

## ULTRASONIC STUDIES ON MOLECULAR INTERACTION IN TERNARY LIQUID MIXTURE OF DIMETHYL SULPHOXIDE AT DIFFERENT FREQUENCIES

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### ABSTRACT

The density ( $\rho$ ) and ultrasonic velocity ( $U$ ) for ternary mixture of dimethyl sulphoxide, benzoic acid and Ethyl Glycol at different frequencies (2MHZ, 4MHZ, 6MHZ, and 8MHZ) have been measured at 308K. Ultrasonic technique is used for studying the acoustic properties for a number of systems including liquids, low melting solids, dilute solutions of organic liquids and their mixtures. Ultrasonic studies in organic liquids and their binary mixtures have been of considerable research interest during the last few decades. 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 ( $RA$ ).

**KEYWORDS:** Molar volume, dimethyl sulphoxide, interaction parameter, ternary mixture.

### INTRODUCTION

Ultrasonic studies in organic liquids and their binary mixtures have been of considerable research interest during the last few decades. A liquid mixture is said to show ideal behavior if the variations in density and sound velocity etc., with mole fraction of the solute are linear. Ultrasonic velocity and adiabatic compressibility, which can be measured accurately, reflect the degree of deviation from ideal behavior. These deviations from the ideal behavior have

been widely used for the study of structural variations and molecular interactions in liquid mixtures.

The present investigation is related on thermodynamic properties of ternary liquid mixture of dimethylsulphoxide which a dipolar aprotic solvent with high is boiling point and good thermal and chemical stability. The study of dimethyl sulphoxide is important because of its utilization in industry and medicine. Dimethyl sulphoxide 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 elasthane fibres, pharmaceuticals, antibiotics and polyimide resins. Benzoic acid is a polar solvent used in rare metal extraction, antibiotics and other pharmaceuticals, cellulose and resin based coatings and adhesives. Ethylene Glycol 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 sulphoxide, benzoic acid and Ethylene Glycol 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.<sup>[5-11]</sup>

### 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  $\rho$ ) 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.

$$AE = A_{\text{exp}} - (X_1A_1 + X_2A_2 + X_3A_3) \quad (6)$$

Where  $X_1$ ,  $X_2$  and  $X_3$  are mole fractions of Dimethyl sulphoxide, benzoic acid and Ethylene Glycol respectively and  $A$  is any acoustic parameter.

Molar sound velocity ( $R$ ), molar compressibility ( $B$ ), Lennard Jones potential repulsive term exponent ( $n$ ), relative association ( $RA$ ) and interaction parameter ( $\chi$ ) 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  $\rho_0$  and  $U_0$  are density and ultrasonic velocity of Dimethyl sulphoxide respectively.

The ideal mixing velocity  $U_{\text{ideal}}$  is given by:

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

## EXPERIMENTAL METHODS

### 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 benzoic acid was kept fixed at 0.4. The mole fractions of Dimethyl sulphoxide and Ethylene glycol were varied from 0 to 0.6 to have the mixture of different compositions.

### 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  $w_1$ ,  $w_2$ ,  $\rho_1$  and  $\rho_2$  are mass of distilled water, mass of liquid mixture, density of distilled water and density of liquid mixture respectively.

### 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. 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.

### 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 5 and displayed graphically in Figures 1 to 9. Experimentally measured density ( $\rho$ ) 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  $\rho$  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.<sup>[11]</sup> Table 2 and Figure 2 show that ultrasonic velocity U in the ternary mixture increases with the increase in mole fraction of DMS. 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.

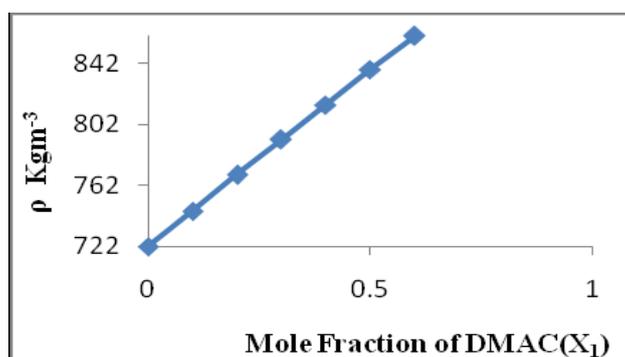
**Table 1: Values of density ( $\rho$ ) and ultrasonic velocity (U) of pure liquids.**

| Organic Liquids        | $\rho$ Kgm <sup>-3</sup> | U ms <sup>-1</sup> |      |      |      |
|------------------------|--------------------------|--------------------|------|------|------|
|                        |                          | 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  |

