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Ultrasonic, volumetric and transport studies of molecular interaction of some amino acids in aqueous dextrose solutions at 303.15 K

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Abstract
Apparent molar compressibilities (φ_k) and apparent molar volume (φ_v) of L-serine and L-alanine in aqueous and aqueous solutions of dextrose at different concentrations have been determined at 303.15K from precise density, ultrasonic velocity and time flow measurements. From these data limiting apparent molar compressibilities (φ_k^o), limiting apparent molar volumes (φ_v^o), and their constants (S_k, Sv), viscosity A & B-coefficients and the corresponding transfer parameters ($\Delta\varphi_k^o$ and $\Delta\varphi_v^o$), and hydration numbers (n_H) have been calculated. The results are interpreted in terms of solute-solute and solute-solvent interaction in these systems. It has also been observed that L-alanine act as a more structure maker than L-serine.

Keywords: apparent molar compressibility, apparent molar volume, transfer parameter, viscosity b-coefficient and hydration number

1. Introduction

In recent years, the studies of acoustical properties of amino acids in aqueous mixed saccharide solutions have been found to be useful in understanding the specific ion-solute and ion-solvent interaction in solutions. A number of researchers have employed the ultrasonic technique to evaluate the thermodynamic properties of amino acids, peptides and proteins [1, 2]. Ultrasonic studies on the physicochemical properties of biomolecules like vitamins, amino acids, sugars and drugs in aqueous solution provide useful information, which is important to understand the complex mechanism of molecular interactions [3]. Protein molecules are very large molecules. It can be broken down into smaller units by hydrolysis. These small units the monomers of proteins are called as amino acids. Interactions are mainly those between the amino acids and the solvent ions. Most of these interactions, such as hydrogen bonding and electrostatic interactions have non-covalent nature [4]. In order to get a better understanding of these interactions, various low molecular weight model compounds, such as amino acids and peptides, have been studied because of the complexities of proteins and infeasibility of direct thermodynamic studies. Proteins, which have enzymatic, structural, and regulatory functions, carry out the vast majority of all biological processes in cells.

Amino acids also play a significant role in metabolism and in many neurochemical response mechanisms, such as memory, appetite control, and pain transmission. They are also used as food additives and have many applications in the pharmaceutical industries [5]. Carbohydrates located at cell surfaces are very important as receptors with regards to the bioactive structure of hormones, enzymes, viruses, antibodies, and so forth. Therefore, in living systems, carbohydrate and protein interactions play an important role in various biochemical processes.

The volumetric and compressibility studies of amino acids in aqueous saccharides solutions have been employed to understand the nature of interactions operative in solutions. The accurate measurement of density, viscosity, ultrasonic velocity and hence the derived parameters will give significant information regarding the state of affairs in a solution [6]. An exhaustive literature review, only some limited contributions have been given recently to the study of amino acids on the stability of sugar in aqueous solutions. Hence, the present investigation focused on interactions between two amino acids (L-serine, and L-alanine) with 0, 0.5 and 1M aqueous dextrose solutions, via density, viscosity, and ultrasonic velocity. Using these data, the adiabatic compressibility, molar hydration number, apparent molar compressibility, apparent molar volume, limiting apparent molar compressibility, limiting apparent molar volume, and their constants, transfer adiabatic compressibility, transfer volume and viscosity A and B coefficient of Jones-Dole equation as a function of solute concentration and temperature at 303.15 K have been evaluated with a view to investigating the solute - solvent and solute-solute interactions and also their nature (hydrophilic - hydrophilic, hydrophilic - hydrophobic and hydrophobic - hydrophobic), occurring in the ternary system (amino acids + saccharides + water) and its effect with constant temperature.

