

Ultrasonic Studies Of Aromatic Amine With Ketone In Nonpolar Solvent at 303.15, 308.15 and 313.15K

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Abstract

The ultrasonic velocity (U), density (ρ) and viscosity (η) measurements were carried out for the three organic ternary mixtures of aniline+1, 4-dioxan+propanone, aniline+1, 4-dioxan+butanone and aniline+1, 4-dioxan+Methyl Iso Butyl Ketone at 303.15, 308.15 and 313.15 K. Thermodynamic parameters namely adiabatic compressibility (β), free length (L_F), free volume (V_F), internal pressure (π_i), Gibbs free energy (ΔG) were evaluated. In addition, excess values of certain above parameters were also calculated. The above parameters were used to discuss the presence of significant interactions between the component molecules in the ternary mixtures. Observed excess values in the mixture indicate the existence of strong interactions in the system.

Keywords: adiabatic compressibility, free length, free volume, internal pressure, Gibbs free energy, excess parameters.

INTRODUCTION

The study of molecular interaction plays a vital role in the development of molecular science. Molecular interactions and structural behavior of molecules and their mixtures can be identified using ultrasonic studies. Ultrasonic waves have been used by many scientists to investigate the nature of molecular interactions and physico-chemical behaviors of pure, binary and ternary liquid mixtures^{1, 2}. Ultrasonic velocity together with density and viscosity data furnish wealth of information about the interaction between ions, dipoles, hydrogen bonding, multipolar and dispersive forces^{3, 4}. Ultrasonic investigations of liquid mixtures are of considerable importance in understanding the intermolecular interactions between the component molecules and find applications in several industrial and technological processes⁵. The nature and rela-

tive strength of the molecular interaction between the components of the liquid mixtures have been successfully investigated by the ultrasonic method⁶.

Although a large number of investigations were carried in liquid mixtures having ketone as one of the components, it is found that no work has done so far to measure the ultrasonic velocity in ternary liquid mixtures of aniline+1, 4-dioxan+propanone,+butanone and+Methyl Iso Butyl Ketone. Aniline molecule is highly polar and self associated through hydrogen bonding of their amine group. Being aromatic, aniline with amino group is comparatively a strong electron donor. The H atoms in the NH₂ group can also play the role of electron-acceptors centers⁷. It is used in the manufacturing of synthetic dyes, drugs and as an accelerator in vulcanization of rubber⁸. Propanone, Butanone and Iso butyl methyl ketone (MIBK) are all aprotic in nature and belong to the same ketone homologous series. Especially they have aliphatic nature and have common carbonyl (C=O) functional group⁹. 1, 4-dioxane is gaining increasing interest and concern in the environmental community. It is a newly regulated compound, it may be present above regulatory levels at existing chlorinated solvent sites, it was used as a stabilizer for solvents such as 1, 1, 1-TCA, typically added ~2% by weight and 1, 4-Dioxane may be concentrated in degreasing waste residuals, especially in degreasers equipped with distillation units.

For a better understanding of the physico-chemical properties and the molecular interactions between the participating components of these mixtures ultrasonic velocities and densities are measured at 303.15K, 308.15K and 313.15K over the entire concentration range for three ternary mixture systems as follows.

System I : Aniline+1, 4-dioxan+propanone

System II : Aniline+1, 4-dioxan+butanone

System III: Aniline+1, 4-dioxan+Methyl IsoButyl Ketone.

MATERIALS AND METHODS

All the chemicals used in the present work were of High purity Analytical reagent (AR) grades. In all the systems, the ternary liquid mixtures were prepared in terms of mole fraction, out of which the mole fraction of the second compound, 1, 4 – dioxan was kept fixed ($X_2 = 0.4$) while the remaining two (X_1 and X_3) were varied from 0.0 to 0.6. The ultrasonic velocity in the liquid mixtures was measured using a single crystal ultrasonic interferometer with an operating frequency of 3MHz supplied by M/s. Mittal Enterprises, New Delhi. The densities of pure liquid and liquid mixtures were determined using a 10 ml specific gravity bottle calibrated with double distilled water and acetone. Densities of the mixtures were ascertained by relative measurement method. An Ostwald's viscometer was used for the viscosity measurements of all the compounds. The flow time of liquid mixtures were measured by a digital stop clock with an accuracy of 0.01s. An electronically operated constant temperature water bath was used to circulate water through the double walled measuring cell made up of steel containing the experimental solution at the desired temperature.

THEORY AND CALCULATIONS

Using the measured values of density (ρ), viscosity (η) and velocity (U), the acoustical parameters such as adiabatic compressibility (β), free length (L_F), free volume (V_F), internal pressure (π_i), and Gibb's free energy (ΔG) have been evaluated using the following standard expressions¹⁰.

