

## Ultrasonic Investigations of Molecular Interactions of Binary Liquid Mixture of Diethylenetriamine (DETA) with 2-Ethoxy Ethanol at temperature 296 K at frequencies 1MHZ, 3MHZ & 5MHZ

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### ABSTRACT:

*Viscosity ( $\eta$ ), Density ( $\rho$ ) and Ultrasonic velocity ( $U$ ) of the binary system of Diethylenetriamine (DETA) and 2-Ethoxy Ethanol are measured over entire composition range. These measurements are done at constant temperature 296 K and at frequencies 1MHZ, 3MHZ & 5MHZ. The acoustic parameters computed using these data values are intermolecular free length ( $L_f$ ), Rao constant ( $R$ ), Wada's constant ( $W$ ), adiabatic compressibility ( $\beta_{ad}$ ), excess intermolecular free length ( $L_f^E$ ), excess adiabatic compressibility ( $\beta_{ad}^E$ ) and excess ultrasonic velocity ( $U^E$ ). These parameters have to be interpreted in terms of intermolecular interactions at frequencies 1MHZ, 3MHZ & 5MHZ at constant temperature 296K. This work aims to evolve the impact of ultrasonic sound over the molecular interaction of binary liquid mixture quoted above at the said conditions.*

**KEYWORDS:** Ethoxy ethanol, Wada's constant, excess adiabatic compressibility, Rao's constant

### INTRODUCTION:

The experimental measurements of ultrasonic velocity, density and viscosity of pure liquids & liquid mixtures are easy & important. These are important in different fields and industries. Many people worked in this fashion for study of molecular interactions of pure liquids & liquid mixtures [1-3]. In the present study the chemicals used are Diethylenetriamine and 2-Ethoxyethanol. DETA is miscible in water [4] is an organic compound with formula  $\text{HN}(\text{CH}_2\text{CH}_2\text{NH}_2)_2$ . This is colorless hygroscopic liquid. It is soluble in water and polar organic solvents. It is not soluble in simple hydrocarbons. It is a weak base & its aqueous solution is alkaline [5]. It is common curing agent for epoxy resins in epoxy adhesives [6]. It has been evaluated for use in countermine system. It would be used to ignite and consume the explosive fill of land mines in beach and surf zones [7]. Its dielectric constant is 12.2 at 1 KHZ and polarity is approximately zero. It is non-polar. It is polydentate ligand.

2-Ethoxy Ethanol is a solvent used widely in commercial and industrial applications. It is clear, colorless, nearly odorless liquid. It is miscible with water, ethanol, diethyl ether,

acetone and ethyl acetate [8]. It has an ideal property as multi-purpose cleaner. Therefore, it is used in products such as varnish removers and degreasing solutions. The chemical formula of 2-Ethoxyethanol is  $C_4H_{10}O_2$  or  $CH_3CH_2OCH_2CH_2OH$ . It plays a role as protic solvent. Its dielectric constant & polarity index is not available. In the present study, density, viscosity and ultrasonic velocity of Diethylenetriamine and 2-Ethoxyethanol binary mixture have been measured and used to compute the acoustic parameters such as intermolecular free length ( $L_f$ ), Rao constant (R), Wada's constant (W), adiabatic compressibility ( $\beta_{ad}$ ), excess intermolecular free length ( $L_f^E$ ), excess adiabatic compressibility ( $\beta_{ad}^E$ ) and excess ultrasonic velocity ( $U^E$ ). These data values are to be used to interpret the intermolecular interactions in this binary mixture for entire mole fraction range of DETA in 2-Ethoxyethanol.

## EXPERIMENTAL:

Chemicals used are obtained from; DETA from Loba Chemicals Pvt. Ltd. Mumbai, and 2-Ethoxyethanol from SDFCL, Mumbai. It is analytical reagent. Density of the pure components and their mixtures were measured by using 10 ml specific gravity bottle up to the accuracy (0.001 g) [9]. The viscosity of pure liquids and their mixtures [10] were measured using Ostwald's viscometer with an accuracy of  $\pm 0.001 \text{ Nsm}^{-2}$ . Ultrasonic sound velocities were measured using multifrequency ultrasonic interferometer MX-3 (H. C. Memorial Scientific Corporation, Ambala Cantonment) with working frequencies 1MHZ, 3MHZ & 5MHZ. The Abbe's refractometer is very popular and owes its popularity to its convenience, its wide range ( $n_D = 1.3$  to  $1.7$ ), and to the minimal sample is needed [11]. The accuracy of the instrument is about  $\pm 0.0002$ ; its precision is half this figure. The improvement in accuracy is obtained by replacing the compensator with a monochromatic source and by using larger and more precise prism mounts. The former provides a much sharper critical boundary and the latter allows a more accurate determination of the prism position. From the measured values of Density ( $\rho$ ), viscosity ( $\eta$ ) and Ultrasonic velocity ( $U$ ), the acoustic parameters intermolecular free length ( $L_f$ ), Rao constant (R), Wada's constant (W), adiabatic compressibility ( $\beta_{ad}$ ), excess intermolecular free length ( $L_f^E$ ), excess adiabatic compressibility ( $\beta_{ad}^E$ ) and excess ultrasonic velocity ( $U^E$ ) were computed using the following equations.

