



Thermodynamic Properties of Electrolytes Solutions in Aqueous Serine and Valine at Different Temperatures

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ABSTRACT

Ultrasonic velocities (u), densities (ρ) and viscosities (η) have been measured for the binary liquid systems (1M serine + 1M NaCl) and (1M valine + 1M MgCl₂). The ultrasonic velocity measurements have been carried out by ultrasonic interferometer technique at 2 MHz frequency and at different temperatures of 303.15, 308.15 and 313.15K. Using these experimental data, derived thermodynamic parameters such as adiabatic compressibility (β_a), intermolecular free length (L_f), acoustic impedance (Z), and relative association (R_A) have been computed using standard formulae. The results have been interpreted on the basis of variations in thermodynamic parameters. The variations in ultrasonic velocity and adiabatic compressibility with concentrations in both the systems show similar trends of increasing ultrasonic velocity and decreasing adiabatic compressibility of constituent electrolytes at different temperatures. This is due to complex formation and co-ordinate covalent bond form between the molecules of liquid solutions. The thermodynamic parameters were highly useful in elucidating solute – solvent interactions in aqueous solutions and binary mixtures.

Keywords: Ultrasonic Velocity(u), Adiabatic Compressibility(β_a), Intermolecular Free Length(L_f), Acoustic Impedance(Z), Relative Association (R_A).

INTRODUCTION

The study of molecular interactions in the liquid mixtures is very much important to study the structural properties of molecules. Ultrasonic energy is used in medicine, engineering, agriculture, defense and industry. In chemical industries ultrasonic energy is found useful in studying the chemical processes and in synthesis of chemical substances. In solution of ionic solute the attraction between the solute and solvent is of ion-dipole type. When electrolyte is dissolved in solvent it causes volume contraction due to interactions between ions and solvent molecules and therefore other acoustical parameters may be affected. Many researchers have used ultrasound to investigate the ion-solvent interactions in aqueous solutions containing electrolytes.

Ultrasonic study and transport properties of electrolytes in aqueous solutions of amino acids [1] are very useful to obtain information about various types of interactions in solutions. Consequently, the characterization of these interactions can assist in understanding the thermodynamic stability of proteins

and their unfolding behavior. Also the interactions of electrolytes in aqueous solution of amino acids and temperature dependence of these interactions play a vital role in understanding the nature of action of bioactive molecules or the thermodynamic behavior of biochemical process in the body system.

Ultrasonic velocity and its related properties have been extensively used to study the physico-chemical behavior and molecular interaction in a variety of liquid mixture [2]. The choice of water for preparing mixed solvent stems from its important and unique role in determining the structure and stability of proteins. Since its presence is known to give rise to hydrophobic forces, which are of prime importance in stabilizing the native globular structure of proteins. Proteins [3] are found in all parts of the body and they have an enormous variety of functions. Amino acids and peptides are used as probe molecules [3-5] to understand the complex nature of protein. It is interesting to study the basic primary structure builders, the amino acids in proteins in aqueous solution. There is information on the zwitter - ionic nature of amino acids in water in the literature [4-7]. The properties of proteins such as their structure, solubility, denaturation activity of enzymes etc. are greatly influenced by electrolytes [8-11].

An electrolyte when dissolved in water, changes the arrangement of water molecules with a strong electric field of its ions. This property of electrolytes is known as structure maker or structure breaker has been widely used to understand the effect of electrolytes on the structure and function of both proteins and nucleic acids [9, 10, 12]. Thermodynamic properties of electrolytes in aqueous amino acid solutions have been studied in order to understand the complex nature of proteins using amino acid-ion interactions [11-13]. There are instances where high concentrations of electrolytes can affect the function and structure of proteins [14-20]. Ion – ion and ion- amino acid interactions dominate in such situations. How the ion-ion and ion-amino acid interactions together with ion –water and amino acid- water ones are altered in electrolytes and amino acid solutions is the object of current investigations. No systematic efforts have been made to study the ion-ion and ion – amino acid interactions in amino acid solutions from thermodynamic properties. Therefore, this investigation presents ultrasonic velocity (u), density (ρ), and viscosity (η) data for systems comprising NaCl, MgCl₂ in aqueous serine & valine. Even though the ultrasonic velocity data as such do not provide significant information about the native and relative strength of various types of intermolecular inter-ionic interactions between the components, their derived parameters such as adiabatic compressibility (β_a), acoustic impedance (Z), intermolecular free length (L_f), and relative association (R_A) throw some light on such interactions.

