

ACOUSTICAL STUDIES OF TERNARY MIXTURES AT DIFFERENT TEMPERATURES

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ABSTRACT

The ternary mixture of crotonaldehyde with iodine in n-hexane has been studied extensively from the view point of their acoustic and molecular interaction studies at 293.15, 298.15, 303.15 and 308.15K. Experimental values are used to compute derived parameters. By using the ultrasonic velocity (U), density (ρ) coefficient of viscosity (η), the other acoustical parameters adiabatic compressibility (κ), free length (L_f), interaction parameter (χ_i) and molar volume (V_m) are calculated. The ultrasonic velocity decreases with concentration at 0.006M. It reveals the weak interaction that takes place at 303.15K of the ternary mixtures. The study reveals that weak interaction takes place at higher temperatures of the ternary mixtures.

Keywords: Ultrasonic velocity-Dipole-dipole interaction, free length, adiabatic compressibility-crotonaldehyde.

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INTRODUCTION

Ultrasonic studies play an important role in the investigation of intermolecular interactions between the molecules. Ultrasonic waves have acquired the status of an important probe for the study of structure and properties of matter. The structural arrangements are influenced by the shape of the molecules as well as by their mutual interactions¹. The significant information of ternary liquid mixtures on the physicochemical behavior of ultrasonic wave propagation influences the physical properties of the medium. The variation in acoustic properties with temperature provides added information regarding the binary and ternary system. The sign and magnitude of non-linear deviations from ideality as a function of composition and temperature may be ascribed to the presence of strong or weak interactions between unlike molecules. Ultrasonic velocity is related to the binding forces between the atoms or the molecules and has been adequately employed in understanding the nature of molecular interaction in pure liquids, binary and ternary mixtures. Many studies have been done to find out the interactions in binary and ternary liquid mixtures by means of acoustic properties, viscometric and thermo dynamical properties of the ternary system²⁻⁷. The present investigation deals with three important liquids namely n-hexane, crotonaldehyde and iodine. The liquids under investigation are very useful chemicals and of industrial significance. Hexane is used in industrial manufacture of benzene, preparation of higher fatty acids and corresponding nitroalkanes, haloalkanes, sulphonic acids and lower alkanes. The variation of ultrasonic velocity and related parameters throw much light upon the structural changes associated with the liquid mixtures having weakly interacting components as well as strongly interacting components⁸. The ultrasonic velocity (U), viscosity (η), internal pressure (π_i), adiabatic compressibility (κ) and free volume (V_f) throws light on the nature and degree of molecular interaction occurring in the ternary system. Hence, the derived parameters listed above have been obtained to explore the interactions⁹. In the present investigation the ultrasonic velocity (U) studies and allied parameters of ternary mixtures of

crotonaldehyde with iodine in n-hexane at 293.15, 298.15, 303.15 and 308.15K over the entire range of concentrations are determined.

EXPERIMENTAL

In the present work, the densities (ρ) and Ultrasonic velocities (U) of ternary mixtures of crotonaldehyde with iodine in n-hexane at 293.15, 298.15, 303.15 and 308.15K over the entire composition range are measured in different concentrations. The solvent has taken as accurately weighted amount of sample is dissolved in suitable solvent to obtain solution in the concentration range 0.001M – 0.01M. The ultrasonic velocity (U) has been measured by ultrasonic interferometer (Model F81 Mittal enterprises, New Delhi) at a frequency of 2MHz with an accuracy of ± 0.1 . The chemicals used in the present work were analytical reagent (AR) and spectroscopic reagent (SR) grade with minimum assay of 99.9 % were obtained from SD Fine Chemicals, India and E-Merck, Germany. The temperatures are controlled by circulating water around the liquid cell from a thermostatically controlled water bath (accuracy $\pm 0.1^\circ\text{C}$).

The various physical parameters are calculated from the measured values of density (ρ), viscosity (η) and ultrasonic velocity (U). Using standard formula the following acoustical parameters have been calculated:

$$\text{Adiabatic compressibility } (\kappa) = 1/U^2 \rho \quad (1)$$

Intermolecular free length (L_f) has been calculated from the relation,

$$L_f = K/\sqrt{U\rho} \text{ \AA} \quad (2)$$

Where K is a temperature dependent constant.

