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RESEARCH ARTICLE

VOLUMETRIC, VISCOMETRIC AND COMPRESSIBILITY STUDIES OF SOME AMINO ACIDS IN AQUEOUS MAGNESIUM NITRATE SOLUTION AT VARYING TEMPERATURES

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ABSTRACT

Measurements of density (ρ), viscosity (η), and ultrasonic velocity (u) were carried out on a-amino acids, L-lysine, L-glutamine, L-arginine (0.1– 0.5 mol kg $^{-1}$), in 0.2 mol kg^{-1} aqueous magnesium nitrate at 298.15, 303.15, 308.15,and 313.15 K. These measurements were carried out to evaluate some important parameters, viz., adiabatic compressibility (β), apparent molar compressibility (φ_K), apparent molar volume (φ_V), limiting apparent molar compressibility (φ_K^0), limiting apparent molar volume (φ_V^0), and their constants (S_K, S_V) viscosity *A* and *B* coefficients of Jones-Dole equation. These parameters were interpreted in terms of solute-solute and solute-solvent interactions and structure-making/breaking ability of solutes in the given solution.

INTRODUCTION

Measurement of ultrasonic velocity, density and viscosity values of amino acids in aqueous electrolyte have been of interest with a view to improve the comprehension about the stability of native proteins and the equilibrium process between folded *versus* unfolded forms of proteins, such as protein hydration, denaturation, and protein aggregation (Yan et al. 2004; Millero et al., 1998).The physicochemical behaviour of amino acids in aqueous as well as aqueous-electrolyte (Badarayani and Kumar, 2002) and carbohydrate (Hindley et al., 2005), media. Amino acids when dissolved in water convert into zwitterionic form because of ionization of their carboxyl ($COOH^-$) and amino (NH_3^+) groups. Salts have a negative effect on the properties and structural stability of proteins that includes their solubility, denaturation, dissociation into subunits and activity of enzymes (Jeneks, 1969). Magnesium in biological chemistry has been branded as a Cinderella element because of its double behaviour (Frausto da Silva, and Willams, 1993). Magnesium is very important for a number of biological functions in the human body. In this study, density (ρ), viscosity (η), and ultrasonic velocity (u) values of L-lysine, L-glutamine, L-arginine (0.1– 0.5 mol kg $^{-1}$), in 0.2 mol kg $^{-1}$ aqueous magnesium nitrate solution at 298.15, 303.15, 308.15, and 313.15 K. These measurements were carried out to evaluate some important parameters, viz., adiabatic compressibility(β), apparent molar compressibility(φ_K), apparent molar volume(φ_V), limiting apparent molar compressibility (φ_K^0) , limiting apparent molar volume (φ_v^0), and their constants (S_K, S_V) viscosity A and B, coefficients of Jones-Dole equation respectively. These parameters were interpreted in terms of solute-solute and solute-solvent interactions occurring between amino acids and aqueous magnesium nitrate solution.

MATERIALS AND METHODS

The amino acids selected for the present study's L-glutamine, L-arginine and L-lysine were obtained from E-Merck, Germany and Sd fine chemicals, India. These, along with magnesium nitrate Sigma Aldrich,India, were used without further purification. All the solutions were prepared by weight. The 0.2 mol kg $^{-1}$ molal solution of magnesium nitrate in water was prepared and used as solvent to prepare 0.1, 0.2, 0.3, 0.4 and 0.5 mol kg^{-1} solutions of L-arginine and L-lysine and L-glutamine (0.2 mol kg^{-1} is the maximum solubility of Glutamine in 0.2 mol kg^{-1} [$Mg(NO_3)_2$] using doubly distilled and deionised water. The weighing was done on an electronic digital balance SHIMADZU AX200 (Swiss make) accurate up to 1.0×10^{-4}g. The densities of mixed solvents and solutions of amino acids in these solvents were measured using a specific gravity bottle by relative measurement method with an accuracy of ±0.01 kg m -3.

