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Density, Ultrasonic Velocity, Isentropic Compressibility, Molar Volumes and Related Excess Parameters Studies on Ethyl Acetate with 1-Ethanol at 303K, 308K, And 313K

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Abstract

A binary liquid mixture that consists of ethyl acetate and 1-ethanol has been prepared at various concentrations by the mole fraction method. The ultrasonic velocity and density have been determined at 303K, 308K and 313K. From the experimental data, the excess isentropic compressibility, excess molar volumes, excess internal pressures, and excess molar enthalpy have been computed. The variations were observed as polynomial and fitted to the Redlich-Kister polynomial functions. By using this function, adjustable parameters and the standard deviations have been calculated. The experimental and theoretical data reveal that the existence of the intermolecular interactions between the selected liquid system. The partial molar compressibility's and partial molar volume also calculated at infinite dilution of the system. In general, the intermolecular forces have tended to the variations in the magnitude and sign of the excess parameters. The excess molar volume (V_m^E), excess isentropic compressibility ($\Delta k_s{}^E$), excess internal pressure (π_i^E) and the enthalpy (H_m^E) show the negative magnitude at the entire range of concentrations and temperatures. The significant variations of these parameters with the mole fraction of ethyl acetate have been analysed. Furthermore, the strength of the intermolecular interactions decreased with increasing the experimental temperatures as 303K > 308K >313K.

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Introduction

The intermolecular forces acting between the molecules take a vital role in the interactions between the functional groups present in the binary as well as the ternary liquid mixtures. Due to the variations of these forces and the temperature of the solutions, thermo dynamical properties of the liquid system are also significantly changed. The study of

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the changes in the ideal and excess parameters at various temperatures in the entire concentrations leads to provoke the nature of intermolecular interactions, molecular structure, and intermolecular forces acting in the liquid systems.[1-3]

In recent years, determination of various physico chemical properties such that molar volume, density, ultrasonic velocity of the pure liquids and the binary systems are used as one of the reliable methods to investigate the strength and nature of the bonding formations among the molecules.[4-6] Moreover, the volumetric properties depend upon the temperature of the systems have reported as a functional indicator of the presence of the intermolecular interactions.[7,8] The excess internal pressure also considerable changes with the interactions, recent reports of various liquid mixtures[9-11] furnish the impact of the excess internal pressure insight of the intermolecular forces acting among the molecules as a function of temperature of the system.

Ethyl acetate is one of the ester group compounds. It acts as a solvent in the column chromatography. Furthermore, ethyl acetate acts as a building block in the synthesis process of bulk materials having spectacular applications in the pharmaceutical, polymers, fragrances. Alcohols are self-associated organic compounds, have widely used in chemical and pharmaceutical and applied research.[12] The 1-ethanol behaves as both H-bond donor and acceptor. The ethyl acetate acts as H- bond acceptor. The prospect of hydrogen bonding between the hydroxyl group of 1-ethanol and the carbonyl group of the ethyl acetate gives the significant importance of the chosen liquid system. The present work is an attempt to study the excess molar volume (V_m^E), excess isentropic compressibility (Δk_s^E), excess internal pressure (π_i^E), excess molar enthalpy(HE), partial molar volume (Vm_1, Vm_2), and partial molar compressibility's (k_1 , k_2) and their excess values (Vm_1^E, Vm_2^E, β_1^E, β_2^E) of ethyl acetate and 1- ethanol over the entire composition at 303K, 308K, and 313K.

Materials and Methods

The various concentrations of ethyl acetate and 1-ethanol solutions were prepared by the mole fraction method. An electronic mass balance (OHAUS- AR 2104) with accuracy of ± 1x 10^{-4} g was used to measure the masses of the solvents. Air tight, sample bottles were used to avoid the evaporation and avoid the atmospheric moisture. The Analytical Reagent (AR grade) chemicals which are used in the study were obtained from Ranboxy Fine Chemicals Pvt Ltd (India). These chemicals were purified by following the standard procedure prescribed by Perrin and Armarego.[13] A pycnometer with a bulb volume of 10 ml and capillary diameter 0.1cm was used to determine the densities of the liquid mixtures and calibrated with the double distilled water. A constant temperature bath, INSREF-India, modelIRI-016C was used to maintain the experimental temperature of the accuracy ±0.1K. The concurrent values of the density were noted with the precision ± 0.001 kgm^{-3}. A single frequency (2MHz) ultrasonic interferometer with the uncertainty ±1 ms^{-1} was used to gauge the ultrasonic velocity of the pure and mixtures. It was calibrated with the double distilled water and carbon tetrachloride.