These two parameters basically depend on the interaction phenomenon and they represent the strength of the interaction present in the component molecules¹⁰⁻¹¹. Free volume (V_f) has been calculated from relation,

$$V_f = (M_{\text{eff}} U / K\eta)^{3/2} \quad (3)$$

Where M_{eff} is the effective molecular weight ($M_{\text{eff}} = \sum m_i x_i$, in which m_i and x_i are the molecular weight and the mole fraction of the individual constituents respectively).

The following equation (4) is used to compute internal pressure (π_i).

$$\pi_i = bRT (K\eta/U)^{1/2} (\rho^{2/3}/M_{\text{eff}}^{7/6}) \quad (4)$$

The acoustic impedance can be related as

$$Z = U\rho \quad (5)$$

Other acoustical parameters such as available volume (v_a), relaxation time (τ), interaction parameter (χ_i), cohesive energy (CE) and Lenord Jones Potential (LJP) are calculated using standard equation formula.

Table-1: Ultrasonic velocity u , density ρ , viscosity η , adiabatic compressibility κ , free length L_f , free volume V_f , internal pressure π_i , molar volume (V_m) cohesive energy and available volume (V_a) for equimolar concentration C of crotonaldehyde with iodine in n-hexane solutions at 293.15K, 298.15K, 303.15K and 308.15K

Conc.	Density (ρ) (Kgm^{-3})				Viscosity(η) (10^{-3}Nsm^{-2})				Velocity(U) (ms^{-1})			
	293.15K	298.15K	303.15K	308.15K	293.15K	298.15K	303.15K	308.15K	293.15K	298.15K	303.15K	308.15K
0.001	669.2	668.1	664.3	663.2	0.5108	0.5031	0.5043	0.5043	1098.5	1077.4	1058.3	1066.3
0.002	668.9	666.4	663.4	663.5	0.5202	0.5151	0.4946	0.4891	1098.0	1076.8	1057.3	1064.9
0.003	669.0	666.3	664.5	661.6	0.5097	0.5119	0.5032	0.4887	1100.1	1078.8	1057.2	1065.4
0.004	670.0	668.0	664.9	662.9	0.5073	0.4931	0.4969	0.4924	1102.0	1076.1	1056.7	1065.9
0.005	670.2	666.0	665.4	662.6	0.5115	0.5018	0.5039	0.5008	1103.4	1075.7	1056.6	1069.0
0.006	668.1	665.7	664.4	663.9	0.5127	0.4997	0.4944	0.4919	1100.4	1076.8	1057.5	1069.1
0.007	672.7	667.1	665.5	661.6	0.5122	0.5126	0.5909	0.4865	1101.3	1075.5	1056.7	1065.8
0.008	669.3	667.9	664.7	664.1	0.5031	0.5148	0.4959	0.4843	1100.5	1072.7	1055.5	1064.9
0.009	669.5	669.2	665.5	663.7	0.5069	0.5011	0.4974	0.4813	1099.3	1077.6	1054.4	1065.9
0.010	670.7	669.0	664.8	663.9	0.5138	0.5009	0.4756	0.5027	1098.9	1073.6	1055.2	1060.1

