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PHYSICOCHEMICAL INVESTIGATION OF DIVERSE INTERACTIONS OF SOME BIOLOGICALLY POTENT MOLECULES IN AQUEOUS GREEN ENVIRONMENTS AT DIFFERENT TEMPERATURES

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ABSTRACT

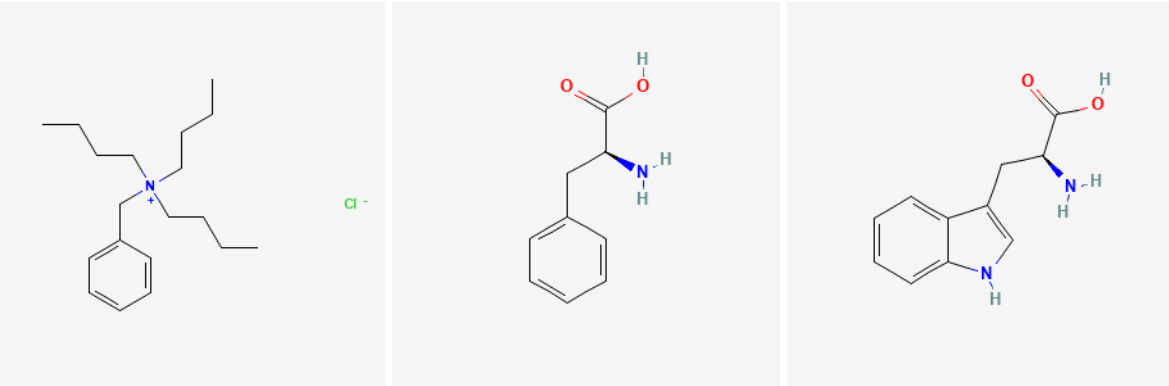
The densities, viscosities, refractive index, conductanceand surface tension of L-phenylalanine and L-tryptophan in aqueous Benzyltributylammonium chloride (BTBACl)solutions have been measured at 298.15K, 303.15K and 308.15K. Apparent molar volumes and viscosity *B*-coefficients are obtained from these data supplemented with densities and viscosities, respectively. The limiting apparent molar volumes and experimental slopesderived from the Masson equation have been interpreted in terms of solute-solvent and solute-solute interactions, respectively. The viscosity data have been analyzed using the Jones-Dole equation, and the derived parameters *B* and *A* have also been interpreted in terms of solute-solvent and solute-solute interactions, respectively. The structure making/breaking capacities of the solutes in the studied solvent systems have been discussed. Lorentz-Lorenz equation was used to calculate the molar refraction. The specific conductance and surface tension values also explained and supported the interaction properties.

KEYWORDS: Ionic Liquid, Amino acids, Solute-Solvent and Solute-Solute Interactions; Structure Maker and Breaker solutes.

INTRODUCTION

Studies on densities, viscosities, refraction, conductance and surface tension are of great use in characterizing the structure and properties of solutions. Various types of interactions exist between the solutes in solutions, and these solute-solute and solute-solvent interactions are of great interest in all branches of chemistry. These interactions provide a better understanding of the nature of the solute and solvent, i.e., whether the solute modifies or distorts the structure of the solvent.

An ionic liquid (IL) is a salt in the liquid state which has melting point below the temperature such as 373.15 K with negligible vapor pressure and they also enhance their importance in industrial process due to the presence of asymmetric cation and anion. In many inorganic and biocatalytic reactions they are used as designer solvents" and "green" alternative for volatile organic solvents. They can be utilized as heat transfer fluids in processing biomass, as conductive liquids in batteries and solar cells and also in analytical equipment. They make up electrolytes in lithium-ion batteries, super capacitors and metal plating baths. Benzyltributylammonium chloride or BTBACl, $C_{19}H_{34}ClN$ is water soluble Ionic Liquid (IL). In many biphasic organic transitions such as in the agrochemicals, polymer and pharmaceutical industries, it can be used as phase transfer catalyst.[1]



Scheme 1: Molecular structures of BTBACl, L-Phenylalanine and L-Tryptophan.

Amino acids, the basic component of protein molecules particularly important in biochemistryare serious to life, and have many functions in metabolism. One particularly important function is to serve as the building blocks of proteins. Due to their central role in biochemistry; amino acids are important in nutrition and are commonly used in food

technology and industry and they arequite helpful in understanding the various interactions in solutions.[2] L-phenylalanine, $C_9H_{11}NO_2$ is an essential amino acid. It is the only form of phenylalanine found in proteins. Major dietary sources of L-phenylalanine include meat, fish, eggs, cheese, and milk. Phenylalanine is most commonly used for a skin disorder that causes white patches to develop on the skin (vitiligo).L-tryptophan, $C_{11}H_{12}N_2O_2$ is an essential amino acid that is necessary for making proteins. It is naturally found in red meat, poultry, eggs, and dairy. L-tryptophan is important for many organs in the body. L-tryptophan is not made by the body and must be consumed from the diet.

In the present work, an attempt has been made to provide an interpretation of solute-solvent and solute-solute interactions of the amino acids prevailing in 0.001m, 0.003m and 0.005m IL solutions at 298.15K, 303.15K and 308.15K.

Experimental Section

Source and Purification of the Chemicals Used

The studied IL, Benzyltributylammonium chloride(M.W. 311.93 g/mole) and amino acids, L-phenylalanine (M.W. 165.9 g/mole) and L-tryptophan (M.W. 204.23 g/mole) of purissgrade were purchased from Sigma-Aldrich India and TCI chemicals India Pvt. Ltd. respectively. The purity of mass of taken salts were ≥0.99 which was confirmed byemploying 1 H NMR spectroscopic studies for pure IL and the amino acids used. The amino acids were dried in vacuum for about 48 h at room temperature. Freshly distilled water was used for the experimental purpose.

Apparatus and Procedure

The vibrating-tube Anton Paar Density-Meter (DMA 4500 M) was used to measure the density with a precision of 0.00001×10^{-3} ($kg \cdot m^{-3}$). The double-distilled water and dry air was used to calibrate the density meter. The temperature was kept constant automatically within range ±0.01 K.

The suspended Ubbelohde viscometer was used to measure the viscosity of the solutions, which was calibrated at room temperature (298.15 K) using doubly distilled water. It was purified with methanol and using viscosity, density data from literature. The viscometer was dried and cleaned perfectly through hot thermostat and then filled with experimental solution, vertically placed in a glass-walled thermostat (Bose–Panda instruments Pvt. Ltd.) This instrument was maintained to 0.01 K. At thermal equilibrium the flow-time were recorded

with the help of a stop watch with an accuracy of ±0.01 s. The uncertainty for the measurement of viscosity was $\pm 0.2 \times 10^{-3}$mPa.s.

Measurement of refractive index has been done with the help of a Digital Refractometer Mettler Toledo instrument. The accuracy for the refractive index measurement was ±0.0002units approximately. Doubly distilled water used to calibrate the refractometer twice for measurement of the refractive indices of experimental solutions.

