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Ultrasonic Study on Ternary Mixture of Dimethyl Acetamide (DMAC) in Diethyl ether and Acetone

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Abstract

The experimental density (ρ) and ultrasonic velocity (U) for ternary mixture containing dimethyl acetamide (DAMC), diethyl ether and acetone at different frequencies (2MH$_Z$, 4MHz 6MH$_Z$, 8MH$_Z$) have been measured at temperature 308K. These data are used to compute adiabatic compressibility (K_s), intermolecular free length (L_f), acoustic Impedance (Z), molar volume (V_m), molar sound velocity(R), molar compressibility (B), available volume (V_a), Lennard Jones potential repulsive term exponent (n), relative association (R_A), interaction parameter (χ) and some excess thermo acoustic parameters for whole range of concentration *of DMAC and are interpreted to elucidate molecular interaction occurring in the liquid mixture.*

Keywords: Relative association, interaction parameter, available volume, ternary mixture.

Introduction

The investigations of intermolecular interaction in binary and ternary mixtures of polar and non- polar liquids play an important role in the development of molecular sciences. A number of studies have been made on the intermolecular interaction in liquid or solid system by different methods like Ultraviolet, Dielectric constant, Infrared, Electron Paramagnetic Resonance, Raman Effect, Nuclear magnetic resonance, Optical and Ultrasonic methods. Recently ultrasonic method is a powerful technique in providing information relating to the physiochemical properties of liquid or solid system[1-7].

The present investigation is related on thermodynamic properties of ternary liquid mixture of dimethyl acetamide (DMAC) which is a dipolar aprotic solvent with high boiling point and good thermal and chemical stability. The ultrasonic investigation of DMAC is important because of its use in industry and medicine. DMAC 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 non-polar solvents and readily suitable to explore solvent solvent interactions. Diethyl ether is a non-polar liquid used as a solvent in the production of cellulose plastics. Acetone is also an important solvent used in industry and pharmaceuticals. In view of these considerations an attempt has been taken to explain the molecular interaction in ternary liquid mixture consisting of dimethyl acetamide (DMAC), diethyl ether and acetone at different frequencies at constant temperature 308K.

Deviation from linearity in the ultrasonic velocity versus mole fraction in liquid mixture is taken as an indication of the presence of molecular interactions between different components. The physiochemical properties of the ternary liquid mixture can be investigated by the non-linear change of ultrasonic velocity and other acoustic parameters with structural changes occurring in a liquid as its mole fraction is changed in the liquid mixture[7-22].

Material and Methods

The chemicals used were of analytical and spectroscopic grade reagents which were obtained from E Merck Ltd (India) and were used as such without further purification. The ternary mixtures of various concentrations were prepared out of which the mole fraction of diethyl ether was kept fixed arbitrarily at 0.4. The mole fractions of DMAC and acetone were varied between 0 to 0.6 to have the mixtures of various compositions. Liquid mixtures of different mole fractions were prepared with a precision of 0.0001g by a digital balance. Density (ρ**)** of pure liquid and liquid mixture was measured by a specific gravity bottle of 10ml capacity. The ultrasonic velocity **(**U**)** was determined by a multi-frequency interferometer (M-82S) with a high degree of accuracy operating at different frequencies (1MHz, to 8MHz). The temperature of the experimental liquid mixture is kept constant at 308K by circulating water through the double walled measuring cell, coming from an electronically operated water bath at constant temperature.

Theory: The thermo acoustic parameters like adiabatic compressibility (K_s) intermolecular free length (L_f), acoustic impedance (Z), molar volume (V_m) and available volume (V_a) are calculated using the measured data from the following relations.

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

$$L_f = k\,(K_S)^{-1/2} \quad (2)$$

$$Z = \rho U \quad (3)$$

$$V_m = M/\rho \quad (4)$$

$$V_a = (M/\rho)\,[1-(U/U_\infty)] \quad (5)$$

Where k is a temperature dependent constant, M is the molecular mass of the liquid mixture and U_∞=1600 m/s.

Their excess values are calculated from the following relation.

$$A^E = A_{exp} - (X_1A_1 + X_2A_2 + X_3A_3) \quad (6)$$

Where X_1, X_1 and X_3 represent mole fractions of DMAC, diethyl ether and acetone respectively and A_{exp}, A_1, A_2 and A_3 represent experimental parameter of the mixture and individual parameters of solvent components respectively.

Molar sound velocity(R), molar compressibility (B), Lennard Jones potential repulsive term exponent (n), relative association (R_A) and interaction parameter (χ) have been calculated from the following relations.

$$R= (M/\rho)\ U^{1/3} \quad (7)$$

$$B= (M/ \rho)\ (K_s)^{-1/7} \quad (8)$$

$$n= (6\ V_m / V_a) - 13 \quad (9)$$

$$R_A= (\rho / \rho_0)\ (U_0/ U) \quad (10)$$

$$\chi = (U/U_{ideal})^2 -1 \quad (11)$$

Where U_0 and ρ_0 are ultrasonic velocity and density of DMAC respectively.