Conc.	Adiabatic compressibility(κ) ($10^{-10} \text{ Kg}^{-1}\text{ms}^2$)				Free length (L_f) (10^{-10}m)				Free Volume(V_f) ($10^{-7} \text{ m}^3 \text{ mol}^{-1}$)			
	0.001	12.384	12.895	13.441	13.263	0.7038	0.7182	0.7332	0.7284	2.851	2.833	2.748
0.002	12.400	12.942	13.485	13.291	0.7043	0.7195	0.7344	0.7291	2.773	2.732	2.826	2.904
0.003	12.351	12.896	13.465	13.317	0.7029	0.7182	0.7339	0.7298	2.868	2.767	2.755	2.912
0.004	12.290	12.928	13.468	13.277	0.7012	0.7191	0.7340	0.7288	2.893	2.913	2.802	2.878
0.005	12.255	12.976	13.462	13.207	0.7002	0.7204	0.7338	0.7268	2.867	2.841	2.749	2.823
0.006	12.361	12.955	13.459	13.178	0.7032	0.7199	0.7337	0.7260	2.847	2.864	2.832	2.901
0.007	12.257	12.959	13.457	13.307	0.7002	0.7200	0.7337	0.7296	2.856	2.753	2.166	2.937
0.008	12.337	13.012	13.503	13.279	0.7025	0.7214	0.7349	0.7288	2.932	2.726	2.813	2.954
0.009	12.360	12.869	13.516	13.261	0.7031	0.7175	0.7353	0.7283	2.894	2.858	2.797	2.987
0.010	12.347	12.968	13.510	13.403	0.7028	0.7202	0.7351	0.7322	2.836	2.844	2.997	2.776
Conc.	Internal pressure (π_i) 10^8 atm				Molar Volume(V_m) $10^{-4} \text{ m}^3 \text{ mol}^{-1}$				Available volume(V_a) 10^{-5}			
	0.001	2.902	2.905	2.924	2.910	1.2881	1.2902	1.2977	1.2999	4.037	4.214	4.393
0.002	2.928	2.935	2.894	2.867	1.2889	1.2938	1.2997	1.2995	4.044	4.231	4.409	4.346
0.003	2.895	2.922	2.921	2.859	1.2891	1.2943	1.2978	1.3035	4.028	4.216	4.403	4.355
0.004	2.891	2.878	2.907	2.875	1.2864	1.2902	1.2962	1.3001	4.004	4.225	4.401	4.340
0.005	2.898	2.895	2.925	2.891	1.2874	1.2955	1.2967	1.3022	3.996	4.245	4.404	4.322
0.006	2.898	2.886	2.893	2.868	1.2917	1.2964	1.2989	1.2998	4.033	4.239	4.404	4.313
0.007	2.908	2.928	3.166	2.849	1.2832	1.2939	1.2971	1.3047	3.999	4.242	4.404	4.356
0.008	2.873	2.939	2.899	2.850	1.2899	1.2927	1.2989	1.3002	4.027	4.260	4.420	4.348
0.009	2.885	2.896	2.906	2.838	1.2899	1.2905	1.2977	1.3012	4.037	4.213	4.425	4.343
0.010	2.908	2.900	2.838	2.909	1.2879	1.2912	1.2993	1.3011	4.033	4.248	4.424	4.390

RESULTS AND DISCUSSIONS

The experimentally determined values of the density (ρ), ultrasonic velocity (U), adiabatic compressibility (κ), intermolecular free length (L_f), free volume (V_f) and internal pressure (π_i) and other acoustical parameters for the ternary liquid systems at 293.15, 298.15, 303.15 and 308.15K are reported in Table-1. In the present investigation, ultrasonic velocity measurement is used to assess molecular interaction between crotonaldehyde with iodine in n-hexane. In all the mixtures, the ultrasonic velocity (U) decreases with increase in concentration and a sudden increase at a concentration of 0.007M at 293.15K. This behavior at such concentrations is different from the ideal mixtures behavior can be attributed to intermolecular interactions in the systems studied¹²⁻¹³.

Table-2: Impedence(Z), Relaxation time(ζ), Interaction parameter(χ_i), Lenord Jones Potential(LJP), Cohesive Energy(CE) and free energy of activation (ΔG^*)for equimolar concentration C of crotonaldehyde with iodine in n-hexane solutions at 293.15K, 298.15K, 303.15K and 308.15K