The conductivity measurement was done by Mettler Toledo Instrument(In Lab730 probe cell). The specification of the cell has measurement of conductivity range(0.01–1000 mS/cm). The cell type 4 graphite having a cell constant 0.56 cm^{-1}. The calibration of the cell has been done with 0.01 N (NaCl solution). The accuracy of the conductance measurement was ±0.5%. The specific conductance of the experimental solution having concentration (0.0010, 0.0025, 0.0040, 0.0055, 0.0070, 0.0085) molality, m was reported at 298.15, 303.15, 308.15 K.

At 298.15 K, a Tensiometer (K9, KRUSS; Germany) was used to search and find the surface tensions of the solutions using the platinum ring detachment technique. The temperature of the device was controlled by circulating auto-thermostatic water through the solution in a double-wall glass vessel. The calibration of the K9 Digital Tensiometer was carried out with doubly distilled water. The measurement was accurate to within ± 0.3 mN/m.

The mixtures were prepared by mixing known volume of solutions in airtight-stoppered bottles. At first, we prepared 0.001mIL, 0.003mIL, 0.005mILin 250 ml water. Then there was preparation of 0.01m L-phenylalanine and 0.01m L-tryptophan in the aqueous ionic liquid solutions. We prepared and used: 30 ml, 0.001m IL +0.01 m amino acid in six sets (0.0010, 0.0025, 0.0040, 0.0055, 0.0070, 0.0085 m by dilution), 0.003m IL + 0.01m amino acid in six sets (0.0010, 0.0025, 0.0040, 0.0055, 0.0070, 0.0085 m by dilution),0.005m IL + 0.01m amino acid in six sets (0.0010, 0.0025, 0.0040, 0.0055, 0.0070, 0.0085 m by dilution) at (298.15, 303.15, 308.15) K for experimental purpose. A Mettler AG-285 electronic balance was used to measure mass of the stock solutions with a precision of $\pm 0.0001 \times 10^{-3}$ kg.[1-3]

RESULTS AND DISCUSSION

Density

The experimental values of density (ρ) and viscosity (η)of different molality (m) of aqueous IL (BTBACl) solution at 298.15K, 303.15K and 308.15K are reported in **Table 1.**Experimental values of refractive index (n_D) of different molality (m) of aqueous IL (BTBACl) solution at 298.15K, 303.15K and 308.15K are shown in **Table 2**. The density (ρ), viscosity (η) and molar refraction (R_M)values of L-phenylalanine and L-tryptophan in aqueous (BTBACl)ionic liquid solutions at 298.15K, 303.15K and 308.15K are given in **Table 3** and **Table 4**, respectively.

The apparent molar volumes (Φ_V) were determined from the solution densities using the following equation:

$$\Phi_V = M / \rho - 1000 (\rho - \rho_0) / (m.\rho.\rho_0) \quad \ldots\ldots\ldots(1)$$

Where M is the molar mass of the solute, m is concentrationin molality of the solution, and ρ_0 and ρ are the densities of the solvent and solution, respectively. The $\Phi_V$values of L-phenylalanine and L-tryptophan solution in 0.001m, 0.003m and 0.005m aqueous BTBACl solution at different temperatures (298.15K, 303.15K, 308.15K) are given in **Table 5** and **Table 6**, respectively.

It was observed that Φ_V increases with increase in temperature as well as increase in concentration of the IL but decreases as the amount of both the amino acids increases in the solution.

The limiting apparent molar volumes (Φ_V^0) were calculated using a least-squares treatment to the plots of $\Phi_V$versus$\sqrt{m}$using the following Masson equation:[4]

$$\Phi_V = \Phi_V^0 + S_V^* \sqrt{m} \quad \ldots\ldots\ldots(2)$$

Where ϕ_V^0 is the partial molar volume at infinite dilution and S_V^*isthe experimental slope. The values of $\Phi_V^0$and S_V^* are reported in **Table 8a** and **Table 8b**and **Figure 1a** and **Figure 1b** gives the plot of Φ_V^0forL-phenylalanine and L-tryptophan solution in the IL, respectively.

As the investigated systems are characterized by hydrogen bonds, the solute-solvent and solute-solute interactions can be interpreted in terms of structural changes, which arise due to hydrogen bond interactions present between various components of the solvent and solution

systems. This indicates the presence of strong solute-solvent interactions, and these interactions are strengthened with a rise in temperature and increase in concentration of the IL but weakened with an increase in the amount of the amino acids in solutions, suggesting larger electrostriction at higher temperature with greater amount of IL but lesser amount of amino acids in the solution mixture.

The S_V^* values are negative for all temperatures at all concentration of the solutions of IL and amino acids. Since S_V^* is a measure of solute-solute interactions, the results indicate the presence of weak solute-solute interactions. These interactions, however, decrease with a rise in temperature and increase in concentration of IL, which is attributed to more violent thermal agitation at higher temperatures, resulting in diminishing the force of solute-solute interactions (ionic dissociation).[5] The higher S_V^* values in L-phenylalanine solutions compared to L-tryptophan solutions is due to a decrease in solvation of ions, i.e., more and more L-phenylalanine solutes are accommodated in the void space left in the packing of large associated benzyltributylammonium chloride solvent molecules with the addition of this amino acid to the mixture.

The variation of $\Phi_V^0$with temperature of the amino acids in IL solvent mixtures follows the polynomial:

$$\Phi_V^0 = a_0 + a_1.T + a_2.T^2 \qquad \ldots\ldots\ldots(3)$$

over the temperature range under investigation where T is the temperature in K.

Values of the empirical coefficients, a_0, a_1,a_2 in the above equation for the amino acids in various compositions of the IL are reported in **Table 9**.

The limiting apparent molar expansibilities (Φ^0_E) can be obtained by the following equation:

$$\Phi^0_E = (\delta\Phi_V^0/\delta T)_P = a_1 + 2\,a_2\,T \qquad \ldots\ldots\ldots(4)$$

The values of Φ^0_E of the studied compoundsat 298.15K, 303.15K and 313.15 K are determined and reported in **Table 10a** and **Table 10b**.

It is found from **Table 10a** and **Table 10b**that the values of Φ^0_Edecrease with a rise in temperature and L-tryptophan solutions in IL are found to have greater values compared to L-phenylalanine solutions, which can be ascribed to the absence of caging or packing effects of L-tryptophan molecules in the IL mixtures.[6]

During the past few years it has been emphasized by different workers that S_V^* is not the sole criterion for determining the structure-making or breaking nature of any solute. Hepler[7] developed a technique of examining the sign of $(\delta^2\Phi_V^0/\delta T^2)_P$ for the solute in terms of long-range structure-making and structure-breaking capacity of the solute in the mixed solvent systems using the general thermodynamic expression:

$$(\delta c_P/\ \delta P)_T = -\ (\delta^{\ 2}\ \Phi V^0/\ \delta\ T^{\ 2})_P \qquad \ldots\ldots\ldots(5)$$

On the basis of this expression, it has been deduced that the structure-making solutes should have positive values, whereas structure-breaking solutes should have negative values. In our present investigation, it is evident from **Table 10a and 10b** that $(\delta^2\Phi_V^0/\delta T^2)_P$ or $(\delta\Phi^0_E/\delta T)_P$values are less negative and even positive at 0.005m IL solution of L-phenylalanine which is acting as a structure-maker solute in this ionic liquid whereas the negative values of L-tryptophan thereby proves that it acts as a structure-breaker in theIL solvent mixtures and the effect being maximum at 0.005m IL concentration solution.

