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Ultrasonic, Volumetric and Viscometric study of Molecular Interaction in Aqueous NaCl Solution

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Abstract: ***Ultrasonic velocity (U), density (ρ) and viscosity (η) have been measured in aqueous NaCl solution at 2MHz frequency and 297 K temperature. These parameters are found to increase with concentration. These experimentally measured data have been used to calculate thermo-acoustic parameters such as free length (L_f), adiabatic compressibility (β), acoustic impedance (z), Rao's constant or molar velocity(R_m), Relaxation time (τ), Gibbs free energy (G), internal pressure (π), free volume (V_f), Molar volume (V_m), Salvation Number (S_n) and Surface tension (S). Our study revels strong ion-dipole interaction between water and ions in the electrolyte solution. This is confirmed by calculating the apparent molar volume of solution and the Jones-Dole coefficient A, B and D.***

Keywords: Adiabatic compressibility; Acoustic impedance; Apparent molar volume; Jones-Dole coefficient; Thermo-acoustic parameter

I. INTRODUTION

In recent years ultrasonic technique has been found to be very effective in studying the properties of any substance, to understand the nature of molecular interaction in pure liquid [1], liquid mixtures [2, 3] and ionic interaction in electrolytic solution [4, 5]. In many fields such as chemical engineering, drug industry etc., it is necessary to use liquid mixtures for better results instead of using pure solvent [6-10]. For this the properties of mixture must be known [11, 12]. The properties of mixture depend on the intermolecular interaction between the molecules of the liquid mixture. The ultrasonic study of solution can be used as a precious source of information to observe association / salvation between the molecules, nature of intermolecular interaction [13-15] and physico-chemical properties of the solution [16].

Since ionic liquid such as Na and K- salt solution are important to human as well as plants and also have been currently used as novel agents in organic synthesis [3], catalysis [5], electrochemistry [13] and chemical industry [14]. So it is important to study the various properties of ionic liquid through ultrasonic studies. Though, a number of investigations had been carried out in aqueous NaCl solution [17-25], but to the best of my knowledge, ultrasonic study of aqueous NaCl solution at higher concentration has been scarcely reported.

In present work we have measured the ultrasonic velocity, density and viscosity of aq. NaCl at different concentration at 297 K and the various thermodynamic parameters are calculated which show the presence of specific molecular interaction in the mixture. The association of molecules in the mixture is attributed to the electrostatic force of attraction between the ions in the mixture.

A. Experimental

1) Chemicals used

a) Water: molecular weight-0.018 Kg/mol, distil supplied by *Molychem* used without further distillation

b) NaCl: molecular weight - 0.05844 Kg/mol of Analytic regent (AR) Grade

2) Measurement

a) Velocity Measurement: The mixtures of various concentrations in mole fraction (x_1: Water; x_2: NaCl) were prepared by taking chemicals at 24 ^{o} C temperature. The ultrasonic velocities in liquid mixtures have been measured using a Mittal type (M-81D) ultrasonic interferometer working at 2MHz frequency with an accuracy of ± 0.1 ms^{-1} and have provision for temperature constancy. Circulating water from thermostatically regulated bath (manufactured by Mittal), around the doubled walled sample holder is used to maintain the temperature of liquid constant with a precision ± 0.1 ^{o} C.

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b) *Density Measurement:* Density of solution is measured with RD bottle of 25ml by weighing the solution using digital electronic balance to an accuracy of ± 1 mg. The average uncertainty in the measured density was ±0.001 kg/m^3.

c) *Viscosity Measurement:* Viscosity of solution is measured with Oswald Viscometer by measuring the time of flow using digital stop watch of accuracy 0.01 s. The viscometer was calibrated at each temperature using redistilled water. The uncertainty in viscosity measurement is up to 0.001 kgm^{-1}s^{-1}.

II. THEORY

From the observed values of ultrasonic wave velocity (u), density (ρ) and viscosity (η), the following thermo-acoustic parameters were calculated using following formulas.

1) The isentropic compressibility (K_S) is determined using Laplace's equation –

$$K_S = \frac{1}{\rho u^2} \quad \ldots(1)$$

2) Intermolecular free length (L_f) is determined using the following formula given by Jacobson,

$$L_f = K_T\sqrt{\kappa_s} \quad \ldots(2)$$

Where, K_T is temperature dependent empirical constant, proposed by Jacobson given as K_T= (93.875+0.375×T)×10^{-8} and T is temperature.

3) The acoustic impedance is determined from relation,

$$Z = \rho\, u \quad \ldots(3)$$

4) Rao's constant is calculated by using following equation,

$$R_m = \left(\frac{M_{eff}}{\rho}\right) u^{1/2} \quad \ldots(4)$$

Where,M_{eff} is the effective molecular mass given as M_{eff}=$\sum M_i X_i$.
M_i and X_i are the mole fraction and molecular weight of the individual component.

