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Ultrasonic study of the molecular interaction parameters of HCl in aqueous ethanolic mixtures

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Abstract: The experimental data pertaining to ultrasonic velocity, density and viscosity have been resolved for Hydrochloric acid in aqueous ethanolic binary mixtures at30ºC. Several acoustical parameters like intermolecular free length(Lf), relative association (R.A), Adiabatic compressibility (β), Specific acoustic impedance (Z), Free volume(V_f) and Wada's constant(W) have been calculated from the velocity, viscosity and density data of HCl in binary mixtures. The variation of these parameters gives interesting and useful information regarding the behavior of molecules in the binary liquid mixtures. Interesting results on the structure making and structure breaking properties of the liquids have been recorded which delve directly into the solute-solvent and solvent-solvent interactions. The results obtained are presented and analyzed to study the molecular interactions that exist between unlike molecules of HCl in the binary mixtures. The method adapted to study the present work involves the usage of a single crystal variable path interferometer. The accuracy of the interferometer ranged to an accuracy of ± 0.05 %. A 10 ml specific gravity bottle was used to measure the density of the liquids. An Oswald viscometer was used to measure the viscosity of the pure liquids and liquid mixtures. The estimated accuracy was valued to 0.01s. The specific conductivity of the triply distilled water used for the study was of the order of $1X10^{-7}Scm^{-1}$.

The work can be extended to probe into the reactivity of several acids, salts and other solutes into various other binary solvent mixtures supported with computational methods that may draw other interesting conclusions. The Density results of HCl in with increased intermolecular interactions between ethanol and water molecules suggest the structure making properties of the solvent due to the added solute. These results are from the inferred decreasing density values of HCl in alcoholic water mixtures. Specific molecular interactions in the mixture are inferred by a cluster model of a stacked ethanol core surrounding the species. This is shown by the increase in ultrasonic velocity values of HCl throughout the study. The results of intermolecular free length and R.A validate the direct impact of strong molecular association due to the impact of intermolecular cohesion existing in the system. The increase in acoustic impedance, free volume and Wada's constant confirm the existence of strong intermolecular interaction. This suggests the presence of a hydrogen bonding between HCl and the liquid mixture the results in common formulates the path for research in solution chemistry as it supplements in understanding the different types of molecular interactions existing in the system.

Keywords: Molecular interaction, Ultrasonic velocity, HCl, acoustical parameters.

1. INTRODUCTION

The studies of intermolecular interactions play a prominent role in determining the behavior of the medium. Molecular interactions are very important in forming biological organisms or even life. The different types of intermolecular interactions may be attractive or repulsive in nature. Numerous studies in liquid systems highlight on the specific kind of interactions that exist in a system. Though methods like the Infra-Red, Raman Effect, Dielectric constant and the nuclear magnetic resonance are available, ultrasonic techniques are becoming highly successful in highlighting the ion-ion interactions of the molecules in the system. This technique probes into the system giving information about the molecular behavior of liquids and solids that characterize the physiochemical properties of the medium. However the results of ultrasonic studies are very useful as it delves into the ion-ion interactions of a system. The physiochemical properties of the solution can be characterized from the behavior of molecules in liquids and solids using the ultrasonic technique.

The present study deals with the study of molecular interaction of HCl in varied combinations of ethanol water mixtures by ultrasonic technique. Most of the derived acoustical parameters throw light on the type and kind of intermolecular interactions in the binary and liquid mixtures[1-6]

The reactivity of Glycine, L-alanine and L-serine in water ethanol mixtures at 298.15 K were discussed in terms of the apparent molar volume calculation by X. Renetal. The results established the structure making property of aminoacid in the liquid mixture. The studies also concluded that with an increasing ethanol percentage up to 45 % there was a decrease in the electrostriction.

The measured thermodynamic properties were discussed in terms of nature of molecular interactions between component molecules. The free energies and amino acid side chain transfer from water to ethanol and dioxane solutions have been measured by Yasuhiko Nosaki*et al.*[7]. The results were discussed based on the stability of the side chains over the reactivity of ethanol and dioxane.

