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Study of thermodynamic and transport properties of ternary liquid mixture at different temperatures

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ABSTRACT

The Ultrasonic velocity, density, and viscosity have been measured for ternary mixture of N-N dimethyl formamide (DMF), Cyclohexane and Chloro-benzene at different temperatures for a constant frequency (6 MHz). These experimental data have been used to estimate the thermodynamic parameters such as adiabatic compressibility (β), free length (L_f), internal pressure (Πi), relaxation time (τ), acoustic impedance (Z), and Gibb's free energy (ΔG) for the solution. The excess values of the parameters are also evaluated and discussed.

Key words: Ultrasonic velocity, intermolecular, free length, internal pressure, free volume, Gibb's free energy.

INTRODUCTION

The studies of thermodynamical and transport properties of multi-component (binary and ternary) liquid mixtures and solutions have found wide application in chemical, textile, leather and nuclear industries. Ultrasonic investigations of liquid mixtures consisting of polar and non-polar components enable to understand the molecular interactions and structural behavior of molecules and their mixtures **[1-3]**. The intermolecular interaction influences the structural arrangement along with the shape of the molecules.

For a better understanding of the physio-chemical properties and the molecular interaction between the participating components of these mixtures, ultrasonic velocities together with density and viscosity are measured at different temperatures for different concentration of the components in the mixture. These data furnish wealth of information about the interaction between ions, dipoles; hydrogen bonding, multipolar and dispersive forces **[4-5]**. In order to understand the nature of molecular interactions between the components of the liquid mixtures, it is of interest to discuss the same in terms of excess parameters rather than the actual values. The dispersion forces which are caused by correlated movements of the electrons in interacting molecules are responsible for positive excess values. Dipole-dipole, dipole-induced dipole, charge transfer interaction and hydrogen bonding between unlike molecules are responsible for possessing negative excess values.

The present paper deals with the study of different parameters of the mixture containing DMF, Cyclohexane and chlorobenzene at different temperature through ultrasonic measurements at high frequency range.

DMF is a versatile compound. It is a non-aqueous solvent which has no hydrogen bonding in pure state. Therefore it acts as an aprotic, protophilic medium with high dielectric constant and it is also considered as a dissociating solvent. DMF being a polar molecule results in dipolar and induced dipolar interaction between it and chlorobenzene in addition to dipolar-dipolar interaction between its molecules.

Cyclohexane is a non-polar, unassociated, inert hydrocarbon possessing globular structure. It is produced in large quantity by hydrogenation of benzene. The principal use of cyclohexane is conversion by oxidation in air to a mixture of cyclohexanol and the ketone, which is then oxidized further to adipic acid for the manufacture of nylon.

Cyclohexane belongs to alicyclic hydrocarbon (closed chain). The packing of carbon atoms in the even numbered alkane groups allows the maximum intermolecular attractions **[6]**. It is highly inert towards an electrophille or a nucleophille at ordinary temperature. Due to the non-polar nature of cyclohexane and its inertness towards electron donors **[7]**, dispersive types of interaction are expected between it and other components.

Chlorobenzene is a poor electron donor towards the electron seeking proton of any group. It has low dielectric constant ($\varepsilon = 5.649$) and dipole moment. ($\mu = 1.69$ D). It is neither acidic nor basic and is more reactive because the chlorine atom is bonded with SP^3 hybridised carbon atom and consequently can be removed easily. Hence the rate of reaction with chlorobenzene is faster. The rate of molecular interactions results in a greater degree of variation with respect to ultrasonic related parameters. The chlorine atom being an electron withdrawing atom attracts the π-electron of benzene ring in C_6H_5Cl and thus a decrease of the electron density of the ring takes place. This makes the benzene ring a relatively poor electron donor towards the Cyclohexane molecules. Hence a weak interaction between chlorobenzene and Cyclohexane is expected. In our system the dipole - induced dipole interaction between DMF and chlorobenzene is significant.

MATERIALS AND METHODS

The chemicals used in the present work were analytical reagent (AR) and spectroscopic reagent (SR) grades with minimum assay of 99.9% were obtained from E-Merk Ltd (India). Various concentrations of the ternary liquid mixtures were prepared in terms of mole fraction, out of which the mole fraction of the second component Cyclohexane ($X_2 = 0.4$) was kept fixed while the mole fractions of remaining two (X_1 and X_3) were varied from 0.0 to 0.6. There is nothing significant in fixing the second component at 0.4.

2.1: Measurements

(i) Velocity Measurement:-

The velocity of ultrasonic wave in the ternary mixture have been measured using multi-frequency ultrasonic interferometer with an high degree of accuracy operating at 11 different frequencies (Model M-84) supplied by M/s Mittal Enterprises, New Delhi. The measuring cell of interferometer is a specially designed double walled vessel with provision for temperature constancy. An electronically operated digital constant temperature bath (Model SSI-03spl) supplied by M/s Mittal Enterprises, New Delhi, operating in the temperature range -10^oc to 85^oc with an accuracy of ± 0.1K has been used to circulate water through the outer jacket of the double walled measuring cell containing the experimental liquid.