Theory

The various physicochemical parameters were determined by the standard relations[14-17] as listed below,

Excess molar volume = $\left(\frac{x_1M_1+x_2M_2}{\rho_{mix}}\right)-\left(\frac{x_1M_1+x_2M_2}{\rho_1+\rho_2}\right)$ ...(1)

Here x_1 and x_2 are mole fractions; M_1 and M_2 are molar masses; ρ_1 and ρ_2 are densities of ethyl acetate and 1-ethanol respectively. ρ is the density of liquid mixtures.

Isentropic compressibility of the solution $k_s = \frac{1}{u^2\rho}$ ...(2)

Here u is the ultrasonic velocity.

Excess isentropic compressibility = $\Delta k_s^E = k_s - k_s^{id}$ ...(3)

Here, k_s^{id} is the ideal value of the isentropic compressibility

Excess internal pressure, $\pi_i^E = \pi_{i.mix} - \sum_{i=1}^{2} x_i\pi_i$ ...(4)

Here, x_i is the mole fraction of the components (In this study, x_1 denotes the mole fraction of ethyl acetate and x_2 represents the mole fraction of 1-ethanol).

Excess enthalpy $H_m^E = \sum_{i=1}^{2} x_i \pi_i V_i - \pi_{mix} V_{mix}$ ...(5)

Here x_i is the internal pressure of the liquid, V_i is the volume of the liquid, π_{mix}, V_{mix} are the internal pressure and volume of the mixture.

Molar isentropic compressibility $K_s = K_s V_s$ ...(6)

Excess molar isentropic compressibility

$$K_s^E = K_s - \sum_{i=1}^{2} x_i K_{s,i} \quad ...(7)$$

The excess parameters were fitted with Redlich-Kister polynomial as the following relations,

$$Y^E = x_1 x_2 \sum_{i=1}^{5} a_i (1-2x)^{i-1} \quad ...(8)$$

The a_i represents the coefficients of the polynomial.

$$\sigma\left(Y^E\right) = \left[\frac{\sum\left(Y_{exp} - Y_{cal}\right)}{n-p}\right]^{1/2} \quad ...(9)$$

Here Y_{exp} is the experimental data and Y_{cal} is the theoretical data. The n denotes the total number of experimental values and p is the number of coefficients.

The average partial molar volume of ethyl acetate ($\bar{V}_{m,1}$) and 1-ethanol($\bar{V}_{m,2}$) are given as below, 1 denotes the first component ethyl acetate and 2 represents the second component 1-ethanol,

$$\bar{V}_{m,1} = V_m^E + V_1^* + x_2 \left(\frac{\partial V_m^E}{\partial x_1}\right)_{T,p} \quad ...(10)$$

$$\bar{V}_{m,2} = V_m^E + V_2^* - x_1 \left(\frac{\partial V_m^E}{\partial x_1}\right)_{T,p} \quad ...(11)$$

The average partial molar isentropic compressibility of the selected components at the constant temperature (T) and atmospheric pressure are given in equ (12) and equ (13) respectively

$$\bar{K}_{m,1}{}^0 = K_s^E + K_{s,1} + x_2 \left(\frac{\partial K_s^E}{\partial x_1}\right)_{T,p} \quad ...(12)$$

$$\bar{K}_{m,2}{}^0 = K_s^E + K_{s,2} - x_1 \left(\frac{\partial K_s^E}{\partial x_2}\right)_{T,p} \quad ...(13)$$

The molar volume at the infinite dilution can be derived from the equ (14) and equ (15) for the ethyl acetate and 1- ethanol respectively.

$$V^{\infty}{}_{m,1} = V_1^* + x_2^2 \sum_{i=0}^{i=n} A_i (1-2x_1)^i - 2x_1 x_2^2 \sum_{i=1}^{i=n} A_i (1-2x_1)^{i-1} \quad ...(14)$$

$$V_{m,2}^{\infty} = V_2^* + x_1^2 \sum_{i=0}^{i=n} A_i (1-2x_1)^i + 2x_1^2\, x_2 \sum_{i=1}^{i=n} A_i (1-2x_1)^{i-1} \quad ...(15)$$