Viscosity

The viscosity data of solutions for the amino acids in the IL solutions have been analyzed using the Jones-Dole[8] equation:

$$(\eta\ /\ \eta_0 - 1)\ /m^{1/2} = A + Bm^{1/2} \quad \text{where } \eta = (k.\ t - \ l/t)\ \rho \qquad \ldots\ldots\ldots(6)$$

Where η_0 and η are the viscosities of solvent and solution, respectively, k and l are the constants for a particular viscometer and t is the flow time of the solvent/solution in seconds. A and B are estimated by least-squares method and obtained from the intercept and slope, respectively by plotting $(\eta/\eta_0 - 1)\ /m^{1/2}$ against $m^{1/2}$. The A and B values are reported in **Table 8a** and **Table 8b** for L-phenylalanine and L-tryptophan, respectively. The representative plots of Bagainst temperature at different concentration of the IL for L-phenylalanine and L-trytophan solutionshave been shown in **Figure 2a** and **Figure 2b**, respectively.

A perusal of **Table 8a** and **Table 8b** shows that the values of the A-coefficient are decreasing whereas the B-coefficient values are increasing with increase in temperature and concentration of IL solutions. These results indicate the presence of weak solute-solute interactions and strong solute-solvent interactions in the studied solvent systems. These conclusions are in excellent agreement with those drawn here from Φ_V^0 values.

It has been reported in a number of studies[9,10] that *dB/dT* is a better criterion for determining the structure-making/breaking nature of any solute rather than simply the value of the *B*-coefficient. The positive*dB/dT* in **Table 8c** and **Table 8d** suggest thatthe amino acids are acting as a structure-maker solutes in this ionic liquid. These conclusions from viscosity measurements are in agreement with those drawn from the density parameters of the studied solution of the amino acids and the IL.

Refractive Index

The molar refraction, $R_M$can be evaluated from the Lorentz–Lorenz relation:[11,12]

$$R_M=\{(n_D^2-1)/(n_D^2+2)\}(M/\rho)\ldots\ldots \quad (7)$$

Where R_M, n_D, M and $\rho$are the molar refraction, the refractive index, the molar mass and the density of the solution, respectively. The refractive index of a substance can be represented as the ratio c_o/c, where $c_o$denotes the speed of light in vacuum and c is the speed of light in the medium. In a more simple way, the refractive index of a substance can be described by its ability to refract light as it moves from one medium to another and thus, the higher the refractive index of a substance, the more the light is refracted.[13] As stated by Deetlefs et al.[14] a substance in which molecules are more tightly bound or denser, it shows higher refractive index. Furthermore, the compactness of the system can also be well measured by refractive index as it is the ability to refract light in a given medium. Table3 and Table 4 shows the molar refraction (R_M) variation of L-phenylalanine and L-tryptophan, respectively in aqueous BTBACl solution at 298.15 K, 303.15 K and 308.15 K. The values seem to increase with increase in temperature only forL-phenylalaninein 0.001m IL but as the amount of the IL is increased in solution R_M decreases at higher temperatures for both the amino acids at all concentrations. Moreover R_M is greater in L-Tryptophan solutions. Table 7a and Table 7b displays the refractive index (n_D) values which seem to decrease with increase in temperature at all concentration of the IL mixtures with higher values in L-Tryptophan solutions. This is in good agreement with the volumetric and viscometric results attained here. The higher refractive index shows that the molecules are more tightly packed in the solution.

The limiting molar refraction, (R_M^0) listed in **Table 8a** and **Table 8b** can be calculated using the following equation:

$$R_M = R_M^0 + R_S\sqrt{m}\ldots\ldots\ldots(8)$$

Where, 'm' is the molality of solution and R_M^0 is the limiting molar refraction that signifies solute–solvent interaction. So, these measurements are used as an extensive tool for studying the molecular interaction in aqueous solution. Regular increase in the values of R_M^0 as shown in **Figure 3a** and **Figure 3b** for L-phenylalanine and L-tryptophan solutions in BTBACl signifies that solute–solvent interaction is predominant over solute-solute interactions.

Electrical conductance

Tables 7a and **Table 7b** show the specific conductance (κ) values and **Table 11** shows the molar conductance (Λ) of L-phenylalanine and L-tryptophan solution in (0.001 m, 0.003 m, 0.005 m) IL at 298.15, 303.15 and 308.15 K. **Figure 4a** and **Figure 4b** gives the variation of molar conductance (Λ)with different concentrations of L–phenylalanine and L-tryptophan in aqueous (0.001 m, 0.003 m, 0.005 m) BTBACl (IL) solution at 298.15K, 303.15K, 308.15 K. The specific conductance (κ) values increase with increase in temperature and increase in concentration of aqueous IL solution as well as the amino acid concentrations where L-Tryptophan is having lower values but the molar conductance (Λ)values increases with the increase in temperature and decrease with the increase in concentration of amino acid solutions. However, the values are lower for L-tryptophan for all concentrations and temperature.

The decrease of molar conductance (Λ) values may be due to the improvement of solute-solvent interaction. The development of molecular assembly empowers the ionic species to lose their independent movement making the ionic species less mobile to exhibit conductivity in solution. Hence, conductometric study also supports the volumetric, viscometric and refractometric studies of the two systems.

Surface Tension

Table 12a gives the surface tension values of L-phenylalanine and L-tryptophan. **Table 12b** gives the limiting slopes ($\partial\sigma/\partial m$) of the surface tension of the aqueous solutions of the amino acids. **Figure 5** is the plot of surface tension (σ) of L-phenylalanine and L-tryptophan as a function of different molality of aqueous BTBACl ionic liquid solutions. Surface tension values are decreasing with increase in concentration of the amino acids. In case of L-phenylalanine the values increase with increase in temperature but for L-tryptophan solutions surface tension is decreasing with increasing temperature. The sign and extent of the limiting slopes ($\partial\sigma/\partial m$) of surfacetension with reference to concentration are associated with thehydrophobic or hydrophilic character of the solute because itreflects the sort of interaction

predominant on the surface.[15,16] The amino acids have negative (∂σ/∂m)values, which are not the characteristicsof electrolytes and very polar hydrophilic compounds. The values seem to decrease with increase in concentration of the IL solutions in both the amino acids.

Table 1: Experimental values of density (ρ) and viscosity (η) of different molality (m) of aqueous IL (BTBACl) solution at 298.15K, 303.15K and 308.15K.

Conc. of Aq. IL (BTBACl) soln. in molality, m (mol.kg^{-1})	ρ×10^{-3}(kg.m^{-3})			η(mPa.s)		
	298.15K	303.15K	308.15K	298.15K	303.15K	308.15K
0.001	.99706	.99568	.99408	1.057	0.816	0.695
0.003	.99710	.99573	.99410	1.230	1.011	0.890
0.005	.99713	.99576	.99413	1.268	1.087	0.974

Table 2: Experimental values of refractive index (n_D) of different molality (m) of aqueous IL (BTBACl) solution at 298.15K, 303.15K and 308.15K.