(ii) Density Measurement:-

The densities of the mixture were measured using a 25ml specific gravity bottle. The specific gravity bottle with the experimental mixture was immersed in a temperature controlled water bath. The density was measured using the formula

$$\rho_2 = (w_2/w_1).\ \rho_1$$

Where, w_1 = weight of distilled water, w_2 = Weight of experimental liquid, ρ_1 = Density of water, ρ_2 = Density of experimental liquid

(iii) Viscosity measurement:-

The viscosities of the ternary mixture were measured using an Oswald's viscometer calibrated with double distilled water. The Oswald's viscometer with the experimental mixture was immersed in a temperature controlled water bath. The time of flow was measured using a digital racer stop watch with an accuracy of 0.1 sec. The viscosity was determined using the relation,

$$\eta_2 = \eta_1\ (t_2/t_1)\ (\rho_2/\rho_1)$$

Where, η_1 = Viscosity of water, η_2 = Viscosity of mixture, ρ_1 = Density of water, ρ_2 = Density of mixture, t_1 = Time of flow of water, t_2 = Time of flow of mixture.

2.2: THEORETICAL ASPECT

The following thermodynamic parameters were calculated from Jacobson's relation **[8–10]**.

(i) Adiabatic Compressibility (β):-

The adiabatic compressibility is the fractional decrease of volume per unit increase of pressure, when no heat flows in or out. It is calculated from the speed of sound (U) and the density (ρ) of the medium by using the equation of Newton Laplace as,

$$\beta = 1/U^{2}\cdot\rho \qquad \text{--------}\ (1)$$

(ii) Intermolecular free length (L_f) :-

The intermolecular free length is the distance between the surfaces of the neighboring molecules. It is calculated by using the relation

$$L_f = K_T\beta^{1/2} \qquad \text{---------}\ (2)$$

Where K_T is the temperature dependent constant and 'β' is the adiabatic compressibility.

(iii) Free Volume (V_f) :- Free volume in terms of ultrasonic velocity(U) and the viscosity(η) of liquid is

$$V_f = (M_{eff}.U / K.\eta)^{1/2} \qquad \text{--------}\ (3)$$

Where 'M_{eff}' is the effective mass of the mixture, 'K' is a dimensionless constant independent of temperature and liquid. Its value is 4.281×10^{9}.

(iv) Internal Pressure (Πi):-

The measurement of internal pressure is important in the study of the thermodynamic properties of liquids. The internal pressure is the cohesive force, which is a resultant o force of attraction and force of repulsion between the molecules. It is calculated by using the relation,

$$\Pi_i = bRT\,(k\eta/U)^{1/2}\,(\rho^{2/3}/M_{eff}^{7/6}) \qquad \text{---------}\ (4)$$

Where, 'b' stands for cubic packing, which is assumed to be '2' for all liquids, ' k' is a dimensionless constant independent of temperature and nature of liquids. Its value is 4.281×10^{9}. ' T ' is the absolute temperature in Kelvin, 'M_{eff}' is the effective molecular weight, 'R' is the Universal gas constant, 'η' is the viscosity of solution in $N.S.m^{-2}$, 'U' is the ultrasonic velocity in $m.s^{-1}$ and 'ρ' is the density in $Kg.m^{-3}$ of solution.

(v) Relaxation time (τ):-

Relaxation time is the time taken for the excitation energy to appear as translational energy and it depends on temperature and impurities. The relaxation time can be calculated from the relation,

$$\tau = 4/3.\,(\beta.\eta) \qquad \text{--------}\ (5)$$

Where 'β' is the adiabatic compressibility and 'η' is the viscosity of the mixture.

(vi) Acoustic impedance (Z):-

The specific acoustic impedance is given by,

$$Z = U.\rho \qquad \text{--------}\ (6)$$

Where 'U' is the velocity and 'ρ' is the density of the mixture.

(vii) Gibb's free energy:-

The Gibb's free energy is calculated by using the relation

$$\Delta G = kT.\mathrm{Ln}\,(kT\tau/h) \qquad \text{---------}\ (7)$$

Where, 'τ' is the viscous relaxation time, '*T*' is the absolute temperature, 'k' is the Boltzmann's constant and '*h*' is the Planck's constant.

RESULTS AND DISCUSSION

The experimental data relating to density, viscosity and velocity at 288 K, 298 K, 308 K and 318 K for the mixture are given in table-3.1.

TABLE – 3.1: Values of Density (ρ), Viscosity (η) and velocity (U) at 288k, 298K, 308 K and 318 K.

Mole fraction		Density (ρ) Kg.m^{-3}				Viscosity (η) (10^{-3} N.S.m^{-2})				Velocity (U) m.s^{-1}			
X_1	X_2	288K	298K	308K	318K	288K	298K	308K	318K	288K	298K	308K	318K
0.0000	0.6000	998.17	991.77	984.93	975.25	1.1087	0.8401	0.6509	0.5486	1280.4	1242	1206	1166.4
0.0999	0.4999	981.86	975.50	968.85	959.40	1.1104	0.8485	0.6655	0.5509	1291.1	1254.8	1218.3	1178.1
0.1998	0.4001	964.20	957.90	951.46	942.25	1.1166	0.8612	0.6755	0.5536	1304.6	1269.6	1233.3	1190.6
0.3001	0.3000	944.93	938.69	932.47	923.53	1.1553	0.8820	0.6852	0.5573	1320.1	1287.3	1246.9	1205.4
0.4000	0.1999	923.88	917.70	911.72	903.08	1.1836	0.8955	0.6899	0.5594	1336.8	1303.5	1263	1220.6
0.4998	0.1001	900.86	894.75	889.04	880.71	1.1910	0.9012	0.6943	0.5615	1355	1321.9	1280.1	1237.6
0.5997	0.0000	875.41	869.38	863.96	855.98	1.2188	0.9153	0.7002	0.5631	1377.2	1350.1	1300.5	1256.4

Ultrasonic velocity increases with increase in mole-fraction. This is due to the structural changes occurring in the mixtures resulting in increase of intermolecular forces. However velocity decreases with increase in temperature because of less ordered structure and more spacing between the molecules.

