

Ultrasonic Studies on Molecular Interaction in Ternary Liquid Mixture of Dimethyl Acetamide at Different Frequencies

Ashok Kumar Dash¹ and Rita Paikaray²

¹Department of Physics, L.N College, Patkura, Kendrapara, Odisha, 754134, India

²Department of Physics, Ravenshaw University Cuttack, Odisha, 753003, India

ashok.phy@rediffmail.com

Abstract

The density (ρ) and ultrasonic velocity (U) for ternary mixture of dimethyl acetamide, isobutyl methyl ketone and diethyl ether at different frequencies (2MHz, 4MHz, 6MHz, 8MHz) have been measured at 308K. These data have been 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 excess values of some of the above parameters for the entire range of mole fraction of DAMC and are interpreted to explain molecular interaction occurring in the liquid mixture.

Keywords: Molar volume, Lennard Jones potential repulsive term exponent, relative association, interaction parameter, ternary mixture

1. Introduction

The study of molecular interaction in binary and ternary liquid mixtures plays an important role in the development of molecular sciences. A large number of studies have been made on the intermolecular interaction in liquid system by various methods like Ultraviolet, Dielectric constant, Infrared, Raman Effect, Nuclear magnetic resonance and Ultrasonic method. In recent years Ultrasonic method has become a powerful tool in providing information regarding the physico-chemical properties of liquid system [1-4].

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 study of DMAC is important because of its utilization 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. It is used as a solvent in the production of acrylic and elastane fibres, pharmaceuticals, antibiotics and polyimide resins. Isobutyl methyl ketone is a polar solvent used in rare metal extraction, antibiotics and other pharmaceuticals, cellulose and resin based coatings and adhesives. Diethyl ether is a non-polar liquid used as a solvent in the production of cellulose plastics. In view of the importance mentioned above an attempt has been made to elucidate the molecular interaction in ternary liquid mixture of dimethyl acetamide (DMAC), isobutyl methyl ketone and diethyl ether at different frequencies at constant temperature 308K.

Departure from linearity in the velocity versus concentration in liquid mixture is taken as an indication of the existence of molecular interactions between different species. The

physico-chemical properties of liquid mixture can be studied by the nonlinear variation of ultrasonic velocity and other related parameters with the variation of concentration in the liquid mixture [5-11].

2. Theory

The acoustical parameters such as adiabatic compressibility (K_s), intermolecular free length (L_f), acoustic Impedance (Z), molar volume (V_m) and available volume (V_a) have been calculated using the measured data (U and ρ) 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 $U_\infty = 1600$ m/s.

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

$$A^E = A_{\text{exp}} - (X_1 A_1 + X_2 A_2 + X_3 A_3) \quad (6)$$

Where X_1 , X_2 and X_3 are mole fractions of DMAC, isobutyl methyl ketone and diethyl ether respectively and A is any acoustic parameter.

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.

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

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

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

$$R_A = (\rho / \rho_0) (U_0 / U)^{1/3} \quad (10)$$

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

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

The ideal mixing velocity U_{ideal} is given by

$$U_{\text{ideal}} = X_1 U_1 + X_2 U_2 + X_3 U_3 \quad (12)$$

3. Experimental Methods

3.1. Materials

The chemicals used were of analytical reagent (AR) and spectroscopic reagent (SR) grade which were obtained from E Merck Ltd (India) and were used as such without further purification. The ternary liquid mixtures of various concentrations were prepared out of which the mole fraction of isobutyl methyl ketone was kept fixed at 0.4. The mole fractions of

DMAC and diethyl ether were varied from 0 to 0.6 to have the mixture of different compositions. Liquid mixtures of different mole fractions were prepared with a precision of 0.0001g using an electronic digital balance.

3.2. Density Measurement

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

$$\rho_2 = (W_2/W_1) \rho_1 \quad (13)$$

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.

3.3. Velocity Measurement

The velocity of ultrasonic waves in the liquid mixture was measured by using 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 ultrasonic velocity was determined using the relation

$$U = \lambda f \quad (14)$$

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

4. Results and discussion

The experimental values of density and ultrasonic velocity at 308K for frequencies 2MHz, 4MHz, 6MHz and 8MHz for pure liquids and ternary liquid mixture were used to calculate the acoustical parameters and the relevant data are presented in Tables 1 to 10 and displayed graphically in Figures 1 to 18. Experimentally measured density (ρ) and ultrasonic velocity (U) of pure liquids at 308K and at different frequencies are shown in Table 1.

Table 2 and Figure 1 show that density ρ increases with the increase in mole fraction of DMAC. The increase in density indicates the presence of solvent-solvent interactions in the ternary mixture which may bring a bonding between them [11]. Table 2 and Figure 2 show that ultrasonic velocity U in the ternary mixture increases with the increase in mole fraction of DMAC. 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 K_s , intermolecular free length L_f , molar volume V_m and acoustic impedance Z with the increase in mole fraction of DMAC are presented in Tables 3 and 4 and shown graphically in Figures 3 to 6. The decrease in adiabatic compressibility, intermolecular free length and

molar volume and increase in acoustic impedance with the increase in concentration of DMAC reveal the presence of specific interactions between the components in the ternary liquid mixture [12].

