

Study of molecular interactions between L-aspartic acid and aqueous 1,2-propanediol solution at $T = 303.15$ K

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ABSTRACT

Densities, speeds of sound, and viscosities of L-aspartic acid (Asp) having fixed molality of 0.05 mol kg^{-1} in water and different concentrations of 1,2-propanediol (PD) (from 10 to 90%) has been measured at 303.15 K. From experimental values of density and speed of sound data, apparent molar volume, V_{ϕ} , isentropic compressibility, k_s , apparent molar isentropic compression, $K_{\phi,s}$, specific acoustic impedance, Z , intermolecular free length, L_f , and relative association, R_A , have been calculated. The viscosity data has also been used to determine relative viscosity, η_r . The calculated parameters are discussed in terms of various solute-solute and solute-solvent interactions prevailing in the solution. Further, a detailed insight into the physicochemical interactions between Asp and aqueous PD, the structure-making tendency has been retrieved through the perusal of these calculated parameters.

Keywords - Apparent molar volume, L-aspartic acid, Density, 1, 2-Propanediol, Transport properties

I. INTRODUCTION

The direct study of interaction between proteins and other components is quite difficult due to their complex conformational and configurational three-dimensional structures. Amino acids, the model compounds for proteins, are important biological active molecules and are useful to investigate the solvation behavior [1,2]. The choice of water for preparing mixed solvent stems from its important and unique role in determining the structure and stability of proteins. Its presence also gives rise to hydrophobic forces, which are of prime importance in stabilizing native globular structure of protein [3-4]. Binary solvents, representing a concentrated mixture of a co-solvent and water as the principal solvent, are of special interest from both the biological and the physico-chemical viewpoints [5-7]. The added co-solvent may exert a stabilizing or destabilizing influence or neutral effect [8] on the native conformation of a protein.

It is known that polyhydric alcohols increase the thermal stability of proteins or reduce the extent of their denaturation by other substances [9,10]. The alkane diols have wide range of applications in pharmacology and cosmetic industry, however, they are not components of living organisms, but they act as a vehicle for pharmaceuticals or cosmetics when introduced into living organisms [11]. Thus, the properties of amino acids in aqueous-polyols solutions are essential in understanding the chemistry of biological systems [12-14].

The literature survey reveals physical and thermodynamic studies on Asp and inorganic salts as a function of temperature [15, 16]. The effect of dielectric constant on protonation equilibria of Asp in one of the alkane diol-

water mixtures has been reported by Rani *et al.* [17]. The earlier reports mention the volumetric, acoustic and viscometric properties of Asp having different molalities in water and in limited percentage range of aqueous PD solutions [18]. But no report seems to be available on thermodynamic and transport properties of PD + water mixture and Asp + PD + water mixture in which concentration of PD varies from 10 to 90%. Hence, we herein report the effect of change in concentration of PD from 10% to 90% and on addition of Asp on thermodynamic and transport properties of solutions.

II. EXPERIMENTAL

Asp and PD with mass fraction purity >99% obtained from Sigma Aldrich were used as such without further purification. Freshly prepared triply distilled with specific conductance less than $1 \times 10^{-6} \text{ S cm}^{-1}$ was used for the preparation of solutions in which 0.5M hydrochloric acid was added to increase the solubility [17]. All the solutions were prepared with care and stored in special airtight bottles to avoid the exposure of solutions to air and evaporation. The solutions were prepared by weighing on an electronic single pan five digit Mettler Balance (Model AE-240) with an accuracy of $\pm 0.01 \text{ mg}$. The densities of the solutions were measured by using a single capillary pycnometer. Prior to use, the pycnometer was calibrated using triply distilled water. After filling the pycnometer with sample, it was immersed in thermostat for about 20 minutes in order to maintain the constant temperature. After 20 minutes, pycnometer was removed from thermostat and weighed on a sensitive single pan five digit balance with an accuracy of $\pm 0.01 \text{ mg}$. The accuracy during the measurement was within $\pm 5 \times 10^{-5} \text{ g cm}^{-3}$. The speeds of sound in the solutions were measured using a single-crystal variable-path multi-frequency ultrasonic interferometer (M-82, Mittal Enterprises) having stainless steel sample cell (with digital micrometer) operating at fixed frequency of 4 MHz. The uncertainty in speeds of sound measurements was found to be within $\pm 0.5 \text{ m s}^{-1}$. The temperature of the sample solutions was maintained to an accuracy of $\pm 0.01 \text{ K}$ in an electronically controlled thermostatic water bath (Model: TIC-4000N, Thermotech, India). The viscosities of the solutions were measured by using Ubbelohde viscometer. The viscometer containing the test liquid was allowed to stand for about 30 minutes in a thermostatic water bath so that the thermal fluctuations in viscometer were minimized. The time of flow was recorded with a digital stopwatch with an accuracy of $\pm 0.01 \text{ s}$. The accuracy of viscosity measurements was found to be $\pm 1 \times 10^{-6} \text{ Pa s}$.