The ideal mixing velocity U_{ideal} is given by.

$$U_{ideal} =X_1U_1+X_2U_2+ X_3U_3 \quad (12)$$

Results and Discussion

For pure liquids and ternary liquid mixture the experimental values of density and ultrasonic velocity at 308K for frequencies 2MHz, 4MHz, 6MHz and 8MHz were used to calculate the thermo acoustic parameters and the relevant data are presented in Tables 1 to 10 and displayed graphically in figures- 1 to 18.

Table-2, figure-1 and figure-2 show that density ρ and ultrasonic velocity U increase as the concentration of DMAC increases. The structural changes occurring in the ternary mixture with the increase in concentration may cause the increase in ultrasonic velocity at a particular frequency which may result in the increase in intermolecular forces. The variations of adiabatic compressibility Ks, intermolecular free length L_f , molar volume V_m and acoustic impedance Z with the increase in concentration of DMAC are presented in tables-3 and 4 and shown graphically in figures-3 to 6. The decrease in adiabatic compressibility and intermolecular free length while opposite trend in acoustic impedance and molar volume with the increase in concentration of DMAC predict the existence of specific interactions among the components in the ternary liquid mixture [23].

Table-1
Values of density and ultrasonic velocity of pure liquids at 308K

Organic Liquids	ρ Kgm^{-3}	U ms^{-1}			
		2MHz	4MHz	6MHz	8MHz
Dimethyl acetamide	925	1488	1472	1464	1440
Diethyl ether	693	928	912	904	880
Acetone	764	1140	1128	1116	1104

Table-2
Values of density and ultrasonic velocity for ternary mixture at 308K

Mole Fraction		ρ Kgm^{-3}	U ms^{-1}			
X_1	X_3		2MHz	4MHz	6MHz	8MHz
0	0.6	736	1020	1012	1000	986
0.1	0.5	752	1086	1074	1062	1046
0.2	0.4	768	1102	1094	1080	1060
0.3	0.3	784	1152	1140	1134	1120
0.4	0.2	800	1192	1178	1164	1144
0.5	0.1	816	1206	1198	1188	1168
0.6	0	832	1230	1224	1212	1200

Table-3
Values of adiabatic compressibility and free length for ternary mixture at 308K

Mole Fraction		K_s×10^{-10} m^{2}N^{-1}				L_f×10^{-10} m			
X_1	X_3	2MHz	4MHz	6MHz	8MHz	2MHz	4MHz	6MHz	8MHz
0	0.6	13.0594	13.2666	13.587	13.9755	0.7234	0.7291	0.7379	0.7484
0.1	0.5	11.2752	11.5285	11.7905	12.154	0.6722	0.6797	0.6874	0.6979
0.2	0.4	10.722	10.8794	11.1633	11.5885	0.6555	0.6603	0.6688	0.6815
0.3	0.3	9.6112	9.8146	9.9187	10.1683	0.6206	0.6271	0.6305	0.6383
0.4	0.2	8.7974	9.0078	9.2258	9.5512	0.5938	0.6008	0.608	0.6187
0.5	0.1	8.4258	8.5387	8.6831	8.983	0.5811	0.585	0.5899	0.6
0.6	0	7.9445	8.0225	8.1822	8.3466	0.5642	0.567	0.5726	0.5783

According to the Eyring Kincaid model for sound propagation [24] ultrasonic velocity increases as intermolecular free length decreases in the liquid mixture and vice versa. Therefore intermolecular free length is one of the important factors for deciding the nature of change in acoustic parameters in the liquid mixture. In the present study the increase in ultrasonic velocity, density and acoustic impedance and opposite trend in adiabatic compressibility are caused due to the the decrease in intermolecular free length as the concentration of DMAC increases in the ternary liquid mixture at a particular frequency. Our experimental results agree well with this model.

The compactness and strength of bonding between the components molecules of the liquid mixture are measured directly by the available volume[25]. Table-4 and figure-7 show that the values of available volume V_a decrease with the increase in concentration of DMAC. The decrease in available volume is caused due to the net packing of molecules inside the shell which may be due to the complexation between unlike molecules though hydrogen bonding in the ternary mixture. Further, the values of V_a increase with the increase in frequency and such increase in V_a indicates the decrease in molecular interaction in the ternary liquid mixture at higher frequency range.

However with the increase in frequency from 2MHz, to 8MHz the ultrasonic velocity decreases at a fixed mole fraction of DMAC. This decrease in ultrasonic velocity may be caused due to the reduction in molecular interaction in the ternary liquid mixture with the increase in frequency. Consequently the values of Ks and Lf increase and the values of Z decrease with the increase in frequency for a particular mole fraction of DMAC.

