

Volumetric and Viscometric Behavior of Arginine in Aqueous-Glucose Solutions at Different Temperatures

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Abstract

Densities and viscosities of Arginine in (glucose + water) mixed solvents have been measured at 303,308&313 K by an oscillating-tube densimeter and Viscometer. Adiabatic compressibility (β), Standard-state partial molar volume(v_{ϕ}^0), hydration number(H_n), Relative Viscosity (η_r) Limiting Apparent Molar Volume (v_{ϕ}^0), Slope s_v and Falkenhagen Coefficient(A) viscosity B-coefficients or Jones – Dole Coefficient (B) of Arginine have been calculated. The transfer volumes from water to (glucose + water) mixed solvents have been obtained and discussed in terms of the structural hydration interaction model. The results indicate that the partial molar volumes of transfer and viscosity B-coefficients of Arginine increase with increasing the mass concentration of glucose, and the hydration number of Arginine increases owing to the interaction of glucose. It is concluded that the magnitude of the enhancement effect on volume and hydration number is related to the number of OH groups and the structure of mixture solvent.

KEYWORDS: Arginine; Aqueous glucose solutions; Partial molar volumes; Viscosity A, B-coefficients.

Introduction

The physicochemical and thermodynamic properties of amino acids are of considerable interest, as these biomolecules are the building blocks of all living organisms, and are found to provide valuable information that leads to a better understanding of proteins (Banipal T S et al., 2008; Pal A et al.,2005; Nain A K et al.,2009; Zhao C et al.,2005; Zhuo K et al.,2006). Since Proteins are large complex molecules, the direct study of protein interactions is difficult. Therefore, one useful approach is to investigate interactions of the model compounds of proteins, e.g., amino acids, in aqueous and mixed-aqueous solutions (Banipal T S et al., 2008; Pal A et al.,2005; Nain A K et al.,2009; Zhao C et al.,2005; Zhuo K et al.,2006). It has also been recognized that in the absence of experimental thermodynamic data of proteins, amino acids can serve as useful models in estimating their properties (Yan Z et al.,2004). The choice of water for preparing mixed solvent stems from its important and unique role in determining the structure and stability of protein. since its presence give rise to hydrophobic forces (Banipal P K et al.,2002) which are of prime importance in stabilizing native globular structure of protein (Thirumaran S et al.,2009). It is known (Li S et al.,2002) that polyhydroxy compounds (carbohydrates) helps in stabilizing the native globular structure of protein and reduce the extent of their denaturation by other substances. Carbohydrates located at cell surfaces, are important as receptors for the bioactive structures of enzymes, hormones, viruses, antibodies, etc. (Banipal TS et al.,2007).The protein-carbohydrate interactions are important for

immunology, biosynthesis, pharmacology, medicine and cosmetic industry (Parfenyuk E V et al., 2004), however, these are not components of living organisms, but they act as a vehicle for pharmaceuticals or cosmetics when introduced into living organisms (Bai T C et al., 2003). Thus, the properties of amino acids in aqueous carbohydrate solutions are important for understanding the chemistry of biological systems (Li S et al., 2002, Pal A et al., 2009). There have been a number of physicochemical studies of some amino acids in aqueous-carbohydrate solutions (Kulikova G A et al., 2008; Ali A, Hyder S et al., 2006; Ali A, Sabir S, et al., 2006; Ali A, Khan S et al., 2006; Ali A, et al., 2007). To the best of our knowledge, no Volumetric, ultrasonic and Viscometric 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, 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, Adiabatic compressibility (β), Standard-state partial molar volume (v_ϕ), hydration number (H_n), Relative Viscosity (η_r) Limiting Apparent Molar Volume (v_ϕ^0), Slope (s_v) and Falkenhagen Coefficient (A) viscosity B-coefficients or Jones – Dole Coefficient (B) of Arginine have been calculated.