III. RESULTS AND DISCUSSION

3.1. Density

The experimental values of density (Table 1) of binary solution (water + PD) and ternary solution (water + PD + Asp) have been measured in which molality of Asp (solute) is fixed at 0.05 mol kg^{-1} for all ternary solutions while concentrations of PD vary from 10 to 90%. The binary solution comprising different concentrations of water and PD (from 10 to 90% PD) is considered as solvent.

The density increases as the concentration of PD in water is varied from 10% to 70% and then decreases with further increase of PD concentration up to 90%. The increase of density with increase in concentration of PD from 10% to

70% binary solution indicates the increase in interaction between PD and water. The -OH group of PD is highly polar; it has a strong electron-pair donating ability which is capable of forming HO...HOH hydrogen bond with water molecules. This leads to shrinkage in the volume resulting in increase in density of solvent up to 70% PD. Further, from 70% to 90% of PD, density decreases which may be attributed to weakening of the molecular forces resulting in increase in volume and, hence, decrease in density at higher concentration.

A comparison between binary solution (i.e., solvent) and ternary solution (i.e., solution) shows that density of solution is greater than solvent for concentrations up to 70%. This change in density may be that on addition of Asp, ion-hydrophilic interactions between -NH_3^+ and -COO^- head groups of Asp and -OH group of PD, and hydrophilic-hydrophilic interactions between $\text{-CH}_2\text{COOH}$ group of Asp and -OH group of PD takes place. In other words, the increase in density may be interpreted to the structure-maker of the solvent due to the added solute.

3.2. Apparent molar volume

The values of density were used to calculate apparent molar volume (V_ϕ) of the solutions using Eq. (1):

$$V_\phi = (M/\rho) - [(\rho - \rho_0) 1000/m\rho\rho_0] \quad (1)$$

where m is the molality (mol kg^{-1}) of the Asp, M is the molar mass of the Asp (kg mol^{-1}), and ρ_0 and ρ are the densities (kg m^{-3}) of the solvent and solution, respectively. The values of apparent molar volume are reported in Table 1, while the curve for later is presented in Fig. 1. The results reveal that the apparent molar volume increases with increase in concentration of PD. This may be attributed to the increase in solvation (release of some solvent molecules from loose solvation layers of the solute in solution) of Asp at higher concentration of PD due to strong attractive interactions [19]. The high value of apparent molar volume for Asp with increasing concentration of PD (10 to 90% PD) as compared to their values in aqueous solution (0% PD) suggests that interactions mentioned in section 3.1 dominates over the ion-hydrophobic, hydrophilic-hydrophobic and hydrophobic-hydrophobic interactions.

It is important to mention here that on addition of Asp to the co-solute there is decrease in the pH of solution, and the side chains of the acidic amino acids remain fully deprotonated [20]. Thus, the amino acid studied mainly exists in zwitterionic form in co-solute solution and their side chains remain fully deprotonated due to hydrolysis. Thus, deprotonated amino acid interacts strongly with aqueous solvent resulting in increase in apparent molar volume.

