



Study of Molecular Interaction in Binary Mixture of Dimethyl Acetamide with Diethyl ether using Ultrasonic and Viscosity Probes

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Abstract

The ultrasonic velocity (U), density (ρ) and coefficient of viscosity (η) of binary mixture of dimethyl acetamide (DMAC) and diethyl ether at temperature 308K have been measured at different frequencies (2MHz, 4MHz, 6MHz and 8MHz). Adiabatic compressibility (K_s), intermolecular free length (L_f), free volume (V_f), internal pressure (π_i) and their respective excess values have been computed for entire range of concentration and are interpreted to explain molecular interaction occurring in the liquid mixture. Relaxation time (τ), excess enthalpy (H^E) and absorption coefficient (α/f^2) have been calculated and discussed.

Key words: Internal pressure, relaxation time, excess enthalpy and absorption coefficient.

Introduction

The acoustical study of liquids plays an important role to understand the nature and strength of molecular interactions. Ultrasonic velocities have been adequately employed in order to explain the nature of molecular interaction in pure liquids, binary and ternary mixture¹⁻¹⁰. The investigation regarding the study of molecular interaction in binary liquid mixture with dimethyl acetamide (DMAC) and diethyl ether as the components is of particular interest, because DMAC is a dipolar aprotic solvent with high boiling point and having good thermal and chemical stability. It has large dipole moment and dielectric constant ($\mu = 3.7$ D and $\epsilon = 37.8$). It is used in industry and medicine. It is also used as solvent for the production of acrylic fibres, elasthane fibres, polyimide resins and various pharmaceuticals. It is an excellent proton donor as well as proton acceptor and hence it is strongly associated through intermolecular hydrogen bond. It is highly soluble in a variety of polar and nonpolar solvents and readily suitable to explore solute solvent interactions. Diethyl ether is a non-polar solvent. It has small dipole moment and dielectric constant ($\mu = 1.15$ D and $\epsilon = 4.34$). It is used in the production of cellulose plastics.

An attempt has been taken to explain the molecular interaction in binary mixture of dimethyl acetamide (DMAC) and diethyl ether at different frequencies at 308K. Departure from linearity in the acoustical parameters versus mole fraction in binary mixture of DMAC is considered as an indication of the presence of molecular interaction between different liquid molecules¹¹⁻²⁰. The physiochemical properties of binary mixture is studied by nonlinear variation of ultrasonic velocity and other acoustical parameters with structural changes occurring in a liquid and the liquid mixture.

Theory: Using the measured data the acoustical parameters such as adiabatic compressibility (K_s), intermolecular free length (L_f), free volume (V_f) and internal pressure (π_i) have been calculated from the following equations.

$$K_s = (U^2 \rho)^{-1} \quad (1)$$

$$L_f = k (K_s)^{1/2} \quad (2)$$

$$V_f = (MU/K\eta)^{3/2} \quad (3)$$

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

Where k , M and R are temperature dependent constant, effective molecular weight and universal gas constant respectively. K is a temperature independent constant equal to 4.28×10^9 where b is the cubic packing factor equal to 2 for all liquid mixtures.

Excess values of the above acoustical parameters have been calculated from the following relations.

$$A^E = A_{exp} - (X_1 A_1 + X_2 A_2) \quad (5)$$

Where X_1 and X_2 are mole fractions of DMAC and diethyl ether respectively and A is any acoustical parameter.

Relaxation time(τ), excess enthalpy(H^E) and absorption coefficient(α/f^2), have been calculated from the following relations.

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

$$H^E = (X_1 \pi_{i1} V_{m1} + X_2 \pi_{i2} V_{m2}) - \pi_i V_m \quad (7)$$

$$\alpha/f^2 = 2\pi^2 \tau / U \quad (8)$$

Material and Methods

Liquid mixtures of various concentrations in mole fraction were prepared by taking chemicals of analytical reagent(AR) and

spectroscopic reagent(SR) grades with minimum assay of 99.9% (E-Merck Ltd, India) which were used as such without further purification. Liquid mixture of different mole fractions were prepared on concentration scale with a precision 0.0001g using an electronic digital balance. The density of liquid mixture was determined by a specific gravity bottle of 10ml capacity. The specific gravity bottle with the liquid mixture was immersed in a temperature controlled water bath. The density was determined using the relation.

$$\rho_2 = (w_2/w_1) \rho_1 \quad (9)$$

Where w_1 , w_2 , ρ_1 and ρ_2 are mass of distilled water, mass of liquid mixture, density of distilled water and density of liquid mixture respectively.

