



Research Article

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Studies on Solute-Solvent Interactions of Amino acids in Water + D-glucose mixtures at Different Temperatures

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ABSTRACT

Density (d) values of the three amino acids, namely glycine, α -alanine and β -alanine have been measured at 298.15, 303.15, 308.15 and 313.15 K in aqueous D-glucose solutions ranging from 0 to 20 mass % of D-glucose. The ultrasonic velocity (U) in these solutions have been measured at 298.15 K only. Acoustical parameters such as isentropic compressibility (K_s), acoustic impedance (Z), solvation number (S_n), apparent molar volume (V_ϕ), limiting apparent molar volume (V_ϕ^0) and limiting apparent molar expansibility (E_ϕ^0) have been computed. The results are discussed in the light of molecular interactions in the mixtures.

Keywords : Amino acids, D-glucose, apparent molar volume, ultrasonic velocity.

INTRODUCTION

The study of carbohydrate-protein interactions is very important for immunology, pharmacology and medicine. Due to the complex molecular structure of proteins direct study is quite difficult. So the amino acids, which are the building blocks of proteins are studied. Although a lot of attention has been given to the behaviour of amino acids in different salt-water mixed solvents, very few studies have been carried out on amino acids in carbohydrate and water mixtures. The present investigation deals with the study of molecular interactions of three amino acids (Glycine, α -alanine and β -alanine) in aqueous D-glucose solutions in mass percentage from 0 to 20 % by 5% increments. Various parameters such as isentropic compressibility (K_s), acoustic impedance (Z), apparent molar expansibility (E_ϕ), limiting apparent molar expansibility (E_ϕ^0), apparent molar volume (V_ϕ) and limiting apparent molar volume (V_ϕ^0) have been calculated from the density (d) and ultrasonic velocity (U) data respectively, which provide qualitative information regarding molecular interactions.

EXPERIMENTAL SECTION

Glycine, α -alanine and β -alanine were of the AnalaR grades and used as such. The solutions of glycine, α -alanine and β -alanine were prepared on the molal basis and conversion of molality to molarity was done by using the standard expression [1] using the density values of the solutions determined at 298.15 K. Conductivity water (Specific conductance $\sim 10^{-6}$ S cm^{-1}) was used to prepare solutions of D-glucose and the solutions were used on the same day. The densities of pure solvents and their solutions were determined by relative measurement method by using a specific gravity bottle (25 ml capacity) as described elsewhere [2]. At least five observations were taken and differences in any two readings did not exceed $\pm 0.02\%$. The ultrasonic velocity was measured by using ultrasonic

interferometer(Mittal Enterprises, New Delhi, Model No.F-81) having frequency 2MHz.The precision of the ultrasonic velocity measurements was within ± 0.5 m/s. The amino acid content in the solutions varied over a concentration range of 0.01 to 0.08 mol dm⁻³ in various solvents. Measurement of density was done in the temperature range 298.15 K to 313.15 K and speed of sound at 298.15 K only.

Theoretical Aspects

From the density data,the apparent molar volume(V_ϕ), was calculated by using equation [3]

$$V_\phi = 1000(cd_0)^{-1}(d_0 - d) + M_2 d_0^{-1} \quad (1)$$

where d_0 is the density of solvent.

It was found that the V_ϕ varied linearly with concentration $c^{1/2}$.

The V_ϕ data were fitted by a method of least squares to Masson equation [3]

$$V_\phi = V_\phi^0 + S_v c^{1/2} \quad (2)$$

to obtain V_ϕ^0 (limiting apparent molar volume) and the slope, S_v .

The apparent molar expansibility, E_ϕ was determined from equation [3]

$$E_\phi = \alpha_0 V_\phi + (\alpha - \alpha_0) 1000 c^{-1} \quad (3)$$

Where $\alpha_0 = -1/d_0(\delta d_0/\delta t)_p$ &

$$\alpha = -1/d(\delta d/\delta t)_p \quad (4)$$

The E_ϕ data were fitted by a method of least squares to the equation [3]

$$E_\phi = E_\phi^0 + S_E c^{1/2} \quad (5)$$

The ultrasonic velocities U of the solutions of the amino acids in mixed solvents at different concentrations were fitted to an equation of the form [4-6]

$$U = U_0 + Fc + Gc^{3/2} + Hc^2 \quad (6)$$

Where U_0 is the sound velocity in pure solvent and U in mixed solvent, and F, G and H are the constants.