Conc.	Impedence(Z) $10^5 \text{ Kg}^{-1} \text{ m}^2 \text{ s}^{-1}$				Relaxation time(ζ) 10^{-13}S				Interaction parameter(χ_i)			
	293.15K	298.15K	303.15K	308.15K	293.15K	298.15K	303.15K	308.15K	293.15K	298.15K	303.15K	308.15K
0.001	7.351	7.198	7.030	7.071	8.434	8.649	9.037	8.919	0.0429	0.0032	-0.0320	-0.0174
0.002	7.345	7.176	7.014	7.065	8.601	8.889	8.893	8.668	0.0418	0.0019	-0.0340	-0.0201
0.003	7.360	7.188	7.025	7.048	8.394	8.803	9.033	8.676	0.0456	0.0055	-0.0343	-0.0193
0.004	7.383	7.188	7.026	7.066	8.314	8.499	8.925	8.717	0.0496	0.0009	-0.0349	-0.0180
0.005	7.395	7.164	7.031	7.083	8.359	8.682	9.044	8.819	0.0516	-0.0005	-0.0357	-0.0129
0.006	7.352	7.168	7.026	7.098	8.451	8.632	8.873	8.643	0.0457	0.0014	-0.0342	-0.0129
0.007	7.408	7.175	7.032	7.051	8.370	8.857	10.60	8.632	0.0473	-0.0012	-0.0358	-0.0191
0.008	7.366	7.165	7.016	7.071	8.275	8.930	8.929	8.576	0.0456	-0.0065	-0.0381	-0.0209
0.009	7.360	7.211	7.017	7.074	8.355	8.598	8.964	8.509	0.0432	0.0024	-0.0403	-0.0192
0.010	7.370	7.182	7.015	7.038	8.458	8.662	8.566	8.984	0.0423	-0.0051	-0.0390	-0.0300
	Lenord Jones Potential (LJP)				Cohesive Energy(CE) 10^4				free energy of activation (ΔG^*)			

Conc.					$10^{-19} \text{KJ mol}^{-1}$							
0.001	6.143	5.370	4.722	4.988	3.739	3.749	3.794	3.782	3.8652	3.9315	3.9986	3.7696
0.002	6.124	5.349	4.689	4.941	3.774	3.797	3.761	3.726	3.8660	3.9327	3.9980	3.7684
0.003	6.204	5.419	4.686	4.957	3.732	3.782	3.791	3.727	3.8650	3.9323	3.9986	3.7684
0.004	6.277	5.324	4.670	4.974	3.719	3.714	3.768	3.738	3.8647	3.9308	3.9981	3.7686
0.005	6.331	5.310	4.667	5.079	3.731	3.750	3.793	3.764	3.8649	3.9317	3.9987	3.7691
0.006	6.215	5.349	4.696	5.083	3.744	3.741	3.757	3.728	3.8653	3.9315	3.9979	3.7683
0.007	6.250	5.303	4.670	4.971	3.732	3.788	4.107	3.718	3.8649	3.9325	4.0053	3.7682
0.008	6.219	5.206	4.631	4.941	3.707	3.799	3.765	3.706	3.8645	3.9329	3.9981	3.7679
0.009	6.173	5.377	4.595	4.974	3.722	3.738	3.772	3.693	3.8649	3.9313	3.9983	3.7676
0.010	6.158	5.237	4.621	4.781	3.746	3.744	3.688	3.784	3.8654	3.9316	3.9965	3.7699

However, the value of compressibility (κ) shows an inverse behavior as compared to the ultrasonic velocity (U). The adiabatic compressibility (κ) increases with increase of concentration and a sudden decrease at 0.007M at 293.15K. It is primarily the compressibility that increases due to structural changes of molecules in the mixture leading to a decrease in ultrasonic velocity¹⁴⁻¹⁶. Ultrasonic velocity decreases with increase in temperature up to 303.15K and it suddenly decreases at higher temperature of 308.15K. At 308.15K, ultrasonic velocity is maximum at the concentration of 0.005M and 0.006M and after that, it gradually decreases. Such a continuous increase in adiabatic compressibility with the increase of molar concentration is due to the effect of hydrogen bonding or dipole-dipole interactions and it indicates significant interaction between crotonaldehyde and iodine forming dipole-dipole interactions. The intermolecular free length (L_f) is a predominant factor in determining existing interactions among the components of the mixture. The parameter basically depends on the interaction phenomenon and they represent the strength of the interaction present in the component molecules¹⁷⁻¹⁸.