MATERIALS AND METHODS

All the chemicals used were of AR grade and dried over anhydrous CaCl₂ in desiccators' before use. All solutions were prepared in deionized and distilled water (degassed by boiling), having specific conductivity 10^{-6} S cm⁻¹. The stock solutions [13] of 1M concentration were prepared by weighing serine and valine, NaCl and MgCl₂ on a digital balance with an accuracy of 1×10^{-5} g. Solutions of NaCl & MgCl₂ were prepared by mass on the molality concentration scale. Uncertainties in solution concentrations were estimated at 1×10^{-5} mol.kg⁻¹ in calculations. The mixture of 1M molal solutions were prepared by volume fraction concentration scale. Solutions were kept in the special air tight bottles and were used within 12 hrs. After preparation to minimize decomposition due to bacterial contamination. Ultrasonic velocity was measured with a single crystal interferometer (F- 81, Mittal Enterprises, NewDelhi) at 2 MHz. The source of ultrasonic waves was a quartz crystal excited by a radio frequency oscillator placed at bottom of a double jacketed metallic cylindrical container. The cell was filled with the desired solution and in the outer jacket constant temperature water was circulated. The cell was allowed equilibrate for 30 minutes prior to making the measurements. The interferometer was calibrated against the ultrasonic velocity of water used at $T = 303.15$ K. The present experimental value is 1508.80 ms⁻¹ which is in good agreement with literature value 1509.55 ms⁻¹. Accuracy in the velocity measurement was 1.0 ms⁻¹. The densities of the mixtures were determined accurately using 25 ml specific gravity bottle and electronic balance. The accuracy of electronic balance is ± 0.1 mg. An average of triple measurements was taken into account. Sufficient care was taken to avoid any air bubble entrapment. Viscosity was measured with

precalibrated Ostwald type viscometer. The flow time was measured with a digital stop watch capable of registering time accurate to 0.1 s. An average of three or four sets of flow of times for each solution was taken for the purpose of calculation of viscosity. The accuracy of the viscosity measurements was 0.5 %. Accuracy in experimental temperature was maintained at 0.1K by means of thermostatic water bath.

RESULTS AND DISCUSSION

Ultrasonic velocity, density and viscosity of the liquid systems have been measured. Using these experimental data, the thermodynamic parameters such as the adiabatic compressibility (β_a), intermolecular free length (L_f), acoustic impedance (Z) and relative association (R_A) were investigated for six different vol. fractions of 1M NaCl & 1M MgCl₂ at frequency 2 MHz and temperatures 303.15, 308.15 and 313.15 K. From the experimental data of ultrasonic velocity (u), density (ρ), and viscosity (η), the thermodynamic parameters have been calculated by using the following relations [21 - 25].

$$\text{Ultrasonic velocity } u = n \times \lambda \quad \dots\dots\dots (1)$$

$$\text{Adiabatic compressibility } \beta_a = 1/u^2 \rho \quad \dots\dots\dots (2)$$

$$\text{Intermolecular free length } L_f = K / u \cdot \rho \quad \dots\dots\dots (3)$$

$$\text{Acoustic impedance } Z = u \rho \quad \dots\dots\dots (4)$$

$$\text{Relative association } R_A = (\rho / \rho_0) \cdot (u_0 / u)^{1/3} \quad \dots\dots\dots (5)$$

Where, K is the temperature dependant Jacobson constant, T is the absolute temperature, ρ_0 , ρ and u_0 , u are the density and ultrasonic velocity of solvent and solution respectively. For the amino acids - electrolytes liquid systems (1M serine + 1M NaCl) and (1M valine + 1M MgCl₂), ultrasonic velocity (u), density (ρ) and the coefficient of viscosity (η) for various volume fractions have been measured at constant frequency of 2 MHz and at different temperatures. The experimental values of u , ρ , η , are given in Tables - 1 and 2. The data obtained are used to evaluate β_a , Z , L_f , and R_A which is included in the same Tables. The graph plotted of thermodynamic parameters versus vol. fraction(x) for two liquid systems are as shown in Fig.1-10.

Table-1: Variation of thermodynamic parameters at different volume fractions (x) and different temperatures for the system (1M NaCl+1M Serine) at 2MHz.