The calculated values of adiabatic compressibility (β) and free length (L_f) are reported in table - 3.2, whereas values of free volume and internal pressure are reported in table -3.3.

TABLE – 3.2: Values of Adiabatic compressibility (β) and free length (L_f) at 288k, 298K, 308 K and 318 K.

Mole fraction		Adiabatic compressibility (β) (10^{-10} N^{-1}.m^2)				Free length (L_f) (10^{-10} m)			
X_1	X_2	288K	298K	308K	318K	288K	298K	308K	318K
0.0000	0.6000	6.1109	6.5365	6.9807	7.5368	0.4846	0.5063	0.5340	0.5576
0.0999	0.4999	6.1099	6.5106	6.9540	7.5099	0.4845	0.5053	0.5330	0.5566
0.1998	0.4001	6.0937	6.4766	6.9099	7.4869	0.4839	0.5039	0.5313	0.5557
0.3001	0.3000	6.0728	6.4286	6.8977	7.4522	0.4831	0.5021	0.5308	0.5544
0.4000	0.1999	6.0569	6.4132	6.8759	7.4324	0.4824	0.5015	0.5300	0.5537
0.4998	0.1001	6.0459	6.3959	6.8642	7.4132	0.4820	0.5008	0.5295	0.5530
0.5997	0.0000	6.0228	6.3105	6.8436	7.4008	0.4811	0.4974	0.5287	0.5525

TABLE – 3.3: Values of free volume (V_f) and internal pressure (Πi) at 288k, 298K, 308 K and 318 K

Mole fraction		Free volume (V_f) (10^{-7} m^3.mol^{-1})				Internal pressure(Πi) (x 10^6 N.m-^{2})			
X_1	X_2	288K	298K	308K	318K	288K	298K	308K	318K
0.0000	0.6000	0.4648	0.6533	0.9167	1.1268	131.98	121.39	111.56	106.81
0.0999	0.4999	0.4352	0.6157	0.8482	1.0708	137.02	125.75	116.27	110.35
0.1998	0.4001	0.4064	0.5760	0.7938	1.0149	142.38	130.58	120.74	114.11
0.3001	0.3000	0.3683	0.5318	0.7403	0.9591	149.40	136.17	125.49	118.09
0.4000	0.1999	0.3383	0.4949	0.6980	0.9082	156.04	141.59	129.92	122.09
0.4998	0.1001	0.3185	0.4664	0.6571	0.8589	161.55	146.55	134.53	126.25
0.5997	0.0000	0.2927	0.4365	0.6168	0.8121	168.53	151.92	139.35	130.46

The intermolecular interaction seems to be stronger than the intra-molecular interaction thus leading to a decrease of volume. Hence free volume decreases with increase in mole fraction. When the temperature is increased there is reduction in molecular interaction as they move away from each other. This reduces the cohesive force. Thus a decrease in internal pressure and increase in free volume occurs with increases in temperature (fig.1-4).

Contraction in volume leads to subsequent decrease in adiabatic compressibility as well as in intermolecular free length. These parameters increase with increase in temperature for a particular concentration. (Fig. 5-8)