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An Oswald's viscometer (10 ml capacity) was used for Viscosity measurement and efflux time was determined using a digital chronometer to within ±0.01s. Sound speeds were determined with an uncertainty of 0.3% using a single crystal variable path ultrasonic interferometer (Mittal Enterprises, New Delhi, India) operating at 3 MHz which was calibrated with water, methanol and benzene at 303.15K. During the measurements the temperature of the test solution was maintained to an accuracy of ±0.01K in an electronically controlled thermostatic water bath (RAAGA INDUSTRIES, INDIA).

RESULTS

Adiabatic compressibility coefficients were derived from the following equation,

$$\beta = \frac{1}{U^2 \rho} \qquad \dots (1)$$

where ρ is the density and U is the velocity of sound in the solution.

The apparent molar compressibility φ_K of the electrolyte solutions was calculated from

$$\varphi_K = \frac{1000}{m\rho_0}(\beta\rho_0 - \rho\beta_0) + \left(\frac{\beta_0 M}{\rho_0}\right) \qquad \dots (2)$$

where β and β_0 are the adiabatic compressibility of solution and solvent, respectively. m is the molal concentration of solution, M is the molar mass of the solute and ρ and ρ_0 are the densities of solution and the solvent respectively. The limiting apparent molar adiabatic compressibility φ_K^0 values were obtained by extrapolating the plots of φ_K *versus* $m^{½}$ of the solution to zero concentration by a least-square method.

$$\varphi_K = \varphi_K{}^0 + S_K m^{½} \qquad \dots (3)$$

were S_K is the experimental slope.

The apparent molal volume φ_V values of the solution were calculated from the density using the following standard expression,

$$\varphi_V = \left(\frac{M}{\rho}\right) - \frac{1000(\rho - \rho_0)}{m\rho\rho_0} \qquad \dots (4)$$

where m is the molal concentration of solution and M is the molar mass of the solute and other symbols have their usual meaning.

The values of limiting apparent molar volumes φ_V^0 and experimental slopes S_V^* at different temperatures were obtained by least-squares from linear plots of φ_V against $C^{½}$ using the Masson equation (1929):

$$\varphi_V = \varphi_V^0 + S_V C^{½} \qquad \dots (5)$$

where S_V^* is a constant dependent on charge and salt type and can be related to ion-ion interactions and φ_V^0 is the limiting apparent molar volume which is related to ion-solvent interaction.

The viscosity data for amino acids in aqueous magnesium nitrate solutions were analysed using the Jones-Dole equation (1929).

$$\eta/\eta_0 = 1 + Am^{½} + Bm \qquad \dots (6)$$

where η_0 and η are the viscosities of solvent mixtures and solution, respectively. A is the constant characteristic of the salt, and B is the Jones-Dole coefficient; both are obtained from the plot of $(\eta/\eta_0 - 1)/C^{½}$ *versus* $C^{½}$.

DISCUSSION

The ultrasonic velocity, density and viscosity values of the L-glutamine or L-arginine or L-lysine + 0.2 mol·L^{-1} in aqueous magnesium nitrate systems have been measured experimentally as a function of molality of amino acid at T = 298.15, 303.15, 308.15 and 313.15K in Table 1. The ultrasonic velocity values increase with an increase in molality of amino acids in the 0.2 mol·L^{-1} aqueous magnesium nitrate solution (Table 1). Amino acid molecules in aqueous solution essentially behave as Zwitterions having NH_3^+ and COO^- groups at two ends of the molecule. The Mg^{2+}, NO_3^- ions furnished by $Mg(NO_3)_2$ in solutions stronger interact electrostatically with NH_3^+ and COO^- end groups of Zwitterions. These interactions comprehensively, introduce the cohesion into solutions understudy. These cohesive forces may further have enhanced on successive additions of amino acid amount in solutions (Rao and Vernal, 1987; Harata and Arakawa, 1972; Magazu et al. 1997; Rohnman and Mahiuddin, 1997; Ragouramane and Srinivasa Rao, 1998; Ernst and Manikowski, 1997). The ultrasonic velocity values for the studied systems increase with an increase in temperature. The rise in temperature may cause the thermal rupture of an icelike open structure to close packed structure of water. It seems that the cohension factor dominates over the thermal expansion factor in solution with an increase in temperature (Rohnman and Mahiuddin, 1997; Hall, 1948; Arakava and Sasiki, 1969). From the Table 2 it is observed the adiabatic compressibility values decrease with an increase in concentration (Table 2). The trend of variation of β_{ad} is in consonance with the variation of ultrasonic velocity with concentration (Hall, 1948; Arakava and Sasiki, 1969). The smaller values of β_{ad} for aqueous electrolyte solutions may be attributed to cation–water dipole and anion-water dipole interactions in water, which ultimately may lead to an overall increase in cohesive forces in water.