## Theory & Calculations:

1. **ULTRASONIC VELOCITY:** It is the velocity of the sound waves propagating through the binary liquid mixture.  $\lambda$  is the wavelength of the sound waves inside the binary liquid mixture.  

$$U = n \lambda \text{ m/s}$$
2. **INTERMOLECULAR FREE LENGTH:** It is the distance covered by sound wave between the surfaces of the neighboring molecules. It is measure of intermolecular attractions between the components in a binary liquid mixture.  

$$L_f = k \beta_{ad}^{1/2} \text{ m}$$

K is a constant known as Jacobson's constant given by  

$$K = (93.875 + 0.375 T \text{ in degree Kelvin}) \times 10^{-8}$$
3. **RAO CONSTANT OR MOLAR SOUND VELOCITY:** It is required in the study of acoustical properties of pure liquids & liquid mixtures. Variation in the values of Rao's constant with molar composition is an evidence of significant interaction between the components of binary or ternary system.

$$R = V_m U^{1/3}$$

4. **WADA'S CONSTANT OR MOLAR COMPRESSIBILITY:** It is required in the study of acoustical properties of pure liquids & liquid mixtures. It is also known as molar compressibility. Its value depends on the structure of pure liquid or liquid mixtures. Variations in Wada's constant with mole fraction of the solute provide evidence of molecular interaction between the components of binary or ternary system.

$$W = \frac{M}{\rho} \frac{1}{\beta_s^{1/7}} \quad \text{J/mol}$$

M is molar mass.

5. **ADIABATIC COMPRESSIBILITY:** It determines the orientation of the solvent molecules around the liquid molecules. The structural change in molecules in a liquid mixture takes place due to the existence of electrostatics field between the interacting molecules. The structural arrangement of the molecules affects the value of adiabatic compressibility. It is defined as fractional degrees of volume per unit increase of pressure when no heat flows in or out. It is therefore a measure of intermolecular association or dissociation or repulsion.

$$\beta_{ad} = \frac{1}{u^2 \rho} \quad \text{m}^2/\text{N}$$

u ultrasonic velocity and  $\rho$  is density of liquid in SI

The values of above quoted excess acoustic parameters are computed using the general relation given below as relation 6.

6. Excess parameters are in general given by the relation

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

where  $A_{\text{id}} = \sum A_i X_i$ ,  $A_i$  is any acoustical parameters &  $X_i$  the mole fraction of that liquid component. The nature and degree of molecular interaction between the component molecules of the liquid mixture have been speculated through the size and extent of deviation of the excess parameters. There will be positive deviation if size of the solvent molecule is increased and if it is decreased then the deviation is negative.

## RESULTS AND DISCUSSION:

The acoustic parameters Ultrasonic velocity (U), Viscosity ( $\eta$ ), Density ( $\rho$ ) intermolecular free length ( $L_f$ ) and adiabatic compressibility ( $\beta_{ad}$ ) are presented in the table I. Other acoustic parameters such as Rao constant (R), Wada's constant (W), excess ultrasonic velocity ( $U^E$ ), excess intermolecular free length ( $L_f^E$ ) and excess adiabatic compressibility ( $\beta_{ad}^E$ ) are shown in table II. These parameters are evaluated at constant temperature 296K and at frequencies 1MHz, 3MHz & 5MHz. The behavior of these parameters with rise of mole fraction of DETA in this binary mixture and with increase of ultrasonic frequencies is shown graphically in different figures from 1 to 10. The change in ultrasonic velocity at constant temperature with increase in mole fraction of DETA and with increase of ultrasonic frequencies is illustrated in figure 1. Perusal of figure 1 indicates that the ultrasonic velocity is increasing with increase of concentration of DETA in the binary mixture of DETA and 2-Ethoxyethanol. The sound velocity is more pronounced at higher frequencies. This indicates that there is strong interaction between the components and the interaction increases with increase of concentration of DETA. Linear variation in ultrasonic velocity for all concentration range reveals that greater specific interaction exists between the components of this binary mixture [12].