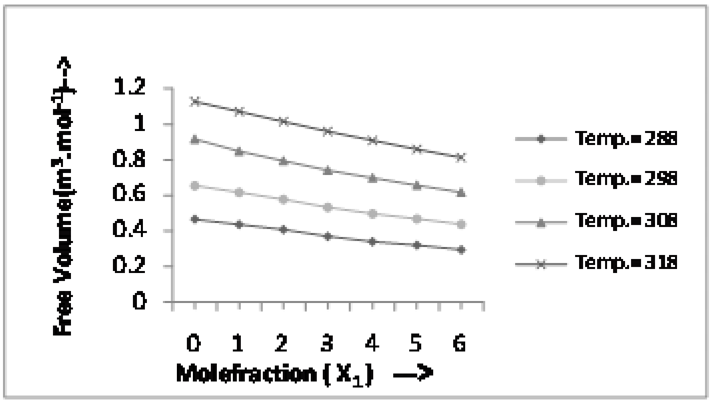

Fig.-1. Variation of free volume with mole fraction

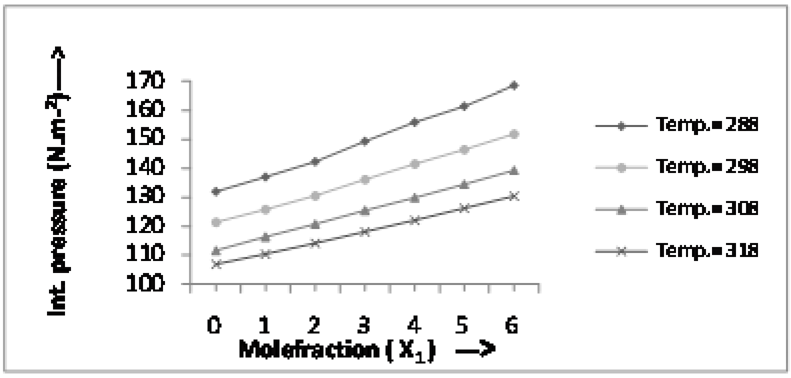

Fig.-2. Variation of internal pressure with mole fraction

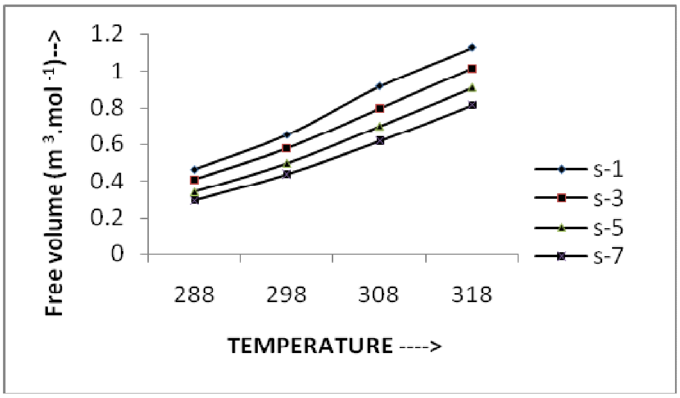

Fig.-3. Variation of free volume with temperature

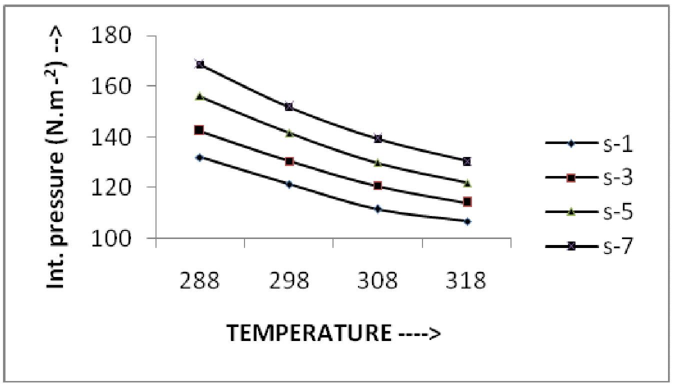

Fig.-4. Variation of internal pressure with temperature

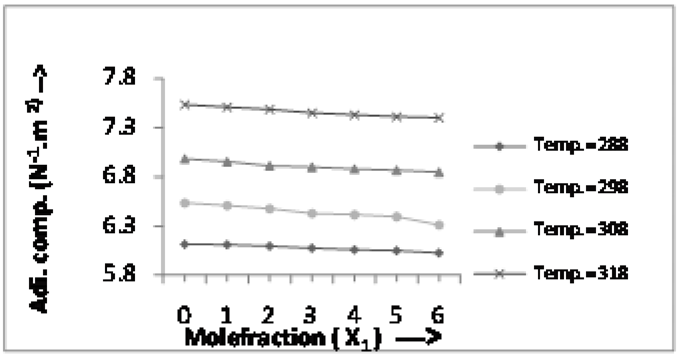

Fig.-5. Variation of adiabatic compressibility with mole fraction

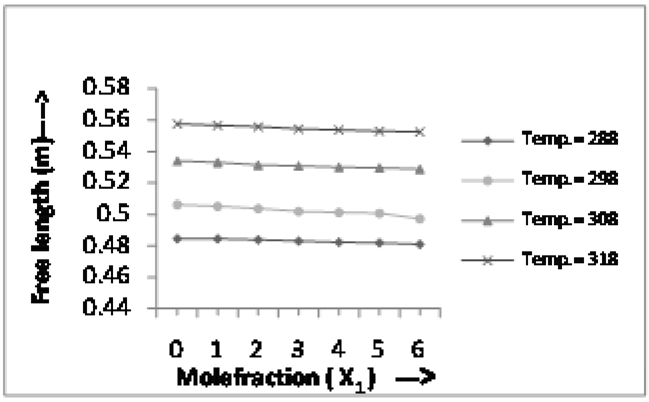

Fig.-6. Variation of free length with mole fraction

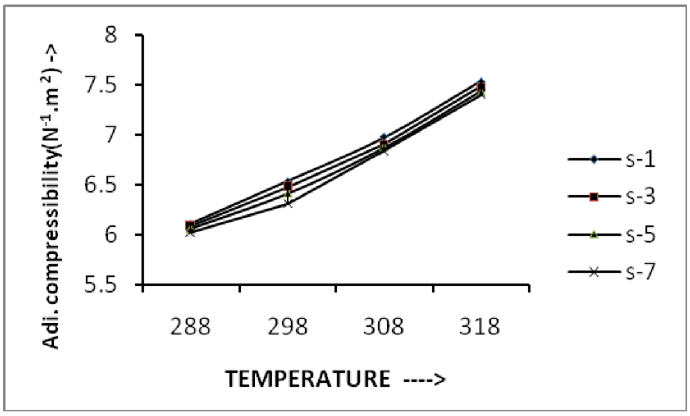

Fig.-7: Variation of adiabatic compressibility with temperature

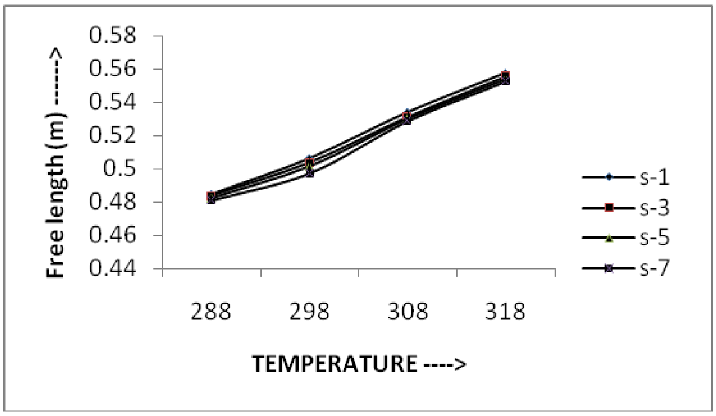

Fig.-8: Variation of Free Volume with temperature

Internal pressure (Π_i), viscous relaxation time (τ) and Gibb's free energy (ΔG) are reported in table -3.3.

