

Viscosity coefficient and activation parameters for viscous flow of a homologous amino acids in aqueous xylose solutions

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ABSTRACT

Values of viscosity(η) have been determined for four amino acids (glycine, L-alanine, L-valine and L-leucine) in aqueous xylose solutions (0.05, 0.10, 0.15, 0.20) M at $T = (298.15, 303.15, 308.15, 313.15)$ K. Viscosity B-coefficients of Jones-Dole equation, B-coefficients transfer (ΔB), temperature dependent of B-coefficient, free energy of activation per mole of solvent ($\Delta\mu_1^{0*}$) and solute ($\Delta\mu_2^{0*}$) and thermodynamic activation parameter, $G_2^\circ(1 \rightarrow 1')$, are estimated using viscosity data. The linear correlations of B and $\Delta\mu_2^{0*}$ for a homologous series of amino acids have been used to calculate the contribution of charged end groups (NH_3^+ , COO^-), CH_2 and other alkyl chain of the amino acids. Our study concludes the existence of strong solute solvent interaction in the studied systems. Furthermore, structure making and breaking behaviour of all the four amino acids have also been assessed in aqueous xylose solutions. The thermodynamics of viscous flow has also been discussed

Keywords - Amino acids, Xylose, B-coefficients, Free energy of activation per mole of solvent ($\Delta\mu_1^{0*}$) and solute ($\Delta\mu_2^{0*}$).

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I. INTRODUCTION

Sugar solutions play an important role in the biological and food industries. It is widely recognized that sugars as cosolutes help in stabilizing biological macromolecules. Experimental findings indicate that this action is performed either as a result of direct interactions between them and/or through alteration of the water structure [1-4]. Since proteins are large complex molecules, the direct estimation of protein-sugar interactions is difficult. Amino acids, being the essential component of proteins, are models well suited for the study of protein. Keeping this in mind, a number of researchers have studied the amino acid/peptide sugar interactions by employing the viscometric technique. Some authors have reported the thermodynamic study of amino acid in aqueous carbohydrate / saccharide solutions viz. Back et al [5] have reported the increasing thermal stability of proteins in the presence of sugars and polyols, A. Pal et al [6] have studied the Partial molar compressibilities of transfer of some amino acids from water to aqueous lactose solutions at different temperatures, C. Zhao et al [7] have estimated partial molar volumes and viscosity B-coefficients of arginine in aqueous glucose, sucrose and L-ascorbic acid solutions at $T=298.15$ K, Ali et al [8] have investigated the volumetric, viscometric, and refractive index behaviour of α -amino acids and their groups' contribution in aqueous D-glucose solution at different temperatures, Riyazuddeen et al [9] have measured the densities, speeds of sound, and viscosities of (L-Proline + Aqueous Glucose) and (L-Proline + Aqueous Sucrose) Solutions in the Temperature Range (298.15 to 323.15) K. Rajiv Bhat et al [10] have recorded the transfer of some amino acids & peptides from water to aqueous glucose & sucrose solutions at 298.15 K., Nain et al [11] have deduced the solute-solute and solute-solvent interactions in (l-arginine + d-xylose/l-arabinose + water) solutions at different temperatures by using volumetric and viscometric methods and [12] volumetric, ultrasonic and viscometric studies of solute-solute and solute-solvent interactions of l-threonine in aqueous-sucrose solutions at different temperatures viz., 298.15 K, 303.15 K, 308.15 K, 313.15 K. The present paper is a continuation of our research project on the interactions study on ternary systems comprising amino acids + drugs / electrolytes / aqueous sugar systems [13-21]. In this paper, we report the viscosity, η , of homologous amino acids in aqueous xylose solutions at different temperatures. Using the viscosity, η , data, the Jones-Dole viscosity B-coefficient, Temperature dependence of B-coefficient, B-coefficients transfer (ΔB), free energy of activation per mole of solvent ($\Delta\mu_1^{0*}$) and solute ($\Delta\mu_2^{0*}$) are estimated. The linear correlations of B and $\Delta\mu_2^{0*}$ for a homologous series of amino acids have been used to calculate the contribution of charged end groups (NH_3^+ , COO^-), CH_2 and other alkyl chain of the amino acids. Our study concludes the existence of strong solute solvent interaction in the studied systems. Furthermore, structure making and breaking behaviour all the four amino acids have also been assessed in aqueous xylose solutions. The thermodynamics of viscous flow has also been discussed.