The ultrasonic velocity, U is related to the density, d of the solution and isentropic compressibility K_s by the relationship [4-6]

$$U = (K_s d)^{-1/2} \quad (7)$$

The values of K_s obtained for solutions of different concentrations were fitted to an equation of the form

$$K_s = K_s^0 + A'c + B'c^{3/2} + C'c^2 \quad (8)$$

where A', B' and C' are the constants and c is the molar concentration.

The apparent isentropic molar compressibility $K_{s,\phi}$ has been computed from equation(9) [4-6]

$$K_{s,\phi} = 1000K_s c^{-1} - K_s^0 d_0^{-1} (1000c^{-1}d - M_2) \quad (9)$$

The $K_{s,\phi}$ data were fitted to equation(10)

$$K_{s,\phi} = K_{s,\phi}^0 + F'c^{1/2} + G'c \quad (10)$$

to obtain $K_{s,\phi}^0$ (the limiting apparent isentropic molar compressibility)

The solvation number, S_n of a solute can be related to the isentropic compressibility by equation (11)

$$S_n = n_1 n_2^{-1} [1 - VK_s (n_1 v_1^0 K_s^0)^{-1}] \quad (11)$$

where V is the volume of the solution containing n_2 moles of solute.

v_1^0 is the molar volume of solvent and n_1 is the number of moles of solvent.

The variation of solvation number with molar concentration of the solute leads to the limiting solvation number, S_n^0 which was obtained from the relation (12)

$$\lim_{c \rightarrow 0} K_{s,\phi} = -S_n^0 V_1^0 K_s^0 \quad (12)$$

From the density and sound velocity values, the magnitude of relative association, R_A was calculated from the relation [4-6]

$$R_A = (d/d_0) (U_0/U)^{1/3} \quad (13)$$

The specific acoustic impedance, Z was calculated by using the relation [4-8]

$$Z = Ud \quad (14)$$

RESULTS AND DISCUSSION

The values of partial molar volume (V_ϕ^0), partial molar expansibility (E_ϕ^0), the slope (S_v) of the plot of V_ϕ vs $c^{1/2}$ and the slope (S_E) of E_ϕ vs $c^{1/2}$ are given in Table 1 for the amino acids in water and water + D-glucose mixtures at 298.15, 303.15, 308.15 and 313.15 K.

The density values of amino acid solutions vary linearly with the concentration in both water and mixed solvents. It was found that the apparent molar volume (V_ϕ) values vary linearly with $c^{1/2}$ for all the amino acids in water and water + D-glucose mixtures at the experimental temperatures. Since V_ϕ^0 indicates the ion-solvent interactions at infinite dilution (as the ion-ion interaction vanishes at infinite dilution), the positive values indicate strong ion-solvent interaction which mostly increase with increase of temperature. As observed, the ion-solvent interaction increases with increasing D-glucose content in water. The strong ion-solvent interaction between the molecules tends to promote structure-making effect of the amino acids in the mixed solvents.

The negative values of S_v (excepting a few cases) are the indicative of weak ion-ion interaction, however, vary with change of temperature and the D-glucose content in water. The negative values of S_v for the amino acids in water may be attributed to increase in solvation indicating the cavity occupation of the crystal lattice of the corresponding amino acids by water molecules.

The values of limiting apparent molar expansibility, E_ϕ^0 are positive (excepting a few cases) and vary slightly with increase of temperature. Ion-ion interactions as seen through the magnitudes of S_v and S_E show a wide diversion for the amino acids in all solvents at all temperatures. As observed, the values of S_E are positive in some cases and negative in others. The results are again in agreement with the above contention that the structure making effect is favoured in all the solvents studied.

Table - 1 (Values of parameters, V_{ϕ}^0 ($\text{m}^3 \text{mol}^{-1}$), S_{ϕ} ($\text{m}^{9/2} \text{mol}^{-3/2}$), E_{ϕ}^0 ($\text{m}^3 \text{mol}^{-1} \text{K}^{-1}$), S_E ($\text{m}^{9/2} \text{mol}^{-3/2} \text{K}^{-1}$) for glycine, α -alanine and β -alanine in aqueous solutions of D-glucose at different temperatures.)

