

A study of transport properties of L-arginine in aqueous solutions of D-maltose monohydrate at 298.15 K: A viscometric approach

Ashwani Kumar and Rajinder K. Bamezai

Abstract: Viscosities of L-arginine in water and in aqueous–D-maltose monohydrate (MM) (2%, 4% and 6% of MM, w/w in water) mixed solvents have been determined as a function of molal concentration of L-arginine at 298.15 K. The viscosity values have been analysed on the basis of the Jones–Dole equation; and viscosity B-coefficients were calculated. The Gibbs free energies of activation of viscous flow per mole of solvent, $\Delta\mu_1^\circ$, per mole of solute, $\Delta\mu_2^\circ$, were also calculated using viscosity data. The calculated Jones-Dole B-coefficients and activation free energy, $\Delta\mu_2^\circ$, results have been interpreted in terms of ionic–hydrophilic, hydrophilic–hydrophilic and hydrophilic–hydrophobic interactions. It has been observed that there exist strong solute–solvent interactions in these systems, which increases with increase in MM concentration in solution.

Key words: Viscosity; L-arginine; D-maltose monohydrate; Viscosity B-coefficients; Activation free energy

1. Introduction

The polyhydroxy compounds play a very important role in stabilizing the native conformations of proteins/enzymes [1-3]. Saccharides are widely distributed in various forms of life as essential moieties of glycoproteins, glycolipids, nucleic acids and polysaccharides. Because of conformational flexibility, saccharides play significant roles in many biological processes such as signaling, cell-cell recognition and molecular and cellular communication [4-6]. The stabilization of native conformations of proteins has been related to various non-covalent interactions like hydrogen bonding, electrostatic and hydrophobic interactions [7, 8].

The study of these interactions provides important insight into the conformational stability and folding/unfolding of globular proteins [9] and are found implicated in several biochemical and physiological processes of a living cell [10-12]. Lee and Timasheff [13] studied the thermal transitions of α -chymotrypsin, chymotrypsinogen and ribonuclease in sucrose and argued that the sucrose stabilize proteins against thermal damage. The complex conformational and configurational factors determining the structure of proteins in sugar solution makes the study of protein-sugar interactions difficult. Therefore interactions of the model compounds of proteins, i.e., amino acids in aqueous saccharide solution are investigated. Since amino acids are the model compounds of protein molecules, their thermodynamic and transport properties in aqueous solutions provide valuable information on solute-solute and solute-solvent interactions that are useful in studying the stability of proteins. Keeping this in mind, a lot of work has been done on amino acid–sugar interactions involving the viscometric technique. Nain et al. have studied the viscometric studies of L-methionine in aqueous D-glucose solutions [14]; L-threonine in aqueous glucose solutions [15]. Pal et al. have studied the viscosities of diglycine in aqueous xylose, L-arabinose, and D-ribose solutions [16]; of glycine, L-alanine, L-valine and L-leucine in aqueous lactose

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solutions [17]; of glyglyglycine in aqueous sucrose and fructose solutions [18]. Palani et al. have studied the viscosities of L-serine, L-glutamine and L-asparagine in aqueous D-glucose solutions [19]. Ali et al. have measured the viscosities of glycine, DL-alanine, L-serine, and DL-valine in aqueous D-glucose solution [20]. Zhao et al. have studied the viscosities of arginine in aqueous D-glucose and sucrose solutions [21]. Riyazuddeen et al. have reported the viscosities for L-alanine, L-threonine and L-glycylglycine in aqueous D-glucose and D-sucrose solutions [22]. But, to the best of our knowledge, there are very few studies on the transport properties of amino acid–sugar interactions with positively charged side chain in aqueous solutions [21,23,24,25] and no viscometric studies have been done on L-arginine in aqueous–D-maltose monohydrate solutions with varying maltose concentrations at 298.15 K. These considerations led us to undertake the viscometric study of L-arginine (with positively charged R group) in aqueous–D-maltose monohydrate solutions.

The present investigation focuses on studying the interactions of L-arginine in water and aqueous–D-maltose monohydrate (MM) (2%, 4% and 6% MM, w/w in water) solvents as a function of molal concentrations of L-arginine at 298.15 K employing the viscometric technique. Using the viscometric data, the Jones–Dole viscosity B-coefficients and activation free energy values have been computed. The obtained viscosity, viscosity B-coefficients and activation free energy values have been discussed in terms of ionic–hydrophilic groups, hydrophilic–hydrophilic groups and hydrophilic–hydrophobic groups interactions operative in the studied systems.