Conc. of Aq. IL (BTBACl) soln. in molality, m (mol.kg^{-1})	n_D		
	298.15K	303.15K	308.15K
0.001	1.3321	1.3315	1.3313
0.003	1.3324	1.3322	1.3320
0.005	1.3332	1.3330	1.3327

Table 3: Density (ρ), viscosity (η) and molar refraction (R_M) of L-Phenylalanine in aqueous (BTBACl)ionic liquid solutions at 298.15K, 303.15K and 308.15K.

Conc. of L-phenylalanine soln. in molality, m (mol.kg^{-1})	ρ×10^{-3}(kg.m^{-3})			η(mPa.s)			R_M		
	298.15K	303.15K	308.15K	298.15K	303.15K	308.15K	298.15K	303.15K	308.15K
0.001m IL									
0.0010	0.99711	0.99572	0.99412	1.064	0.823	0.703	33.9386	33.9674	34.0034
0.0025	0.99719	0.99579	0.99420	1.074	0.834	0.714	33.9741	34.0122	34.0390
0.0040	0.99728	0.99587	0.99429	1.083	0.844	0.726	34.0092	34.0381	34.0742
0.0055	0.99737	0.99596	0.99439	1.093	0.855	0.739	34.0330	34.0639	34.1001
0.0070	0.99747	0.99606	0.99450	1.102	0.865	0.750	34.0609	34.0898	34.1260
0.0085	0.99757	0.99616	0.99462	1.110	0.875	0.762	34.0770	34.1060	34.1516
0.003mIL									
0.0010	0.99714	0.99576	0.99413	1.238	1.020	0.898	34.0043	33.9949	33.9763
0.0025	0.99722	0.99582	0.99421	1.248	1.033	0.913	34.0491	34.0305	34.0119
0.0040	0.99731	0.99590	0.99430	1.259	1.047	0.928	34.0750	34.0563	34.0470
0.0055	0.99741	0.99599	0.99441	1.271	1.061	0.943	34.1008	34.0822	34.0729
0.0070	0.99752	0.99609	0.99453	1.281	1.075	0.958	34.1267	34.1081	34.0988
0.0085	0.99764	0.99620	0.99467	1.291	1.089	0.973	34.1428	34.1243	34.1150
0.005mIL									
0.0010	0.99716	0.99578	0.99415	1.275	1.095	0.983	34.0043	33.9856	33.9670

0.0025	0.99724	0.99584	0.99422	1.287	1.110	1.001	34.0398	34.0212	34.0026
0.0040	0.99733	0.99592	0.99432	1.299	1.125	1.019	34.0656	34.0470	34.0377
0.0055	0.99744	0.99602	0.99444	1.312	1.140	1.037	34.0915	34.0729	34.0543
0.0070	0.99756	0.99613	0.99457	1.324	1.155	1.055	34.1081	34.0895	34.0801
0.0085	0.99770	0.99624	0.99471	1.336	1.171	1.072	34.1243	34.1057	34.0964

Table 4: Density (ρ), viscosity (η) and molar refraction (R_M) of L-Tryptophan in aqueous (BTBACl)ionic liquid solutions at 298.15K, 303.15K and 308.15K.

Conc. of L-tryptophan soln. in molality ($mol.kg^{-1}$)	$\rho\times10^{-3}(kg.m^{-3})$			η(mPa.s)			R_M		
	298.15K	303.15K	308.15K	298.15K	303.15K	308.15K	298.15K	303.15K	308.15K
0.001m IL									
0.0010	0.99713	0.99574	0.99413	1.066	0.825	0.705	42.0278	42.0163	41.9703
0.0025	0.99725	0.99585	0.99422	1.079	0.838	0.720	42.0948	42.0488	42.0142
0.0040	0.99738	0.99597	0.99434	1.092	0.851	0.734	42.1498	42.0922	42.0577
0.0055	0.99752	0.99611	0.99446	1.102	0.864	0.749	42.1817	42.1242	42.0897
0.0070	0.99766	0.99626	0.99460	1.115	0.878	0.764	42.2132	42.1558	42.1213
0.0085	0.99781	0.99641	0.99475	1.127	0.890	0.780	42.2447	42.1988	42.1528
0.003mIL									
0.0010	0.99716	0.99578	0.99414	1.240	1.024	0.900	42.0509	42.0394	42.0048
0.0025	0.99727	0.99588	0.99423	1.255	1.042	0.920	42.0833	42.0833	42.0488
0.0040	0.99740	0.99600	0.99435	1.269	1.060	0.939	42.1268	42.1152	42.0922
0.0055	0.99754	0.99613	0.99449	1.284	1.079	0.957	42.1587	42.1472	42.1242
0.0070	0.99768	0.99628	0.99464	1.299	1.098	0.976	42.1902	42.1787	42.1443
0.0085	0.99784	0.99644	0.99481	1.312	1.118	0.995	42.2103	42.2103	42.1758
0.005m IL									
0.0010	0.99718	0.99580	0.99416	1.280	1.098	0.987	42.1199	42.0739	42.0394
0.0025	0.99728	0.99590	0.99425	1.296	1.120	1.011	42.1523	42.1178	42.0833
0.0040	0.99741	0.99602	0.99437	1.313	1.143	1.035	42.1843	42.1613	42.1268
0.0055	0.99755	0.99616	0.99450	1.331	1.164	1.058	42.2162	42.1932	42.1587
0.0070	0.99770	0.99631	0.99466	1.349	1.187	1.083	42.2477	42.2247	42.1902
0.0085	0.99786	0.99648	0.99484	1.366	1.211	1.112	42.2677	42.2447	42.2218

Table 5: Apparent molar volume, (Φv) and $(\eta/\eta^0 - 1)/\sqrt{m}$ of L-phenylalanine solution in 0.001m, 0.003m and 0.005m aqueousBTBACl solution at different temperatures (298.15K, 303.15K, 308.15K).

Conc. of L-phenylalanine soln. in molality, m ($mol.kg^{-1}$)	Φv x 10^6 ($m^3.mol^{-1}$)	$(\eta/\mathring{\eta}-1)/\sqrt{m}$ ($kg^{1/2}.mol^{-1/2}$)	Φv x 10^6 ($m^3.mol^{-1}$)	$(\eta/\mathring{\eta}-1)/\sqrt{m}$ ($kg^{1/2}.mol^{-1/2}$)	Φvx 10^6 ($m^3.mol^{-1}$)	$(\eta/\mathring{\eta}-1)/\sqrt{m}$ ($kg^{1/2}.mol^{-1/2}$)
0.001m IL	**298.15K**		**303.15K**		**308.15K**	
0.0010	160.7207	0.209	161.9460	0.271	162.2257	0.357

