Viscometric and Compressibility behavior of Arginine in Aqueous-Glucose solutions

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Abstract— Densities , Ultrasonic Speeds and Viscosity of aqueous-glucose (5, 10, and 15% of glucose, in water) and of solutions of Arginine in three aqueous-glucose solvents were measured at 303, 308, and 313 K. From these experimental data Compressibility, Apparent Molar Compressibility , Limiting Apparent Molar Compressibility and the Slope , Transfer Compressibility , Falkenhagen coefficient, Jones Dole coefficient, Free energy of activation of Viscous flow per mole of solvent and Viscous flow per mole of solute were calculated. The results are interpreted in terms of solute_solvent and solute_ solute interactions in Arginine systems. It is observed that there exist strong solute_solvent interactions, which increases with increase in glucose concentration.

Index Terms— Arginine, Apparent Molar Compressibility, Transfer Compressibility, ultrasonic speeds, Viscosity.

1 INTRODUCTION

KNOWLEDGE of the interactions is responsible for stabilizing the native state of a globular protein in aqueous solution, which is essential to understand its structure and function. The study of these interactions provides an important insight into the conformational stability and unfolding behavior of globular proteins [1]. Hydration of proteins plays a significant role in the stability, dynamics, structural characteristics, and functional activity of these biopolymers. Since proteins are large complex molecules, the direct study of protein–water interactions is difficult. Therefore, one useful approach is to investigate interactions of the model compounds of proteins, i.e., amino acids in aqueous and mixed aqueous solutions [2],[3],[4],[5]. Since amino acids are the building blocks of all living organisms and incorporate structural features of proteins, their physicochemical and thermodynamic properties in aqueous solutions are found to provide valuable information on solute–solute and solute–solvent interactions that are important in understanding the stability of proteins. Some of these interactions are found implicated in several biochemical and physiological processes in a living cell [6]. The choice of water for preparing mixed solvent is important and unique role in determining the structure and stability of protein since its presence is known to give rise to hydrophobic forces [7], which are of prime importance in stabilizing the native globular structure of protein [8].

Due to complex structure of proteins, the study of conformational stability and unfolding behavior of globular proteins has proved quite challenging and still remains a subject of extensive investigations [9,10]. Therefore, protein model compounds such as amino acids, which are basic components of proteins, have been investigated in detail with respect to their thermodynamic properties in aqueous and mixed aqueous solutions [11], [12], [13], [14], [15], [16].

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Salt solutions are known to influence the stability and structure of proteins [13],[14]. Remarkable experimental work has been reported on the thermodynamic and transport properties of amino acids in aqueous salt solutions [11],[12], but very few studies exist on the volumetric and compressibility properties of amino acids in aqueous organic salt solutions [15], [16], [17], [18], [19], [20], [21], [22] probably due to the complex nature of their interactions. Moreover, no systematic studies are available on the thermodynamic and transport properties of amino acids having polar side group (chain) in the presence of organic salt solutions. To the best of our knowledge, no Viscometric and compressibility studies have been reported on Arginine in aqueous-glucose solutions.

In the present paper, we report the Densities (ρ), Ultrasonic speeds (u) and Viscosities (η) of aqueous-glucose (5, 10, and15% of glucose, in water) and of solutions of Arginine in three aqueous-glucose solvents were measured at 303, 308, and 313 K. From these experimental data Compressibility(β),Apparent Molar Compressibility ($K_{s,\phi}$), Limiting Apparent Molar Compressibility ($K°_{s,\phi}$) and the Slope (S_k), Transfer Compressibility ($K°_{s,\phi,tr}$), free energy of activation of Viscous flow per mole of solvent ($\Delta\mu_1^{\circ\#}$) and Viscous flow per mole of solute, ($\Delta\mu_2^{\circ\#}$) for Arginine in aqueous-glucose solutions were also calculated. The thermodynamics of viscous flow has also been discussed.

1 Experimental

Analytical reagent grade Arginine, used after recrystallization twice from (ethanol + water) mixtures. Glucose (analytical reagent) was dried over P_2O_5 in vacuum desiccators for 72 hrs at room temperature before use. Water used in the experiments was deionized and distilled, and was degassed prior to making solutions. Solutions of Glucose were prepared by mass and used on the day they were prepared. The mass percentage of Glucose in these solutions ranged from 5% to 10% by 15% increments. Solutions of amino acids were prepared by mass on the molality scale with an accuracy of 0 to .1 g. The density of all compounds was measured by 10 ml specific gravity bottle calibrated with double distilled water and ace-

tone.