2. Materials and Methods

Analytical reagent (AR) and spectroscopic reagent (SR) grades that minimum assay of 99.9% of L-serine, L-alanine and dextrose was obtained from Sd fine chemicals, India, which are used as such without further purification. Doubly distilled degassed water was used for the preparation of all solutions. Solutions of dextrose (0.5 and1.0 mol.dm^{-3}) were prepared by volume and used on the day they were prepared. Solutions of amino acids within the concentration range of (0 – 1.0 mol.dm^{-3}) were created by volume on the molarity concentration scale with a preciseness of ± 0.1mg on an electronic digital balance (Model: SHIMADZU AX - 200). The density was determined using a specific gravity bottle by relative measurement method with an accuracy of ± 0.01kgm^{-3}. An ultrasonic interferometer having the frequency of 2 MHz (Mittal Enterprises, New Delhi, Model: F-81) with an overall accuracy of ± 0.2 ms^{-1} has been used for velocity measurements. An electronically digital operated constant temperature bath (JULABA F25) has been used to circulate water through the double walled measuring cell made up of steel containing the experimental solution at the desired temperature. The accuracy in the temperature measurement is ± 0.1 K. Solution viscosities were measured by Ostwald type capillary Viscometer, which was placed in a water thermostat having temperature stability. Flow time measurements were performed using digital chronometer within ± 0.01s (Model: CASIO HS -10W).The average of at least six readings was used as the final efflux time. The measured viscosity values have an uncertainty of ± 0.001m.Pa.s.

3. Theory and Calculations

Using the measured data, the following volumetric, compressibility and transport parameter have been calculated using the standard relations.

$$\text{Adiabatic compressibility } \beta = \frac{1}{U^2\rho} \qquad (1)$$

Molar hydration number has been computed using the relation

$$n_H = \left(\frac{n_1}{n_2}\right)\left(1 - \frac{\beta}{\beta o}\right) \qquad (2)$$

where, β and βo are adiabatic compressibility's of solution and solvent respectively, n_1 and n_2 are number of moles of solvent and solute respectively.

The apparent molar compressibility has been calculated from relation,

$$\varphi_k = \frac{1000}{M\rho_0}(\rho_o\beta - \rho\beta o) + \left(\frac{\beta o M_w}{\rho_o}\right) \qquad (3)$$

where β, ρ and βo, ρ_o are the adiabatic compressibility and density of solution and solvent respectively, M is the molar concentration of the solute and M_w the molecular weight of the solute. φ_k is the function of M as obtained by Gucker (1993)[7] from Debye Huckel [8] and is given by

$$\varphi_k = \varphi_k^o + S_k M^{1/2} \qquad (4)$$

where, $\varphi^o{}_k$ is the limiting apparent molar compressibility at infinite dilution and S_k is a constant. $\varphi^o{}_k$ and S_k of equation 4 have been evaluated by least square method.

The apparent molar volume φ_v has been calculated using the relation:

$$\varphi_v = \left(\frac{M_w}{\rho}\right) - \left(\frac{1000(\rho-\rho_o)}{M\rho\rho_o}\right) \qquad (5)$$

The apparent molar volume φ_v has been found to differ with concentration according to empirical relation as:

$$\varphi_v = \varphi_v^o + S_v M^{1/2} \qquad (6)$$

where, $\varphi^o{}_v$ is the limiting apparent molar volume at infinite dilution and S_v is a constant and these values were determined by least square method.

The viscosity A and B coefficients for the amino acids in aqueous dextrose solutions were calculated from the Jones-Dole equation [9].

$$\left(\frac{\eta}{\eta_o}\right) = 1 + AM^{1/2} + BM \qquad (7)$$

Where, η and η_o are the viscosities of the solution and solvent respectively and M are the molar concentration of the solute. A is determined by the ionic attraction theory of Falkenhagen – Vernon [10] and therefore also called Falkenhagen coefficient B or Jones-Dole coefficient is an empirical constant determined by ion-solvent interactions.

Transfer adiabatic compressibility ($\Delta\varphi^o{}_k$) and transfer volume ($\Delta\varphi^o{}_v$) of each amino acid from water to aqueous dextrose solutions have been calculated as:

$$\Delta\varphi_y^o = \varphi_y^o \text{ (in aqueous dextrose solution) - } \varphi_y^o \text{ (in water)} \qquad (8)$$

where, $\varphi^o{}_y$ denotes limiting apparent molar compressibility $\varphi^o{}_k$, limiting apparent molar volume $\varphi^o{}_v$ and viscosity coefficient B.