The excess molar volume, and excess isentropic compressibility at the infinite dilutions are given as,

$$V_{m,1}^{\infty\, E} = \bar{V}_{m,1} - V_1^* \quad ...(16)$$

$$V_{m,2}^{\infty\, E} = \bar{V}_{m,2} - V_2^* \quad ...(17)$$

$$K_{m,1}^{\infty\, E} = \bar{K}_{m,1} - K_1^* \quad ...(18)$$

$$K_{m,2}^{\infty\, E} = \bar{K}_{m,2} - K_2^* \quad ...(19)$$

Results and Discussions

The experimental observations of density (ρ), ultrasonic velocity (U), and the calculated the isentropic compressibility of the entire concentrations of ethyl acetate and 1-ethanol are listed in Table1. In this study, the density (ρ) of the liquid mixtures increased with the ethyl acetate concentrations. Moreover, densities decreasing with increase the experimental temperature. This trend suggested the dissociation of the dipoles in the liquid system.

In general, the ultrasonic velocity varies with the medium and intermolecular forces acting between the solute- solvent systems. Furthermore, the increasing cluster of molecules with the increasing mole fraction of the ethyl acetate leads to the increasing ultrasonic velocity (U) linearly in the liquid mixtures. Since, the increasing temperature rupture the molecular forces, the ultrasonic velocities, decreased with the increasing the entire temperature range. Obviously, the isentropic compressibility inversely related with the density and ultrasonic velocity of the liquid system.[18-20] Thus, the isentropic compressibility values are increasing with the temperature from 303K to 313K.

The molar volume, partial molar volume of the pure liquids and the partial excess molar volume at the infinite solutions with their excess values are listed in Table 2.

Table 1: Density (ρ), Ultrasonic velocity (U), and isentropic compressibility (ks) of ethyl acetate and 1-ethanol at 303K, 308K, and 313K

X_1	ρ (kgm^{-3})	U (ms^{-1})	ks 10^{-10} (m^2N^{-1})	ρ (kgm^{-3})	U (ms^{-1})	ks 10^{-10} (m^2N^{-1})	ρ (kgm^{-3})	U (ms^{-1})	ks 10^{-10} (m^2N^{-1})
	303 K			**308K**			**313K**		
0.0000	784	1123	10.1140	779	1115	10.3255	774	1107	10.5430
0.1012	795	1124	9.9564	790	1116	10.1635	786	1109	10.3446
0.2026	806	1125	9.8030	800	1118	10.0006	795	1111	10.1907
0.3028	817	1126	9.6539	811	1119	9.8473	804	1112	10.0585
0.4035	828	1127	9.5087	822	1120	9.6982	814	1114	9.8993
0.5034	839	1128	9.3674	832	1122	9.5475	825	1116	9.7324
0.6037	850	1128	9.2461	843	1123	9.4062	833	1118	9.6044
0.7029	861	1129	9.1119	854	1124	9.2685	844	1120	9.4454
0.8020	872	1130	8.9810	864	1125	9.1450	852	1121	9.3401
0.9083	884	1131	8.8435	875	1127	8.9980	864	1123	9.1776
1.0000	894	1132	8.7291	885	1128	8.8805	872	1125	9.0611

Table 2: The molar volume, partial molar volume, excess partial molar volume at infinite dilution of ethyl acetate and 1-ethanol at 303K, 308K and 313K

Temperature	V_1^* $(m^3\ mol^{-1})$	$\bar{V}_{m,1}$ $(m^3\ mol^{-1})$	$V_{m,1}^{\infty\ E}$ $(m^3\ mol^{-1})$	V_2^* $(m^3\ mol^{-1})$	$\bar{V}_{m,2}$ $(m^3 mol^{-1})$	$V_{m,2}^{\infty\ E}$ $(m^3\ mol^{-1})$
303K	100.88	99.75	-1.13	93.95	92.61	-1.34
308K	103.59	101.95	-1.64	96.8	93.59	-3.21
313K	105.45	103.35	-2.1	97.7	94.53	-3.17

In this present work, the partial molar volume at the infinite dilutions ($\bar{V}_{m,1}, \bar{V}_{m,2}$) are relatively smaller than that of the molar volume of the pure liquid components ($V_1^* . V_2^*$) respectively. This result suggested that the pure component's molar volume is the resultant of the actual molar volume and free molar volume that arises due to the formation of self association and the intramolcular forces acting within the molecules.[21,22] Similar trends observed in the molar isentropic compressibility and the partial molar isentropic compressibility's and their excess values are obtained at the entire temperature ranges as specified in Table3.