0.0025	160.4951	0.320	161.5463	0.439	161.4320	0.543
0.0040	160.2416	0.388	161.1939	0.541	160.9812	0.703
0.0055	160.1123	0.458	160.8120	0.643	160.5903	0.852
0.0070	159.8966	0.508	160.5009	0.717	160.2174	0.944
0.0085	159.7639	0.543	160.2770	0.783	159.8597	1.044
0.003m IL	**298.15K**		**303.15K**		**308.15K**	
0.0010	161.6984	0.202	162.9276	0.278	163.2093	0.281
0.0025	160.9001	0.291	162.3305	0.432	161.8237	0.513
0.0040	160.4467	0.372	161.6813	0.562	161.2250	0.673
0.0055	160.0537	0.447	161.2005	0.666	160.5868	0.801
0.0070	159.6787	0.495	160.7767	0.755	160.0720	0.912
0.0085	159.3191	0.537	160.3854	0.835	159.5051	1.010
0.05mIL	**298.15K**		**303.15K**		**308.15K**	
0.0010	162.6863	0.171	163.9127	0.231	164.1912	0.289
0.0025	161.2925	0.298	162.6682	0.419	162.5251	0.553
0.0040	160.6902	0.386	161.9242	0.550	161.4670	0.729
0.0055	160.0484	0.467	161.1952	0.656	160.5814	0.870
0.0070	159.5307	0.527	160.6291	0.746	159.9247	0.993
0.0085	158.9606	0.581	160.1731	0.837	159.2313	1.090

Table 6: Apparent molar volume, (Φv) and (η/η^0 –1) /√m of L-tryptophan solution in 0.001m, 0.003m and 0.005m in aqueous(BTBACl) solution at different temperatures (298.15K, 303.15K, 308.15K).

Conc. of L-tryptophan solution in molality, m ($mol.kg^{-1}$)	Φv x 10^6 ($m^3.mol^{-1}$)	($\eta/\mathring{\eta}$-1)/√m ($kg^{1/2}.mol^{-1/2}$)	Φv x 10^6 ($m^3.mol^{-1}$)	($\eta/\mathring{\eta}$-1)/√m ($kg^{1/2}.mol^{-1/2}$)	Φvx 10^6 ($m^3.mol^{-1}$)	($\eta/\mathring{\eta}$-1)/√m ($kg^{1/2}.mol^{-1/2}$)
0.001m IL	**298.15K**		**303.15K**		**308.15K**	
0.0010	197.8788	0.267	199.2428	0.346	200.5091	0.451
0.0025	197.2718	0.416	198.4405	0.537	199.9086	0.718
0.0040	196.8595	0.523	197.9810	0.676	199.0068	0.884
0.0055	196.4790	0.573	197.3993	0.791	198.5866	1.046
0.0070	196.2482	0.654	196.9110	0.907	198.0505	1.185
0.0085	195.9803	0.717	196.5936	0.982	197.5847	1.325
0.003mIL	**298.15K**		**303.15K**		**308.15K**	
0.0010	198.8635	0.254	200.1524	0.401	201.4923	0.351
0.0025	198.0590	0.405	199.1531	0.612	200.2999	0.673
0.0040	197.3491	0.500	198.3968	0.765	199.2501	0.867
0.0055	196.8335	0.591	197.8615	0.906	198.4016	1.014
0.0070	196.5256	0.670	197.2579	1.028	197.7614	1.153
0.0085	196.0895	0.722	196.7479	1.146	197.1105	1.278
0.005m IL	**298.15K**		**303.15K**		**308.15K**	
0.0010	199.8505	0.295	201.1367	0.317	202.4734	0.417
0.0025	198.8485	0.440	199.5435	0.606	200.6890	0.759
0.0040	197.8410	0.559	198.6388	0.811	199.4913	0.987

0.0055	197.1903	0.669	197.8549	0.954	198.7562	1.161
0.0070	196.6622	0.761	197.2513	1.097	197.8971	1.337
0.0085	196.2010	0.837	196.6234	1.236	197.1039	1.533

Table 7(a): Refractive index (n_D) and specific conductance (κ) of L-phenylalanine in aqueous IL (BTBACl)solution at 298.15K, 303.15K and 308.15K.

Conc. of L-phenylalanine soln. in molality, m (mol.kg^{-1})	n_D			κx10^2(mS/m)		
	298.15K	303.15K	308.15K	298.15K	303.15K	308.15K
0.001mIL						
0.0010	1.3315	1.3313	1.3311	0.106	0.114	0.127
0.0025	1.3319	1.3318	1.3315	0.107	0.115	0.128
0.0040	1.3323	1.3321	1.3319	0.108	0.116	0.129
0.0055	1.3326	1.3324	1.3322	0.109	0.117	0.131
0.0070	1.3329	1.3327	1.3325	0.111	0.119	0.132
0.0085	1.3331	1.3329	1.3328	0.112	0.121	0.134
0.003mIL						
0.0010	1.3317	1.3316	1.3314	0.284	0.313	0.349
0.0025	1.3322	1.3320	1.3318	0.285	0.315	0.351
0.0040	1.3325	1.3323	1.3322	0.287	0.317	0.352
0.0055	1.3328	1.3326	1.3325	0.288	0.321	0.354
0.0070	1.3331	1.3329	1.3328	0.291	0.323	0.357
0.0085	1.3333	1.3331	1.3330	0.295	0.327	0.363
0.005mIL						
0.0010	1.3317	1.3315	1.3313	0.455	0.508	0.557
0.0025	1.3321	1.3319	1.3317	0.458	0.512	0.561
0.0040	1.3324	1.3322	1.3321	0.459	0.514	0.562
0.0055	1.3327	1.3325	1.3323	0.461	0.516	0.565
0.0070	1.3329	1.3327	1.3326	0.463	0.518	0.568
0.0085	1.3331	1.3329	1.3328	0.466	0.521	0.571

Table 7b: Refractive index (n_D) and specific conductance (κ) of L-tryptophan in aqueous IL (BTBACl)solution at 298.15K, 303.15K and 308.15K.

Conc. of L-tryptophan soln. in molality, m (mol.kg^{-1})	n_D			κx10^2(mS/m)		
	298.15K	303.15K	308.15K	298.15K	303.15K	308.15K
0.001mIL						
0.0010	1.3316	1.3315	1.3311	0.102	0.109	0.125
0.0025	1.3322	1.3318	1.3315	0.103	0.111	0.126
0.0040	1.3327	1.3322	1.3319	0.104	0.112	0.129
0.0055	1.3330	1.3325	1.3322	0.105	0.113	0.131
0.0070	1.3333	1.3328	1.3325	0.106	0.114	0.132
0.0085	1.3336	1.3332	1.3328	0.108	0.115	0.133

0.003mIL						
0.0010	1.3318	1.3317	1.3314	0.281	0.309	0.342
0.0025	1.3321	1.3321	1.3318	0.287	0.315	0.348
0.0040	1.3325	1.3324	1.3322	0.289	0.317	0.349
0.0055	1.3328	1.3327	1.3325	0.291	0.319	0.352
0.0070	1.3331	1.3330	1.3327	0.293	0.321	0.354
0.0085	1.3333	1.3333	1.3330	0.301	0.327	0.362
0.005mIL						
0.0010	1.3324	1.3320	1.3317	0.446	0.495	0.545
0.0025	1.3327	1.3324	1.3321	0.452	0.501	0.549
0.0040	1.3330	1.3328	1.3325	0.453	0.503	0.551
0.0055	1.3333	1.3331	1.3328	0.454	0.505	0.553
0.0070	1.3336	1.3334	1.3331	0.456	0.507	0.556
0.0085	1.3338	1.3336	1.3334	0.458	0.511	0.559

Table 8a: Limiting apparent molar volumes(Φv^0), Limiting molar refraction(R_M^0), experimental slopes (S_V^*), viscosity A, B-coefficients of L-phenylalanine solution in IL at different temperatures.

