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Ultrasonic velocity, density, viscosity for the ternary mixture of (benzene + chloroform + cyclohexane) at different temperatures

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ABSTRACT

Ultrasonic velocity (U), density (ρ), viscosity (η) for the ternary mixture of benzene + chloroform + cyclohexane in the whole range of composition has been carried out at various temperatures 303.15, 308.15 and 313.15K. From the measured parameters U, ρ and η, some derived parameters β, L_f, V_f, π_i, Z, R,W with molar enthalpy (H_m) and apparent molar volume (ϕ_v) are also estimated. The decreasing trend of η, R,W, H_m, and ϕ_v with concentrations and other parameters are in increasing trend were observed. The trend of acoustical and physico-chemical parameters confirm the dynamics of molecules at higher temperature and the magnitude of intermolecular interactions among the constituents of the mixtures always reflects the nature of substance. The role of enthalpy and apparent molar volume has been used to determine their thermal response. The sign and magnitude of these properties are evident for the nature of interactions between component molecules.

1. Introduction

In recent years transport, acoustic and thermodynamic behavior in many binary, ternary liquid mixtures has been analyzed by several researchers more than 5 decades. The analysis have may relevance in the area of biochemical, pharmaceutical, geological natures in bio-fluids, oils, petroleum and etc, including both in experimental and theoretical approach (Grolier et al., 1974; Semeniur, and Wachnik, 1998; Vanathi et al., 2013a,b; Rathnam et al., 2012). There is some considerable interest to aware the intermolecular interaction in liquid mixtures. The main usage of organic mixtures have used for processing and further formulations of product. Organic liquid used for synthesis of organic compounds, for coupling and dispersion agents in pharmaceuticals dye and etc., Cyclohexane and its derivates are used for making pharmaceuticals, drugs, dye, pesticides and etc. The high electro negativity and larger dipole moment of polar molecules forming complex through H-bonds as a acceptors (Battino, 1971; Wang, 2006; Sundaram and Palaniappan, 2005; Kolska et al., 2011). The major advantages of inorganic liquids having some peculiar properties is attracted the attention to chemist, physicist and materials rescued. Find it is electrochemical application (Chno, 2005; Gardas and Coutinho, 2008).

Physicochemical properties of pure and mixtures of organic liquids are having great importance in the field of science and industrial engineering. In particular tension on the surface of film caused by some attraction (surface tension), density affects some important steps in the production process such as absorption on surface (adsorption) and separation from the mixtures (extraction). The need of viscosity can useful to detect various types of fluids mainly liquids on molecular and atomic (nano fluids)basis. Temperature dependence of viscous/sticky behavior of inorganic mixture were play on important role for characteristics the materials (Okotuno and VanderNoot, 2004; Singh et al., 2010).

The volumetric properties like free volume and internal pressure can classify the various intermolecular interactions between the different species exist in solution (Riddick et al., 1986; Vogel, 1989; Jain, 2006). The analysis towards the geometrical properties (topology) of components of solution that have been used (1) to know the state of components in pure as well as combined form of mixed state with existence of molecular interaction among them (1,2) further to find the physico-chemical behaviors of liquid mixtures (Mullainathan and Nithiyanantham, 2010; Vanathi et al., 2013a,b). The increasing/decreasing trends with linearity of parameters are helpful to decide the type, strength and magnitude of interaction in the liquid mixtures.

In this present work, we reported three basic measured parameters and some measured parameters such as adiabatic compressibility (β), free length (L_f), free volume (V_f) internal pressure (π_i) acoustical impedance (Z) Rao's constant (R) Wada's constant(W), apparent molar volume (ϕ_v)

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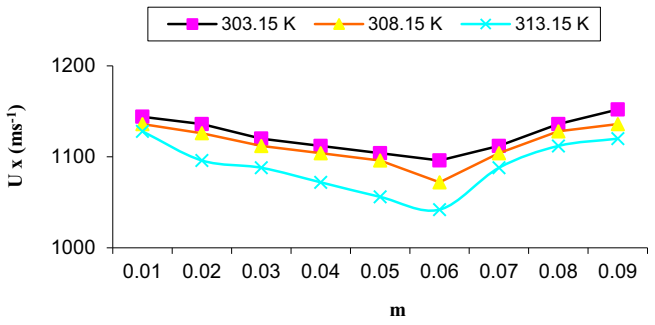

Fig. 1. Ultrasonic velocity (U) vs molality (m) of (BEN + CHF + CYH) at various temperatures.

