



Research Article

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## Study of thermodynamic and transport properties of ternary liquid mixture at different frequencies

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### ABSTRACT

The Ultrasonic velocity, density and viscosity have been measured for ternary mixture of N-N dimethylformamide (DMF), Cyclohexane and Chloro-benzene at different frequencies (2 MHz, 4 MHz, 6 MHz and 8 MHz) for a constant temperature (318 K). These experimental data have been used to estimate the thermodynamic parameters such as adiabatic compressibility ( $\beta$ ), free length ( $L_f$ ), internal pressure ( $\Pi_i$ ), relaxation time ( $\tau$ ), acoustic impedance ( $Z$ ), Gibb's free energy ( $\Delta G$ ) and absorption coefficient for the mixture. The excess values of the parameters are also evaluated and discussed.

**Key Words:** Ultrasonic velocity, intermolecular free length, internal pressure, free volume, Gibb's free energy.

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### INTRODUCTION

The practical importance of liquid mixture rather than single component liquid systems has gained much importance during the last two decades in assessing the nature of molecular interactions and investigating the physio-chemical behavior of such systems [1,2]. Ultrasonic investigation of liquid mixtures consisting of polar and non-polar components is of considerable importance in understanding the physical nature and strength of molecular interaction in the liquid mixtures. [3].

For a better understanding of the physio-chemical properties and the molecular interaction between the participating components of the mixtures, ultrasonic velocity together with density and viscosity are measured at different frequencies for different concentrations of the components in the mixture. These data's furnish wealth of information about the interaction between ions, dipoles; hydrogen bonding, multi-polar and dispersive forces [4-5]. In order to understand the nature of molecular interactions between the components of liquid mixtures, it is of interest to discuss the same in terms of excess parameters rather than the actual values.

The present paper deals with the study of different parameters of the mixture containing Dimethylformamide (DMF), Cyclohexane and chloro-benzene at different frequencies through ultrasonic measurements.

DMF ( $C_3H_7NO$ ) is a versatile compound. It is a non-aqueous solvent which has no hydrogen bonding in pure state. Therefore it acts as an aprotic, protophilic medium with high dielectric constant and it is also considered as a dissociating solvent. DMF being a polar molecule results in dipolar and induced dipolar interaction between it and chlorobenzene in addition to dipolar-dipolar interaction between its molecules. DMF is primarily used as an industrial solvent. DMF solutions are used to process polymer fibers, films and surface coating to permit easy spinning of acrylic fibers to produce wire enamels and as a crystallization medium in the pharmaceutical industry.

Cyclohexane ( $C_6H_{12}$ ) is a non-polar, unassociated, inert hydrocarbon possessing globular structure. It is produced in large quantity by hydrogenation of benzene. The principal use of cyclohexane is conversion by oxidation in air to a mixture of cyclohexanol and the ketone, which is then oxidized further to adipic acid for the manufacture of nylon.

Cyclohexane belongs to alicyclic hydrocarbon (closed chain). The packing of carbon atoms in the even numbered alkane groups allows the maximum intermolecular attractions [6]. It is highly inert towards an electrophile or a nucleophile at ordinary temperature. Due to the non-polar nature of cyclohexane and its inertness towards electron donors [7], dispersive types of interaction are expected between it and other components.

Chlorobenzene (C<sub>6</sub>H<sub>5</sub>Cl) is a poor electron donor towards the electron seeking proton of any group. It has low dielectric constant ( $\epsilon = 5.649$ ) and dipole moment. ( $\mu = 1.69$  D). It is neither acidic nor basic and is more reactive because the chlorine atom is bonded with SP<sup>3</sup> hybridised carbon atom and consequently can be removed easily.. Hence the rate of reaction with chlorobenzene is faster. The rate of molecular interactions results in a greater degree of variation with respect to ultrasonic related parameters. The chlorine atom being an electron withdrawing atom attracts the  $\pi$ -electron of benzene ring in C<sub>6</sub>H<sub>5</sub>Cl and thus a decrease of the electron density of the ring takes place. This makes the benzene ring a relatively poor electron donor towards the Cyclohexane molecules. Hence a weak interaction between chlorobenzene and Cyclohexane is expected. In our system the dipole - induced dipole interaction between DMF and chlorobenzene is significant. Chlorobenzene is used as an intermediate in the production of commodities such as herbicides, dyestuffs and rubber. It is also used as a high-boiling solvent in many industrial applications as well as in the laboratory.

### EXPERIMENTAL SECTION

The chemicals used in the present work were analytical reagent (AR) and spectroscopic reagent (SR) grades with minimum assay of 99.9% were obtained from E-Merk Ltd (India). Various concentrations of the ternary liquid mixtures were prepared in terms of mole fraction, out of which the mole fraction of the second component Cyclohexane ( $X_2 = 0.4$ ) was kept fixed while the mole fractions of remaining two ( $X_1$  and  $X_3$ ) were varied from 0.0 to 0.6. There is nothing significant in fixing the second component at 0.4.

#### (i) Velocity Measurement:-

The velocity of ultrasonic wave in the ternary mixture have been measured using multi-frequency ultrasonic interferometer with an high degree of accuracy operating at 11 different frequencies (Model M-84) supplied by M/s Mittal Enterprises, New Delhi. The measuring cell of interferometer is a specially designed double walled vessel with provision for temperature constancy. An electronically operated digital constant temperature bath (Model SSI-03spl) supplied by M/s Mittal Enterprises, New Delhi, operating in the temperature range -10°C to 85°C with an accuracy of  $\pm 0.1$ K has been used to circulate water through the outer jacket of the double walled measuring cell containing the experimental liquid.

#### (ii) Density Measurement:-

The densities of the mixture were measured using a 25ml specific gravity bottle. The specific gravity bottle with the experimental mixture was immersed in a temperature controlled water bath. The density was measured using the formula

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

where,  $w_1$  = weight of distilled water,  $w_2$  = Weight of experimental liquid,  
 $\rho_1$  = Density of water,  $\rho_2$  = Density of experimental liquid.