Materials and Methods

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 sucrose 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 densities of solutions were measured using a single stem pycnometer having a total volume of 8 cm^3 and an internal diameter of the capillary of about 0.1 cm. The details pertaining to calibration, experimental setup and operational procedures have been described in earlier paper (Badarayani R et al., 2005). The pycnometer was calibrated at $T = (303, 308, 313\text{ K})$ with twice distilled water (Badarayani R et al., 2003) which was filled to the marks on the stem. Its working was checked by measuring the densities of aqueous NaCl solutions at 298.15 K and good agreement was found with the literature values (Bhat I et al., 2004 ; Lark B S et al., 2004). The mass of the dried pycnometer along with a Teflon cap was first taken. Subsequently, the test solution was introduced into it by means of a hypodermic syringe. Sufficient care was taken to avoid any air bubble entrapment. The limb of the pycnometer was then closed with Teflon cap and weighed. The pycnometer with the test solution was equilibrated in a water bath ($\pm 0.01\text{ K}$) of the desired temperature until no further change in the level of the solution in the capillary was observed. This level was noted and used in the calculation of density. Further readings on the same solution were taken at the other temperature, achieved by putting the pycnometer in two separate baths maintained at the required temperatures. The evaporation losses remained insignificant during the time of actual measurements. 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 313 K. 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.

Results and discussion

The measured values of density for Arginine in aqueous glucose solutions are given in table 1. The apparent molar volume V_{ϕ} were calculated from the equation

$$V_{\phi} = \frac{1000(\rho_0 - \rho)}{m\rho\rho_0} + \frac{M}{\rho} \quad \dots(1)$$

Where m is the molar concentration of the solute (Arginine), ρ and ρ_0 are the densities of the solution and the solvent (aqueous-glucose), respectively; M is the molar mass of the solute (Arginine). Since, for Arginine studied, the apparent molar volume was found to be a linear function of the molality over the range studied, the standard-state (infinite dilution) partial molar volumes

$$V_{\phi} = V_{\phi}^0 + S_v m \quad \dots(2)$$

Where the intercepts, V_{ϕ}^0 by definition are free from solute-solute interactions and therefore provide a measure of solute-solvent interactions, whereas the experimental slope, S_v provides information regarding solute-solute interaction. The values of V_{ϕ} , V_{ϕ}^0 , S_v , along with β for Arginine in aqueous-glucose solutions at different temperatures are given in Table 1,2.

$$V_{\phi, tr}^0 = V_{\phi, aq, glucose}^0 - V_{\phi, water}^0 \quad \dots(3)$$

Where $V_{\phi, water}^0$ is the limiting apparent molar volume of Arginine in water. The $V_{\phi, tr}^0$ values for Arginine from water to aqueous-glucose solutions are included in Table 2 indicates that V_{ϕ}^0 of Arginine in aqueous-glucose positive.

The value of $V_{\phi, tr}^0$ is by definition free from solute - solute interactions and therefore provides information regarding solute-solvent interactions (Yan Z et al.,2004). In Table 2 the values of $V_{\phi, tr}^0$ for Arginine are positive and increase monotonically with the mass fractions of glucose in aqueous glucose solutions. These results can be explained by the co-sphere overlap model. (Friedman et al.,1973). Properties of the water molecules in the hydration cosphere depend on the nature of the solute species. The types of the interaction occurring between the Arginine and glucose molecules can be classified as (Nain A K et al.,2009):

- (1) Hydrophilic-ionic group interactions between the OH groups of glucose, and the zwitterionic center of Arginine.
- (2) Hydrophilic-hydrophilic group interactions between the OH groups of glucose NH_2 groups of Arginine mediated through the hydrogen bonding.
- (3) Hydrophilic-hydrophobic group interactions between the OH groups of glucose and the non-polar ($-\text{CH}_2$) groups of the Arginine.
- (4) Hydrophobic-hydrophobic group interactions between the non-polar groups of glucose, and the non-polar ($-\text{CH}_2$) groups of the Arginine.

The interaction of type (1) leads to a positive contribution to the transfer volume owing to the overlap of the hydration cosphere of the ion (COO^- and NH_3^+) and a hydrophilic OH group, which leads to a decrease in the structure-breaking tendency of the ion and a reduction in the electrostriction of the water caused by these ions. The

interaction type (2) also makes a positive contribution to the transfer volume, since the overlap of the hydration co-sphere of NH₂ and OH groups leads to a increase in the magnitude of the hydrogen bonding interaction. On the contrary, the interaction of type (3) between the OH groups of glucose, and the non-polar (-CH₂) groups of the Arginine leads to a decrease in the transfer volume as a result of their co-sphere overlap. The interaction type (4) also makes a negative contribution to the transfer volume.