5) Relaxation time (τ)is calculated using the relation

$$\tau = \frac{4}{3}\frac{\eta}{\rho u^2} \quad \ldots(5)$$

6) The absolute rate theory based Eyring's kinematic viscosity model gives Gibbs free energy relation as

$$\Delta G = -K_B T \log\left(\frac{h}{K_B T \tau}\right) \quad \ldots(6)$$

Where, h is Plank's constant and K_B is Boltzmann constant.

7) The formula for Internal pressure (π)

$$\pi = bRT\left(\frac{K\eta}{u}\right)^{\frac{1}{2}}\left(\frac{\rho^{\frac{3}{2}}}{M_{eff}^{\frac{7}{6}}}\right) \quad \ldots(7)$$

K is a dimensionless temperature-independent constant having a value of 4.28×10^9. Vanderwaal constant b is given by

$$b = \left(\frac{M_{eff}}{\rho}\right)\left[1 - \left(\frac{RT}{M_{eff}u^2}\right)\left\{\left(1 + \frac{M_{eff}u^2}{3RT}\right)^{1/2} - 1\right\}\right]$$

8) According to Suryanarayana and Kuppusamy free volume (V_f) is given by

$$V_f = \left[\frac{M_{eff}\, u}{K\eta}\right]^{3/2} \quad \ldots(8)$$

9) The Molar Volume (V_m) is determined using he following formula

$$V_m = \frac{M_{eff}}{\rho} \quad \ldots(9)$$

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*10)*Salvation Number (S_n) is calculated by using the following formula

$$S_n = \frac{M}{M_o}\left(1-\frac{\beta}{\beta_o}\right)\left(\frac{100-x}{x}\right) \quad \ldots(10)$$

Where M, M_o, β, β_o and x are molecular weight of solution, molecular weight of solvent, adiabatic compressibility of solution, adiabatic compressibility of solvent and number of grams of salt dissolved in 100gm of the solution respectively.

*11)*Surface tension (S) is determined from Auerbach relation-

$$S = 6.3 \times 10^{-4} \rho U^{3/2} \quad \ldots(11)$$

*12)*Volumetric Study is done by using Apparent Molal volume ($\emptyset_V$) [26] given as

$$\emptyset_V = \frac{M}{\rho_0} - 1000\left(\frac{\rho-\rho_0}{m\rho\rho_0}\right) \quad \ldots(12)$$

Where M ($g\ mol^{-1}$) is the molar mass of the solute, m ($mol\ Kg^{-1}$) is the molality of the solutions; $\rho(g\ cm^{-3})$ and ρ_0 ($g\ cm^{-3}$) are the densities of solution and solvent, respectively.

Masson's eq. [27] for the estimation of limiting apparent molal volume is

$$\emptyset_V = \emptyset_V^o + S_v C^{1/2} \quad \ldots(13)$$

$\emptyset_V^o$, which is the limiting apparent molal volume (equal to partial molal volume at infinite dilution and obtained by least square fitting method) and interaction parameter S_v (the experimental slope of the graph $\emptyset_V$ versus $C^{1/2}$) give idea about solute-solute interaction, are considered to be volumetric pairwise interaction coefficient.

*13)*The viscosity data has been analyzed on the basis of Jones- Dole equation [28]

$$\frac{\eta_S}{\eta_o} = 1 + AC^{1/2} + BC \quad \ldots(14)$$

Where η_S and η_o are viscosities of solution and solvent respectively, C is the molar concentration, 'A' and 'B' are constants. Parameter 'A' of Jones-Dole equation represents the contribution from solute-solute interactions i.e. ion-ion interactions. 'A' depends on long range columbic forces and is always positive.

The 'B' parameter which measures the structure making/breaking capacity of an electrolyte in a solution also contain a contribution from structural effects such as size, shape and charge of ions and is responsible for solute-solvent interactions in a solvent. Structure-maker will have positive 'B' and structure-breaker will have negative 'B' [29].

The Jones- Dole equation is limited to very low concentration of electrolyte (<0.1 mole/kg) while for higher concentrations, additional parameter can be added [30]

$$\frac{\eta_S}{\eta_o} = 1 + AC^{1/2} + BC + DC^2 \quad \ldots(15)$$

Additional parameter 'D' is generally positive and related to solute-solute association and volume effects. 'D'-coefficient signifies the hydrodynamic effects due to large size of ions and cation-cation interaction. 'D'-coefficient variations in pre and post miceller region are attributed to the combination of coulombic interaction, cation-cation pairwise interaction and cation-anion-cation triplet interaction effect, affecting the flow properties of solution in a characteristic way.