In fact, the negative values of apparent molar compressibility (φ_K) are attributed to the strong attractive interactions due to the hydration of ions and low temperatures (Table 2). By increasing temperature, ion-solvent interactions are weaken (Sadeghi and Baharak Goodavzi, 2008), and therefore, at a high temperature, the

Table 1 Values of Density (ρ), viscosity (η) and velocity (U) for temperature

DENSITY $\rho/(kg/m^3)$				**VISCOSITY** $\eta/(\times10^{-3}\ NSm^{-2})$				**VELOCITY** U/ms^{-1}	
Temperature (K)									
298.15	**303.15**	**313.15**	**323.15**	**298.15**	**303.15**	**313.15**	**323.15**	**298.15**	**303.15**
L-lysine + aqueous magnesium nitrate solution									
1016.60	1015.30	1012.50	1011.12	0.9482	0.8424	0.7623	0.6981	1512.80	1523.70
1019.91	1018.50	1015.70	1014.22	0.9604	0.8523	0.7701	0.7043	1526.10	1537.20
1022.61	1021.21	1018.31	1016.82	0.9726	0.8622	0.7779	0.7105	1533.30	1545.20
1025.41	1023.92	1020.92	1019.42	0.9848	0.8721	0.7857	0.7167	1540.50	1553.10
1028.12	1026.63	1023.53	1022.02	0.9970	0.8820	0.7935	0.7229	1547.70	1561.30
1030.75	1029.24	1026.04	1024.52	1.0092	0.8919	0.8013	0.7291	1554.90	1569.20
L-arginine + aqueous magnesium nitrate solution									
1016.60	1015.30	1012.50	1011.12	0.9482	0.8424	0.7623	0.6981	1512.80	1523.70
1019.30	1018.00	1015.10	1013.62	0.9955	0.8796	0.7929	0.7218	1524.60	1536.50
1021.51	1020.10	1017.20	1015.74	1.0428	0.9168	0.8235	0.7455	1531.70	1542.40
1023.72	1022.21	1019.32	1017.83	1.0901	0.9540	0.8541	0.7692	1536.10	1547.20
1025.93	1024.30	1021.40	1019.93	1.1374	0.9912	0.8847	0.7929	1542.20	1554.40
1028.04	1026.32	1023.41	1021.88	1.1847	1.0284	0.9153	0.8166	1549.10	1560.70
L-glutamine + aqueous magnesium nitrate solution									
1016.60	1015.30	1012.50	1011.12	0.9482	0.8424	0.7623	0.6981	1512.80	1523.70
1019.53	1017.94	1015.00	1013.53	0.9878	0.8746	0.7860	0.7208	1521.10	1533.10
1021.32	1019.81	1017.10	1015.71	1.0284	0.9124	0.8307	0.7538	1529.20	1541.90

Table 2 Values of Adiabatic compressibility (β), apparent molal compressibility (φ_k) and apparent molal volume (φ_V) for malality