Wt % of D-glucose	Temp (K)	$V_{\phi}^0 \times 10^6$ ($\text{m}^3 \text{mol}^{-1}$)	$S_{\phi} \times 10^3$ ($\text{m}^{9/2} \text{mol}^{-3/2}$)	$E_{\phi}^0 \times 10^6$ ($\text{m}^3 \text{mol}^{-1} \text{K}^{-1}$)	$S_E \times 10^3$ ($\text{m}^{9/2} \text{mol}^{-3/2} \text{K}^{-1}$)
Glycine					
0	298.15	59.69	-59.57	0.26	0.23
	303.15	61.12	-58.72	0.21	3.73
	308.15	61.93	-39.00	1.05	-3.53
	313.15	67.14	-56.30	0.56	-0.02
5	298.15	13.15	90.48	3.33	-6.72
	303.15	29.79	57.39	0.84	-3.35
	308.15	34.09	40.49	1.09	-1.83
	313.15	39.69	31.07	1.78	-4.02
10	298.15	14.40	97.40	4.28	-12.42
	303.15	35.65	36.60	0.34	-1.26
	308.15	37.50	29.59	0.50	0.93
	313.15	40.00	34.75	1.73	-4.32
15	298.15	25.85	83.24	0.22	-0.49
	303.15	26.81	81.50	1.68	-7.02
	308.15	35.60	45.26	0.71	-0.74
	313.15	39.57	40.48	0.90	-2.83
20	298.15	27.10	85.29	0.11	0.73
	303.15	27.90	88.22	1.56	-6.30
	308.15	35.56	57.14	0.87	-3.99
	313.15	39.72	37.92	0.80	-3.04
α -alanine					
0	298.15	84.85	-81.53	-0.67	-0.80
	303.15	85.60	-85.86	2.36	-2.10
	308.15	97.19	-95.21	0.37	-2.14
	313.15	98.97	-105.46	0.74	-1.90
5	298.15	55.32	44.77	0.05	-0.10
	303.15	55.43	44.87	0.01	0.03
	308.15	55.53	44.95	0.01	0.07
	313.15	55.69	45.08	0.03	-0.01
10	298.15	57.01	41.78	0.38	0.18
	303.15	58.65	43.61	0.14	-0.07
	308.15	59.47	43.11	1.07	-2.69
	313.15	64.75	30.00	0.54	-0.90
15	298.15	59.07	41.15	0.92	-3.59
	303.15	63.46	23.95	0.23	-0.19
	308.15	63.78	26.77	0.52	-1.54
	313.15	66.73	18.01	0.50	-1.53
20	298.15	62.08	25.20	0.62	-0.95
	303.15	65.40	19.89	0.24	-0.76
	308.15	66.45	16.78	0.22	0.46
	313.15	67.33	19.99	0.31	-0.22
β -alanine					
0	298.15	84.13	-72.85	0.68	-2.50
	303.15	85.56	-85.70	0.27	-0.15
	308.15	88.76	-85.41	0.61	-1.70
	313.15	91.72	-93.47	0.57	-1.63
5	298.15	7.17	119.90	1.79	2.79
	303.15	16.08	134.59	3.15	-11.34
	308.15	32.02	77.49	0.78	0.03
	313.15	32.13	94.58	1.66	-1.72
10	298.15	10.40	143.05	4.95	-19.91
	303.15	35.02	44.00	1.79	0.92
	308.15	44.17	48.63	0.56	-2.59
	313.15	46.96	36.04	2.47	-7.30

Contd.

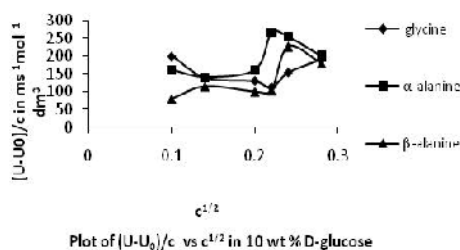
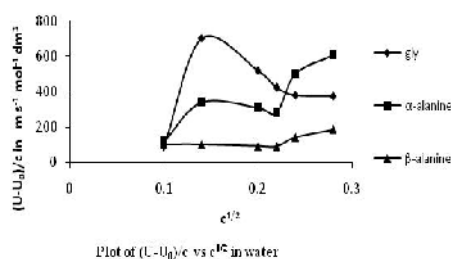
15	298.15	12.53	141.54	6.27	-19.73
	303.15	43.88	43.20	0.09	1.65
	308.15	44.71	50.61	1.78	-5.34
	313.15	54.05	22.68	2.77	-7.96
20	298.15	38.08	62.11	1.34	-6.48
	303.15	45.07	28.90	0.54	2.76
	308.15	47.63	43.53	1.56	-3.85
	313.15	55.29	25.10	1.10	-2.35