TABLE – 3.3: Values of viscous relaxation time (τ) and Gibb's free energy (ΔG) and acoustic impedance (Z) at 288k, 298K, 308 K and 318 K.

Mole fraction		Viscous relaxation time(τ) (x 10^{-12} s)				Gibb's free energy (ΔG) (x 10^{-20} k.J.mol^{-1})				Acoustic impedance (Z) (x 10^{6} Kg.m^{2}.s^{-1})			
X_1	X_2	288K	298K	308K	318K	288K	298K	308K	318K	288K	298K	308K	318K
0.0000	0.6000	0.8854	0.7321	0.6058	0.5513	0.6641	0.6230	0.5774	0.5688	1.2781	1.2318	1.1878	1.1375
0.0999	0.4999	0.8964	0.7366	0.6170	0.5516	0.6690	0.6255	0.5852	0.5690	1.2677	1.2241	1.1804	1.1303
0.1998	0.4001	0.9072	0.7437	0.6224	0.5526	0.6738	0.6294	0.5889	0.5698	1.2579	1.2161	1.1734	1.1218
0.3001	0.3000	0.9354	0.7559	0.6302	0.5538	0.6860	0.6362	0.5942	0.5708	1.2474	1.2084	1.1627	1.1132
0.4000	0.1999	0.9558	0.7657	0.6325	0.5544	0.6946	0.6415	0.5957	0.5712	1.2350	1.1962	1.1515	1.1023
0.4998	0.1001	0.9601	0.7685	0.6354	0.5550	0.6963	0.6430	0.5977	0.5718	1.2207	1.1828	1.1381	1.0900
0.5997	0.0000	0.9787	0.7701	0.6389	0.5556	0.7040	0.6438	0.6000	0.5723	1.2056	1.1737	1.1236	1.0755

Relaxation time increase with increase in mole fraction of DMF. Such situation suggests that, the molecules get rearranged due to co-operation process **[11]**. When temperature increases relaxation time decreases indicating the reverse process. (Fig. 9-10)

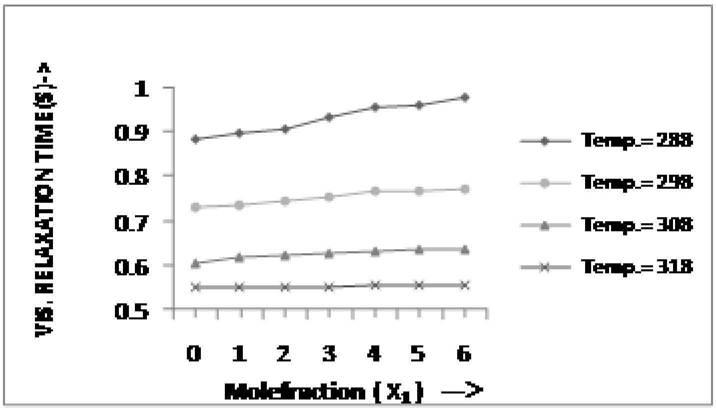

Fig.-9. Variation of Viscous Relaxation Time with mole fraction

Gibbs' free energy increases with increase in concentration of DMF. Increasing value of Gibbs function suggests that the closer approach of unlike molecule is due to hydrogen bonding **[12-13]**. The increase in Gibbs' free energy also suggests shorter time for rearrangement of the molecules in the mixtures. The case is just reverse when temperature increases keeping the concentration of DMF constant. (Fig. 11-12)

