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Der Chemica Sinica, 2014, 5(1): 81-88



# Acoustical study in binary liquid mixture containing dimethyl acetamide using ultrasonic and viscosity probes

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# ABSTRACT

Ultrasonic velocity (U), density ( $\rho$ ) and coefficient of viscosity ( $\eta$ ) of binary mixture of dimethyl acetamide (DAMC) and acetone at different frequencies (2MH<sub>Z</sub>, 4MH<sub>Z</sub> 6MH<sub>Z</sub>, 8MH<sub>Z</sub>) have been measured at temperature 308K. Adiabatic compressibility ( $K_s$ ), intermolecular free length ( $L_f$ ), free volume ( $V_f$ ), internal pressure ( $\pi_i$ ) and their respective excess values have been computed for entire range of mole fraction and are interpreted to explain molecular interaction occurring in the liquid mixture. Relaxation time ( $\tau$ ), excess enthalpy ( $H^E$ ) and absorption coefficient ( $\alpha/f^2$ ) have been calculated and discussed.

Key words: Free volume, internal pressure, relaxation time, excess enthalpy and absorption coefficient

# INTRODUCTION

The acoustical study of liquids plays an important role in understanding the nature and strength of molecular interactions. Ultrasonic velocities have been adequately employed to understand the nature of molecular interaction in pure liquids, binary and ternary mixture[1-6]. The study of molecular interaction in binary liquid mixture with dimethyl acetamide (DMAC) and acetone as the components is of particular interest, since DMAC is a dipolar aprotic solvent with high boiling point and good thermal and chemical stability. It is used in industry and medicine. It is also used as solvent for the production of acrylic fibres, elasthane fibres, polymide 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 non-polar solvents and readily suitable to explore solute solvent interactions. Acetone is also an important polar solvent used in industry and pharmaceuticals. In view of these considerations, an attempt has been made to explain the molecular interaction in binary liquid mixture of dimethyl acetamide (DMAC) and acetone at different frequencies at constant temperature 308K.Departure from linearity in the acoustical parameter versus concentration in liquid mixture of DMAC is taken as an indication of the existence of interaction between different liquid molecules[7-18]. The physiochemical properties of liquid mixture can be studied by the non-linear variation of ultrasonic velocity and other acoustical parameters with structural changes occurring in a liquid and the liquid mixture.

# MATERIALS AND METHODS

The liquid mixtures of various concentrations in mole fraction were prepared by taking chemicals of analytical grade (E-Merck) which were used as such without further purification. Liquid mixtures of different mole fractions were prepared on concentration scale with a precision 0.0001g using an electronic digital balance.

# **Experimental Part**

Density of pure liquids and liquid mixture was determined by a specific gravity bottle of 10ml capacity. Coefficient of viscosity of pure liquids and liquid mixture was determined by an Ostwald's viscometer. The ultrasonic velocity was measured by a multi-frequency interferometer (Model M-82S) with a high degree of accuracy operating at different frequencies supplied by Mittal Enterprises, New Delhi,India. An electronically operated constant temperature water bath is used to circulate water through the double walled measuring cell made up of steel containing the experimental liquid mixture at the desired temperature of 308K.

# **Computational Part**

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 ( $\pi_i$ ) have been calculated from the following relations.

$$K_{s} = (U^{2}\rho)^{-1}$$
(1)

$$L_{\rm f} = k \left( K_{\rm S} \right)^{1/2}$$
 (2)

$$V_{\rm f} = (MU/K\eta)^{3/2} \tag{3}$$

$$\pi_{\rm i} = b R T(K \eta/U)^{1/2} (\rho^{2/3}/M^{7/6}) \tag{4}$$

Where k is a temperature dependent constant, M is the effective molecular weight, K is a temperature independent constant which is equal to  $4.28 \times 10^9$  for all liquids. R is universal gas constant and b is the cubic packing factor which is equal to 2 for all liquid mixtures.

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

$$A^{E} = A_{exp} - (X_{1}A_{1} + X_{2}A_{2})$$
(5)

Where X<sub>1</sub>, and X<sub>1</sub> are mole fractions of DMAC and acetone respectively and A is any acoustical parameter.