Table 1. Values of density (ρ) and ultrasonic velocity (U) of pure liquids

Organic Liquids	ρ Kgm ⁻³	U ms ⁻¹			
		2MHz	4MHz	6MHz	8MHz
Dimethyl acetamide	925	1488	1472	1464	1440
Isobutyl methyl ketone	765	1160	1152	1140	1120
Diethyl ether	693	928	912	904	880

Table 2. Values of density(ρ) and ultrasonic velocity(U) for DMAC + isobutyl methyl ketone + diethyl ether

Mole Fraction		ρ Kgm ⁻³	U ms ⁻¹			
X ₁	X ₃		2MHz	4MHz	6MHz	8MHz
0	0.6	722	1034	1016	1008	1000
0.1	0.5	745	1078	1066	1056	1036
0.2	0.4	769	1138	1128	1116	1108
0.3	0.3	792	1172	1156	1140	1128
0.4	0.2	815	1214	1200	1194	1180
0.5	0.1	838	1264	1252	1248	1244
0.6	0	860	1328	1320	1308	1296

The experimental results are in good agreement with the Eyring Kincaid model for sound propagation [13]. According to which the ultrasonic velocity increases with the decrease in intermolecular free length in the liquid mixture and vice versa. Therefore, intermolecular free length is one of the predominating factors for deciding the nature of variation in ultrasonic parameters in the liquid mixture. In the present study the decrease in intermolecular free length causes increase in ultrasonic velocity, decrease in adiabatic compressibility, increase in density and acoustic impedance as the concentration of DMAC increases.

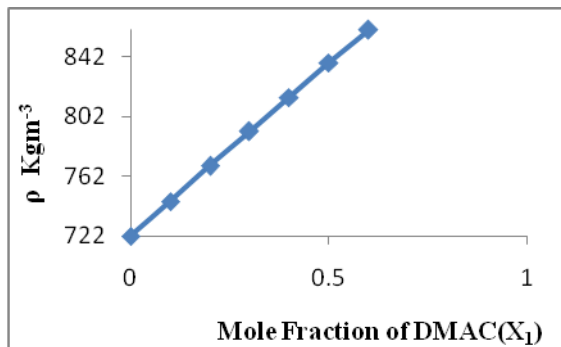


Figure 1. Variation of ρ Versus X₁

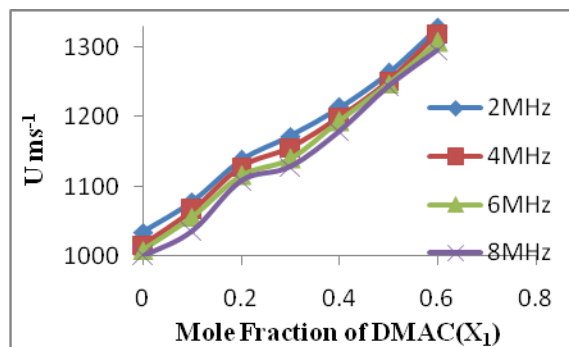


Figure 2. Variation of U Versus X_1

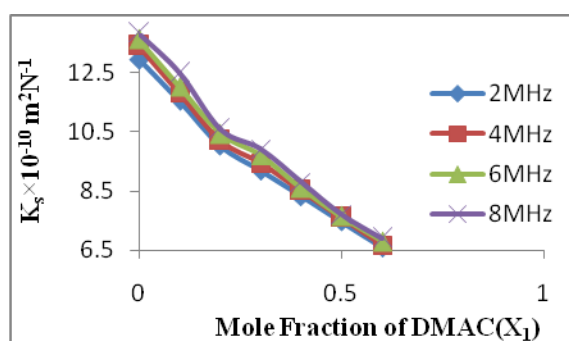


Figure 3. Variation of K_s Versus X_1

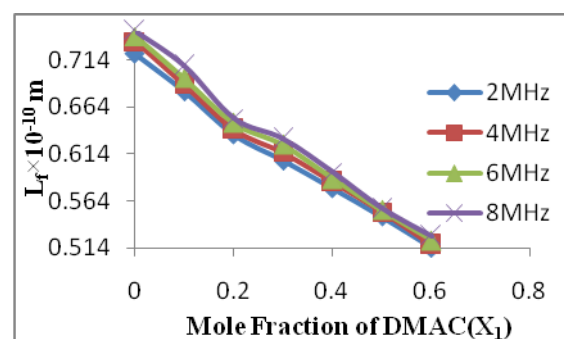


Figure 4. Variation of L_f Versus X_1

Table 4 and Figure 7 show that available volume V_a decreases with the increase in mole fraction of DMAC. Available volume is a direct measure of compactness and strength of bonding between the molecules of the liquid mixture [14]. The decrease in V_a is due to the net packing of molecules inside the shell which may be formed by complexation between unlike molecules through hydrogen bonding in the ternary mixture.