Intermolecular free length (L_F) was calculated using the expression

$$L_F = K \beta^{1/2} \quad (1)$$

Where K is a temperature dependent constant known as Jacobson constant and β is the adiabatic compressibility that can be calculated from the velocity of sound (U) and the density of the medium (ρ) as

$$\beta = \frac{1}{U^2 \rho} \quad (2)$$

The relation for free volume in terms of ultrasonic velocity and the viscosity (η) of the liquid as

$$V_F = \left(\frac{M_{eff} U}{\eta K} \right)^{3/2} \quad (3)$$

Expression for the determination of internal pressure by the use of free volume as

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

Where b stands for cubic packing which is assumed to be 2 for liquids and k is a dimensionless constant independent of temperature and nature of liquids and its value is 4.281×10^9 , T is the absolute temperature and M_{eff} is the effective molecular weight.

The Gibbs free energy of activation flow in the mixtures can be calculated from the relation

$$\Delta G = -KT \log (h/KT\tau) \quad (5)$$

Where K is Boltzmann's constant.

Relaxation time can be calculated using viscosity and adiabatic compressibility

$$\tau = (4/3) \eta \beta \quad (6)$$

Excess parameter (A^E) has been calculated from the relation

$$A^E = A_{\text{exp}} - A_{\text{id}} \quad (7)$$

Where, $A_{\text{id}} = \sum_{i=1}^n A_i X_i$, A_i is any acoustical parameter and X_i the mole fraction of the liquid component.

RESULT AND DISCUSSION

The experimentally measured values of ultrasonic velocity, density and viscosity for pure liquids and the liquid mixtures at 303, 308 and 313K were listed in the Table-1 and Table-2 respectively for the ternary liquid mixtures of aniline+1, 4-dioxan+propanone,+butanone and+Methyl Iso Butyl Ketone. The computed values of adiabatic Compressibility, free length and free volume for all the mixtures were depicted in Table 3. Table 4 represents the calculated values of internal pressure and Gibbs free energy for the mixtures. The computed values of excess adiabatic compressibility, excess free volume, excess free length and excess internal pressure for liquid mixtures were represented in fig.1, fig.2 and fig.3 respectively.

Table.1 Values of density (ρ), viscosity (η) and ultrasonic velocity (U) of pure liquids at 303.15, 308.15 and 313.15 K

Organic liquids	$\rho \text{ Kg m}^{-3}$			$\eta \times 10^3 \text{ N s m}^{-2}$			$U \text{ ms}^{-1}$		
	303.15 K	308.15 K	313.15 K	303.15 K	308.15 K	313.15 K	303.15 K	308.15 K	313.15 K
aniline	1013	1008	1004	3.3070	2.6150	2.4430	1620	1604	1582
1, 4-dioxan	1018	1014	1007	1.0370	0.9348	0.885	1312	1282	1268
propanone	774.2	764.0	761.5	0.3573	0.3320	0.3138	1120	1103	1091
butanone	794.2	785.1	780.7	0.3989	0.3728	0.3545	1173	1146	1125
Methyl Iso Butyl Ketone	789.5	785.5	780.5	0.5316	0.5000	0.4837	1176	1149	1118

Table.2 Values of density (ρ), viscosity (η) and ultrasonic velocity (U) of ternary liquid mixtures at 303.15, 308.15 and 313.15 K

Mole fraction		ρ Kgm ⁻³			$\eta \times 10^3$ Nsm ⁻²			U ms ⁻¹		
X ₁	X ₃	303.15K	308.15K	313.15K	303.15K	308.15K	313.15K	303.15K	308.15K	313.15K
Aniline+1, 4-dioxan+propanone										
0.5990	0.0000	1021.95	1020.74	1015.6	2.6380	2.3514	2.0644	1522	1510	1475
0.4990	0.1000	1003.11	999.9	995.93	1.5767	1.4798	1.3952	1499	1464	1449
0.3997	0.1999	982.69	979.45	975.48	1.2187	1.1487	1.0584	1432	1415	1391
0.3004	0.2991	961.88	959.78	953.85	0.9432	0.8978	0.8515	1373	1362	1351
0.1999	0.4000	937.94	936.17	931.43	0.7304	0.6797	0.6396	1328	1308	1293
0.0999	0.4999	913.96	909.82	901.53	0.6063	0.5589	0.5324	1270	1252	1223
0.0000	0.6000	884.15	879.14	870.06	0.4973	0.4664	0.4421	1207	1194	1170
Aniline+1, 4-dioxan+butanone										
0.5999	0.0000	1021.95	1020.74	1015.6	2.6380	2.3514	2.0644	1522	1510	1475
0.5001	0.1000	1000.36	997.93	992.78	1.5724	1.449	1.2953	1470	1446	1428
0.3999	0.2003	979.16	976.69	971.15	1.2567	1.1454	1.0537	1413	1409	1388
0.2999	0.3001	955.21	951.52	947.55	0.9366	0.8635	0.8069	1379	1359	1336
0.2000	0.3999	933.61	929.09	924.34	0.8015	0.7325	0.6954	1321	1312	1287
0.0999	0.5001	905.74	901.56	897.99	0.6379	0.603	0.5727	1271	1253	1225
0.0000	0.6162	879.83	877.17	869.67	0.5472	0.5182	0.486	1200	1181	1169
Aniline+1, 4-dioxan+Methyl IsoButyl Ketone										
0.5999	0.0000	1021.9	1020.7	1015.6	2.6380	2.3514	2.0644	1522	1510	1475
0.5000	0.10011	991.19	987.50	983.46	1.5723	1.5028	1.3372	1450	1435	1413
0.3999	0.1998	936.60	960.27	956.62	1.3618	1.2066	1.1299	1397	1379	1359
0.3001	0.2999	936.41	933.04	928.96	1.0803	0.9900	0.9058	1349	1333	1314
0.2000	0.4001	910.45	907.04	904.15	0.9191	0.8231	0.7823	1305	1284	1260
0.0999	0.5002	886.10	882.65	877.30	0.7455	0.6824	0.6471	1257	1233	1212
0.0000	0.6001	862.17	858.68	854.12	0.6414	0.5994	0.5605	1207	1199	1175