The coefficient of viscosity (η) of pure liquids and liquid mixture was determined by an Ostwald's viscometer calibrated with distilled water. The Ostwald's viscometer containing the liquid was immersed in a temperature controlled water bath and the time of flow was measured by a Racer stop watch. The coefficient of viscosity was determined using the relation.

$$\eta_2 = (\eta_1 / t_1 \rho_1) t_2 \rho_2 \quad (10)$$

Where η_1 , η_2 , t_1 , and t_2 are coefficient of viscosity of distilled water, coefficient of viscosity of liquid mixture, time of flow of distilled water and time of flow of liquid mixture respectively.

The velocity of ultrasonic waves in the binary liquid mixture was determined by using a multi frequency interferometer (Model M-82S) with a high degree of accuracy operating at different frequencies supplied by Mittal Enterprises, New Delhi. The measuring cell of the interferometer is a specially designed double walled vessel with provision to circulate water at constant temperature i.e.308K. The high frequency generator excites a quartz crystal fixed at the bottom of the vessel, at its resonant frequency. A fine micrometer screw at the top of the cell is used to raise or lower the reflector plate in the liquid through a known distance. The measuring cell is connected to the output terminals of the high frequency generator through a cable. Ultrasonic waves normal to quartz crystal are reflected

from the reflector plate. Stationary waves are formed in the region between reflector plate and the quartz crystal. The micrometer is slowly moved till a number of maximum readings (N) of the anode current is passed. The total distance (d) moved by the micrometer is noted. The velocity of ultrasonic waves in the binary liquid mixture was determined using the relation.

$$U = \lambda f \quad (11)$$

Where $\lambda = 2d/N =$ wavelength of the ultrasonic waves in the binary liquid mixture and f is the frequency of the generator.

Results and Discussion

Density ρ , coefficient of viscosity η and ultrasonic velocity U increase with the increase in mole fraction of DMAC as shown in table-1 and in figure-1, figure-2 and figure-3 respectively. The increase in density with the increase in mole fraction of DMAC indicates that dipole-induced dipole interactions increases with the increase in concentration of DMAC in the liquid mixture. The increase in ultrasonic velocity at a particular frequency indicates the presence of dipole-induced dipole interactions in the liquid mixture. The increase in coefficient of viscosity with the increase in mole fraction of DMAC indicates the presence of solute-solvent interactions in the liquid mixture.

DMAC is a polar molecule and when it is associated with nonpolar solvent diethyl ether, the diethyl ether molecule tends to break the DMAC- DMAC dipolar association and releases several DMAC dipoles. Free DMAC dipoles would induce moments in the neighboring diethyl ether molecules resulting dipole-induced dipole interactions in the liquid mixture. Consequently the binary liquid mixture has dipole-dipole interactions between DMAC molecules as well as dipole-induced dipole interactions between DMAC and diethyl ether molecules. This leads to contraction in volume and it causes decrease in adiabatic compressibility K_s and intermolecular free length L_f with the increasing molar concentration of DMAC which are depicted in table-2 and shown graphically in figures-4 and 5.

Table-1
Values of density, coefficient of viscosity and ultrasonic velocity at 308K

Mole Fraction of DMAC(X_1)	ρ Kgm ⁻³	$\eta \times 10^{-3}$ Nsm ⁻²	U ms ⁻¹			
			2MHz	4MHz	6MHz	8MHz
0	693	0.279	928	912	904	880
0.219	743	0.346	1032	1024	1008	992
0.327	768	0.387	1094	1076	1068	1056
0.428	792	0.471	1152	1136	1116	1104
0.529	815	0.567	1198	1182	1160	1150
0.631	839	0.631	1252	1240	1224	1216
0.733	862	0.725	1296	1288	1272	1264
0.826	883	0.809	1356	1344	1332	1328
0.917	904	0.863	1416	1396	1386	1372
1	925	0.946	1488	1472	1464	1440