303.15K							
Vol. Fra(x)	u ms ⁻¹	ρ kg m ⁻³	η Nm ⁻² s	β _a x10 ⁻¹⁰ m ² N ⁻¹	z x10 ⁶ Nm ⁻²	L _f A ⁰	R _A
0.0	1554.50	1056	1.0428	3.9188	1.64155	0.41088	1.05025
0.2	1561.70	1058	1.0507	3.8754	1.65228	0.40869	1.05062
0.4	1568.00	1060	1.0578	3.8371	1.66208	0.40657	1.05120
0.6	1570.04	1062	1.0605	3.8199	1.66738	0.40566	1.05272
0.8	1575.10	1064	1.0710	3.7883	1.67590	0.40398	1.05358
1.0	1586.08	1066	1.0810	3.7220	1.69076	0.40080	1.05312
308.15K							
Vol. Fra(x)	u ms ⁻¹	ρ kg m ⁻³	η Nm ⁻² s	β _a x10 ⁻¹⁰ m ² N ⁻¹	z x10 ⁶ Nm ⁻²	L _f A ⁰	R _A
0.0	1568.00	1054	0.9478	3.5899	1.65267	0.41141	1.04988
0.2	1571.70	1055	0.9561	3.8371	1.65814	0.41025	1.05005
0.4	1574.00	1058	0.9541	3.8151	1.66529	0.40907	1.05252
0.6	1579.08	1060	0.9591	3.7834	1.67382	0.40737	1.05338
0.8	1581.00	1062	0.9853	3.7671	1.67902	0.40649	1.05494
1.0	1588.07	1064	0.9911	3.7267	1.68970	0.40443	1.05536

313.15K							
Vol. Fra(x)	u ms ⁻¹	ρ kg m ⁻³	η Nm ⁻² s	β _a x10 ⁻¹⁰ m ² N ⁻¹	z x10 ⁶ Nm ⁻²	L _f A ⁰	R _A
0.0	1572.88	1052	0.9427	3.8423	1.67297	0.41360	1.04934
0.2	1578.04	1053	0.9640	3.8136	1.70748	0.41205	1.04919
0.4	1582.07	1055	0.9681	3.7870	1.74472	0.41061	1.05029
0.6	1584.88	1058	0.9802	3.7629	1.77020	0.40930	1.05266
0.8	1586.53	1060	0.9825	3.7480	1.80087	0.40849	1.05428
1.0	1588.07	1063	0.9909	3.7302	1.84258	0.40752	1.05692

Where u, ultrasonic velocity; ρ, density of the solution ; η, viscosity of solution; β_a, adiabatic compressibility; L_f, intermolecular free length; R_A, relative association; z, acoustic impedance.

Table-2: Variation of thermodynamic parameters at different volume fraction (x) and different temperatures for the system (1M MgCl₂ +1M Valine) at 2MHz.

303.15K							
Vol. Fra(x)	u ms ⁻¹	ρ kg m ⁻³	η Nm ⁻² s	β _a x10 ⁻¹⁰ m ² N ⁻¹	z x10 ⁶ Nm ⁻²	L _f A ⁰	R _A
0.0	1558.80	1056	1.0628	3.8972	1.64609	0.40975	1.04928
0.2	1566.77	1063	1.0871	3.8323	1.66548	0.40632	1.05654
0.4	1586.08	1070	1.1492	3.7151	1.69710	0.40006	1.05707
0.6	1501.10	1080	1.1541	3.6119	1.72919	0.39447	1.06360
0.8	1508.20	1088	1.2688	3.5538	1.74972	0.39128	1.06360
1.0	1612.17	1096	1.3170	3.5105	1.76694	0.38889	1.07689
308.15K							
Vol. Fra(x)	u ms ⁻¹	ρ kg m ⁻³	η Nm ⁻² s	β _a x10 ⁻¹⁰ m ² N ⁻¹	z x10 ⁶ Nm ⁻²	L _f A ⁰	R _A
0.0	157820	1054	0.9871	3.8092	1.66342	0.40875	1.04761
0.2	1584.10	1062	1.0421	3.7524	1.68231	0.40569	1.05425
0.4	1591.00	1075	1.0732	3.6749	1.71032	0.40148	1.06561
0.6	1606.17	1086	1.1282	3.5693	1.74430	0.39597	1.07312
0.8	1612.80	1092	1.1780	3.5206	1.76118	0.39269	1.07757
1.0	1620.08	1097	0.2931	3.4731	1.77723	0.39030	1.08088
313.15K							
Vol. Fra(x)	u ms ⁻¹	ρ kg m ⁻³	η Nm ⁻² s	β _a x10 ⁻¹⁰ m ² N ⁻¹	z x10 ⁶ Nm ⁻²	L _f A ⁰	R _A
0.0	1584.06	1052	0.8812	3.7883	1.66643	0.41068	1.04689
0.2	1591.40	1060	0.9278	3.7251	1.68688	0.40724	1.05320
0.4	1594.00	1066	0.9632	3.6920	1.69620	0.40543	1.05859
0.6	1608.00	1076	1.0078	3.5943	1.73021	0.40003	1.06541
0.8	1618.18	1084	1.1111	3.5230	1.75411	0.39604	1.07108
1.0	1624.10	1094	1.1838	3.4654	1.77677	0.39280	1.07965