The ultrasonic velocity was measured by a single crystal interferometer with a high degree of accuracy operating at a frequency of 3 MHz (Model F-05, with digital micrometer) at 303, 308 and 313K. The viscosity was measured by Ostwald's viscometer. An electronically operated constant temperature water bath is used to circulate water through the double walled measuring cell made up of steel containing the experimental solution at the desired temperature.

3 RESULTS AND DISCUSSIONS

The experimental values of Densities (ρ), ultrasonic speeds (u) and viscosities (η) of Arginine solutions in water and in aqueous-glucose solvents as functions of Arginine concentration and temperature are listed in Table 1.

The apparent molar compressibility, $K_{S,\phi}$ of the Arginine solutions in aqueous glucose were calculated by using the relations

$$K_{S,\varphi} = \frac{1000\,(k_s\rho_0 - k_s^0\rho)}{m\rho\rho_0} + \frac{k_s^0 M}{\rho} \quad (1)$$

where m is the molar concentration of the solute (Arginine), ρ and ρ_o are the densities of the solution and the solvent (aqueous-glucose), respectively; M is the molar mass of the solute (Arginine), and κ_s and $\kappa_s°$ are the isentropic compressibilities of the solution and the solvent (aqueous-glucose) respectively, calculated using the relation

$$k_s = \frac{1}{u^2\rho} \quad (2)$$

It is observed that, for Arginine in all the three aqueous-glucose solvents, $K_{s,\phi}$ is linear in the studied concentration range and at each investigated temperature (Table 2).

The values of $K_{s,\phi}$ are negative for Arginine aqueous-glucose solutions, indicating that the water molecules around ionic charged groups of amino acids are less compressible than the water molecules in the bulk solution [22]. The values $K°_{s,\phi}$ were obtained using the relations [12]

$$K_{S,\varphi} = K_{S,\varphi}^0 + S_K m \quad (3)$$

where the intercepts, $K°_{s,\phi}$, by definition are free from solute–solute interactions and therefore provide a measure of solute–solvent interactions, whereas the experimental slope S_k provides information regarding solute–solute interaction. The values of $K°_{s,\phi}$, and S_k for Arginine in aqueous-glucose solutions at different temperatures are given in Table 3.

Limiting apparent molar properties provide qualitative as well as quantitative information regarding solute–solvent interactions without taking into account the effects of solute–solute interactions [20]. In general, the types of interactions occurring between Arginine and glucose can be classified as follows [15], [16], [21]:

(a) The hydrophilic–ionic interaction between OH groups of glucose and zwitterions of Arginine.

(b) Hydrophilic–hydrophobic interaction between the OH groups of glucose molecule and non-polar (–CH_2) in side chain of Arginine molecule.

A perusal of Table (3) reveals that the values of $K°_{s,\phi}$, S_k are negative for Arginine in aqueous-glucose solutions indicating the presence strong solute–solvent interactions in these systems. The trends observed in $K°_{s,\phi}$ values can be due to their hydration behavior [13],[14],[15],[16],[17] which comprises of following interactions in these systems: (a) The terminal groups of zwitterions of amino acids, NH_3^+ and COO^- are hydrated in an electrostatic manner whereas, hydration of R group depends on its nature, which may be hydrophilic, hydrophobic or amphiphilic; and (b) the overlap of hydration co-spheres of terminal NH_3^+ and COO^- groups and of adjacent groups results in volume change. The $K°_{s,\phi}$ values increase due to reduction in the electrostriction at terminals, whereas it decreases due to disruption of side group hydration by that of the charged end.

The increase in $K°_{s,\phi}$ values with increase in temperature for Arginine in aqueous-glucose solutions can be explained by considering the size of primary and secondary solvation layers around the zwitterions. At higher temperatures the solvent from the secondary solvation layer of Arginine zwitter ions is released into the bulk of the solvent, resulting in the expansion of the solution, as inferred from larger $K°_{s,\phi}$, S_k values at higher temperatures [18], [19].Similar trends have also been reported [12] for amino acids in aqueous-glucose solutions.

