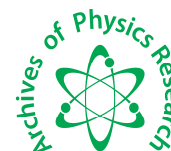




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Ultrasonic Study of Ternary Liquid Mixture Containing Substituted Benzene

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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 Nitrobenzene at different temperatures (288K, 298K and 308K) for a constant frequency (2 MHz). Acoustic parameters like adiabatic compressibility (β), free length (L_f), free volume (V_f), available volume (V_a), Vander Waals constant (b), internal pressure (π_i), relaxation time (τ), acoustic impedance (Z), and Gibb's free energy (ΔG) for the solution have been computed. The excess values of the parameters are also evaluated and discussed.

Key Words: Ultrasonic velocity, adiabatic compressibility, internal pressure, free volume, Vander Waals constant, Gibb's free energy.

INTRODUCTION

Ultrasonic investigations of liquid mixtures consisting of polar and non-polar components enable to understand the molecular interactions and structural behavior of molecules in the mixture [1]. 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 the mixture, ultrasonic velocity together with density and viscosity are measured at different temperature and different concentrations of the components in the mixture keeping the frequency constant.

The trends of variation of the derived thermo-acoustic properties such as isentropic compressibility, available volume, acoustic impedance, intermolecular free length and their excess properties which are obtained from the measured ultrasonic velocity and density of the mixture provide valuable information about the molecular environments and could be helpful in assessing the degree of association between the molecules [2-4].

The present paper deals with the study of different parameters of the mixture containing DMF, Cyclohexane and nitrobenzene at a constant ultrasonic frequency but different concentration of the components and at different temperatures.

DMF (C_3H_7NO) is a versatile compound. It is a non-aqueous solvent which has no hydrogen bonding in pure state. It acts as an aprotic, protophilic medium with high dielectric constant and is a dissociating solvent. DMF is a polar molecule. When it is associated with substituted benzenes, in addition to the intramolecular dipole-dipole interaction there is intermolecular dipole-induced dipole interaction. The intermolecular interaction seems to be stronger than intramolecular interaction. This leads to a decrease of volume, when concentration of DMF increases. In addition, the relatively small size of DMF and its linear aliphatic configuration may be another factor contributing to the volume contraction of the mixtures. DMF and nitrobenzene are characterized by the interaction of the π -electron cloud of benzene ring with the delocalized π -electron cloud over the nitrogen and oxygen atoms of the nitro-group of nitrobenzene resulting in the formation of the weak interaction between them. Cyclohexane has a closed chain structure and is non-polar in nature. Hence it is not involved in any interaction with DMF or nitrobenzene. However

dispersion forces caused by correlated movements of electrons in interactive molecules are possible between cyclohexane and the other components.

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 mole fraction of second component at 0.4.

(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°C to 85°C with an accuracy of $\pm 0.1\text{K}$ 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 10ml 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

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

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,

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

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.

The following thermodynamic parameters were calculated.

(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 [5],

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

(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 [6]

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

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 [7]

$$V_f = (M_{\text{eff}} \cdot U / k \cdot \eta)^{3/2} \text{ ----- (3)}$$

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 x 10⁹.

(iv) Internal Pressure (Π_i):-

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 of force of attraction and force of repulsion between the molecules. It is calculated by using the relation [8],

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

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 4.281x10⁹. 'T' is the absolute temperature in Kelvin, 'M' 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,

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

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,

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

Where 'U' and 'ρ' are velocity and density of the mixture respectively.

(vii) Gibb's free energy:-

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

$$\Delta G = KT \cdot \text{Ln} (KT\tau/h) \text{ ----- (7)}$$

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.

(viii) Available volume:-

The available volume is calculated by using the relation

$$V_a = (V_m - V_o) = V_m \cdot [1 - U/U_\infty] \text{ ----- (8)}$$

Where, V_m = M/ρ, is the molar volume, U = velocity, V_o = M/ρ_o = molar volume at absolute zero temperature and U_∞ = Schaaf's limiting value taken as 1600 m/s for liquids.

(ix) Vander Waal's constant:-

Vander Waal's constant is calculated by using the relation [9]

$$b = (M/\rho) \cdot [1 - (RT/MU^2)] \{ (1 + MU^2/3RT)^{1/2} - 1 \} \text{ ----- (9)}$$

Where, M = molecular weight, R = 8.3143 JK⁻¹mol⁻¹, is the gas constant, ρ = density.

