_2O$ are found to increase with increasing temperature. This behavior is similar to that of pure water, where ultrasonic velocity increases with temperature. As the temperature increase, the hydrogen bonds among the water molecules break and more monomeric water molecules are formed. These broken water,

Tables 1: Densities(ρ), Ultrasonic velocities(U), adiabatic compressibility (β_{ad}) apparent molar volume (V_{ϕ},), limiting apparent molar volume (V_{ϕ}) and viscosity (η) partial molar volumes of transfer (ΔV^0_{ϕ}), experimental slope(Sv) and B-coefficient of some amino acid in 0.02 M $LiClO_4.3H_2O$ solutions at 298.15K.

C	ρ	U	β_{ad}	η	Vϕ	V^0_{ϕ}	Sv	ΔV^0_{ϕ}	B
Glycine									
0.00	1.01083	1528	4.2372	0.9473	-	44.61	-1.43	1.99	0.146
0.025	1.01157	1537	4.1846	0.9584	44.35	-	-	-	-
0.075	1.01305	1555	4.0823	0.9654	44.25	-	-	-	-
0.125	1.01455	1568	4.0090	0.9711	44.16	-	-	-	-
0.175	1.01605	1584	3.9226	0.9767	44.02	-	-	-	-
0.225	1.01750	1600	3.8388	0.9810	43.89	-	-	-	-
L-Alanine									
0.00	1.01083	1528	4.2372	0.9473	-	60.80	-1.56	0.31	0.253
0.025	1.01149	1538	4.1795	0.9852	60.52	-	-	-	-
0.075	1.01282	1558	4.0675	0.9907	60.40	-	-	-	-
0.125	1.01416	1578	3.9599	0.9963	60.29	-	-	-	-
0.175	1.01551	1592	3.8853	1.0018	60.14	-	-	-	-
0.225	1.01688	1610	3.7938	1.00141	60.03	-	-	-	-
L-valine									
0.00	1.01083	1528	4.2372	0.9473	-	89.98	-1.48	-1.00	0.423
0.025	1.01147	1540	4.1687	1.0226	89.72	-	-	-	-
0.075	1.01276	1564	4.0366	1.0281	89.60	-	-	-	-
0.125	1.01407	1586	3.9204	1.0336	89.50	-	-	-	-
0.175	1.01538	1608	3.8089	1.0391	89.38	-	-	-	-
0.225	1.01672	1628	3.7110	1.0698	89.24	-	-	-	-
L-leucine									
0.00	1.01083	1528	4.2372	0.9473	-	106.87	-1.43	-0.90	0.578
0.025	1.01138	1540	4.1691	0.9280	106.60	-	-	-	-
0.075	1.01249	1564	4.0377	1.0982	106.52	-	-	-	-
0.125	1.01361	1589	3.9073	1.1037	106.42	-	-	-	-
0.175	1.01474	1606	3.8208	1.1134	106.28	-	-	-	-
0.225	1.01589	1626	3.7237	1.1268	106.14	-	-	-	-
L-phenyl alanine									
0.00	1.01083	1528	4.2372	0.9473	-	119.74	-1.34	-1.58	0.581
0.025	1.01189	1541	4.1616	1,0350	119.50	-	-	-	-
0.075	1.01402	1565	4.0265	1.0414	119.40	-	-	-	-
0.125	1.01615	1588	3.9025	1.0563	119.28	-	-	-	-
0.175	1.01830	1608	3.7980	1.0670	119.18	-	-	-	-
0.225	1.02045	1632	3.6793	1.1114	119.08	-	-	-	-

molecules enter the vacant space present in the cage like water structures and thus get "trapped". As a result the number of close-packed water structure increase with the increase in temperature. This increase in close packed water structure forms the material medium for the propagation of ultrasonic waves. Thus the ultrasonic velocity increases with the increase in temperature for pure water as well as aqueous $LiClO_4.3H_2O$. In addition, in aqueous $LiClO_4.3H_2O$ solution, the polar perchlorate molecules form more compact structure with the solvent water molecules. This effect favors a decrease in compressibility. With increase in temperature, however, the mean distance between the molecules tends to increase with a corresponding increase in compressibility. These two opposing tendencies results in a net decrease in compressibility and hence increase in ultrasonic velocity.