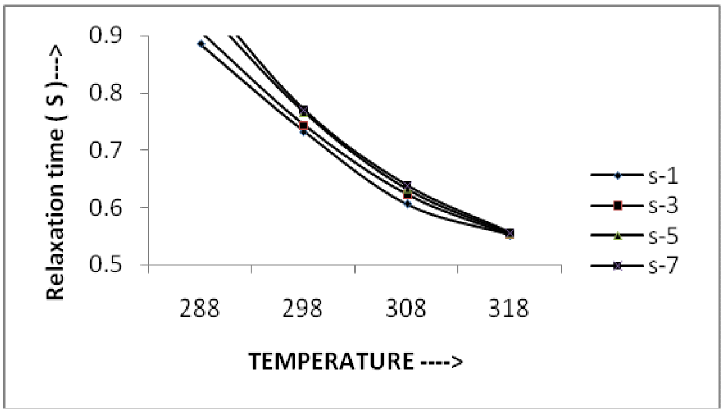

Fig.-10. Variation of Viscous Relaxation Time with temperature

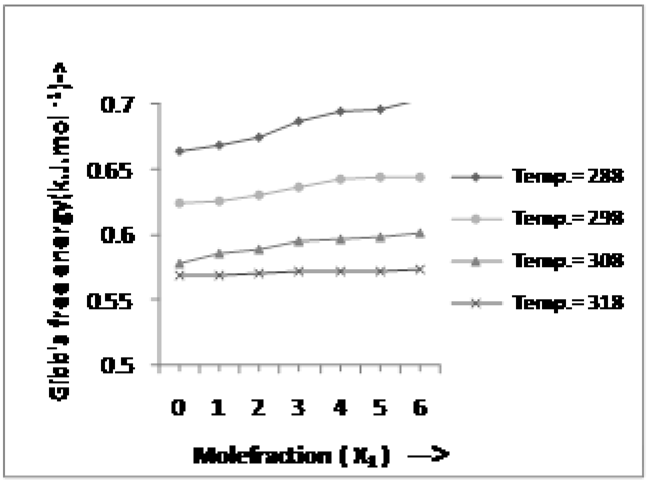

Fig-11. Variation of Gibb's Free Energy with mole fraction

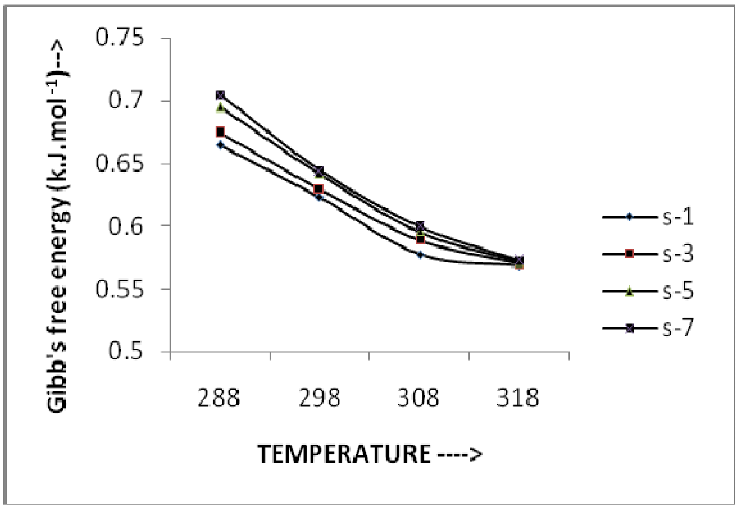

Fig-12. Variation of Gibb's Free Energy with temperature

The ratio of instantaneous excess pressure of any particle to the instantaneous velocity of that particle is known as specific acoustic impedance of the medium. This acoustic impedance decreases with increase in concentration of

DMF as well as with increase in temperature. This indicates that there is a decrease in molecular concentration in both the cases. (Fig. 13-14).

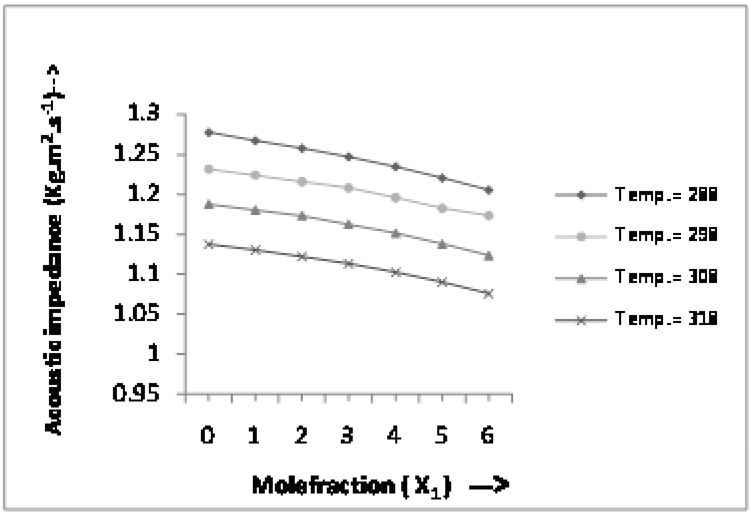

Fig.-13. Variation of Acoustic Impedance with mole fraction

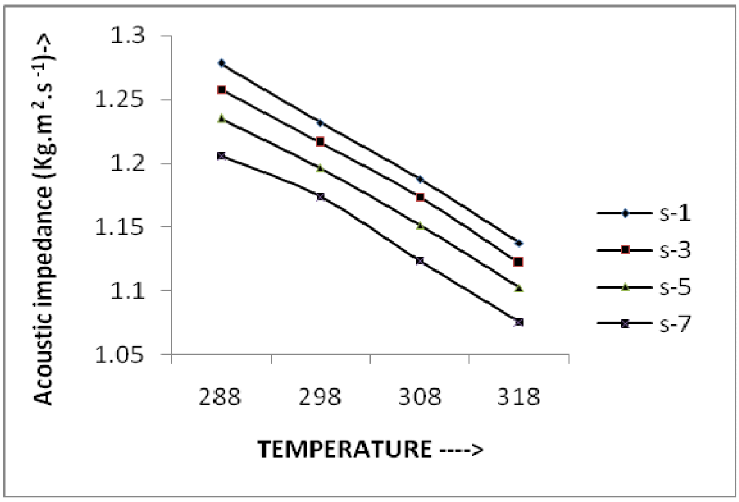

Fig.-14. Variation of Acoustic Impedance with temperature

Excess values of adiabatic compressibility (β^E), excess free length (L_f^E) and free volume (V_f) are reported in table-3.5. Excess internal pressure ($\boldsymbol{\Pi_i}^E$) and excess Gibb's free energy (ΔG^E) are reported in table-3.6.