The observed increasing positive transfer volumes in glucose mixed solvents suggest that in the ternary solutions, the ion-hydrophilic and hydrophilic-hydrophilic group interactions are predominant over hydrophilic-hydrophobic group interactions. It may be inferred that in the ternary solution, the increased concentrations of the glucose lead to the greater ion-hydrophilic and hydrophilic – hydrophilic interactions that are not compounded by hydrophilic-hydrophobic interactions.

The standard partial molar volumes of Arginine can be expressed from a simple model (Pal A et al.,2005)

$$V_{\phi}^0 = V_{\phi}^0(\text{int}) + V_{\phi}^0(\text{elect}) \quad \dots(4)$$

Where $V_{\phi}^0(\text{int})$ is the intrinsic partial molar volume of Arginine, and $V_{\phi}^0(\text{elect})$ is the electrostriction partial molar volume due to the hydration of Arginine. The $V_{\phi}^0(\text{int})$ is made up of two terms, the van der Waals volume and the volume due to packing effects. According to the suggestion of Millero et al.,1978, the values of $V_{\phi}^0(\text{int})$ for Arginine can be estimated from crystal molar volumes

$$V_{\phi}^0(\text{int}) = \left(\frac{0.7}{0.634}\right) V_{\phi}^0(\text{crystal}) \quad \dots(5)$$

Where 0.7 is the packing density for molecules in organic crystals, and 0.634 is the packing density for random packing spheres. The crystal molar volume can be calculated by the following equation:

$$V_{\phi}^0(\text{crystal}) = \frac{M_s}{\rho(\text{crystal})} \quad \dots(6)$$

where $V_{\phi}^0(\text{cryst})$ is the crystal density of arginine (Ali A, et al.,2007). The $V_{\phi}^0(\text{elect})$ can be estimated from the experimentally measured V_{ϕ}^0 values .

$$V_{\phi}^0(\text{elect}) = V_{\phi}^0 - V_{\phi}^0(\text{int}) \quad \dots(7)$$

The decrease in volume due to electrostriction can be related to the number of water molecules (N_H) hydrated to arginine (Badarayani R et al.,2005)

$$N_H = V_{\phi}^0(\text{elect}) / (V_E^0 - V_B^0) \quad \dots(8)$$

where V_E^0 is the molar volume of electrostricted water and V_B^0 is the molar volume of bulk water at T = 298.15 K. Following the procedure described by Millero et al. 1978.

$$(V_E^0 - V_B^0) = -3.3 \text{ cm}^3 \cdot \text{mol}^{-1} \quad \dots(9)$$

Therefore, as an approximation, the hydration number of water molecules can be obtained as

$$N_H = V_\phi^0(\text{elect})/(-3.3) \quad \dots(10)$$

The N_H value of Arginine in aqueous glucose solutions is shown in Table 1. It can be seen that N_H of Arginine increases with increasing concentration of glucose. This shows that glucose have a dehydration effect on the Arginine. The viscosity data for the Arginine in glucose, at $T = 303, 308, 313$ K are collected in tables 1. The relative viscosity η_r can be represented as

$$\eta_r = \frac{\eta}{\eta_0} = 1 + Am^{1/2} + B \quad \dots(11)$$

Where η_r is the relative viscosity of the solution, η and η_0 are the viscosities of solution and the solvent (aqueous–glucose), respectively, A and B are the Falkenhagen and Jones–Dole coefficients, respectively (Table 3). Coefficient A accounts for the solute–solute interactions and B is a measure of structural modifications induced by the solute–solvent interactions.

The viscosity B-coefficient is valuable to provide information concerning the solvation of the solutes and their effects on the structure of the solvent in the near environment of the solute Molecules. Larger and positive B values indicate a structure to allow the solute to act on solvents. From table 3, Viscosity B-coefficients increase with increasing concentration of glucose, and the Coefficient A decrease with increasing concentration of glucose. The reason is that the friction increases to prevent water flowing with increasing concentration of glucose.

Conclusion

The results indicate that the partial molar volumes of transfer and viscosity B-coefficients of Arginine increase with increasing the mass concentration of grouse, and the hydration number of Arginine increases owing to the interaction of grouse. It is concluded that the magnitude of the enhancement effect on volume and hydration number is related to the number of OH groups and the structure of mixture solvent.