III. RESULT AND CONCLUSION

Experimentally measured values of ultrasonic velocity (u), density (ρ) and viscosity (η) for NaCl solution at 297 K, at 2 MHz frequency are shown in Table 1.

The values of free length ($\boldsymbol{L_f}$), adiabatic compressibility (β), acoustic impedance ($\boldsymbol{z}$), Relaxation time (τ), Gibbs free energy (G), Rao's constant or molar velocity ($\mathbf{R_m}$) are calculated and presented in Table 2. The parameters internal pressure (π), free volume (V_f), Molar volume (V_m), Salvation Number (S_n) and Surface tension (S) for water-NaCl solution at 297 K, at 2 MHz frequency are shown in Table 3.

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Table 1: Measured values of ultrasonic velocity (u), density (ρ) and viscosity (η) for aq. NaCl solution at 297 K, at 2 MHz frequency

x_2	u Ms^{-1}	ρ Kgm^{-3}	$\eta\times10^{-3}$ $kgm^{-1}s^{-1}$
0.01	1520.513	1030.96	0.9722
0.02	1560.381	1051.60	1.0087
0.03	1592.800	1072.20	1.0430
0.04	1619.380	1095.04	1.0771
0.05	1659.710	1111.56	1.1084
0.06	1686.260	1135.44	1.1507
0.07	1718.880	1155.16	1.2082
0.08	1766.250	1173.32	1.2399
0.09	1770.200	1191.00	1.2941

x_1: Water; x_2:NaCl

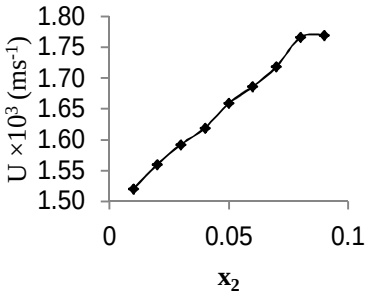

Figure 1: variation of ultrasonic velocity vs. concentration of aq. NaCl solution

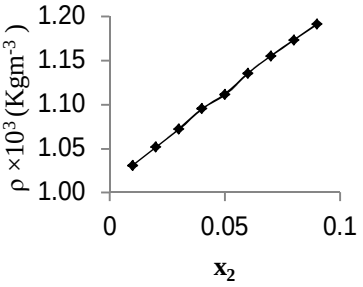

Figure 2: variation of density vs. concentration of aq. NaCl solution

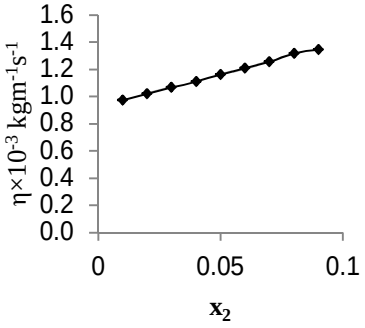

Figure 3: variation of viscosity vs. concentration of aq. NaCl solution

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From Fig.1, Fig.2 and Fig.3 we observe that Ultrasonic velocity (U), density (ρ) and viscosity (η) of NaCl solution increases with increasing molar concentration. This shows that the addition of salt in water leads to strong interaction between the components which increase the close packing structure. Increase in viscosity with concentration suggest the interaction between the different layers of solution thereby decreasing mobility and making dynamic flow of solution slow.

From Fig.4, Fig.5, Fig.7 and Fig.8 we see that the adiabatic compressibility (K_s), Free length (L_f), Relaxation time (τ) and Gibbs free energy (ΔG) decreases with increasing concentration of in aq. salt solution whereas Fig.6 and Fig.9 show that acoustic impedance (Z) and Rao's constant or molar velocity (R) increase with concentration. The decrease in adiabatic compressibility (K_s) is indicative of a more ordered/ compact structure due to well-fitting in solution compared to that of the pure solvent. According to Fort and Moore a decrease in adiabatic compressibility is due to strong hetero-molecular interaction in the liquid mixtures which is attributable to charge transfer, dipole-dipole, dipole-induced dipole interactions, and hydrogen bonding between unlike components [31].

Table 2 : Calculated Thermo-Acoustic parameters for aq. NaCl solution at 297 K, at 2 MHz frequency

x_2	$K_s \times 10^{-10}$ $N^{-1}m^2$	$L_f \times 10^{-11}$ m	z $kgm^{-2}s^{-1}$	$\tau \times 10^{-13}$ s	$-\Delta G \times 10^{-20}$ $KJmol^{-1}$	$R_m \times 10^{-3}$ $m^{10/3}s^{-1/3}mol^{-1}$
0.01	4.1954	4.2041	1567588.08	5.438	0.497	0.20528
0.02	3.9056	4.0563	1640896.66	5.312	0.487	0.20745
0.03	3.6762	3.9354	1707800.16	5.226	0.481	0.20927
0.04	3.4823	3.8302	1773285.88	5.153	0.475	0.21038
0.05	3.2659	3.7092	1844867.25	5.063	0.468	0.21326
0.06	3.0973	3.6122	1914647.05	4.981	0.461	0.21413
0.07	2.9300	3.5133	1985581.42	4.904	0.455	0.21601
0.08	2.7320	3.3925	2072376.45	4.795	0.445	0.21877
0.09	2.6794	3.3597	2108308.20	4.808	0.446	0.21979