The excess values of parameters has been calculated by using the relation

$$A^E = A_{\text{exp}} - A_{\text{id}}$$

Where, $A_{\text{exp}} = \sum A_i X_i$, $i = 1$ to n , A_i is any acoustical parameters and X_i is the mole fraction of the liquid component.

RESULT AND DISCUSSION

The tables 1 and 2 provide the experimentally determined values of density (ρ), viscosity (η) and ultrasonic velocity (U) of pure liquids and for the ternary system at 288 K, 298 K and 308 K. Adiabatic compressibility, free length, internal pressure, free volume, available volume, Vander waal's constant, viscous relaxation time, Gibbs' free energy and acoustic impedance are tabulated in tables 3, 4 and 5. The excess vales of adiabatic compressibility, free length, free volume and internal pressure are listed in tables 6 and 7.

TABLE – 1: Density (ρ), Viscosity (η) and velocity (U) of pure liquids at 288k, 298k and 308k.

Organic Liquids	Density (ρ) Kg.m ⁻³			Viscosity (η) 10 ⁻³ N.s.m ⁻²			Velocity (U) m.s ⁻²		
	288K	298K	308K	288K	298K	308K	288K	298K	308K
DMF	948.96	943.05	938.62	1.0063	0.8267	0.7007	1510	1467	1429.3
Cyclohexane	779.63	773.45	766.75	1.0800	0.8266	0.6259	1299.9	1252	1206
Nitrobenzene	1206.50	1196.50	1193.0	1.8846	1.4973	1.2087	1490	1456	1421

TABLE – 2: Density (ρ), Viscosity (η) and velocity (U) of the ternary mixture, DMF + Cyclohexane + Nitrobenzene at 288k, 298k and 308k.

Mole fraction		Density (ρ) Kg.m ⁻³			Viscosity (η) 10 ⁻³ N.s.m ⁻²			Velocity (U) m.s ⁻²		
X ₁	X ₂	288K	298K	308K	288K	298K	308K	288K	298K	308K
0.0000	0.6000	869.94	864.22	859.31	1.0233	0.9333	0.8353	1284.2	1245.9	1210.2
0.0999	0.4999	865.48	862.36	858.46	0.9948	0.9034	0.8295	1297.1	1259.8	1221.3
0.1998	0.4001	862.69	857.71	853.81	0.9728	0.8938	0.8137	1308.8	1275.1	1239.5
0.3001	0.3000	857.12	853.99	850.09	0.9512	0.8854	0.7973	1325.6	1294.2	1252.2
0.4000	0.1999	846.91	844.70	840.80	0.9323	0.8701	0.7762	1343.6	1310	1273.6
0.4998	0.1001	836.51	835.34	833.37	0.9169	0.8548	0.7556	1362.8	1332.5	1287
0.5997	0.0000	827.69	826.61	819.93	0.8880	0.8323	0.7355	1379.5	1352.5	1313.1

TABLE – 3 : Adiabatic compressibility (β), Free length (L_f) and internal pressure (Π_i) of the ternary mixture, DMF + Cyclohexane + Nitrobenzene at 288k, 298k and 308k.

Mole fraction		Adiabatic compressibility (β) (10 ⁻¹⁰ N ⁻¹ .m ²)			Free length (L_f) (10 ⁻¹⁰ m)			Internal pressure (Π_i) (x 10 ⁶ N.m ⁻²)		
X ₁	X ₂	288K	298K	308K	288K	298K	308K	288K	298K	308K
0.0000	0.6000	6.9702	7.4544	7.9457	0.5175	0.5407	0.5697	343.86	343.47	339.48
0.0999	0.4999	6.8675	7.3065	7.8097	0.5137	0.5353	0.5648	355.48	354.81	355.81
0.1998	0.4001	6.7670	7.1709	7.6233	0.5099	0.5303	0.5581	370.10	370.46	369.41
0.3001	0.3000	6.6395	6.9911	7.5021	0.5051	0.5236	0.5536	384.95	387.97	385.66
0.4000	0.1999	6.5407	6.8985	7.3323	0.5013	0.5201	0.5473	400.77	404.99	399.73
0.4998	0.1001	6.4368	6.7422	7.2444	0.4973	0.5142	0.5440	419.12	423.07	417.65
0.5997	0.0000	6.3488	6.6134	7.0734	0.4939	0.5093	0.5376	437.98	442.73	434.19

TABLE – 4: Free volume (V_f), available volume (V_a) and Vander Waals' constant (b) of the ternary mixture, DMF + Cyclohexane + Nitrobenzene at 288k, 298k and 308k.