Where u, ultrasonic velocity; ρ, density of the solution ; η, viscosity of solution; β_a, adiabatic compressibility; L_f, intermolecular free length; R_A, relative association; z, acoustic impedance.

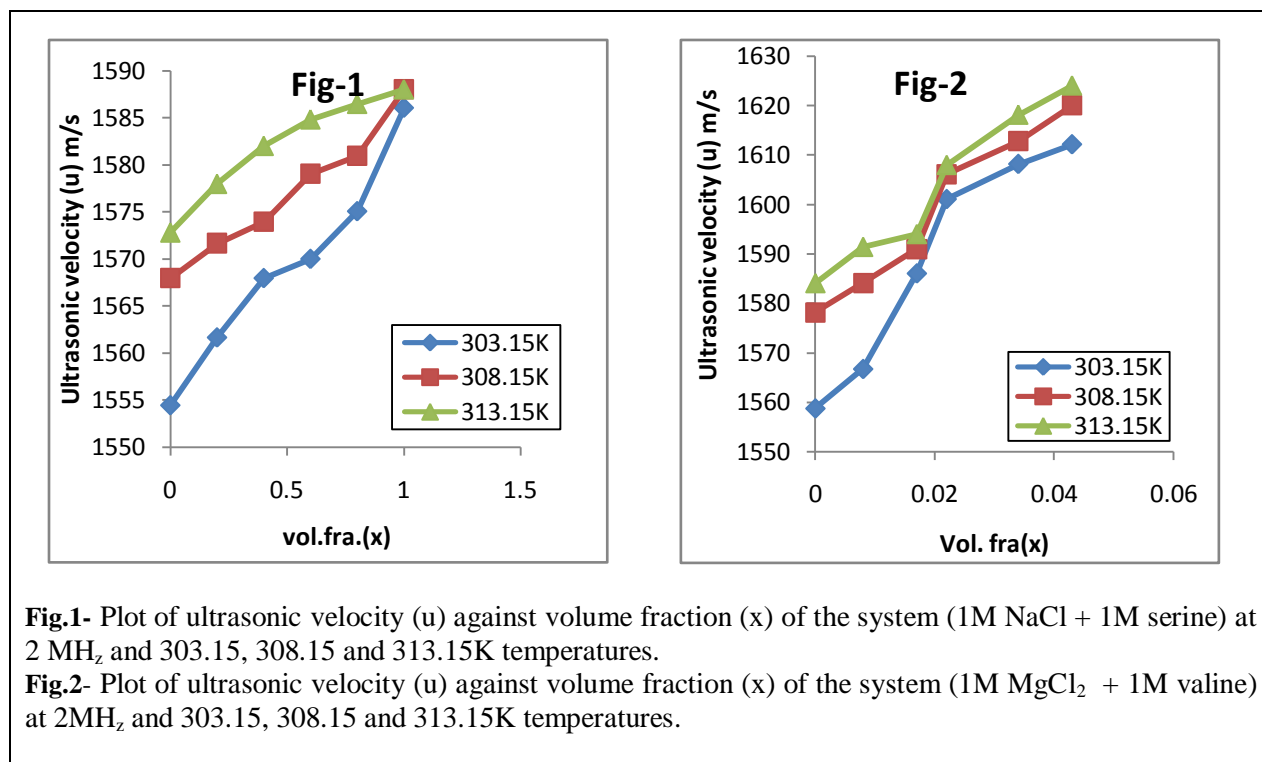
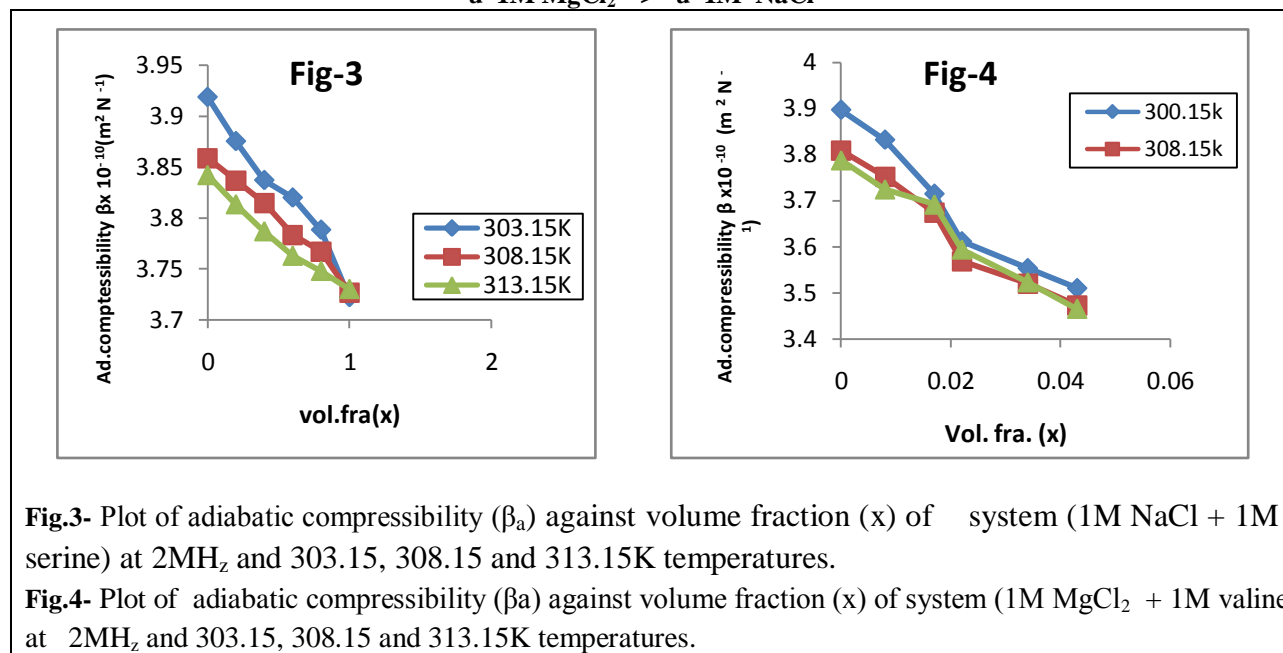