x_1: Water; x_2:NaCl

The decrease in Free length (L_f) show that component of mixture get closer, there is interstitial accommodation and orientational ordering between components, making the structure compact (Similar trends in L_f was reported by D Sravana Kumar et al [32] for binary mixtures). According to Eyring and Kincaid, the non-linear trends in free length indicate the presence of structural rearrangements between the components of the molecules [33].

The decrease in Gibbs free energy (ΔG) indicates that less time is needed for rearrangement of molecules (decreasingτ), which leads to dissociation in molecular structure. Increase in acoustic impedance (Z) shows that the elasticity of medium is reduced i.e. rigidity or bulkiness of medium is increased with increasing concentration of salt. The increasing value of Rao's constant (R) also indicates association of components in the solution resulting into strong interaction between them.

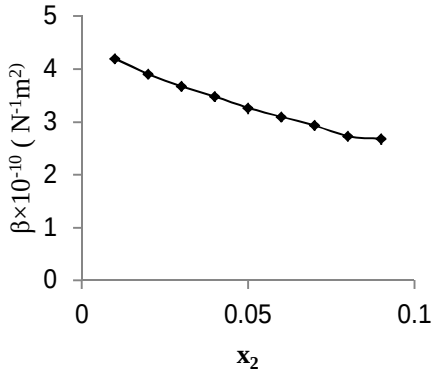

Figure 4: variation of adiabatic compressibility vs. concentration of aq. NaCl solution

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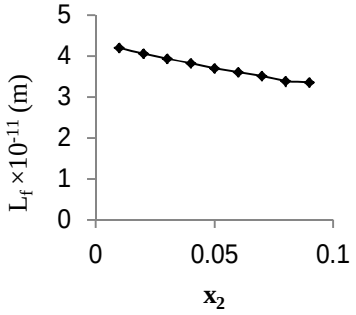

Figure 5 : variation of Free Length vs. concentration of aq. NaCl solution

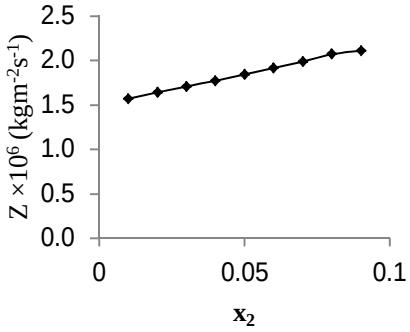

Figure 6 : variation of Acoustic impedance vs. concentration of aq. NaCl solution

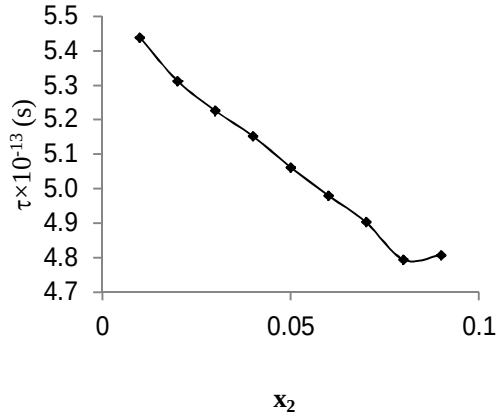

Figure 7: variation of Relaxation Time vs. concentration of aq. NaCl solution

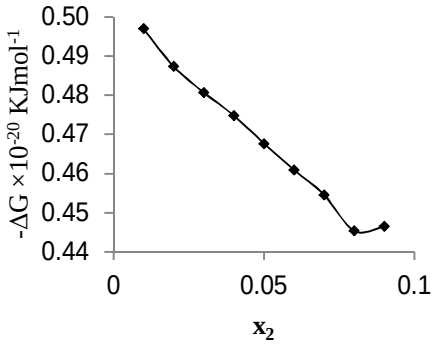

Figure 8 : variation of Gibbs Free Energy vs. concentration of aq. NaCl solution

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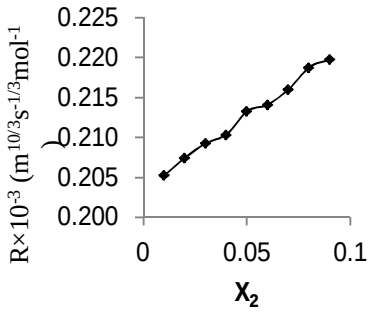

Figure 9 : variation of Rao's constant vs. concentration of aq. NaCl solution

Internal pressure (π) is the measure of cohesive forces which are result of attractive and repulsive forces between the molecules. Fig. 10 show that the internal pressure (π) increases with concentration. This indicates a strong attractive type force, the electrostatic field of the ions, causing orientation of the solvent molecules around the ions (Similar trends in π was reported by G. S. Gayathri et al [34] for binary mixtures).