This further supports the conclusion that the hydrophilic–ionic group interactions between OH groups of glucose with zwitter ions dominate in these systems. The values of $K°_{s,\phi}$ show a minimum for lower glucose concentrations (between 0 and 5%) at higher temperatures (313K). This increase in $K°_{s,\phi}$ may be due to higher expansion of solutions for lower glucose concentrations, as the interactions are less pronounced at lower glucose concentration range as mentioned above. The values of $K°_{s,\phi}$ increase with increase in temperature, indicating release of more water molecules from the secondary solvation layer of Arginine zwitterions into the bulk, thereby, are making the solutions more compressible.

The compressibility of transfer of Arginine from water to aqueous-glucose solutions, $K°_{s,\phi,tr}$ were calculated by using the relation

$$K_{s,\varphi,tr}^0 = K_{s,\varphi,aq,glucose}^0 - K_{s,\varphi,water}^0 \quad (4)$$

where $K°_{s,\phi,water}$ is the limiting apparent molar volume of Arginine in water. The $K°_{s,\phi,tr}$ values for Arginine from water to aqueous glucose solutions are included in Table 3. Table 3 indicates that $K°_{s,\phi,tr}$ values are positive. The observed positive $K°_{s,\phi,tr}$ values suggest that the hydrophilic–ionic groups interactions dominate in these systems. The $K°_{s,\phi,tr}$ values increase with increase in glucose concentration in the solutions (Table 3). This may be due to greater hydrophilic–ionic group interactions with increased concentrations of glucose. The observed trends in $K°_{s,\Phi}$ and $K°_{s,\phi,tr}$ further support the conclusions.

The viscosity data were analyzed by using Jones–Dole equation of the form

$$\eta_r = \frac{\eta}{\eta_0} = 1 + Am^{1/2} + Bm \quad (5)$$

where η_r is the relative viscosity of the solution, η and η_o

are the viscosities of solution and the solvent (aqueous–glucose), respectively, A and B are the Falkenhagen and Jones–Dole coefficients, respectively. Coefficient A accounts for the solute–solute interactions and B is a measure of structural modifications induced by the solute– solvent interactions [20], [21]. The values of A and B are listed in Table 4. The values of A- and B-coefficients are positive, however, the A-coefficients are much larger in magnitude as compared to B-coefficients, suggesting weak solute–solute and strong solute–solvent interactions in these solutions. The B-coefficients values increase with increasing concentration of glucose also indicate a structure to allow the co-solute (glucose) to act on solvent [11].

B-coefficients increase when the water is replaced by glucose, i.e., glucose modifies water structure through H-bonding [11]. The B-coefficients increase with rise in temperature indicating increased solute–solvent interactions at higher temperatures in these systems. Thus, the values of coefficients A and B support the behaviors of $K°_{s,\Phi}$, and $K°_{s,\phi,tr}$, which suggest strong solute–solvent interactions as compared to solute–solute interactions in these solutions.

The viscosity data have also been examined in the light of transition state theory of the relative viscosity proposed by [20], [22]. According to this theory, the B-coefficient is given by the following relation

$$B = \frac{(\bar{V}_1^0 - \bar{V}_2^0) + \bar{V}_1^0 (\Delta\mu_2^{0\#} - \Delta\mu_1^{0\#})/RT}{1000} \quad (6)$$

where $V°_1$ is the apparent (partial) molar volume of the solvent (aqueous–glucose) and $V°_2$ (= $V°_\Phi$) is the limiting apparent (partial) molar volume of the solute, respectively. The free energy of activation per mole of solvent ($\Delta\mu_1^{\circ\#}$) has been calculated by using the Eyring viscosity relation

$$\Delta\mu_1^{0\#} = RT \ln\left(\frac{\eta_0 \bar{V}_1^0}{hN}\right) \quad (7)$$

where h and N are Planck's constant and Avogadro number, respectively. Eq. (9) rearranges to give free energy of activation per mole of the solute, $\Delta\mu_2^{\circ\#}$

$$\Delta\mu_2^{0\#} = \Delta\mu_1^{0\#} + \frac{RT}{\bar{V}_1^0}\left[1000\,B - (\bar{V}_1^0 - \bar{V}_2^0)\right] \quad (8)$$

The values of $\Delta\mu_1^{\circ\#}$ and $\Delta\mu_2^{\circ\#}$ are included in Table 5. It is evident from Table 5 that for Arginine in aqueous-glucose solutions, the $\Delta\mu_2^{\circ\#}$ values are positive and much larger than those of $\Delta\mu_1^{\circ\#}$ in aqueous-glucose solvents.