4. Result and Discussion

The experimental values of density (ρ), viscosity (η) and ultrasonic velocity (u) for different molarity composition of each of the two amino acids viz., L-serine, and L-alanine in aqueous and aqueous dextrose solutions (0.5 and 1.0 mol.dm^{-3}) at 303.15 K are shown in the Table-1. Further, the values of adiabatic compressibility (β), molar hydration number (n_H), apparent molar compressibility (φ_k), apparent molar volume (φ_v), limiting apparent molar compressibility($\varphi^o{}_k$), limiting apparent molar volume($\varphi^o{}_v$), and their constants (S_k, S_v), transfer adiabatic compressibility ($\Delta\varphi^o{}_k$), transfer volume ($\Delta\varphi^o{}_v$), and viscosity A & B coefficients are shown in Tables 2,3.

In all the two amino acids system the values of density and ultrasonic velocity (Table 1) increases with increase in molar concentration of amino acids as well as dextrose content. This increasing trend suggests a moderate strong electrolytic nature in which the solutes (amino acids) tend to attract the solvent (aqueous dextrose) molecules. Molecular interaction is thus responsible for the observed increase in density and ultrasonic velocity in these mixtures. The factors apparently responsible for such behaviour may be due to the presence of interactions caused by the proton transfer reactions of amino acids in aqueous dextrose mixtures. The increase in ultrasonic velocity in these solutions may be attributed to the cohesion brought about by the ionic hydration.

Table 1: Values of density (ρ), viscosity (η) and ultrasonic velocity (U) of amino acids in aqueous dextrose solutions at 303.15K for

M/(mol.dm^{-3})	**ρ / (kgm^{-3})**			**η / (x 10^{-3} Nsm^{-2})**			**U / (m.s^{-1})**		
	water + dextrose								
	0.0M	**0.5M**	**1.0M**	**0.0M**	**0.5M**	**1.0M**	**0.0M**	**0.5M**	**1.0M**
System - I: water + dextrose + L-serine									
0.0	995.7	1011.6	1027.5	0.7972	0.9818	1.1587	1512.5	1542.4	1569.4
0.2	996.2	1015.3	1041.6	0.8681	1.0432	1.2592	1530.6	1543.5	1572.2
0.4	997.7	1023.9	1043.4	0.8965	1.0795	1.2988	1555.0	1557.0	1579.4
0.6	1000.9	1052.2	1046.9	0.9013	1.1382	1.3379	1558.8	1589.3	1595.9
0.8	1008.0	1055.8	1055.8	0.9212	1.2106	1.3629	1577.9	1590.9	1600.4
1.0	1015.1	1062.1	1057.6	0.9310	1.2951	1.4027	1588.7	1601.8	1627.6
System – II: water + dextrose + L-alanine									
0.0	995.7	1011.6	1027.5	0.7972	0.9818	1.1587	1512.5	1542.4	1569.4
0.2	996.0	1013.3	1034.6	0.8179	1.0335	1.2105	1515.8	1543.5	1581.2
0.4	996.9	1015.1	1039.9	0.8291	1.0642	1.2502	1520.2	1555.9	1589.8
0.6	998.0	1027.5	1041.6	0.8539	1.1250	1.2980	1525.9	1558.7	1595.1
0.8	1000.1	1031.0	1045.2	0.8852	1.1852	1.3526	1526.0	1564.9	1601.3
1.0	1002.2	1034.6	1054.0	0.9152	1.2166	1.3880	1535.8	1572.6	1606.9