Table.3 Values of Adiabatic compressibility (β), Free length (LF) and Free Volume (V_f) of ternary liquid mixtures at 303.15, 308.15 and 313.15 K

Mole fraction		$\beta \times 10^{10} \text{ Pa}^{-1}$			$L_f \times 10^{10} \text{ m}$			$V_f \times 10^7 \text{ m}^3 \text{ mol}^{-1}$		
X1	X3	303.15K	308.15K	313.15K	303.15K	308.15K	313.15K	303.15K	308.15K	313.15K
Aniline+1, 4-dioxan+propanone										
0.5990	0.0000	4.2242	4.2989	4.5282	0.4101	0.4170	0.4320	0.4295	0.5041	0.5916
0.4990	0.1000	4.4354	4.6615	4.7791	0.4202	0.4342	0.4438	0.8582	0.9115	0.9801
0.3997	0.1999	4.9604	5.0927	5.2959	0.4444	0.4539	0.4672	1.1094	1.1916	1.3123
0.3004	0.2991	5.5172	5.6149	5.7457	0.4687	0.4766	0.4866	1.4343	1.5268	1.6323
0.1999	0.4000	6.0451	6.2465	6.4228	0.4906	0.5027	0.5145	1.8739	2.0394	2.1966
0.0999	0.4999	6.7741	7.0084	7.4123	0.5193	0.5324	0.5527	2.1634	2.3909	2.4828
0.0000	0.6000	7.7603	7.9819	8.3926	0.5558	0.5682	0.5881	2.5061	2.7133	2.8535
Aniline+1, 4-dioxan+butanone										
0.5999	0.0000	4.2242	4.2989	4.5282	0.4101	0.4170	0.4320	0.4302	0.5050	0.5926
0.5001	0.1000	4.6254	4.7912	4.9382	0.4291	0.4402	0.4511	0.8569	0.9452	1.0975
0.3999	0.2003	5.1180	5.1529	5.3448	0.4514	0.4565	0.4694	1.0903	1.2490	1.3831
0.2999	0.3001	5.5217	5.6837	5.9082	0.4689	0.4795	0.4935	1.5761	1.7422	1.8793
0.2000	0.3999	6.0139	6.2489	6.5345	0.4944	0.5028	0.5190	1.7961	2.0362	2.1369
0.0999	0.5001	6.8334	7.0637	7.4257	0.5216	0.5345	0.5532	2.2978	2.4473	2.5544
0.0000	0.6162	7.8889	8.1625	8.4071	0.5605	0.5746	0.5887	2.6545	2.8141	3.0497
Aniline+1, 4-dioxan+Methyl IsoButyl Ketone										
0.5999	0.0000	4.2242	4.2989	4.5282	0.4101	0.4170	0.4320	0.4295	0.5050	0.5926
0.5000	0.10011	4.7965	4.9115	5.0928	0.4370	0.4457	0.4582	0.8797	0.9274	1.0786
0.3999	0.1998	5.3175	5.4714	5.6634	0.4601	0.4704	0.4831	1.0436	1.2279	1.2000
0.3001	0.2999	5.8726	6.0263	6.2318	0.4835	0.4937	0.5068	1.4167	1.5882	1.7754
0.2000	0.4001	6.4534	6.6789	6.9632	0.5069	0.5198	0.5357	1.7372	2.0033	2.1005
0.0999	0.5002	7.1436	7.4558	7.7559	0.5333	0.5492	0.5654	2.2740	2.5220	2.6636
0.0000	0.6001	7.9628	8.1076	8.4787	0.5630	0.5727	0.5912	2.7110	2.9696	3.1884

Table.4 Values of Internal pressure (π_i) and Gibb's Free Energy (ΔG) of ternary liquid mixtures at 303.15, 308.15 and 313.15 K