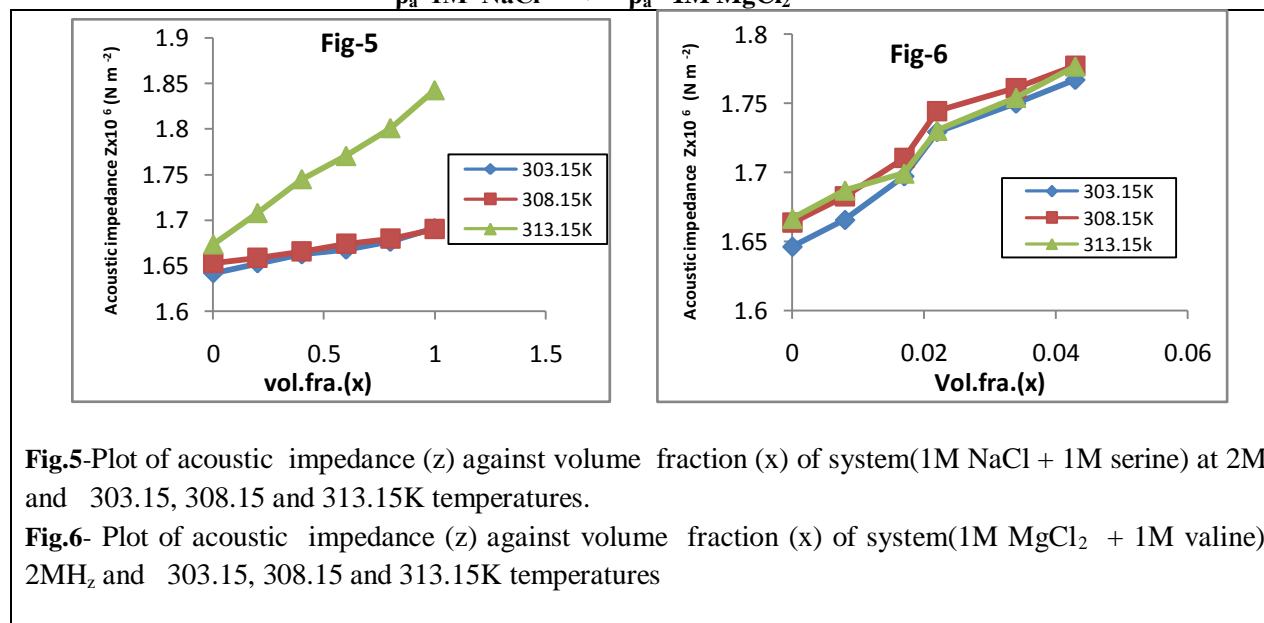
Fig.1- Plot of ultrasonic velocity (u) against volume fraction (x) of the system (1M NaCl + 1M serine) at 2 MHz and 303.15, 308.15 and 313.15K temperatures.