Table 3: Calculated Thermo-Acoustic parameters for aq. NaCl solution at 297 K, at 2 MHz frequency

x_2	π Nm^{-2}	S_n	S Nm^{-1}	$V_m \times 10^{-5}$ m^3mol^{-1}	$V_f \times 10^{-8}$ m^3mol^{-1}
0.01	22581.9	77.02	38509.47	1.7852	1.744
0.02	22654.4	141.50	40835.43	1.7886	1.742
0.03	22739.8	187.25	42939.62	1.7919	1.736
0.04	22805.4	223.32	44956.62	1.7915	1.729
0.05	22913.4	255.75	47350.20	1.8013	1.725
0.06	22949.3	281.44	49532.65	1.7990	1.723
0.07	23021.2	301.86	51862.21	1.8033	1.720
0.08	23112.5	320.00	54870.04	1.8098	1.717
0.09	23171.2	326.52	55883.79	1.8169	1.715

x_1: Water; x_2:NaCl

The Surface tension(S), Salvation number (Sn) and Molar Volume (Vm) (Fig.11, Fig.12 and Fig.13) are found to increase with concentration. Increase in salvation number reveals that compressibility of solution is less than that of the solvent. Increase in surface tension indicates stronger molecular interaction between adjacent molecules. The molar volume is also seen to increase with concentration of salt which shows that interaction among components of solution tends to rearrange the geometry of the cluster in such a way that the volume of cluster increases (Similar trends in V_m was reported by Anil Kumar et al [35] for binary mixtures). This will lead to decrease in free volume in the solution (Fig.14). We can also say that the component molecules come more close to each other in the liquid mixture and are allowed to fit into each other's structure i.e. the interstitial accommodation thereby decreasing the free space (Similar trends in V_f was reported by Mishra Sujata et al [36] for binary mixtures).

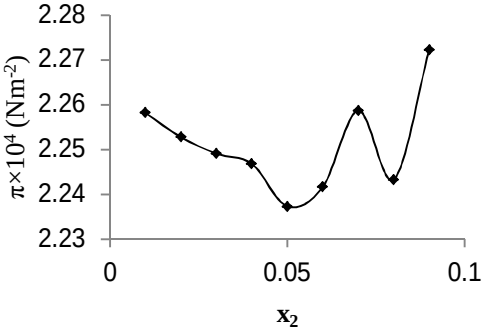

Figure 10: variation of internal pressure vs. concentration of aq. NaCl solution

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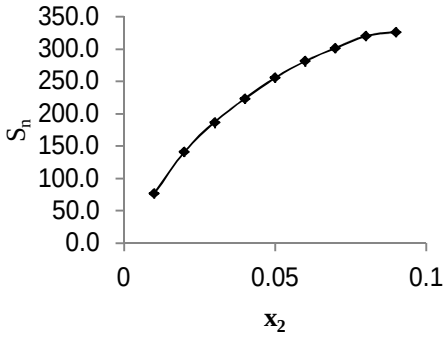