Table-4
Values of molar volume, acoustic impedance (Z) and available volume (V_a) for ternary mixture at 308K

Mole Fraction		V_m $m^3\,mol^{-1}$	$Z\times10^5\,Kgm^2s^{-1}$				$V_a\,m^3mol^{-3}$			
X_1	X_3		2MHz	4MHz	6MHz	8MHz	2MHz	4MHz	6MHz	8MHz
0	0.6	0.0876	7.5072	7.4483	7.36	7.2569	0.0317	0.0321	0.0328	0.0336
0.1	0.5	0.0896	8.1667	8.0764	7.98624	7.8659	0.0287	0.0294	0.0301	0.031
0.2	0.4	0.0915	8.4633	8.4019	8.2944	8.1408	0.0284	0.0289	0.0297	0.0308
0.3	0.3	0.0933	9.0316	8.9376	8.8905	8.7808	0.0261	0.0268	0.0271	0.0279
0.4	0.2	0.0951	9.536	9.424	9.312	9.152	0.0242	0.025	0.0259	0.0271
0.5	0.1	0.0968	9.8409	9.7756	9.694	9.5308	0.0238	0.0243	0.0249	0.0261
0.6	0	0.0984	10.2336	10.1836	10.0838	9.984	0.0227	0.0231	0.0238	0.0246