2. MATERIALS AND METHODS

Chemicals of extremely high purity and analytical grade [E. Merck., India] were used in the present study. The density of the liquids was measured using a10ml specific gravity bottle. An Oswald viscometer with an estimated accuracy of 0.01s was used to measure the viscosity of the liquid mixture. A single crystal variable path interferometer with an accuracy of ± 0.05 % was used to measure the ultrasonic velocity. The specific conductivity of the triply distilled water used in the present study was analyzed to be $1X10^{-7}Scm^{-1}$

2.1 Purification methods: Purification was done by standard procedure [8-10] and the solvent was redistilled before use. The density of the purified solvent used was $0.7806gcm^{-3}$ with a coefficient of viscosity of 0.9590mPa.s at room temperature.

2.2 Statistical analysis: Several concentrations of HCl, 0.001 N, 0.002 N, 0.003 N, 0.004 N, 0.005 N, 0.006 N, 0.008 N, and 0.010N in ethanol/water mixtures ranging from 5%, 10%, 15%, 20%, 25%, 50% and 80% were prepared individually and investigated.

3. RESULTS AND DISCUSSION

The data pertaining to density, viscosity and ultrasonic velocity are given in **Tables 1-2**.From Table 1, it is evident that as the concentration of HCl is increased there is a very slight increase in density for any given ethanol water mixture. The results are graphically seen in **Figure 1.**From the figure, it is also seen that the density decreases with the increase of alcohol content. This trend suggests the possibility of an increased intermolecular interaction between the ethanol and water molecules as seen in literature [11, 12]. The mixing of ethanol and water molecules depicts the greater association of water compared to liquid alcohol.

Table No. 1. Experimental profile of Density, Velocity and Viscosity of HCl in ethanol water mixtures.

	Concentration (M)	**Velocity (m/s)**	**Density (Kg/m^3)**	**Viscosity(Pa.S.) or $Kg\ m^{-1}\ s^{-1}\times10^{-3}$**
5%	0.001098	1530	0.698	55.5
	0.002196	1530.2	0.699	56.22
	0.003294	1533.7	0.700	57.06
	0.004392	1539.4	0.701	57.16
	0.00549	1545.6	0.702	57.32
	0.006588	1549.7	0.703	58.28
	0.00878	1564	0.704	59.03

	0.01098	1565.7	0.705	60.04
10%	0.001098	1549.3	0.689	62.32
	0.002196	1564.5	0.691	62.47
	0.003294	1572.5	0.693	62.56
	0.004392	1576.5	0.694	62.97
	0.00549	1582.2	0.695	63.13
	0.006588	1586.8	0.697	63.15
	0.00878	1588	0.698	63.6
	0.01098	1595.3	0.699	65.97
15%	0.001098	1558.8	0.681	63.59
	0.002196	1570.6	0.687	63.87
	0.003294	1573	0.688	66.25
	0.004392	1578	0.691	67.81
	0.00549	1584.5	0.692	68.78
	0.006588	1591.2	0.694	68.84
	0.00878	1612.4	0.695	68.93
	0.01098	1655	0.696	69.41
20%	0.001098	1565.4	0.682	72.28
	0.002196	1573	0.6821	78.53
	0.003294	1588	0.6822	78.56
	0.004392	1608	0.6824	79.28
	0.00549	1609.7	0.6826	80.47
	0.006588	1610	0.683	80.62

	0.00878	1621.3	0.6848	80.85
	0.01098	1699.4	0.6878	81.28
25%	0.001098	1577.4	0.6812	87.72
	0.002196	1580.6	0.6814	88
	0.003294	1588.6	0.6819	88.44
	0.004392	1614.6	0.6838	88.63
	0.00549	1619.2	0.6844	88.75
	0.006588	1623.6	0.6845	88.88
	0.00878	1631.3	0.6856	89.66
	0.01098	1706.6	0.6862	90.47
50%	0.001098	1580	0.6535	117.06
	0.002196	1615.2	0.6542	117.4
	0.003294	1638.3	0.6544	117.8
	0.004392	1683	0.6559	118.28
	0.00549	1689.3	0.6567	118.75
	0.006588	1704	0.657	118.87
	0.00878	1714.8	0.6574	118.91
	0.01098	1758.6	0.6577	119.06
80%	0.001098	1470	0.5881	96.75
	0.002196	1518.6	0.5883	98.62
	0.003294	1604.8	0.5887	100.09
	0.004392	1618	0.5888	101.72
	0.00549	1635	0.589	103.13

	0.006588	1642.6	0.5892	112.97
	0.00878	1655	0.5897	114.5
	0.01098	1682.2	0.5899	114.91