Molality m ($mol.Kg^{-1}$)	Adiabatic compressibility (β)				Apparent molal compressibility $-\varphi_k(\times10^{-8}\ m^2\ N^{-1})$				Apparent molal volume $\varphi_v(\times m^2\ mol^{-1})$			
	Temperature (K)											
	298.15	**303.15**	**313.15**	**323.15**	**298.15**	**303.15**	**313.15**	**323.15**	**298.15**	**303.15**	**313.15**	**323.15**
L-lysine + aqueous magnesium nitrate solution												
0.0	4.2982	4.2424	4.1721	4.1028	…	…	…	…	…	…	…	…
0.1	4.2099	4.1551	4.0864	4.0123	10.2230	10.0590	9.8830	10.2961	11.1412	11.2589	11.2814	11.3911
0.2	4.1594	4.1012	4.0332	3.9686	8.2021	8.2840	8.1362	7.8600	11.4052	11.4654	11.5386	11.6051
0.3	4.1094	4.0489	3.9809	3.9255	7.5287	7.6435	7.5226	7.0266	11.4396	11.5136	11.6042	11.6564
0.4	4.0605	3.9959	3.9296	3.8830	7.1533	7.3390	7.1929	6.5942	11.4637	11.5223	11.6221	11.6671
0.5	4.0127	3.9457	3.8795	3.8309	6.9111	7.0991	6.9689	6.5259	11.4821	11.5357	11.6412	11.6820
L-arginine + aqueous magnesium nitrate solution												
0.0	4.2982	4.2424	4.1721	4.1028	…	…	…	…	…	…	…	…
0.1	4.2207	4.16090	4.0877	4.0260	8.8828	9.2663	9.4984	8.6849	14.4845	14.4997	14.6312	14.7466
0.2	4.1726	4.1206	4.0532	3.9809	7.3092	7.0818	6.9078	7.0244	14.6891	14.7595	14.8437	14.9009
0.3	4.1398	4.0866	4.0225	3.9366	6.2760	6.1454	5.9164	6.4400	14.7359	14.8222	14.8871	14.9415
0.4	4.0983	4.0406	3.9811	3.8929	5.9771	5.9763	5.6848	6.1330	14.7433	14.8432	14.9035	14.9439
0.5	4.0535	4.0001	3.9300	3.8504	5.8618	5.7651	5.7406	5.9227	14.7556	14.8581	14.9157	14.9642
L-glutamine + aqueous magnesium nitrate solution												
0.0	4.2982	4.2424	4.1721	4.1028	…	…	…	…	…	…	…	…
0.1	4.2392	4.1796	4.1045	4.0470	7.1249	7.3626	7.7805	6.5401	11.5080	11.8030	11.9664	12.0583
0.2	4.1871	4.1245	4.0567	3.9932	6.5486	6.8301	6.7122	6.4046	12.0369	12.1532	12.1359	12.1543

Table 3. Limiting apparent molal compressibility ($\varphi_K{}^0$) and limiting apparent molal volume ($\varphi_V{}^0$) and their constants S_K and S_V of amino acids

Amino Acids	$-\varphi_k{}^0(\times10^{-8}\ m^2\ N^{-1})$				$S_K/(\times10^{-8}\ N^{-1}\ m^{-1}\ mol^{-1})$				$\varphi_v{}^0(\times m^3\ mol^{-1})$				$S_V/(N^{-1}\ m^{-1}\ mol^{-1})$			
	Temperature (K)															
	298.15	**303.15**	**313.15**	**323.15**	**298.15**	**303.15**	**313.15**	**323.15**	**298.15**	**303.15**	**313.15**	**323.15**	**298.15**	**303.15**	**313.15**	**323.15**
L-lysine	1.2348	1.1976	1.1780	1.2698	8.1897	7.3372	7.2380	9.4999	10.98	11.38	11.40	11.50	7.557	-3.5352	-3.5525	-3.4796
L-arginine	1.1036	1.1479	1.1794	1.0476	7.8718	8.7337	9.5127	6.8519	14.35	14.31	14.49	14.64	6.1883	8.3830	6.5231	4.7023
L-glutamine	0.7580	0.8255	1.0340	0.5324	2.3003	3.1779	8.1041	-3.1023	11.31	11.41	11.54	11.57	16.2854	16.7831	13.4665	13.1381

Table 4. Falkenhagen coefficient A, Jones-Dole coefficient B of amino acids in aqueous magnesium nitrate solution at different temperatures