Table 3. Values of adiabatic compressibility (K_s) and free length (L_f) for DMAC + isobutyl methyl ketone + diethyl ether

Mole Fraction		$K_s \times 10^{-10} \text{ m}^2 \text{N}^{-1}$				$L_f \times 10^{-10} \text{ m}$			
X_1	X_3	2MHz	4MHz	6MHz	8MHz	2MHz	4MHz	6MHz	8MHz
0	0.6	12.9545	13.4176	13.6314	13.8504	0.7205	0.7333	0.7391	0.745
0.1	0.5	11.5506	11.8122	12.0369	12.5062	0.6804	0.688	0.6945	0.7079
0.2	0.4	10.0413	10.2201	10.4411	10.5924	0.6343	0.64	0.6469	0.6515
0.3	0.3	9.1922	9.4484	9.7155	9.9233	0.6069	0.6153	0.624	0.6306
0.4	0.2	8.3254	8.5207	8.6066	8.812	0.5776	0.5843	0.5873	0.5942
0.5	0.1	7.4689	7.6125	7.6617	7.711	0.5471	0.5523	0.5541	0.5559
0.6	0	6.5933	6.6735	6.7965	6.9229	0.514	0.5171	0.5219	0.5267

Table 4. Values of molar volume (V_m), acoustic impedance (Z) and available volume (V_a) for DMAC + isobutyl methyl ketone+ diethyl ether

Mole Fraction		V_m $\text{m}^3 \text{mol}^{-1}$	$Z \times 10^5 \text{ Kg m}^2 \text{ s}^{-1}$				$V_a \text{ m}^3 \text{mol}^{-1}$			
X_1	X_3		2MHz	4MHz	6MHz	8MHz	2MHz	4MHz	6MHz	8MHz
0	0.6	0.117	7.4654	7.3355	7.2777	7.22	0.0413	0.0427	0.0432	0.0438
0.1	0.5	0.1152	8.0311	7.9417	7.8672	7.7182	0.0375	0.0384	0.0391	0.0406
0.2	0.4	0.1133	8.7512	8.6743	8.582	8.5205	0.0327	0.0334	0.0342	0.0348
0.3	0.3	0.1116	9.2822	9.1555	9.0288	8.9337	0.0298	0.0309	0.032	0.0329
0.4	0.2	0.1101	9.8941	9.78	9.7311	9.617	0.0265	0.0275	0.0279	0.0289
0.5	0.1	0.1086	10.5923	10.4917	10.4582	10.4247	0.0228	0.0236	0.0238	0.0241
0.6	0	0.1073	11.4208	11.352	11.2488	11.1456	0.0182	0.0187	0.0195	0.0203

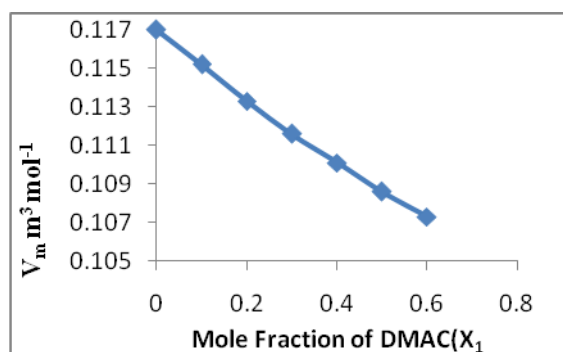


Figure 5. Variation of V_m Versus X_1

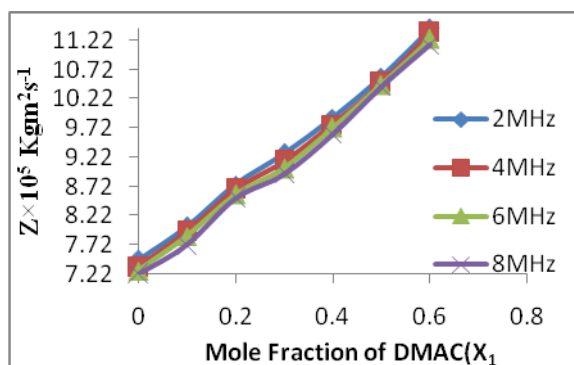


Figure 6. Variation of Z Versus X_1

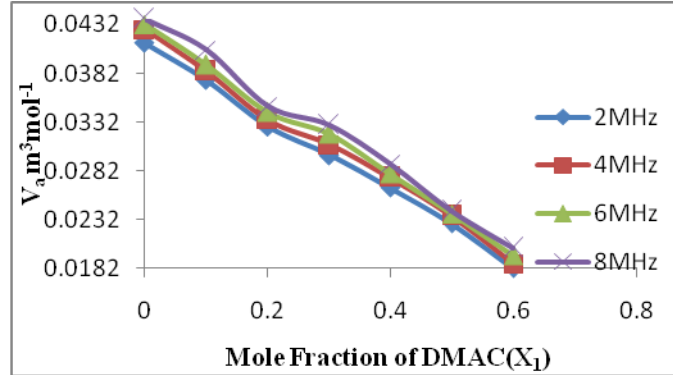


Figure 7. Variation of V_a Versus X_1

However, with the increase in frequency from 2MHz to 8MHz ultrasonic velocity decreases at a fixed mole fraction of DMAC. This decrease in ultrasonic velocity may be due to the decrease in molecular interaction in the ternary mixture with the increase in frequency. Consequently, adiabatic compressibility, intermolecular free length and available volume increase and acoustic impedance decreases with the increase in frequency for a particular mole fraction of DMAC.