Analyzing the respective Tables 1 and 2, it is noticed that the free length (L_f) decreases in the liquid systems with the increasing concentration of crotonaldehyde. Free length is increasing at the concentration of 0.005M at 293.15K. But, a sudden increase at 0.006M and again it increases with concentration. The additions of interacting molecules break up the molecular structure and releasing several dipoles for interaction. This is due to the addition of interacting molecules; there will be an increase of cohesive energy hence the free length (L_f) increases in the mixture after mixing.

Table-3: Excess velocity U^E , excess compressibility κ^E , excess free length L_f^E and excess molar volume V_m^E for equimolar concentration of crotonaldehyde and iodine in n-hexane at 303.15 K

Conc	U^E/ms^{-1}	$\kappa^E/10^{-11} \text{Kg}^{-1} \text{ms}^2$	$L_f^E/10^{-12} \text{m}$	$V_m^E/10^{-4} \text{m}^3 \text{mol}^{-1}$
0.001	-17.4	6.238	1.765	-2.224
0.002	-18.5	6.699	1.893	-2.221
0.003	-18.6	6.521	1.848	-2.222
0.004	-18.9	6.495	1.833	-2.225
0.005	-19.4	6.539	1.858	-2.223
0.006	-18.6	6.537	1.861	-2.220
0.007	-19.4	6.538	1.864	-2.221
0.008	-20.7	7.026	2.000	-2.219
0.009	-21.9	7.173	2.043	-2.219
0.010	-21.2	7.134	2.035	-2.217

Similarly, free length is maximum at 298.15K and 303.15K and it is decreased with further increase in concentration and it releases several dipoles for interaction. In view of greater force of interaction

between the crotonaldehyde and iodine, there will be an increase in cohesive energy and hence an increase in free length (L_f) is observed. Similar observation is noticed by earlier workers¹⁹. Free volume and the internal pressure shows reverse trend at 0.007M.

The internal pressure (π_i) is the resultant of force of attraction and force of repulsion per unit area between the components. In this liquid systems, the internal Pressure (π_i) increases at the concentration of 0.007M at 303.15 and 308.15K.

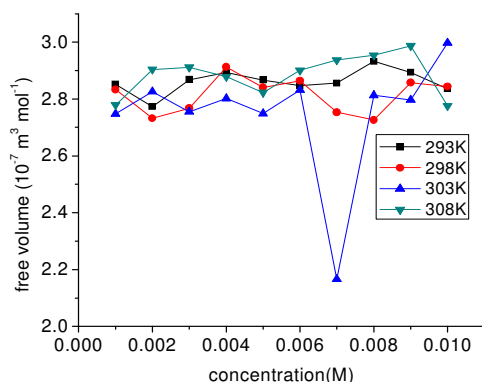


Fig.-1: Free volume Vs Concentration

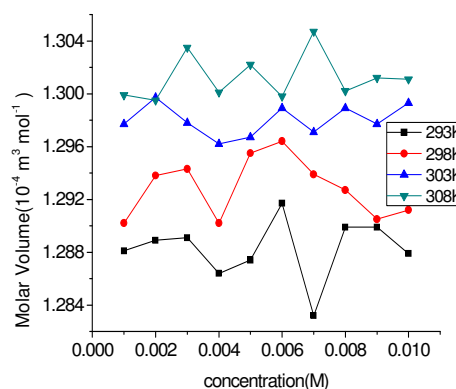


Fig.-2: Molar volume Vs Concentration

The free volume decreases with increase in concentration of crotonaldehyde with iodine in n-hexane mixtures as shown in Figure-1. This is due to the intermolecular association between the solvent and solute with information of bigger molecules. At all the concentrations, due to increase in temperature, the internal pressure increases as in decrease of free volume. It is characterized by structure-formation due to the presence of ions of greater charge. The increase in free volume with temperature is because the effect of temperature is to disturb the equilibrium of H-bonded structure of molecules. This suggests that the close packing of molecules inside the Shield, which may brought about by the increasing magnitude of interactions²⁰⁻²¹. The acoustical impedance (Z) is another parameter, is the product of velocity and density of mixtures. Further, in the liquid systems, the value of acoustic impedance (Z) is found to be decreased, which are listed in Table-2.