Temperature(K)	$\Phi v^0 \times 10^6$ (m^3. mol^{-1})	R_M^0	$Sv^* \times 10^6$ (m^3. $mol^{-3/2}.kg^{1/2}$)	B ($kg^{1/2}$. $mol^{-1/2}$)	A (kg. mol^{-1})
0.001m IL					
298.15	161.3	33.86	-5.038	5.5869	0.0358
303.15	162.9	33.89	-8.8956	8.4617	0.0073
308.15	163.4	33.92	-12.042	11.605	-0.0263
0.003m IL					
298.15	162.9	33.93	-12.110	5.7125	0.0131
303.15	164.4	33.92	-13.428	9.329	-0.0282
308.15	165.0	33.89	-18.607	12.062	-0.0984
0.005m IL					
298.15	164.4	33.94	-18.715	6.8537	-0.0478
303.15	165.8	33.92	-19.429	9.9749	-0.0858
308.15	166.6	33.89	-25.517	13.267	-0.1213

Table 8b: Limiting apparent molar volumes(Φv^0), Limiting molar refraction(R_M^0), experimental slopes (S_V^*), viscosity A, B-coefficients of L-tryptophan solution in IL at different temperatures.

Temperature (K)	$\Phi v^0 \times 10^6$ (m^3. mol^{-1})	R_M^0	$Sv^* \times 10^6$ (m^3. $mol^{-3/2}.kg^{1/2}$)	B ($kg^{1/2}$. $mol^{-1/2}$)	A (kg. mol^{-1})
0.001m IL					
298.15	198.8	41.91	-09.849	7.3128	0.0428
303.15	200.7	41.91	-13.896	10.628	0.0055
308.15	202.2	41.86	-15.494	14.299	-0.0079
0.003m IL					
298.15	200.3	41.95	-14.430	7.828	0.0073
303.15	201.9	41.94	-17.567	12.318	-0.003
308.15	203.9	41.91	-22.962	15.143	-0.1101

0.005m IL					
298.15	201.8	42.03	-19.332	9.0857	-0.0062
303.15	203.3	41.97	-23.064	15.017	-0.1539
308.15	205.1	41.93	-27.358	18.062	-0.1597

Table 8c: dB/dT of L-phenylalanine solutions in BTBACl at different temperatures.

Temperature (K)	0.001m IL B	0.003m IL B	0.005m IL B	dB/dT
298.15	5.5869	5.7125	6.8537	0.601
303.15	8.4617	9.329	9.9749	0.635
308.15	11.605	12.062	13.267	0.641

Table 8d: dB/dT of L-tryptophan solutions in BTBACl at different temperatures.

Temperature (K)	0.001m IL B	0.003m IL B	0.005m IL B	dB/dT
298.15	7.312	7.828	9.085	0.698
303.15	10.628	12.318	15.017	0.731
308.15	14.299	15.143	18.062	0.897

Table 9: The empirical coefficient values (a_0, a_1 and a_2) of L-phenylalanine solution & L-tryptophan in different concentration of the IL(0.001m,0.003m,0.005m) at 298.15K, 303.15K and 308.15K.

Conc. of aq. IL soln. in molality, m ($mol.kg^{-1}$)	$a_0 \times 10^6$ ($m^3.mol^{-1}$)	$a_1 \times 10^6$ ($m^3. mol^{-1}. K^{-1}$)	$a_2 \times 10^6$ ($m^3. mol^{-1}. K^{-2}$)	$a_0 \times 10^6$ ($m^3. mol^{-1}$)	$a_1 \times 10^6$ ($m^3. mol^{-1}. K^{-1}$)	$a_2 \times 10^6$ ($m^3. mol^{-1}. K^{-2}$)
	L-phenylalanine			L-tryptophan		
	298.15K	303.15K	308.15K	298.15K	303.15K	308.15K
0.001	-138.18	1.7501	-0.0025	-94.823	1.6111	-0.0021
0.003	59.19	0.4673	-0.0004	-755.55	5.9785	-0.0093
0.005	242.45	-0.7385	0.0016	-811.48	6.3798	-0.01

Table 10a: Values of limiting molar expansibilities (Φ_E^0) for L-phenylalanine solution in IL(BTBACl) at different temperatures.

Conc. of aq. IL soln. in molality, m ($mol.kg^{-1}$)	$\Phi_E^0 \times 10^6$ ($m^3. mol^{-1}. K^{-1}$)			$(\delta\Phi_E^0/\delta T)_P \times 10^6$ ($m^3. mol^{-1}. K^{-2}$)
	298.15K	303.15K	308.15K	
0.001	0.25935	0.23435	0.20935	-0.0050
0.003	0.22878	0.22478	0.22078	-0.0008
0.005	0.21558	0.23158	0.24785	0.0032

Table 10b: Values of limiting molar expansibilities (Φ_E^0) for L-tryptophan solution in IL (BTBACl) at different temperatures.

Conc. of aq. IL soln. in molality, m (mol.kg^{-1})	$\Phi_E^0 \times 10^6$ (m^3. mol^{-1}. K^{-1})			$(\delta\Phi_E^0/\delta T)_P \times 10^6$ (m^3. mol^{-1}. K^{-2})
	298.15K	303.15K	308.15K	
0.001	0.35887	0.33787	0.31687	-0.0042
0.003	0.43291	0.33991	0.24691	-0.0186
0.005	0.4168	0.3168	0.2168	-0.0200

Table 11: Molar conductance (Λ) of L-phenylalanine and L-tryptophan solution in (0.001m, 0.003m, 0.005m) IL at 298.15K, 303.15K and 308.15K.

Concentration of amino acid solutions in molality, m (mol.kg^{-1})	Molar Conductance of L-phenylalanine solution Λ x 10^{-4} (S.m^2.mol^{-1})			Molar conductance of L-tryptophan solution Λ x 10^{-4} (S.m^2.mol^{-1})		
	298.15K	303.15K	308.15K	298.15K	303.15K	308.15K
0.001m IL						
0.0010	105.68	113.65	126.62	101.69	108.67	124.62
0.0025	42.68	45.87	51.06	41.08	44.27	50.26
0.0040	26.91	28.90	32.15	25.92	27.90	32.15
0.0055	19.78	21.22	23.77	19.05	20.50	23.77
0.0070	15.80	16.93	18.79	15.09	16.22	18.79
0.0085	13.13	14.18	15.71	12.66	13.48	15.59
0.003m IL						
0.0010	283.15	312.06	347.95	280.15	308.07	340.97
0.0025	113.68	125.64	140.01	114.48	125.64	138.81
0.0040	71.52	78.99	87.71	72.02	78.99	86.97
0.0055	52.25	58.23	64.22	52.79	57.87	63.86
0.0070	41.42	45.97	50.82	41.71	45.69	50.39
0.0085	34.58	38.33	42.55	5.28	38.33	42.43
0.005mIL						
0.0010	453.63	506.48	555.33	444.66	493.51	543.36
0.0025	182.69	204.22	223.77	180.30	199.84	218.99
0.0040	114.38	128.08	140.04	112.88	125.34	137.30
0.0055	83.64	93.61	102.50	82.37	91.61	100.33
0.0070	65.91	73.73	80.85	64.91	72.17	79.15
0.0085	54.62	61.07	66.93	53.69	59.89	65.53

Table 12a: Surface tension values of L-Phenylalanine and L-Tryptophan.