#### (iii) Viscosity measurement:-

The viscosities of the ternary mixture were measured using an Oswald's viscometer calibrated with double distilled water. The Oswald's viscometer with the experimental mixture was immersed in a temperature controlled water bath. The time of flow was measured using a digital racer stop watch with an accuracy of 0.1 sec. The viscosity was determined using the relation,

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

Where,  $\eta_1$  = Viscosity of water,  $\eta_2$  = Viscosity of mixture,  $\rho_1$  = Density of water,  $\rho_2$  = Density of mixture,  $t_1$  = Time of flow of water,  $t_2$  = Time of flow of mixture.

### THEORETICAL ASPECT

The following thermodynamic parameters were calculated from Jacobson's relation [8 – 10].

#### (i) Adiabatic Compressibility ( $\beta$ ):-

The adiabatic compressibility is the fractional decrease of volume per unit increase of pressure, when no heat flows in or out. It is calculated from the speed of sound (U) and the density ( $\rho$ ) of the medium by using the equation of Newton Laplace as,

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

**(ii) Intermolecular free length ( $L_f$ ) :-**

The intermolecular free length is the distance between the surfaces of the neighboring molecules. It is calculated by using the relation

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

where,  $K_T$  is the temperature dependent constant and ' $\beta$ ' is the adiabatic compressibility.

**(iii) Free Volume ( $V_f$ ) :-** Free volume in terms of ultrasonic velocity( $U$ ) and the viscosity( $\eta$ ) of liquid is

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

Where ' $M_{\text{eff}}$ ' is the effective mass of the mixture, ' $K$ ' is a dimensionless constant independent of temperature and liquid. Its value is  $4.281 \times 10^9$ .

**(iv) Internal Pressure ( $\Pi_i$ ):-**

The measurement of internal pressure is important in the study of the thermodynamic properties of liquids. The internal pressure is the cohesive force, which is a resultant of force of attraction and force of repulsion between the molecules. It is calculated by using the relation,

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

Where, ' $b$ ' stands for cubic packing, which is assumed to be '2' for all liquids, ' $k$ ' is a dimensionless constant independent of temperature and nature of liquids. Its value is  $4.281 \times 10^9$ . ' $T$ ' is the absolute temperature in Kelvin, ' $M$ ' is the effective molecular weight, ' $R$ ' is the Universal gas constant, ' $\eta$ ' is the viscosity of solution in  $\text{N.S.m}^{-2}$ , ' $U$ ' is the ultrasonic velocity in  $\text{m.s}^{-1}$  and ' $\rho$ ' is the density in  $\text{Kg.m}^{-3}$  of solution.

**(v) Relaxation time ( $\tau$ ):-**

Relaxation time is the time taken for the excitation energy to appear as translational energy and it depends on temperature and impurities. The relaxation time can be calculated from the relation,

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

Where, ' $\beta$ ' is the adiabatic compressibility and ' $\eta$ ' is the viscosity of the mixture.

**(vi) Acoustic impedance ( $Z$ ):-**

The specific acoustic impedance is given by,

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

Where, ' $U$ ' and ' $\rho$ ' are velocity and density of the mixture.

**(vii) Gibb's free energy:-**

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

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

Where, ' $\tau$ ' is the viscous relaxation time, ' $T$ ' is the absolute temperature, ' $k$ ' is the Boltzmann's constant and ' $h$ ' is the Planck's constant.

**(viii) Absorption coefficient:-**

Absorption coefficient or attenuation coefficient is a characteristic of the medium and it depends on the external condition like temperature, pressure and frequency of measurement. It is given by the following relation [11].

$$\alpha = 8\pi^2 \eta f^2 / 3\rho U^2 \quad \text{----- (8)}$$

Where, ' $\eta$ ' is the viscosity in  $\text{NS.m}^{-2}$ , ' $U$ ' is the ultrasonic velocity in  $\text{m.s}^{-2}$ , ' $\rho$ ' in  $\text{Kg.m}^{-3}$  and ' $f$ ' is the frequency in Hz.

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

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

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

## RESULTS AND DISCUSSION

The experimental data relating to density, viscosity and velocity at 318 K for frequencies 2MHz, 4 MHz, 6 MHz, 8 MHz for the mixture are given in table-I. The calculated values of adiabatic compressibility ( $\beta$ ), free length ( $L_f$ ) and free volume ( $V_f$ ) are reported in table-2. Internal pressure ( $\Pi_i$ ), viscous relaxation time ( $\tau$ ) and Gibb's free energy ( $\Delta G$ ) are reported in table-3. Further acoustic impedance ( $Z$ ) and absorption coefficient, for the mixture are presented in table-4. The variation of different parameters with frequency and concentration of DMF are shown in fig.1 to 16.

**TABLE – 1 : Values of Density ( $\rho$ ), Viscosity ( $\eta$ ) and velocity ( $U$ ) at 2 MHz, 4 MHz, 6 MHz & 8 MHz**

Mole fraction		Density ( $\rho$ ) $\text{Kg.m}^{-3}$	Viscosity ( $\eta$ ) $(10^{-3} \text{ N.s.m}^{-2})$	Velocity ( $U$ ) $\text{m.s}^{-2}$			
$X_1$	$X_3$			2 MHz	4 MHz	6 MHz	8 MHz
0.0000	0.6000	975.254	0.5486	1172.2	1169.2	1166.4	1165.9
0.0999	0.4999	959.400	0.5509	1184.7	1181.6	1178.1	1176.9
0.1998	0.4001	942.248	0.5536	1198.9	1194.6	1190.6	1188.1
0.3001	0.3000	923.528	0.5573	1215.0	1209.6	1205.4	1202.0
0.4000	0.1999	903.075	0.5594	1230.6	1224.4	1220.6	1216.0
0.4998	0.1001	880.712	0.5615	1247.0	1241.8	1237.6	1233.0
0.5997	0.0000	855.982	0.5631	1266.1	1261.6	1256.4	1251.5