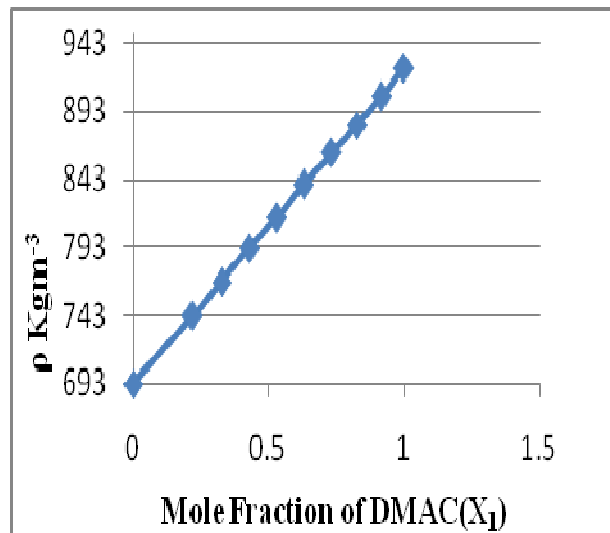


Figure-1
Variation of ρ Versus X_1

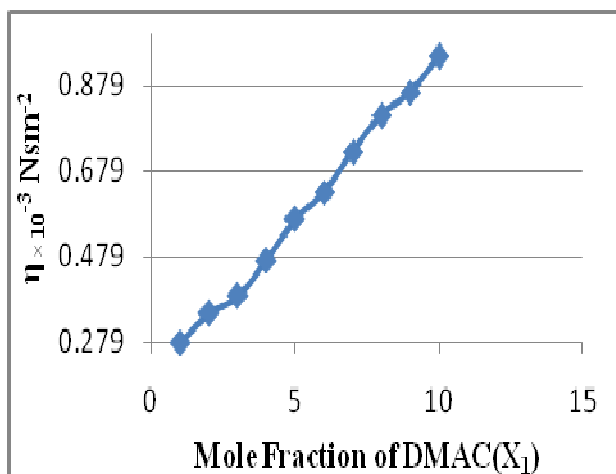


Figure-2
Variation of η Versus X_1

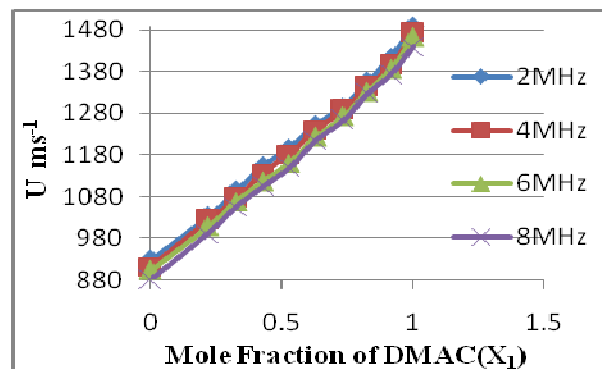


Figure-3
Variation of U Versus X_1

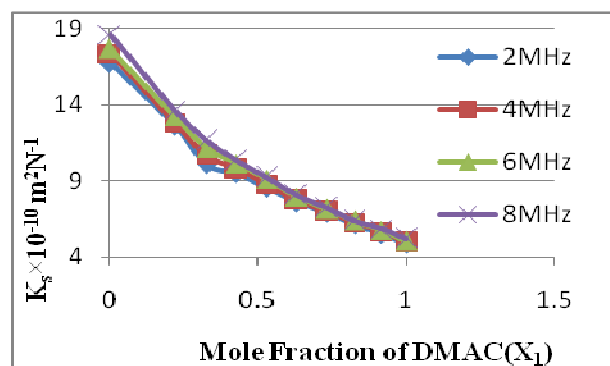


Figure-4
Variation of K_s Versus X_1

Table-3 and figure-6 show that the values of free volume V_f decrease with the increase in mole fraction of DMAC for a particular frequency. The decrease in free volume with the increase in mole fraction of DMAC is because of (i) contraction due to the free volume difference of unlike molecules. (ii) contraction due to the hydrogen bond formation between unlike molecules. (iii) specific interactions between unlike molecules in the binary liquid mixture.

Table-2
Values of adiabatic compressibility and free length at 308K

Mole Fraction of DMAC(X_1)	$K_s \times 10^{-10} \text{ m}^2 \text{ N}^{-1}$				$L_f \times 10^{-10} \text{ m}$			
	2MHz	4MHz	6MHz	8MHz	2MHz	4MHz	6MHz	8MHz
0	16.756	17.3491	17.6575	18.6338	0.8195	0.8338	0.8412	0.8642
0.219	12.6372	12.8355	13.2462	13.6769	0.7116	0.7172	0.7286	0.7403
0.327	10.8794	11.2464	11.4155	11.6765	0.6603	0.6713	0.6688	0.6764
0.428	9.5141	9.784	10.1379	10.3594	0.6175	0.6262	0.6374	0.6443
0.529	8.5492	8.7822	9.1185	9.2778	0.5853	0.5932	0.6045	0.6097
0.631	7.6037	7.7516	7.9556	8.0606	0.552	0.5573	0.5646	0.5683
0.733	6.9068	6.9929	7.1699	7.261	0.5261	0.5294	0.536	0.5394
0.826	6.1591	6.2696	6.383	6.4216	0.4968	0.5012	0.5057	0.5073
0.917	5.517	5.6762	5.7584	5.8765	0.4702	0.4769	0.4804	0.4853
1	4.8826	4.9893	5.044	5.2135	0.4423	0.4471	0.4496	0.4571