2. Experimental

L-Arginine and D-Maltose monohydrate, herein abbreviated as MM, was used as such (Table 1) without further purification, except drying in an oven for 24 h. The aqueous-saccharide solutions (2%, 4% and 6% of MM, w/w in water) were prepared using triple distilled water with specific conductance less than $1 \times 10^{-6} \text{ S cm}^{-1}$. The saccharide solution was used as solvent to prepare L-arginine solution of eight different molal concentrations

(ranging from 0.0 m to 0.2 m). An electronic single pan five digit analytical balance (Mettler; Model AE-240) with a precision of $\pm 0.00001 \text{ g}$ was used for weighing. All the solutions were prepared with care and stored in special airtight bottles to avoid the exposure of solution to air and evaporation. The possible error in the mole fraction is calculated to be less than $\pm 1 \times 10^{-4}$. The densities of solutions were measured using a single-capillary pycnometer (made of Borosil glass) having a bulb capacity of $\sim 10 \text{ mL}$. The capillary, with graduated marks had a uniform bore which could be closed by a well-fitted glass cap. The pycnometer was calibrated by measuring the density of triply distilled water at 298.15 K. The uncertainty in density measurements was within $\pm 0.03 \text{ kg m}^{-3}$. The viscosities of the solutions were measured by using Ubbelohde type suspended level viscometer. The test solution in the viscometer was allowed to stand for about 30 min in a thermostatic water bath so that the thermal fluctuations in viscometer were minimized. The time of flow was performed using an electronic watch with the resolution of 0.01 s. The average of at least four readings reproducible within 0.1 s was used as the final efflux time. The viscosities of water at different temperatures were taken from the literature [24]. The uncertainty in viscosity measurements was within $\pm 1 \times 10^{-6} \text{ N s m}^{-2}$. The accuracies in measurements of the viscosity values have been ascertained by comparing the measured values for the water with the corresponding literature values at different temperatures. The temperature of the sample solutions was maintained to an accuracy of $\pm 0.02 \text{ K}$ in an electronically controlled thermostatic water bath (Model: TIC-4000N, Thermotech, India).

3. Results and discussion

The experimental values for density, ρ of L-arginine in water and aqueous-MM (2%, 4% and 6% MM, w/w in water) solvents as a function of molal concentrations of L-arginine at 298.15 K are listed in the Table 2. The experimental values for viscosity of L-arginine in water and aqueous-MM (2%, 4% and 6% MM, w/w in water) solvents as a function of molal concentrations of L-arginine at 298.15 K are listed in the Table 3.

Table 1. Provenance and purity of the chemical samples studied.

Chemical name	Provenance	Purification method	Final mass fraction purity
L-Arginine	Sigma Aldrich, India	Used as received	> 0.998
D-Maltose Monohydrate (MM)	Sigma Aldrich, India	Used as received	> 0.998

Table 2. Densities ($\rho/\text{kg m}^{-3}$) of solutions of L-arginine in water, aqueous-MM (2%, 4% and 6%) as function of molality (m/ mol kg^{-1}) of L-arginine at 298.15 K.

m	L-Arginine in water (MM)	L-Arginine in 2% (MM)	L-Arginine in 4% (MM)	L-Arginine in 6% (MM)
0.000	997.07	1004.26	1011.64	1020.59
0.025	998.35	1005.50	1012.86	1021.78
0.050	999.63	1006.74	1014.08	1022.97
0.075	1000.91	1007.98	1015.30	1024.16
0.100	1002.19	1009.22	1016.52	1025.35
0.125	1003.47	1010.46	1017.74	1026.54
0.150	1004.75	1011.70	1018.96	1027.73
0.175	1006.03	1012.94	1020.18	1028.92
0.200	1007.31	1014.18	1021.40	1030.11

Table 3. Viscosities ($\eta \times 10^3/\text{N s m}^{-2}$) of solutions of L-arginine in water, aqueous-MM (2%, 4% and 6%) as functions of molality (m/ mol kg^{-1}) of L-arginine at 298.15 K.