Relaxation time( $\tau$ ),excess enthalpy(H<sup>E</sup>) and absorption coefficient( $\alpha/f^2$ ), have been calculated from the following relations.

$$\tau = (4/3) \operatorname{K}_{\mathrm{s}} \eta \tag{6}$$

$$\mathbf{H}^{\mathrm{E}} = (\mathbf{X}_{1} \, \pi_{i1} \, \mathbf{V} \mathbf{m}_{1} + \mathbf{X}_{2} \, \pi_{i2} \mathbf{V}_{\mathrm{m}2}) - \pi_{i} \mathbf{V}_{\mathrm{m}} \tag{7}$$

$$\alpha/f^2 = 2\pi^2 \tau/U \tag{8}$$

The experimental values of density  $\rho$ , coefficient of viscosity  $\eta$  and ultrasonic velocity U are represented in Table-1 and the values of adiabatic compressibility Ks, intermolecular free length  $L_f$ , free volume  $V_f$  and internal pressure  $\pi_i$  are depicted in Tables-2 and 3.

Table-1-Values of density, coefficient of viscosity and ultrasonic velocity for the binary mixture at 308K

Male Freedom of $DMAC(\mathbf{X})$	- V3	-3 NJ2		U ms <sup>-1</sup>				
Mole Fraction of $DMAC(X_1)$	ρKgm	$\eta \times 10^{-1}$ NSM	2MHz	4MHz	6MHz	8MHz		
0	764	0.36	1140	1128	1116	1104		
0.081	776	0.388	1162	1150	1137	1126		
0.164	789	0.412	1184	1172	1158	1148		
0.299	811	0.465	1240	1228	1212	1200		
0.395	826	0.546	1258	1248	1236	1224		
0.494	842	0.618	1293	1284	1272	1256		
0.598	859	0.677	1333	1320	1308	1296		
0.705	876	0.766	1358	1344	1335	1324		
0.819	895	0.835	1406	1392	1368	1360		
1	925	0.946	1488	1472	1464	1440		

The values of excess coefficient of viscosity  $\eta^E$ , excess velocity  $U^E$  excess adiabatic compressibility  $K_s^{\ E}$ , excess intermolecular free length  $L_f^{\ E}$ , excess free volume  $V_f^{\ E}$  and excess internal pressure  $\pi_i^{\ E}$  are shown in Tables-4 to 6.

Table-2-Values of adiabatic compressibility and intermolecular free length for the binary mixture at 308K

Mole Fraction of DMAC(X <sub>1</sub> )	K <sub>s</sub> ×10 <sup>-10</sup> m <sup>2</sup> N <sup>-1</sup>				L <sub>f</sub> ×10 <sup>-10</sup> m				
	2MHz	4MHz	6MHz	8MHz	2MHz	4MHz	6MHz	8MHz	
0	10.0715	10.2869	10.5094	10.7391	0.6353	0.6421	0.649	0.656	
0.081	9.5439	9.7441	9.9682	10.1639	0.6184	0.6249	0.632	0.6382	
0.164	9.041	9.2271	9.4516	9.6169	0.6019	0.6081	0.6154	0.6208	
0.299	8.0192	8.1767	8.394	8.5628	0.5669	0.5724	0.58	0.5858	
0.395	7.6499	7.773	7.9247	8.0805	0.5537	0.5581	0.5635	0.569	
0.494	7.1038	7.2037	7.3402	7.5285	0.5335	0.5373	0.5423	0.5493	
0.598	6.5515	6.6812	6.8044	6.931	0.5124	0.5174	0.5222	0.527	
0.705	6.19	6.3197	6.4052	6.512	0.498	0.5032	0.5066	0.5108	
0.819	5.652	5.7663	5.9704	6.0408	0.4759	0.4807	0.4891	0.492	
1	4.8826	4.9893	5.044	5.2135	0.4423	0.4471	0.4496	0.4571	

#### Table-3-Values of free volume and internal pressure for the binary mixture at 308K

Mole Fraction of DMAC(X <sub>1</sub> )	V <sub>f</sub> m <sup>3</sup> mol <sup>-1</sup>				$\pi_i \times 10^4 \text{ Nm}^{-2}$				
	2MHz	4MHz	6MHz	8MHz	2MHz	4MHz	6MHz	8MHz	
0	0.0089	0.00876	0.00862	0.00843	13.671	13.84	13.914	13.989	
0.081	0.00868	0.00854	0.0084	0.00828	13.672	13.743	13.821	13.889	
0.164	0.00865	0.00852	0.00837	0.00826	13.484	13.551	13.633	13.692	
0.299	0.00847	0.00835	0.00818	0.00806	13.283	13.347	13.435	13.502	
0.395	0.00723	0.00714	0.00704	0.00694	13.791	13.846	13.913	13.981	
0.494	0.00665	0.00658	0.00649	0.00637	13.981	14.03	14.096	14.185	
0.598	0.00645	0.00636	0.00627	0.00619	13.925	13.993	14.057	14.122	
0.705	0.00586	0.00577	0.00574	0.00564	14.183	14.257	14.305	14.364	
0.819	0.00577	0.00568	0.00554	0.00549	14.069	14.139	14.263	14.305	
1	0.0057	0.0056	0.0055	0.0054	13.826	13.901	13.939	14.055	