**TABLE – 2: Values of Adiabatic compressibility ( $\beta$ ), Free length ( $L_f$ ) and free volume ( $V_f$ ) at 2 MHz, 4 MHz, 6 MHz & 8 MHz**

Mole fraction		Adiabatic compressibility ( $\beta$ ) $(10^{-10} \text{ N}^{-1} \cdot \text{m}^2)$				Free length ( $L_f$ ) $(10^{-10} \text{ m})$				Free volume ( $V_f$ ) $(10^{-7} \text{ m}^3 \cdot \text{mol}^{-1})$			
$X_1$	$X_3$	2 MHz	4 MHz	6 MHz	8 MHz	2 MHz	4 MHz	6 MHz	8 MHz	2 MHz	4 MHz	6 MHz	8 MHz
0.0000	0.6000	7.4624	7.5007	7.5368	7.5432	0.5542	0.5562	0.5576	0.5578	1.1372	1.1308	1.1268	1.1261
0.0999	0.4999	7.4264	7.4655	7.5099	7.5252	0.5531	0.5551	0.5566	0.5571	1.0809	1.0751	1.0708	1.0692
0.1998	0.4001	7.3836	7.4368	7.4869	7.5184	0.5518	0.5539	0.5557	0.5569	1.0255	1.0199	1.0149	1.0117
0.3001	0.3000	7.3349	7.4005	7.4522	7.4944	0.5500	0.5525	0.5544	0.5560	0.9706	0.9642	0.9592	0.9551
0.4000	0.1999	7.3121	7.3863	7.4324	7.4887	0.5492	0.5520	0.5537	0.5558	0.9194	0.9125	0.9082	0.9031
0.4998	0.1001	7.3018	7.3631	7.4132	7.4686	0.5488	0.5511	0.5530	0.5550	0.8686	0.8633	0.8589	0.8541
0.5997	0.0000	7.2878	7.3399	7.4008	7.4588	0.5482	0.5505	0.5525	0.5547	0.8215	0.8166	0.8121	0.8073

The intermolecular free length as well as relaxation time are properties of liquid mixtures which mainly affect the ultrasonic velocity. Intermolecular free length as well as relaxation time decrease with increase in ultrasonic velocity. In low frequency range the molecules find greater time for interaction, for which they move with low velocities. When frequency increases, the molecules become less interacting with each other and move with larger velocities. The change of free length and relaxation time with frequency confirm the above behavior (Graph 2 & 5).

**TABLE – 3: Values of Internal pressure ( $\Pi_i$ ), viscous relaxation time ( $\tau$ ) and Gibb's free energy ( $\Delta G$ ) at 2 MHz, 4 MHz, 6 MHz & 8 MHz**

Mole fraction		Internal pressure ( $\Pi_i$ ) $(\times 10^6 \text{ N.m}^{-2})$				Viscous relaxation time ( $\tau$ ) $(\times 10^{-12} \text{ s})$				Gibb's free energy ( $\Delta G$ ) $(\times 10^{-20} \text{ kJ.mol}^{-1})$			
$X_1$	$X_3$	2 MHz	4 MHz	6 MHz	8 MHz	2 MHz	4 MHz	6 MHz	8 MHz	2 MHz	4 MHz	6 MHz	8 MHz
0.0000	0.6000	106.49	106.69	106.81	106.84	0.5445	0.5486	0.5513	0.5517	0.5634	0.5667	0.5688	0.5692
0.0999	0.4999	110.01	110.21	110.35	110.41	0.5447	0.5487	0.5516	0.5527	0.5635	0.5667	0.5690	0.5699
0.1998	0.4001	113.71	113.92	114.11	114.23	0.5450	0.5490	0.5526	0.5549	0.5637	0.5670	0.5698	0.5717
0.3001	0.3000	117.62	117.88	118.09	118.25	0.5451	0.5499	0.5538	0.5569	0.5638	0.5677	0.5708	0.5732
0.4000	0.1999	121.60	121.90	122.09	122.32	0.5454	0.5509	0.5544	0.5586	0.5641	0.5685	0.5712	0.5746
0.4998	0.1001	125.77	126.03	126.25	126.48	0.5467	0.5513	0.5550	0.5592	0.5651	0.5688	0.5718	0.5750
0.5997	0.0000	129.96	130.21	130.46	130.71	0.5472	0.5515	0.5556	0.5600	0.5655	0.5690	0.5722	0.5757

Since the association of the interacting molecules varies with the frequency of the ultrasonic wave, the cohesive force between the molecules also varies. Cohesive force as well as internal pressure increases as frequency increases.

Gibb's free energy increases with increase in frequency as well as mole fraction of DMF. Increasing value of Gibb's free energy suggests that the closer approach of unlike molecules is due to hydrogen bonding [12-13]. The increase in Gibb's free energy also suggests shorter time for rearrangement of the molecules in the mixture.

**TABLE – 4: Values of Acoustic impedance (Z) and absorption coefficient ( $\alpha$ ) at 2 MHz, 4 MHz, 6 MHz & 8 MHz**

Mole fraction		Acoustic impedance (Z) ( $\times 10^6 \text{ Kg.m}^2.\text{s}^{-1}$ )				Absorption coefficient ( $\alpha$ ) np.m <sup>-1</sup>			
X <sub>1</sub>	X <sub>3</sub>	2 MHz	4 MHz	6 MHz	8 MHz	2 MHz	4 MHz	6 MHz	8 MHz
0.0000	0.6000	1.1446	1.1403	1.1375	1.1370	43.0068	173.3245	391.8547	697.2282
0.0999	0.4999	1.1374	1.1332	1.1303	1.1291	43.0222	173.3439	392.0791	698.4516
0.1998	0.4001	1.1297	1.1255	1.1218	1.1195	43.0418	173.4381	392.7965	701.2466
0.3001	0.3000	1.1221	1.1171	1.1132	1.1101	43.0491	173.7372	393.6375	703.7646
0.4000	0.1999	1.1113	1.1057	1.1023	1.0981	43.0762	174.0542	394.0642	705.8688
0.4998	0.1001	1.0982	1.0937	1.0900	1.0859	43.1775	174.1597	394.5234	706.6180
0.5997	0.0000	1.0838	1.0795	1.0754	1.0713	43.2149	174.2329	394.9626	707.6648

Attenuation coefficient which is a characteristic of the medium also increases with increase in frequency. This shows a similar behavior to that of the passage of electromagnetic waves through a conductor where the skin depth decreases with frequency of the incoming wave. Decrease in skin depth means increase in absorption.