In order to know the nature of interactions between the component molecules of the ternary liquid mixture, it is of interest to discuss the same in terms of the excess values of acoustical parameters rather than the actual values. It is learnt that dispersive forces are responsible for weak interaction between unlike molecules. This leads to positive excess values of adiabatic compressibility, intermolecular free length, molar volume and available volume and negative excess values of velocity and acoustic impedance. The attractive forces are responsible for strong interaction between unlike molecules which leads to negative excess values of adiabatic compressibility, intermolecular free length, molar volume and available volume and positive excess values of velocity and acoustic impedance. Non-ideal liquid mixtures show considerable deviation from linearity from their physical properties with respect to mole fraction and these have been interpreted as the presence of both strong and weak interactions.

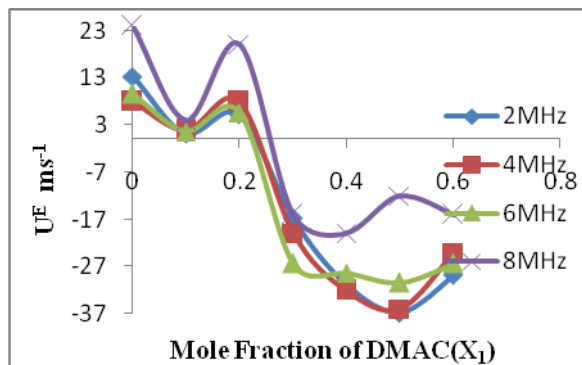


Figure 8. Variation of U^E Versus X_1

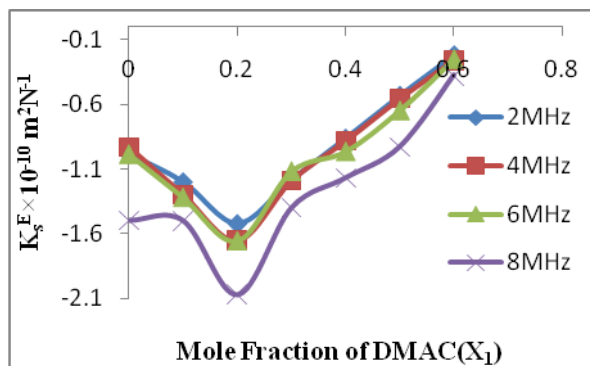


Figure 9. Variation of K_s^E Versus X_1

Table 5 and Figure 8 show that the values of excess velocity U^E are positive for the mole fraction of DMAC between 0 to 0.2 and negative for the mole fraction of DMAC between 0.3 to 0.6 for all frequencies. The positive values of U^E indicate the presence strong molecular interactions at lower concentration of DMAC. The negative values of U^E indicate the presence dispersive forces between unlike molecules in the ternary mixture.

The values of K_s^E are negative as shown in Table-6 and Figure 9 for the entire range of mole fraction of DMAC for frequencies 2MHz, 4MHz, 6MHz and 8MHz. The negative value of K_s^E is associated with a structure forming tendency but the positive value is associated with a structure breaking tendency due to hetero-molecular interaction between the component molecules of liquid mixture [15]. In the present investigation the negative values of K_s^E predict the existence of strong molecular interactions in the ternary liquid mixture.

Table 6 and Figure 10 show that the values of L_f^E are negative for the entire range of mole fraction of DMAC for all frequencies. The positive excess values of free length are attributed to the dispersive forces and the negative excess values of L_f^E are due to due to charge transfer, formation of hydrogen bond, dipole-induced dipole and dipole-dipole interactions. In the present study the negative excess values of L_f^E predict the existence of strong molecular interactions in the ternary mixture due to charge transfer, formation of hydrogen bond, dipole-induced dipole and dipole-dipole interactions.

Table 5. Values of excess velocity (U^E) the ternary mixture for DMAC + isobutyl methyl ketone + diethyl ether

Mole Fraction		$U^E \text{ ms}^{-1}$			
X_1	X_3	2MHz	4MHz	6MHz	8MHz
0	0.6	13.2	8	9.6	24
0.1	0.5	1.2	2	1.6	4
0.2	0.4	5.2	8	5.6	20
0.3	0.3	-16.8	-20	-26.4	-16
0.4	0.2	-30.8	-32	-28.4	-20
0.5	0.1	-36.8	-36	-30.4	-12
0.6	0	-28.8	-24	-26.4	-16

The values of excess molar volume V_m^E are negative for the of mole fraction of DMAC between 0.1 to 0.6 for all frequencies and positive in the absence of DMAC as shown in Table 7 and Figure 11. Excess molar volume is influenced by (i) the loss of dipolar association and the difference in size and shape (ii) dipole-dipole and dipole-induced dipole interactions or charge transfer complexation between the unlike molecules. The former effect

leads to expansion of volume and the latter effect leads to contraction of volume. In the present investigation the negative values of V_m^E may be interpreted as the contraction of volume of the mixture. 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 charge transfer complexation between the unlike molecules.