Concentration of amino acid solutions in molality, m (mol.kg^{-1})	L-phenylalanine			L-Tryptophan		
	0.001m IL	0.003m IL	0.005m IL	0.001m IL	0.003m IL	0.005m IL
0.0010	63.5	65.7	68.3	75.1	73.9	72.7
0.0025	62.2	64.3	67.6	72.3	70.3	68.8
0.0040	61.9	63.6	66.1	70.9	68.8	67.2
0.0055	61.3	63	65.1	69.2	66.0	64.3
0.0070	60.7	62.9	64.1	66.9	64.9	61.8
0.0085	60.0	61.1	62.3	63.5	60.6	57.9

Table 12b: Limiting Slopes (∂σ/∂m) of the Surface Tension of the Aqueous Solutions of Amino Acids.

Aqueous IL Mixture/ mol. kg⁻¹	(∂σ/∂m) / mN m⁻¹.kg. mol⁻¹	
	L- phenylalanine + Aq. BTBACl	L-tryptophan + Aq. BTBACl
0.001	-43.048	-144.57
0.003	-52.952	-162.86
0.005	-79.048	-186.48

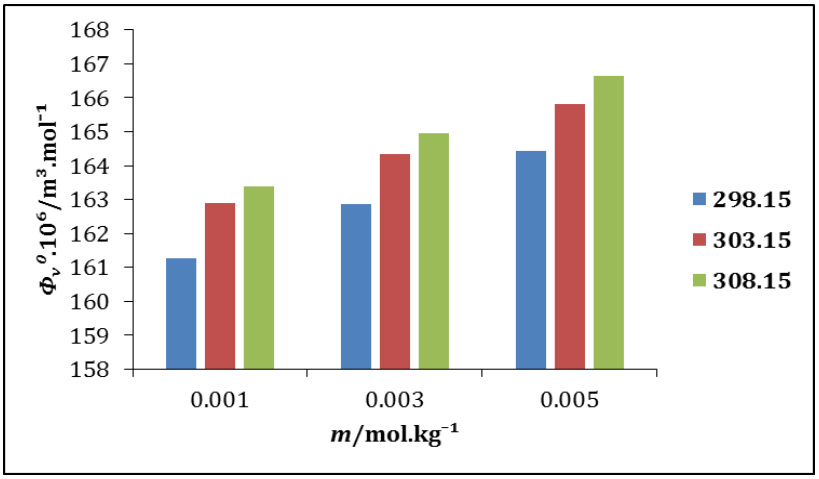


Figure 1a: Variation of $\Phi_v{}^o$ of L-Phenylalanine inaqueousBTBACl with different concentration at 298.15K, 303.15K, 308.15K

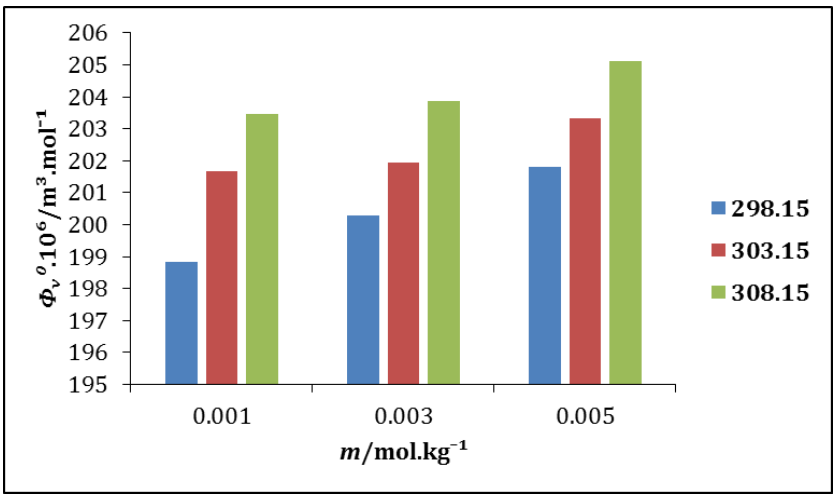


Figure 1b: Variation of $\Phi_v{}^o$ of L-Tryptophan inaqueousBTBACl with different concentration at 298.15K, 303.15K, 308.15K.

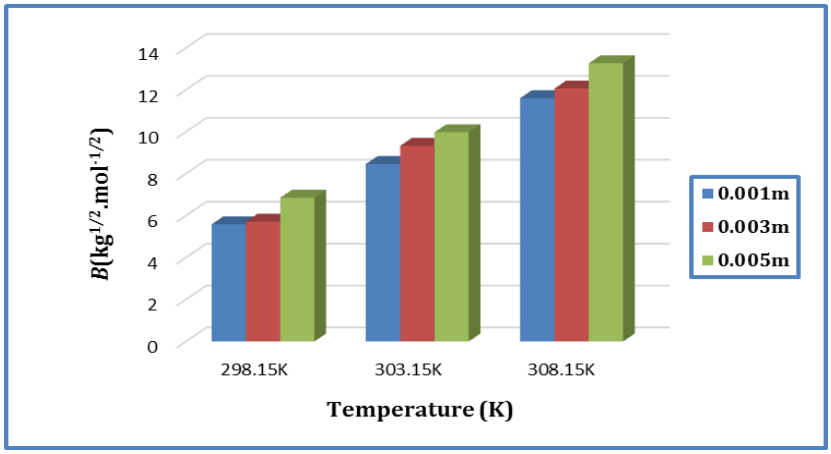


Figure 2a: Variation of *B* values of L-Phenylalanine at 298.15K, 303.15K and 308.15K for 0.001 m IL, 0.003 m IL and 0.005 m IL solutions.

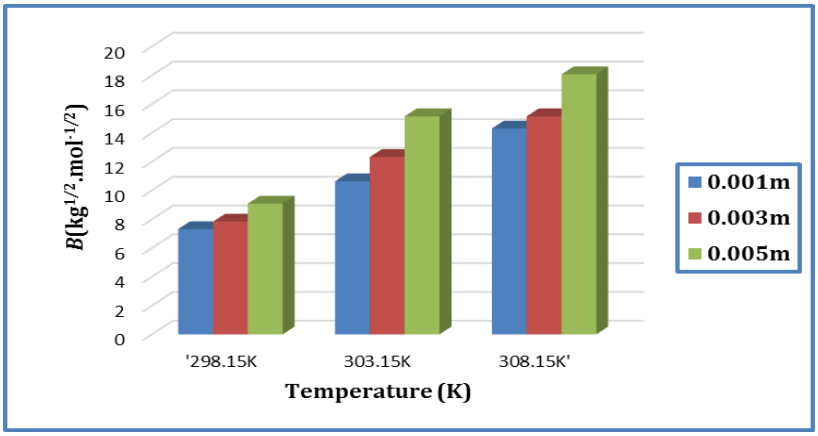


Figure 2b: Variation of *B* values of L-Trytophan at 298.15K, 303.15K and 308.15K for 0.001 m IL, 0.003 m IL and 0.005 m IL solutions.