Mole fraction		$\pi_i \times 10^{-6}$ Pa			$\Delta G^* \times 10^{20}$ KJmol ⁻¹		
X1	X3	303.15K	308.15K	313.15K	303.15K	308.15K	313.15K
Aniline+1, 4-dioxan+propanone							
0.5990	0.0000	721.52	694.73	667.08	0.9359	0.9169	0.9050
0.4990	0.1000	580.32	576.94	570.76	0.7411	0.7545	0.7590
0.3997	0.1999	539.96	534.77	524.82	0.6802	0.6844	0.6840
0.3004	0.2991	502.67	499.71	494.59	0.6175	0.6212	0.6253
0.1999	0.4000	466.44	460.37	454.18	0.5488	0.5482	0.5498
0.0999	0.4999	450.09	441.18	440.05	0.5186	0.5139	0.5325
0.0000	0.6000	433.75	427.05	423.81	0.4925	0.4923	0.5058
Aniline+1, 4-dioxan+butanone							
0.5999	0.000	721.52	694.73	667.08	0.9359	0.9169	0.9050
0.5001	0.1000	558.07	563.28	542.74	0.7575	0.7572	0.7411
0.3999	0.2003	515.59	514.16	502.99	0.7061	0.6882	0.6861
0.2999	0.3001	455.54	459.62	454.17	0.6149	0.6097	0.6141
0.2000	0.3999	436.45	436.69	435.21	0.5942	0.5802	0.5934
0.0999	0.5001	400.51	409.54	409.20	0.5435	0.5496	0.5648
0.0000	0.6162	385.82	383.80	377.55	0.5394	0.5466	0.5475
Aniline+1, 4-dioxan+Methyl IsoButyl Ketone							
0.5999	0.0000	721.52	694.73	667.08	0.9359	0.9169	0.9050
0.5000	0.10011	553.45	551.38	531.37	0.7727	0.7832	0.7681
0.3999	0.1998	510.47	490.37	484.71	0.7557	0.7358	0.7413
0.3001	0.2999	450.04	439.31	428.91	0.7004	0.6928	0.6871
0.2000	0.4001	410.60	397.01	396.29	0.6723	0.6580	0.6717
0.0999	0.5002	366.81	359.28	357.07	0.6272	0.6251	0.6363
0.0000	0.6001	338.01	332.41	328.73	0.6097	0.6056	0.6127

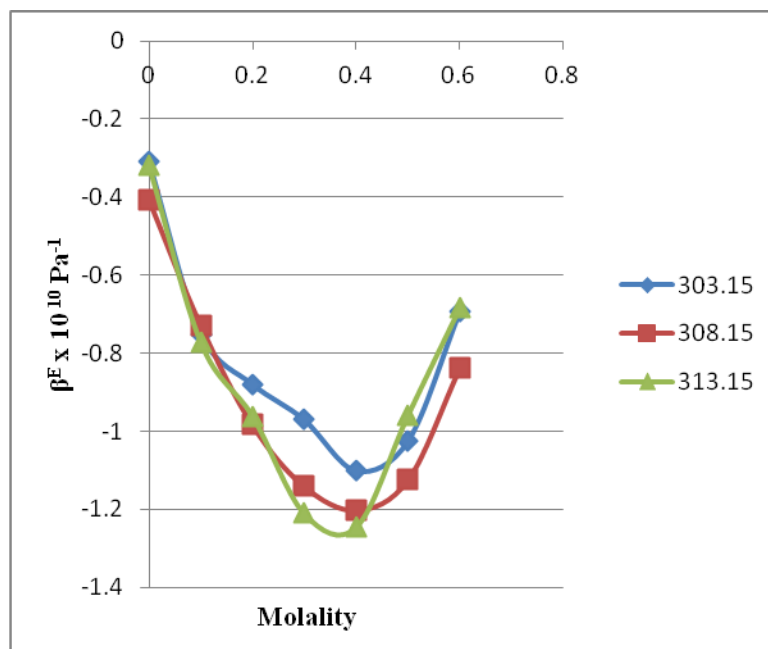


Fig 1.a. variation of Excess Adiabatic Compressibility Vs Mole fraction of acetone at 303.15, 308.15 and 313.15 K

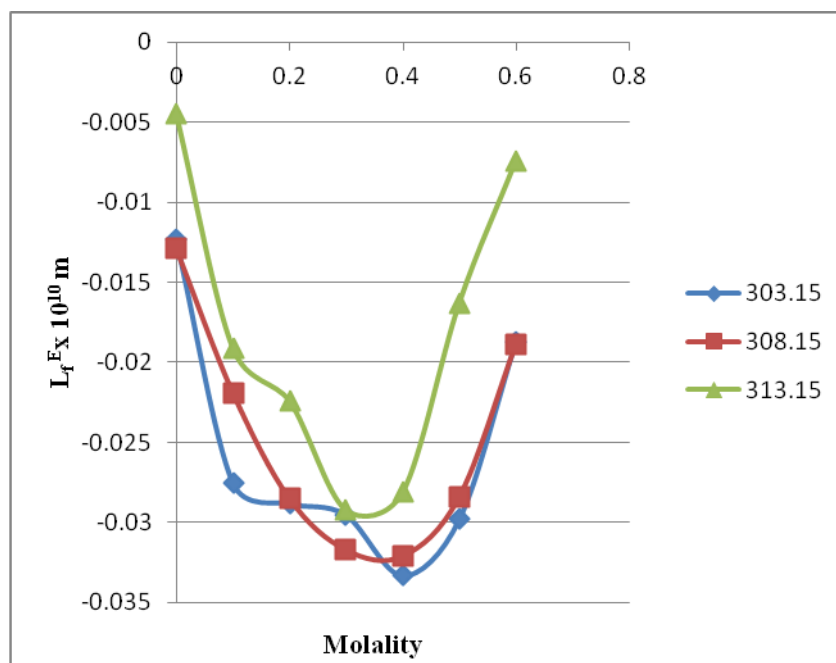


Fig 1.b. variation of Excess Free Length Vs Mole fraction of acetone at 303.15, 308.15 and 313.15 K