**Table I:**

The acoustic parameters presented in table I are ultrasonic velocity, viscosity, density, intermolecular free length and adiabatic compressibility. These are evaluated at constant temperature 296K and at ultrasonic frequencies 1MHz, 3MHz and 5MHz.

| Mole fraction of DETA in 2-Ethoxyethanol | U (m/s) | (poise)  | $\rho$ (Kg/m <sup>3</sup> ) | $L_f \cdot 10^{-11}$ (m) | $Q_{ad} \cdot 10^{-10}$ (m <sup>2</sup> /N) |
|--|---------|----------|-----------------------------|--------------------------|---|
| <b>T=296°K and Frequency = 1MHZ</b>      |         |          |                             |                          |   |
| 0  | 1324.6  | 0.025    | 930                         | 5.0732                   | 6.12841                                     |
| 0.088535                                 | 1378.2  | 0.026371 | 933.125                     | 4.8677                   | 5.64203                                     |
| 0.184771                                 | 1432    | 0.027856 | 936.25                      | 4.677                    | 5.20861                                     |
| 0.289757                                 | 1486.2  | 0.029469 | 939.375                     | 4.499                    | 4.81955                                     |
| 0.404743                                 | 1530.5  | 0.031222 | 942.5                       | 4.3615                   | 4.52952                                     |
| 0.53123                                  | 1584.8  | 0.033132 | 945.625                     | 4.2051                   | 4.21049                                     |
| 0.671035                                 | 1636    | 0.035217 | 948.75                      | 4.0668                   | 3.93805                                     |
| 0.826378                                 | 1676    | 0.037499 | 951.875                     | 3.9632                   | 3.74  |
| 1  | 1706.6  | 0.04     | 955                         | 3.8858                   | 3.59528                                     |
| <b>T=296°K and Frequency = 3MHZ</b>      |         |          |                             |                          |   |
| 0  | 1338    | 0.025    | 930                         | 5.02239                  | 6.00627                                     |
| 0.088535                                 | 1393.02 | 0.026371 | 933.125                     | 4.81594                  | 5.52262                                     |
| 0.184771                                 | 1447.8  | 0.027856 | 936.25                      | 4.62598                  | 5.09555                                     |
| 0.289757                                 | 1491.96 | 0.029469 | 939.375                     | 4.48158                  | 4.78241                                     |
| 0.404743                                 | 1543.98 | 0.031222 | 942.5                       | 4.32341                  | 4.45077                                     |
| 0.53123                                  | 1593    | 0.033132 | 945.625                     | 4.18344                  | 4.16725                                     |
| 0.671035                                 | 1647.6  | 0.035217 | 948.75                      | 4.03813                  | 3.8828                                      |
| 0.826378                                 | 1684.98 | 0.037499 | 951.875                     | 3.94206                  | 3.70025                                     |
| 1  | 1725    | 0.04     | 955                         | 3.8443                   | 3.51899                                     |
| <b>T=296°K and Frequency = 5MHZ</b>      |         |          |                             |                          |   |
| 0  | 1349    | 0.025    | 930                         | 4.9814                   | 5.90872                                     |
| 0.088535                                 | 1398    | 0.026371 | 933.125                     | 4.7988                   | 5.48335                                     |
| 0.184771                                 | 1447    | 0.027856 | 936.25                      | 4.6285                   | 5.10119                                     |
| 0.289757                                 | 1496    | 0.029469 | 939.375                     | 4.4695                   | 4.75661                                     |
| 0.404743                                 | 1544.8  | 0.031222 | 942.5                       | 4.3211                   | 4.44605                                     |
| 0.53123                                  | 1594    | 0.033132 | 945.625                     | 4.1808                   | 4.16202                                     |
| 0.671035                                 | 1642    | 0.035217 | 948.75                      | 4.0519                   | 3.90933                                     |
| 0.826378                                 | 1691.6  | 0.037499 | 951.875                     | 3.9266                   | 3.67134                                     |
| 1  | 1744    | 0.04     | 955                         | 3.8024                   | 3.44274                                     |

Table II:

The acoustic parameters listed in table II are Rao's constant, Wada's constant, excess ultrasonic velocity, excess intermolecular free length and excess adiabatic compressibility. These parameters are evaluated at constant temperature 296K and at frequencies 1MHz, 3MHz and 5MHz.