With increase in mole fraction of DMF, ultrasonic velocity, internal pressure, relaxation time and Gibb's free energy increases where as adiabatic compressibility, free length, free volume and acoustic impedance decreases. According to a model proposed by Eyring Kincaid [14], ultrasonic velocity should increase if the intermolecular free length decreases as a result of mixing of components. The decrease in adiabatic compressibility suggests that there is significant interaction between unlike molecules. The value of free volume decreases where as internal pressure increases due to the various dispersive interactions and the columbic interaction existing between the component molecules.

When an acoustic wave travels in a medium, there is a variation of pressure from particle to particle. The ratio of instantaneous pressure excess at any particle of the medium to the instantaneous velocity of that particle is known as "specific acoustic impedance" of the medium. This factor is governed by the inertial and elastic properties of the medium. In the present investigation acoustic impedance decreases with increase in concentration of DMF, Such a decreasing value of 'Z' further supports the possibility of molecular interaction between the unlike molecules.

Viscous relaxation time and Gibb's free energy increases with increase in mole fraction of DMF. The viscous relaxation time shows the presence of molecular interaction by addition of solvent concentration and the same is confirmed by Gibb's free energy parameter.

In order to understand the nature of molecular interactions between the components of the liquid mixture, it is of interest to discuss the same in terms of excess parameter rather than actual values. Excess values of adiabatic compressibility ( $\beta^E$ ), excess free length ( $L_f^E$ ) and free volume ( $V_f$ ) are reported in table-5. Excess internal pressure ( $\Pi_i^E$ ), excess relaxation time ( $\tau^E$ ) and excess Gibb's free energy ( $\Delta G^E$ ) are reported in table-6 and excess value of acoustic impedance ( $Z^E$ ) is reported in table-7. Also the variations of excess values of parameters with frequency are shown in fig. 17 to 23.

**TABLE – 5: Excess values of adiabatic compressibility ( $\beta$ ), free length ( $L_f$ ) and free volume ( $V_f$ ) at 2 MHz, 4 MHz, 6 MHz & 8 MHz**

Mole fraction		Excess adiabatic compressibility ( $\beta^E$ ) ( $10^{-10} \text{ N}^{-1}.\text{m}^2$ )				Excess free length ( $L_f^E$ ) ( $10^{-10} \text{ m}$ )				Excess free volume ( $V_f^E$ ) ( $10^{-7} \text{ m}^3.\text{mol}^{-1}$ )			
X <sub>1</sub>	X <sub>3</sub>	2 MHz	4 MHz	6 MHz	8 MHz	2 MHz	4 MHz	6 MHz	8 MHz	2 MHz	4 MHz	6 MHz	8 MHz
0.0000	0.6000	1.2901	0.8123	0.2989	0.2562	0.5305	0.3369	0.1488	0.1223	-0.1155	-0.447	-0.198	-0.250
0.0999	0.4999	1.2582	0.8141	0.3410	0.3097	0.5187	0.3402	0.1587	0.1465	-0.1498	-0.528	-0.051	-0.089
0.1998	0.4001	1.2200	0.8120	0.3853	0.3726	0.5039	0.3411	0.1785	0.1733	-0.1837	-0.608	-0.100	-0.082
0.3001	0.3000	1.1655	0.8043	0.4175	0.4179	0.4826	0.3397	0.1936	0.1933	-0.2170	-0.692	-0.248	-0.243
0.4000	0.1999	1.1389	0.8221	0.4667	0.4837	0.4727	0.3488	0.2161	0.2220	-0.2463	-0.734	-0.345	-0.355
0.4998	0.1001	1.1238	0.8297	0.5153	0.5338	0.4667	0.3536	0.2378	0.2443	-0.2754	-0.753	-0.429	-0.440
0.5997	0.0000	1.1061	0.8443	0.5720	0.5956	0.4602	0.3615	0.2630	0.2714	-0.3006	-0.744	-0.485	-0.500

**TABLE – 6: Excess values of Internal pressure ( $\pi_i^E$ ), Viscous relaxation time ( $\tau^E$ ) and Gibb's free energy ( $\Delta G^E$ ) at 2 MHz, 4 MHz, 6 MHz & 8 MHz .**