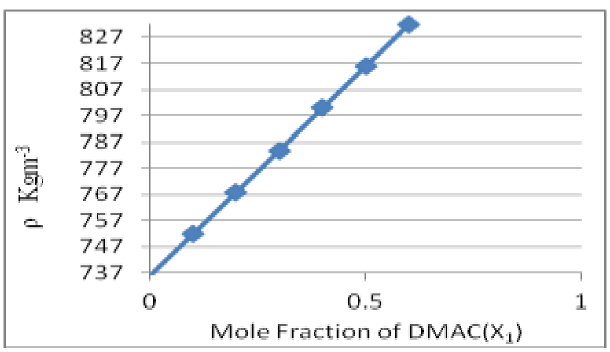

Figure-1
Variation of ρ Versus X_1

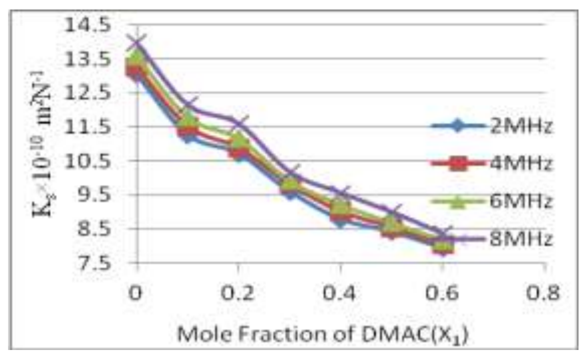

Figure-3
Variation of K_s Versus X_1

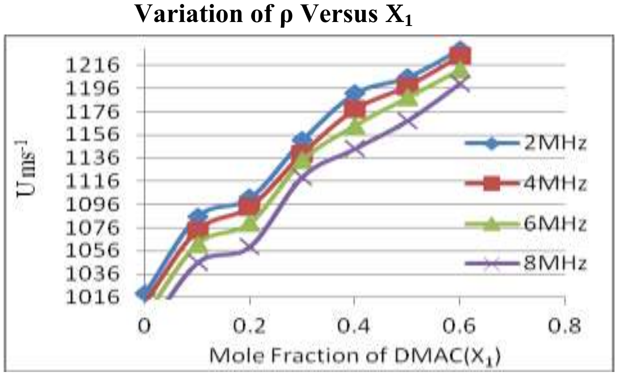

Figure-2
Variation of U Versus X_1

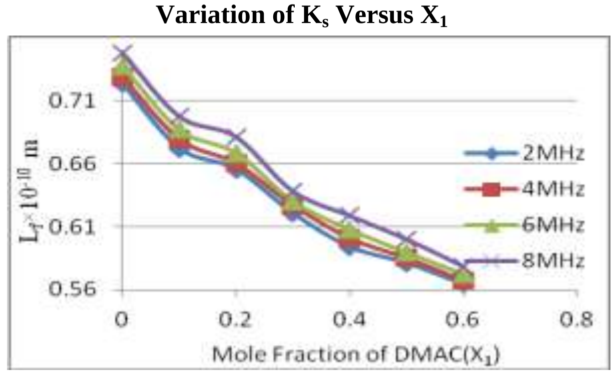

Figure-4
Variation of L_f Versus X_1

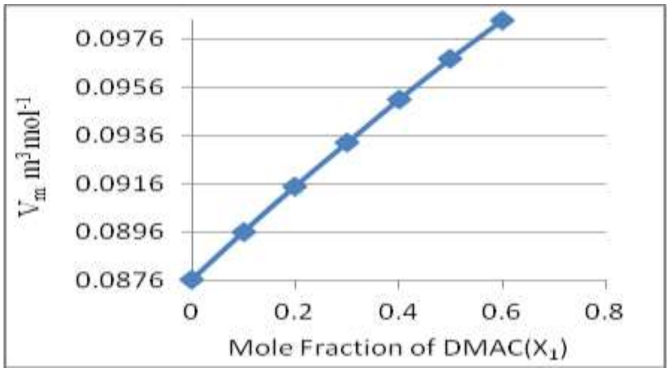

Figure-5
Variation of V_m Versus X_1

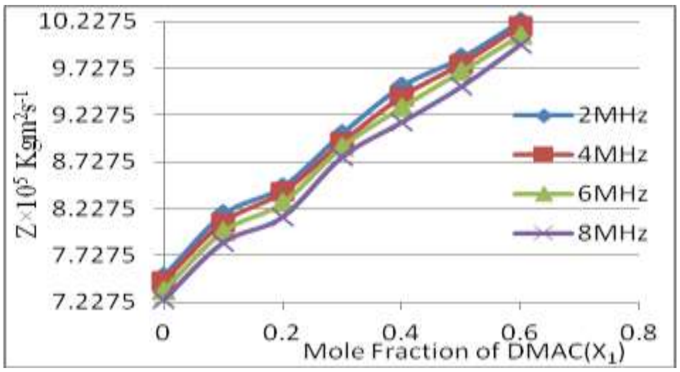

Figure-6
Variation of Z Versus X_1

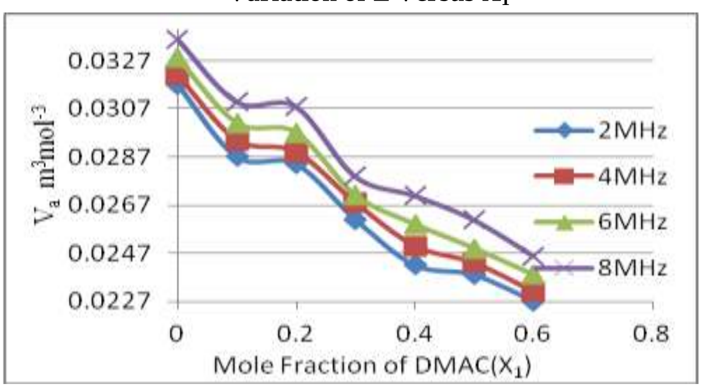

Figure-7
Variation of V_a Versus X_1

In understanding the nature of molecular interactions between the components in the ternary mixture, it is better to explain the same in terms of the excess parameters rather than the actual values. It is seen that the weak interaction between unlike molecules is due to the dispersive forces. This leads to positive excess values of adiabatic compressibility, intermolecular free length, molar volume and available volume and negative excess values of ultrasonic velocity and acoustic impedance. 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 and available volume and positive excess values of ultrasonic velocity and acoustic impedance. 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.

Table-5 and figure-8 show that the values of excess velocity U^E are negative for the entire range of mole fraction of DMAC for all frequencies. The negative values of U^E indicate the presence dispersive forces between unlike molecules in the ternary liquid mixture.

The values of K_s^E are negative as shown in Table-6 and Figure-9 when the mole fraction of DMAC is between 0.1 to 0.6 for frequencies 2MHz, 4MHz, 6MHz and 8MHz but K_s^E is positive in the absence of DMAC. The structure forming tendency is associated with the negative values of K_s^E but the structure breaking tendency due to hetero-molecular interaction between the component molecules of liquid mixture is associated with its positive values[26]. In our present investigation the negative values of K_s^E predict the presence of strong molecular interactions in the ternary liquid mixture.