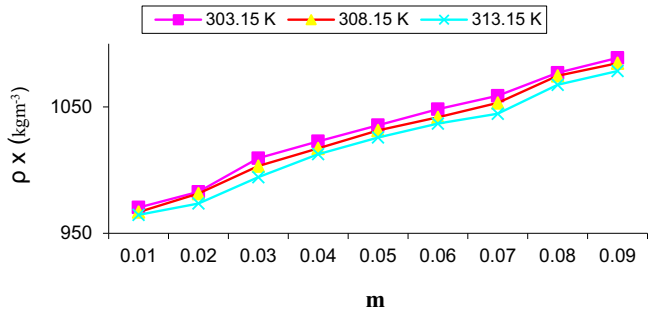

Fig. 2. Density (ρ) vs molality (m) of (BEN + CHF + CYH) at various temperatures.

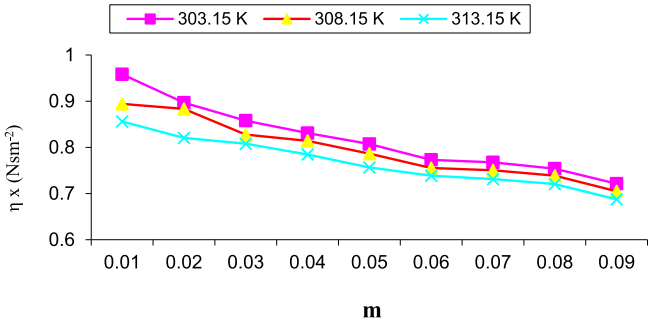

Fig. 3. Viscosity vs molality of (BEN + CHF + CYH) at various temperatures.

and molar enthalpy (H_m) of ternary mixtures at three different temperatures (Nithiyanantham and Palaniappan, 2016; Punitha et al., 2019).

2. Experimental

All the studied chemicals which were purchased from S.D Fine chemicals, Mumbai, India, with on purity of 99.7 % were used as such without further purification. In 100 ml of solutions of mixture contains (Benzene + Chloroform + Cyclohexane), among this component 2(chloroform) is same (fixed) concentrations. Component 2 (Benzene) gradually increased, Component 3 (cyclohexane) mutually decreased. The solutions were prepared for 0.01, 0.02, 0.03 upto 0.1 in steps of 0.01 M concentrations and weighing of samples through GR -202r, setup which are kept in air tight bottles to avoid evaporation and etc. The dialysis of the sample solutions were measured by using specific gravity bottles having bulk capacity of 10 cm^3. The graduates mark, and will be glassed by well-fitting glass cap. The uncertainties in density measurements were within ±0.1 Kg m^{-3}. The acoustical measurements were carried out through ultrasonic interferometer (Model M-8 IPS, Mittal Enterprises, India) operating at 3MHz with an uncertainty of ±0.1 ms^{-1}. This is the principle based on the reflection from the bottom and top of the cell. The reflection and the conversion of mechanical energy converted into electrical energy with help of piezo electric crystal (transducer) situated at the bottom of the ultrasonic cell. The viscosity measurement was done through the Oswald's viscometer. The time of flow noted (3-trials) with a digital stop watch and the precision intensity measurements was within the ±0.001 $mNsm^{-2}$. The temperature of the samples were maintained constant to an accuracy of ±0.1K using a thermostatically controlled digital water bath.

3. Result and discussion

The experimental values of density (ρ), ultrasonic velocity (U), and viscosity (η), of ternary mixtures of various concentrations at different temperatures are shown in Figs. 1, 2 and 3.

3.1. Ultrasonic velocity, density and viscosity

The Ultrasonic velocity, density and viscosity of solutions were determined through the factors.

$$U = n\lambda \tag{1}$$

Where 'n' and 'λ' are the frequency and wavelength of ultrasonic waves.

$$\rho = \frac{W}{W_s} X\ \rho_s \tag{2}$$

Where ρ, ρ_s are the density of solution and solvent, W, Ws are the weight of solution and solvent respectively.

$$\eta = \eta_s \frac{\rho t}{\rho_s t_s} \tag{3}$$

Where, η and η_s, are the viscosity of solution and solvent, and t and t_0 is the time of flow of solutions and solvent respectively (Vanathi et al., 2013a,b; Nithiyanantham and Palaniappan, 2016).