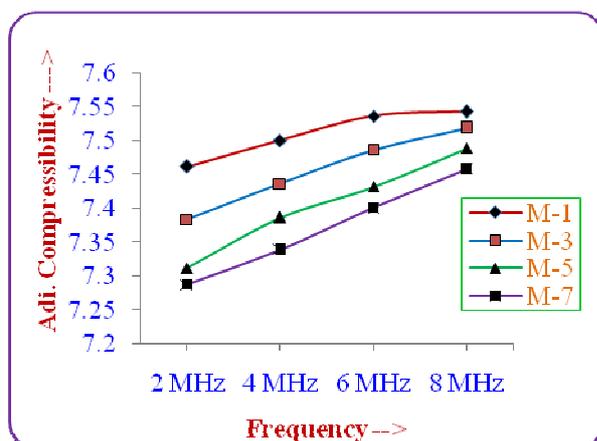
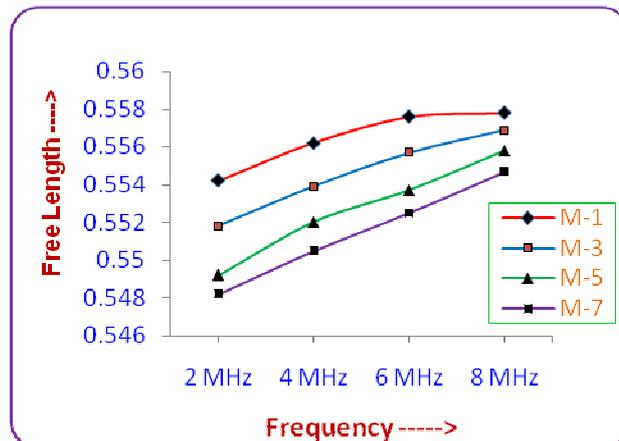
Mole fraction		Excess internal pressure ( $\Pi_i^E$ ) ( $\times 10^6 \text{ N.m}^{-2}$ )				Excess viscous relaxation time ( $\tau^E$ ) ( $\times 10^{-12} \text{ s}$ )				Excess Gibb's free energy ( $\Delta G^E$ ) ( $\times 10^{-20} \text{ kJ.mol}^{-1}$ )			
$X_1$	$X_3$	2 MHz	4 MHz	6 MHz	8 MHz	2 MHz	4 MHz	6 MHz	8 MHz	2 MHz	4 MHz	6 MHz	8 MHz
0.0000	0.6000	5.085	3.125	1.169	1.005	0.7769	0.41	0.0171	0.016	0.7002	0.3628	0.0838	0.066
0.0999	0.4999	4.251	2.436	0.637	0.511	0.7801	0.4419	0.0827	0.0579	0.7031	0.3939	0.0941	0.0736
0.1998	0.4001	3.574	1.913	0.295	0.237	0.7825	0.4748	0.1537	0.143	0.7050	0.4253	0.1584	0.1489
0.3001	0.3000	3.068	1.605	0.145	0.138	0.783	0.5138	0.2262	0.2256	0.7052	0.4616	0.2238	0.2221
0.4000	0.1999	2.693	1.422	0.086	0.147	0.7876	0.5551	0.2946	0.3067	0.7093	0.5002	0.2864	0.2946
0.4998	0.1001	2.496	1.325	0.153	0.223	0.8008	0.5887	0.3624	0.3761	0.7200	0.5324	0.3482	0.3575
0.5997	0.0000	2.329	1.298	0.296	0.390	0.8068	0.6223	0.431	0.4488	0.7251	0.5648	0.4110	0.4234

**TABLE – 7: Excess values of Acoustic impedance ( $Z^E$ ) at 2 MHz, 4 MHz, 6 MHz & 8 MHz**

Mole fraction		Excess acoustic impedance ( $Z^E$ ) ( $\times 10^6 \text{ Kg.m}^2.\text{s}^{-1}$ )			
$X_1$	$X_3$	2 MHz	4 MHz	6 MHz	8 MHz
0.0000	0.6000	-1.266	-0.7781	-0.3253	-0.2872
0.0999	0.4999	-1.222	-0.7803	-0.3733	-0.3443
0.1998	0.4001	-1.187	-0.7942	-0.4366	-0.4223
0.3001	0.3000	-1.152	-0.8163	-0.5033	-0.4988
0.4000	0.1999	-1.143	-0.8622	-0.5878	-0.5962
0.4998	0.1001	-1.161	-0.9181	-0.6889	-0.6987
0.5997	0.0000	-1.189	-0.9923	-0.8093	-0.8233

It is learnt that the dispersion forces are responsible for possessing positive excess values, while dipole-dipole, dipole-induced dipole, charge transfer interaction and hydrogen bonding between unlike molecules are responsible for possessing negative excess values [15].

In the present investigation excess values of adiabatic compressibility, free length, internal pressure, and Gibb's free energy are positive and excess acoustic impedance is negative. This suggests that in addition to dipole-dipole and dipole-induced dipole interactions, dispersive forces are also operative in the liquid mixture. Positive values of adiabatic compressibility and free length indicate weak molecular interaction between the components of the mixture, which results from dispersive forces [16-17]. The excess values of free volume and acoustic impedance are negative, which confirms the same.

**Fig.-1: Variation of adiabatic Compressibility with frequency****Fig.-2: Variation of free length with frequency**

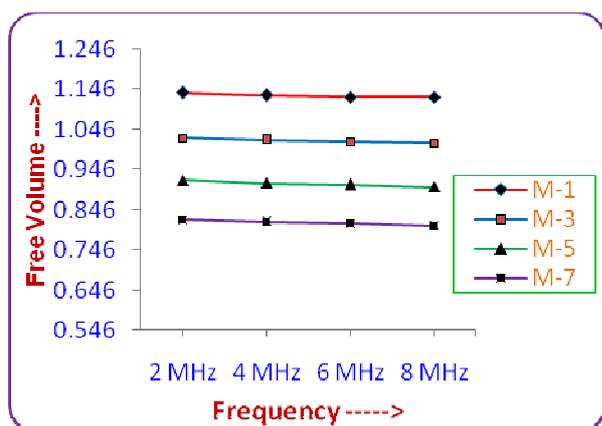


Fig.-3: Variation of free volume with frequency

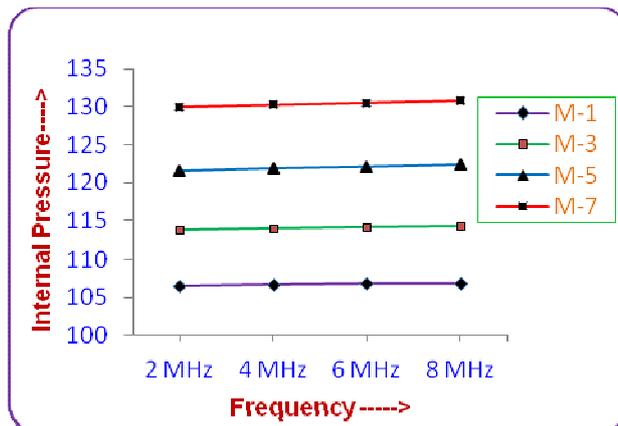


Fig.-4: Variation of Internal Pressure with frequency

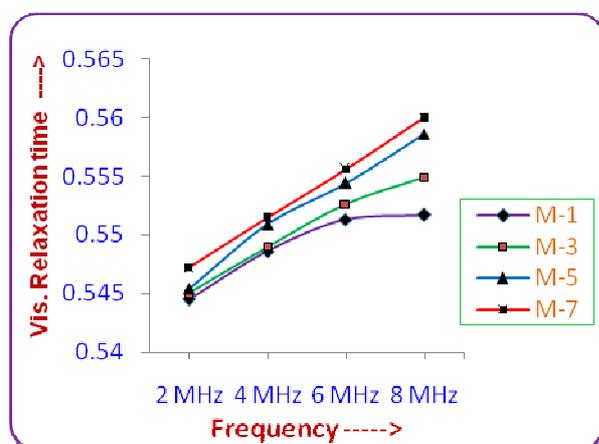


Fig.-5: Variation of Viscosity relaxation time with frequency.