II. EXPERIMENTAL

Xylose (mass fraction purity > 0.990) is procured from S.D. Fine. Chem. Ltd. Mumbai, Glycine (99.7% assay), is procured from Merck Ltd. Mumbai, L-valine (99% assay) and L-leucine (+99 % assay) are obtained from ALDRICH (U.S.A). L-alanine (99% assay) is procured from SISCO, Mumbai. The amino acids and xylose are used after drying in a desiccator over P₂O₅ for 72 hrs before use. Aqueous xylose solutions of molality 0.05M, 0.10M, 0.15M and 0.20M are prepared using doubly deionized distilled water with a conductivity of 1.5×10⁻⁴ Ω⁻¹·m⁻¹ and are used as solvents to prepare the molal concentrations of amino acids. For glycine, L-alanine, L-valine, molalities of (0.02, 0.04, 0.06, 0.08 and 0.1) M and for L-leucine, molalities of (0.02, 0.04, 0.05, 0.06 and 0.08) M are used respectively. The mass measurements are made using a high precision and electronic balance (Model HR 300, Japan) with a precision of ± 0.1 mg. Viscosity is measured by means of a suspended level Ubbelohde viscometer and flow times have been measured using a Racer digital stopwatch having an accuracy of ±0.01s. An average of three sets of flow time reading has been taken for each solution for calculation of viscosity[22]. The overall experimental reproducibility is estimated to be within ± 2×10⁻³ m Pa·s. The temperatures of the solutions are maintained to an uncertainty of ± 0.01 K in an electronically controlled thermostatic water bath (Eurotherm, Mittal enterprises, New Delhi).

III. RESULTS

The experimental values of viscosity (see table 1) is used to evaluate the relative viscosities of homologous amino acids in various concentrations of aqueous xylose solutions at different temperatures using the following equation (1).

$$\eta_r = \eta / \eta_0 \quad (1)$$

Table 1 Viscosity (η) of α – amino acids in aqueous xylose solutions at different temperatures

<i>η / m Pa s at various m_s / mol kg⁻¹</i>					
<i>m</i> (mol kg ⁻¹)	0.00 (Water)	0.05	0.10	0.15	0.20
T = 298.15 K					
Glycine					
0	0.8905	0.9054	0.9212	0.9352	0.9495
0.02	0.8943	0.9081	0.9243	0.9388	0.9535
0.04	0.8968	0.9108	0.9271	0.9419	0.9571
0.06	0.8998	0.9133	0.9301	0.9452	0.9608
0.08	0.9026	0.9160	0.9330	0.9486	0.9644
0.10	0.8905	0.9054	0.9212	0.9352	0.9495
Alanine					
0	0.8905	0.9054	0.9212	0.9352	0.9495
0.02	0.8958	0.9101	0.9262	0.9406	0.9554
0.04	0.9003	0.9147	0.9310	0.9460	0.9610
0.06	0.9051	0.9192	0.9359	0.9513	0.9668
0.08	0.9096	0.9236	0.9407	0.9568	0.9725
0.10	0.9139	0.9278	0.9454	0.9619	0.9783
Valine					
0	0.8905	0.9054	0.9212	0.9352	0.9495
0.02	0.8985	0.9139	0.9302	0.9447	0.9595
0.04	0.9060	0.9220	0.9385	0.9532	0.9683
0.06	0.9137	0.9299	0.9469	0.9620	0.9776
0.08	0.9215	0.9380	0.9551	0.9707	0.9868
0.10	0.9288	0.9456	0.9634	0.9793	0.9956
Leucine					
0	0.8905	0.9054	0.9212	0.9352	0.9495
0.02	0.9000	0.9146	0.9310	0.9455	0.9603
0.04	0.9087	0.9240	0.9406	0.9554	0.9705
0.05	0.9128	0.9285	0.9454	0.9605	0.9759
0.06	0.9174	0.9330	0.9500	0.9653	0.9811
0.08	0.9260	0.9417	0.9593	0.9750	0.9910
T = 303.15 K					
Glycine					
0	0.7969	0.8095	0.8235	0.8363	0.8480
0.02	0.8000	0.8117	0.8261	0.8392	0.8513