| Mole fraction of DETA in 2-Ethoxyethanol | $R$<br>( $m^3/mol$ ) | $W$<br>(J/mol) | $U^E$<br>(m/s) | $L_f^E * 10^{-13}$<br>(m) | $Q_{ad}^E * 10^{-12}$<br>( $m^2/N$ ) |
|--|----------------------|----------------|----------------|---------------------------|--------------------------------------|
| <b>T=296°K and Frequency = 1MHZ</b>      |                      |                |                |                           |                                      |
| 0  | 0.001064             | 0.002006       | -0.4           | 0.153153                  | 0.369962                             |
| 0.088535                                 | 0.001092             | 0.002057       | 10.4915        | -7.0692                   | -19.8058                             |
| 0.184771                                 | 0.001121             | 0.002108       | 20.45408       | -12.462                   | -33.934                              |
| 0.289757                                 | 0.00115              | 0.00216        | 29.6424        | -16.225                   | -42.844                              |
| 0.404743                                 | 0.001177             | 0.002209       | 27.70862       | -15.547                   | -41.0365                             |
| 0.53123                                  | 0.001208             | 0.002263       | 34.50229       | -16.366                   | -41.2809                             |
| 0.671035                                 | 0.001237             | 0.002316       | 36.87014       | -14.961                   | -35.9817                             |
| 0.826378                                 | 0.001265             | 0.002366       | 26.65589       | -9.6525                   | -22.3233                             |
| 1  | 0.001291             | 0.002414       | 5.6            | -1.2793                   | -2.37116                             |
| <b>T=296°K and Frequency = 3MHZ</b>      |                      |                |                |                           |                                      |
| 0  | 0.0010678            | 0.002012       | 13             | -4.92763                  | -11.8437                             |
| 0.088535                                 | 0.0010964            | 0.002063       | 25.3115        | -12.2479                  | -31.7468                             |
| 0.184771                                 | 0.0011252            | 0.002115       | 36.25408       | -17.5664                  | -45.2404                             |
| 0.289757                                 | 0.0011518            | 0.002162       | 35.4024        | -17.9623                  | -46.5582                             |
| 0.404743                                 | 0.0011808            | 0.002214       | 41.18862       | -19.3552                  | -48.9111                             |
| 0.53123                                  | 0.0012096            | 0.002266       | 42.70229       | -18.5301                  | -45.6045                             |
| 0.671035                                 | 0.0012403            | 0.002321       | 48.47014       | -17.8246                  | -41.5074                             |
| 0.826378                                 | 0.0012673            | 0.00237        | 35.63589       | -11.7647                  | -26.2991                             |
| 1  | 0.0012956            | 0.002421       | 24             | -5.42406                  | -10.0002                             |
| <b>T=296°K and Frequency = 5MHZ</b>      |                      |                |                |                           |                                      |
| 0  | 0.001071             | 0.002017       | 24             | -9.02298                  | -0.0000003                           |
| 0.088535                                 | 0.001098             | 0.002065       | 30.2915        | -13.9634                  | -14.5266                             |
| 0.184771                                 | 0.001125             | 0.002114       | 35.45408       | -17.3106                  | -23.9923                             |
| 0.289757                                 | 0.001153             | 0.002164       | 39.4424        | -19.1725                  | -28.9288                             |
| 0.404743                                 | 0.001181             | 0.002215       | 42.00862       | -19.584                   | -29.6631                             |
| 0.53123                                  | 0.00121              | 0.002266       | 43.70229       | -18.7926                  | -26.9088                             |
| 0.671035                                 | 0.001239             | 0.002319       | 42.87014       | -16.4474                  | -20.1521                             |
| 0.826378                                 | 0.001269             | 0.002373       | 42.25589       | -13.3074                  | -11.0178                             |
| 1  | 0.0013               | 0.002429       | 43             | -9.61223                  | 0.00000046                           |

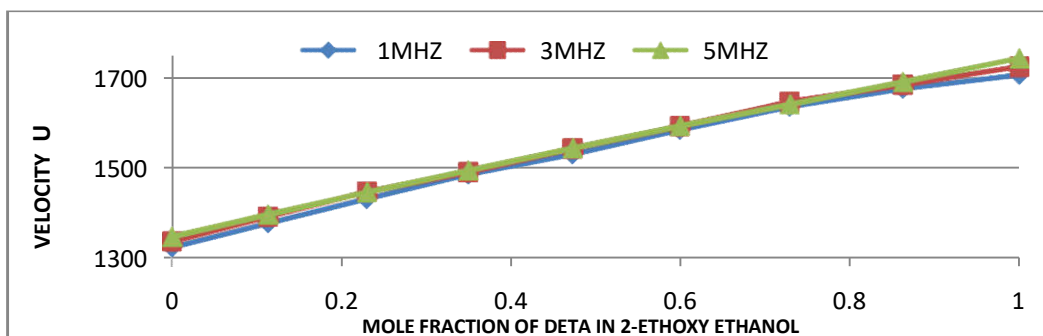


Fig 1 Graph between variation of ultrasonic velocity & mole fraction of DETA in 2-Ethoxyethanol at constant temperature 296K and frequencies 1MHz, 3MHz & 5MHz