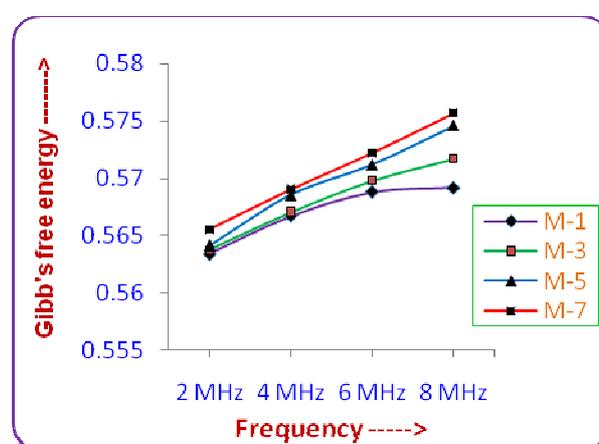


Fig.-6: Variation of Gibb's free energy with frequency.

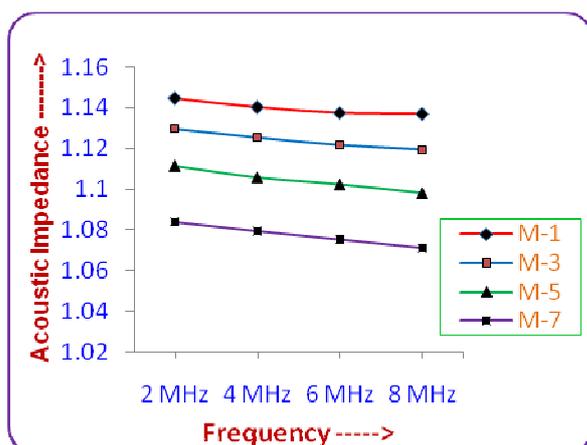


Fig.-7: Variation of Acoustic impedance with frequency.

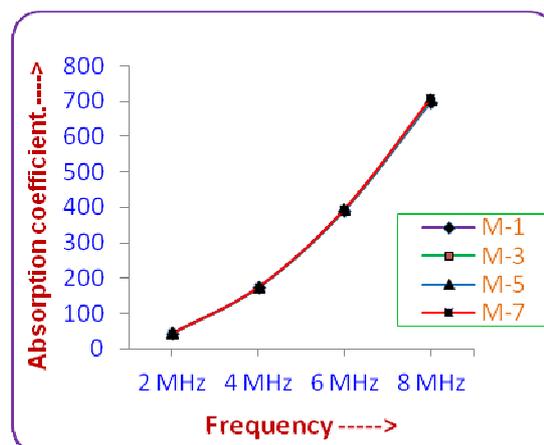


Fig.-8: Variation of absorption coefficient with frequency.

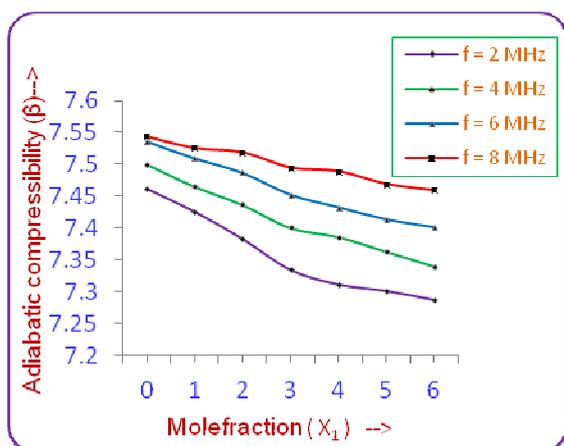


Fig.-9: Variation of adiabatic compressibility with mole fraction.

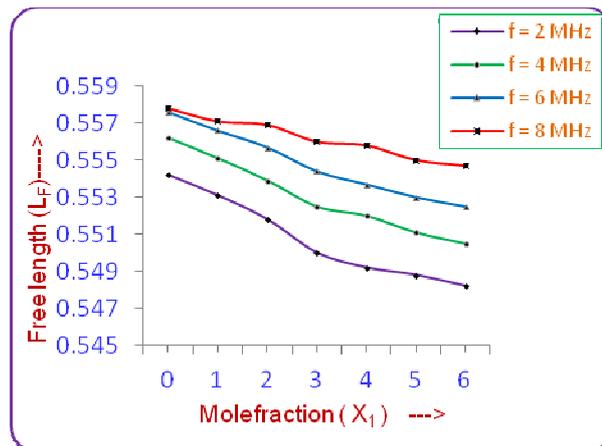


Fig.-10: Variation of free length with mole fraction.

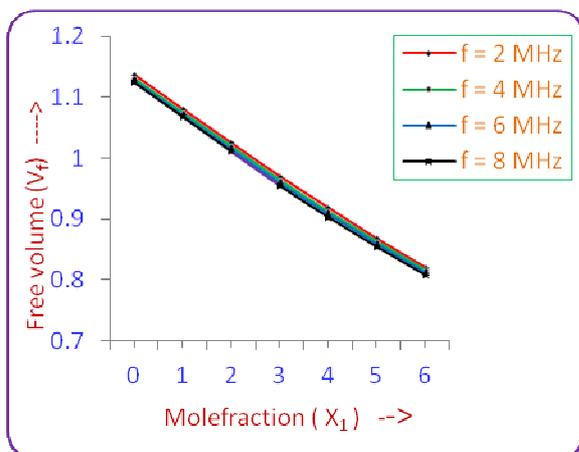


Fig.-11: Variation of free volume with mole fraction.