0.04	0.8024	0.8139	0.8284	0.8418	0.8542
0.06	0.8048	0.8161	0.8309	0.8446	0.8573
0.08	0.8072	0.8182	0.8333	0.8474	0.8604
0.10	0.8096	0.8203	0.8358	0.8502	0.8634
Alanine					
0	0.7969	0.8095	0.8235	0.8363	0.8480
0.02	0.8016	0.8135	0.8277	0.8409	0.8529
0.04	0.8057	0.8173	0.8319	0.8455	0.8579
0.06	0.8095	0.8211	0.8359	0.8500	0.8627
0.08	0.8137	0.8248	0.8401	0.8547	0.8676
0.10	0.8175	0.8286	0.8441	0.859	0.8724
Valine					
0	0.7969	0.8095	0.8235	0.8363	0.8480
0.02	0.8039	0.8173	0.8317	0.8449	0.8571
0.04	0.8107	0.8245	0.8391	0.8526	0.8651
0.06	0.8174	0.8318	0.8467	0.8606	0.8732
0.08	0.8242	0.8391	0.8543	0.8685	0.8815
0.10	0.8307	0.8460	0.8617	0.8762	0.8897
Leucine					
0	0.7969	0.8095	0.8235	0.8363	0.8480
0.02	0.8051	0.8181	0.8325	0.8457	0.8579
0.04	0.8130	0.8264	0.8411	0.8545	0.8671
0.05	0.8168	0.8306	0.8455	0.8594	0.8720
0.06	0.8207	0.8346	0.8497	0.8637	0.8767
0.08	0.8280	0.8426	0.8581	0.8724	0.8857
T = 308.15 K					
Glycine					
0	0.7190	0.7300	0.7426	0.7539	0.7640
0.02	0.7210	0.7318	0.7447	0.7564	0.7668
0.04	0.7234	0.7337	0.7467	0.7588	0.7694
0.06	0.7254	0.7354	0.7487	0.7611	0.7720
0.08	0.7275	0.7372	0.7508	0.7634	0.7745
0.10	0.7294	0.7390	0.7528	0.7657	0.7771
Alanine					
0	0.7190	0.7300	0.7426	0.7539	0.7640
0.02	0.7231	0.7333	0.7462	0.7578	0.7682
0.04	0.7268	0.7367	0.7498	0.7618	0.7726
0.06	0.7301	0.7398	0.7533	0.7657	0.7766
0.08	0.7337	0.7431	0.7568	0.7695	0.7809
0.10	0.7372	0.7464	0.7604	0.7735	0.7850
Valine					
0	0.7190	0.7300	0.7426	0.7539	0.7640
0.02	0.7250	0.7372	0.7501	0.7618	0.7722
0.04	0.7314	0.7437	0.7568	0.7687	0.7795
0.06	0.7372	0.7504	0.7638	0.7759	0.7868
0.08	0.7431	0.7570	0.7707	0.7833	0.7945
0.10	0.7490	0.7634	0.7775	0.7903	0.8020
Leucine					
0	0.7190	0.7300	0.7426	0.7539	0.7640
0.02	0.7260	0.7380	0.7509	0.7626	0.7732
0.04	0.7334	0.7455	0.7588	0.7707	0.7814
0.05	0.7368	0.7493	0.7628	0.7749	0.7861
0.06	0.7401	0.7530	0.7666	0.7790	0.7901
0.08	0.7464	0.7604	0.7743	0.7870	0.7986
T = 313.15 K					
Glycine					
0	0.6523	0.6631	0.6729	0.6833	0.6932

0.02	0.6542	0.6646	0.6747	0.6853	0.6955
0.04	0.6561	0.6661	0.6764	0.6873	0.6977
0.06	0.6579	0.6675	0.6781	0.6892	0.6999
0.08	0.6598	0.6691	0.6797	0.6911	0.7022
0.10	0.6616	0.6704	0.6815	0.6931	0.7042
Alanine					
0	0.6523	0.6631	0.6729	0.6833	0.6932
0.02	0.6554	0.6659	0.6759	0.6867	0.6969
0.04	0.6589	0.6687	0.6791	0.6901	0.7006
0.06	0.6620	0.6715	0.6820	0.6934	0.7043
0.08	0.6650	0.6743	0.6850	0.6969	0.7081
0.10	0.6680	0.6770	0.6880	0.7001	0.7115
Valine					
0	0.6523	0.6631	0.6729	0.6833	0.6932
0.02	0.6576	0.6697	0.6798	0.6905	0.7007
0.04	0.6633	0.6757	0.6859	0.6970	0.7075
0.06	0.6687	0.6818	0.6923	0.7037	0.7144
0.08	0.6740	0.6877	0.6986	0.7104	0.7213
0.10	0.6790	0.6938	0.7050	0.7169	0.7279
Leucine					
0	0.6523	0.6631	0.6729	0.6833	0.6932
0.02	0.6584	0.6705	0.6806	0.6912	0.7016
0.04	0.6649	0.6774	0.6878	0.6987	0.7091
0.05	0.6677	0.6809	0.6916	0.7027	0.7131
0.06	0.6706	0.6845	0.6950	0.7063	0.7168
0.08	0.6767	0.6910	0.7021	0.7136	0.7248

Where η and η_0 are the viscosities of the solution(AAs +xylose + Water) and solvent (xylose + Water). The B-coefficients values are evaluated by fitting the η_r values to the Jones-Dole equation (2) by a least squares method [23] as follows.

$$\eta_r = \eta / \eta_0 = 1 + B \cdot c \tag{2}$$

Where ‘c’ is the molarity (calculated from molality data). The values of B-coefficients along with the standard deviations of linear regression, σ are summarized in table 2. It is seen from the table 2 that, the values of B-coefficients for amino acids in water agree very well with available literature values [20, 24-30], thus validating our experimental procedures..

The B-coefficients data in aqueous xylose solutions have been used to calculate the corresponding (ΔB) transfer function as follows:

$$\Delta B = \Delta B \text{ (in W + X)} - \Delta B \text{ (in W)} \tag{3}$$

Where W stands for water and X stands for xylose. The ΔB values as a function of molality of the solute amino acid at all the studied temperatures are given in table 3

The sign of temperature dependence of B- coefficient viz. (dB/dT) gives the information of structure making/breaking property of the solute in the solvent media [31] and the evaluated dB/dT for all four amino acids in aqueous xylose solutions are included in table 4.