A study of ultrasonic behaviour of solutions of amino acids in water and water + D-glucose(5,10,15 and 20 wt%) mixed solvents at 298.15 K reveals that the sound velocity increases and the isentropic compressibility(K_s) decreases as the D-glucose content in water increases. The values of U_0 and the constants F,G,H are given in Table 2 .

Table-2 (Values of U_0 (ms^{-1}) and the constants F,G,H at 298.15K.)

Aminoacid	Solvent	Wt% of D-glucose	$U_0(\text{ms}^{-1})$	F	G	H
Glycine	Water	0	1502.0	316.41	500.46	-1898.89
		5	1520.0	333.7	-689.8	53.76
	D-glucose	10	1541.6	169.8	-79.43	341.62
		15	1568.8	445.1	-1021.0	-126.9
		20	1583.9	1022.0	-2731.0	512.18
α -alanine	Water	0	1502.0	-78.25	2224.2	64.5
		5	1520.0	386.3	-271.4	-840.5
	D-glucose	10	1541.6	98.14	498.6	16.66
		15	1568.8	27.99	91.84	24.76
		20	1583.9	329.8	-315.0	-325.22
β -alanine	Water	0	1502.0	40.92	387.69	223.34
		5	1520.0	428.8	-1335.0	537.90
	D-glucose	10	1541.6	14.73	607.8	75.23
		15	1568.8	66.82	263.8	-37.26
		20	1583.9	1923.0	-5555.0	1426.8

Such changes are also reported by other workers in other aquo-organic solvent mixtures like water+methanol [9],water+DMSO [10] and water+PG [11].As observed, the sound velocity increases with increase in the concentration of the solutions in all the solvents .The sound velocities of amino acids are in the order: α -alanine > glycine > β -alanine in water and water + D-glucose mixtures. As the amino acids contain one carboxylate ion each, the variation of sound velocity is due to the increase in the mass of the $-\text{NH}_2$ substituted aliphatic carbon residue (typical plots of $(U-U_0)/c$ vs $c^{1/2}$ in water and water+10 wt% D-glucose mixtures are shown in Figure 1 and 2 respectively).



The values of K_s^0 and the constants A', B', C' are given in Table 3. The value of K_s decreases with increase in the concentration of the solution (a typical plot of $(K_s - K_s^0)/c$ vs $c^{1/2}$ is given in Figure 3).

The decrease in K_s with concentration may be due to the fact that the interstitial spaces of water are occupied by the solute molecules making the medium less compressible, i.e providing greater electrostriction. Further, the decrease in compressibility with increase in the D-glucose content in water may be due to the filling of the interstitial spaces of water molecules by organic co-solvent, D-glucose molecules thereby making a tight structure.

Since the values of K_s follow the reverse order to that of the sound velocity, it would be correct to conclude that in case of amino acids, the K_s depends on the solvation layer formed around the aliphatic carbon residue (since, the other part is the same, i.e. carboxylate group). As a tight solvation layer formed around a group leads to a decrease in K_s , the lowering of K_s is due to the increased tightness of solvation layer formed around the corresponding aliphatic carbon residue of the amino acids. This also supports the variation of K_s with the size of the aliphatic carbon residue.