m	L-Arginine in water (MM)	L-Arginine in 2% (MM)	L-Arginine in 4% (MM)	L-Arginine in 6% (MM)
0.000	0.8903	0.9355	0.9838	1.0365
0.025	0.9005	0.9486	1.0008	1.0580
0.050	0.9108	0.9618	1.0179	1.0796
0.075	0.9210	0.9750	1.0350	1.1012
0.100	0.9313	0.9883	1.0521	1.1229
0.125	0.9416	1.0015	1.0693	1.1446
0.150	0.9519	1.0148	1.0865	1.1663
0.175	0.9623	1.0282	1.1037	1.1881
0.200	0.9727	1.0415	1.1209	1.2100

The viscosities of L-arginine in water and aqueous-MM (2%, 4% and 6% MM, w/w in water) solvents at 298.15 K are plotted in Fig. 1. The Jones–Dole [26] equation describes the relative viscosities of solutions as functions of their concentration and viscosity results were analysed by using equation of the form

$$\eta_r = \frac{\eta}{\eta_0} = 1 + Am^{1/2} + Bm \dots\dots\dots 1$$

where η_r is the relative viscosity of the solution; m is the molal concentration of solution; η and η_0 are the viscosities of solution and solvent, respectively; A, the Falkenhagen coefficient, reflects the solute–solute interactions

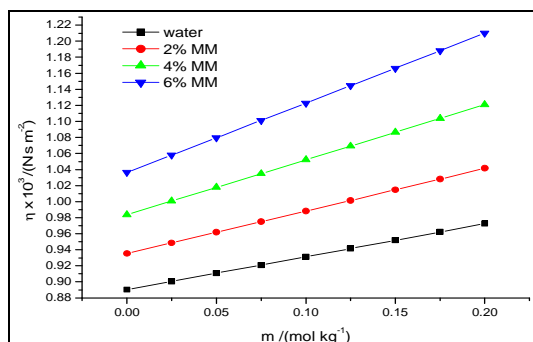


Fig. 1. Variation of viscosity (η) with molality (m) of L-arginine in water, and MM + water solutions at 298.15 K

associated with the size and shape of solute, and B , the Jones-Dole coefficient, is a measure of structural effect induced by solute–solvent interactions [27,28]. The B -coefficients have been given in Table 4.

Table 4. B -coefficients ($\text{dm}^3 \text{mol}^{-1}$) of L-arginine in water and aqueous-MM (2%, 4% and 6% maltose, w/w in water) solutions at 298.15 K.

Solute	Solvent	298.15 K
L-Arginine	Water	0.465
L-Arginine	2% aq. MM solution	0.570
L-Arginine	4% aq. MM solution	0.700
L-Arginine	6% aq. MM solution	0.841

The Falkenhagen A -coefficients which represents solute–solute interactions, are found to be much smaller in magnitude as compared to Jones-Dole B -coefficients and therefore can be considered negligible in case of non-electrolytes [22,29]; therefore, it is often ignored in non-electrolyte systems. Hence $A = 0$ has been taken in this study. The plots of $(\eta_r - 1)/m^{1/2}$ versus $m^{1/2}$ (Fig 2) have been found to be linear at all temperatures for the studied systems in accordance with the Jones–Dole equation.

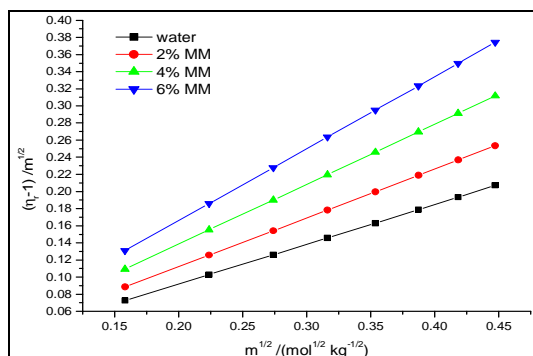


Fig. 2. Variation of $[(\eta_r - 1)/m^{1/2}]$ with molality (m) of L-arginine in water, and MM + water solutions.

The Jones-Dole B -coefficient has significance in the description and understanding of ionic processes, e.g., solvation effects of cations and anions. It is an empirical term which measures the size, shape and charge effects as well as the structural effect induced by solute–solvent interactions.