Table 6. Excess values of adiabatic compressibility (K_s^E) and free length (L_f^E) for DMAC + isobutyl methyl ketone+ diethyl ether

Mole Fraction		$K_s^E \times 10^{-10} \text{ m}^2 \text{ N}^{-1}$				$L_f^E \times 10^{-10} \text{ m}$			
X_1	X_3	2MHz	4MHz	6MHz	8MHz	2MHz	4MHz	6MHz	8MHz
0	0.6	-0.9849	-0.9318	-0.9864	-1.4982	-0.0207	-0.0183	-0.0195	-0.032
0.1	0.5	-1.2014	-1.3012	-1.3195	-1.5003	-0.0231	-0.0249	-0.025	-0.0283
0.2	0.4	-1.5234	-1.6573	-1.654	-2.0721	-0.0315	-0.0342	-0.0334	-0.044
0.3	0.3	-1.1851	-1.193	-1.1182	-1.3992	-0.0212	-0.0202	-0.0172	-0.0242
0.4	0.2	-0.8646	-0.8848	-0.9652	-1.1684	-0.0127	-0.0126	-0.0147	-0.0199
0.5	0.1	-0.5338	-0.557	-0.6493	-0.9274	-0.0055	-0.0059	-0.0087	-0.0175
0.6	0	-0.222	-0.26	-0.2532	-0.3735	-0.0009	-0.0024	-0.0018	-0.006

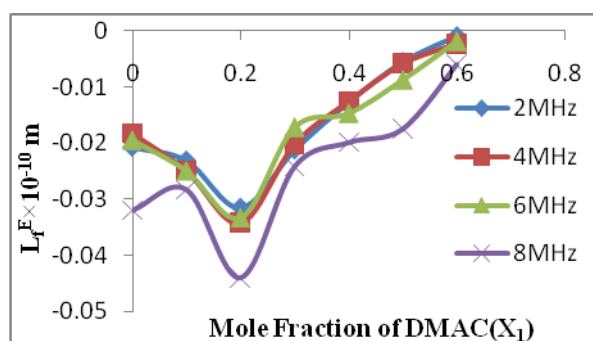


Figure 10. Variation of L_f^E Versus X_1

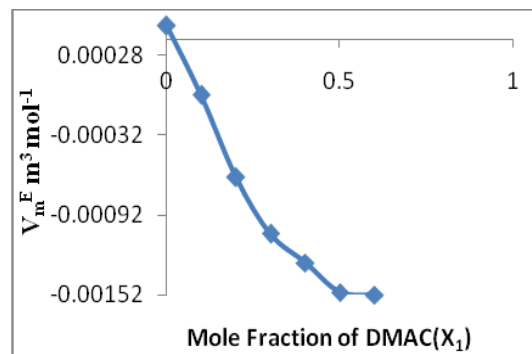


Figure 11. Variation of V_m^E Versus X_1

Table 7. Excess Values of molar volume V_m^E , acoustic impedance (Z^E) and available volume (V_a^E) for DMAC + isobutyl methyl ketone + diethyl ether

Mole Fraction		V_m^E $m^3 mol^{-1}$	$Z^E \times 10^5 \text{ Kgm}^2 s^{-1}$				$V_a^E \text{ m}^3 mol^{-1}$			
X_1	X_3		2MHz	4MHz	6MHz	8MHz	2MHz	4MHz	6MHz	8MHz
0	0.6	0.0005	0.0572	0.0183	0.0304	0.1337	0.00006	0.00052	0.00026	-0.00074
0.1	0.5	-0.00002	-0.1174	-0.105	-0.1077	-0.0902	0.00009	0.00005	0.00002	-0.00007
0.2	0.4	-0.00064	-0.1236	-0.102	-0.1206	-0.01	-0.00088	-0.0011	-0.00102	-0.002
0.3	0.3	-0.00106	-0.3259	-0.3504	-0.4016	-0.319	0.00005	0.00024	0.00064	-0.00003
0.4	0.2	-0.00128	-0.4473	-0.4555	-0.427	-0.5504	0.00058	0.00068	0.0004	-0.00016
0.5	0.1	-0.0015	-0.4824	-0.4734	-0.4276	-0.2723	0.00071	0.00062	0.00016	-0.00109
0.6	0	-0.00152	-0.3872	-0.3427	-0.3648	-0.2736	-0.00006	-0.0004	-0.0002	-0.001

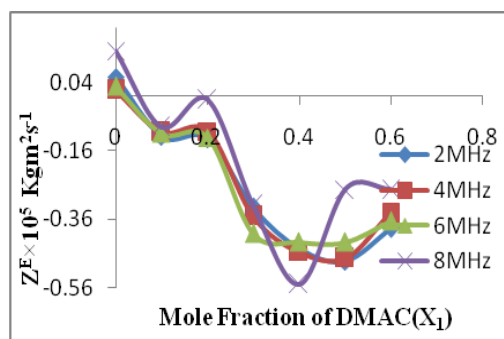


Figure 12. Variation of Z^E Versus X_1

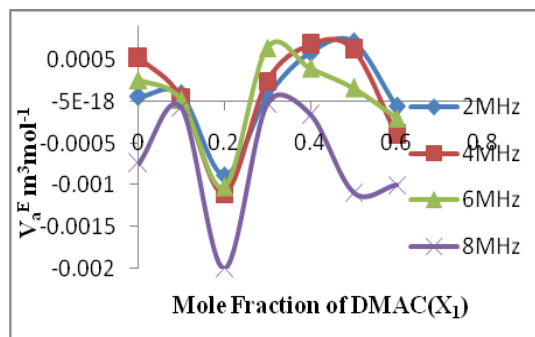


Figure 13. Variation of V_a^E Versus X_1

The values of excess acoustic impedance Z^E are negative for the mole fraction of DMAC between 0.1 to 0.6 as shown in Table 7 and Figure 12 for all frequencies and positive in the absence of DMAC in the liquid mixture. The negative values of Z^E indicate the presence of dispersive forces between unlike molecules in the ternary mixture.