Table-4-Excess values of coefficient of viscosity and ultrasonic velocity for the binary mixture at 308K

Mole Fraction of $\mathbf{DMAC}(\mathbf{Y}_{i})$	E -3 NJ				
Mole Fraction of DMAC( $X_1$ )	$\eta \times 10$ INSIII	2MHz	4MHz	6MHz	8MHz
0	0	0	0	0	0
0.081	-0.0191	-5.048	-4.736	-6.072	-4.112
0.164	-0.0437	-11.932	-11.932	-13.956	-10
0.299	-0.0698	-2.912	-1.728	-9.036	-3.36
0.395	-0.0451	-18.326	-14.752	-16.344	-11.616
0.494	-0.0311	-16.772	-12.808	-14.796	-12.88
0.598	-0.033	-13.964	-12.584	-14.988	-7.824
0.705	-0.0067	-26.2	-25.398	-25.224	-15.776
0.819	-0.0045	-17.872	-16.606	-31.896	-18.08
1	0	0	0	0	0

The values of relaxation time  $\tau$ , excess enthalpy H<sup>E</sup> and absorption coefficient  $\alpha/f^2$  are depicted in Tables-7 and 8.

Table-5-Excess values of adiabatic compressibility and intermolecular free length for the binary mixture at 308K

Mole Fraction of DMAC(X <sub>1</sub> )	$K_{s}^{E} \times 10^{-10} \text{ m}^{2} \text{N}^{-1}$				L <sub>f</sub> <sup>E</sup> ×10 <sup>-10</sup> m				
	2MHz	4MHz	6MHz	8MHz	2MHz	4MHz	6MHz	8MHz	
0	0	0	0	0	0	0	0	0	
0.081	-0.0972	-0.1033	-0.0879	-0.1167	-0.0006	-0.0006	-0.0001	-0.001	
0.164	-0.1694	-0.1803	-0.1509	-0.2052	-0.001	-0.0012	-0.0002	-0.0018	
0.299	-0.4906	-0.5159	-0.4706	-0.5133	-0.01	-0.0105	-0.0087	-0.01	
0.395	-0.3619	-0.4109	-0.4152	-0.4652	-0.0047	-0.0061	-0.0059	-0.0077	
0.494	-0.3943	-0.4558	-0.4587	-0.4701	-0.0057	-0.0076	-0.0075	-0.0077	
0.598	-0.4068	-0.4274	-0.4261	-0.4929	-0.0067	-0.0071	-0.0068	-0.0093	
0.705	-0.2132	-0.222	-0.2405	-0.3207	-0.0005	-0.0004	-0.0011	-0.0042	
0.819	-0.1596	-0.1715	-0.0522	-0.162	-0.0006	-0.0007	-0.0033	-0.0003	
1	0	0	0	0	0	0	0	0	

Mole Fraction of DMAC(X <sub>1</sub> )		$\pi_i^E \times 10^4 \text{ Nm}^{-2}$						
	2MHz	4MHz	6MHz	8MHz	2MHz	4MHz	6MHz	8MHz
0	0	0	0	0	0	0	0	0
0.081	0.00005	0.00005	0.00005	0.0001	0.002	-0.088	-0.081	-0.091
0.164	0.00028	0.00028	0.00027	0.00033	-0.198	-0.285	-0.271	-0.293
0.299	0.00053	0.00054	0.0005	0.00054	-0.42	-0.497	-0.472	-0.492
0.395	-0.0004	-0.00036	-0.00034	-0.00028	0.072	-0.004	0.003	-0.02
0.494	-0.00066	-0.00061	-0.00058	-0.00055	0.247	0.173	0.183	0.177
0.598	-0.00053	-0.0005	-0.00048	-0.00042	0.174	0.13	0.141	0.197
0.705	-0.00078	-0.00075	-0.00067	-0.00065	0.416	0.387	0.387	0.342
0.819	-0.0005	-0.00048	-0.00052	-0.00045	0.284	0.262	0.342	0.275
1	0	0	0	0	0	0	0	0