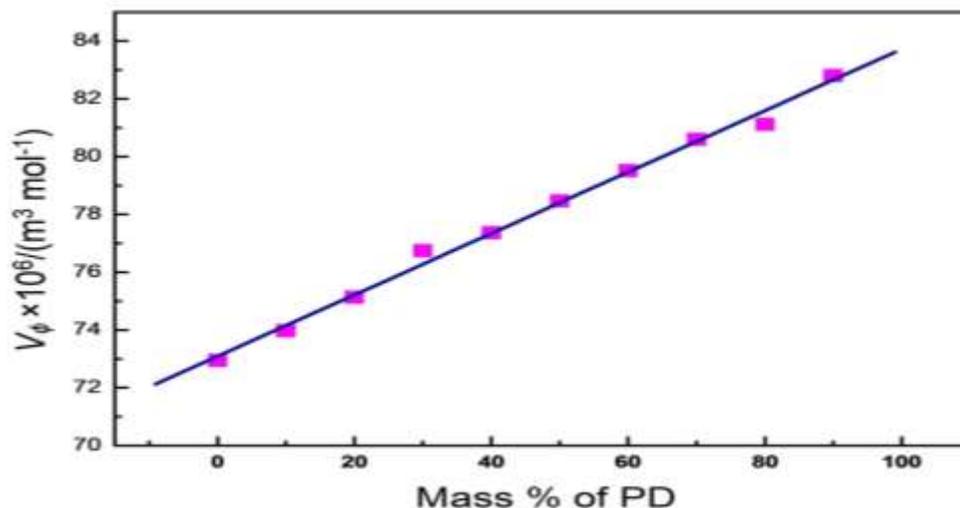


Fig.1: Plot of apparent molal volume, V_{ϕ} , v/s mass % of PD in 0.05 mol kg^{-1} Asp at 303.15 K.

Table 1: Density of solvent (water + PD), solution (water + PD+ Asp) and apparent molar volume of solution at 303.15 K.

Concentration of PD (%)	$\rho / (\text{kg m}^{-3})$		$V_{\phi} \times 10^6 / (\text{m}^3 \text{mol}^{-1})$
	Solvent	Solution	
0	1003.655	1006.648	72.96
10	1010.447	1013.383	73.98
20	1017.292	1020.163	75.14
30	1024.427	1027.206	76.75
40	1030.975	1033.712	77.37
50	1037.416	1040.086	78.47
60	1042.779	1045.384	79.52
70	1047.945	1050.482	80.60
80	1045.743	1048.256	81.12
90	1042.015	1044.443	82.81

3.3. Speed of sound

The experimental values of speed of sound for both solvent and solution are reported in Table 2. It is observed that the speed of sound in water-PD mixture (from 10 to 90% binary solution) is higher than that of water (0% PD). The

speed of sound increases as the concentration of PD in water is varied from 10% to 60% and then decreases with further increase of PD concentration up to 90%. This decrease in speed of sound may be due to the reason that the increase in concentration of the PD weakens the molecular forces and hence the abrupt change in speed of sound is observed at higher concentration. The increase in speed of sound up to 60% with the increase in PD concentration may be attributed to the formation of hydrogen bond between water and PD having two hydroxyl groups. As PD concentration is increased, the number of OH groups available for hydrogen bond formation will increase resulting in large number of O-H-O bonds between water and PD molecules. In other words, this suggests that there is strong intermolecular association between the solute and solvent molecules because of which the close-packed structures of water absorb the sound energy and hence the speed of sound increases (up to 60%). This probably explains the observed increase in speed of sound as PD concentration is increased. When amino acid (Asp with fixed molality) is added to the solvent, speed of sound showed same trend as discussed above.

Upon comparison between solvent and solution, it has been observed that speed of sound in solution is more than that observed in solvent for concentrations up to 60%. This increase in speed of sound in solution may be attributed to the intermolecular interactions among Asp, PD and water molecules. As a result of these interactions, overall compressibility of the medium decreases and hence speed of sound increases. But above a particular concentration (from 70% to 90%) of PD, although the interaction of Asp with PD (Asp-PD) and water molecules (Asp-water) increases but simultaneously the molecular interactions between PD and water molecules (PD-water) decreases. As a result overall compressibility of the medium increases and hence speed of sound decreases.

3.4. Isentropic compressibility

The speed of sound data has been used to calculate isentropic compressibility by using following equation:

$$k_s = 1 / (u^2 \rho) \quad (2)$$

where k_s and k_s^0 are isentropic compressibilities of solution and solvent, respectively.