When an ion is added to a solvent, it attracts certain solvent molecules towards itself by wrenching the molecules from bulk of the solvent due to the force of electrostriction. Because of this, the available solvent molecule for the next incoming ion get decreased, this process is called as compression. Every solvent has a limit for the compression called the limiting compressibility value. The compressibility of a solvent is higher than that of a solution and it decrease with increase in concentration of the solution. With increase in ionic solute concentration, their electrostrictive forces cause the water structure to break and the solute surrounded water molecule are more compactly packed. This hydration effect in turn, results in reducing the compressibility with increasing ionic solute concentrations.

Adiabatic compressibility β_{ad} ($N^{-1}m^2$) were calculated from both density and sound velocity values using the following equation.

$$\beta_{ad} = 100/u^2\rho \qquad ...(2)$$

Where u=Sound velocity of solution (m/sec) and r = density of the solution at the same temperature (Kg/m^3).

In the present study adiabatic compressibility decrease with the increase in temperature and the concentration of amino acids in 0.02 M $LiClO_4.3H_2O$. This confirms the presence of solute solvent interaction through dipole-dipole interaction of the ClO_4 ion of $LiClO_4$ with the surrounding water molecule[201]. The variation of adiabatic compressibility of amino acids with respect to temperature are shown in figures. The apparent molar volumes (V_ϕ) was calculated from the density data using well known expression.

$$V_\phi = \frac{1000\,(\rho_0 - \rho_s)}{C.\rho_0} + \frac{M}{\rho_0} \qquad ...(3)$$

Where $\rho_0$and ρ_s are densities of solutions and solvent respectively. C is molarity and M is the molar mass of solute. The resulting values of apparent molar volumes (V_ϕ) and the molar concentration (M) of the amino acids in 0.02M $LiClO_4.3H_2O$ at 298.15 K are reported in Table.1. Comparison with earlier results (A Pal et al. 2005) shows that Values of V_f increase with increase in temperature in 0.02 M $LiClO_4.3H_2O$. It is also found that V_ϕ increases linearly with increase in size of alkyl side chain of the amino acids in 0.02 M $LiClO_4.3H_2O$. It indicates that the solute-solvent interaction increases with increase in size of the alkyl side chain of amino acids and with the temperature. It is accordance with earlier results[10].

The variation of apparent molar volumes with the square root of molar concentration can be represented by Mason's equation[11].

$$V_\phi = V_\phi^0 + Sv.C^{3/2} \qquad ...(4)$$

Where v^0_ϕ is the limiting value of the apparent molar volume (equal to the partial molar volume at infinite dilutions) and S_v is the experimental slope. The Values of and S_v obtained by least square fitting of the V_ϕ values to equation 3 for various amino acids in 0.02 M $LiClO_4.3H_2O$ at different temperatures, which are reported in Table. 1.

Limiting apparent molar Volume (v^0_ϕ) and the experimental slope have been employed to indicate the type of interaction [12-13]. The effect of

temperature on v^0_ϕ has been interpreted in terms of the solute-solvent interactions while that on experimental slope in terms of solute-solute and solute-solvent interaction. The positive value of S_v suggesting that the added electrolyte behave as a structure maker in the solvent while negative value of S_v suggest the structure breaker capacity in the solvent.

All the amino acids studied have positive value in binary aqueous solution of 0.02 M $LiClO_4.3H_2O$. It is also found that increase linearly with the size of alkyl side chain of amino acids and increases with increase in temperature. It indicates that the co-solute-solvent interaction increase both on increasing temperature and the size of alkyl side chain of amino acids [10]

Since S_v is related to solute-solute interaction. It is evident from Table 1. that the values of the slope S_v for all amino acids in 0.02 M $LiClO_4.3H_2O$ at different temperatures are negative suggesting weak solute-solute interaction in the system. The experimental slope S_v increases with increase in temperature, suggest that more and more solute is accommodated in void space left in packing the large associated solvent molecule and such enhance the structure of the solvent. However S_v is found to decrease with increase in temperature of L-valine in 0.02 M $LiClO_4.3H_2O$, suggesting the decrease in solute-solute interaction with the rise in temperature indicating structure breaking effect of L-valine in 0.02 M $LiClO_4.3H_2O$. It is found from table 1. that the values of S_v are more negative for L-alanine and L-valine at different temperature when compared with the values of S_v of other amino acids.