The value of ultrasonic velocity (U) is a decreasing trend up to 0.07m and further it is increases with concentration (Table 1). Further it is

Table 1
Measured values of ultrasonic velocity (U), density (ρ) and viscosity (η) of Benzene + Chloroform + Cyclohexane at different temperatures.

S.No	Molality (m)	U (ms^{-1})			ρ (Kgm^{-3})			η (Nsm^{-3})		
		303.15K	308.15K	313.15K	303.15 K	308.15K	313.15K	303.15K	308.15K	313.15K
1	0.01	1144.0	1136.1	1128.3	970.4	966.8	964.6	0.9582	0.8942	0.8560
2	0.02	1136.2	1126.5	1096.8	982.8	981.4	973.6	0.8968	0.8832	0.8204
3	0.03	1120.5	1112.6	1088.4	1009.4	1003.2	994.4	0.8579	0.8276	0.8079
4	0.04	1112.3	1104.4	1072.6	1022.8	1017.2	1012.6	0.8310	0.8142	0.7847
5	0.05	1104.2	1096.2	1056.0	1035.5	1031.4	1025.8	0.8072	0.7864	0.7565
6	0.06	1096.3	1072.3	1042.0	1048.2	1041.8	1036.8	0.7730	0.7553	0.7387
7	0.07	1112.0	1104.0	1088.1	1058.8	1053.2	1044.8	0.7676	0.7504	0.7312
8	0.08	1136.6	1128.0	1112.2	1077.0	1074.6	1067.5	0.7539	0.7387	0.7205
9	0.09	1152.1	1136.2	1120.4	1088.8	1084.6	1078.4	0.7213	0.7049	0.6874

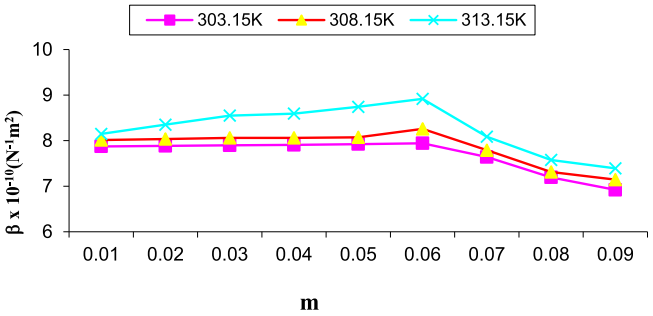

Fig. 4. Adiabatic compressibility vs molality of (BEN + CHF + CYH) at various temperatures.

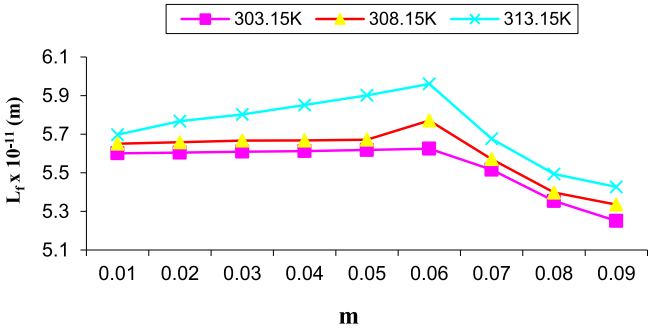

Fig. 5. Free length vs molality of (BEN + CHF + CYH) at various temperatures.

decrease with temperature is indicates the existence molecular interaction is shown in Fig. 1. The measurement of density and velocity of Benzene and Chloroform + Cyclohexane have been patterned were the temperature ranges from 303.15 K to 313.15 K are shown in Figs. 2 and 3 respectively. It can be observed that from the same Figs. 2 and 3 that density (ρ) of solutions increases with the concentration of solute (m) in the solution for all the system studied. The similar kind of behavior is found by the researcher at various temperatures (Gardas and Coutinho, 2008). Further the density of ternary mixture decreases slightly with respect to temperature for all the systems (Pal and Kumar, 2004; Dhondge et al., 2012).

From Fig. 3, the viscosity values decreases with both (i) concentration of solute and (ii) temperature. So, it is reverse trend with 'm' and temperature. It is peculiar type of observation, rather than negative trend with density values. This leads to the possibility of high stability and weak solute – solvent interaction (Rita Mehra and Poncholi Meenakshi, 2007; Santhosh and Krishna Bhat, 2011). When the temperature increased the values of (U), (ρ) and (η) were decreases, this trend reveals that at increasing temperature the molecular interaction between the molecules decreased and it shown low in higher temperatures (Kolhe and Bhosale, 2017).