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Table (1)

Densities (ρ), viscosities (η), Adiabatic compressibility [$\beta \times 10^{-10} \text{ m}^2/\text{N}$], Apparent Molar Volume [$V_\phi / 10^{-6} \text{ m}^3 \text{ mol}^{-1}$] Hydration Number [H_n], and Relative Viscosity [$\eta_r \times 10^{-3} \text{ N s/m}^2$] of Arginine solutions with glucose + water (5, 10, and 15% of glucose, in water) solutions under different temperature.

m (mol/Kg)	Arginine in 5% & Glucose 95%			Arginine in 10% & Glucose 90%			Arginine in 15% & Glucose 85%		
	[$\rho \text{ Kg/m}^3$]								
	303 K	308 K	313 K	303 K	308 K	313 K	303 K	308 K	313 K
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
	[$\eta \times 10^{-3} \text{ N s/m}^2$]								
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
	[$\beta \times 10^{-10} \text{ m}^2/\text{N}$]								
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
	[$V_\phi / 10^{-6} \text{ m}^3 \text{ mol}^{-1}$]								
0.0195	-0.0422	-0.0474	-0.0597	-0.0400	-0.0465	-0.0589	-0.0331	-0.0402	-0.0480
0.0402	-0.0467	-0.0544	-0.0869	-0.0405	-0.0519	-0.0668	-0.0349	-0.0487	-0.0497
0.0597	-0.0528	-0.0584	-0.0937	-0.0468	-0.0599	-0.0694	-0.0369	-0.0491	-0.0548
0.0804	-0.0580	-0.0596	-0.0965	-0.0502	-0.0622	-0.0743	-0.0381	-0.0529	-0.0569
0.1033	-0.0592	-0.0631	-0.1677	-0.0569	-0.0644	-0.0799	-0.0402	-0.0544	-0.0598

	[H _n]								
0.0195	0.0226	0.025	0.0281	0.042	0.0499	0.0542	0.0867	0.0993	0.1095
0.0402	0.0495	0.0521	0.0613	0.079	0.0913	0.0954	0.1126	0.1877	0.1965
0.0597	0.0692	0.0695	0.0846	0.1256	0.1319	0.1494	0.189	0.1953	0.2574
0.0804	0.0912	0.0916	0.0998	0.1485	0.1491	0.1623	0.2523	0.2677	0.3164
0.1033	0.1149	0.1152	0.1297	0.1963	0.2088	0.2886	0.3125	0.3479	0.4772
	[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 (2)

Limiting Apparant Molar Volume [$V_{\phi}^0 / 10^{-6} \text{ m}^3 \text{ mol}^{-1}$] Slope [$S_v / 10^{-6} \text{ m}^3 \text{ mol}^{-1} \text{ Kg}^{-1}$] and Transfer Volume [$V_{\phi, \text{tr}}^0 / 10^{-6} \text{ m}^3 \text{ mol}^{-1}$] of arginine solutions with glucose + water (5, 10, and 15% of glucose, in water) solutions under different temperature.

Arginine in Glucose	[$V_{\phi}^0 / 10^{-6} \text{ m}^3 \text{ mol}^{-1}$]			[$S_v / 10^{-6} \text{ m}^3 \text{ mol}^{-1} \text{ Kg}^{-1}$]			[$V_{\phi, \text{tr}}^0 / 10^{-6} \text{ m}^3 \text{ mol}^{-1}$]		
	303 K	308 K	313 K	303 K	308 K	313 K	303 K	308 K	313 K
5%	-0.0415	-0.0506	-0.0617	-0.0240	-0.0423	-0.0612	1.14	1.54	2.02
10%	-0.0421	-0.0529	-0.0652	-0.0367	-0.0591	-0.0682	1.50	1.86	2.61
15%	-0.0457	-0.0639	-0.0691	-0.0481	-0.0599	-0.0690	2.19	2.23	3.30

Table (3)

Falkenhagen Coefficient, [$A / 10^{-5} \text{ Kg}^{1/2} \cdot \text{mol}^{-1/2}$] and Jones – Dole Coefficient [$B / 10^{-4} \text{ Kg} \cdot \text{mol}^{-1}$] of arginine solutions with glucose + water (5, 10, and 15% of glucose, in water) solutions under different temperature.

Arginine in Glucose	[$A / 10^{-5} \text{ Kg}^{1/2} \cdot \text{mol}^{-1/2}$]			[$B / 10^{-4} \text{ Kg} \cdot \text{mol}^{-1}$]		
	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