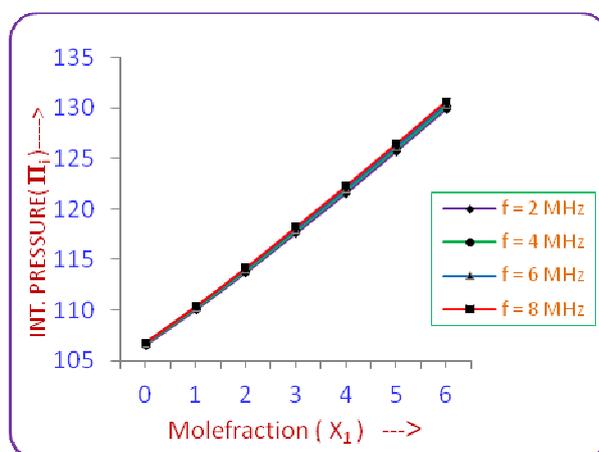


Fig.—12: Variation of Internal pressure with mole fraction.

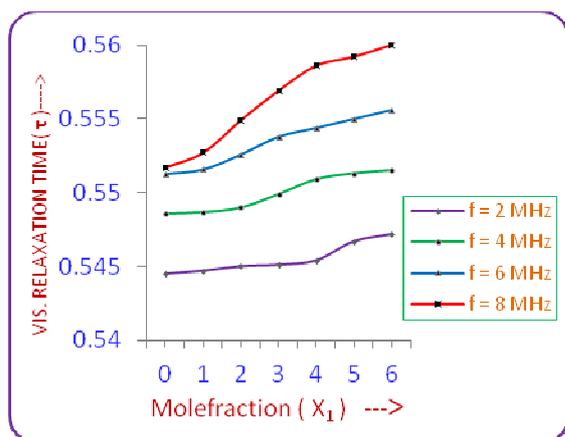


Fig.-13: Variation of Viscosity relaxation time with mole fraction.

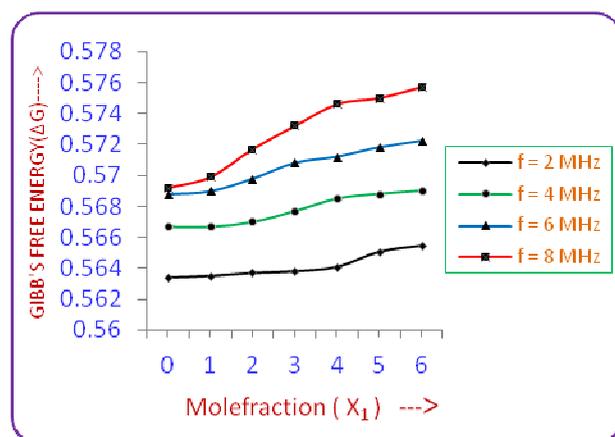


Fig.-14: Variation of Gibb's free energy with mole fraction.

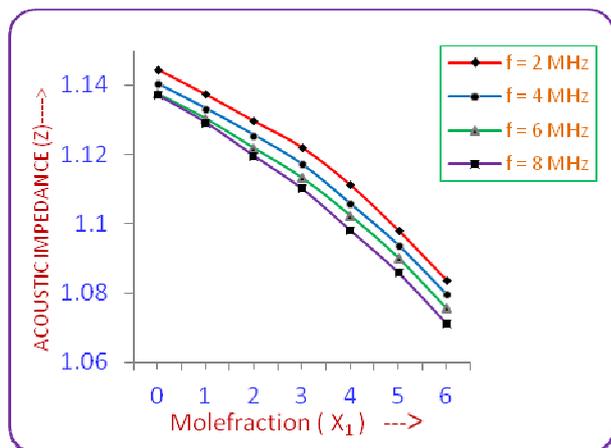


Fig.-15: Variation of acoustic impedance with mole fraction

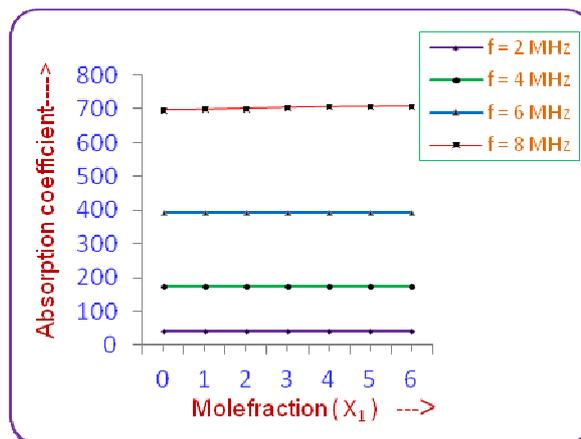


Fig.-16: Variation of absorption coefficient with mole fraction

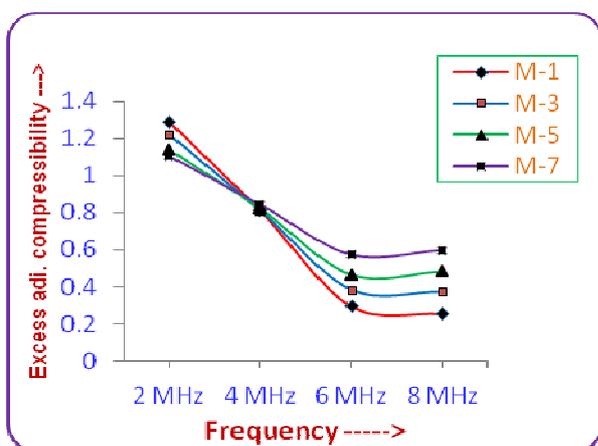


Fig.-17: Variation of Excess adiabatic compressibility with Frequency.