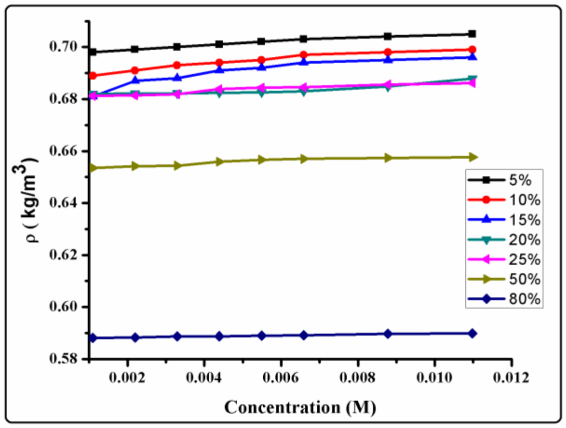

Fig.1: Plot of Density Vs.Concentration of HCl in ethanol water mixtures

The combination of ethanol and water results in a considerable release of heat of the solution, in measurable form. The molecules thus start spreading out as the solutions gets warmer. The solution becomes less dense and the specific gravity gets lowered. Liquid alcohols associates less strongly and produce polymeric H-bonded chains, instead of large clusters. Thus a closer packing of the molecules is expected with an overall decrease in size of the hydrated alcohol. As the hydrated alcohol fits in a number of molecules into a given space the structure tends to become volume-compact[13].The decreasing density values with increasing concentration show the shrinkage in volume of the mixture. This must be due to the presence of solute molecules. This also suggests the structure making property of the solvent.

The experimental ultrasonic velocity data of HCl in the ethanol water systems is seen in Table 1.The results are graphically depicted in **Figure 2.**The figure depicts the increase in ultrasonic velocity values with increase in ethanol. Reported studies account for the fact that associates of ethanol molecules start to grow at X EtOH>0.1. At this stage the 3D water structure breaks down in ethanol-water mixtures. When the X EtOH>0.15, the liquid mixtures show the formation of a large number of water-ethanol bonds, and the water-water bonds are broken.

It is seen that when a pure organic solvent is added into the medium, the micro-heterogeneous structure of the binary solvent undergoes large and nonlinear changes[14].This trend also suggests the possibility of ionic hydration or hydrogen bonding of the solutes resulting in a greater association of the molecules again. The inferred structure is hence described by a cluster model of a stacked ethanol core surrounded by a thin water shell[15, 16]. The relatively small ions present in the liquid medium reduce the compressibility with high ultrasonic velocity values as reported in literature [17]. Thus the presence of specific molecular interactions is closely expected in the binary liquid medium.

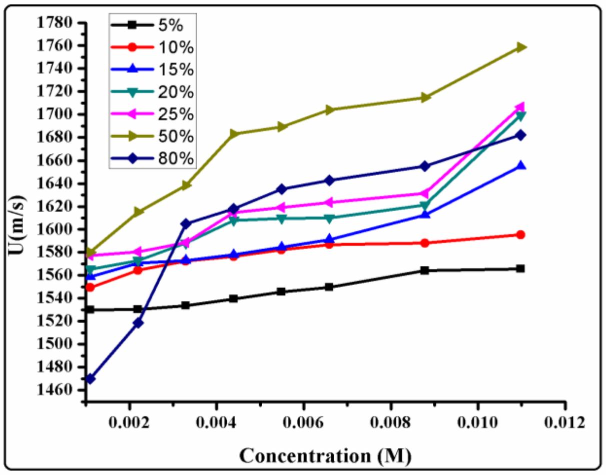

Fig.2: Plot of Velocity Vs.Concentration of HCl in ethanol water mixtures

The decrease in adiabatic compressibility values suggest the complex formation tendency of the liquid mixture which might arise due to a expected contraction on mixing. It is also expected that the solvent molecules aggregate around the solute molecules indicating the possibility of solute-solvent interactions[18, 19]. It is seen that the solvent molecules present in the solution surround the hydrochloric acid due to the combined effect of electrostriction and wrenching. Hence the space automatically gets reduced for the next incoming ion which is clearly seen in **figure 3.**