Amino acids	**A (× $dm^{-3/2}$ $m^{-1/2}$)**				**B (×dm^3 mol^{-1})**			
	Temperature (K)							
	298.15	**303.15**	**313.15**	**323.15**	**298.15**	**303.15**	**313.15**	**323.15**
L-lysine	-0.0130	-0.0118	-0.0103	-0.0090	0.2520	0.2301	0.2004	0.1739
L-arginine	-0.0503	-0.0445	-0.0405	-0.0342	0.9768	0.8647	0.7860	0.6648
L-glutamine	-0.0120	-0.0115	-0.0111	-0.0105	3.1704	3.0240	2.9360	2.7685

water molecule around the ions become more compressible than those at a low temperature (Millero et al. 1974). The extent of φ_K is in the order: L-glutamine > L-arginine > L-lysine. Table 3 contains the negative values of φ_K^0 suggests either strong solute-solvent interactions or a higher number of water molecules within the solvated shell of solute or combination of both. The observed negative values of φ_K^0 indicates strong magnesium nitrate solutions in the dilute region due to structure effects in ion-solvent interactions (Nashiour Rohman and Sekh Mahiuddin, 1997). The positive limiting experimental slopes (S_K) results (Table 3) can be interpreted in terms of the loss of compressibility of water due to electrostrictive forces in the vicinity of ions (Palani and Saravanan, 2008). The values of limiting apparent molar volume (φ_V^0) are positive(Table 3) for in L-glutamine, L-arginine and L-lysine indicating the presence of strong solute-solvent interactions (Yasmin Akhtar, 2007). The strong solute-solvent interactions are indicating the presence of strong interactions between NH_3^+ and COO^- for amino acids and aqueous magnesium nitrate and due to the electrostiction effect (Yasmin Akhtar, 2007).

The increases of φ_V^0 values with the increases in temperature may also be attributed to the reduction in electrostiction with temperature (Ali et al., 2007). The increase in the positive φ_V^0 values of the studied amino acids with the increase in the concentration of $Mg(NO_3)_2$ may further be attributed to the formation of non-covalent ion passes between the charged groups of the amino acids and the cation (Mg^{2+}) and the anion(NO^-) of the electrolyte. The increase in the apparent molar volume of the amino acids decrease the electrostiction of water around amino acids in the presence of $Mg(NO_3)_2$. Further, the formation of the ion pairs also decreases the hydrophobicity of amino acids molecules arising rom the interactions of the hydrocarbon portion of amino acids with water molecules (Hall, 1948). The increase in φ_V^0 values is in the order: L-arginine >L-glutamine > L-lysine indicating the trend of the strength of solute-solvent interactions.

Table 3 shows the values of S_V amino acids are found to be positive and less negative suggesting strong solute-solute interactions. The values of φ_V^0 for L-glutamine in aqueous magnesium nitrate are less than the values of S_V, indicating the solute-solvent interactions are greater than the solute-solute interactions in the systems. The values of φ_V^0 for L-lysine and L-arginine in aqueous magnesium nitrate are greater then the values of S_V, suggesting the solute-solute interactions are greater than the solute-solvent interaction in the system. The positive and less negative S_V values are generally associated with the solutes showing an overall hydrophilic character (Ali et al. 2007) as in the present investigation.

The value of the viscosity A-coefficient of the Jones-Dole equation is a measure of long range ion-ion interactions whereas B reflects the effect of ion-solvent interactions (Wadi and Ramasami, 1997). Table 4 the positive B-coefficients suggest Kosmostropes (structure-making) because strongly hydrated solutes exhibit a larger change in viscosity with concentration. However, the B-coefficients may not be indicative, especially for large hydrophobic solutes. The positive values of B at all temperature indicating structure making ability water (Iqbal and Matecullah, 1990). The magnitude of B values is in the order of L-glutamine>L-arginine> L-lysine, which supports the behaviour of S_V and φ_K^0 data.

CONCLUSION

From density, viscosity and ultrasonic velocity studies of amino acids in aqueous solutions of magnesium nitrate at different temperatures, it is concluded that,

i. The positive values of apparent molar compressibility (φ_K) indicate a strong solute-solvent interaction.
ii. The negative values of limiting apparent molar compressibility (φ_K^0) for amino acid in aqueous magnesium nitrate solution at all temperature studied also indicate the strong ion-solvent interactions.
iii. The values of limiting apparent volume (φ_V^0) at all temperature and suggest the strong ion-solvent interactions in amino acids in aqueous magnesium nitrate solutions at all temperature studied also indicate the strong ion-solvent interactions.
iv. The positive and less negative S_V values are generally associated with the solutes showing an overall hydrophilic character.
v. The value of Jones-Dole coefficient B is positive. It is indicative of water structuring.

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