Table-6 and figure-10 show that the values of L_f^E are negative when the mole fraction of DMAC is between 0.1 to 0.6 for frequencies 2MHz, 4MHz, 6MHz and 8MHz but L_f^E is positive in the absence of DMAC. The dispersive forces lead to the positive values of L_f^E and the strong molecular interactions due to charge transfer and formation of hydrogen bond lead to negative values of L_f^E. In our present investigation the negative excess values of free length L_f^E predict the existence of strong molecular interactions in the ternary liquid mixture.

Table-5
Values of excess velocity the ternary mixture for ternary mixture at 308K

Mole Fraction		U^E ms^{-1}			
X_1	X_3	2MHz	4MHz	6MHz	8MHz
0	0.6	-35.2	-29.6	-31.2	-28.4
0.1	0.5	-4	-2	-4	-2
0.2	0.4	-22.8	-16.4	-20.8	-21.6
0.3	0.3	-7.6	-4.8	-1.6	-1.2
0.4	0.2	-2.4	-1.2	-6.4	-4.8
0.5	0.1	-23.2	-15.6	-17.2	-14.4
0.6	0	-34	-24	-28	-16

Table-6
Excess values of adiabatic compressibility and free length for ternary mixture at 308K

Mole Fraction		$K_s^E \times 10^{-10}\ m^2N^{-1}$				$L_f^E \times 10^{-10}$ m			
X_1	X_3	2MHz	4MHz	6MHz	8MHz	2MHz	4MHz	6MHz	8MHz
0	0.6	0.3141	0.1548	0.2183	0.0785	0.0144	0.0103	0.012	0.0091
0.1	0.5	-0.9512	-1.0535	-1.0316	-1.1904	-0.0174	-0.0195	-0.0185	-0.0214
0.2	0.4	-0.9855	-1.1728	-1.1126	-1.2033	-0.0148	-0.0194	-0.0172	-0.018
0.3	0.3	-1.5774	-1.7079	-1.8103	-2.071	-0.0304	-0.0331	-0.0355	-0.0413
0.4	0.2	-1.8723	-1.9849	-1.9566	-2.1355	-0.0379	-0.0399	-0.0381	-0.041
0.5	0.1	-1.725	-1.9242	-1.9528	-2.1511	-0.0313	-0.0362	-0.0362	-0.0398
0.6	0	-1.6874	-1.9107	-1.9072	-2.235	-0.0289	-0.0347	-0.0336	-0.0416