Table-3 (Values of K_s^0 (pa^{-1}) and the constants A', B', C')

Amino acid	Solvent	Wt% of D-glucose	$10^{-10} \times K_s^0 (\text{m s}^{-2} \text{kg}^{-1})$	$10^{-10} (A')$	$10^{-10} (B')$	$10^{-9} (C')$
Glycine	Water	0	4.45	-3.00	-0.7	2.69
		5	4.26	-2.00	5.00	-1.004
	D-glucose	10	4.06	-0.7	-0.5	-0.25
		15	3.85	-3.00	8.00	-0.30
		20	3.70	-5.00	10.0	2.2
α -alanine	Water	0	4.45	-0.2	-10.0	-0.52
		5	4.26	-2.00	2.00	-0.74
	D-glucose	10	4.06	-0.2	-4.00	-0.047
		15	3.85	-0.4	-0.2	-0.17
		20	3.70	-2.00	2.00	1.09
β -alanine	Water	0	4.45	-0.9	-0.4	-0.058
		5	4.26	-3.00	8.00	1.09
	D-glucose	10	4.06	-0.03	-4.00	-0.003
		15	3.85	-0.08	-0.06	-0.048
		20	3.70	-9.00	30.0	-3.08

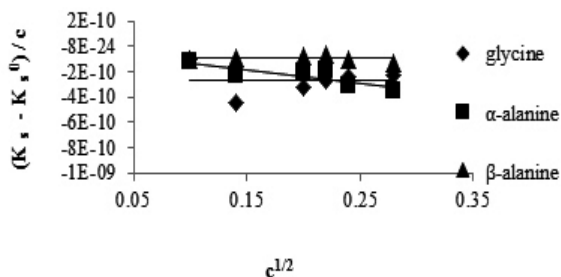


Figure 3 (i) : Plot of $(K_s - K_s^0)/c$ vs $c^{1/2}$ in water at 298.15 K

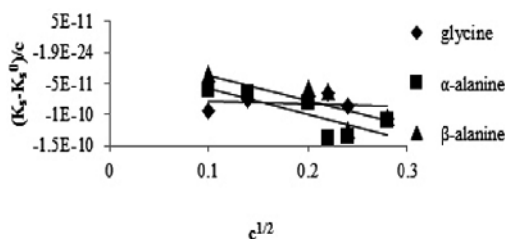


Figure 3(ii) : Plot of $(K_s - K_s^0)/c$ vs $c^{1/2}$ in 10wt % D-glucose at 298.15K

A typical plot of $K_{s,\phi}$ vs $c^{1/2}$ is shown in Figure 4.

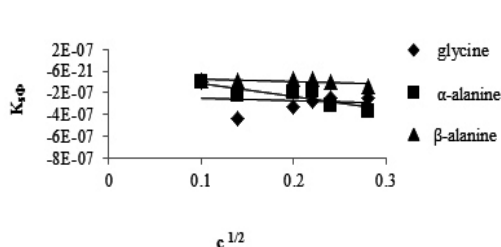


Figure 4 (i) : Plot of $K_{s,\phi}$ vs $c^{1/2}$ in water at 298.15K

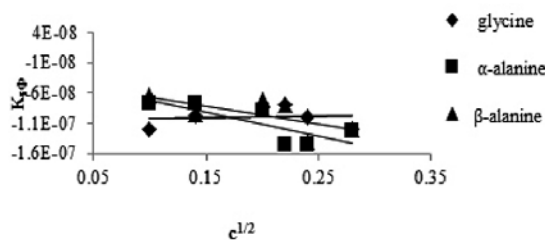
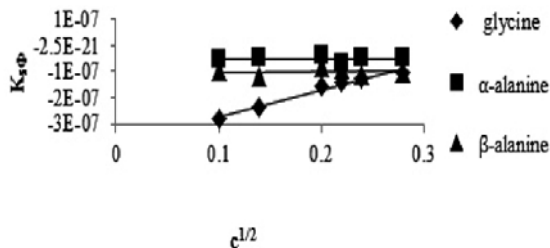
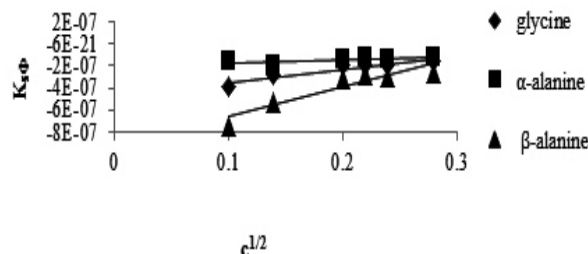


Figure 4(ii) : Plot of $K_{s,\phi}$ vs $c^{1/2}$ in 10wt % D-glucose

Figure 4(iii) : Plot of $K_{s,\phi}$ vs $c^{1/2}$ in 15wt% D-glucoseFigure 4(iv) : Plot of $K_{s,\phi}$ vs $c^{1/2}$ in 20 wt% D-glucose

The values of $K_{s,\phi}$ are negative and so also the values of $K_{s,\phi}^0$ [12]. The negative values may be explained by means of two different phenomena, viz., electrostriction and hydrophobic solvation. The loss of compressibility of the surrounding solvent molecules due to strong electrostrictive forces at the carboxyl group causes electrostrictive solvation. In other words, a tight solvation layer is formed around the ion for which the medium is little compressed by the application of pressure.