Thermodynamic transfer functions of amino acids may be expressed by the McMillan-Mayer Theory [32] of solutions that permits the formal separation of the effects due to interactions involving two or more solute molecules. This approach has been further discussed by Friedman and Krishnan [33] and Frank et al. [34] in order to include solute - cosolute interactions in the solvation spheres. According to this treatment, a thermodynamic transfer function at infinite dilution can be expressed as

$$\Delta B_\phi^0 = 2 \eta_{AB} m_B + 3 \eta_{ABB} m_B^2 + \dots \tag{4}$$

Where, in η_{AB} and η_{ABB} , A stands for aminoacid and B stands for xylose and m_B is the molality of xylose (cosolute). The constants η_{AB} and η_{ABB} are pair and triplet interaction parameters, obtained by fitting ΔB_ϕ^0 data to equation (11). The parameters η_{AB} and η_{ABB} for viscosity are listed in Table 5.

The viscosity data are used to estimate the free energy of activation per mole of the solvent ($\Delta\mu_1^{0*}$) and solute ($\Delta\mu_2^{0*}$) as suggested by Feakins et al. [35] and Eyring et al. [36] using the following equations (5-7).

$$B = (\bar{V}_1^0 - \bar{V}_2^0) / 1000 + \bar{V}_1^0 / 1000 RT (\Delta\mu_2^{0*} - \Delta\mu_1^{0*}) \tag{5}$$

$$\Delta \mu_1^{0*} = RT \ln (\eta_0 \bar{V}_1^0 / hN) \quad (6)$$

Equation (6) can be rearranged as

$$\Delta \mu_2^{0*} = \Delta \mu_1^{0*} + RT / \bar{V}_1^0 [1000 B - (\bar{V}_1^0 - \bar{V}_2^0)] \quad (7)$$

Where $\bar{V}_1^0 = (\sum x_i m_i / \rho)$ is the mean volume of the solvent and $\bar{V}_2^0 = V_\phi^0$ is the partial molar volume at infinite dilution of the solute. The terms x_i and m_i denote the mole fractions and molecular weights of water (1) and xylose (2) and ρ is the density of the solvent mixture (xylose + water), 'h' is the Planck's constant, 'N' is the Avogadro's number, η_0 is the viscosity of the solvent and 'R' is the gas constant. The values of $\Delta \mu_1^{0*}$, \bar{V}_1^0 and $\Delta \mu_2^{0*}$ are reported in tables 6 and 7.

According to transition state theory [35], every solvent molecule in one mole of solution must pass through the transition state and also interact more or less strongly with solute molecules. Thus, the activation free energy $\Delta \mu_2^{0*}$ include the free energy transfer of solute from ground state to transition state solvents [ΔG_2° (1→1')] and the free energy of solute through its own viscous transition state [ΔG_2° (2→2')]. The [ΔG_2° (1→1')] values, calculated by using methods similar to those reported elsewhere [37] are also given in Table 8.

The linear regression analysis of D_ϕ^0 values as a function of number of carbon atoms (n_c) in the alkyl chain of the amino acids can be represented as[38]

$$D_\phi^0 = D_\phi^0(\text{NH}_3^+, \text{COO}^-) + n_c D_\phi^0(\text{CH}_2) \quad (8)$$

Where $D_\phi^0(\text{NH}_3^+, \text{COO}^-)$ and $D_\phi^0(\text{CH}_2)$ are the zwitterionic end groups and the methylene group contribution to D_ϕ^0 , respectively. Here D_ϕ^0 stands for B and $\Delta \mu_2^{0*}$ respectively. As suggested by Hakin et al.[39,40] the contributions of the other alkyl chain of the α -amino acids are calculated as follows:

$$D_\phi^0(\text{CH}_3) = 1.5 D_\phi^0(\text{CH}_2) \quad (9)$$

$$D_\phi^0(\text{CH}) = 0.5 D_\phi^0(\text{CH}_2) \quad (10)$$

The alkyl chains of homologous series of the α -amino acids studied in this work are CH_2 -(gly), CH_3CH -(Ala), $\text{CH}_3\text{CH}_2\text{CH}$ -(Val) and $\text{CH}_3\text{CH}_2\text{CH}_2\text{CH}$ -(Leu), the values of $D_\phi^0(\text{CH}_2)$ obtained by this procedure characterize the mean contribution of CH- and CH_3 - groups to D_ϕ^0 of the α -amino acids[41]. The results are given in tables 9 and 10.

IV. DISCUSSION

The experimental measurements of viscosity data of homologous amino acids in aqueous xylose solution at different temperatures yield information on viscosity B-Coefficients of Jones-Dole equation that will reflect size and shape effects as well as structural effects induced by solute solvent interaction[42,43]. Further, the B- Coefficients provide information about the solvation of the solutes and effects on the structure of the solvent in the near environment of solute molecules.