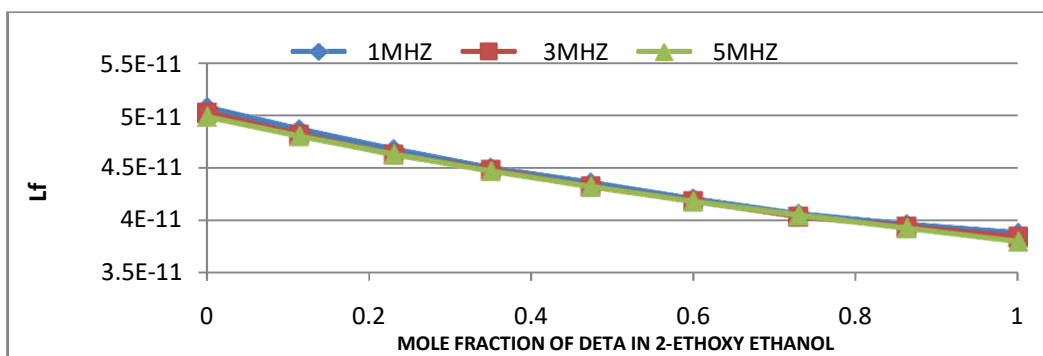


Fig 2 Graph between variation of intermolecular free length & mole fraction of DETA in 2-Ethoxyethanol at constant temperature 296K and frequencies 1MHz, 3MHz & 5MHz

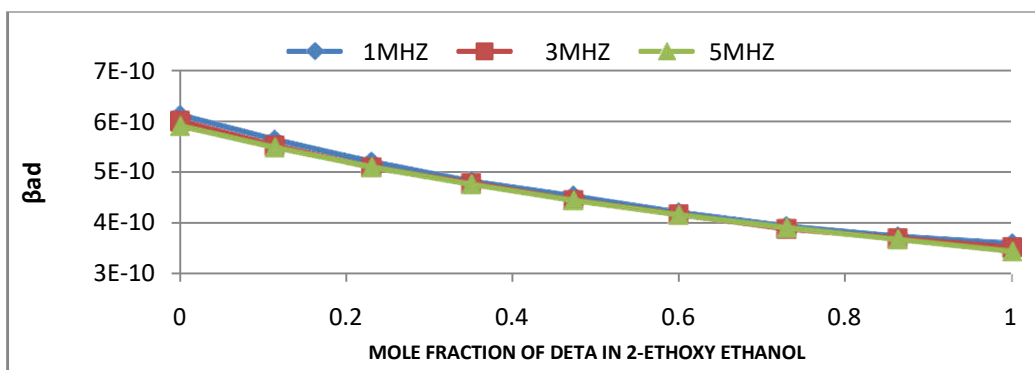


Fig 3 Graph between variation of adiabatic compressibility & mole fraction of DETA in 2-Ethoxyethanol at constant temperature 296K and frequencies 1MHz, 3MHz & 5MHz

The behavior of intermolecular free length and adiabatic compressibility with increase in mole fraction of DETA and with increase of ultrasonic frequencies are shown in figures 2 & 3 respectively. Observation of figures 3 & 4 show that the intermolecular free length and adiabatic compressibility both decrease with rise of concentration of DETA in the binary system for all the three frequencies. This behavior of intermolecular free length and adiabatic compressibility is exactly opposite to ultrasonic velocity. This is in accordance with Eyring Kincaid model. According to which decrease in intermolecular free length and adiabatic

compressibility increases the ultrasonic velocity. Intermolecular free length  $L_f$  is measure of intermolecular attraction between the components of the binary mixture. Behavior of  $L_f$  indicates strong interaction between the components of the binary system. The adiabatic compressibility  $\beta_{ad}$  is defined as fractional degrees of volume per unit increase of pressure when no heat flows in or out.  $\beta_{ad}$  is therefore a measure of intermolecular association or dissociation. The behavior of  $\beta_{ad}$  leads to increase in sound velocity through the mixture and also suggests strong forces of cohesion between the components of the binary system [13].