Fig.2- Plot of ultrasonic velocity (u) against volume fraction (x) of the system (1M MgCl₂ + 1M valine) at 2MHz, and 303.15, 308.15 and 313.15K temperatures.

Ultrasonic Velocity (u): The ultrasonic velocity (u) for amino acid electrolytes solutions at 2MHz frequency and at different temperatures have been determined using relation (1) and presented in Tables - 1, 2. From Figures - 1, 2, the variations in ultrasonic velocity in liquid mixtures depend on concentrations (x) of solutes and temperatures. Ultrasonic velocity (u) is related to intermolecular free length. As the free length decreases due to the increase in concentrations of solutes, the ultrasonic velocity has to increase. The experimental results support the above statement in both liquid systems. Consequently, ultrasonic velocity of system increases depending on the structural properties of solutes. The solute that increases the ultrasonic velocity is structure maker. From Tables - 1, 2, ultrasonic velocity increases with increase in concentrations of solutes (1M NaCl, 1M MgCl₂) in liquid systems investigated such as (1M serine+1M NaCl) and (1M valine +1M MgCl₂). The value of ultrasonic velocity of (1M serine + 1M NaCl) is less as compared to the value of ultrasonic velocity in (1M valine +1M MgCl₂). When NaCl or MgCl₂ is dissolved in solution, the sodium ion (Na⁺) or (Mg⁺⁺) has a structure breaking effect, would disrupt the water structure[1, 3]. This makes the liquid medium less compressible and hence the ultrasonic velocity increases above that of pure value.

$$\mu \text{ 1M MgCl}_2 > \mu \text{ 1M NaCl}$$


Adiabatic compressibility (β_a): When an aqueous electrolytes solution is added to a 1M serine & valine (solvent), it attracts certain solvent molecules towards itself by wrenching the molecules from bulk of the solvent due to the forces of electrostriction. Due to this the available solvent molecule for the next incoming ion gets decreased. From Figures .3 and 4, the compressibility of a solvent is higher than that of a solution and it decreases with increase in concentrations. The adiabatic compressibility is calculated using equation (2). The calculated values of (a) have been presented in Tables 1 and 2. The presence of hydrophobic hydration in due to presence of methyl group causes serine & valine to be under a higher electrostriction effect (Methyl group tightens the water molecules around itself). Hence the values of adiabatic compressibility for valine decreases. The relation between adiabatic compressibility, for two systems is as follows,

$$\beta_a \text{ 1M NaCl} > \beta_a \text{ 1M MgCl}_2$$


Acoustic impedance (z): Acoustic impedance (z) is found to be almost inversely to the adiabatic compressibility (α). Specific acoustic impedance is calculated by using standard relation (3). The calculated values of z are mention in Tables 1, 2, From Figures –5, 6; it is observed that acoustic impedance (z) increases for different vol. fractions. Acoustic impedance becomes either maximum or minimum depending on the concentrations and different temperatures. This is the stage where complex formation is taking place in the liquid system due to increased electrolytes – amino acids interaction. For a given concentration the values of acoustic impedance (z) increases with increase in concentration liquid systems (1M serine + 1M NaCl) and (1M valine+ 1M MgCl₂). It is in good agreement with the theoretical requirements because ultrasonic velocity increases with increase in concentrations of solutes in liquid mixtures. The increase in (z) with the increase in concentrations of solutes can be explained in terms of inter and intra molecular interactions between the molecules [2, 3] of liquid mixtures. This indicates significant interactions in the liquid systems. Similar behaviour is also observed in amino acid based liquid systems. From Tables 1 & Table-2 it is seen that values of acoustic impedance (z) are maximum in 1M MgCl₂ compared to that in 1M NaCl.