Table 2 Viscosity B – Coefficient of α – amino acids in aqueous xylose solutions at different temperatures

$B * 10^3 / \text{m}^3 \text{mol}^{-1}$ at various $m_s / \text{mol kg}^{-1}$						
Amino Acid	0.00 (Water)		0.05	0.10	0.15	0.20
	Present Work	Literature				
T = 298.15 K						
Glycine	0.159 (0.024)	0.153 ^a	0.145 (0.009)	0.158 (0.009)	0.175 (0.017)	0.192 (0.011)
Alanine	0.259 (0.002)	0.258 ^b	0.246 (0.009)	0.262 (0.012)	0.286 (0.015)	0.301 (0.012)
Valine	0.433 (0.033)	0.447 ^c , 0.423 ^d	0.443 (0.010)	0.454 (0.007)	0.466 (0.007)	0.479 (0.006)
Leucine	0.493 (0.007)	0.487 ^b	0.504 (0.017)	0.516 (0.012)	0.529 (0.009)	0.542 (0.018)
T = 303.15 K						
Glycine	0.153 (0.025)		0.133 (0.023)	0.148 (0.009)	0.165 (0.016)	0.179 (0.012)
Alanine	0.253 (0.021)		0.234 (0.006)	0.250 (0.011)	0.272 (0.018)	0.287 (0.010)
Valine	0.427 (0.021)		0.450 (0.016)	0.461 (0.008)	0.472 (0.012)	0.483 (0.020)
Leucine	0.487 (0.043)		0.511 (0.007)	0.523 (0.018)	0.537 (0.016)	0.551 (0.025)
T = 308.15 K						
Glycine	0.148 (0.024)	0.148 ^e	0.123 (0.023)	0.137 (0.007)	0.154 (0.016)	0.168 (0.028)
Alanine	0.248 (0.015)	0.247 ^e	0.225 (0.022)	0.240 (0.015)	0.260 (0.018)	0.275 (0.025)
Valine	0.422 (0.037)	0.418 ^a	0.456 (0.017)	0.468 (0.019)	0.479 (0.034)	0.491 (0.036)
Leucine	0.480 (0.052)	0.423 ^c	0.518 (0.014)	0.531 (0.028)	0.545 (0.017)	0.559 (0.019)
T = 313.15 K						
Glycine	0.144 (0.033)	0.144 ^f	0.111 (0.049)	0.126 (0.024)	0.142 (0.024)	0.158 (0.041)
Alanine	0.244 (0.045)	0.247 ^g	0.211 (0.029)	0.225 (0.022)	0.246 (0.052)	0.266 (0.039)
Valine	0.418 (0.107)	0.413 ^h	0.461 (0.010)	0.475 (0.023)	0.486 (0.021)	0.496 (0.057)
Leucine	0.473 (0.065)	0.480 ^h	0.525 (0.049)	0.540 (0.036)	0.553 (0.045)	0.562 (0.054)

^aRef 24, ^bRef 25, ^cRef 26, ^dRef 27, ^eRef 28, ^fRef 29, ^gRef 30, ^hRef 20

The observed large and positive B coefficient values (see table 2) for all the four amino acids in aqueous xylose solution increase with increase in concentration of xylose suggest the presence of stronger solute–solvent interactions as compared to solute–solute interactions. However the decrease in B coefficient values with increase of temperature indicate the rupturing effect of thermal energy on the solute-solvent interactions of the studied systems. The viscosity B coefficient values for amino acids in aqueous xylose solutions at particular concentration and temperatures show the following order; glycine < alanine < valine < leucine.

Table 3 shows that the transfer B coefficient, ΔB (W → Xyl) in going from water to xylose are calculated by taking the difference between viscosity B coefficient in aqueous xylose and in water for each amino acid studied. The increase in B coefficients (i.e., positive ΔB values, with the exemption of glycine and alanine at low concentration.) from water to xylose is due to interactions of xylose with either R groups or charged centers of amino acids. In other words the main contribution to ΔB (W → Xyl) values comes from the interactions between charged centers of amino acids and xylose molecules, rather than from interactions between R groups of amino acids and xylose molecules cause an increase in viscosity and thus in B coefficient. Similar results are available in literature [44].

Table 3 Viscosity B - Coefficient transfer (ΔB) α - amino acids in aqueous xylose solutions at different temperatures

Amino Acids	various m_s / mol kg ⁻¹			
	0.05	0.10	0.15	0.20
	$\Delta B \cdot 10^3$ m ³ mol ⁻¹	$\Delta B \cdot 10^3$ m ³ mol ⁻¹	$\Delta B \cdot 10^3$ m ³ mol ⁻¹	$\Delta B \cdot 10^3$ m ³ mol ⁻¹
T = 298.15 K				
Glycine	-0.014	-0.001	0.016	0.033
Alanine	-0.013	0.003	0.027	0.042
Valine	0.010	0.021	0.033	0.046
Leucine	0.011	0.023	0.036	0.049
T = 303.15 K				
Glycine	-0.020	-0.005	0.012	0.026
Alanine	-0.019	-0.003	0.019	0.034
Valine	0.023	0.034	0.045	0.056
Leucine	0.024	0.036	0.05	0.064
T = 308.15 K				
Glycine	-0.025	-0.011	0.006	0.020
Alanine	-0.023	-0.008	0.012	0.027
Valine	0.034	0.046	0.057	0.069
Leucine	0.038	0.051	0.065	0.079
T = 313.15 K				
Glycine	-0.033	-0.018	-0.002	0.014
Alanine	-0.032	-0.019	0.002	0.022
Valine	0.043	0.057	0.068	0.078
Leucine	0.052	0.067	0.080	0.089