The values of $K_{s,\phi}^0$ are given in Table 4. As observed, the values of $K_{s,\phi}^0$ of the amino acids follow the order : $K_{s,\phi}^0(\alpha\text{-alanine}) > K_{s,\phi}^0(\beta\text{-alanine}) > K_{s,\phi}^0(\text{glycine})$ in water, $K_{s,\phi}^0(\alpha\text{-alanine}) \sim K_{s,\phi}^0(\text{glycine}) > K_{s,\phi}^0(\beta\text{-alanine})$ in 5 wt% D-glucose, $K_{s,\phi}^0(\alpha\text{-alanine}) > K_{s,\phi}^0(\text{glycine}) > K_{s,\phi}^0(\beta\text{-alanine})$ in 20 wt% D-glucose.

Table - 4 (Values of $K_{s,\phi}^0$ ($\text{m}^3 \text{mol}^{-1} \text{pa}^{-1}$) and the constants F' , G' and S_n^0)

Amino acid	Solvent	D-glucose	$K_{s,\phi}^0$ ($\text{m}^3 \text{mol}^{-1} \text{pa}^{-1}$) $\times 10^{-7}$	$10^{-1}(F')$	$10^{-1}(G')$	$10^{-2}(S_n^0)$
Glycine	Water	0	-2.00	-2.00	-0.74	6.21
		5	-2.00	5.00	-17	2.49
	D-glucose	10	-1.00	0.3	-3.85	2.49
		15	-4.00	10.0	9.84	3.75
		20	-5.00	10.0	19.4	4.92
α -alanine	Water	0	0.1	-10.0	-12.0	6.21
		5	-2.00	1.00	-4.08	3.74
	D-glucose	10	-0.3	-4.0	-0.52	2.49
		15	-0.5	-0.03	-0.07	1.12
		20	-2.00	3.00	-1.57	2.46
β -alanine	Water	0	-0.5	-2.00	-2.85	2.49
		5	-3.00	7.00	12.7	2.49
	D-glucose	10	-0.4	-3.00	0.95	2.49
		15	-1.00	0.2	-1.40	2.50
		20	-9.00	30.0	-38.1	7.39

This trend in $K_{s,\phi}^0$ values implies that α -alanine shows strongly electrostriction as compared to other amino acids in all the solvents. In other words, the solvation layer formed around the α -alanine is thick and / or hard in the solvent concerned.

Another property [13,14] which also can be studied to understand the ion-ion or ion-solvent interaction is the relative association, R_A . It is influenced by two factors: (i) breaking up of the associated solvent molecules on addition of the solute to it, and (ii) the solvation of solute molecules. The former leads to the decrease and the latter to the increase of relative association. In the present study, R_A decreases with increase in the solute concentration for all the amino acids in all solvents (a typical plot of R_A vs c in water and water+ 20 wt% D-glucose for all the amino acids is shown in Figure 5).

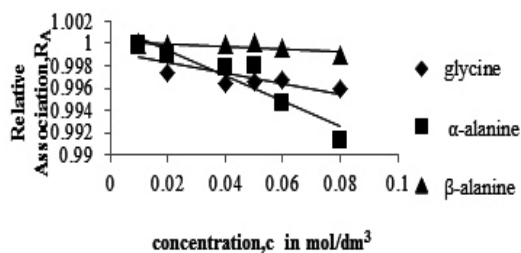


Figure 5(i) : Plot of R_A vs conc. in water

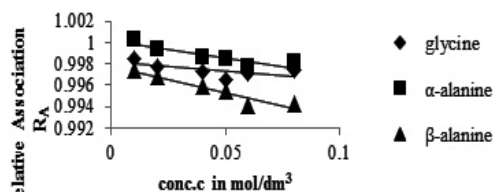


Figure 5(ii) : Plot of R_A vs conc. in 20 wt% D-glucose

This implies that the breaking up of the associated solvent molecules on addition of the solute takes place in all solvents.