Mole fraction		Free volume (V_f) (10 ⁻⁷ m ³ .mol ⁻¹)			Available volume (V_a) (10 ⁻⁷ m ³ .mol ⁻¹)			Vander Waals' constant (b) (10 ⁻⁷ m ³ .mol ⁻¹)		
X ₁	X ₂	288K	298K	308K	288K	298K	308K	288K	298K	308K
0.0000	0.6000	1.7691	1.9409	2.1943	0.1236	0.1244	0.1251	0.1233	0.1241	0.1248
0.0999	0.4999	1.7439	1.9289	2.0927	0.1184	0.1188	0.1194	0.1182	0.1186	0.1191
0.1998	0.4001	1.6962	1.8520	2.0436	0.1130	0.1137	0.1142	0.1128	0.1134	0.1139
0.3001	0.3000	1.6527	1.7755	1.9776	0.1079	0.1083	0.1088	0.1077	0.1081	0.1086
0.4000	0.1999	1.5986	1.7072	1.9423	0.1033	0.1036	0.1041	0.1031	0.1034	0.1038
0.4998	0.1001	1.5333	1.6470	1.8811	0.0986	0.0988	0.0990	0.0984	0.0985	0.0988
0.5997	0.0000	1.4913	1.5954	1.8374	0.0936	0.0937	0.0945	0.0934	0.0935	0.0943

When concentration of DMF increases at a particular temperature, the experimental density of the mixture decreases, where as the velocity increases, hence adiabatic compressibility decreases. When temperature increases at fixed concentration of DMF, velocity as well as density decreases hence adiabatic compressibility increases [fig-1]. These variations in adiabatic compressibility indicate variation in the molecular forces with concentration and temperature.

The variation of ultrasonic velocity in a solution depends upon the increase or decrease of intermolecular free length after mixing the components. As the ultrasonic velocity for the ternary mixtures increases with increasing mole fraction of DMF, the adiabatic compressibility, free length and free volume show a reverse trend.

Intermolecular free length is the distance between the surfaces of two molecules. It depends on intermolecular as well as intramolecular interaction existing in and between components of the mixture. When adiabatic compressibility decreases, free length decreases and vice versa [10-11]. Variation in free length indicates variation in the molecular forces in the mixture, which depends on the experimental density as well as the temperature of the mixture [fig-2].

Since the intermolecular interaction seems to be stronger than intramolecular interaction, it leads to a decrease of volume when the concentration of DMF increases. When temperature increases, there is reduction in molecular interaction [fig-3 and 4]. This reduces the cohesive force or internal pressure and hence free volume increases [12].

Available volume is a direct measure of the compactness and strength of bonding between the molecules of the liquid mixture. When temperature increases, V_m increases and hence V_a increases. With increase in concentration of DMF, V_m decreases and hence V_a decreases. This has been given in table - 4.

Vander Waals' constant (b), called the co-volume in the Vander Waals' equation varies in a similar manner to that of available volume. Concentration remaining constant as the temperature increases, 'b' increases. Temperature remaining constant, as the concentration of DMF increases, 'b' decreases. This has also been given in table - 4.