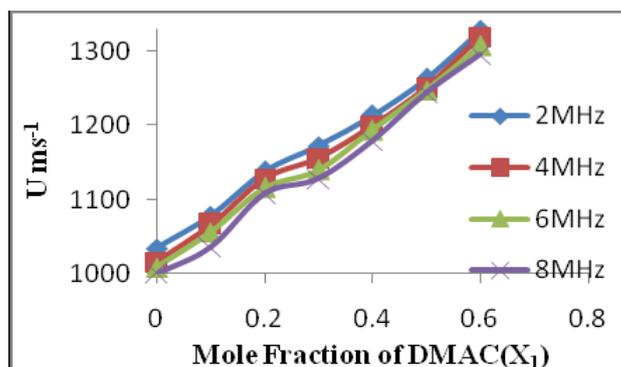
**Table 2: Values of density ( $\rho$ ) and ultrasonic velocity (U) for DMS + benzoic acid + Ethylene glycol.**

| Mole Fraction  |                | $\rho$ Kgm <sup>-3</sup> | U ms <sup>-1</sup> |      |      |      |
|----------------|----------------|--------------------------|--------------------|------|------|------|
| X <sub>1</sub> | X <sub>3</sub> |                          | 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 |

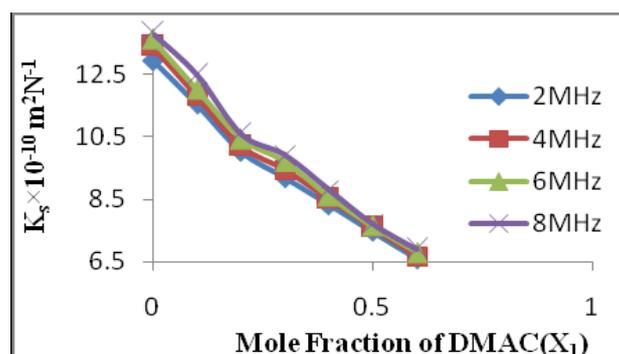
The experimental results are in good according to which the ultrasonic velocity increases with the decrease in intermolecular free length in the liquid mixture and vice versa. Therefore, intermolecular free length is one of the predominating factors for deciding the nature of variation in ultrasonic parameters in the liquid mixture. In the present study the decrease in intermolecular free length causes increase in ultrasonic velocity, decrease in adiabatic compressibility and increase in density and acoustic impedance as the concentration of DMS increases.



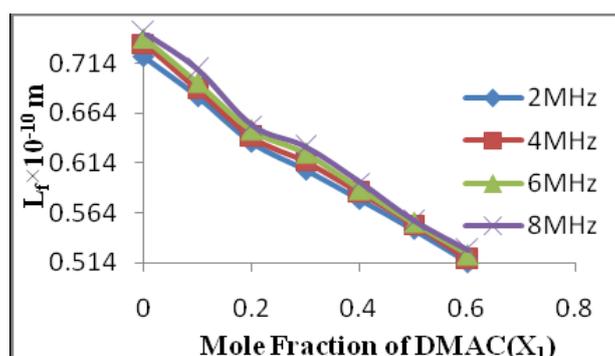
**Figure 1: Variation of  $\rho$  versus  $X_1$ .**



**Figure 2: Variation of U Versus  $X_1$ .**



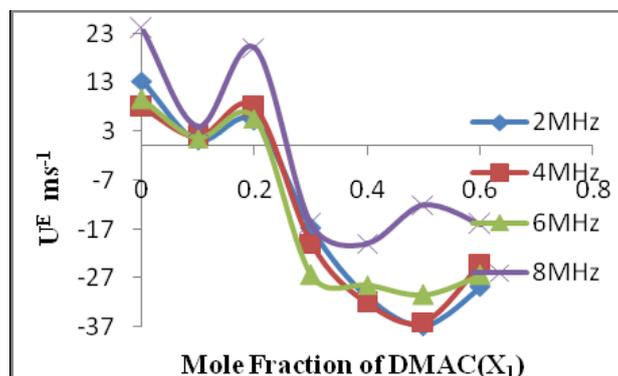
**Figure 3: Variation of  $K_s$  Versus  $X_1$ .**



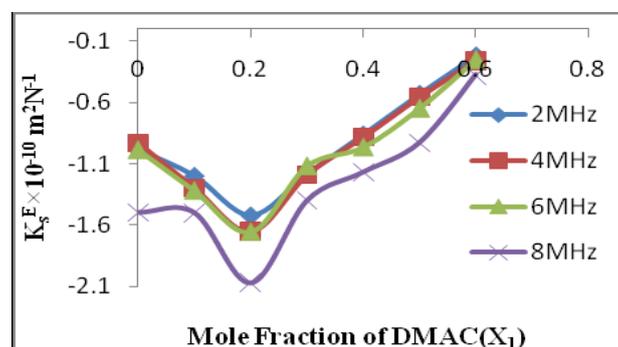
**Figure 4: Variation of  $L_f$  Versus  $X_1$ .**

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 7: Variation of UE Versus  $X_1$ .**



**Figure 8: Variation of  $K_s^E$  Versus  $X_1$ .**

Table 5 and Figure 7 show that the values of excess velocity UE are positive for the mole fraction of DMS between 0 to 0.2 and negative for the mole fraction of DMS between 0.3 to 0.6 for all frequencies. The positive values of UE indicate the presence strong molecular interactions at lower concentration of DMS. The negative values of UE 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 8 for the entire range of mole fraction of DMS 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.<sup>[15]</sup> 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 9 show that the values of  $L_f^E$  are negative for the entire range of mole fraction of DMS 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 (UE) the ternary mixture for DMS + benzoic acid+ Ethylene glycol.**

| 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 DMS between 0.1 to 0.6 for all frequencies and positive in the absence of DMS as shown in Table 7 and Figure 10. 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.