The values of isentropic compressibilities for both solvent and solution are reported in Table 2. It is observed that the isentropic compressibility for both solvent and solution decreases as the concentration of PD in water is varied from 10% to 60%, followed by an increase up to 90% concentration. This decrease of isentropic compressibility of solvent as well as solution with increase in concentration of PD from 10% to 60% may be attributed to the electrostatic interaction between solute and solvent which makes the solution rather incompressible and, hence, electrostrictive compression of solvent around the solute molecule increases. Thus, the decrease in the k_s value with the increase in the PD concentration is an indicative of the presence of solute-solvent interactions. At higher concentration, isentropic compressibility increases as we increase the concentration of PD. It may be due to the fact that the solute-solvent interactions get weakened with the increase in concentration of PD which may be attributed to the decrease in solvation of ions by water.

3.5. Apparent molar isentropic compression

The apparent molar isentropic compression, $K_{\phi,s}$, has been calculated by using Eq. (3).

$$K_{\phi,s} = (Mk_s/\rho) - [(k_s^0\rho - k_s\rho_0)/m\rho\rho_0] \quad (3)$$

The magnitude of negative values of apparent molar isentropic compression decreases with increase in the concentration of PD in solutions as shown in Table 2. The linear variation of apparent molar isentropic compression with concentration of PD is shown in Fig. 2 which indicates that as the concentration of PD increases, magnitude of negative values of apparent molar isentropic compression of solution decreases. The negative $K_{\phi,s}$ values show that water molecules around solute are less compressible than those in the bulk which may be attributed to strong solute-solvent interactions between Asp and PD.

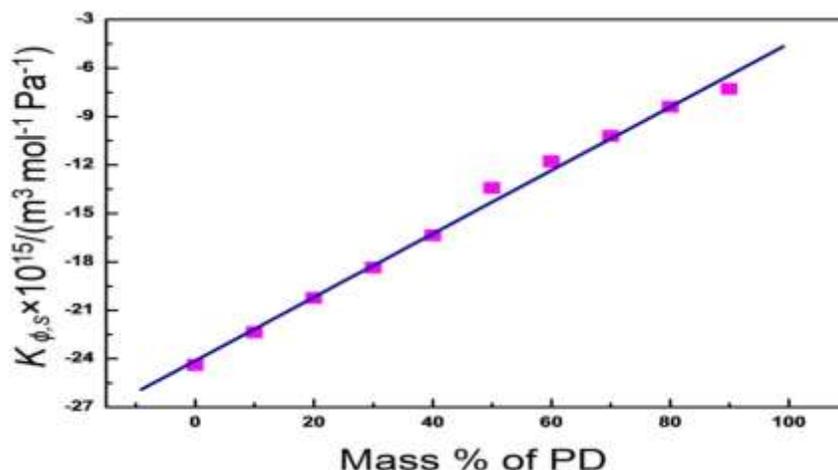


Fig. 2: Plot of apparent molal isentropic compression, $K_{\phi,s}$ v/s mass % of PD in 0.05 mol kg⁻¹ Asp at 303.15 K.

Table 2: Speed of sound, isentropic compressibility of solvent, solution and apparent molar isentropic compression of solution at 303.15 K.

Concentration of PD (%)	$u / (\text{m s}^{-1})$		$k_s \times 10^{10} / (\text{Pa}^{-1})$		$K_{\phi,s} \times 10^{15} / (\text{m}^3 \text{mol}^{-1} \text{Pa}^{-1})$
	Solvent	Solution	Solvent	Solution	
0	1510.50	1513.13	4.36690	4.33878	-24.37
10	1568.00	1570.85	4.02526	3.99901	-22.36
20	1613.91	1616.92	3.77394	3.74932	-20.23
30	1656.91	1659.20	3.55957	3.53626	-18.36
40	1683.60	1686.80	3.42195	3.39995	-16.36

50	1692.57	1695.55	3.36475	3.34417	-13.43
60	1696.01	1698.97	3.33387	3.31398	-11.79
70	1679.55	1682.39	3.38277	3.36321	-10.22
80	1632.53	1635.03	3.58798	3.56845	-08.40
90	1595.13	1597.51	3.77167	3.75168	-07.30