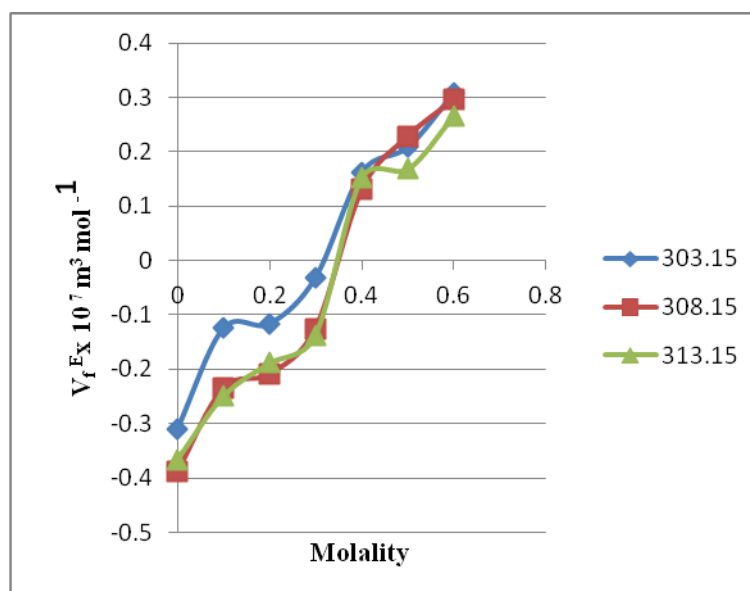


Fig 1.c. variation of Excess Free Volume Vs Mole fraction of acetone at 303.15, 308.15 and 313.15 K

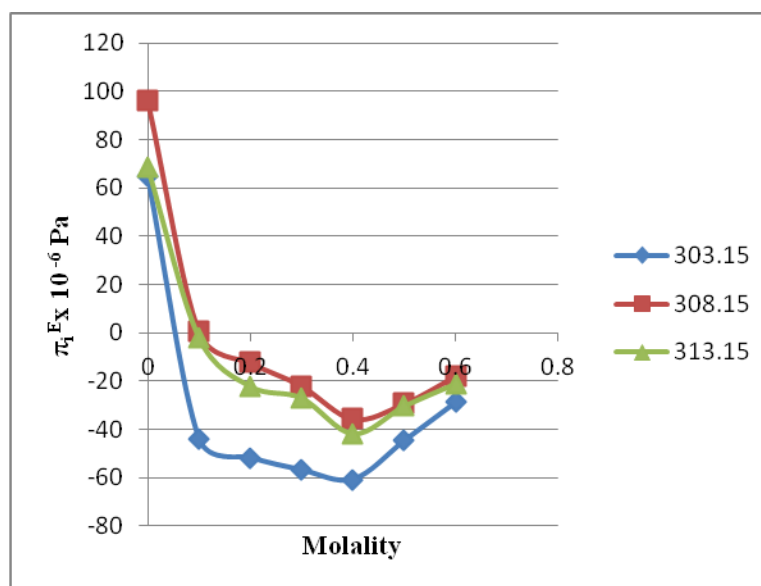


Fig 1.d. variation of Excess Internal Pressure Vs Mole fraction of acetone at 303.15, 308.15 and 313.15 K

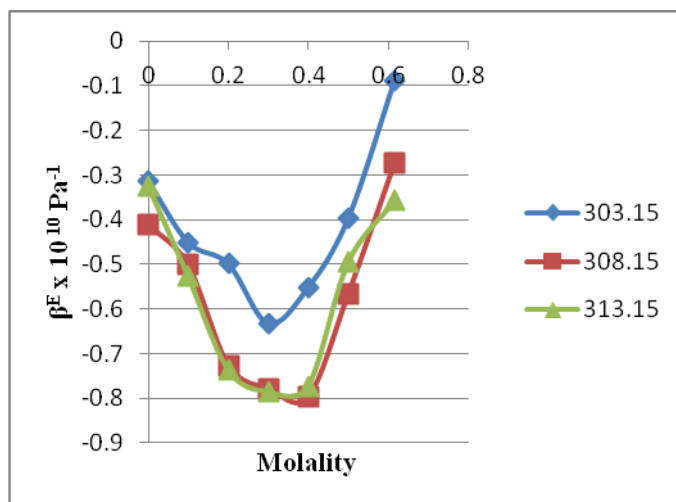


Fig 2.a. variation of Excess Adiabatic Compressibility Vs Mole fraction of butanone at 303.15, 308.15 and 313.15 K