3.2. Derived parameters

The parameters computed from the measured parameters said above, adiabatic compressibility, free length, free volume, internal pressure (Rita Mehra and Poncholi Meenakshi, 2007) and etc.

The adiabatic compressibility (β) is obtained from the following expression

$$\beta = \left({}^{1}/_{U^2\rho}\right) \tag{4}$$

And the related parameters, have an intermolecular free length (L_f) is

$$L_f = K_T \beta^{1/2} \tag{5}$$

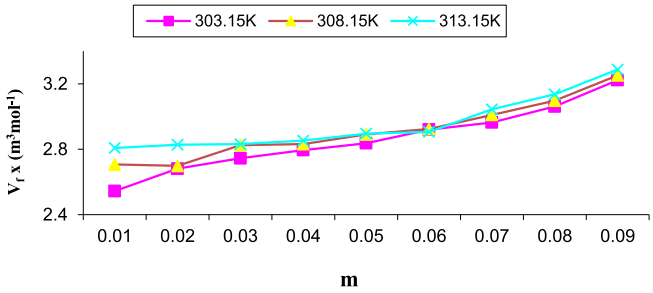

Fig. 6. Free volume vs molality of (BEN + CHF + CYH) at various temperatures.

Where K_T (2.131×10^{-6}) is Jacobson's constant referred as temperature dependent constant (Mullainathan and Nithiyantham, 2011).

The adiabatic compatibility (β) and intermolecular free length (L_f) are shown in Figs. 4 and 5. The values are gradually increase and upto 0.07m, after these are decrease with molarity. The additions of interacting molecules break up the molecular clustering of the other, then they releasing many dipoles for interaction. This may cause due to the electrostatic field energy exist in the mixture will affects through interaction and extend the cohesive energy and the structure changed. The temperature will enhance the structural changes by disturbing the bonding between the components of the mixtures, are also observed.

Both obtained values of (β) and (L_f) follow similar trend in the concentration in all the systems were investigated and there is no linearity observed for these values. This confirm the presence of specific interactions are evident among the solute and solvent molecules, the small value of β, because of large magnitude of solute–solvent interaction which make the system incompressible in the range of concentrations 0.07m–0.10m. Similar trend of least value of L_f of these concentration indicates the increase in the more attractive which leads the compactness of the system due to the components are much more close, and these closer molecular causes a strong molecular contraction (Ali et al., 2004; Karthikeyan and Palaniappan, 2005; Kannappan et al., 2008; Kumar et al., 2011). This is in accordance with the view that the ultrasonic velocities increases with decrease in free length (L_f) and vice versa (Eyring and Kincaid, 1938). It is negative in the free volume increases in the concentration. This indicates that there is a specific close packing due to increasing strength of interaction between the components of the mixtures.

$$V_f = \left(\frac{M_{eff}\,U}{\eta K}\right)^{3/2} \tag{6}$$

$$\pi_i = bRT\left(\frac{K\eta}{U}\right)^{1/2}\left(\frac{\rho^{2/3}}{M^{7/6}}\right) \tag{7}$$

[T- Experimental temperature, b-cubic paking factor, assumed to be 2 for

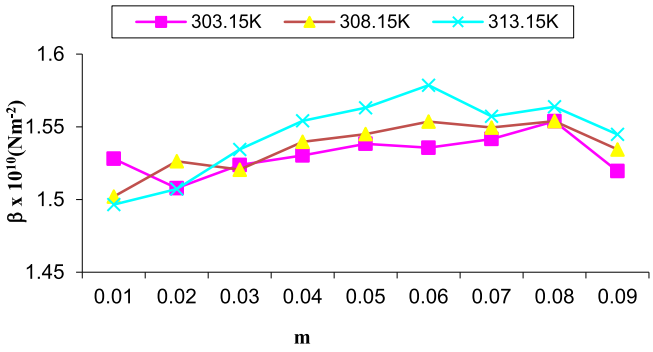

Fig. 7. Internal pressures molality of (BEN + CHF + CYH) at various temperatures.