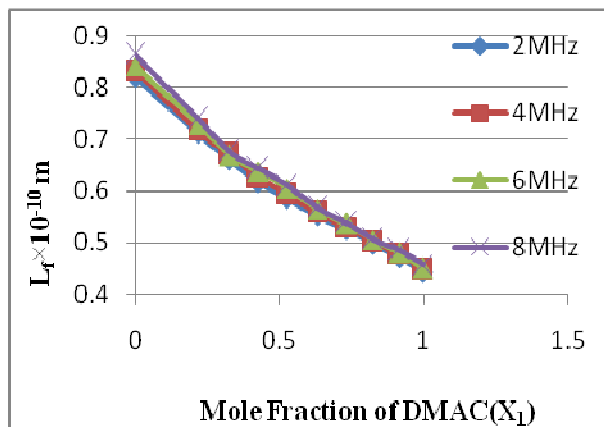


Figure-5
 Variation of L_f Versus X_1

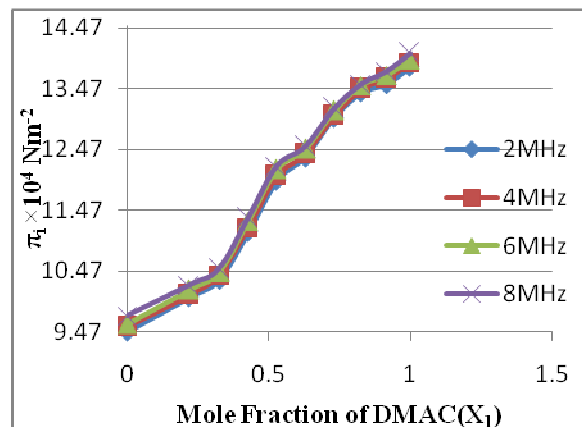


Figure-7
 Variation of π_i Versus X_1

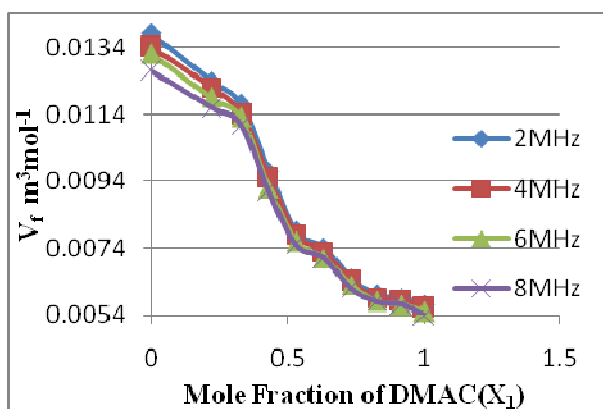


Figure-6
 Variation of V_f Versus X_1

Table-3 and figure-7 show that the values of internal pressure π_i increase with the increase in mole fraction of DMAC for a particular frequency. The increase in internal pressure with the increase in mole fraction of DMAC indicates the increase in cohesive forces in the binary liquid mixture.

With the increase of frequency from 2MHz, to 8MHz ultrasonic velocity decreases at a fixed concentration of DMAC. The decrease in ultrasonic velocity is perhaps due to the decrease in molecular interaction in the binary liquid mixture at higher frequencies. Consequently, the values of adiabatic compressibility, intermolecular free length and internal pressure increase but free volume decreases with the increase in frequency for a particular mole fraction of DMAC.

Table-3

Values of free volume and internal pressure at 308K

Mole Fraction of DMAC(X_1)	$V_f \text{ m}^3 \text{ mol}^{-1}$				$\pi_i \times 10^4 \text{ Nm}^{-2}$			
	2MHz	4MHz	6MHz	8MHz	2MHz	4MHz	6MHz	8MHz
0	0.0138	0.0134	0.0132	0.0127	9.47	9.553	9.595	9.725
0.219	0.0124	0.0122	0.0119	0.0116	10.037	10.072	10.155	10.237
0.327	0.0117	0.0114	0.0113	0.0111	10.319	10.405	10.444	10.503
0.428	0.0097	0.0095	0.0092	0.0091	11.108	11.184	11.284	11.345
0.529	0.0079	0.0078	0.0076	0.0075	11.95	12.03	12.144	12.196
0.631	0.0074	0.0073	0.0071	0.0071	12.336	12.395	12.476	12.517
0.733	0.0065	0.0064	0.0063	0.0062	12.988	13.029	13.11	13.152
0.826	0.006	0.0059	0.0058	0.0058	13.404	13.463	13.524	13.544
0.917	0.0059	0.0058	0.0057	0.0057	13.541	13.637	13.686	13.756
1	0.0057	0.0056	0.0055	0.0054	13.826	13.901	13.939	14.055