Table 3: The molar isentropic compressibility, partial compressibility, excess partial molar compressibility at infinite dilution of ethyl acetate and 1-ethanol at 303K, 308K and 313K

Temperature	$K_{s,1}$ 10^{-10} (m^2N^{-1})	$\bar{K}_{m,1}^{0}$ 10^{-10} (m^2N^{-1})	$K_{m,1}^{\infty\ E}$ 10^{-10} (m^2N^{-1})	$K_{s,2}$ 10^{-10} (m^2N^{-1})	$\bar{K}_{m,2}^{0}$ 10^{-10} (m^2N^{-1})	$\overline{K_{m,2}^{\infty\ E}\ 10^{-10}}$ (m^2N^{-1})
303K	10.11	10.15	-0.04	8.73	8.76	-0.03
308K	10.32	10.39	-0.07	8.88	8.94	-0.06
313K	10.54	10.57	-0.03	9.06	9.17	-0.11

The Redlich-Kister[23] polynomial coefficients (a_1, a_2, a_3, a_4, a_5) are calculated along with the standard deviations $\sigma(Y^E)$ of the excess values of the selected parameters are as shown in Table 4.

Table 4: Redlich-Kister polynomial coefficients with the standard deviations of the excess parameters ($V_m^E, \Delta k_s^E, \pi_i^E, H_m^E$) of ethylacetate and 1-ethanol at 303K, 308K , and 313K