TABLE – 3.5 : Excess values of adiabatic compressibility (β), free length (L_f) and free volume (V_f) at 288k, 298K, 308 K and 318 K.

Mole fraction		Adiabatic compressibility(β) ($10^{-10}N^{-1}.m^2$)				Free length (L_f) (10^{-10} m)				Free volume (V_f) (10^{-7} $m^3.mol^{-1}$)			
X_1	X_2	288K	298K	308K	318K	288K	298K	308K	318K	288K	298K	308K	318K
0.0000	0.6000	.0162	.157	.306	.2989	.0218	.0725	.1392	.1488	-0.1176	-0.2431	-0.1555	-0.1978
0.0999	0.4999	.0911	.2143	.3343	.341	.0554	.0989	.1533	.1587	-0.2515	-0.3925	-0.2099	-0.0570
0.1998	0.4001	.1492	.2618	.3437	.3853	.0816	.1207	.1594	.1785	-0.3783	-0.5652	-0.4378	-0.1000
0.3001	0.3000	.2024	.2951	.3844	.4175	.1056	.1367	.1772	.1936	-0.5971	-0.7821	-0.6563	-0.2480
0.4000	0.1999	.2624	.3629	.4177	.4667	.1334	.167	.1932	.2161	-0.7352	-0.9239	-0.7589	-0.3453
0.4998	0.1001	.3262	.4275	.4599	.5153	.1621	.1957	.2121	.2378	-0.7707	-0.9842	-0.8506	-0.4290
0.5997	0.0000	.3789	.4253	.4944	.572	.1868	.1985	.2285	.263	-0.8666	-1.0555	-0.9350	-0.4849

TABLE – 3.6 : Excess values of internal pressure (Πi), Acoustic impedance (Z) and Gibb's free energy (ΔG) at 288k, 298K, 308 K and 318 K.

Mole fraction		Internal pressure(Πi) (x 10^6 N.m-^{2})				Gibb's free energy (ΔG) (x 10^{-20} k.J.mol^{-1})			
X_1	X_2	288K	298K	308K	318K	288K	298K	308K	318K
0.0000	0.6000	2.600	3.103	1.197	1.168	0.1201	0.1018	0.0950	0.0838
0.0999	0.4999	3.257	3.418	1.860	0.636	0.2482	0.2092	0.1682	0.0941
0.1998	0.4001	4.187	4.174	2.233	0.294	0.2729	0.2692	0.2648	0.1584
0.3001	0.3000	6.746	5.657	2.875	0.145	0.4716	0.4771	0.3774	0.2238
0.4000	0.1999	8.998	7.028	3.248	0.085	0.6364	0.6123	0.4543	0.2864
0.4998	0.1001	10.10	7.931	3.784	0.153	0.7316	0.7084	0.5343	0.3482
0.5997	0.0000	12.68	9.254	4.544	0.295	0.8871	0.7993	0.6188	0.4110

Positive excess values of adiabatic compressibility, indicates the existence of dispersive forces between molecules of mixtures. It also indicates loosely packed molecules in the mixtures. The positive excess values of internal pressure clearly confirm this prediction. (Fig.-15-16).

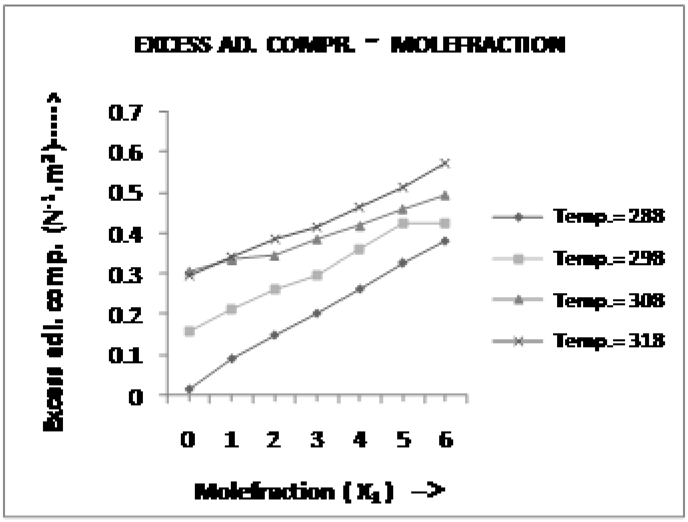

Fig.-15. Variation of Excess Adiabatic Compressibility with mole fraction

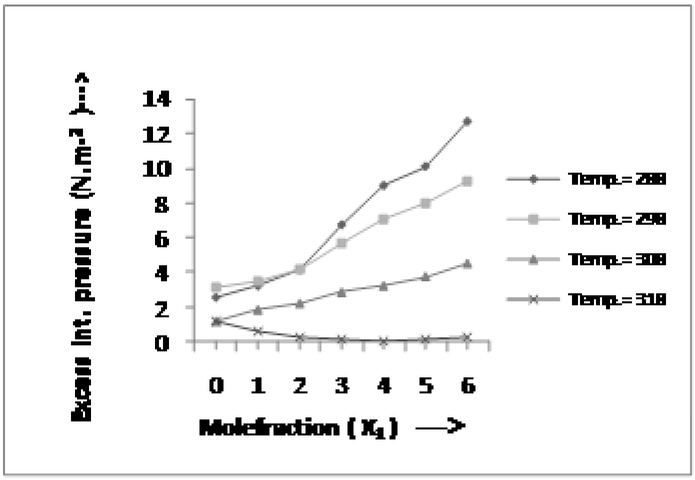

Fig.-16. Variation of Excess internal pressure with mole fraction

The positive value of excess free length shows the existence of dispersive forces between molecules of mixture. Spencer et. al **[2]** has also reported a similar observation. (Fig.-17)