Figure 11: variation of Solvation Number vs. concentration of aq. NaCl solution

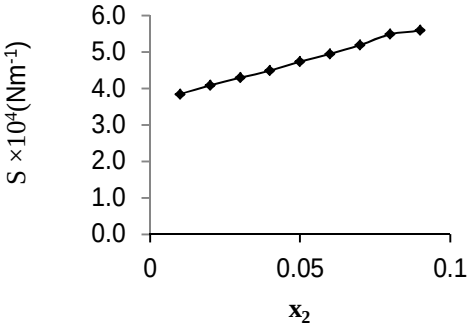

Figure 12: variation of Surface Tension vs. concentration of aq. NaCl solution

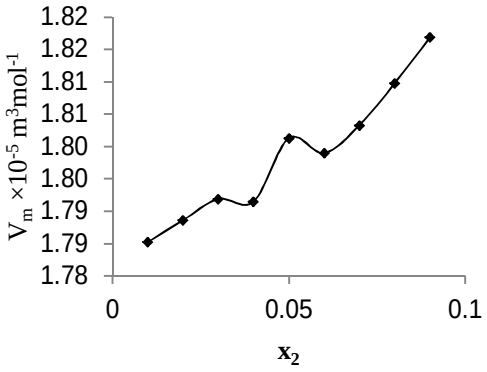

Figure 13: variation of Molar Volume vs. concentration of aq. NaCl solution

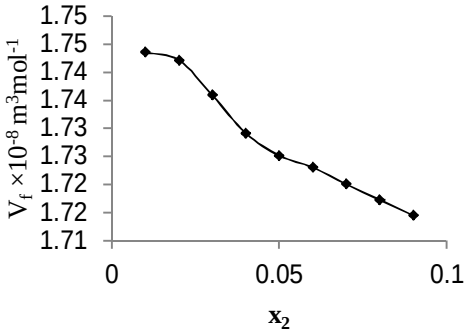

Figure 14: variation of free volume vs. concentration of aq. NaCl solution

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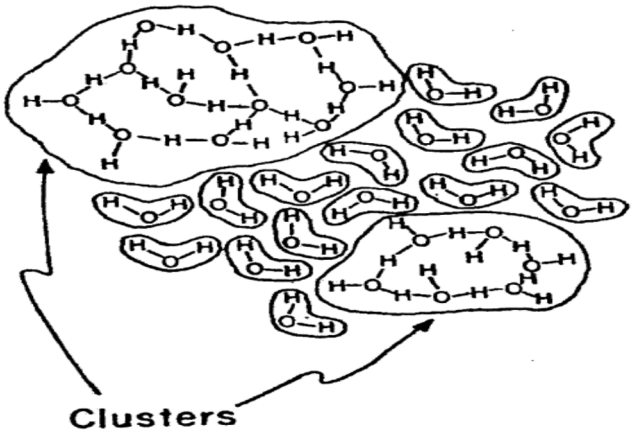

Figure 15 : Flickering Cluster Model of Water

The structure of water molecule is shown in Fig.15 known as "Flickering Cluster" model proposed by Frank and Wen [37]. It is supposed to consist of H-bonded cluster and unbounded water molecules [38]. The H-bonded structure gets disturbed by the addition of the electrolyte. On adding salt into water, the salt gets dissociated into positive and negative ions. These charged ions interact with water molecules (dipole with dipole moment 1.85 d) via ion-dipole interaction and enhance the cluster breaking which is a cooperative process. At the same time the electrostatic field of ions tries to orient the water dipole into hydration shell, making the solution more ordered [39-41]. This ordered geometry of solution with increase in concentration is due to strong ion-dipole interaction which results in increase in acoustic velocity and density.

Hydration shell results in increase in effective size of ions, making solution bulky. The bulkiness and strong forces between water molecules and ions cause increase in viscosity and molar volume. Initially when the concentration of solute is low the number of ions to form hydration shell is low. As concentration increases more ions become available to form hydration shell. Increased numbers of hydration shell cause increase in viscosity and molar volume with concentration.

Increase in salt concentration in water results in more ionization thereby increasing the number of ions in the solution. Repulsion between Like ions causes expansion of free volume while electrostatic attraction between water molecules and ions cause contraction in volume. Decreasing trend of free volume shows that electrostatic force of attraction dominates over the repulsive force.

Due to strong attraction between water and ions, water molecules are surrounded by ions Fig.16 [42] more the concentration of salt, more ions become available to interact with water molecules, causing increase in internal pressure, surface tension and salvation number.

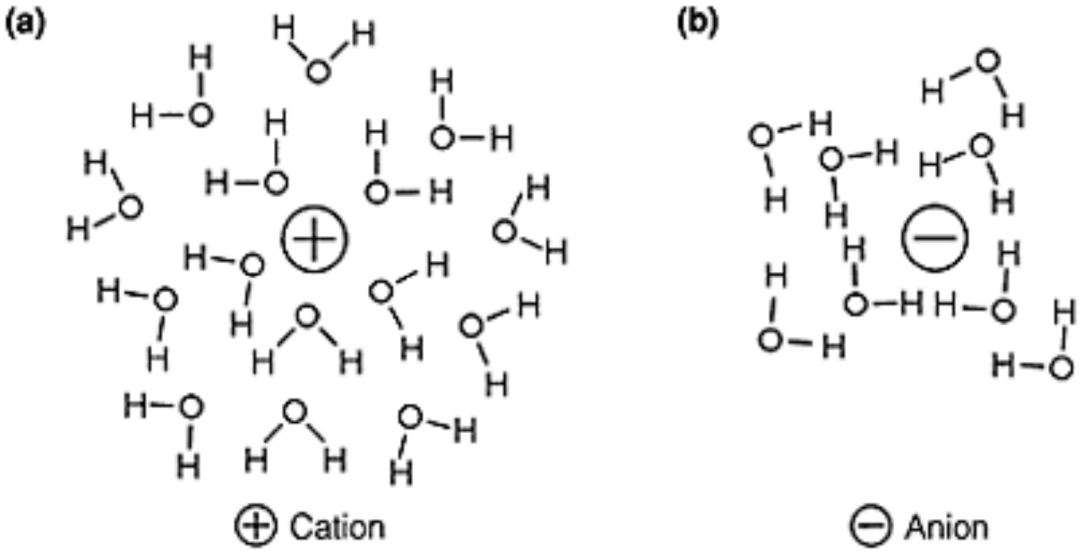

Figure 16: The interaction between water and a cation and water and an anion in solution