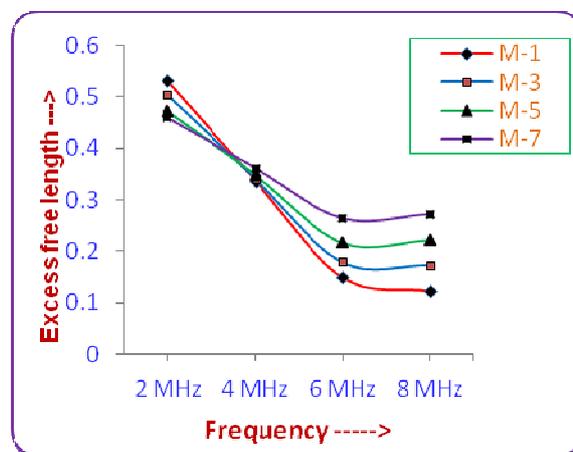


Fig.-18: Variation of Excess free length with frequency.

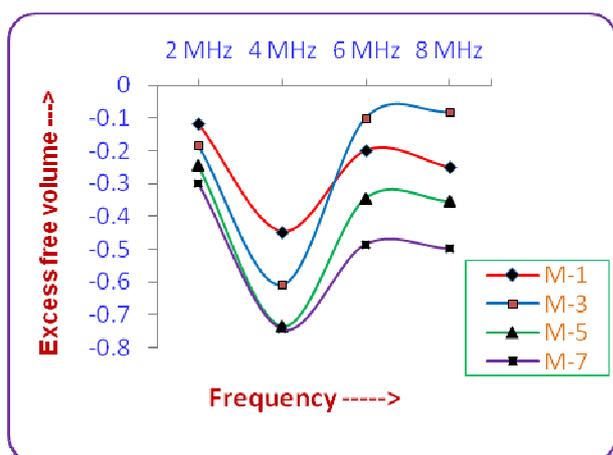


Fig.-19: Variation of Excess free volume with frequency.

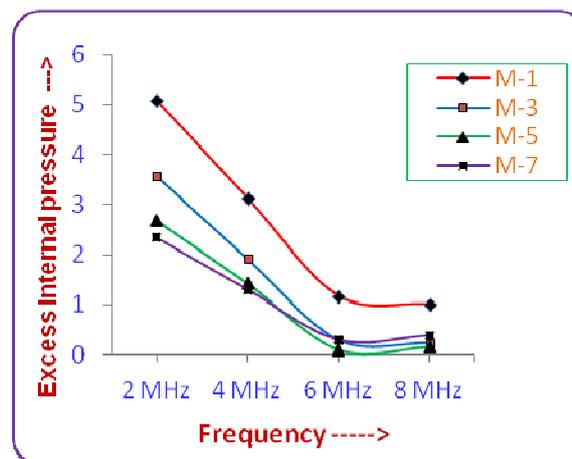


Fig.-20: Variation of Excess internal pressure with frequency.

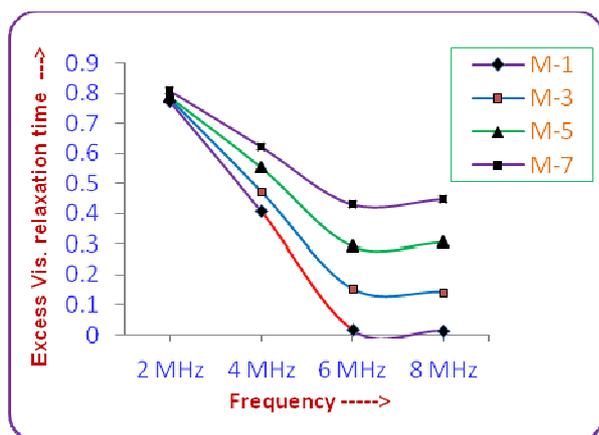


Fig.-21: Variation of Excess relaxation time with frequency.

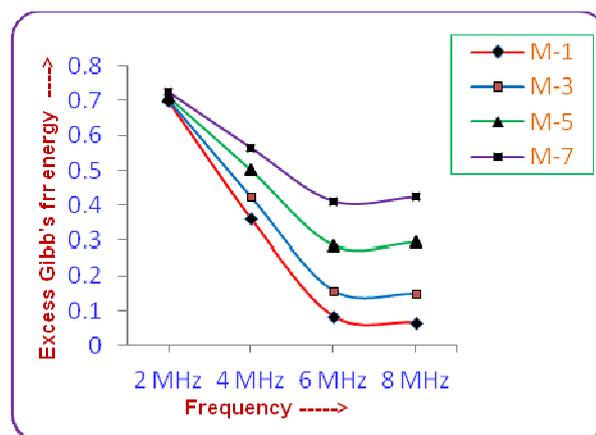


Fig.-22: Variation of Excess Gibb's free energy with frequency.

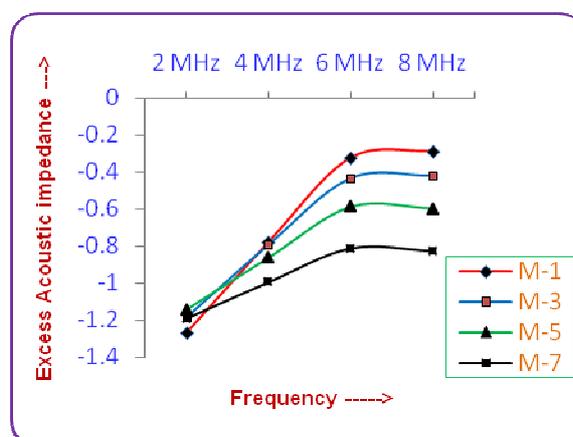


Fig.-23: Variation of Excess acoustic impedance with Frequency.

## CONCLUSION

When frequency of the incoming waves is large, the response of the medium to the changes is relatively less. Hence it is observed that the acoustic parameters and their excess values are highly affected at low frequency range as compared to the high frequency range. It may be concluded that in describing the thermal and acoustic parameters, the variation of the frequency plays a vital role.

Further the non-linear variation of acoustical parameters with concentration reveals the complex formation, which is also strongly supported by the excess parameters in the ternary liquid mixtures.

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