Table 7 and Figure 13 show that the values of excess available volume V_a^E are both negative and positive which indicate the presence of strong interactions and dispersive forces in the liquid mixture [16]. The excess acoustic parameters are also changed with the increase in frequency in the ternary mixture.

Table 8, Figures 14 and 15 show that molar sound velocity R and molar compressibility B change non-linearly with the increase in mole fraction of DMAC. The non-linear change in molar sound velocity and molar compressibility indicates the presence of molecular

interactions in the ternary liquid mixture with complex formation. The values of R and B decrease with the increase in frequency at a particular concentration of DMAC in the liquid mixture. This decrease in R and B also supports the decrease in molecular interaction with the increase in frequency in the ternary mixture.

Table 8. Values of molar sound velocity (R) and molar compressibility (B) for DMAC + isobutyl methyl ketone + diethyl ether

Mole Fraction		R				B			
X ₁	X ₃	2MHz	4MHz	6MHz	8MHz	2MHz	4MHz	6MHz	8MHz
0	0.6	1.1831	1.1762	1.1731	1.17	2.1768	2.1659	2.1611	2.1561
0.1	0.5	1.1812	1.1768	1.1731	1.1656	2.1788	2.1718	2.166	2.1542
0.2	0.4	1.1828	1.1794	1.1752	1.1724	2.1861	2.1806	2.174	2.1695
0.3	0.3	1.1766	1.1712	1.1658	1.1617	2.1807	2.1721	2.1635	2.157
0.4	0.2	1.1745	1.1699	1.168	1.1634	2.182	2.1748	2.1717	2.1644
0.5	0.1	1.1742	1.1704	1.1692	1.1679	2.1859	2.18	2.178	2.176
0.6	0	1.1794	1.177	1.1734	1.1698	2.1986	2.1948	2.189	2.1833

Table 9. Values of Lennard Jones potential repulsive term exponent (n) for DMAC + isobutyl methyl ketone + diethyl ether

Mole Fraction		n			
X ₁	X ₃	2MHz	4MHz	6MHz	8MHz
0	0.6	3.9975	3.4402	3.25	3.0273
0.1	0.5	5.432	5	4.6777	4.0246
0.2	0.4	7.7889	7.3532	6.8771	6.5344
0.3	0.3	9.4697	8.6699	7.925	7.3525
0.4	0.2	11.9283	11.0218	10.6774	9.8581
0.5	0.1	15.5789	14.6101	14.3781	14.0373
0.6	0	22.3736	21.4278	20.0153	18.7142

Lennard-Jones potential $\phi(r)$ is given by the relation [17].

$$\phi(r) = -Ar^{-6} + Dr^{-n} \quad (15)$$

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. Larger the value of n smaller is the second term. Thus large value of n indicates the dominance of attractive forces over repulsive forces. The values of n increase with the increase in mole fraction of DMAC as shown in Table 9 and Figure 16 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 [18].

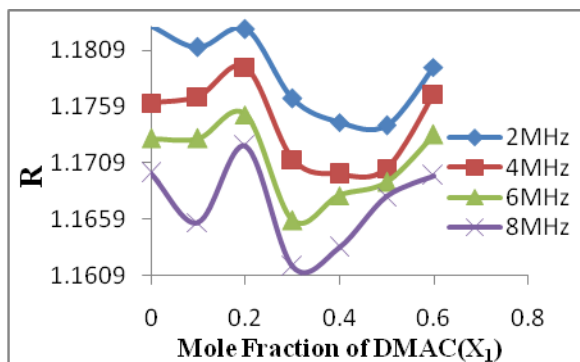


Figure 14. Variation of R Versus X_1

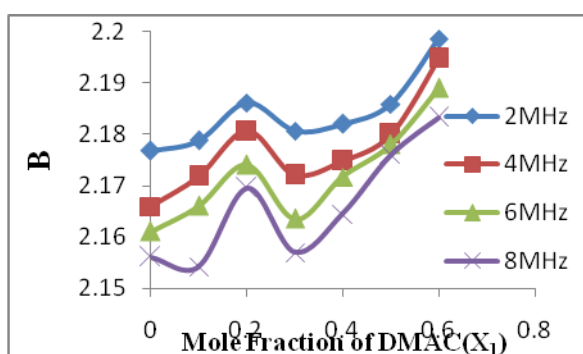


Figure 15. Variation of B Versus X_1

Table 10. Values of relative association (R_A) and interaction parameter (χ) for DMAC + isobutyl methyl ketone + diethyl ether

Mole Fraction		R_A				χ			
X_1	X_3	2MHz	4MHz	6MHz	8MHz	2MHz	4MHz	6MHz	8MHz
0	0.6	0.8812	0.8832	0.8839	0.8814	0.026	0.0159	0.0193	0.0497
0.1	0.5	0.8967	0.8968	0.898	0.8988	0.0022	0.0037	0.003	0.0077
0.2	0.4	0.909	0.9084	0.91	0.9072	0.0092	0.0143	0.0101	0.0371
0.3	0.3	0.9271	0.928	0.9306	0.9288	-0.028	-0.0337	-0.0447	-0.0277
0.4	0.2	0.9429	0.9431	0.943	0.9415	-0.0488	-0.0512	-0.0459	-0.033
0.5	0.1	0.9565	0.9561	0.9554	0.9512	-0.0557	-0.0551	-0.0469	-0.019
0.6	0	0.9656	0.9641	0.9653	0.9629	-0.042	-0.0354	-0.0391	-0.0242