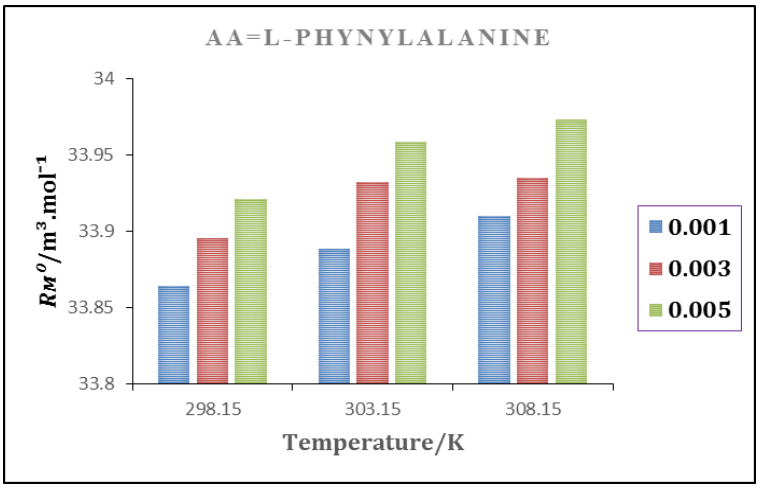


Figure 3a: Variation of limiting molar refraction(R_M^0) of L-Phenylalanine with temperature as a function of concentration of aq. BTBACl solutions.

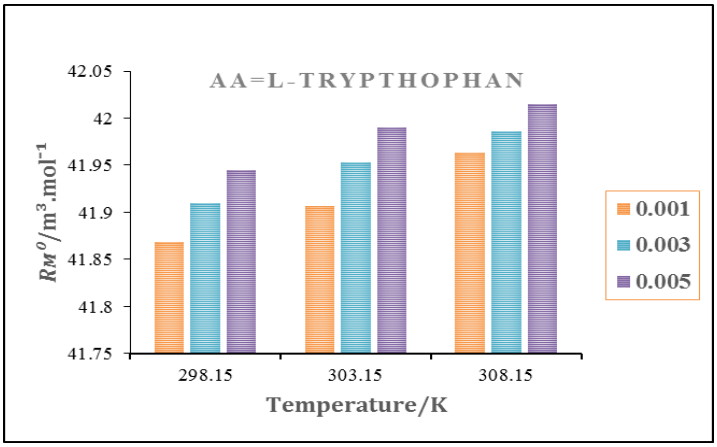


Figure 3b: Variation of limiting molar refraction($R_M{}^0$) of L-Tryptophan with temperature as a function of concentration of aq. BTBACl solutions.

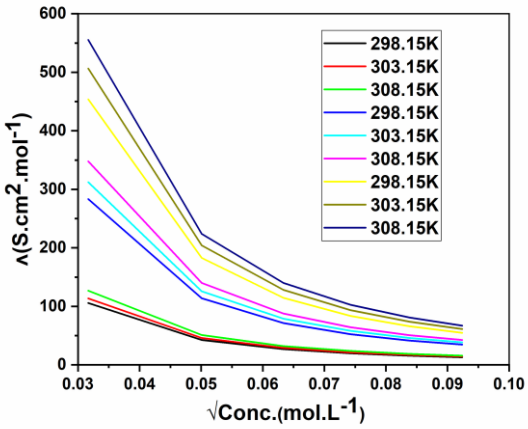


Figure 4a: Variation of molar conductance (*Λ*) against concentration of L-Phenylalanine in (0.001m, 0.003m, 0.005m) aq. BTBACl solutions at 298.15K, 303.15K, 308.15K.

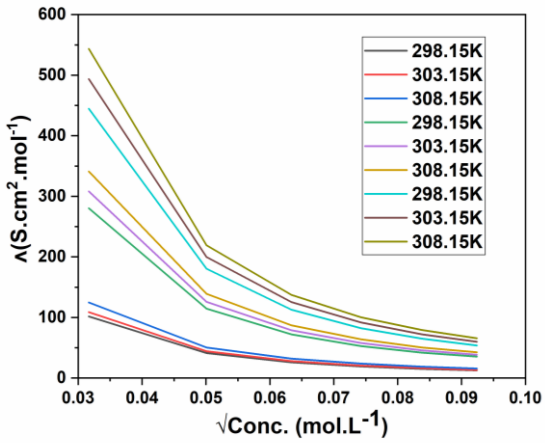


Figure 4b: Variation of molar conductance (*Λ*) against concentration of L-Trytophan in (0.001m, 0.003m, 0.005m) aq. BTBACl solutions at 298.15K, 303.15K, 308.15K.

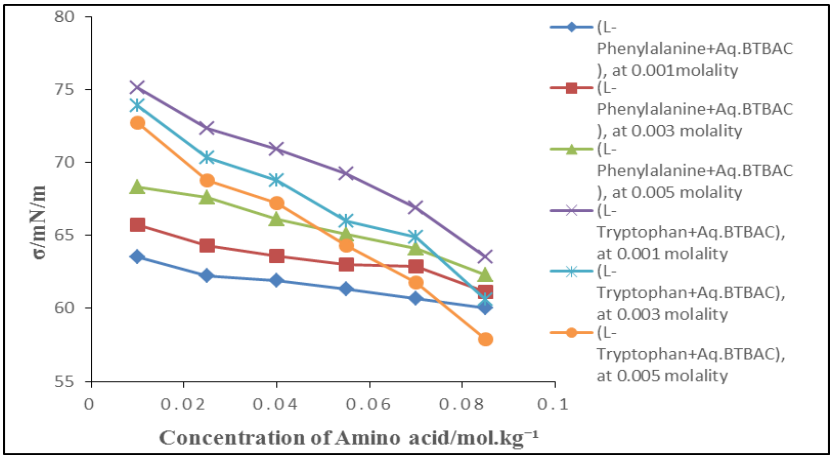


Figure 5: Plot of Surface Tension(σ) of L-phenylalanine and L-tryptophan as a function of different molality of aqueous BTBACl ionic liquid solutions.

CONCLUSION

The studied system of Ionic Liquid, Benzyltributylammonium chloride and Amino Acids, L-Phenylalanine and L-Tryptophan exhibit strong solute-solvent interactions with a rise in temperature and increase in concentration of the IL which is weakened with an increase in the amount of the amino acids in solutions, suggesting larger electrostriction at higher temperature with greater amount of IL but lesser amount of amino acids in the solution mixture. There is presence of weak solute-solute interactions which decrease with a rise in temperature and increase in concentration of IL. Moreover, the structure-making effect is strongest for L-phenylalanine in 0.005m IL solution whereas L-tryptophan acts as a structure-breaker in theIL solvent mixtures and the effect being maximum at 0.005m IL concentration solution.

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