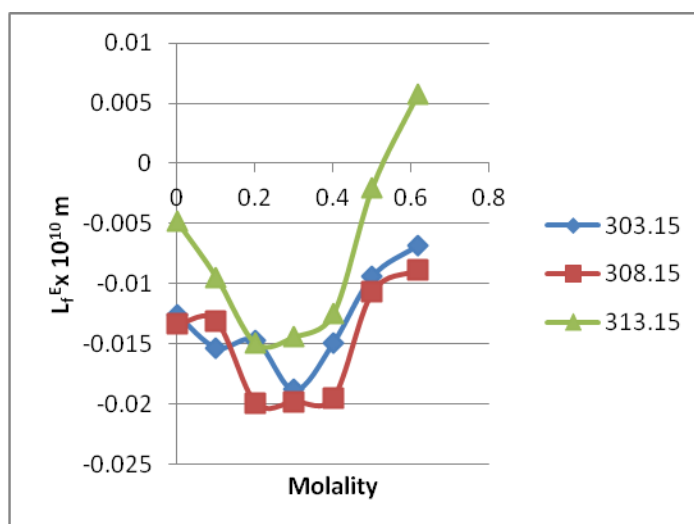


Fig 2.b. variation of Excess Free Length Vs Mole fraction of butanone at 303.15, 308.15 and 313.15 K

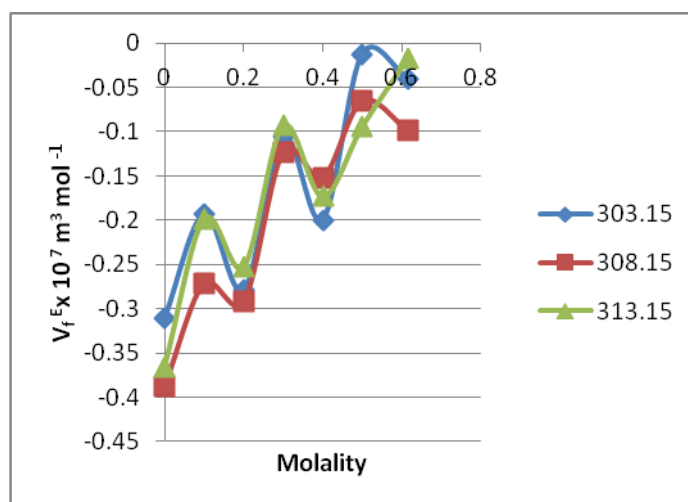


Fig 2.c. variation of Excess Free Volume Vs Mole fraction of butanone at 303.15, 308.15 and 313.15 K

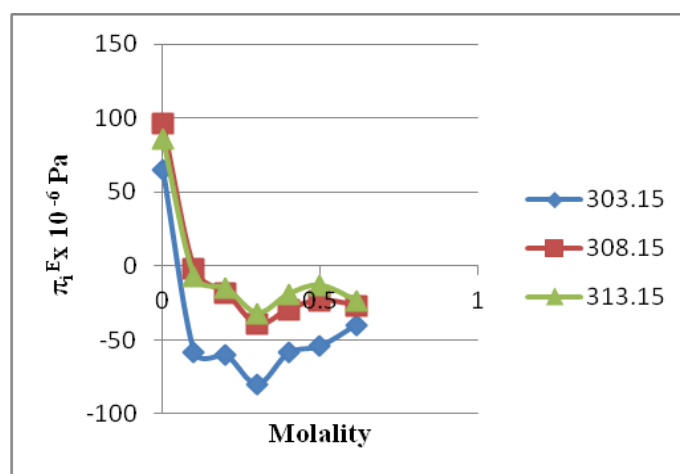


Fig 2.d. variation of Excess internal pressure Vs Mole fraction of butanone at 303.15, 308.15 and 313.15 K

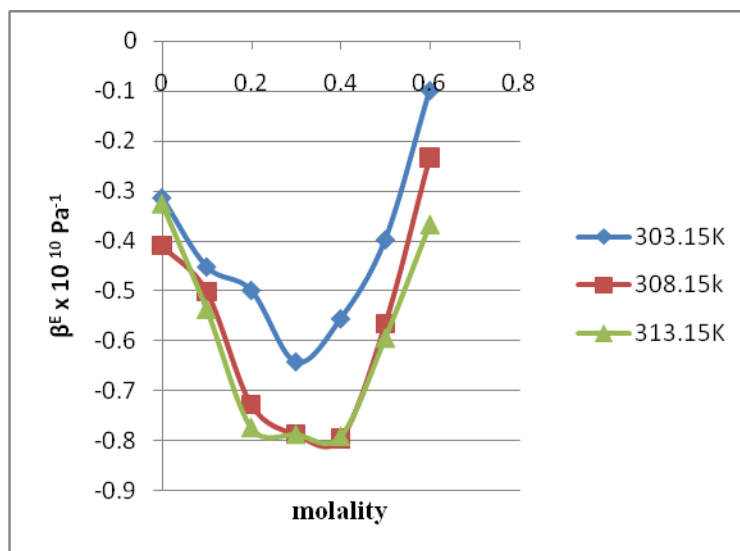


Fig 3.a. variation of Excess Adiabatic Compressibility Vs Mole fraction of MIBK at 303.15, 308.15 and 313.15 K