The partial molar volumes of transfer (ΔV^0_ϕ) from water to aqueous 0.02 M $LiClO_4.3H_2O$ have been calculated by using the equation 4 . Banipal et al reported values of in water were used to calculate .

$$\Delta V^0_\phi = V^0_\phi(\text{in } LiClO_4.\ 3H_2O) - V^0_\phi(\text{in water}) \quad ...(5)$$

The value of ΔV^0_ϕ at 298.15 are illustrated in table 1. The more positive value for glycine and L-alanine indicates the dominance of the charged group NH^+_3 and COO^- while negative values in case of L-valine L- leucine and L-phenyl alanine indicates the effect of hydrophobic parts. That is the interactions between the $LiClO_4.3H_2O$ and zwitter ionic center of amino acids increase with increase in temperature. For L-valine the interaction between non-polar group of L-valine and $LiClO_4.3H_2O$ are predominant. The overall effect is that the charged end group of glycine and L-alanine influence electrostatically the surrounding water molecule the so called electrostriction (A Pal 2005). In other word, the hydration co-sphere of COO^-. Which are more hydrated than that of $LiClO_4.3H_2O$ will be affected to greater extent than the later. The positive values results that the dehydration of solute and co-solute occurs more in case of glycine and L-alanine.

Now to explain partial molar volume data, different models have been used. Franks et. al[14] have shown that partial molar volume at infinite dilution of a non electrolyte is a combination of two factors by the following equation.

$$\Delta V^0_\phi = V_{int} + V_s \quad ...(6)$$

Where is V_{int} the intrinsic molar volume of the non-hydrated solute V_s is the contribution due to the interaction of the solute with water. Some workers [15-16] have suggested that the V_{int} is made of the following type of contribution.

$$V_{int} = V_{v,w} + V_{void} \quad ...(7)$$

Where V_{V1} is the Van der walls volume[17-18] and V_{void} is the volume associated with void or empty space. For electrolyte zwitter ionic solutes, this equation was modified by Shahidi et. al[15]. to find contribution of one molecule to partial molar volume of a hydrophobic solutes as

$$V^0_\phi = V_{v,w} + V_{void} - V_{shrinkage} \quad ...(8)$$

Where $V_{shrinkage}$ is the volume due to shrinkage this is due to interaction of hydrogen bonding sites with water molecules. Assuming thatV_{v1w} and V_{void} have the same magnitude in water and aqueous 0.02M $LiClO_4.3H_2O$ positive ΔV^0_ϕ

values of glycine and L-alanine might arise from the decrease in $V_{shrinkage}$ in 0.02m $LiClO_4.3H_2O$. The interaction $LiClO_4.3H_2O$ with the zwitter ionic center of amino acids (glycine and L-alanine) reduces the effect electrostriction of water, thereby causing a decrease in $V_{shrinkage}$. In other words some water molecule may be released as bulk water in presence of $LiClO_4.3H_2O$. It brings about the increase in volume of the solvent [19] there by the reducing the strong interactions between amino acids and water. This result in positive volume of transfer from water to aqueous $LiClO_4.3H_2O$ solution observed in case of glycine and L-alanine. Thus a positive for glycine and L-alanine results from the decreased effect of LiClO4.3H2O and glycine or L-alanine on water structure which arises due to glycine or L-alanine-$LiClO_4.3H_2O$. The negative values of L-valine, L-Leucine and L-phenyl alanine might arise from the smaller decrease in $V_{shrinkage}$ in $LiClO_4.3H_2O$ solution.

Values can further be rationalized by co-sphere overlap model developed by Gurney [20]and Frank and Evans[21]. According these properties of water molecules in the hydration co sphere depend on the nature of solute molecule. When two solute particles come close enough such that their co-sphere over lap. Some of the co sphere material is displaced and this is accompanied by changes in the thermodynamic parameters.

The interaction between $LiClO_4.3H_2O$ and amino acids can be classified as follows 1) hydrophilic–ionic interaction occurring between zwitter ionic centers of amino acids and dipolar parts of $LiClO_4.3H_2O$ 2) Hydrophilic- hydrophobic interaction occurring between non polar parts of amino acids and hydrophobic parts of $LiClO_4.3H_2O$.