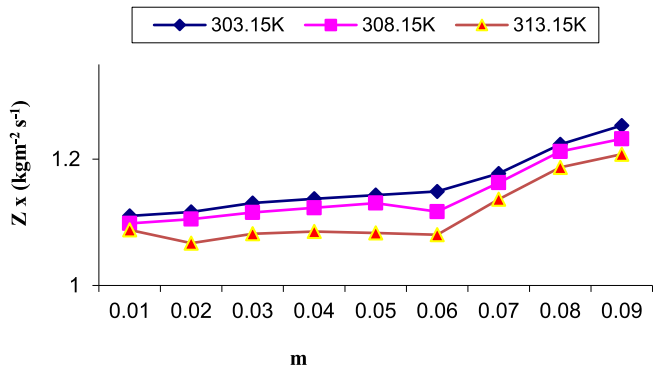

Fig. 8. Acoustic impedance vs molality of (BEN + CHF + CYH) at various temperatures.

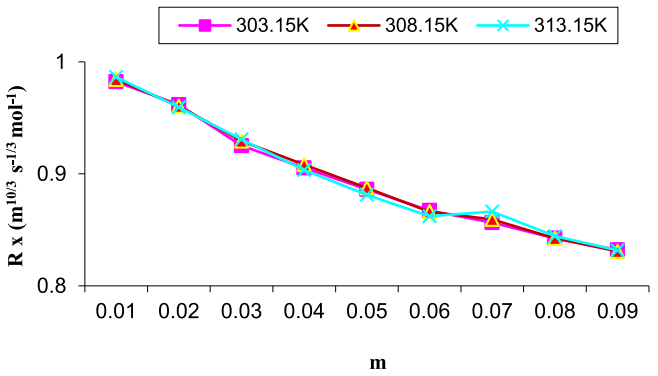

Fig. 9. Molar sound velocity vs molality of (BEN + CHF + CYH) at various temperatures.

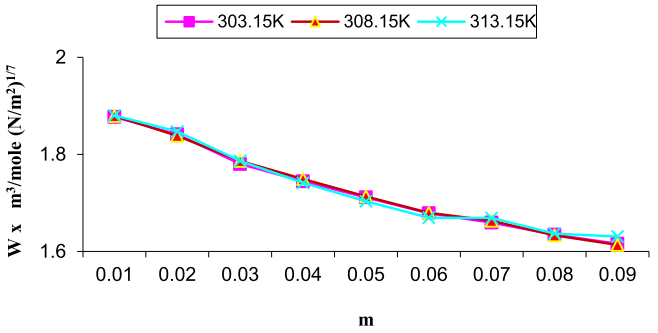

Fig. 10. Molar compressibility vs molality of (BEN + CHF + CYH) at various temperatures.

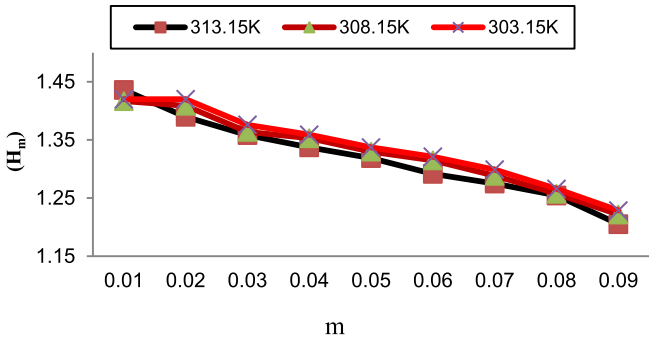

Fig. 11. Molar enthalpy vs molality of (BEN + CHF + CYH) at various temperatures.

all liquids, *K*- temperature independent constant for liquids (4.281 x 109) and *R*- Universal gas constant (Dhondge et al. 2012).

Free volume is average volume in which the centre of a molecule can move due to the repulsion surrounding molecules (Jain, 2006). The free volume between the components is increasing trend with cyclohexane is seen from Fig. 6. The increases in free space between the components offer independent nature and mutual forces of attractive and repulsive forces retain their original values (Kannappan et al. 2008; Eyring and Kincaid, 1938).

It is peculiar; an unexpected same increasing trend of their values for the free volume (V_f) is up to 0.09 m (Fig. 6), bur for internal pressure (π_i) is only up to 0.07m, after 0.07m reverse trend with free volume from 0.07m to 0.09m (Fig. 7). The observed increased trend in internal pressure indicates that the existence of strong interaction. The grown adhesions between the solute molecules leads to viscous force which in reflected by the internal pressure were observed (Ali et al., 2004; Kumar et al., 2011; Vanathi et al., 2013a,b). The existences of strong molecular interaction between the benzene with cyclohexane are evident besides the dipole-dipole interaction (Glory et al., 2009).