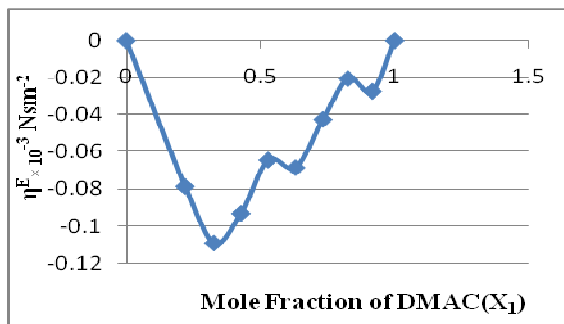


Figure-8
 Variation of η^E Versus X_1

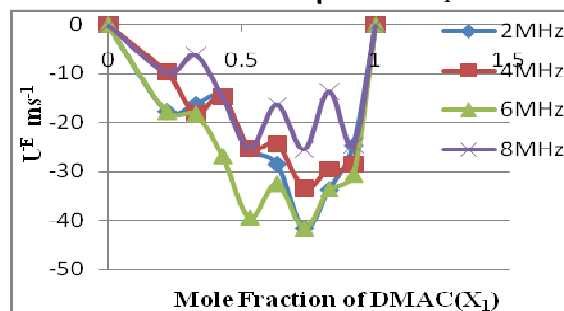


Figure-9
 Variation of U^E Versus X_1

Table-4, figure-8 and figure-9 show that the excess values of coefficient of viscosity η^E and excess values of U^E are negative for the entire range of concentration of DMAC for all frequencies. The negative excess values of η^E indicate the presence of dispersion, induction and dipolar forces²¹ and the negative excess values of U^E indicate the presence of dispersion interactions in the binary liquid mixture.

It is evident from table-5, figure-10 and figure-11 that the values of excess adiabatic compressibility and excess free length are negative for the entire range of concentration of DMAC for frequencies 2MHz, 4MHz, 6MHz and 8MHz. The negative excess values of adiabatic compressibility (K_s^E) indicate the existence of strong interactions in the binary liquid mixture. It also indicates tightly packed molecules in the liquid mixture. The negative excess values of free length (L_f^E) predict the existence of strong interactions in the binary liquid mixture due to charge transfer, dipole-induced dipole, dipole-dipole interactions, interstitial accommodation and orientational ordering.

Table-4
 Excess values of coefficient of viscosity and velocity at 308K

Mole Fraction of DMAC(X_1)	$\eta^E \times 10^{-3} \text{ Nsm}^{-2}$	$U^E \text{ ms}^{-1}$			
		2MHz	4MHz	6MHz	8MHz
0	0	0	0	0	0
0.219	-0.0787	-17.712	-9.728	-17.736	-9.76
0.327	-0.1093	-16.192	-18.208	-18.216	-6.24
0.428	-0.0932	-14.752	-14.768	-26.776	-14.8
0.529	-0.0645	-25.312	-25.328	-39.336	-25.36
0.631	-0.0686	-28.432	-24.448	-32.456	-16.48
0.733	-0.0426	-41.552	-33.568	-41.576	-25.6
0.826	-0.0206	-33.632	-29.648	-33.656	-13.68
0.917	-0.0273	-24.592	-28.608	-30.616	-20.69
1	0	0	0	0	0

Table-5
 Excess values of adiabatic compressibility and free length at 308K

Mole Fraction of DMAC(X_1)	$K_s^E \times 10^{-10} \text{ m}^2 \text{ N}^{-1}$				$L_f^E \times 10^{-10} \text{ m}$			
	2MHz	4MHz	6MHz	8MHz	2MHz	4MHz	6MHz	8MHz
0	0	0	0	0	0	0	0	0
0.219	-1.5017	-1.7894	-1.6312	-1.9992	-0.0244	-0.031	-0.026	-0.0338
0.327	-1.9772	-2.0437	-2.3519	-2.5502	-0.035	-0.0352	-0.0359	-0.0461
0.428	-2.1433	-2.2577	-2.1033	-2.5118	-0.0397	-0.0412	-0.0353	-0.0448
0.529	-1.909	-2.0112	-1.8488	-2.238	-0.0338	-0.0352	-0.0287	-0.0382
0.631	-1.6434	-1.7811	-1.7251	-2.0863	-0.0286	-0.0316	-0.0286	-0.0381
0.733	-1.1292	-1.2791	-1.2242	-1.517	-0.016	-0.0201	-0.0173	-0.0255
0.826	-0.7727	-0.8529	-0.838	-1.1084	-0.0103	-0.0123	-0.0112	-0.0197
0.917	-0.3343	-0.3216	-0.3148	-0.4322	-0.0025	-0.0014	-0.0008	-0.0047
1	0	0	0	0	0	0	0	0