The viscosity B -coefficient values are positive for L-arginine in water and aqueous-MM (2%, 4% and 6% MM, w/w in water) solvents at all the molal concentrations and at temperature of study. The viscosity B -coefficients of L-arginine in 2.5%, 5% aqueous D-xylose and 2.5%, 5% aqueous L-arabinose solutions at 298.15 K are 0.508, 0.582 and 0.525, 0.599 [24] ($\text{dm}^3 \text{mol}^{-1}$), respectively, while in aqueous medium it has been reported as 0.404 [24] ($\text{dm}^3 \text{mol}^{-1}$). The viscosity B -coefficients of L-arginine in 0.5 M aqueous sucrose [30], 1.0 m aqueous glucose, 0.3 m aqueous glucose, 0.3 m aqueous ascorbic acid [21], 0.3 m aqueous galactose, 0.3 m aqueous maltose, 0.3 m aqueous lactose [31] solutions at the same temperature are reported as 0.826, 0.752, 0.731, 0.649, 0.763, 0.357, 0.348 ($\text{dm}^3 \text{mol}^{-1}$), respectively.

The viscosity B -coefficient gives valuable information regarding the solvation of the solutes and their effects on the structure of solvent in the surrounding of the solute molecules. Large and positive B values indicate a structure to allow the solute to act on the solvents. B coefficient increase when water is replaced by MM; i.e., these saccharides are acting as water structure-maker by hydrogen bonding. The viscosity B -coefficients increase with increasing concentration of MM (Fig. S1 of supplementary material) due to increase in friction that may prevent water flow with increasing concentration of maltose. Thus, the inference drawn from the values of viscosity B -coefficients supports the behaviour that there exist strong solute–solvent interactions as compared to solute–solute interactions in these solutions.

Thermodynamics of viscous flow

According to Eyring's simple model [34], the average activation free energy of a single solute in a pure solvent can be calculated from the following equation.

$$\eta_0 = \left(\frac{hN_A}{V_1^{\circ}} \right) \exp \left(\frac{\Delta\mu_1^{\circ\#}}{RT} \right) \dots\dots\dots 2$$

This can be rearranged as

$$\Delta\mu_1^{\circ\#} = RT \ln \left(\frac{\eta_0 V_1^{\circ}}{hN_A} \right) \dots\dots\dots 3$$

where h , N_A , T , and R are Planck's constant, Avogadro's number, the temperature, and universal gas constant, respectively. V_1° ($= M/\rho_0$) is the apparent (partial) molar volume of the solvent (aqueous-MM); M is the molar mass of solvent; ρ_0 is

the density of the solvent; and $\Delta\mu_1^{\circ\#}$, is chemical potential or Gibbs free energy of activation per mole of the solvent at temperature of 298.15 K. The $\Delta\mu_1^{\circ\#}$, and V_1° values are listed in Table 5. The activation Gibbs free energy, $\Delta\mu_1^{\circ\#}$, for the viscous flow of L-arginine in the solvent is related to viscosity B-coefficients as reported by Feakins et al. [35,36] in the following way.

$$B = \frac{(V_1^{\circ} - V_2^{\circ}) + V_1^{\circ}(\Delta\mu_2^{\circ\#} - \Delta\mu_1^{\circ\#})/RT}{1000} \dots\dots 4$$

Table 5. Apparent (partial) molar volume of solvent (V_1°), free energy of activation for solvent ($\Delta\mu_1^{\circ\#}$) and free energy of activation for solution ($\Delta\mu_2^{\circ\#}$) at 298.15 K.

T/298.15 K	$V_1^{\circ} \times 10^6$ ($\text{m}^3 \text{mol}^{-1}$)	$\Delta\mu_1^{\circ\#}$ (kJmol^{-1})	$\Delta\mu_2^{\circ\#}$ (kJmol^{-1})
L-Arginine in water	18.07	9.16	87.30
L-Arginine 2% aq. MM solution	18.29	9.32	100.99
L-Arginine 4% aq. MM solution	18.51	9.47	117.38
L-Arginine 6% aq. MM solution	18.72	9.63	135.11