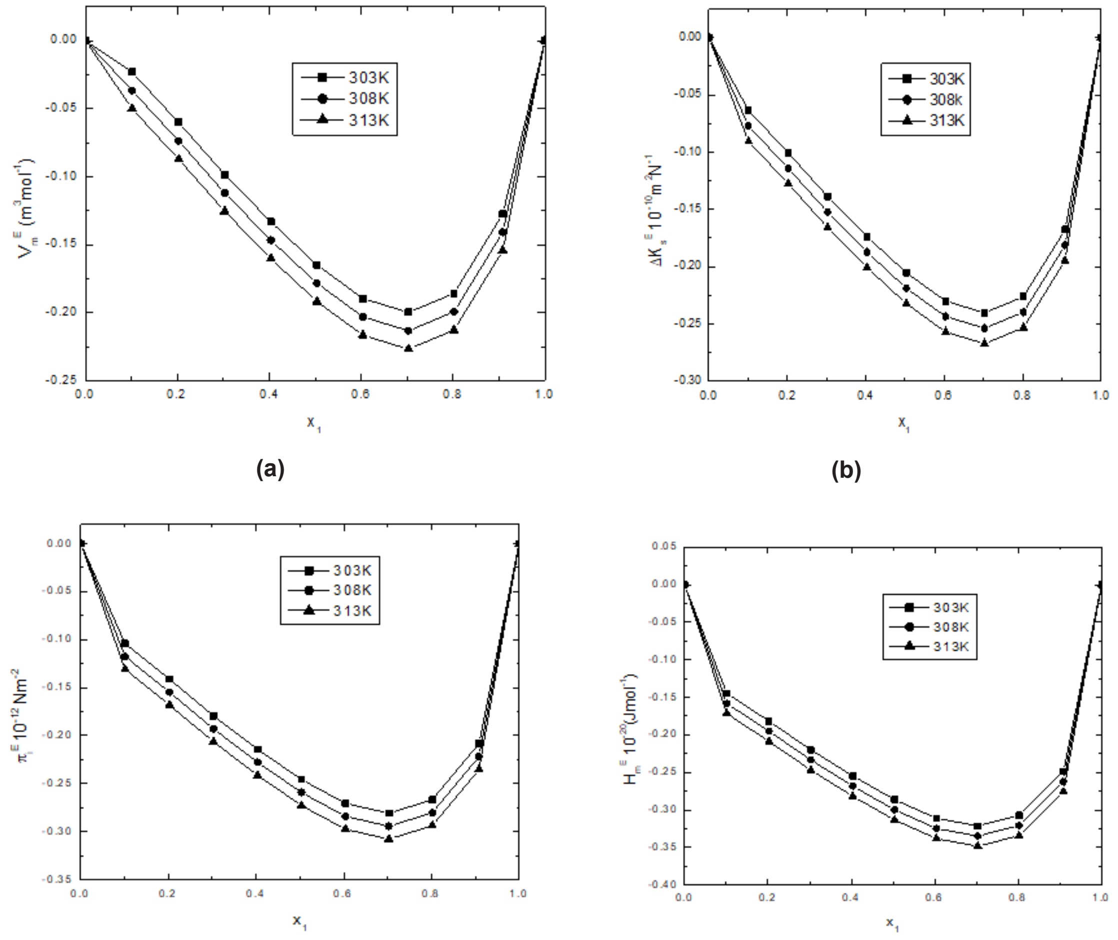
Temperature (K)	Parameters	a_1	a_2	a_3	a_4	a_5	$\sigma(Y^E)$
303K	V_m^E (m^3mol^{-1})	-0.471	-0.311	-0.231	-0.451	-0.095	0.033
	Δk_s^E 10^{-10} (m^2N^{-1})	-0.341	-0.329	-0.113	-0.321	-0.023	0.025
	π_i^E 10^{-12} Nm^{-2}	-0.341	-0.385	-0.120	-0.321	-0.016	0.032
	H_m^E 10^{-20}($J\ mol^{-1}$)	-1.027	-0.464	0.163	-1.007	-0.299	0.045
308K	V_m^E (m^3mol^{-1})	-0.789	-0.160	0.185	-0.769	-0.321	0.037
	Δk_s^E 10^{-10} (m^2N^{-1})	-1.187	-0.503	0.291	-1.167	-0.427	0.046
	π_i^E 10^{-12} Nm^{-2}	-0.272	-0.642	0.525	-0.252	-0.661	0.034
	H_m^E 10^{-20}($J\ mol^{-1}$)	-0.326	-1.022	0.956	-0.306	-1.092	0.040
313K	V_m^E (m^3mol^{-1})	-0.358	-1.480	1.380	-0.338	-1.516	0.048
	Δk_s^E 10^{-10} (m^2N^{-1})	-0.110	-0.128	-0.150	-0.090	-0.014	0.031
	π_i^E 10^{-12} Nm^{-2}	-0.044	-0.106	-0.237	-0.024	-0.101	0.035
	H_m^E 10^{-20}($J\ mol^{-1}$)	-0.010	-0.051	-0.346	-0.030	-0.210	0.030

All the coefficients are found in negative magnitude and the standard deviations are in negligible values are obtained in the entire temperature range. This data validated the precision of the measurements and theoretical calculations.

The deviations of the excess molar volume (V_m^E), excess isentropic compressibility (Δk_s^E), excess internal pressure (π_i^E) and the enthalpy (H_m^E) at 303K, 308K, and 313K are plotted against the mole fraction of ethyl acetate as displayed in Fig 1(a-d) respectively.

The deviations of the excess parameters are useful to interpret the intermolecular interactions among the unlike molecules. In general, the positive values of the excess values revealed that the dispersion forces. More over, the negative values of the excess parameters suggested that the dipole-dipole, charge transfer interaction, and hydrogen bonding formation between the liquid system.[24]

The Fig 1(a-d) signifies that all the excess parameters show the negative sign, and the magnitudes vary with the entire concentrations and temperature range. At a critical mole fraction (x_1~0.7), the negative excess values increased with the increasing the concentrations of ethyl acetate. The change in the variations suggested the 1:1 complex formation of the ethyl acetate and 1-ethanol at the entire range of temperatures. The changes in the experimental and the excess parameters confirmed that (i) Breakdown of the self association takes place with increasing 1-ethanol concentrations, (ii) While increasing the temperature, dispersive force increased between the molecules, (iii) At a critical mole fraction (x_1~0.7), the negative excess deviation decreased with increasing the concentrations of ethyl acetate. This trend reveals the weakened hydrogen bonding formation between(C=O….H-O) functional group present in the liquid mixtures. The negative excess parameters were decreasing with increase the experimental temperature. This result suggested that the strength of intermolecular interactions decreased with the increase the temperature and found in the order303K>308K>313K.