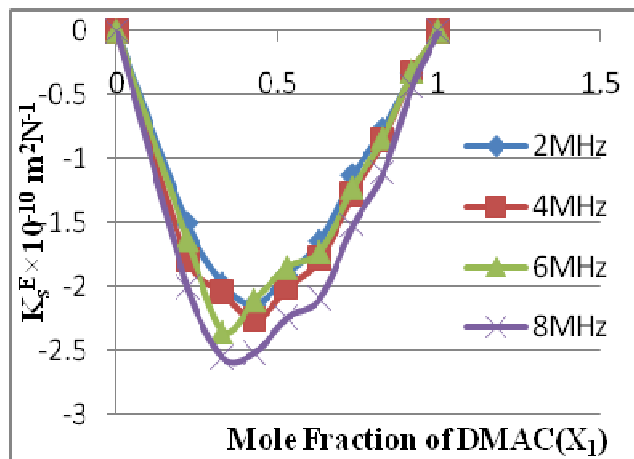


Figure-10
 Variation of K_s^E Versus X_1

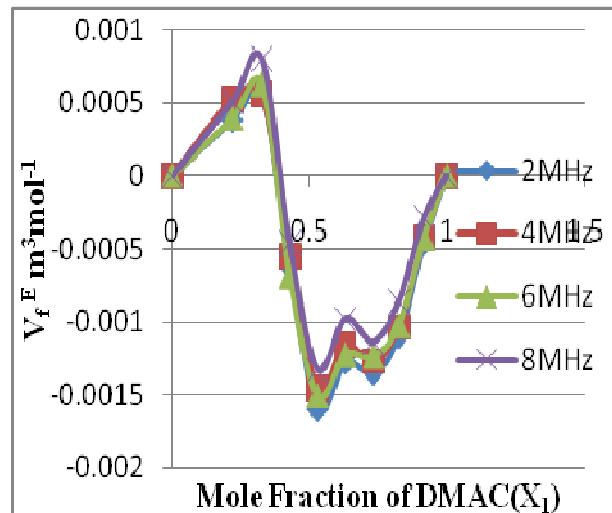


Figure-12
 Variation of V_f^E Versus X_1

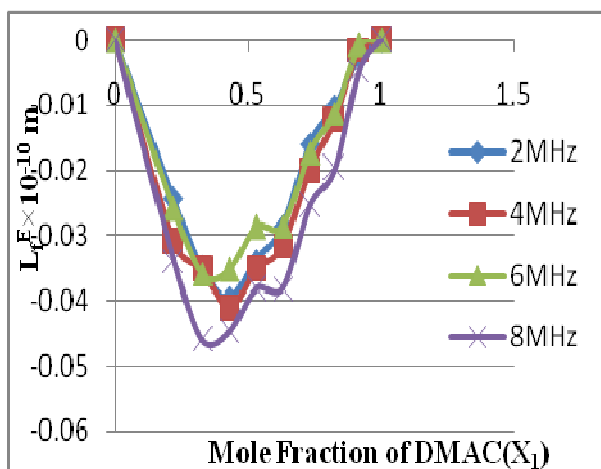


Figure-11
 Variation of L_r^E Versus X_1

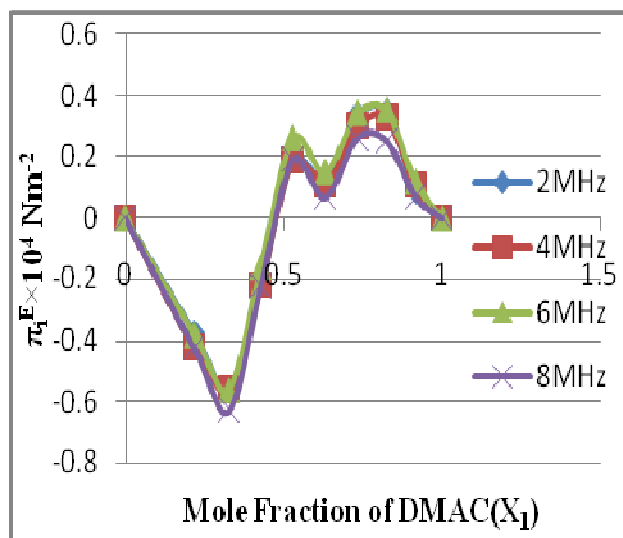


Figure-13
 Variation of π_i^E Versus X_1

Table-6 and figure-12 show that the values of excess free volume V_f^E are positive between 0 to 0.327 mole fraction of DMAC and negative between 0.428 to 1 mole fraction of DMAC. The values of excess free volume are influenced by i. the specific interactions between the component molecules and weak physical forces like dipole-dipole or dipole-induced dipole interactions or vander Waal's forces ii. The dispersive forces, steric hindrance of component molecules, unfavorable geometric fitting and electrostatic repulsion. The former effect leads to contraction of volume and the latter effect leads to expansion of volume. In the present investigation the positive values of V_f^E may be interpreted as the expansion of volume and the negative values of V_f^E may be interpreted as the contraction of volume of the binary liquid mixture of DMAC and diethyl ether. The positive values of V_f^E at lower concentration of DMAC are favorable for the latter effect which accounts for the weak molecular interactions and the negative values of V_f^E at higher concentration of DMAC are favorable for the former effect which accounts for the strong molecular interactions in the binary liquid mixture²².