3.6. Specific acoustic impedance

Specific acoustic impedance (Z) is calculated by using standard relation [21]:

$$Z = u \times \rho \tag{4}$$

The values of specific acoustic impedance are reported in Table 3. These are found to be almost inversely proportional to the isentropic compressibility (k_s), the proportionality constant being reciprocal of corresponding speed of sound. The specific acoustic impedance increases linearly as the concentration of PD in water is varied from 10% to 60% and then decreases with further increase of concentration up to 90%. This increase of specific acoustic impedance with increase in concentration of PD from 10% to 60% confirms the presence of molecular association between solute and solvent molecules through intermolecular hydrogen bonding. The addition of Asp has similar effects when various concentration of binary solution is used. That is the increase in specific acoustic impedance with addition of Asp may be due to the presence of bulkier solute (Asp) due to solute-solvent interactions which restricts the free flow of sound waves.

3.7. Relative association

The values of density and speed of sound enable also to calculate the relative association R_A , using the equation:

$$R_A = (\rho/\rho_o) (u_o/u)^{1/3} \tag{5}$$

where symbols have their usual meanings.

The relative association is the function of speed of sound which is influenced by two factors: (a) The breaking up of the associated solvent structures on addition of solute to it, and (b) the solvation of solute molecule. The first factor results in decrease while the second one results in increase in the value of R_A . From Table 3, it is observed that the relative association of ternary solution decreases with increase in the concentration suggesting formation of complex.

3.8. Intermolecular free length

The intermolecular free length has been calculated using the following equation:

$$L_f = K (k_s)^{1/2} \tag{6}$$

where K is the temperature dependant Jacobson constant [22] which is calculated from the relation $(91.368+0.3565T) \times 10^{-8}$, where T is the absolute temperature.

The intermolecular free length depends on isentropic compressibility and shows behaviour similar to that of isentropic compressibility and inverse to that of speed of sound. Intermolecular free length is a predominant factor in determining the variation of speed of sound in liquid mixtures.

It has been observed from Table 3 that the intermolecular free length in water-PD mixture (from 10% to 90% binary solution) and also upon addition of solute, i.e., Asp, is lower than that of water. It decreases as the concentration of PD in water is varied from 10% to 70%, followed by an increase in concentration up to 90%. The various reasons for decrease in intermolecular free length with increase in concentration may be attributed to (i) decrease in gap between two species (water and PD, and between Asp and water + PD), (ii) gain of dipolar association, and/or (iii) making up of hydrogen bonds in liquid mixtures. This may be an indication of significant interactions happening between water and PD, and between Asp and solvent (water + PD), thereby, suggesting the structure promoting behaviour of Asp up to a fixed concentration. The reverse of parameters discussed in (i) to (iii) may be responsible for increase in intermolecular free length.

Table 3: Specific acoustic impedance, mean free length of solvent as well as solution and relative association of solution at 303.15 K

Concentration of PD (%)	$Z \times 10^6 / (\text{kg m}^{-2} \text{s}^{-1})$		$L_f \times 10^{11} / \text{m}$		R_A
	Solvent	Solution	Solvent	Solution	
0	1.51602	1.52319	4.16771	4.15427	1.00239
10	1.58438	1.59187	4.00136	3.98830	1.00229
20	1.64181	1.64952	3.87444	3.86177	1.00219
30	1.69738	1.70434	3.76279	3.75044	1.00206
40	1.73574	1.74366	3.68932	3.67745	1.00201
50	1.75589	1.76352	3.65836	3.64715	1.00198
60	1.76856	1.77608	3.64153	3.63066	1.00191
70	1.76008	1.76732	3.66816	3.65753	1.00185
80	1.70721	1.71393	3.77777	3.76748	1.00189
90	1.66214	1.66851	3.87326	3.86299	1.00183

3.9. Viscosity

The viscosity measurement has been analyzed in terms of Jones-Dole [23] equation:

$$\eta_r = \eta/\eta_0 = 1 + BC \quad (7)$$

where C is the molarity (calculated from molality) of solution, η_r is the relative viscosity, η_o and η are the viscosities of solvent and solution, respectively. The value of viscosity B-coefficients (B), calculated using least squares fit, depends upon the size of solute and nature of solute-solvent interactions and is definite for solute-solvent system. The viscosity of a solvent or solution is a measure of cohesiveness or rigidity present between either ions or ion-solvent or solution. Table 4 shows that viscosity of binary solution increases with increase in the concentration (from 10% to 90% binary solution) of PD which indicates that molecular interactions exist between solute and solvent. This table also shows that viscosity of ternary solution also increases with increase in the concentration of PD. It also evidences the existence of molecular interactions between Asp and solvent.