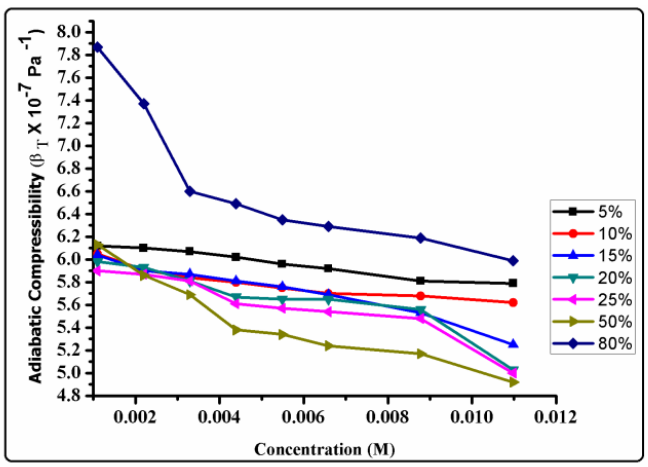

Fig.3: Plot of Adiabatic Compressibility Vs.Concentration of HCl in ethanol water mixtures

From **Table 2,** it is seen that the intermolecular free length decreases with increase in concentration. The results denote the presence of some type of interaction, either the solute-solute interaction or the solute-solvent interaction. This may be due to the compression of liquid, which indicates that the molecules are coming closer to each other; hence the intermolecular cohesion is stronger leading to strong molecular association[19]. According to Eyring and Kincaid[20] the regular fall in free length causes a rise in sound velocity in the mixture. The expected decrease in adiabatic compressibility values and increases in ultrasonic velocity values indicate the associative effect of the protic species that endsup into a clustering together which form miniature cage like agglomerates [21]. The results are graphically seen in **Fig.4.**The experimental values of acoustic impedance (Z) are represented in **Table-2**.

Table2: Experimental profile of Adiabatic compressibility(β),intermolecular free length(LF) specific acoustic impedance(Z) relative association(R.A), Rao's constant (R) and Wada's constant (W) values of HCl in Ethanol Water mixtures

%of Ethanol	**Concentration (M)**	**Adiabatic Compressibility (βS x 10-7)Pa-es**	**Acoustic Impedance Z x 10^{6}($Kgm^{-2}S^{-1}$)**	**Free length Lf x 10^{-13} m**	**Wada's Constant (W)×10^{-3}**	**Relative Association (RA)**	**Free Volume (Vf) ×10^{-7}**
5%	0.001	6.12	1067.9	1.56	3.61	0.2844	4.2
	0.0021	6.11	1069.6	1.56	3.97	0.2848	4.53
	0.0032	6.073	1073.6	1.55	4.39	0.2859	4.85
	0.0043	6.02	1079.1	1.55	4.81	0.2873	5.23
	0.0054	5.963	1085	1.54	5.10	0.2889	5.62
	0.0065	5.923	1089.4	1.54	5.36	0.2901	5.91
	0.0087	5.807	1101.1	1.52	6.10	0.2932	6.64
	0.0109	5.786	1103.8	1.52	6.84	0.2939	7.26
10%	0.001	6.047	1067.5	1.55	7.03	0.2842	7.22
	0.0021	5.912	1081.1	1.53	5.26	0.2879	7.22
	0.0032	5.836	1089.7	1.52	7.67	0.2902	7.63
	0.0043	5.798	1094.1	1.52	7.94	0.2913	8.01
	0.0054	5.748	1099.6	1.51	8.39	0.2928	8.33
	0.0065	5.698	1106	1.51	9.33	0.2945	8.69
	0.0087	5.681	1108.4	1.5	9.42	0.2952	9.06
	0.0109	5.621	1115.1	1.5	1.00	0.2969	9.7