In all the two systems the value of adiabatic compressibility (Table 2) are decreases with increase in concentration of amino acids as well as dextrose content. The decrease in adiabatic compressibility is attributed to the influence of the electrostatic field of ions on the surrounding solvent molecules so called electrostriction. The magnitude of β values is larger in L-alanine than L-serine. The larger β value which shows molecular associations/interactions is greater in L-alanine than L-serine. Amino acid molecules in the neutral solution exist in the dipolar form and thus have stronger interaction with the surrounding water molecules. The increasing electrostrictive compression of water around the molecules results in a large decrease in the compressibility of the solutions [11]. The interaction between the solute and water molecules in the solvent is termed hydration. From the Table 2 it is observed that the positive values of n_H indicate an appreciable solvation of solutes. This is an added support not only for the structure promoting tendency of the solutes but also for the presence of appreciable dipole-dipole interactions between solute and water molecules. This also leads further suggestion that the compressibility of the solution will be less than that of the solvent. As a result, solutes will gain mobility and hence there will be more probability of conducting solvent molecules. This may further enhance the interaction between Solute and solvent molecules. The nonlinear behaviour of n_H shows the existence of strong interaction in the mixtures [12].

The following observations have been made on φ_k and φ_v (Table 2) of the two amino acids in aqueous dextrose solutions at 303.15 K.

1. The values of φ_k and φ_v are all negative over the entire range of the molarity.
2. The negative values of φ_k and φ_v are varying nonlinearly with the increase in concentration of amino acids as well as dextrose contents.
3. The magnitude of φ_k is in the order:
   L-alanine > L-serine

The above observations clearly suggest that the negative values of φ_k and φ_v in all systems indicate the presence of solute-solvent interactions. The observed behaviour of φ_k and φ_v reveals that strengthening of the solute-solvent interaction in all systems studied. The negative values of φ_v indicate electrostrictive solvation of ions [13]. From the magnitude of φ_k, it can be concluded that stronger molecular association is found in L-alanine than L-serine and hence L-alanine is a more effective structure maker.

Table 2: Values of adiabatic compressibility (β), molar hydration number (n_H), apparent molar compressibility (φ_k) and apparent molar volume (φ_v) of amino acids in aqueous dextrose solutions at 303.15 K for

M/ (mol.dm^{-3})	**β / (x 10^{-10} m^2 N^{-1})**			**n_H**			**-φ_k / (x 10^{-7} m^2 N^{-1})**			**- φ_v / (x 10^{-3} m^3 mol^{-1})**		
	water + dextrose											
	0.0M	**0.5M**	**1.0M**	**0.0M**	**0.5M**	**1.0M**	**0.0M**	**0.5M**	**1.0M**	**0.0M**	**0.5M**	**1.0M**
System - I: water + dextrose + L-serine												
0.0	4.3903	4.1552	3.9514	--	--	--	--	--	--	--	--	--
0.2	4.2847	4.1346	3.8839	6.6790	1.2655	3.9814	0.5410	0.1804	0.6095	5.42	17.91	66.10
0.4	4.1450	4.0286	3.8421	7.8222	3.8917	3.2216	0.6356	0.4436	0.4265	9.07	29.78	37.02
0.6	4.1115	3.7627	3.7503	5.9104	8.0818	3.9500	0.5036	0.9327	0.4598	8.78	63.58	30.05
0.8	3.9844	3.7425	3.6983	6.4446	6.3644	3.7282	0.5756	0.7428	0.4525	15.31	51.65	32.52
1.0	3.9032	3.6694	3.5697	6.1613	5.9684	4.4974	0.5723	0.6935	0.4974	19.11	46.96	27.58

System – II: water + dextrose + L-alanine												
0.0	4.3903	4.1552	3.9514	--	--	--	--	--	--	--	--	--
0.2	4.3699	4.1422	3.8660	1.2902	0.7991	5.0303	0.1099	0.1011	0.5631	99.83	84.97	33.22
0.4	4.3404	4.0696	3.8051	1.5779	2.6291	4.3081	0.1383	0.2505	0.4847	40.81	85.06	28.88
0.6	4.3033	4.0059	3.7732	1.8341	3.0571	3.4985	0.1624	0.3577	0.3793	15.09	25.43	21.95
0.8	4.2937	3.9605	3.7313	1.5273	2.9900	3.2409	0.1453	0.3431	0.3601	89.48	23.21	20.51
1.0	4.2304	3.9084	3.6743	2.0225	2.5200	3.2641	0.1889	0.3412	0.3791	1.69	21.88	24.41