Table-6 and figure-13 show that the values of excess internal pressure π_i^E are negative within 0 to 0.428 mole fraction of DMAC and positive within 0.529 to 1 mole fraction of DMAC. The negative values of π_i^E at lower concentration of DMAC indicate the presence of dispersion and dipolar forces but the positive values of π_i^E at higher concentration of DMAC indicate the existence of strong interactions in the binary liquid mixture²³.

Tables-4, 5, and 6 show that the excess values of velocity, coefficient of viscosity, adiabatic compressibility, free length, free volume and internal pressure are changed with the increase in frequency due to the decrease in ultrasonic velocity in the binary liquid mixture.

Table-7 and figure-14 show that the relaxation time τ varies nonlinearly with the increase in concentration of DMAC for a fixed frequency. The relaxation time τ increases with the increase in frequency for a fixed mole fraction DMAC. The relaxation time is in the order of 10^{-12} s may be due to the structural relaxation process showing the existence of molecular interactions and in such a case it is suggested that the molecules are rearranged due to co-operative process.

The values of excess enthalpy H^E are positive for 0 to 0.428 mole fraction of DMAC and negative for 0.529 to 1 mole fraction of DMAC for frequencies 2MHz, 4MHz, 6MHz and 8MHz as shown in Table-8 and Figure-15. The positive excess values of H^E at lower concentration of DMAC indicate the existence of dispersion interactions and the negative excess values of H^E at higher concentration of DMAC indicate the presence of strong interactions in the liquid mixture²⁴.

Table-6
 Excess values of free volume and internal pressure at 308K

Mole Fraction of DMAC(X_1)	$V_f^E \text{ m}^3 \text{ mol}^{-1}$				$\pi_i^E \times 10^4 \text{ Nm}^{-2}$			
	2MHz	4MHz	6MHz	8MHz	2MHz	4MHz	6MHz	8MHz
0	0	0	0	0	0	0	0	0
0.219	0.00038	0.00052	0.0004	0.00051	-0.377	-0.423	-0.381	-0.426
0.327	0.00056	0.00056	0.00063	0.0008	-0.565	-0.56	-0.561	-0.628
0.428	-0.00062	-0.00055	-0.00069	-0.00046	-0.216	-0.22	-0.16	-0.223
0.529	-0.0016	-0.00146	-0.00151	-0.00133	0.185	0.186	0.26	0.19
0.631	-0.00128	-0.00116	-0.00123	-0.00098	0.126	0.107	0.149	0.069
0.733	-0.00135	-0.00127	-0.00124	-0.00114	0.334	0.298	0.34	0.262
0.826	-0.0011	-0.00104	-0.00103	-0.00086	0.345	0.328	0.35	0.252
0.917	-0.00046	-0.00043	-0.00043	-0.00029	0.086	0.106	0.117	0.07
1	0	0	0	0	0	0	0	0

Table-7
 Values of relaxation time at 308K

Mole Fraction of DMAC(X_1)	$\tau \times 10^{-12} \text{ s}$			
	2MHz	4MHz	6MHz	8MHz
0	0.623	0.645	0.656	0.693
0.219	0.582	0.591	0.61	0.63
0.327	0.52	0.551	0.575	0.602
0.428	0.597	0.614	0.636	0.65
0.529	0.646	0.663	0.689	0.701
0.631	0.639	0.652	0.669	0.677
0.733	0.667	0.675	0.692	0.701
0.826	0.664	0.676	0.688	0.692
0.917	0.634	0.652	0.662	0.676
1	0.615	0.629	0.636	0.657

Table-8
 Values of excess enthalpy and absorption coefficient at 308K

Mole Fraction of DMAC(X_1)	$H^E \text{ J mol}^{-1}$				$(\alpha/f^2) \times 10^{-15}$			
	2MHz	4MHz	6MHz	8MHz	2MHz	4MHz	6MHz	8MHz
0	0	0	0	0	13.264	13.975	14.348	15.557
0.219	0.036	0.041	0.037	0.042	11.156	11.418	11.972	12.562
0.327	0.054	0.053	0.053	0.06	9.399	10.118	10.65	11.267
0.428	0.017	0.017	0.013	0.019	10.242	10.68	11.251	11.638
0.529	-0.023	-0.021	-0.028	-0.021	10.654	11.092	11.735	12.045
0.631	-0.015	-0.013	-0.017	-0.009	10.09	10.387	10.799	11.013
0.733	-0.035	-0.032	-0.036	-0.028	10.173	10.365	10.761	10.966
0.826	-0.037	-0.035	-0.037	-0.027	9.675	9.937	10.208	10.301
0.917	-0.01	-0.012	-0.013	-0.008	8.853	9.24	9.441	9.733
1	0	0	0	0	8.174	8.442	8.582	9.018