Table-7
Excess Values of molar volume, acoustic impedance, and available volume, for ternary mixture at 308K

Mole Fraction		V_m^E $m^3\,mol^{-1}$	$Z^E \times 10^5\ Kgm^2s^{-1}$				$V_a^E\ m^3mol^{-3}$			
X_1	X_3		2MHz	4MHz	6MHz	8MHz	2MHz	4MHz	6MHz	8MHz
0	0.6	-0.0007	-0.2909	-0.2504	-0.2616	-0.2431	0.0007	0.0003	0.0004	0.0002
0.1	0.5	-0.0005	-0.1369	-0.1221	-0.1369	-0.1227	-0.0007	-0.0009	-0.0007	-0.0009
0.2	0.4	-0.0004	-0.3457	-0.2965	-0.3303	-0.3363	0.0004	-0.0001	0.0003	0.0002
0.3	0.3	-0.0004	-0.2828	-0.2606	-0.2358	-0.1849	-0.0003	-0.0005	-0.0007	-0.0012
0.4	0.2	-0.0005	-0.2839	-0.274	-0.3159	-0.3022	-0.0006	-0.0008	-0.0004	-0.0006
0.5	0.1	-0.0006	-0.4844	-0.4222	-0.4355	-0.412	0.0004	-0.00005	0.0001	-0.0001
0.6	0	-0.0008	-0.5972	-0.514	-0.5472	-0.4473	0.0008	0.0002	0.0004	-0.0002

Table-8
Values of molar sound velocity, and molar compressibility for ternary mixture at 308K

Mole Fraction		R				B			
X_1	X_3	2MHz	4MHz	6MHz	8MHz	2MHz	4MHz	6MHz	8MHz
0	0.6	0.8818	0.8794	0.876	0.8718	1.6586	1.6551	1.6498	1.6463
0.1	0.5	0.9209	0.9175	0.9141	0.9095	1.7105	1.7052	1.7008	1.6927
0.2	0.4	0.9451	0.9428	0.9387	0.9329	1.7655	1.761	1.7557	1.7521
0.3	0.3	0.978	0.9746	0.9729	0.9689	1.8136	1.8082	1.801	1.7936
0.4	0.2	1.0083	1.0043	1.0003	0.9946	1.8558	1.8521	1.8466	1.8373
0.5	0.1	1.0303	1.028	1.0252	1.0194	1.9091	1.9054	1.8962	1.8906
0.6	0	1.0542	1.0525	1.0491	1.0456	1.9632	1.9605	1.9555	1.9494

The values of V_m^E are negative for the whole range of mole fraction of DMAC as shown in table-7 and figure-11. The values of V_m^E are affected by (i) the difference in shape and size and the loss of dipolar association (ii) dipole-dipole and dipole-induced dipole interactions or formation of complexes due to charge transfer between the unlike molecules. The first effect causes expansion of volume and the second effect causes contraction of volume. In our present investigation the negative values of V_m^E may be interpreted as the contraction of volume of the ternary liquid mixture of DMAC,diethyl ether and acetone. The negative values of V_m^E are favourable for the latter effect which accounts for the dipole-dipole and dipole-induced dipole interactions or formation complexes due to charge transfer between the unlike molecules.

Table-7 and figure-12 show that the values of Z^E are negative for the entire composition range of DMAC for frequencies 2MHz, 4MHz, 6MHz and 8MHz.The negative values of Z^E predict the existence of dispersive interactions between unlike molecules in the ternary liquid mixture.

The strong interactions and dispersive forces are present in the ternary liquid mixture as the values of excess available volume V_a^E are both negative and positive as shown in table-7 and Figure-13 for some mole fractions of DMAC [27].

The values of K_s^E, L_f^E and V_m^E are more negative at about higher concentration region of DMAC which indicate that the dipole-induced dipole interaction is predominant in this region. At lower concentration region of DMAC there exists dipole-dipole interaction among unlike molecules in the ternary mixture.

Tables-5, 6 and 7 show that the values of U^E, K_s^E, L_f^E and Z^E are changed with the increase in frequency due to the decrease in ultrasonic velocity in the ternary liquid mixture.