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A. Volumetric and Viscomitric study of Electrolyte (NaCl) and water solution

Table 4 : Apparent molal volume and relative viscosity data

m	ρ gcm^{-3}	$\eta_s \times 10^{-3}$ $kgm^{-1}s^{-1}$	$-\varphi_v$ m^3mol^{-1}	η_s/η_o
0.561167	1.03096	0.972167	58.3293	1.067494
1.133787	1.05160	1.008727	45.6461	1.107639
1.718213	1.07220	1.043013	40.7425	1.145288
2.314815	1.09504	1.077100	38.6377	1.182717
2.923977	1.11156	1.108410	35.2216	1.217096
3.546099	1.13544	1.150682	34.3722	1.263514
4.181601	1.15516	1.208228	32.7381	1.326703
4.830918	1.17332	1.239940	31.1058	1.361524
5.494505	1.19100	1.294124	29.6466	1.421021

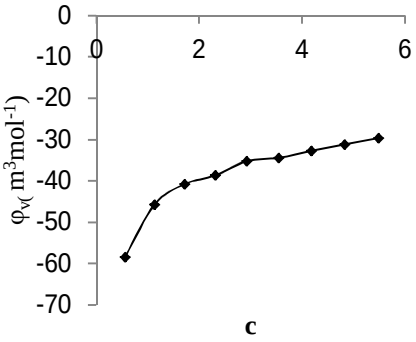

Figure 17: concentration C vs. apparent molal volume graph

The nonlinear variation of Apparent Molal volume ($\emptyset_V$) indicates the existence of ion-solvent interaction. The low and positive value of interaction parameter S_V represents the weak solute-solute interaction the negative value of limiting apparent molal volume at infinite dilution $\emptyset_V^o$ represent the larger electrostatic interaction. Science $\emptyset_V^o > S_v$ (from Table 5) represent that the ion-solvent interaction dominates over the ion-ion interaction.

Table 5 : The values of limiting apparent molar volume $\emptyset_V^o$, interaction parameter S_v and Jones-Dole coefficients A, B, D

Parameter	Estimated Value
φ_v^o m^3mol^{-1}	-64.81667
S_v (molal. $M^3Kg^{1/2}$ $mole^{-3/2}$)	15.97926
A ($Kg^{1/2}mol^{-1/2}$)	0.07388
B (Kg mol^{-1})	0.01929
D (Kg^2 mol^{-2})	0.00473

Jones-Dole equation explains the contribution of long range inter-ionic attraction to the viscosity of electrolyte. Very small and positive value of Jones-Dole equation parameter A represents the presence of long range columbic force. Positive value of the B parameter represents the structure-making tendency of solute. Parameter D>0, shows that at higher concentration the hydrodynamic effect cause the variation of viscosity from Jones-Dole equation.

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IV. CONCLUSION

The systematic study of Sodium Chloride solution has been done by us at different concentration at 2 MHz frequency and 297 K temperature. The result of our study reveals that ultrasonic velocity, density, viscosity and other thermodynamic parameter are highly dependent on the concentration of salt in the solution. The nonlinear variation in these parameters shows the presence of solute-solute, solute-solvent and solvent- solvent interaction. But our volumetric study shows that ion-dipole i.e. solute-solvent interaction dominates over ion-ion interaction. The ions formed in the solution makes the water structure. This is also confirmed by the positive value of Jones-Dole B coefficient.

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1) Figure Captions

Figure 18: variation of ultrasonic velocity vs. concentration of aq. NaCl solution
Figure 19: variation of density vs. concentration of aq. NaCl solution
Figure 3: variation of viscosity vs. concentration of aq. NaCl solution
Figure 20: variation of adiabatic compressibility vs. concentration of aq. NaCl solution
Figure 21 : variation of Free Length vs. concentration of aq. NaCl solution
Figure 22 : variation of Acoustic impedance vs. concentration of aq. NaCl solution
Figure 23: variation of Relaxation Time vs. concentration of aq. NaCl solution
Figure 24 : variation of Gibbs free energy vs. concentration of aq. NaCl solution
Figure 25 : variation of Rao's constant vs. concentration of aq. NaCl solution
Figure 26 : variation of internal pressure vs. concentration of aq. NaCl solution
Figure 27 : variation of Solvation Number vs. concentration of aq. NaCl solution
Figure 28: variation of Surface Tension vs. concentration of aq. NaCl solution
Figure 29: variation of Molar Volume vs. concentration of aq. NaCl solution
Figure 30: variation of free volume vs. concentration of aq. NaCl solution
Figure 31 : Flickering Cluster Model of Water
Figure 32 : The interaction between water and a cation and water and an anion in solution
Figure 33 : concentration C vs. apparent molal volume graph