This suggests that the interactions between Arginine and solvent (aqueous-glucose) molecules in the ground state are stronger than in the transition state. Hence, in the transition state the solvation of the solute molecules is less favored in free energy terms.

The $\Delta\mu_2^{\circ\#}$ values increase with increase in temperature, indicating that solute–solvent interaction increase with rise in temperature making the flow of solute molecules difficult. Thus, the conclusions drawn from $\Delta\mu_2^{\circ\#}$ are in agreement with those drawn from the trends of, $K°_{s,\Phi}$, $K_{,\Phi,tr}$ and B values.

4 Conclusion

The results are interpreted in terms of solute–solvent and solute– solute interactions in Arginine systems. It is observed that there exist strong solute–solvent interactions, which increases with increase in glucose concentration. It is observed that Arginine act as structure-breaker in aqueous-glucose solvents. The thermodynamics of viscous flow has also been discussed. This suggests that the interactions between Arginine and solvent (aqueous-glucose) molecules in the ground state are stronger than in the transition state.

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Table 1

Densities, Ultrasonic Speeds and Viscosities

m (mol/Kg)	Arginine in 5% & Glucose 95%			Arginine in 10% & Glucose 90%			Arginine in 15% & Glucose 85%		
	303 K	308 K	313 K	303 K	308 K	313 K	303 K	308 K	313 K
	Densities (ρ Kg/m^3)								
0	1013.45	1011.51	1009.00	1032.72	1030.48	1028.19	1049.12	1048.39	1046.19
0.0195	1014.30	1012.26	1010.19	1033.14	1031.24	1029.41	1050.91	1049.90	1047.22
0.0402	1015.39	1013.76	1012.58	1034.47	1032.71	1030.19	1051.12	1050.11	1048.31
0.0597	1016.71	1015.10	1014.74	1035.72	1033.67	1031.77	1052.22	1051.63	1049.14
0.0804	1017.53	1016.45	1015.32	1036.19	1034.11	1032.84	1053.41	1052.21	1050.71
0.1033	1018.29	1017.17	1016.19	1037.94	1035.40	1033.91	1054.88	1053.47	1051.42
	ultrasonic speeds (U m/s)								
0	1525.60	1535.10	1545.30	1541.10	1551.40	1560.90	1552.20	1561.70	1570.60
0.0195	1526.90	1536.70	1546.40	1542.70	1552.20	1561.10	1553.70	1562.20	1571.10
0.0402	1528.40	1537.90	1547.90	1543.40	1553.90	1562.40	1554.40	1563.30	1572.20
0.0597	1529.10	1538.40	1548.30	1544.60	1554.60	1563.70	1556.10	1564.40	1573.60
0.0804	1530.40	1539.30	1549.20	1545.30	1555.10	1564.90	1557.30	1656.70	1574.40
0.1033	1531.90	1540.40	1550.30	1546.20	1556.40	1565.30	1558.20	1566.10	1575.90
	viscosities (η x 10^{-3} Ns/m^2)								
0	0.8604	0.7696	0.6944	0.9906	0.8843	0.7902	1.1467	1.0161	0.8941
0.0195	0.8694	0.7714	0.7067	1.0021	0.8910	0.8004	1.1551	1.0223	0.9004
0.0402	0.8784	0.7832	0.7144	1.0142	0.9014	0.8121	1.1617	1.0385	0.9183
0.0597	0.8812	0.7917	0.7229	1.0271	0.9172	0.8241	1.1742	1.0445	0.9223
0.0804	0.8951	0.8014	0.7361	1.0381	0.9266	0.8373	1.1810	1.0561	0.9337
0.1033	0.9028	0.8148	0.7453	1.0418	0.9317	0.8414	1.1941	1.0671	0.9491