15%	0.001	6.043	1061.5	1.55	1.04	0.2827	10.1
	0.0021	5.901	1079	1.53	1.07	0.2873	10.2
	0.0032	5.874	1082.2	1.53	1.06	0.2882	10.5
	0.0043	5.812	1090.4	1.52	1.12	0.2904	10.9
	0.0054	5.756	1096.5	1.51	1.05	0.292	10.8
	0.0065	5.691	1104.3	1.51	1.18	0.2941	10.9
	0.0087	5.534	1120.6	1.48	1.23	0.2984	11.1
	0.0109	5.246	1151.9	1.45	1.33	0.3067	11.5
20%	0.001	5.984	1067.6	1.54	1.36	0.2843	12.3
	0.0021	5.925	1072.9	1.54	1.23	0.2857	13.2
	0.0032	5.813	1083.3	1.52	1.41	0.2885	13.5
	0.0043	5.667	1097.3	1.5	1.41	0.2922	12.3
	0.0054	5.654	1098.8	1.5	1.47	0.2926	11.6
	0.0065	5.648	1099.6	1.5	1.49	0.2928	12.0
	0.0087	5.555	1110.3	1.4	1.56	0.2956	12.3
	0.0109	5.034	1168.8	1.4	1.62	0.3112	12.5
25%	0.001	5.9	1074.5	1.5	1.77	0.2861	12.7
	0.0021	5.87	1077	1.5	1.77	0.2868	13.3
	0.0032	5.811	1083.3	1.53	1.71	0.2885	14.5
	0.0043	5.61	1104.1	1.49	1.75	0.294	14.6
	0.0054	5.57	1108.2	1.49	1.87	0.2951	12.7
	0.0065	5.54	1111.4	1.49	1.97	0.2959	12.9
	0.0087	5.48	1118.4	1.48	1.80	0.2978	13.2

	0.0109	5.00	1171.1	1.41	2.11	0.3118	13.6
50%	0.001	6.13	1032.5	1.56	3.39	0.2749	13.9
	0.0021	5.85	1056.7	1.53	3.70	0.2814	14.2
	0.0032	5.69	1072.1	1.51	3.90	0.2855	14.6
	0.0043	5.38	1103.9	1.46	4.41	0.2939	15.7
	0.0054	5.33	1109.4	1.46	3.83	0.2954	15.8
	0.0065	5.24	1119.5	1.44	3.78	0.2981	18.8
	0.0087	5.17	1127.3	1.43	3.91	0.3002	19.4
	0.0109	4.91	1156.6	1.4	3.85	0.308	19.8
80%	0.001	7.86	864.51	1.77	8.48	0.2302	20.5
	0.0021	7.37	893.39	1.71	7.16	0.2379	20.7
	0.0032	6.59	944.75	1.62	8.02	0.2516	21.0
	0.0043	6.48	952.68	1.61	6.60	0.2537	21.6
	0.0054	6.35	963.02	1.59	6.85	0.2564	22.5
	0.0065	6.29	967.82	1.58	6.99	0.2577	23.5
	0.0087	6.19	975.95	2.75	7.18	0.2599	33.8
	0.0109	5.99	992.33	1.54	7.80	0.2642	34.5

It is observed that the values of Z increase with increase in concentration. It is hence expected that there should be a hydrogen bonding between HCl and the liquid medium. As seen from literature the Z values are directly proportional to the ultrasonic velocity values and inversely related to the values of adiabatic compressibility.

The values of Relative Association (R.A) are seen to decrease with an increase in concentration. This indicates the breaking of the solvent molecules on addition of HCl, which is in concordance with literature. The combined effect of breaking followed by solvation suggests the decrease in structure forming tendency between the solute and solvent molecules.

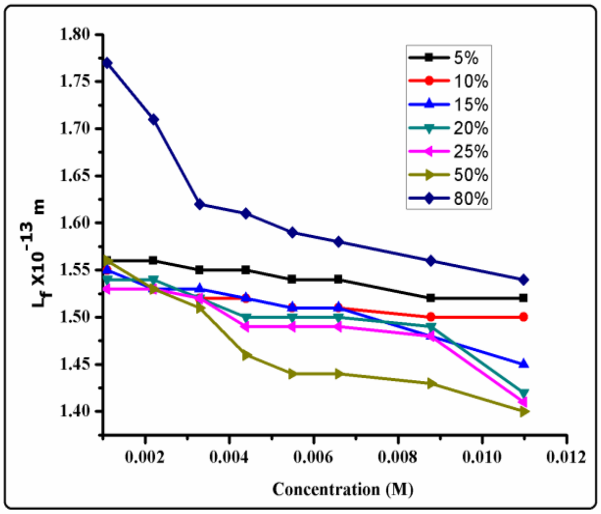

Fig.4: Plot of Intermolecular free length (L_F) Vs. Concentration of HCl in ethanol water mixtures