**Table 6: Excess values of adiabatic compressibility ( $K_s^E$ ) and free length ( $L_f^E$ ) for DMS + Benzoic acid+ Ethylene glycol.**

| 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  |

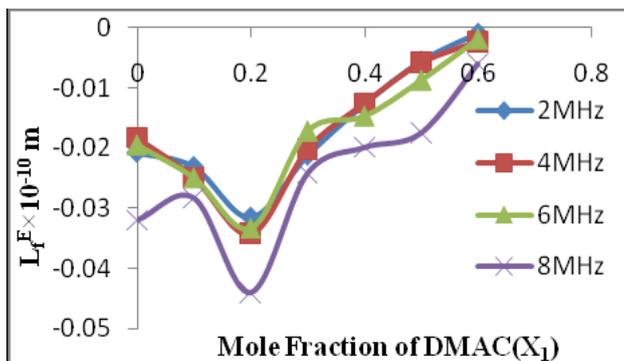


Figure 9: Variation of  $L_f^E$  Versus  $X_1$ .

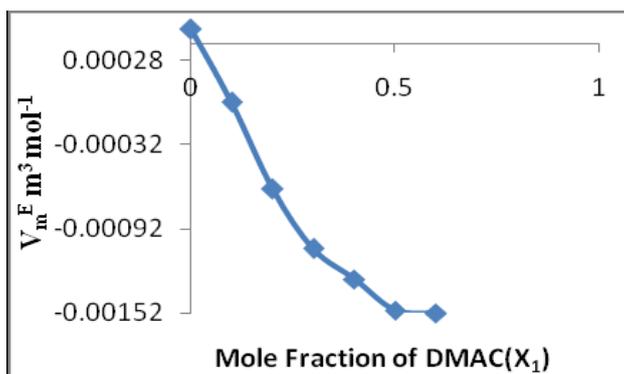


Figure 10: Variation of  $V_m^E$  Versus  $X_1$ .

Table 7: Excess Values of molar volume  $V_m^E$ , acoustic impedance (ZE) and available volume (VaE) for DMS + Benzoic acid + Ethylene glycol.

| Mole Fraction |       | $V_m^E$<br>$m^3 mol^{-1}$ | $Z^E \times 10^5 Kgm^2s^{-1}$ |         |         |         | $V_a^E m^3mol^{-1}$ |         |          |          |
|---------------|-------|---------------------------|-------------------------------|---------|---------|---------|---------------------|---------|----------|----------|
| $X_1$         | $X_3$ |                           | 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   |

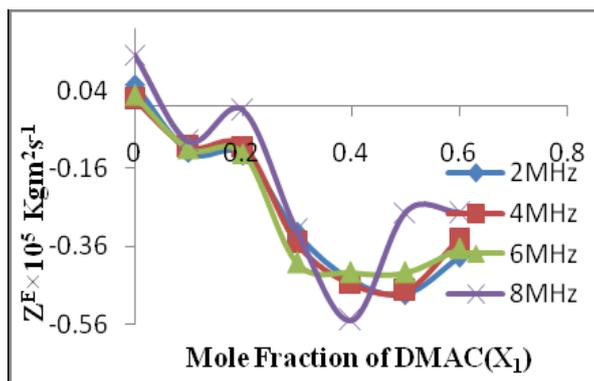
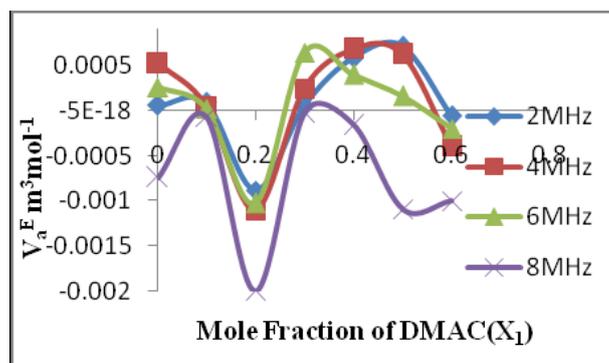


Figure 11: Variation of ZE Versus  $X_1$ .



**Figure 12: Variation of  $V_a^E$  Versus  $X_1$ .**

The values of excess acoustic impedance  $Z^E$  are negative for the mole fraction of DMS between 0.1 to 0.6 as shown in Table 7 and Figure 11 for all frequencies and positive in the absence of DMS 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 12 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.<sup>[16]</sup> The excess acoustic parameters are also changed with the increase in frequency in the ternary mixture.

## 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 DMS, Benzoic acid and Ethylene glycol. 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.

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