Table 2

Adiabatic Compressibility, Apparent Molar Compressibility & Relative Viscosity

m (mol/Kg)	Arginine in 5% & Glucose 95%			Arginine in 10% & Glucose 90%			Arginine in 15% & Glucose 85%		
	303 K	308 K	313 K	303 K	308 K	313 K	303 K	308 K	313 K
	Adiabatic compressibility (β x 10^{-10} m^2/N)								
0	4.2395	4.1952	4.1503	4.0771	4.0319	3.9918	3.9562	3.9109	3.8748
0.0195	4.2287	4.1834	4.1395	4.0670	4.0248	3.9861	3.9418	3.9028	3.8686
0.0402	4.2159	4.1706	4.1217	4.0581	4.0102	3.9764	3.9375	3.8965	3.8591
0.0597	4.2065	4.1624	4.1108	4.0469	4.0029	3.9637	3.9248	3.8854	3.8492
0.0804	4.1960	4.1520	4.1037	4.0469	3.9986	3.9536	3.9143	3.8768	3.8396
0.1033	4.1847	4.1432	4.0944	4.0299	3.9870	3.9474	3.9043	3.8702	3.8297
	Apparent Molar Compressibility $K_{s,\phi}$ /(10^{-11} m^{5}N^{-1}mol^{-1})								
0.0195	-7.2508	-7.5466	-7.9602	-5.8290	-5.9192	-6.0201	-4.9142	-5.2219	-5.4267
0.0402	-7.1915	-7.4759	-7.8381	-5.7732	-5.8381	-5.9792	-4.8238	-5.1019	-5.3631
0.0597	-7.0932	-7.3231	-7.7191	-5.6267	-5.7181	-5.8381	-4.7173	-5.0912	-5.2212
0.0804	-7.0519	-7.2521	-7.6323	-5.5510	-5.6763	-5.7081	-4.6001	-5.0027	-5.1917
0.1033	-7.0091	-7.1002	-7.5119	-5.4243	-5.5790	-5.6382	-4.5508	-4.9214	-5.0092
	Relative Viscosity (η_r x 10^{-3} Ns/m^2)								
0.0195	0.0749	0.0667	0.0585	0.0882	0.0773	0.0645	0.0992	0.0819	0.0757
0.0402	0.0812	0.0717	0.0612	0.0912	0.0831	0.072	0.1214	0.0905	0.0805
0.0597	0.0971	0.0823	0.0573	0.1291	0.0952	0.084	0.2924	0.1215	0.0917
0.0804	0.1213	0.0941	0.0424	0.2914	0.1257	0.0996	0.3067	0.2028	0.1724
0.1033	0.1309	0.1221	0.0313	0.3039	0.2342	0.1215	0.4757	0.3537	0.2315

Table 3

Limiting Apparant Molar Compressibility, Slope, Transfer Compressibility

Arginine in Glucose	Limiting Apparant Molar Compressibility ($-K^0_{s,\phi}$ x10^{-11} m^{3}N^{-1}mol^{-1})			Slope (- S_K x10^{-11} m^{5}N^{-1}mol^{-1})			Transfer Compressibility ($K^0_{s,tr}$ x10^{-11} m^{5}N^{-1}mol^{-1})		
	303 K	308 K	313 K	303 K	308 K	313 K	303 K	308 K	313 K
5%	7.677	8.288	8.761	0.986	1.312	2.190	0.68	0.49	0.36
10%	5.377	5.685	6.296	1.540	2.944	3.658	0.88	0.69	0.49
15%	4.425	4.771	5.219	2.279	3.257	4.124	1.10	0.95	0.82

Table 4

Falkenhagen and Jones – Dole Coefficient

Arginine in Glucose	Falkenhagen A/(10^{-5}Kg$^{1/2}$.mol$^{-1/2}$)			Jones – Dole Coefficient B/(10^{-4}Kg.mol^{-1})		
	303 K	308 K	313 K	303 K	308 K	313 K
5%	1.1770	1.0957	1.0625	0.4328	0.8807	0.9933
10%	1.0950	1.0940	1.0530	0.4713	0.9807	1.0575
15%	1.0890	1.0730	1.0457	0.4954	0.1833	1.1938

Table 5.

Free Energies of Activation of Viscous Flow per Mole of Solvent and Solute

Arginine in Glucose	Solvent $\Delta\mu_1^{\circ\,\#}$/(kJ·mol^{-1})			Solute $\Delta\mu_2^{\circ\,\#}$/(kJ·mol^{-1})		
	303 K	308 K	313 K	303 K	308 K	313 K
5%	8.75	8.46	8.3	363.27	436.55	512.77
10%	9.15	8.85	8.72	409.02	486.98	558.87
15%	10.25	9.95	9.55	445.1	512.9	584.53