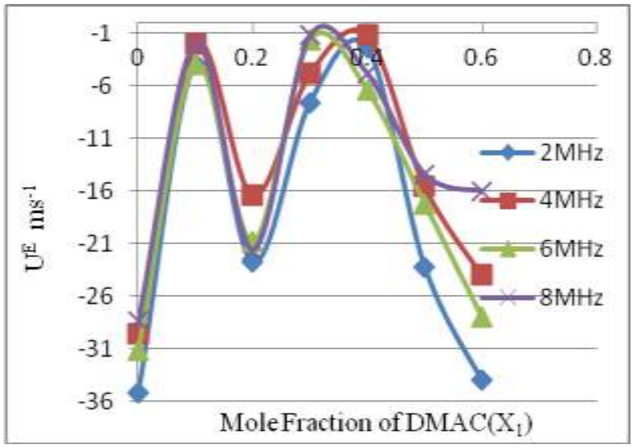

Figure-8
Variation of U^E Versus X_1

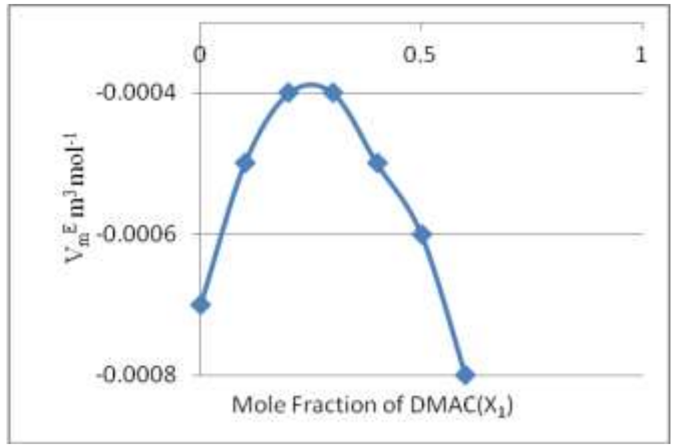

Figure-11
Variation of V_m^E Versus X_1

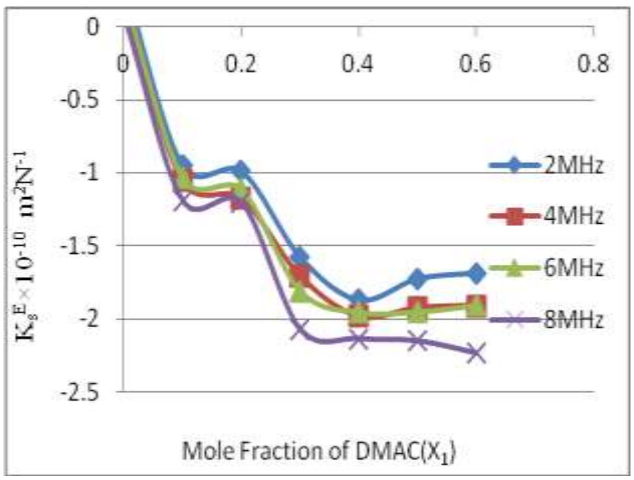

Figure-9
Variation of K_s^E Versus X_1

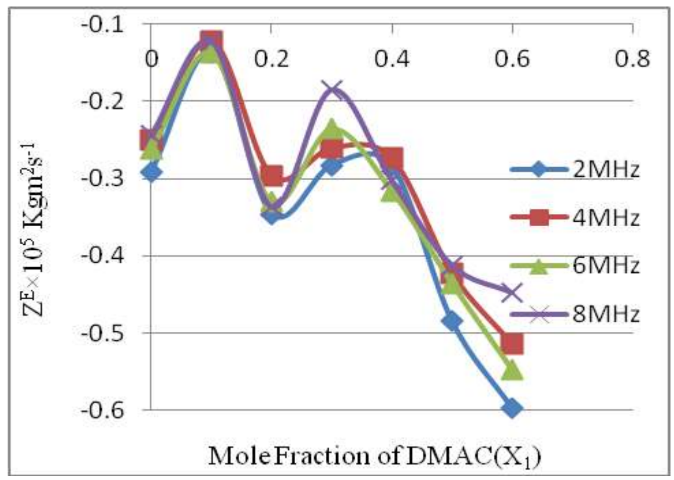

Figure-12
Variation of Z^E Versus X_1

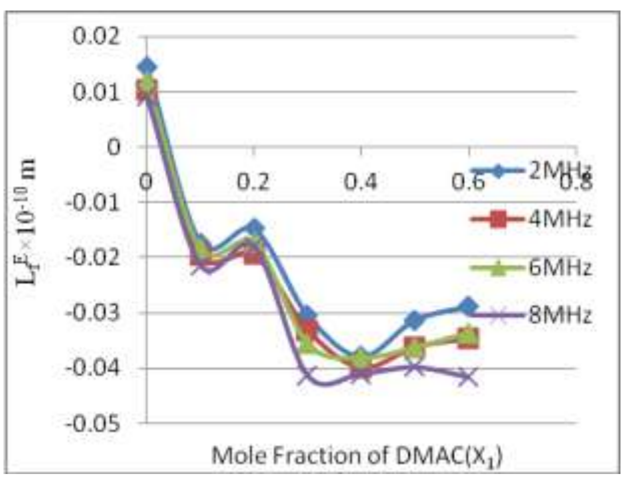

Figure-10
Variation of L_f^E Versus X_1

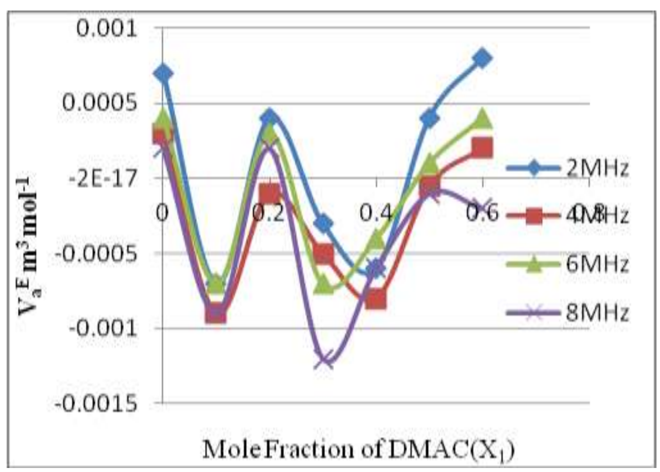

Figure-13
Variation of V_a^E Versus X_1

Table-9
Values of Lennard Jones potential repulsive exponent for ternary mixture at 308K

Mole Fraction		n			
X_1	X_3	2MHz	4MHz	6MHz	8MHz
0	0.6	3.5804	3.3738	3.0243	2.6428
0.1	0.5	5.7317	5.2857	4.8604	4.3419
0.2	0.4	6.3309	5.9965	5.4848	4.8246
0.3	0.3	8.4482	7.888	7.6568	7.0645
0.4	0.2	10.5785	9.824	9.0308	8.0553
0.5	0.1	11.4033	10.9012	10.3253	9.2528
0.6	0	13.0085	12.5584	11.8067	11

Table-10
Values of relative association and interaction parameter for ternary mixture at 308K

Mole Fraction		R_A				χ			
X_1	X_3	2MHz	4MHz	6MHz	8MHz	2MHz	4MHz	6MHz	8MHz
0	0.6	0.9024	0.9015	0.9034	0.9027	-0.0656	-0.056	-0.0596	-0.0552
0.1	0.5	0.904	0.903	0.9047	0.9043	-0.0073	-0.0037	-0.0074	-0.0038
0.2	0.4	0.9176	0.9166	0.9188	0.9195	-0.0401	-0.0293	-0.0374	-0.0395
0.3	0.3	0.923	0.9229	0.9228	0.9216	-0.013	-0.0083	-0.0028	-0.0021
0.4	0.2	0.9312	0.9315	0.9335	0.9338	-0.004	-0.002	-0.0109	-0.0083
0.5	0.1	0.9461	0.9448	0.9457	0.9459	-0.0373	-0.0255	-0.0283	-0.0242
0.6	0	0.9584	0.9565	0.9579	0.9558	-0.053	-0.038	-0.0446	-0.0261