The relative viscosity calculated as the ratio of viscosity of solution (η) and corresponding solvent (η_o), i.e., $\eta_r = \eta/\eta_o$, is also displayed in Table 4. The linear variation of relative viscosity with concentration of PD, shown in Fig. 3, indicates that as the concentration of PD increases, relative viscosity of solution also increases which suggest that there exist strong solute-solvent interactions in the system which immobilizes the solvent molecules, present obstruction to viscous flow of solution, thus, increasing the relative viscosity.

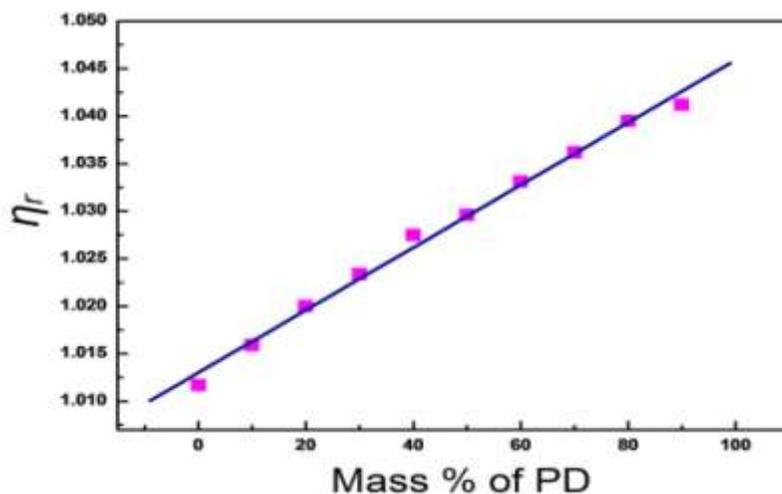


Fig. 3: Plot of relative viscosity, η_r , v/s mass % of PD in 0.05 mol kg⁻¹ Asp at 303.15 K.

Table 4: Viscosity of solvent, solution and relative viscosity of solution at 303.15 K.

Concentration of 1,2- propanediol (%)	$\eta \times 10^3 / (\text{Pa s})$		η_r
	Solvent	Solution	
0	0.8241	0.8338	1.0117
10	0.9999	1.0158	1.0159
20	1.3404	1.3673	1.0200

30	1.7855	1.8273	1.0234
40	2.5432	2.6132	1.0275
50	3.5741	3.6802	1.0296
60	5.0259	5.1924	1.0331
70	6.8801	7.1298	1.0362
80	10.5177	10.9334	1.0395
90	15.7852	16.4368	1.0412

IV CONCLUSION

Density, speed of sound and viscosity of PD + water and PD + water + Asp solutions measured at 303.15 K have been reported in this study in which concentration of PD (mass % of PD) have been varied from 10 to 90%. From the experimental data, various parameters like apparent molar volume, V_{ϕ} , isentropic compressibility, k_s , apparent molar isentropic compression, $K_{\phi,ss}$, specific acoustic impedance, Z , intermolecular free length, L_f , and relative association, R_A , viscosity, η , and, relative viscosity, η_r , have been calculated. The results of apparent molar volume indicates that ion-hydrophilic interactions between $-NH_3^+$ and $-COO^-$ head groups of Asp and $-OH$ group of PD, and hydrophilic-hydrophilic interactions between $-CH_2COOH$ group of Asp and $-OH$ group of PD dominates over the ion-hydrophobic, hydrophilic-hydrophobic and hydrophobic-hydrophobic interactions. The negative values of apparent molar isentropic compression, $K_{\phi,ss}$, are indicative of strong solute-solvent interactions between Asp and PD. The decrease in values of intermolecular free length upto 70% suggest that significant interactions happening between water and PD in case of binary solution, and between Asp and PD in case of ternary solution, thereby, suggesting the structure promoting behaviour of Asp. The relative viscosity, η_r , of solution increases which suggest that there exist strong solute-solvent interactions in the system which immobilizes the solvent molecules, present obstruction to viscous flow of solution.

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