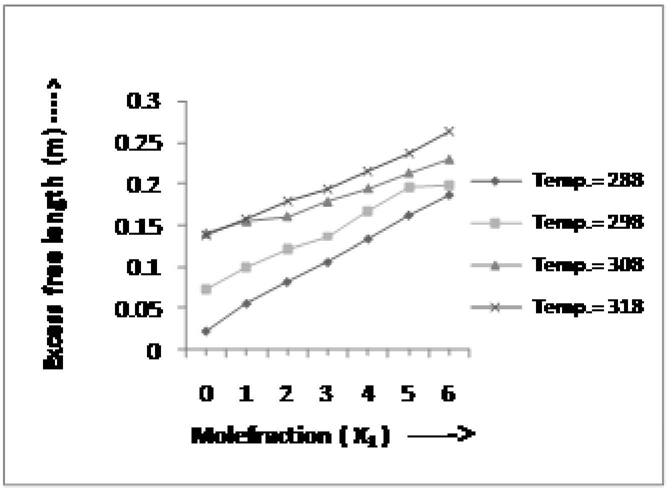

Fig.-17. Variation of Excess free length with mole fraction

Excess free volume exhibiting negative values over the entire range of composition clearly suggests the existence of strong interactive forces present in the mixtures. Negative free volume increases with increasing concentration of DMF. This also indicates strong molecular association between the unlike molecules. (Fig.-18)

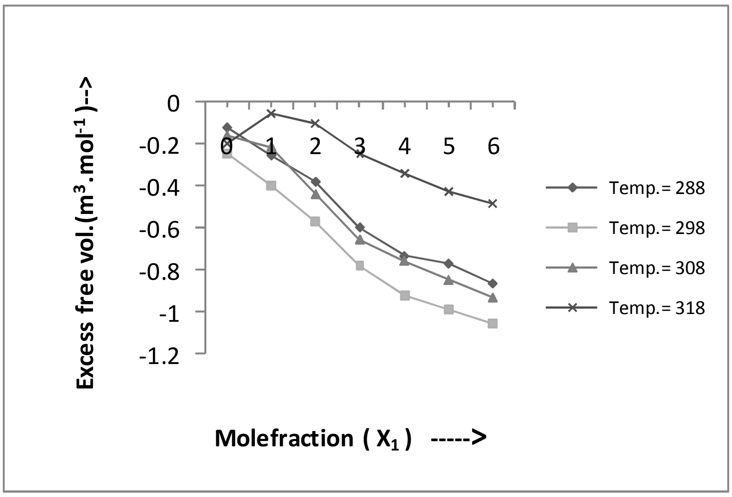

Fig.-18. Variation of Excess free volume with mole fraction

The variations of excess Gibb's free energy are found to be positive and it increases with increase in mole fraction but it decreases with the rise in temperature. According to Reed et. al **[14]** the positive value of excess Gibb's free energy may be attributed to specific interactions like hydrogen bonding and charge transfer. In the present investigation positive values of excess Gibb's free energy shows the strong interaction between the unlike molecules. (Fig. – 19)

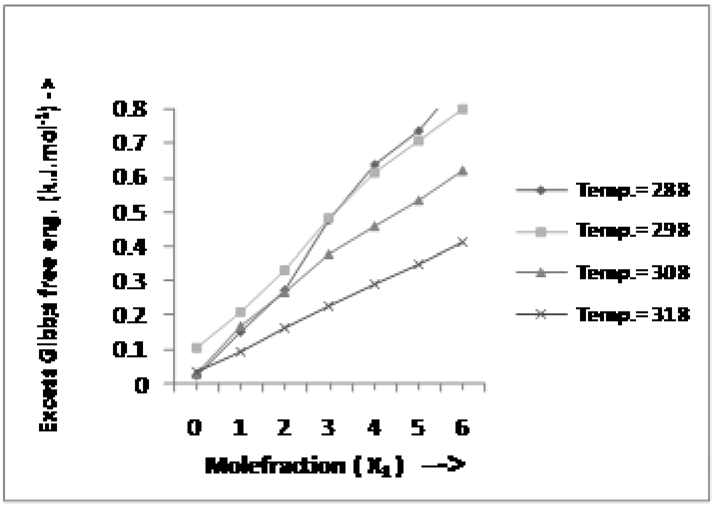

Fig.-19. Variation of Excess Gibb's free energy with mole fraction

CONCLUSION

Ultrasonic velocity studies on the system of ternary mixture of DMF with cyclohexane and chlorobenzene shows that, when concentration of DMF increases, more rigid structure is formed due to bonding between the unlike molecules through dipole-dipole and dipole-induced dipole interaction. Ultrasonic velocity increases with increase in concentration of DMF and decreases with increase in temperature, adiabatic compressibility decreases with concentration of DMF but increases with Increase in temperature, the same variation also occur in case of free length and free volume. Internal pressure, relaxation time and Gibb's free energy increase with increase in concentration of DMF, and decreases with increase in temperature. Acoustic impedance decreases with increase in concentration of DMF, and also with increase in temperature. So acoustic parameters are highly affected at different temperatures due to dipole-dipole and dipole-induced dipole interaction between molecules. Dispersive forces are also found to exist between the components of the mixture.

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