Acoustic impedance (Z) is the notable parameter to give information about the solution and it is responsible for the propagation of ultrasonic waves.

$$Z = U\rho \tag{8}$$

The acoustic impedance is obtained through the reaction (Jain, 2006; Kannappan et al., 2008), where U is the velocity of sound and ρ is the density of mixtures. From Fig. 8 there is an increasing trend with concentration are gradual increases up to 0.07m are further to increase linearly with molarity of cyclohexane (Syal et al. 1995, 1998). The increase in Z suggests that strong solute-solvent interactions through hydrogen bonding (Singh et al., 2010; Kharat, 2010). The decrease in nature with larger temperature meant for changes in structure and less propagation leads to decreasing/weakening the interactions (Singh and Kalsh, 1991; Kharat, 2010; Romero et al., 2017). And this is in agreement for the theoretical requirements as U and ρ both increase with increase of concentration. As increasing Z values indicating that unlike interactions are strong rather than compared to like interactions.

The molar sound function (R) and molar sound velocity (W) were computed using the following relations;

$$R = \left(m/\rho\right)U^{1/3} \tag{9}$$

$$W = \left(m/\rho\right)\beta^{-1/7} \tag{10}$$

It has been noticed that molar sound values (R) and molar sound velocity (W) are independent of sound and concentration is indicates that there is a weak association between the molecules (Singh and Kalsh, 1991). In the present investigation both R and W are similar decreasing trend is observed from Figs. 9 and 10. But in the present case it is dependent with temperature (Romero et al., 2017). The non-linear variation in R and W values with molality suggested the presence of complex formation in the mixture (Vanathi et al., 2013a,b). And R and W dependent with temperature is confirming the interaction between the benzene and cyclohexane molecules.

The molar enthalpy (H_m) the apparent molar values (ϕ_v) are calculated using the following expression.

$$H_m = \pi_i V_m \tag{11}$$

$$\varphi_v = \frac{1000}{m\rho_o}(\rho_o - \rho) + \left(\frac{m}{\rho_o}\right) \tag{12}$$

From Fig. 11, the decreasing value of molar enthalpy indicates the presence of weak interactions among the solute-solvent molecules and strong interaction among the solute-solute molecules (Singh and Kalsh,

Table 2
Apparent molar volume (ϕ_v) vs molality (m) of (BEN + CHF + CYH) at various temperatures.

Molality (m)	Apparent molar volume (ϕ_v) x 10^6 (m^3 mol^{-1})		
	303.15 K	308.15 K	313.15 K
0.00	396.992	392.714	383.248
0.01	194.137	191.771	188.747
0.02	123.906	123.283	121.398
0.03	90.844	90.264	88.139
0.04	71.094	70.427	68.824
0.05	57.928	57.601	56.181
0.07	48.710	48.349	47.443
0.08	41.205	40.625	39.682
0.09	35.811	35.413	34.498

1991; Kalyanasundaram et al., 1997; Dhondge et al., 2012; Pal and Kumar 2004). The variation in (ϕ_v) values of solute against molarity of solution at various temperatures is shown in Table 2 and it can be observed that (ϕ_v) is a linear function within the concentration range standard in the present work but decreasing trend. This also represents the magnitude of ion-solvent interaction. Further, it is observed that (ϕ_v) decreases with temperature for this system. This suggest that the at least possibility of solute – solvent and ion/ion interaction (Kalyanasundaram et al., 1997).

4. Conclusion

In summary, the ultrasonic velocity, density and viscosity measurements on ternary organic mixtures at various temperatures are carried out. The strong molecular interactions are confirmed. The trends and deviations in derived parameters were confirmed further the existence of strong interactions and dipole-dipole interaction. The nonlinearity in β and L_f concluded the specific interaction between the solute and solvent molecules. At lower values the strong molecular interaction through *H*-bonding resulted from *Z* values. But in the limit after 0.7m the reverse case is observed and it is confirmed that low value of molar enthalpy shows the weak interaction among the solute-solvent interaction. The non-linear relation between the *R* and *W* suggested the presence of complex formation in the mixture.

Declarations

Author contribution statement

V. Vanathi, S. Mullainathan, S. Nithiyanatham, V. Ramasamy, L. Palaniappan: Conceived and designed the experiments; Performed the experiments; Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data; Wrote the paper.

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Competing interest statement

The authors declare no conflict of interest.

Additional information

No additional information is available for this paper.

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