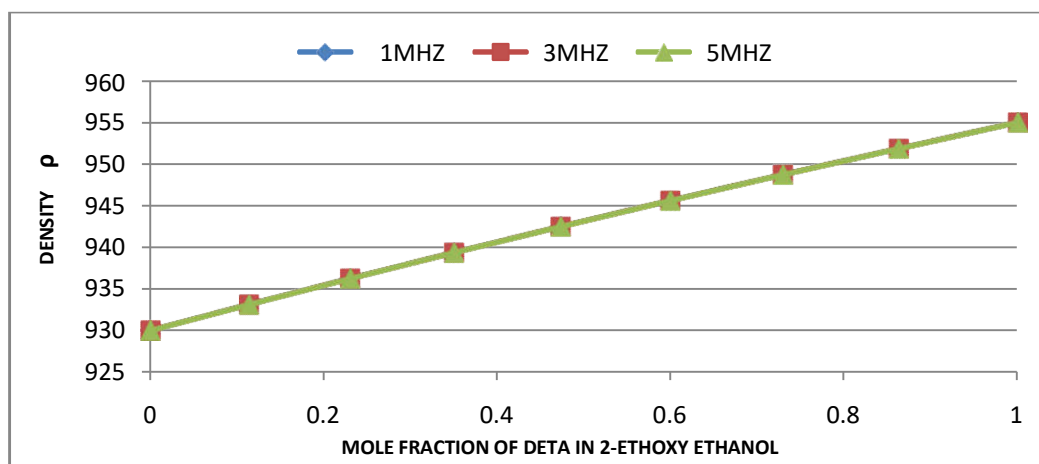


Fig 4 Graph between variation of density & mole fraction of DETA in 2-Ethoxyethanol at constant temperature 296K and frequencies 1MHz, 3MHz & 5MHz

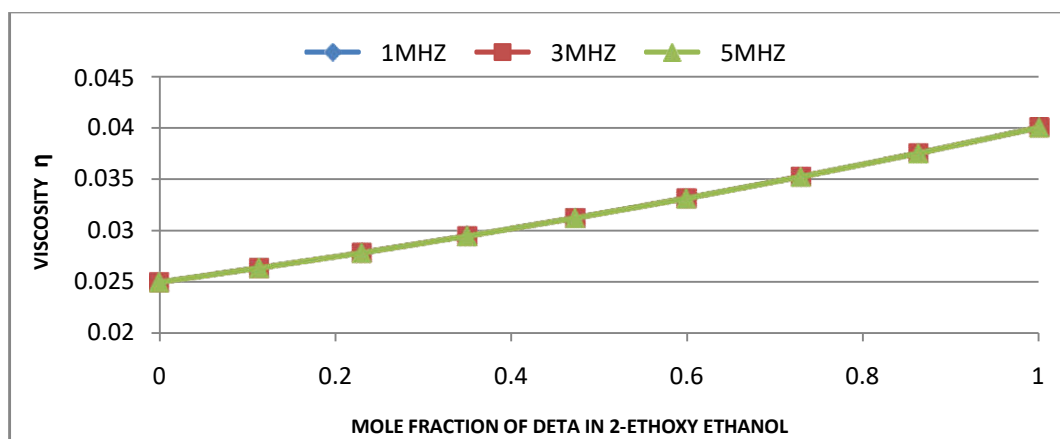


Fig 5 Graph between variation of viscosity & mole fraction of DETA in 2-Ethoxyethanol at constant temperature 296K and frequencies 1MHz, 3MHz & 5MHz

The behavior of density and viscosity of the binary mixture with increase in mole fraction of DETA and with increase of ultrasonic frequencies are shown in figures 4 & 5 respectively. Perusal of figures 4 & 5 shows increase in both density and viscosity with increase in concentration of the solute DETA for all the three frequencies. Density is a measure of solvent-solvent interactions. Increase in density with concentration indicates the increase in solvent-solvent and solute-solute interactions. Increase in density with concentration is due to shrinkage in the volume which in turn is due to presence of solute

molecules. This is structure-making. It suggests strong interaction in the components of binary system. Viscosity is also important parameter for understanding the structure as well as molecular interactions occurring in the binary liquid mixture. Viscosity variation is attributed to the structural changes. The density and viscosity both are independent of ultrasonic frequencies. Both are dependent on temperature. The above behavior of viscosity indicates that there is existence of interaction between the constituents of the mixture [14]. Thus there is existence of strong molecular interaction between the constituents of this binary system.

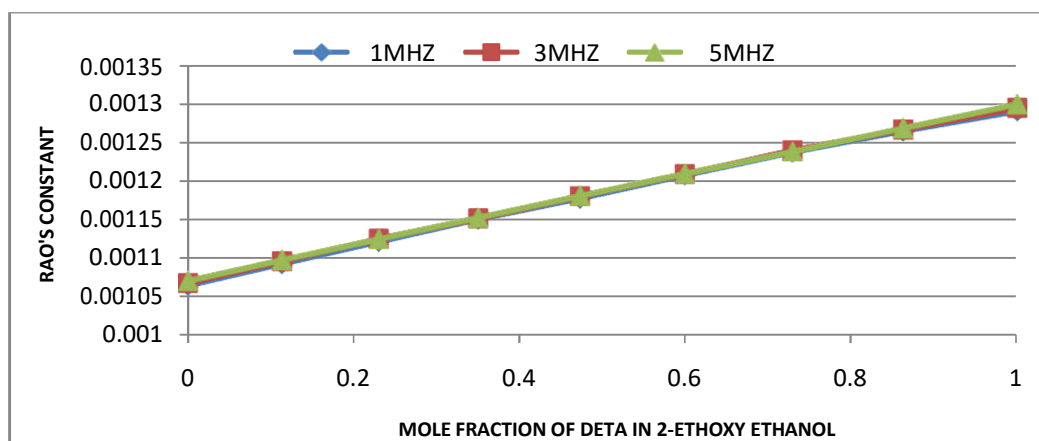


Fig 6 Graph between variation of Rao's constant & mole fraction of DETA in 2-Ethoxyethanol at constant temperature 296K and frequencies 1MHz, 3MHz & 5MHz