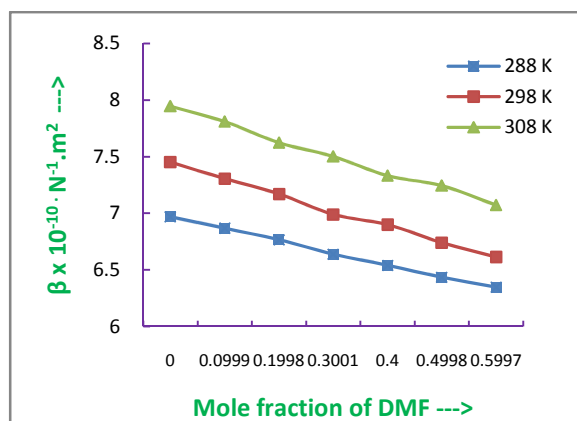


Fig-1: Variation of adiabatic compressibility with mole fraction

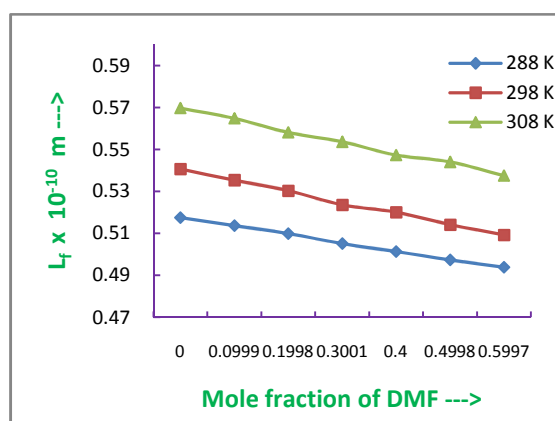


Fig-2: Variation of free length with mole fraction

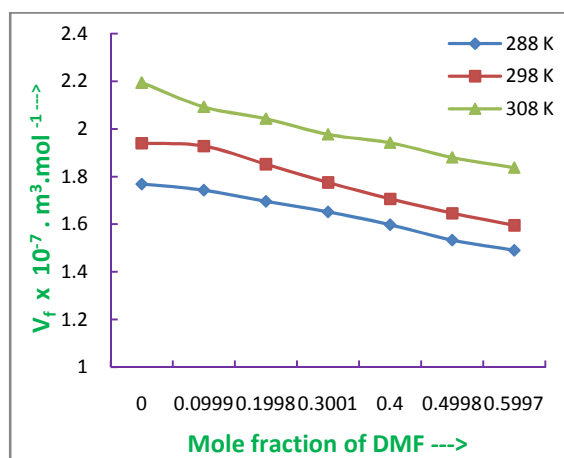


Fig-3: Variation of free volume with mole fraction

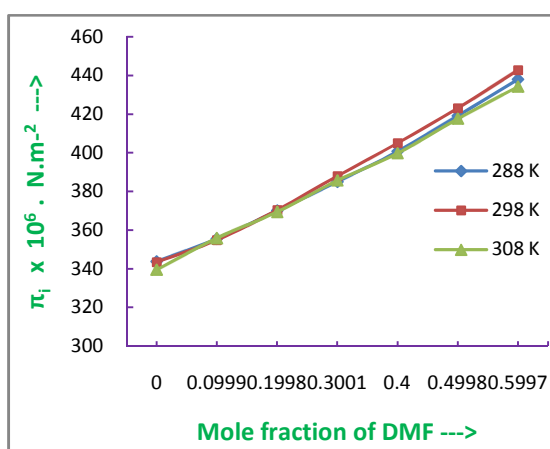


Fig-4: Variation of internal pressure with mole fraction.

TABLE – 5: Viscous relaxation time (τ), Gibb's free energy (ΔG) and acoustic impedance (Z) of the ternary mixture, DMF + Cyclohexane + Nitrobenzene at 288k, 298k and 308k.

Mole fraction		Viscous relaxation time (τ) ($\times 10^{-12}$ s)			Gibb's free energy(ΔG) ($\times 10^{-20}$ k.J.mol ⁻¹)			Acoustic impedance(Z) ($\times 10^6$ Kg.m ² .s ⁻¹)		
X ₁	X ₂	288K	298K	308K	288K	298K	308K	288K	298K	308K
0.0000	0.6000	0.9510	0.9276	0.8849	0.6925	0.7204	0.7386	1.1172	1.0767	1.0399
0.0999	0.4999	0.9109	0.8800	0.8637	0.6754	0.6987	0.7282	1.1226	1.0864	1.0484
0.1998	0.4001	0.8777	0.8546	0.8271	0.6606	0.6867	0.7098	1.1291	1.0937	1.0583
0.3001	0.3000	0.8421	0.8253	0.7975	0.6442	0.6723	0.6943	1.1362	1.1052	1.0645
0.4000	0.1999	0.8131	0.8003	0.7588	0.6302	0.6596	0.6732	1.1379	1.1066	1.0708
0.4998	0.1001	0.7869	0.7684	0.7298	0.6172	0.6429	0.6566	1.14	1.1131	1.0725
0.5997	0.0000	0.7517	0.7339	0.6936	0.5990	0.6240	0.6350	1.1418	1.118	1.0767

Relaxation time is the time taken for the excitation energy to appear as translational energy and it depends on temperature and impurities. In the present case, relaxation time increases with increase in concentration of DMF and decreases with increase in temperature [fig-5]. The former indicates the presence of molecular interaction in the mixture, where as the latter shows the instantaneous conversion of excitation energy to translational energy, when temperature is increased.