### Table-6-Excess values of free volume and internal pressure for the binary mixture at 308K

#### Table-7-Values of relaxation time for the binary mixture at 308K

Male Exaction of $\mathbf{DMAC}(\mathbf{V})$	$\tau \times 10^{-12} s$							
Mole Fraction of $DMAC(\mathbf{x}_1)$	2MHz	4MHz	6MHz	8MHz				
0	0.483	0.493	0.504	0.515				
0.081	0.493	0.503	0.515	0.525				
0.164	0.496	0.506	0.519	0.528				
0.299	0.497	0.506	0.52	0.53				
0.395	0.556	0.565	0.576	0.588				
0.494	0.585	0.593	0.604	0.62				
0.598	0.591	0.602	0.614	0.625				
0.705	0.632	0.645	0.654	0.664				
0.819	0.629	0.641	0.664	0.672				
1	0.615	0.629	0.636	0.657				

Table-8-Values of excess enthalpy and absorption coefficient for the binary mixture at 308K

Mole Exection of $\mathbf{DMAC}(\mathbf{X})$		H <sup>E</sup> J 1	mol <sup>-1</sup>		$(\alpha/f^2) \times 10^{-15}$				
Mole Fraction of DWAC(A)	2MHz	4MHz	6MHz	8MHz	2MHz	4MHz	6MHz	8MHz	
0	0	0	0	0	8.375	8.644	8.927	9.22	
0.081	-0.004	0.002	0.001	0.002	8.391	8.656	8.956	9.221	
0.164	0.008	0.015	0.014	0.016	8.284	8.54	8.854	9.087	
0.299	0.0244	0.03	0.028	0.029	7.918	8.153	8.49	8.736	
0.395	-0.018	-0.012	-0.013	-0.011	8.742	8.954	9.217	9.491	
0.494	-0.0335	-0.027	-0.028	-0.028	8.941	9.129	9.389	9.753	
0.598	-0.026	-0.023	-0.024	-0.021	8.761	9.023	9.273	9.533	
0.705	-0.048	-0.049	-0.046	-0.042	9.193	9.483	9.677	9.92	
0.819	-0.033	-0.031	-0.039	-0.032	8.837	9.108	9.588	9.761	
1	0	0	0	0	8.174	8.442	8.582	9.018	

### **RESULTS AND DISCUSSION**

The values of density  $\rho$ , coefficient of viscosity  $\eta$  and ultrasonic velocity U increase with the increase in mole fraction of DMAC as shown in Figure-1, Figure -2 and Figure -3. The increase in density with the increase in mole fraction of DMAC indicates the presence of dipole-dipole interactions which may lead to the formation of hydrogen bond between DMAC and acetone molecules. The increase in ultrasonic velocity at a particular frequency may be due to the structural changes occurring in the liquid mixture resulting in the increase in intermolecular forces. The increase in coefficient of viscosity with the increase in mole fraction of DMAC indicates the presence of solute-solvent interactions.





Figure-1: Variation of  $\rho$  with  $X_1$ 

Figure-2: Variation of  $\eta$  with  $X_1$ 

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The variations of adiabatic compressibility Ks, intermolecular free length  $L_f$ , free volume  $V_f$  and internal pressure  $\pi_i$ with the increase in mole fraction of DMAC are shown graphically in figures-4 to 7. The decrease in adiabatic compressibility, intermolecular free length, and free volume and the non-linear variation of internal pressure with the increase in concentration of DMAC reveal the presence of specific interactions between the components in the binary liquid mixture.

Figure-6 shows that the values of free volume V<sub>f</sub> decrease with the increase in mole fraction of DMAC for a particular frequency. The decrease in free volume with the increase in concentration 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.

Figure-7 shows that internal pressure  $\pi_i$  changes non-linearly with the increase in mole fraction of DMAC for a particular frequency. The non-linear variation of internal pressure with the increase in concentration of DMAC indicates the presence of molecular interactions in the binary liquid mixture.