The limiting apparent molar compressibility φ^o_k provides information regarding solute-solvent interactions and S_K, that of solute-solute interactions in the solution. From the Table 3, it is observed that φ^o_k values are negative and it increases with increasing the concentration of dextrose in all systems studied. Appreciable negative values of φ^o_k, and its behaviour for all systems reinforce our earlier view regarding existence of solute-solvent interaction in the mixtures. Further, from the above table it found that the values of φ^o_v are negative in L-alanine, whereas it can be found as positive in L-serine. The values of φ^o_v increases with the addition of dextrose contents with respect to aqueous medium in all the systems studied. The increase in φ^o_v may be attributed to the decreased hydrophobicity / non-polar character of the side chain of the amino acids. The magnitude of φ^o_v is in the order: L-alanine < L-serine. It is evident from the Table 3 that S_k and S_v are both positive and negative in all the two systems suggesting the presence of solute-solute interactions.

Table 3: Values of limiting apparent molar compressibility (φ^o_k), limiting apparent molar volume (φ^o_v) and their constants S_k and S_v, transfer adiabatic compressibility ($\Delta\varphi^o_k$), transfer volumes ($\Delta\varphi^o_v$) of amino acids in aqueous dextrose solutions at 303.15 K for

Amino acids	Dextrose M/(mol. dm^{-3})	$-\varphi^o_k$ /(×10^{-7} m^{2}N^{-1})	φ^o_v /(×10^{-3} m^3.mol^{-1})	S_k /(×10^{-7} N^{-1} m^{-1}.mol^{-1})	S_v /(×10^{-3} m^3 L^{½}. mol$^{-3/2}$)	$\Delta\varphi^o_k$ /(×10^{-7} m^2 N^{-1})	$\Delta\varphi^o_v$ /(×10^{-3} m^3. mol^{-1})	A / (dm$^{3/2}$. mol$^{-1/2}$)	B / (dm$^{3/2}$ mol^{-1})
L-serine	0.0	0.6200	-662.96	0.1746	828.31	--	--	-2.8313	4.0183
	0.5	0.5629	6.29	-0.0035	-0.02	0.0571	669.25	-0.0273	0.3251
	1.0	0.1823	524.49	-1.0416	-699.65	0.4377	1187.45	0.1793	0.0254
L-alanine	0.0	0.7136	-780.07	0.3740	974.65	--	--	-0.0534	1.8931
	0.5	0.0524	-0.15	-0.4417	0.14	0.6612	779.93	-1.2374	0.2005
	1.0	0.0595	-0.04	-0.1192	0.02	0.6541	780.03	0.0129	0.1867

The values of transfer adiabatic compressibility $\Delta\varphi^o_K$ and transfer volume $\Delta\varphi^o_V$ (Table 3) are positive and it increases with increase in the concentration of dextrose in all the two amino acid systems which suggest the existence of strong solute-solvent interactions in the mixtures. The magnitude of $\Delta\varphi^o_V$ in order: L-alanine < L-serine. Generally, the interactions between amino acids and aqueous dextrose can be classified as:

a) The hydrophilic - ionic interaction between OH groups of dextrose and zwitterions of amino acids (NH_3^+ and COO^-).
b) Hydrophilic-hydrophilic interaction between OH groups of dextrose and NH groups in the side chain of amino acids.
c) Hydrophilic-hydrophobic interaction between OH groups of dextrose and non-polar ($-CH_2$) in side chain of amino acids.
d) Hydrophobic- hydrophobic group interaction between the non-polar group of dextrose and non-polar ($-CH_2$) in side chain of amino acids.