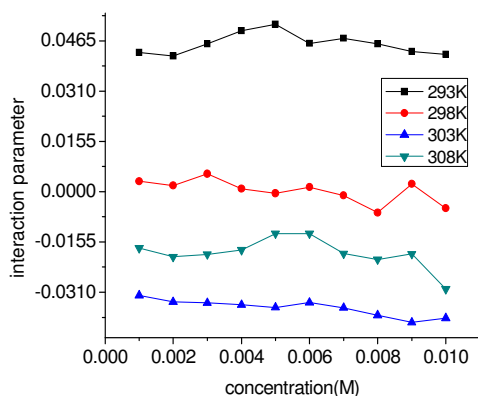


Fig.-3: Interaction parameter Vs Concentration

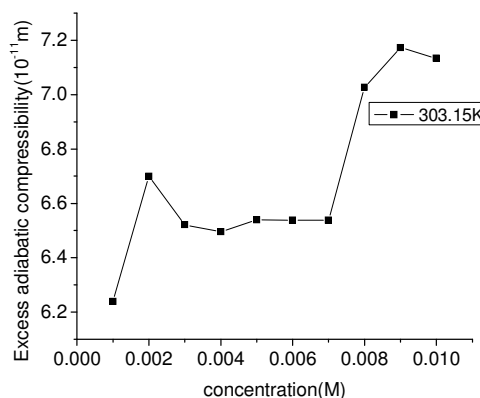
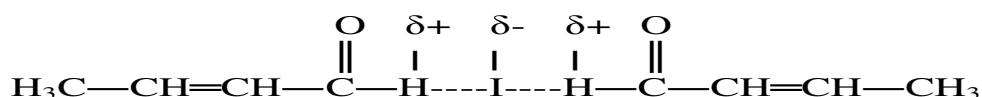


Fig.-4: Excess Vadiabatic compressibility Vs Concentration

When an acoustic wave travels in a medium, there is a variation of pressure from particle to particle. The ratio of the instantaneous pressure excess at any particle of the medium to the instantaneous velocity of that particle is known as 'specific acoustic impedance' of the medium. This factor is governed by the inertial and elastic properties of the medium. It is important to examine specific acoustic impedance in relation to concentration and temperature. When a plane ultrasonic wave is set up in a liquid, the pressure and hence density and refractive index show specific variations with distance from the source along the direction of propagation. In the present investigation, it is observed that the acoustic impedance (Z) increase with increasing concentration of crotonaldehyde and it is maximum at 0.005M at 298.15K, while it is minimum at 0.006M. Such a decrease in acoustic impedance (Z) is further supports the possibility of strong molecular interactions due to dipole-dipole interaction between iodine and crotonaldehyde. When the temperature is increased, the acoustic impedance is decreasing. It is increased at 308.15K. This suggests that the presence of ion-solvent interaction in the systems. This report supports our view that ion-solvent interaction increases with increase in concentration in the solvent mixtures. The free energy of activation (ΔG^*) and relaxation time (τ) are intrinsic properties of charge transfer complex²². It increases with temperature up to 303.15K and it is minimum at 308.15K. From the Figure 2, the molar volume (V_m) of the components is studied. It increases with increase in concentration.

A perusal on the magnitude of LJP indicates that it is in the range of hydrogen bonding type of interaction.



The values of available volume increases with increase in concentration. The interstitial accommodation because of differences between the molecular sizes of the mixing components might be another factor behind the interaction observed. The relaxation time (τ) shows the decreasing trend. Figure 3 shows the interaction parameter (χ_i) are found to be negative which strongly confirm the presence of weak between the unlike molecules at 303.15K and 308.15K.