Equation (4) rearranges to give the Gibbs free energy of activation per mole of the solute, $\Delta\mu_2^{\circ\#}$,

$$\Delta\mu_2^{\circ\#} = \Delta\mu_1^{\circ\#} + \left(\frac{RT}{V_1^{\circ}} \right) [1000B - (V_1^{\circ} - V_2^{\circ})] \dots\dots 5$$

where $V_2^{\circ} = V_{\phi}^{\circ}$, is the limiting apparent (partial) molar volume of the solute. $\Delta\mu_2^{\circ\#}$, is Gibbs free energy of activation per mole of the solute for viscous flow of solution. The $\Delta\mu_2^{\circ\#}$, values are given in Table 5. According to Feakins, if a solute is completely coordinated in the ground-state solvent, formation of the transition state involves solute-solvent bond breaking and a reduction in the coordination number of the solute takes place. The $\Delta\mu_2^{\circ\#}$ values are large and positive than those of $\Delta\mu_1^{\circ\#}$ in L-arginine in water and aqueous-MM (2%, 4% and 6% MM, w/w in water) solvents indicating stronger solute-solvent interactions and suggesting that formation of transition state is less favoured in the presence of L-arginine. This implies that the formation of the transition state is accompanied by the rupture and distortion of the intermolecular bonds in the solvent structure. This further suggests that the interactions between L-arginine and aqueous-MM (2%, 4% and 6% MM, w/w in water)

solvents molecules in the ground state are stronger than in the transition state. The $\Delta\mu_2^{\circ\#}$, values increases with rise in % of maltose for L-arginine solutions (Fig. S2 of supplementary material), indicating that solute-solvent interactions increases with rise in % of maltose thus making the flow easier [37]. Therefore, the conclusions drawn from $\Delta\mu_2^{\circ\#}$ are in agreement with those drawn from the trends of B values.

4. Conclusions

The viscosities, η , of solutions of L-arginine in aqueous-MM solvents 2%, 4% and 6% of MM, w/w in water, were measured at 298.15 K. From the experimental results, viscosity B-coefficients, the Gibbs free energies of activation of viscous flow per mole of solvent, $\Delta\mu_1^{\circ\#}$, per mole of solute, $\Delta\mu_2^{\circ\#}$ were calculated. The viscosity B-coefficients of L-arginine in water and aqueous-MM (2%, 4% and 6% MM, w/w in water) solutions have been found to be positive for all the systems. The positive and large $\Delta\mu_2^{\circ\#}$ values of L-arginine in water and aqueous-MM (2%, 4% and 6% MM, w/w in water) solutions than those of $\Delta\mu_1^{\circ\#}$ at 298.15 K suggest that solute-solvent interactions are stronger than solvent-solvent interactions in all the studied systems.

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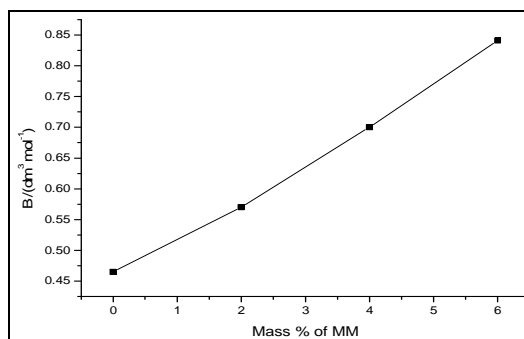


Fig. S1. Variation of Jones-Dole coefficient (B) with mass % of MM for L-arginine in water and aqueous - MM solution at 298.15 K.

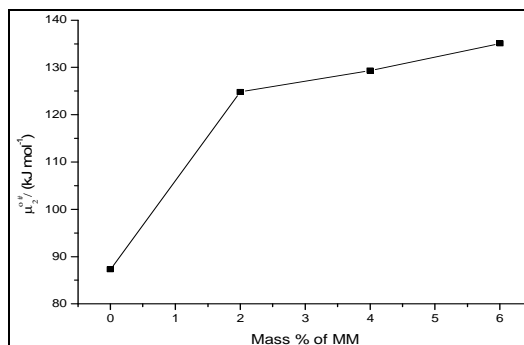


Fig. S2. Variation of Gibbs free energy of activation per mole of the solute ($\Delta\mu_2^\ddagger$) with mass % of MM for L-arginine in water and aqueous - MM solution at 298.15 K.