$$z \text{ 1M MgCl}_2 > z \text{ 1M NaCl}$$

Intermolecular free length (L_f): The values of intermolecular free length for (1M serine + 1M NaCl) and (1M valine + 1M MgCl₂) systems have been calculated using equation (4). Increase in concentrations leads to decrease in gap between two species which is referred by intermolecular free length (L_f). With the increase in concentrations of solutes, intermolecular free length (L_f) has to decrease. Intermolecular free length (L_f) is a predominant factor in determining the variations of ultrasonic velocity in liquid mixtures. From Figures 7, 8 and Tables 1, 2, it has been observed that, in the present investigation, intermolecular free length decreases linearly on increasing vol. fractions. The decrease in L_f with increase of vol. fractions in solution indicates that there are significant interactions between solute and solvent suggesting the structure promoting behaviour of solutes. Ultrasonic velocity increases with vol. fractions of solutes indicates stronger the intermolecular forces in the solution. This gives increase in closed packed structure of aqueous amino acids, i.e. enhancement of the closed structure. This provides the cohesion between amino acids and water molecules increases. The reduction in degree of dissociation among the liquid molecules of the mixture. Thus, the inter molecular distance decreases with concentration. The decrease in free length may due to the gain of dipolar association, making up of hydrogen bonds in the molecules of the liquid mixtures. Similar behavior is observed in (1M MgCl₂ +1M valine). The variation in intermolecular free length in both systems with temperature shows following trend.

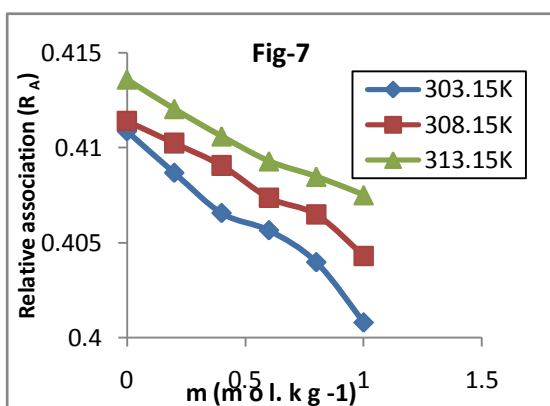


Fig.7- Plot of intermolecular free length (L_f) against volume fraction (x) of the system (1M NaCl + 1M serine) at 2MHz and 303.15, 308.15 and 313.15K temperatures.

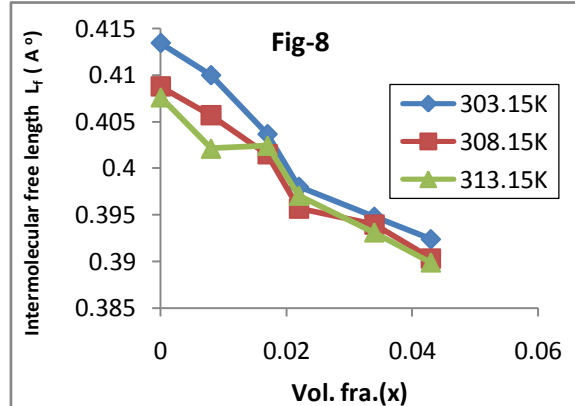


Fig.8- Plot of intermolecular free length (L_f) against volume fraction (x) of the system (1M MgCl₂ +1M valine) at 2MHz and 303.15, 308.15 and 313.15K temperatures.