The values of relative association R_A increase with the increase in mole fraction of DMAC for a particular frequency as shown in Table 10 and Figure 17 and this increase in relative association indicates the close association of component molecules and there may exist specific or chemical interactions like charge transfer, hydrogen bond formation, and other complex forming interactions between component molecules in the ternary liquid mixture [19]. Relative association remains almost constant for all frequencies at a particular concentration.

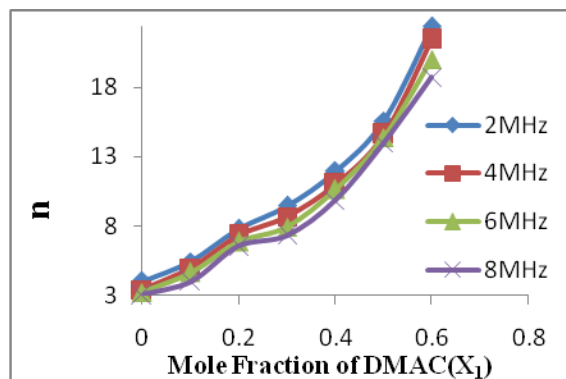


Figure 16. Variation of n Versus X_1

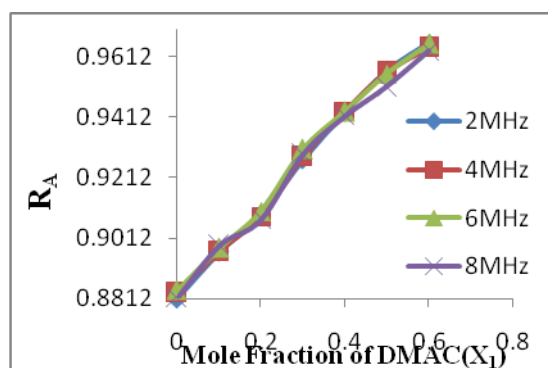


Figure 17. Variation of R_A Versus X_1

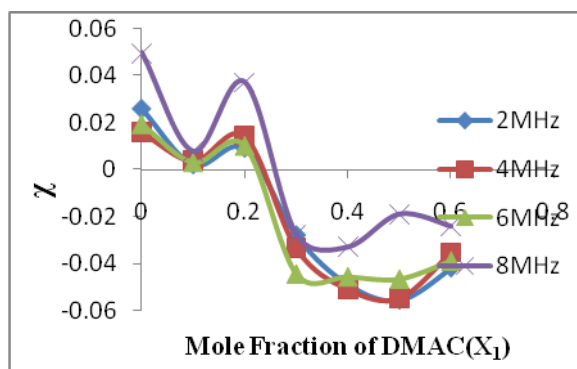


Figure 18. Variation of χ Versus X_1

Table 10 and Figure 18 show that the values of interaction parameter χ are positive for the mole fraction DMAC between 0 to 0.2 for all frequencies. The positive values of χ indicate the existence of strong interactions in the ternary liquid mixture. The values of interaction parameter χ are negative for the mole fraction DMAC between 0.3 to 0.6 for all frequencies indicating the existence of weak interactions in the ternary liquid mixture [20]. Further interaction parameter shows irregular trend with the change in frequency.

5. Conclusion

On the basis of the experimental values of density, ultrasonic velocity, related acoustical parameters and some of their excess values for the ternary liquid mixture, it is concluded that there exists molecular association and molecular interaction between components in the ternary liquid mixture of DMAC, isobutyl methyl ketone and diethyl ether. The negative excess values of adiabatic compressibility and intermolecular free length indicate a strong molecular interaction in the ternary mixture due to charge transfer, formation of hydrogen bond, dipole-induced dipole and dipole-dipole interactions. The negative values of excess acoustic impedance indicate the presence of dispersive forces between component molecules in the liquid 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.