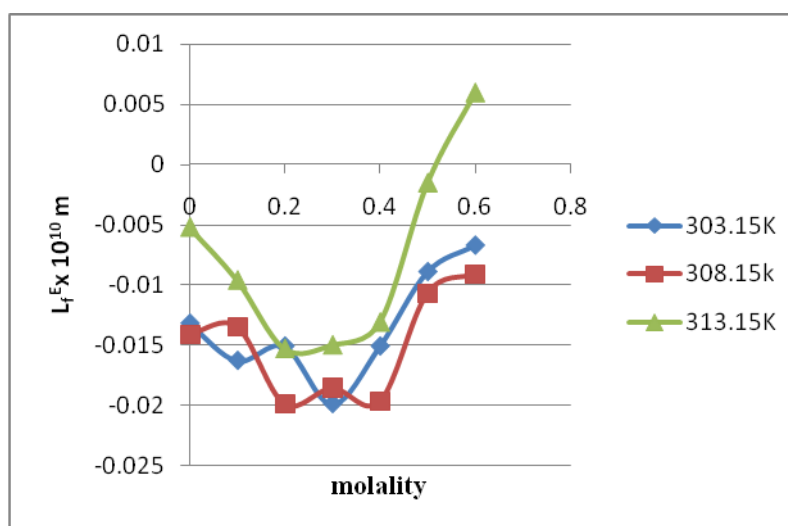


Fig 3.b. variation of Excess Free Length Vs Mole fraction of MIBK at 303.15, 308.15 and 313.15 K

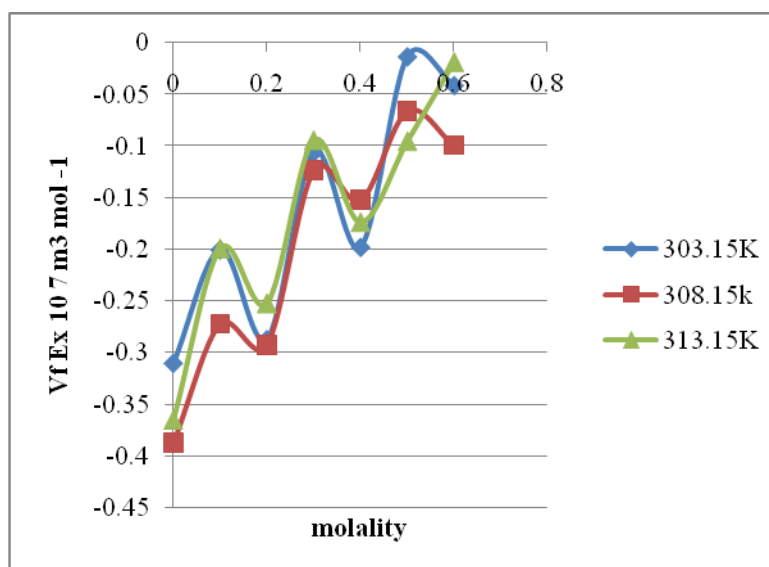


Fig 3.c. variation of Excess Free Volume Vs Mole fraction of MIBK at 303.15, 308.15 and 313.15 K

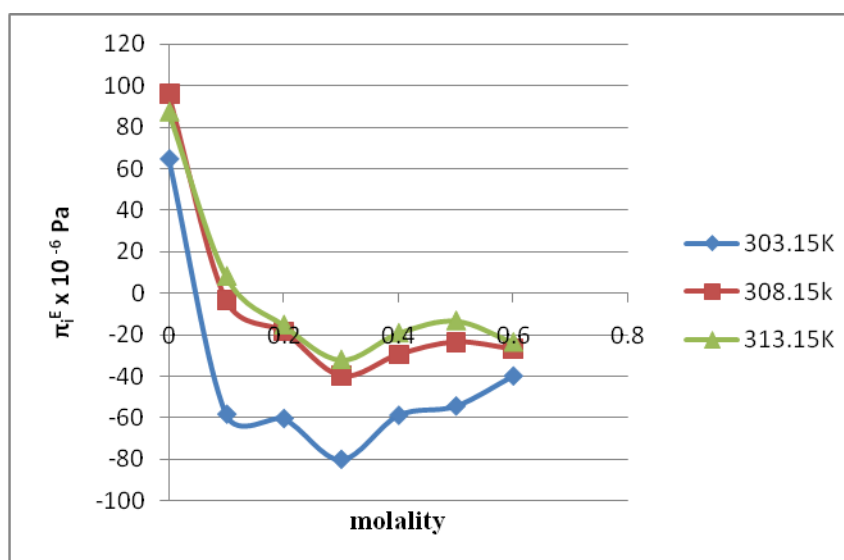


Fig3.d. variation of Excess internal pressure Vs Mole fraction of MIBK at 303.15, 308.15 and 313.15 K

From the Table-2 it was found that the density, viscosity and the ultrasonic velocity of the ternary liquid mixtures decreases with increasing mole fraction of ketone for all the three systems. The adiabatic compressibility and intermolecular free length are the deciding factors of ultrasonic velocity in the ternary liquid mixtures¹¹. The decrease in velocity is due to structural changes occurring in the mixtures resulting in weakening of intermolecular force¹². Further the sound speed decreases with increase

in temperature at any concentration as rise in temperature leads to less disordered structure and more spacing between the molecules. Also from the Table-2 it was observed that the density and viscosity of the ternary liquid mixtures decrease with increasing mole fraction of ketone. It was found that propanone has maximum values compared to butanone and MIBK. The decrease in density and viscosity with temperature indicates the decrease in intermolecular forces due to increase in thermal vibrations of the system, which causes an increase in volume expansion and hence increase in free length¹².