The variation of B with temperature, dB/dT provides (see table 4) direct evidence regarding structure-making or breaking ability of the solute in the solution. When dB/dT is negative, the solute acts as structure-maker and when dB/dT is positive it acts as structure breaker. The negative values of (dB/dT) for glycine and alanine (see table 4) indicate [45] that, these two amino acids act as structure-maker while positive (dB/dT) values for valine and leucine supports the structure breaking ability of these amino acids in aqueous-xylose solutions.

Table 4 Temperature Coefficient (dB/dT) of α – amino acids in aqueous xylose solutions at different temperatures.

Amino Acids	dB/dT / m ³ mol ⁻¹ K ⁻¹ at various m_s / mol kg ⁻¹			
	0.05	0.10	0.15	0.20
Glycine	-0.0022	-0.0021	-0.0022	-0.0023
Alanine	-0.0023	-0.0024	-0.0026	-0.0023
Valine	0.0012	0.0014	0.0013	0.0012
Leucine	0.0014	0.0016	0.0016	0.0014

Table 5 Values of pair (η_{AB}) and triplet (η_{ABB}) of α – amino acids in aqueous xylose at different temperatures

Glycine		Alanine		Valine		Leucine	
$10^3 \eta_{AB}/$ $m^3 \cdot mol^{-2} \cdot kg$	$10^3 \eta_{ABB}/$ $m^3 \cdot mol^{-3} \cdot kg^2$	$10^3 \eta_{AB}/$ $m^3 \cdot mol^{-2} \cdot kg$	$10^3 \eta_{ABB}/$ $m^3 \cdot mol^{-3} \cdot kg^2$	$10^3 \eta_{AB}/$ $m^3 \cdot mol^{-2} \cdot kg$	$10^3 \eta_{ABB}/$ $m^3 \cdot mol^{-3} \cdot kg^2$	$10^3 \eta_{AB}/$ $m^3 \cdot mol^{-2} \cdot kg$	$10^3 \eta_{ABB}/$ $m^3 \cdot mol^{-3} \cdot kg^2$
T = 298.15 K							
-0.184	0.968	-0.175	1.040	0.0950	0.0667	0.106	0.057
T = 303.15 K							
-0.245	1.147	-0.240	1.204	0.2450	-0.3867	0.250	-0.338
T = 308.15 K							
-0.303	1.300	-0.284	1.319	0.3688	-0.9011	0.409	-0.781
T = 313.15 K							
-0.393	1.571	-0.405	1.680	0.4750	-1.2667	0.576	-1.281

From table 5, it is seen that the values of pair interaction coefficient are negative and that of the triplet interaction coefficient are positive indicating the predominance of triplet interactions over the pair interactions in all the studied systems at different temperatures. similar results are available in literature.

It is evident from table 6 and 7 that $\Delta\mu_2^{0*}$ values are positive and much larger than $\Delta\mu_1^{0*}$ suggesting that the interactions between solute (amino acids) and solvent (water +xylose) molecules in the ground state are stronger than in the transition state, i.e., the interactions between solute (Amino acids) and solvent (xylose+ water) molecules in the ground state are stronger than in the transition state. This means that the formation of transition state is accompanied by the rupture and distortion of the intermolecular forces in solvent structure. Thus, the solvation of the solute in the transition state is unfavourable in free energy terms. Further, $\Delta\mu_2^{0*}$ increases from glycine to leucine at a particular temperature, indicating that the solvation of amino acid molecules becomes increasingly unfavourable as the hydrophobicity or the number of carbon atoms of the side chain increases from glycine to leucine. This feature is similar to that observed for glycine in aqueous solutions of transition metal chlorides [46]. The $\Delta\mu_2^{0*}$ values at particular concentration and temperature follow the order, Glycine < Alanine < Valine < Leucine. This reflects that the solute having a longer alkyl chain requires more energy in transferring from the ground state to the transition state. Similar results were also reported for glycine, alanine, and valine in aqueous urea [47] and also for glycine, alanine, serine, and valine in aqueous glucose solutions [8].