Gibb's free energy increases with increase in concentration of DMF and decrease with increasing temperature [fig-6]. The increase in Gibb's free energy indicates the need for shorter time for the co-operative process to take place or for the rearrangement of molecules in the mixture. This indicates the easier flow of ternary mixture compared with the behavior of pure components.

Acoustic impedance is the ratio of the effective sound pressure at a point to the effective particle velocity at that point. The pressure is measured by the totality of the forces of dispersion, repulsion, ionic and dipolar. In the present investigation, acoustic impedance increases with increase in concentration of DMF [fig-7]. This indicates the possibility of strong molecular interaction between the components of the mixture [13-15].

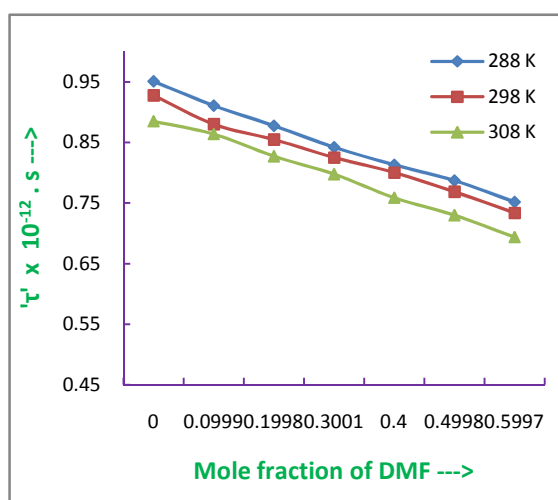


Fig-5: Variation of relaxation time with mole fraction

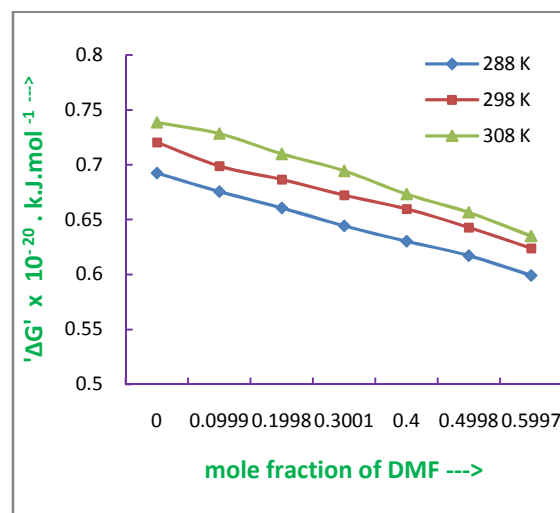


Fig-6: Variation of Gibbs free energy with mole Fraction

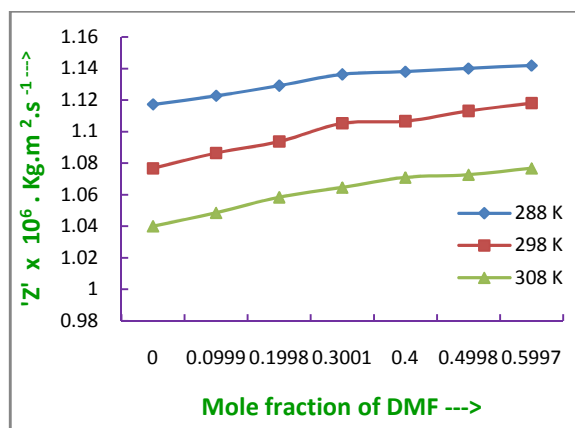


Fig-7: Variation of acoustic impedance with mole fraction

TABLE – 6: Excess values of adiabatic compressibility (β^E) and free length (L_r^E) of the ternary mixture, DMF + Cyclohexane + Nitrobenzene at 288k, 298k and 308k.

Mole fraction		Excess adiabatic compressibility (β^E) ($10^{-10} \text{N}^{-1} \cdot \text{m}^2$)			Excess free length (L_r^E) (10^{-10}m)		
X_1	X_2	288K	298K	308K	288K	298K	308K
0.0000	0.6000	1.6939	1.7897	1.8683	0.0742	0.0773	0.0806
0.0999	0.4999	1.5032	1.5442	1.6268	0.0662	0.0673	0.0708
0.1998	0.4001	1.3136	1.3098	1.3337	0.0572	0.0581	0.0598
0.3001	0.3000	1.0962	1.0304	1.1049	0.0480	0.0492	0.0504
0.4000	0.1999	0.9094	0.8402	0.8296	0.0378	0.0382	0.0393
0.4998	0.1001	0.7168	0.5857	0.6356	0.0258	0.0277	0.0300
0.5997	0.0000	0.5408	0.3592	0.3590	0.0179	0.0182	0.0196

TABLE – 7: Excess values of free volume (V_r^E) and internal pressure (Π_i^E) of the ternary mixture, DMF + Cyclohexane + Nitrobenzene at 288k, 298k and 308k.