From the Table-3 it was observed that as the concentration of ketone increases, there was an increase in adiabatic compressibility, free length and free volume. The adiabatic compressibility shows an inverse behaviour compare to the ultrasonic velocity in the mixtures with increase in concentration. It is primarily the compressibility that changes with the structure and this lead to the change in ultrasonic velocity. In view of greater force of interaction between the molecules there will be an increase in cohesive energy and the occurrence of structural changes take place due to the existence of electrostatic field. Thus structural arrangement of molecules results in increasing adiabatic compressibility there by showing progressively intermolecular interactions. Similar results in some liquid mixtures where also reported by Bhatti and Singh¹³. Also from Table-4 it was noticed that the internal pressure decreases with increasing mole fraction of ketone. The internal pressure may give information regarding the nature and strength of forces existing between the molecules.

The excess parameter studies shows the intricate dependance of molecular interactions on various parameters such as size and shape of components and also the structural changes of components¹⁴. Non ideal liquid mixture shows considerable departure from linearity from their physical properties with respect to mole fraction and these are interpreted to explain the existence of strong or dispersive interactions.

From fig.1.a, fig2.a and fig.3.a it was observed that the values of excess adiabatic compressibility were negative. Fort et.al¹⁵, found that the negative value of excess adiabatic compressibility indicates greater interaction between the components of the mixtures. The strong interaction in the liquid mixture is due to the attractive forces. This leads to negative excess values of adiabatic compressibility, intermolecular free length, molar volume. The negative value of β^E is associated with structure forming tendency for some ternary liquid mixtures.

From fig.1.b, fig2.b and fig.3.b it was observed that the values of excess free volume were negative. Fort et.al¹⁵ noticed that the negative excess free volume tends to decrease as the strength of the interaction between the unlike molecules increases. Negative values of excess free volume may be attributed to the formation of charge transfer complexes between the molecules¹⁶. Fort et.al¹⁵ notice that the negative excess free volume tends to decreases the strength of the interaction between the unlike molecules increases although they do not parallel with the excess compressibility.

Also from fig.1.c, fig2.c and fig.3.c it was found that the values of excess free length are negative. According to Ramamoorthy et.al¹⁷ negative values of excess intermolecular free length L_F^E indicates that sound waves cover longer distances due to decrease in intermolecular free length ascribing the dominant nature of hydrogen bonding interaction between unlike molecules. The strong molecular interactions due

to charge transfer and formation of hydrogen bond lead to negative values of L_F^E . The negative excess values of free length L_F^E predict the existence of strong molecular interactions in the ternary liquid mixture. Fort et.al¹⁵ indicated that values of excess intermolecular free length should be due to charge transfer, dipole –induced dipole and dipole-dipole interactions.

It is inferred that carbonyl compounds are co-ordinated to amine through strong H-bond. Aniline is having a relatively higher dielectric constant^{15, 18} and basically electron donor nature, when it mixes with aliphatic ketones exhibits significant interactions like a dipole-dipole interaction and hydrogen bonding between participating molecules.

From fig.1.d, fig2.d and fig.d.a it was found that the excess internal pressure (π_i^E) value is negative and it decreases with increase in mole fraction of butanone as well as temperature. The negative excess internal pressure indicates that only dispersive and dipolar forces are operating with complete absence of specific interaction¹⁹. The negative excess internal pressure over the entire range of mole fraction also supports the presence of interaction.

From the various acoustical parameters and excess parameters, the trend in these parameters with concentration establishes that (i) strong intermolecular interactions exist in these mixtures and (ii) formation of charge transfer complexes through hydrogen bonding between primary amine and aromatic ketones. The formation of complexes has also been confirmed by optical spectroscopy at 303K²⁰. It is observed that the stability of the complexes is influenced by the structure of the component molecules.

The Gibb's free energy (ΔG) increases with increase in mole fraction of the solute, which may due to intermediate compound formation between the ternary liquid mixtures which also confirm the presence of interaction.

CONCLUSION

Ultrasonic method is a powerful probe for characterising the physico-chemical properties and existence of molecular interaction in the mixture. From the ultrasonic velocity and other related acoustic parameters and excess parameters, the trend in these parameters with concentration establishes that (i) strong intermolecular interactions exist in these mixtures and (ii) formation of charge transfer complexes through hydrogen bonding between primary amine and aromatic ketones. Further the strength of interaction tends to be weaker with rise in temperature due to weak intermolecular forces and thermal dispersion forces. From the magnitude of excess parameter the existence of molecular interaction is in the order Propanone > Butanone > Methyl Iso Butyl Ketone.

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