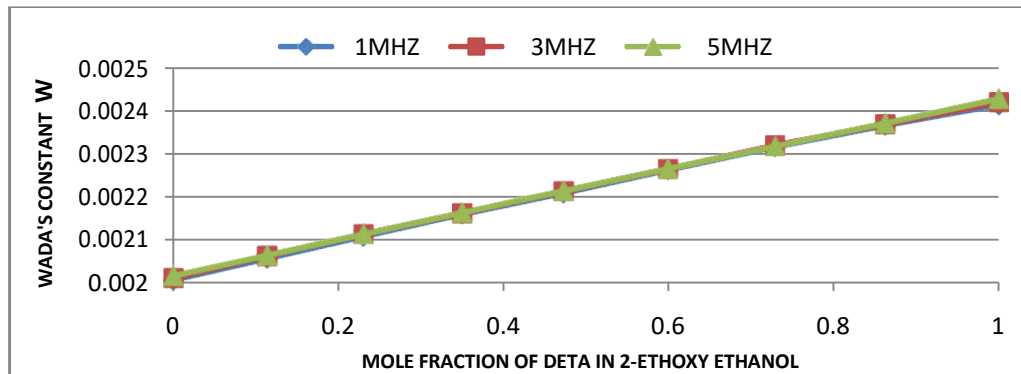


Fig 7 Graph between variation of Wada's constant & mole fraction of DETA in 2-Ethoxyethanol at constant temperature 296K and frequencies 1MHz, 3MHz & 5MHz

The variations in Rao's constant and Wada's constant with increase in mole fraction of DETA and with increase of ultrasonic frequencies are shown in figures 6 & 7 respectively. Perusal of figures 6 & 7 indicates that both Rao's & Wada's constant are increasing linearly with increase of concentration of DETA for all the three frequencies. These are slightly more pronounced at higher frequency. The increase in Rao's and Wada's constant with increase in concentration of DETA in 2-Ethoxyethanol is the evidence for presence of solute-solvent interaction between the constituents of the binary system [15].



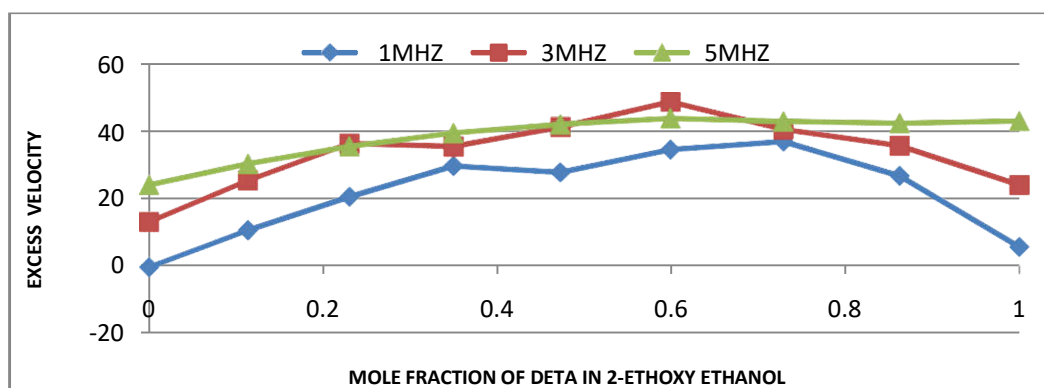


Fig 8 Graph between variation of excess ultrasonic velocity & mole fraction of DETA in 2-Ethoxyethanol at constant temperature 296K and frequencies 1MHz, 3MHz & 5MHz