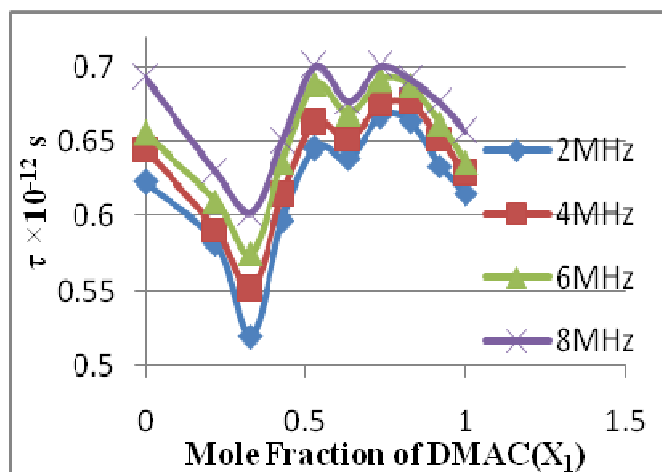


Figure-14
 Variation of τ Versus X_1

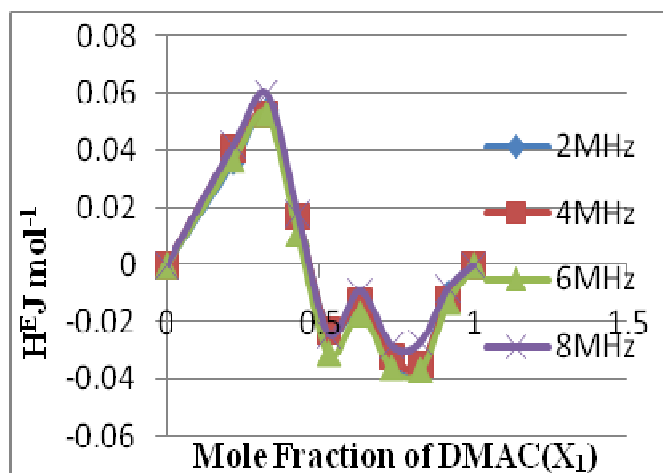


Figure -15
 Variation of H^E Versus X_1

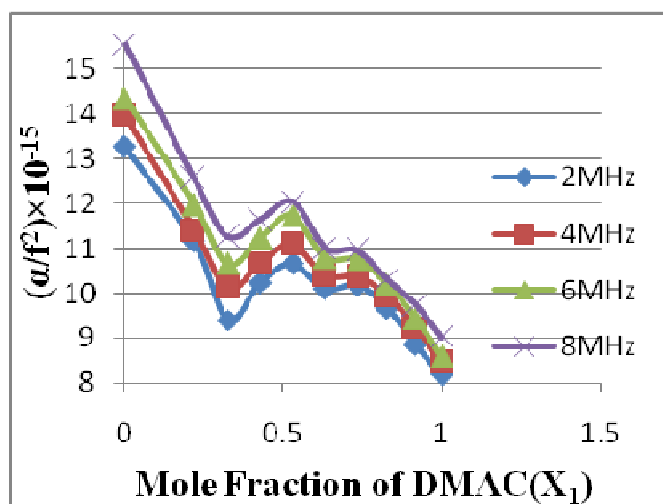


Figure-16
 Variation of α/f^2 Versus X_1

Table-8 and figure-16 show that the values of absorption coefficient α/f^2 decrease non-linearly with the increase in mole fraction of DMAC for a fixed frequency indicating the increase in molecular interaction. The increase in absorption coefficient with the increase in frequency for a fixed concentration of DMAC indicates the reduction in molecular interaction in the binary liquid mixture²⁵ at higher frequencies.

Conclusion

On the basis of the experimental values of density, ultrasonic velocity, related acoustical parameters and their excess values for the binary liquid mixture, it is concluded that there exists dipole-dipole, dipole-induced dipole and dispersion interactions in the binary liquid mixture of DMAC diethyl ether. Further, it is concluded that the molecular interaction increases with the increase in mole fraction of DMAC for a fixed frequency and decreases with the increase in frequency for a fixed concentration of DMAC in the binary mixture.

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