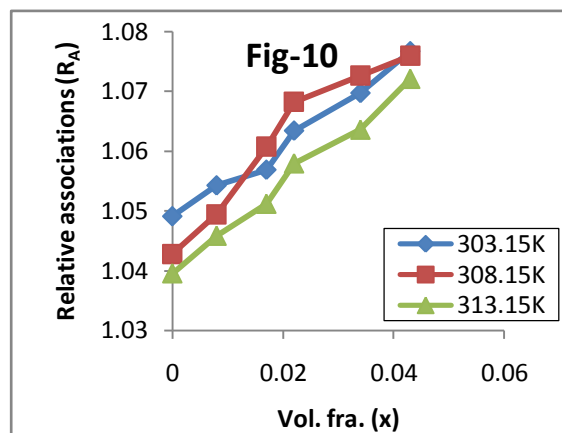
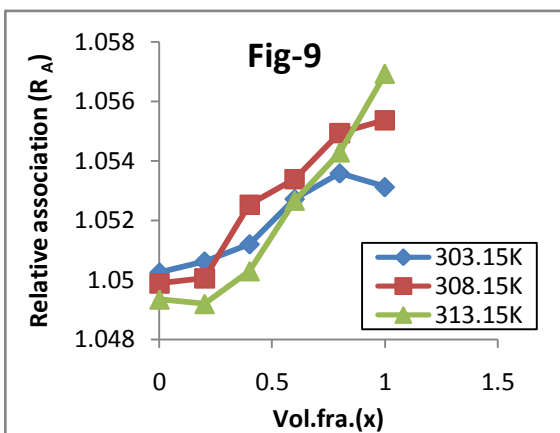


Fig.9- Plot of relative association (R_A) against volume fraction (x) of the system (1M NaCl + 1M serine) at 2MHz and 303.15, 308.15 and 313.15K temperatures.

Fig.10- Plot of relative associations (R_A) against volume fraction (x) of the system (1M MgCl₂ + 1M valine) at 2MHz and 303.15, 308.15 and 313.15K temperatures.

Relative association (R_A): The values of relative association (R_A) for liquid systems (1M serine + 1M NaCl) and (1M valine + 1M MgCl₂) have been estimated using relation (5). The property which can be studied to understand the interaction is the relative association (R_A). It is influenced by two factors: (i) Breaking up of the associated solvent molecules on addition of solute in it and (ii) The salvation of solute molecule. The former leads to the decrease and later to the increase of relative association. From Figures – 9, 10 and Tables 1, 2, it is observed that, R_A increases with increase in the vol. fractions (x). The values of 1M MgCl₂ is more as compared to 1M NaCl. The variation relative association (R_A) in both systems with temperature shows following trend.

$$R_{A \text{ 1M MgCl}_2} > R_{A \text{ 1M NaCl}}$$

APPLICATIONS

The thermodynamic properties of a binary mixture such as viscosity and density are important for practical and theoretical points of view to understand liquid theory. Their properties are extremely useful for the design of process equipment in chemical industries and the data on some of the properties associated with the liquids and liquid mixtures like density and viscosity find extensive application in solution theory and molecular dynamics.

CONCLUSIONS

Ultrasonic velocity, density and viscosity have been measured for 1M NaCl in aqueous serine and 1M MgCl₂ in aqueous valine solution at 303.15, 308.15 and 313.15K. The variation in ultrasonic velocity (u), density (ρ) and viscosity (η) and other related thermodynamic parameters such as adiabatic compressibility (β_a), acoustic impedance (z), intermolecular free length (L_f) and relative association (R_A) of 1M NaCl & 1M MgCl₂ at various concentrations and temperatures in both the amino acid – based systems, shows the non-linear increase or decrease behavior. The non linearity confirms the presence of solute-solvent, ion-ion, dipole-dipole, ion-solvent interactions. The observed molecular interaction, complex formation, hydrogen bond formation are responsible for the hetero-molecular interaction in the liquid mixture. This provides useful information about inter and intra molecular interactions of liquid systems. It is also concluded that ultrasonic velocity of system increases depending on the structural properties of solutes. It is well known that solutes causing electrostriction lead to decrease in the compressibility of the solution. Hydrophilic solutes often show negative compressibility, due to ordering

that is induced by them in water structure. The solute that increases the ultrasonic velocity is of structure maker (SM). It has been observed that intermolecular free length decreases linearly on increasing concentrations of solutes in the systems. The variation in ultrasonic velocity, density and viscosity as well as the related thermodynamic parameters such as adiabatic compressibility (β_a), acoustic impedance (z), intermolecular free length (L_f), relative association (R_A), are more in the system containing $MgCl_2$ as compared to the system having $NaCl$ due to the divalent cation (Mg^{2+}) of $MgCl_2$. As a matter of fact, the effect of $MgCl_2$ on the electrostriction of water molecules is stronger than that of 1:1 electrolytes i.e. $NaCl$. Thus the addition of $MgCl_2$ to valine enhances the transfer volumes more strongly than mono-valent cation Na^+ .

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