References

- [1] A. K. Dash and R. Paikaray, "Acoustical study in binary liquid mixture containing dimethyl acetamide using ultrasonic and viscosity probes", *Der Chem. Sinica*, vol. 5, no. 1, (2014), pp. 81-88.
- [2] R. Palani, S. Saravanan and R. Kumar, "Ultrasonic studies on some ternary organic liquid mixtures at 303, 308 and 313K", *RASAYAN J. Chem.*, vol. 2, no. 3, (2009), pp. 622-629.
- [3] R. Paikaray and N. Mohanty, "Evaluation of Thermodynamical Acoustic Parameters of Binary mixture of DBP with Toluene at 308K and at Different Frequencies", *Research Journal of Chemical Sciences*, vol. 3, no. 5, (2013), pp. 71-82.
- [4] A. A. Mistry, V. D. Bhandakkar and O. P. Chimankar, "Acoustical studies on ternary mixture of toluene in cyclohexane&nitrobenzene at 308k using ultrasonic technique", *J. of Chem.and Pharm.Res.*, vol. 4, no. 1, (2012), pp. 170-174.
- [5] R. Uvarani and J. Sivapragasam, "Intermolecular interaction studies on ternary liquid mixtures at 303K", *J. of Chem. and Pharm. Res.*, vol. 4, no. 1, (2012), pp. 653-655.
- [6] C. Shanmuga Priya, S. Nithya, G. Velraj and A. N. Kanappan, "Molecular interactions studies in liquid mixture using ultrasonic technique", *Int. J. of Adv. Sc.and Tech.*, vol. 18, (2010), pp. 59-73.
- [7] M. K. Praharaj, P. R. Mishra, S. Mishra and A. Satapathy, "Study of thermodynamic and transport properties of ternary liquid mixture at different temperatures", *Adv. in Appl. Sc. Res.*, vol. 3, no. 3, (2012), pp. 1518-1530.
- [8] S. K. Pradhan, S. K. Dash, L. Moharana and B. B. Swain, "Molecular interaction parameters of binary mixtures of diethyl ether and apolar solvents using ultrasonic probe" *Indian J Pure and Appl. Phys.*, vol. 50, (2012), pp.161-166.
- [9] R. Kubendram, F. L. Ali Khan, J. Asghar, M. Aravinthraj and J. Udayaseelan, "Ultrasonic studies of N, N-dimethylacetamide and N-methylacetamide with alkoxyethanols in carbon tetrachloride at different temperatures", *Arch. in Appl. Sc. Res.*, vol. 3, no. 2, (2011) pp. 568-576.
- [10] V. Kanappan and J. R. Santhi, "Ultrasonic study of induced dipole-dipole interactions in binary liquid mixtures", *Indian J Pure and Appl. Phys.*, vol. 43, (2005) pp. 750-754.
- [11] R. Natrajan and P. Ramesh, "Ultrasonic velocity determination in binary liquid mixtures", *J. Pure appl. And Ind. Phys.*, vol. 1, no. 4, (2011), pp. 252-258.
- [12] D. Bhatnagar, D. Joshi, R.Gupta, Y. Kumar, A. Kumar and C. L. Jain, "Studies on Thermo acoustic Parameter in Binary Liquid Mixtures of MIBK with 1-Propanol and 1-pentanol at 303.15K-A new approach by Direct Measurement of Acoustic Impedance", *Res. J. of Chem. Sc.*, vo. 1, no. 5, (2011), pp. 6-13.
- [13] H. Eyring and J. F. Kincaid, "Free Volumes and Free Angle Ratios of Molecules in Liquids", *J. Chem. Physics Chem. Physics*, vol. 6, (1938), pp. 620-629.
- [14] M. K. Praharaj, P. R. Mishra, S. Mishra and A. Satapathy, "Ultrasonic study of ternary liquid mixture containing substituted benzene", *Arch.of. Phys. Res.*, vol. 3. No.3, (2012), pp. 192-200.
- [15] S. Thirumarani and M. Rajeswari, "Acoustical studies on binary liquid mixtures of some aromatic hydrocarbons with dimethylsulphoxide (DMSO) at 303.15K", *Arch.of. Phys. Res.*, vol. 2, no. 2, (2011), pp. 149-156.
- [16] K. Rajagopal and S. Chentilnath, "Excess thermodynamic studies of binary mixtures of 2-methyl 2-propanol with ketones", *Indian J. Pure and Appl. Phys.*, vol. 48, (2010), pp. 326-333.
- [17] A. K. Gupta, K. Kumar and B. K. Karn, "Studies of binary liquid mixtures of o-cresol with ethylmethyl ketone, acetone acetophenone and ethylacetate", *J. Ind. Coun. Chem.*, vol. 26, (2009) pp. 77-81.

- [18] A. K. Dash and R. Paikaray, "Ultrasonic Study on Ternary Mixture of Dimethyl Acetamide (DMAC) in Diethyl ether and Acetone", Res. J. of Phys. Sc., vol. 1, no. 3, (2013), pp. 12-20.
- [19] A. K. Dash and R. Paikaray, "Study of molecular interaction in binary liquid mixture of dimethyl acetamide and acetone using ultrasonic probe", Adv. in Appl. Sc. Res., vol. 4, no. 3, (2013), pp. 130-139.
- [20] A. K. Dash and R. Paikaray, "Acoustical study on ternary mixture of dimethyl acetamide (DMAC) in diethyl ether and isobutyl methyl ketone at different frequencies", Phys. and Chem. of Liquids, vol. 51, no. 6, (2013), pp. 749-763.

Authors

Ashok Kumar Dash is currently doing Ph.D at Ravenshaw University, Cuttack, India in the field of Ultrasonics under the guidance of Dr.(Mrs)Rita Paikaray. He has completed M.Sc from National Institute of Technology, Rourkela, India. He is currently working as a Lecturer in Physics in L.N College, Patkura, Odisha, India

Dr(Mrs).Rita Paikaray is working as an Associate Professor in Physics in Ravenshaw University, Cuttack, India. She has completed M.Sc. from Utkal University, Bhubaneswar, India, M.Phil and Ph.D from University of Delhi, India. She is currently working in the field of Ultrasonics and Plasma Physics. She is an executive member of Ultrasonic Society of India, Life member of Acoustical Society of India and Power beam Society of India.