Table 6 Free energy of activation of solvent ($\Delta\mu_1^{0*}$) and mean volume of solvent (\bar{v}_1^0) of α – amino acids aqueous xylose solution at different temperatures

m_s $mol\ kg^{-1}$	$\Delta\mu_1^{0*}\ kJ\ mol^{-1}$	$\bar{v}_1^0\ m^3\ mol^{-1}$	$\Delta\mu_1^{0*}\ kJ\ mol^{-1}$	$\bar{v}_1^0\ m^3\ mol^{-1}$
	T = 298.15 K		T = 303.15 K	
0	9.16	18.07	9.04	18.09
0.05	9.21	18.14	9.09	18.16
0.1	9.27	18.21	9.14	18.23
0.15	9.31	18.28	9.19	18.30
0.2	9.36	18.35	9.24	18.38
$mol\ kg^{-1}$	T = 308.15 K		T = 313.15 K	
0	8.93	18.12	8.83	18.16
0.05	8.98	18.19	8.88	18.23
0.1	9.03	18.26	8.93	18.30
0.15	9.08	18.34	8.98	18.37
0.2	9.13	18.41	9.03	18.44

Table 7 Free energy of activation of solute ($\Delta\mu_2^{0*}$) of α – Amino acids in aqueous xylose solution at different temperatures

Amino acids	$\Delta\mu_2^{0*} / kJ\ mol^{-1}$ at various $m_s / mol\ kg^{-1}$				
	0.00 (Water)	0.05	0.10	0.15	0.20
T = 298.15 K					
Glycine	34.40	31.74	33.55	35.87	38.15
Alanine	50.49	48.07	50.19	53.39	55.39
Valine	78.52	79.28	80.61	82.03	83.67

Leucine	89.16	90.01	91.45	92.96	94.46
T = 303.15 K					
Glycine	33.90	30.45	32.56	34.93	36.86
Analine	50.21	46.97	49.15	52.14	54.16
Valine	78.64	81.29	82.63	83.93	85.30
Leucine	89.44	92.22	93.66	95.33	96.96
T = 308.15 K					
Glycine	33.51	29.40	31.42	33.81	35.77
Analine	50.13	46.31	48.38	51.14	53.22
Valine	78.94	83.23	84.70	86.03	87.53
Leucine	89.59	94.44	96.02	97.72	99.35
T = 313.15 K					
Glycine	33.26	28.08	30.27	32.57	34.86
Analine	50.19	45.13	46.97	49.90	52.71
Valine	79.31	84.97	86.74	88.09	89.31
Leucine	89.70	96.67	98.53	100.12	101.05

Table 8 Values of, $\Delta G_2^\circ(1 \rightarrow 1')$ from ground state to transition state of α – amino acids in aqueous xylose solution at different temperatures

Amino acids	$\Delta G_2^\circ(1 \rightarrow 1') / \text{kJ mol}^{-1}$ at various $m_s / \text{mol kg}^{-1}$				
	0.00 (Water)	0.05	0.10	0.15	0.20
T = 298.15 K					
Glycine	25.24	22.53	24.28	26.55	28.79
Analine	41.33	38.85	40.93	44.08	46.03
Valine	69.36	70.06	71.34	72.72	74.31
Leucine	80.00	80.80	82.18	83.64	85.09
T = 303.15 K					
Glycine	24.86	21.36	23.42	25.73	27.62
Analine	41.17	37.88	40.01	42.95	44.93
Valine	69.60	72.20	73.49	74.74	76.06
Leucine	80.40	83.13	84.52	86.14	87.73
T = 308.15 K					
Glycine	24.58	20.42	22.38	24.73	26.65
Analine	41.20	37.33	39.35	42.06	44.09
Valine	70.01	74.25	75.67	76.95	78.40
Leucine	80.66	85.46	86.98	88.64	90.22
T = 313.15 K					
Glycine	24.43	19.20	21.34	23.59	25.83
Analine	41.37	36.25	38.04	40.92	43.68
Valine	70.48	76.09	77.81	79.11	80.28
Leucine	80.88	87.79	89.60	91.14	92.03

It is seen from table 8 that, the thermodynamic activation parameter [$\Delta G_2^\circ(1 \rightarrow 1')$] values increase from glycine to L-leucine at particular concentration and temperature, indicating the requirement of more energy for the amino acids with longer alkyl side chains for the transfer from ground state solvent to transition state solvent. This effectively means that more solute- solvent bonds must be broken to form transition state.

Table 9 Group contributions of B-coefficient of α – amino acids in aqueous xylose solutions at different temperatures.