According to the co sphere model in terms of solute-cosolute interactions, hydrophilic-ionic group interaction contributes positively, whereas hydrophilic-hydrophobic group interaction contributes negatively to the values. In case of glycine and L-alinine, the former type of interactions are predominant over the latter and for L-valine, L-leucine, L-phenyl alanine hydrophilic-hydrophobic group interaction are dominating over the hydrophilic-ionic group interaction. It may noted from A. Pal and S. Kumar's[22] findings that values of L alanine are less than those of glycine and L-valine in solutions. This is in line with the earlier conclusion drawn on the basis of volume of shrinkage that varies solute-co-solute interactions occur in these systems which contribute to different extents, depending on the particular amino acids solution. The overall effect is that the solute – co solute interactions are predominant over the solute-solvent interaction as obtained in glycine and L-alanine .

The viscosity values were used to determine the relative viscosities (v^0_ϕ) using the relation

$$\eta_r = \frac{\eta_s}{\eta_0} \qquad ...(9)$$

where η_0 = viscosities of solvent
η_s = viscosities of solution

In the present study the relative viscosity goes on increasing with increase in concentration of amino acids and temperature in presence of 0.02M $LiClO_4.3H_2O$. The experimental results of relative viscosities (h_r) for the amino acids in aqueous 0.02M $LiClO_4.3H_2O$ have been analyzed by using Jones-Dole[23] equation in the following form in order to evaluate β-coefficient

$$\eta_r = \frac{\eta_s}{\eta_0} = 1 + Bc \qquad ...(10)$$

where η_0 and η_s are the viscosities of solution and solvent respectively, B is a constant and characteristic of solute-solvent interactions and C is concentration in moles per unit volume. The values of B-coefficient obtained by the least square method are presented in Table 1. It has been suggested [24]that the positive B-value indicate structure making and negative B-value indicate structure breaking tendencies of added electrolytes. Recently it has been emphasized [25-26]that the sign of the temperature coefficient of the viscosity B-coefficient (dB/dT) is better criteria [26-28] than the magnitude of B it self for determining the ion-solvent interactions. According to this, electrolyte which are structure maker will have negative value (dB/dT) of and those will have positive (dB/dT) are structure breaker.

The B-coefficient values are positive and increase from glycine to L-phenyl alanine since B-coefficient are sensitive to size and shape of the molecule and structural effect induced by solute-solvent interaction [29]the increase in relative magnitude of B-coefficient with the increase in non-polar alkyl side chain and molar mass are suggestive of the structural changes from glycine to L-phenyl alanine B-coefficient of amino acid in 0.02M $LiClO_4.3H_2O$ are positive indicating that the ion-solvent interactions are strong. Also in the present study the value of are negative showing that $LiClO_4.3H_2O$ has structure making capacity.

Conclusion

In this paper a systematic study of volumetric and viscometric of glycine, L-alanine, L-valine, L-Leucine and L-phenyl alanine in 0.02M, $LiClO_4.3H_2O$ solutions at 298.15 K has been carried out at different concentration of amino acids. The experimental volumetric and viscometric give valuable information regarding the solute-solvent interactions in aqueous 0.02M, $LiClO_4.3H_2O$. It is also found that $V^0\phi$ increase linearly with the size of alkyl side chain of amino acids and increases with increase in temperature. It indicates that the co-solute-solvent interaction increase both on increasing temperature and the size of alkyl side chain of amino acids. The slope S_v for all amino acids in 0.02 M $LiClO_4.3H_2O$ at different temperatures are negative suggesting weak solute-solute interaction in the system . . The more positive value for glycine and L-alanine indicates the dominance of the charged group NH^+_3 and COO^- while negative $\Delta V^0\phi$ values in case of L-valine L- leucine and L-phenyl alanine indicates the effect of hydrophobic parts. B-coefficient of amino acid in 0.02M $LiClO_4.3H_2O$ are positive indicating that the ion-solvent interactions are strong. Also in the present study the value of (dB/dT) are negative showing that $LiClO_4.3H_2O$ has structure making capacity.

These conclusion give scope for further studies on the thermodynamic properties of the system.

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