2) Table Caption

Table 6: Measured values of ultrasonic velocity (U), density (ρ) and viscosity (η) for aq. NaCl solution at 297 K, at 2 MHz frequency
Table 7 : Calculated Thermo-Acoustic parameters for aq. NaCl solution at 297 K, at 2 MHz frequency
Table 8: Calculated Thermo-Acoustic parameters for aq. NaCl solution at 297 K, at 2 MHz frequency
Table 9 : Apparent molal volume and relative viscosity data
Table 10 : The values of limiting apparent molar volume $\emptyset_V^o$, interaction parameter S_v and Jones-Dole coefficients A, B, D

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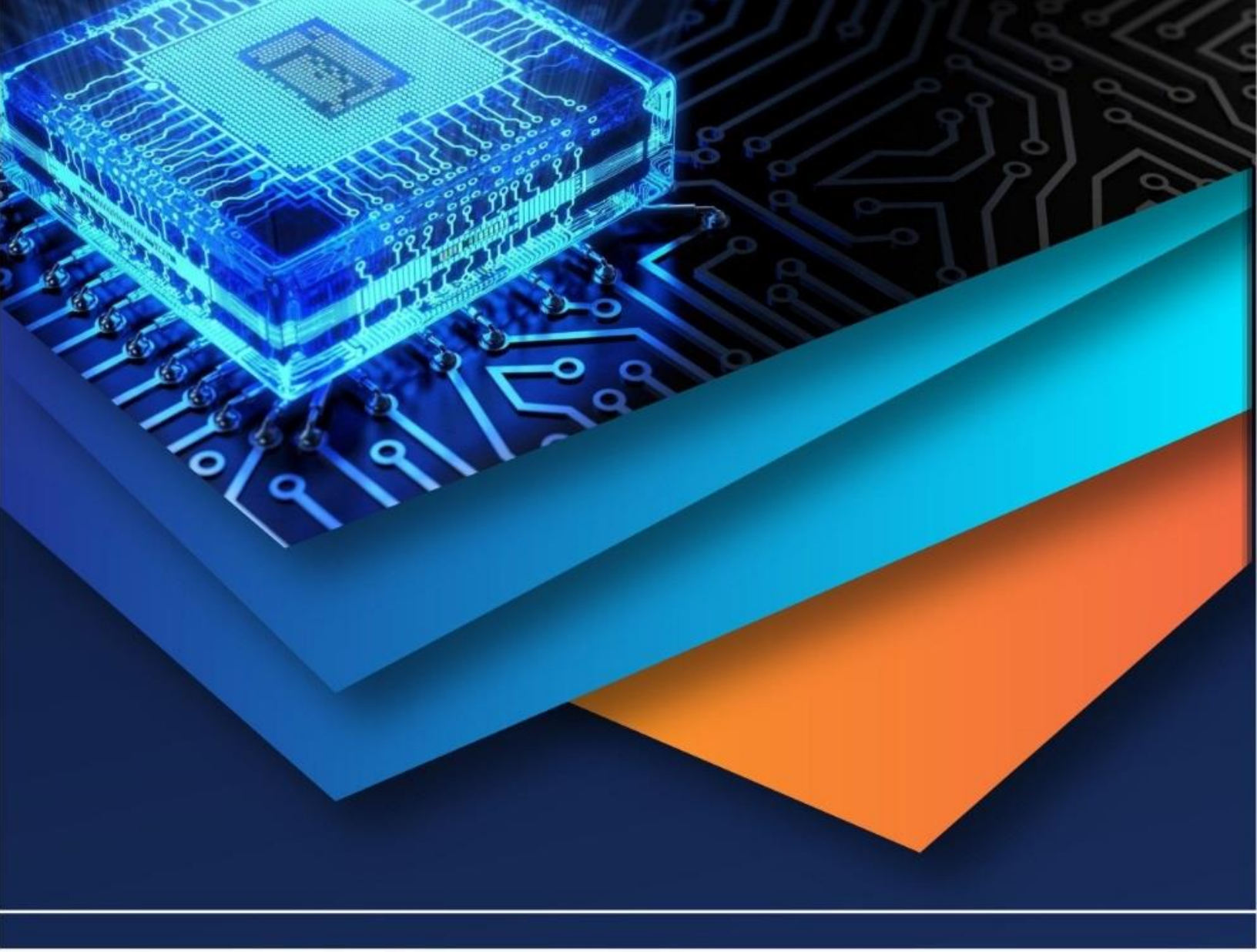

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