The $\Delta\varphi^o_V$ values can also be explained on the basis of co-sphere overlap model [14] in terms of solute-cosolute interactions. According to this model, hydrophilic - ionic interactions and hydrophilic - hydrophilic interactions contribute positively, whereas hydrophilic - hydrophobic interactions and hydrophobic- hydrophobic contribute negatively to the $\Delta\varphi^o_V$ values.

Therefore, from Figures 1 & 2, the positive $\Delta\varphi^o_K$ and $\Delta\varphi^o_V$ values observed in all the two amino acids suggest that the interaction contribution of type (i) and (ii) is stronger than that of type (iii) and (iv).

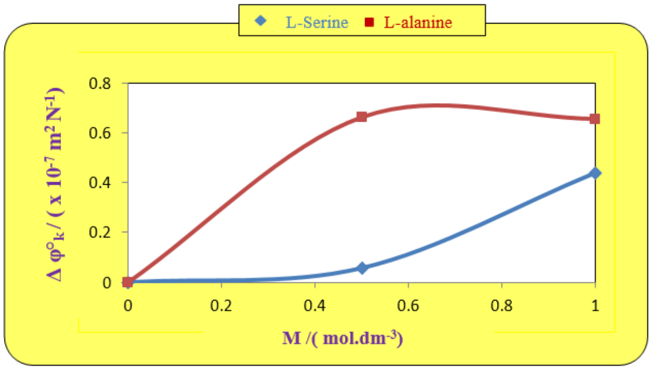

Fig 1: Variation of transfer adiabatic compressibility ($\Delta\varphi^o_k$) of some amino acids with molarity of aqueous dextrose solutions at 303.15 K

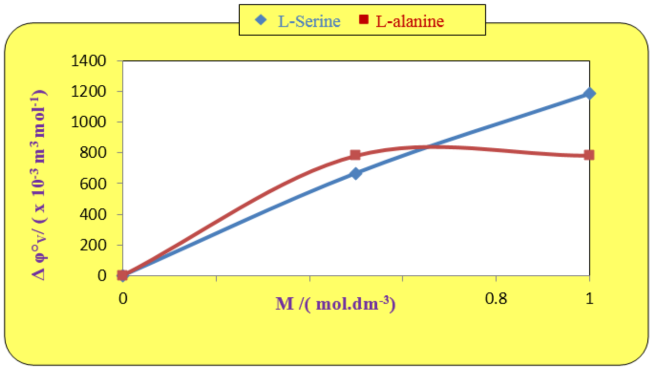

Fig 2: Variation of transfer volume ($\Delta\varphi^o_v$) of some amino acids with molarity of aqueous dextrose solutions at 303.15 K

From Table 1, it is observed that the values of viscosity increases with increase in molar concentration of amino acids as well as dextrose content. This increasing trend indicates the existence of solute-solvent interaction occurring in these systems. In order to shed more light on this, the role of viscosity coefficients have been obtained. From the Table 3, it is observed that the values of A coefficient is both positive and negative for all the system indicating the presence of ion-ion interactions [15]. Further, it is found that the values of the B-coefficients are also positive in all the systems are studied. It is also a measure of solute-solvent interaction and relative size of the solute and solvent molecules. The behaviour of B-coefficient in all the two systems implying the existence of strong solute-solvent interaction.

5. Conclusion

In the present investigation, volumetric, compressibility and transport parameters of L-serine and L-alanine in aqueous and aqueous dextrose solutions (0.5 and 1.0 mol.dm^{-3}) at 303.15 K were obtained using density, viscosity and ultrasonic velocity data and the results have been used to study the existence of solute-solute and solute-solvent interactions. From the magnitude of φ_k, φ^o_v and $\Delta\varphi^o_v$, it is concluded that L-alanine possesses strong solute-solvent interaction than L-serine. The transfer adiabatic compressibility $\Delta\varphi^o_K$ and transfer volume $\Delta\varphi^o_v$ data suggest that hydrophilic-ionic and hydrophilic-hydrophilic interactions are dominating over the hydrophilic – hydrophobic and hydrophobic- hydrophobic interactions.

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