Group	$B * 10^3 / \text{m}^3 \text{mol}^{-1}$ at various $m_s / \text{mol kg}^{-1}$			
	0.05	0.10	0.15	0.20
T=298.15 K				
NH ₃ ⁺ COO ⁻	0.060	0.075	0.097	0.115
CH ₂	0.091	0.090	0.088	0.087
CHCH ₃	0.182	0.180	0.176	0.174
CHCH(CH ₃) ₂	0.364	0.360	0.352	0.348
CHCH ₂ CH(CH ₃) ₂	0.455	0.450	0.440	0.435
T=303.15 K				
NH ₃ ⁺ COO ⁻	0.040	0.057	0.078	0.093

CH ₂	0.097	0.096	0.094	0.094
CHCH ₃	0.194	0.192	0.188	0.188
CHCH(CH ₃) ₂	0.388	0.384	0.376	0.376
CHCH ₂ CH(CH ₃) ₂	0.485	0.480	0.470	0.470
T=308.15 K				
NH ₃ ⁺ COO ⁻	0.024	0.039	0.059	0.073
CH ₂	0.102	0.101	0.100	0.099
CHCH ₃	0.204	0.202	0.200	0.198
CHCH(CH ₃) ₂	0.408	0.404	0.400	0.396
CHCH ₂ CH(CH ₃) ₂	0.510	0.505	0.500	0.495
T=313.15 K				
NH ₃ ⁺ COO ⁻	0.003	0.018	0.038	0.059
CH ₂	0.107	0.107	0.106	0.103
CHCH ₃	0.214	0.214	0.212	0.206
CHCH(CH ₃) ₂	0.428	0.428	0.424	0.412
CHCH ₂ CH(CH ₃) ₂	0.535	0.535	0.530	0.515

The group contribution values of viscosity *B*-coefficient and $\Delta\mu_2^{0*}$ are shown in tables 9 &10 It is observed from table 9 that viscosity *B*-coefficient increases with increase in number of carbon atoms (*n_c*), in the alkyl chain of the amino acids. Moreover, the magnitudes of *B* (NH₃⁺, COO⁻) systematically increase with the increase in the concentration of xylose at all temperatures. The variation of *B* (CH₂) with concentration of xylose is almost insensitive. Similar results on group contribution to *B*-coefficient are available in literature [32, 48,49]. The magnitude of the values indicates the predominance of the interactions between zwitterionic centers of the amino acids with xylose molecules in the solution. It is interesting to add that the contribution to the *B*-coefficient increase with the increase in chain length.

Table 10 Group contributions free energy of activation of solute ($\Delta\mu_2^{0*}$) of α – Amino acids in aqueous xylose solution at different temperatures

Group	$\Delta\mu_2^{0*}$ / kJ mol ⁻¹ at various <i>m_s</i> / mol kg ⁻¹			
	0.05	0.10	0.15	0.20
T=298.15 K				
NH ₃ ⁺ COO ⁻	17.95	20.08	23.21	25.64
CH ₂	14.77	14.62	14.28	14.09
CHCH ₃	29.54	29.24	28.56	28.18
CHCH(CH ₃) ₂	59.08	58.48	57.12	56.36
CHCH ₂ CH(CH ₃) ₂	73.85	73.10	71.40	70.45
T=303.15 K				
NH ₃ ⁺ COO ⁻	15.37	17.80	20.80	22.91
CH ₂	15.78	15.56	15.26	15.13
CHCH ₃	31.56	31.12	30.52	30.26
CHCH(CH ₃) ₂	63.12	62.24	61.04	60.52
CHCH ₂ CH(CH ₃) ₂	78.90	77.80	76.30	75.65
T=308.15 K				
NH ₃ ⁺ COO ⁻	13.24	15.47	18.36	20.52
CH ₂	16.70	16.55	16.27	16.14
CHCH ₃	33.40	33.10	32.54	32.28
CHCH(CH ₃) ₂	66.80	66.20	65.08	64.56
CHCH ₂ CH(CH ₃) ₂	83.50	82.75	81.35	80.70
T=313.15 K				
NH ₃ ⁺ COO ⁻	10.53	12.74	15.68	18.78
CH ₂	17.71	17.62	17.32	16.90
CHCH ₃	35.42	35.24	34.64	33.80
CHCH(CH ₃) ₂	70.84	70.48	69.28	67.60
CHCH ₂ CH(CH ₃) ₂	88.55	88.10	86.60	84.50

It is seen from table 10, that the contribution of $\Delta\mu_2^{0*}$ (NH₃⁺, COO⁻) group increases with the increasing concentration of the xylose but decreases with increase in temperatures. However the contribution of (CH₂) group decreases with increasing concentration of xylose and increases with increase in temperatures. This

indicates that the interactions between co-solute and charged end groups (NH_3^+ , COO^-) of amino acids are much stronger than those between the co-solute and (CH_2) groups.

V. CONCLUSION

The viscosities, η of some homologous amino acids in aqueous xylose solvents (0.05, 0.10, 0.15, 0.20) M have been experimentally measured and are reported at different temperatures. Using the experimental data, various parameters, viz., A, Jones–Dole coefficient, B, Gibbs energy of activation per mole of solvent, $\Delta\mu_1^{0*}$ and Gibbs energy of activation per mole of the solute, $\Delta\mu_2^{0*}$ are deduced and reported. The positive values of viscosity B-coefficient of the amino acids in aqueous xylose solutions show that the interactions involving the charged centers of amino acid with the OH group of aqueous xylose are dominating and indicating the presence of strong- solute solvent interactions. From the temperature dependence of B- coefficient data, it is found that glycine and alanine act as structure maker while valine and leucine as structure breaker in aqueous xylose solutions.

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