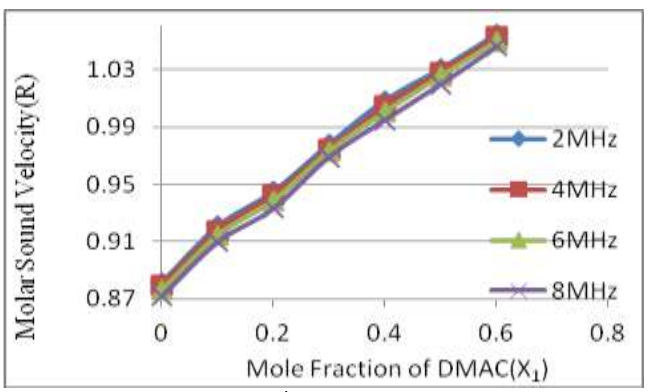

Figure-14
Variation of R Versus X_1

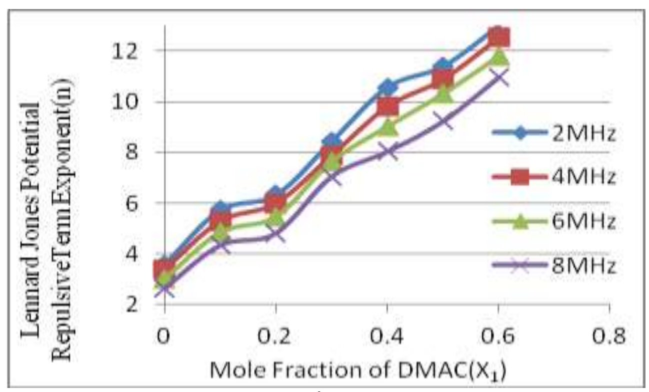

Figure-16
Variation of n Versus X_1

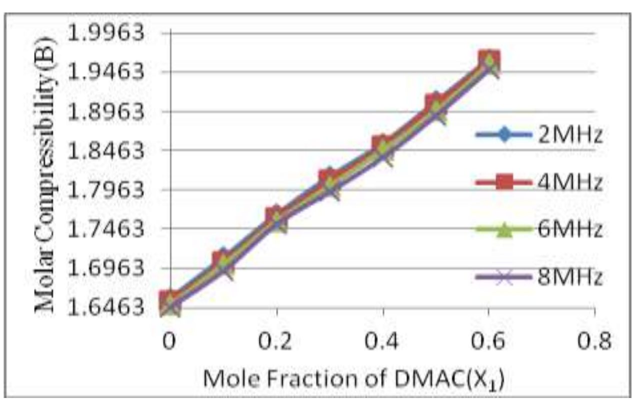

Figure-15
Variation of B Versus X_1

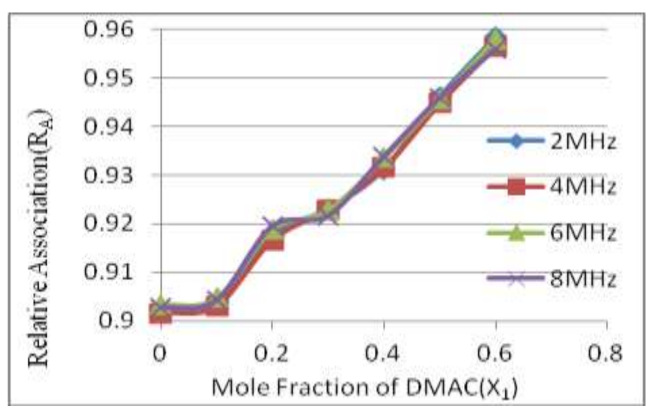

Figure-17
Variation of R_A Versus X_1

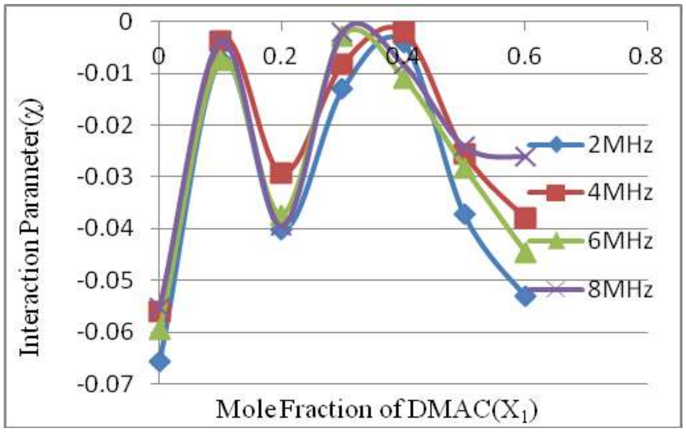

Figure-18
Variation of χ Versus X_1

Table-8, figure-14 and figure-15 show that the values of R and B increase as the concentration of DMAC increases. The increase in R and B indicates the presence of strong interaction in ternary liquid mixture due to molecular association among unlike molecules without complex formation. The values of R and B decrease with the increase in frequency at a particular concentration of DMAC. The decrease in R and B also supports the decrease in molecular interaction with the increase in frequency in the liquid mixture.

Lennard-Jones potential φ(r) is given by the relation [28].

$\varphi(r) = -Ar^{-6} + Dr^{-n}$ (13)

Where r and n are intermolecular distance and Lennard-Jones potential repulsive term exponent respectively. A and D are constants. The first term arises from attractive forces while the second term arises from repulsive forces. It is clear that larger the value of n small is the second term. Thus large value of n indicates the dominance of attractive forces over repulsive forces. Table-9 and figure-16 show that the values of n increase as the concentration of DMAC increases for a fixed frequency. The increase in n indicates the increasing dominance of attractive force over repulsive forces in the ternary liquid mixture. Further, the values of n decrease with the increase in frequency for a particular concentration which indicates the increase in repulsive forces due to reduction in molecular interaction in the ternary mixture. This also explains the cause for the decrease in ultrasonic velocity with the increase in frequency.

Table-10 and figure-17 show that the values of relative association R_A increase with the increase in mole fraction of DMAC for a particular frequency and this increase in relative association hints to the existence of molecular interaction between unlike molecules in the liquid mixture. Such increase in relative association supports the idea that the liquid system is in a more compressed state and the component molecules are much closer to each other at higher concentration of DMAC and there may exist specific interactions between component molecules in the ternary liquid mixture. Relative association remains almost constant for all frequencies at a particular concentration.

Table-10 and figure-18 show that the values of interaction parameter χ are negative for the whole range of mole fraction of DMAC for all frequencies which indicate the existence of dispersive forces with weak dipole-induced dipole interactions in the ternary liquid mixture. Further interaction parameter shows irregular trend with the change in frequency.

Conclusion

From the experimental density, ultrasonic velocity, thermo acoustic parameters and some excess parameters for the ternary mixture it is found that there exists molecular association and intermolecular interaction between components in the ternary liquid mixture of DMAC, diethyl ether and acetone. The negative values of K_s^E, L_f^E and V_m^E indicate strong interactions in the ternary mixture. The negative values of some excess acoustical parameters like velocity and acoustic impedance indicate the existence of dispersive forces in the liiquid mixture. Further, it is concluded that the molecular interaction decreases with the increase in frequency for a fixed concentration of DMAC in the ternary mixture.

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