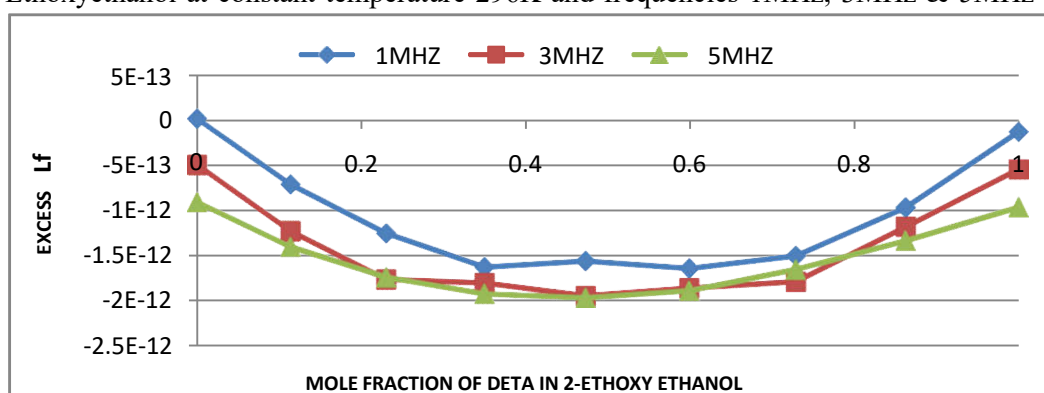


Fig 9 Graph between variation of excess intermolecular free length & mole fraction of DETA in 2-Ethoxyethanol at constant temperature 296K and frequencies 1MHz, 3MHz & 5MHz

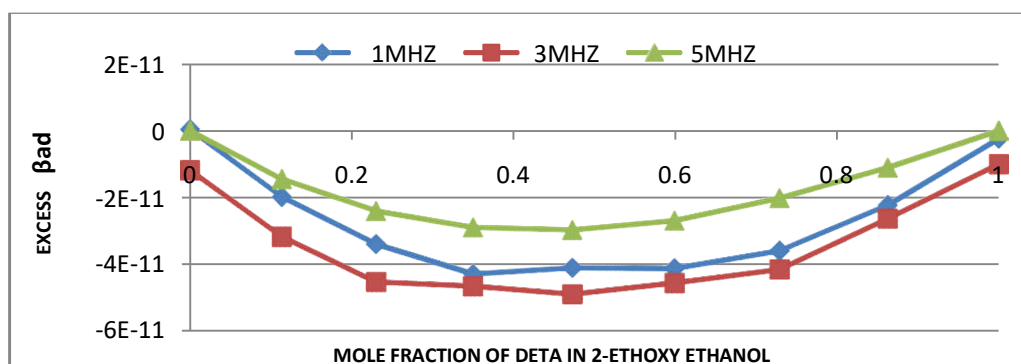


Fig 10 Graph between variation of excess adiabatic compressibility & mole fraction of DETA in 2-Ethoxyethanol at constant temperature 296K and frequencies 1MHz, 3MHz & 5MHz

The deviations in excess ultrasonic velocity, excess intermolecular free length and excess adiabatic compressibility with increase in mole fraction of DETA and with increase of ultrasonic frequencies are shown in figures 8, 9 & 10 respectively. Figure 8 shows positive deviation in excess ultrasonic velocity for all the three frequencies, with increase in concentration of DETA in the binary system. Figure 9 shows negative deviation in excess

intermolecular free length for all the three frequencies, with increase in concentration of DETA in the binary system and figure 10 shows negative deviation in excess adiabatic compressibility, for all the three frequencies, with increase in concentration of DETA in the binary system. This behavior of excess sound velocity, excess intermolecular free length and excess adiabatic compressibility indicates that there is strong molecular interaction between constituents of the binary mixture. This is due to closer packing inside the shell between the constituents of the binary mixture [16]. This change is observed at all the three ultrasonic frequencies. Thus the behavior of excess parameters supports strong molecular interaction between the constituents of the binary mixture of DETA and 2-Ethoxyethanol.

## CONCLUSION:

In this work we have studied the acoustic parameters Ultrasonic velocity ( $U$ ), Viscosity ( $\eta$ ), Density ( $\rho$ ), intermolecular free length ( $L_f$ ), adiabatic compressibility ( $\beta_{ad}$ ), Rao constant ( $R$ ), Wada's constant ( $W$ ), excess ultrasonic velocity ( $U^E$ ), excess intermolecular free length ( $L_f^E$ ) and excess adiabatic compressibility ( $\beta_{ad}^E$ ). Velocity, density, viscosity, Rao's constant and Wada's constant of this binary mixture show steady increase with increase in concentration of DETA whereas intermolecular free length and adiabatic compressibility show continuous decrease with increase in concentration of DETA in the mixture. Similarly excess ultrasonic velocity shows positive deviation whereas excess intermolecular free length and excess adiabatic compressibility show negative deviation with increase in concentration of DETA. The behavior of all the above parameters is in support of strong molecular interaction and forces of cohesion. Therefore, it may be concluded that there is existence of strong interaction between the constituents of this binary mixture at all the three ultrasonic frequencies at constant temperature.

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