Figure-5: Variation of L<sub>f</sub> with X<sub>1</sub>

Figure-3: Variation of U with X<sub>1</sub>

Figure-6: Variation of Vf with X1

Figure-4: Variation of K<sub>s</sub> with X<sub>1</sub>



Figure-7: Variation of  $\pi_i$  with  $X_1$ 

1.5

However the ultrasonic velocity decreases with the increase in frequency from 2MHz, to 8MHz 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. Consequently the values of adiabatic compressibility, intermolecular free length, internal pressure increase and free volume decreases with the increase in frequency for a particular mole fraction of DMAC

The variations of excess coefficient of viscosity  $\eta^E$ , excess velocity  $U^E$  excess adiabatic compressibility  $K_s^E$ , excess intermolecular free length  $L_f^E$ , excess free volume  $V_f^E$  and excess internal pressure  $\pi_i^E$  with the increase in mole fraction of DMAC are shown graphically in Figures-8 to 13. Figure-8 shows that the excess values of coefficient of viscosity  $\eta^E$  are negative for the entire range of mole fraction of DMAC for all frequencies. The negative values of  $\eta^E$  indicate the presence dispersion, induction and dipolar forces in the binary liquid mixture [19].



Figure-12:Variation of  $V_f^{E}$  with  $X_1$ 

Figure-13:Variation of  $\pi_i^E$  with  $X_1$ 

Figure-9 shows that the values of excess velocity  $U^E$  are negative for the entire range of concentration of DMAC for all frequencies. The negative values of  $U^E$  indicate the presence dispersive forces between unlike molecules in the binary liquid mixture.

The values of  $K_s^E$  are negative as shown in Figure-10, for the whole range of mole fraction of DMAC for all frequencies which indicate the existence of strong molecular interactions in the binary liquid mixture due to the formation of hydrogen bonds. It also indicates tightly packed molecules in the liquid mixture.

Figure-11 shows that the values of  $L_f^E$  are negative for the whole range of composition of DMAC for all frequencies which indicate the existence of strong molecular interactions in the binary liquid mixture due to charge transfer, dipole-induced dipole, dipole-dipole interactions and formation of hydrogen bonds.

It is seen from Figure-12, that the values of excess free volume  $V_f^E$  are positive between 0 to 0.299 mole fraction of DMAC and negative between 0.395 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  $V_f^E$  at lower concentration of DMAC are favorable for the latter effect which account 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 account for the former effect which account for the strong molecular interactions in the binary liquid mixture of DMAC and acetone [20].





Figure-16:Variation of  $\alpha/f^2$  with  $X_1$ 

The values of excess internal pressure  $\pi_i^E$  are negative within 0 to 0.299 mole fraction of DMAC and positive within 0.395 to 1 mole fraction of DMAC as shown in Figure-13. The negative values of  $\pi_i^E$  at lower concentration of DMAC indicate the presence of dispersion and dipolar forces but the positive values of  $\pi_i^E$  at higher concentration of DMAC indicate the presence of specific interactions in the binary liquid mixture [21].

It is observed that the excess values of velocity, 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.

The variations of relaxation time  $\tau$ , excess enthalpy  $H^E$  and absorption coefficient  $\alpha/f^2$  with the increase in mole fraction of DMAC are shown graphically in Figures-14 to 16. Figure-14 shows that the relaxation time  $\tau$  varies non-

linearly with the increase in mole fraction of DMAC for a fixed frequency. The relaxation time  $\tau$  increases with the increase in frequency for a fixed mole fraction DMAC. The relaxation time is of the order of  $10^{-12}$ s may be due to the structural relaxation process showing the presence of molecular interactions and in such a case it is suggested that the molecules get rearranged due to co-operative process.

The values of excess enthalpy  $H^E$  are positive or slightly negative for 0 to 0.299 mole fraction of DMAC and negative for 0.395 to 1 mole fraction of DMAC as shown in Figure-15 for frequencies 2MHz, 4MHz ,6MHz and 8MHz. The positive excess values of  $H^E$  at lower concentration of DMAC indicate the presence of dispersive forces and the negative excess values of  $H^E$  at higher concentration of DMAC indicate the presence of specific interactions in the binary liquid mixture [22].

Figure-16 shows that the values of absorption coefficient  $\alpha/f^2$  vary non-linearly with the increase in mole fraction of DMAC for a fixed frequency which indicate the presence of 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 mixture with increasing frequency [23].

### CONCLUSION

On the basis of experimental values of density, ultrasonic velocity, related acoustical parameters and some of their excess values for the binary liquid mixture it is found that there exists hydrogen bonding, charge transfer, dipole-dipole and dipole-induced dipole interactions in the binary liquid mixture of DMAC and acetone. Dispersion forces are also found to exist between component molecules in the liquid mixture. Further, it is concluded that the molecular interaction increases with the increase in concentration 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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