Mole fraction		Excess free volume (V_r^E) ($10^{-7} \text{m}^3 \cdot \text{mol}^{-1}$)			Excess internal pressure (Π_i^E) ($\times 10^6 \text{N} \cdot \text{m}^{-2}$)		
X_1	X_2	288K	298K	308K	288K	298K	308K
0.0000	0.6000	0.6587	0.4043	0.0829	-105.95	-72.09	-46.06
0.0999	0.4999	0.6124	0.3736	-0.0274	-100.49	-67.49	-37.17
0.1998	0.4001	0.5433	0.2776	-0.0858	-92.16	-58.70	-31.14
0.3001	0.3000	0.4781	0.1817	-0.1613	-83.66	-48.12	-22.53
0.4000	0.1999	0.4029	0.0948	-0.2053	-74.00	-37.83	-15.91
0.4998	0.1001	0.3164	0.0156	-0.2754	-61.88	-26.58	-5.50
0.5997	0.0000	0.2533	-0.0546	-0.3278	-49.18	-13.65	3.58

In order to understand the nature of molecular interactions between the components of the ternary liquid mixture, it is of interest to discuss the same in terms of excess parameters rather than the actual values. It is learnt that the dispersion forces are responsible for weak interaction between unlike molecules. This leads to positive excess value of the parameters, while dipole-dipole, dipole-induced dipole, charge transfer interaction and hydrogen bonding between unlike molecules yield negative excess values of the parameters [16]. The negative excess values account for closely packed molecules and strong molecular interaction.

The sign of excess adiabatic compressibility and excess free length play a vital role in assessing the compactness due to molecular interaction in liquid mixtures. Positive excess adiabatic compressibility as well as positive excess free length decreases with increase of concentration and increases with increase of temperature [fig-8 and 9]. This indicates that there is a strong molecular interaction between unlike molecules as the concentration of DMF increases and the interaction decreases as temperature increases [17].

Positive value of excess volume decreases with concentration showing a stronger interaction between the molecules [fig-10]. Negative excess internal pressure increases with concentration also showing a stronger interaction between the molecules [fig-11]. Positive value of excess Gibbs free energy and excess relaxation time also decreases with concentration, confirming the above fact [fig-12 and 13].