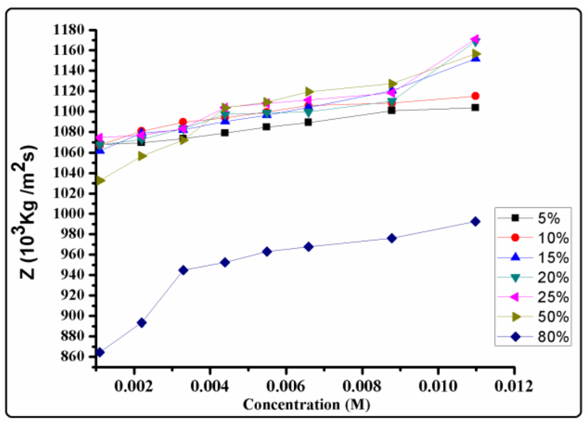

Fig.5: Plot of Specific acoustic Impedance Vs. Concentration of HCl in ethanol water mixtures

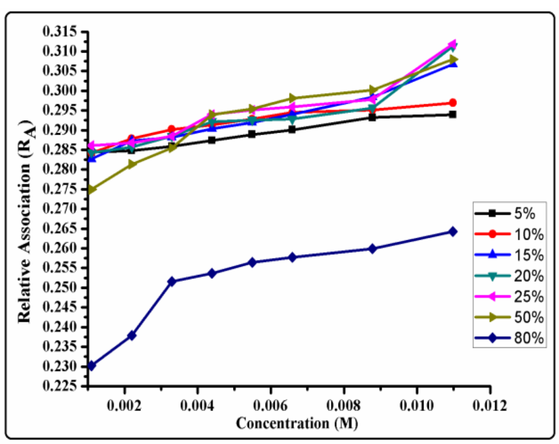

Fig.6: Plot of Relative Association Vs. Concentration of HCl in ethanol water mixtures

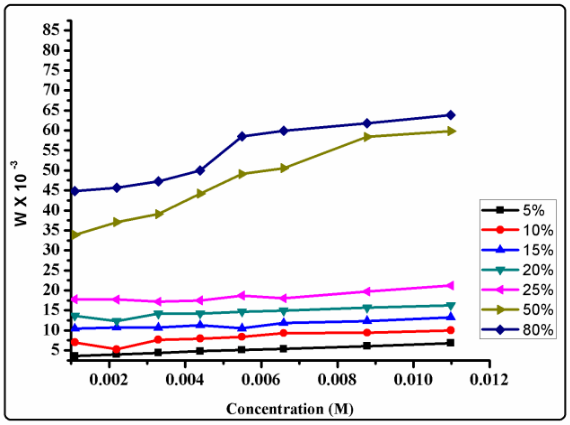

Fig.7: Plot of Wada's constant Vs. Concentration of HCl in ethanol water mixtures

The results of **Fig.7**indicate the increase in free volume with increase in concentration. This is focused on the weakening of molecular interactions due to the lose packing of the molecules inside the liquid medium. The results of Wada's constant from **Figure 8** confirm the prevalence of strong intermolecular interactions existing in the liquid mixture.

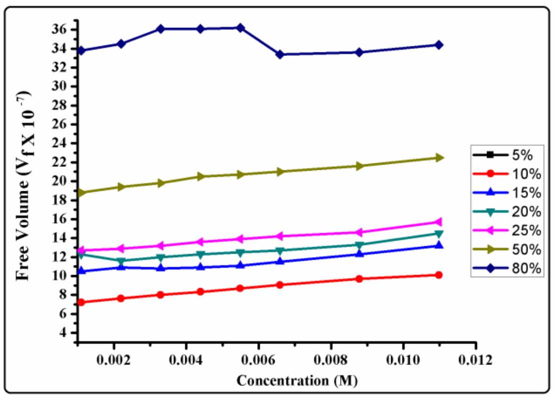

Fig.8: Plot of Free Volume Vs.Concentration of HCl in ethanol water mixtures

4. CONCLUSION

Hence from the inference drawn from the studies it is seen that when HCl is added to aqueous ethanol mixtures the system is characterized by increased intermolecular interactions. Structural changes due to hydrogen bond interactions are hence interpreted in the solute-solvent mixtures. Our results give a straight forward interpretation as a hydrophobic reagent could be hidden inside the cluster and it seems to be unavailable for reaction. The studies on such kind of mixed solvent study would be of tremendous utility in understanding the different types of molecular interactions existing in the system. This phenomenon also formulates the path for research and investigation in solution chemistry that unravels the nature of interaction that is highlighted through the experimental data collected.

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