It is observed that the value of acoustic impedance, Z varies with the increase in solute concentration [15-17]. In the present study, the plot of Z vs $c^{1/2}$ is almost linear (a typical plot of Z vs $c^{1/2}$ is shown in Figure 6 for all the amino acids in water and water + 20wt% D-glucose).

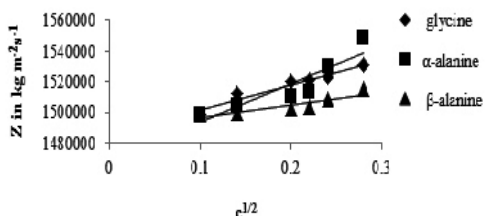


Figure 6(i) : Plot of Z vs $c^{1/2}$ in water

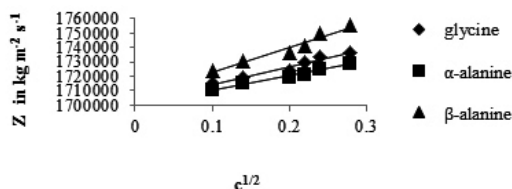


Figure 6(ii) : Plot of Z vs $c^{1/2}$ in 20 wt% D-glucose

A typical plot of S_n vs c is shown in Figure 7. It is found that the limiting solvation number, S_n^0 of the amino acids is larger in water than in water+ D-glucose mixtures.

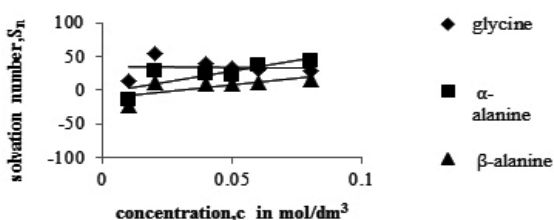


Figure 7(i) : Plot of S_n vs conc. in water

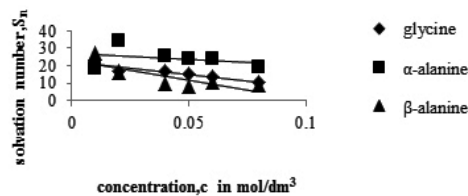


Figure 7(ii) : Plot of S_n vs conc. in 5wt% D-glucose

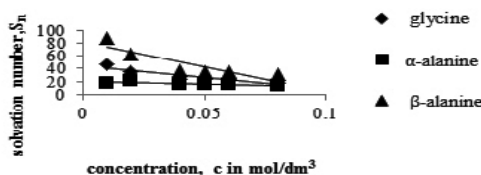


Figure 7(iii) : Plot of S_n vs conc. in 20wt% D-glucose

The decrease in S_n^0 value in the mixed solvent medium indicates a structure breaking process predominating in the medium. In different solvents, S_n^0 value differs and follows the order of S_n value. Higher S_n^0 value indicates strong electrostriction in water as compared to water + D-glucose mixtures. It is considered that the solvation layer formed around the ion is thick and/or hard in water than in water + D-glucose mixtures. However, the variation of S_n^0 as well as of S_n values predicts the degree of hard electrostrictive solvation. It represents the structural effect of the solute on the solvent in a solution.

CONCLUSION

The results of the present investigation on the amino acids in water and water+ D-glucose reveal that amino acids show strong ion-solvent interactions in all solvents, and the structure making effect is very much favoured in these solvents. The variation in sound velocity in the solutions of amino acids is due to the change of the aliphatic carbon part in the amino acids, and increase in sound velocity is due to the increase in their mass. The decrease in the value of isentropic compressibility (K_s) with increase in the solute concentration may be due to the occupation of the interstitial spaces of water by the solute molecules thereby making the medium less compressible. Lower K_s value of one amino acid as compared to others implies that the former provides more electrostriction than the latter. The decrease in the relative association (R_A) values with increase in the concentration of the solutions points to the fact that the breaking up of the associated solvent molecules on addition of the solute takes place in all solvents. The variation of S_n^0 values with the amino acids predicts the degree of hard electrostrictive solvation, i.e, it represents the structural effect of the amino acid on the solvent in the solution.

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