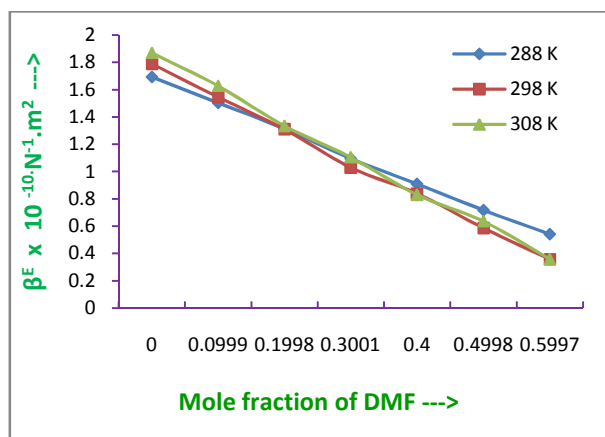


Fig-8: Variation of excess adiabatic compressibility with mole fraction

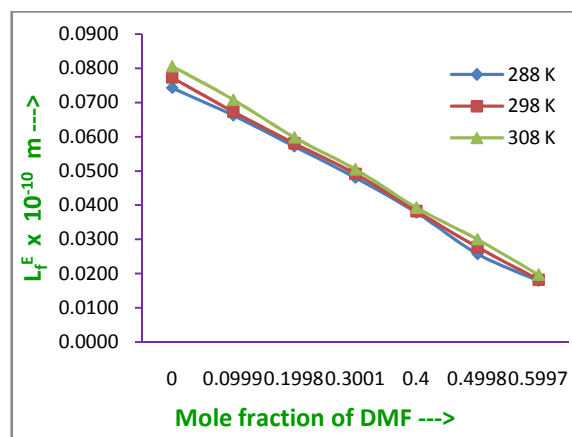


Fig-9: Variation of excess free length with mole fraction

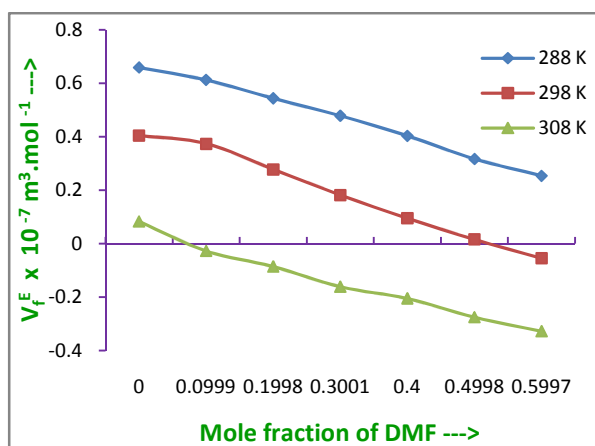


Fig-10: Variation of excess free volume with mole fraction.

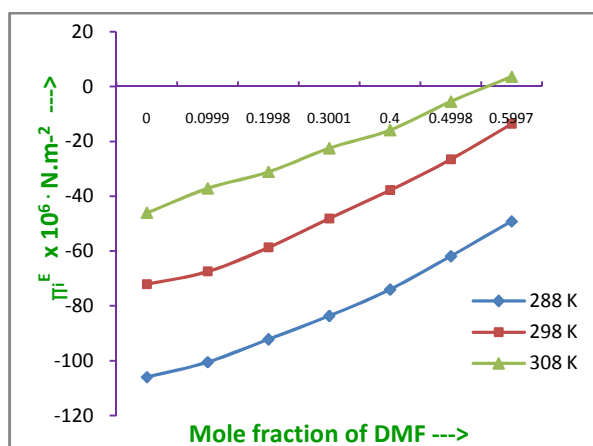


Fig-11: Variation of excess internal pressure with mole fraction.

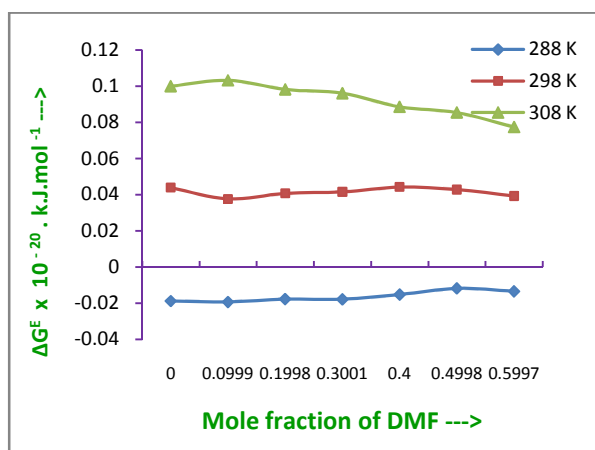


Fig-12: Variation of excess Gibbs free energy with mole fraction

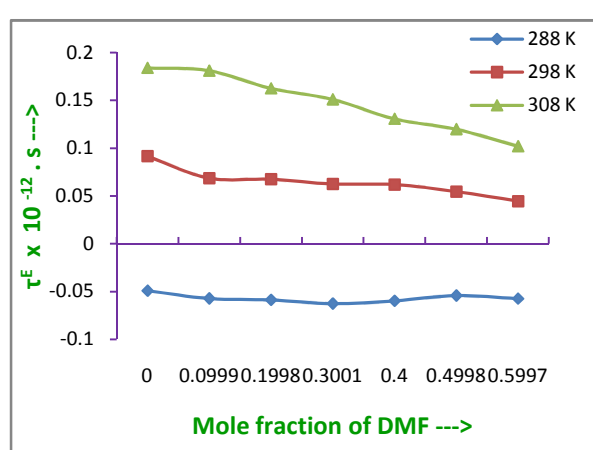


Fig-13: Variation of excess relaxation time with mole fraction.

CONCLUSION

From the above study, it is seen that there exists molecular association between the components of the mixture. In particular the change in the excess parameters indicate that there is a strong molecular interaction between unlike molecules as the concentration of DMF increases and the interaction decreases as temperature increases.

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