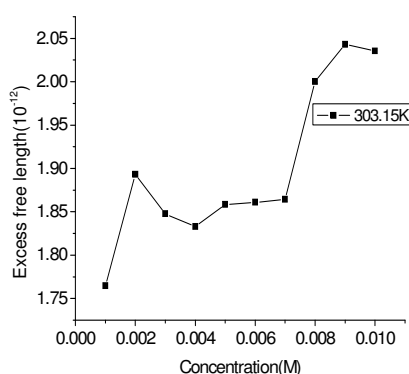


Fig.-5: Excess free length Vs Concentration

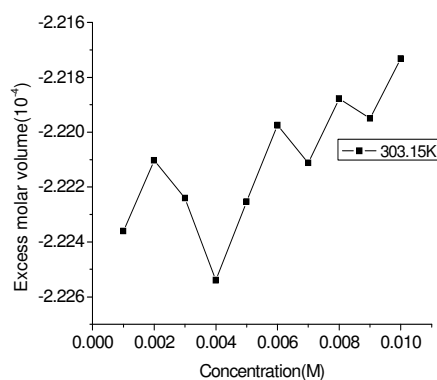


Fig.-6: Free volume Vs Excess molar volume

Excess Parameters

The thermodynamic excess functions are found to be very sensitive towards mutual interactions between the component molecules of the liquid mixtures. The sign and the extent of deviation of these functions from ideality depend on the strength of interactions between unlike molecules. The excess functions are calculated by using the following general equation,

$$Y^E = Y - [(1-x)Y_1 + xY_2]$$

Where 'Y' represents the acoustical parameter and 'x' be the mole fraction of solute

The deviation of acoustical parameters from their ideal behavior is calculated and the excess values for velocity (u^E), compressibility (κ^E), free length (L_f^E) and molar volume (V_m^E) are listed in Table 3 for the liquid mixtures at 303.15K. The values of excess velocity are negative through the whole range all the concentration. The plot of excess velocity (u^E) shows the non linear trend with concentration in all the liquid mixtures which reveals that there is some specific interactions between components of the mixtures. The intermolecular interactions and the formation of CT complexes are evident from the negative deviation of u^E at specific concentrations in the systems. The change of sign of u^E is an evidence for the lesser magnitude of interactions between solute molecules. The disruption of dipolar association of like molecules and the existence of strong solute – solvent interactions may be the reason for the negative deviation in ultrasonic velocity.

Figure-4 shows the variation of excess compressibility (κ^E) for the ternary systems. The sign and magnitude of κ^E is used to explain the intermolecular interaction. A positive value of κ^E indicates weak interaction between the components of the mixtures. When the strength of interaction between unlike molecules decreases, κ^E becomes increasingly positive. From the Figure 5, the variation producing in the excess free length (L_f^E) is observed. The positive value of L_f^E indicates that sound waves cover short distances due to increase in intermolecular free length describing the dominant nature of dispersive forces between unlike molecules. The positive excess values of free length should be attributed to the dispersive forces is indicated by Fort and Moore²³.

The negative excess values should be due to charge transfer and hydrogen bond formation. In the present investigation, the positive contribution of L_f^E in this system prevails the existence of weak interaction. The values of κ^E and L_f^E shows an exact reverse trend as that of u^E . In the present study, the excess adiabatic compressibility and excess free lengths values which are to be increasing values over the whole composition ranges for all concentration of a mixture and it shows the weaker interaction between the components of the mixtures. The dissociation of like molecular attraction and the existence of weak interactions lead to the positive contribution to these excess functions. The excess molar volume (V_m^E) values which are furnished in Table-3 whose negative deviation clearly supports the existence of specific interactions as shown in Figure 6. The comparison between the free volume (V_f) and the internal pressure shows the reverse trend and it will be maximum at 0.007M. It is also found to be increased with increasing concentration and leads to expansion of volume. It indicates dipole-dipole interaction at all the concentrations between the unlike molecules.

CONCLUSION

From the above observations, it may be concluded that the existence of intermolecular interaction. The free length (L_f) increases with increase in the concentration of solute at 303.15 and 308.15K indicating that there is a weak solute-solvent interaction. This may also imply the increase in number of free ions, showing the occurrence of ionic dissociation due to weak solute-solute interaction, while the free length (L_f) in the percentage of organic solvent indicating solute-solvent interaction. This may be due to the decrease in number of free ions, showing the occurrence of ionic association due to solute-solvent interaction.

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