(a) (b)

(c) (d)

Fig 1: (a) Excess molar volume, (b) Excess isentropic compressibility, (c) Excess internal pressure, (d) Excess enthalpy against the mole fraction of ethyl acetate at 303K, 308K, and 313K

Conclusion

The ethyl acetate and 1-ethanol mixtures were prepared at various concentrations at 303K,308K, and 313K. Physico chemical parameters such that the density, ultrasonic velocity of the selected components were measured. Using the experimental results, the excess isentropic compressibility, excess molar volumes, excess internal pressures, and excess molar enthalpy were calculated. More over the excess parameters also evaluated. The excess parameters were fitted with the Redlich-Kister polynomial functions. The hydrogen bonding between the functional groups was stronger at the 1-ethanol rich concentrations and weaker with ethyl acetate molecules. The strength of the intermolecular interactions were decreasing with the temperature as 303K>308K>313K.

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Conflict of Interest

The authors do not have any conflict of interest.

References

1. T. Savitha Jyostna, B. Satheesh, D. Sreenu, G. Ramesh and E. Jayanthi Rani, The study of thermo-physical properties of binary liquid mixtures of isoamyl alcohol with amines at 298.15–308.15K. Phys. *Chem. Liq,* 58, 349-363 (2020).
2. L. Palaniappan and S.Nithyanantham, Molecular interactions from the experimental and validation with estimated theoretical sound velocity. *Chemistry Africa*. 3, 277-285 (2020).
3. D. Zhao, Y. Zhuang, C. Fan, F. Yang, Y. Chen and X. Zhang, Surface tension of binary mixtures composed of N, N-dimethylcyclohexylamine and alcohols at different temperatures. *J.Chem. Thermodyn.*143, 106041(2020).
4. A. S. Mora, R. Tayouo, B. Boutevin, G. David and S. Caillol, A perspective approach on the amine reactivity and the hydrogen bonds effect on epoxy-amine systems. *Eur. Polym. J.* 123, 109460 (2020) .
5. M.V. Alexander, G. Meenakshi, Investigation of ultrasonic speeds of sound and excess parameters in binary liquid mixtures of N-alkanes with octan-2-ol using different empirical theories at 298.15 K. *J Math Chem* 56, 2963–2981 (2018).
6. S. Vani Latha, G. Little Flower, K. Rayapa Reddy, C.V.N Rao and A. Ratnakar, Densities, ultrasonic velocities, excess properties and IR spectra of binary liquid mixtures of organic esters (ethyl lactate, some organic carbonates). *J. Sol. Chem*. 46, 305–330 (2017).
7. A. Shakila, S. Ravikumar, V. Pandiyan and R. Gaba, Thermodynamic properties of binary liquid mixtures containing aromatic alcohol and aliphatic amines at different temperatures. *J. Mol. Liq.* 2851, 279-287 (2019).
8. I. N. Mezhevoi and V. G. Badelin, Thermochemical analysis of intermolecular interaction of aliphatic amino acids with propanediol-1,3 in aqueous media. *Russ J. Gen Chem.* 84, 223–226 (2014).
9. H. Wang, Theoretical study on the molecular structure, intermolecular interaction and spectral features of 2-aminopyridine/ 2,3-dichloro-5,6-dicyano-1,4-benzoquinone complex. *J. Chem Sci.* 129, 775–782 (2017).
10. F. S. Mjalli and O. U. Ahmed, Characteristics and intermolecular interaction of eutectic binary mixtures: Reline and Glyceline. *Korean J. Chem. Eng*. 33, 337–343 (2016).
11. V.V. Kompaneets and I.A.Vasileva, The influence of terminal substituents of diphenylbutadiene on the parameters of intra-and intermolecular interactions. *Opt. Spectrosc.* 122, 615–624 (2017).
12. S. Elangovan and S. Mullainathan, Intermolecular interactions in methyl formate–ethanol mixtures at 303–313 K according to ultrasonic data. *Russ J. Phys Chem*. 88, 601-606 (2014).
13. D. D. Perrin, W.L.F. Armarego, Purification of Laboratory Chemicals. Pergamon Press, Oxford (1980).
14. H. Eyring and J. F.Kincaid, Free volumes and free angle ratios of molecules in liquids. *J. Chem Phys.* 6, 620-629 (1938).
15. S.Singh, S.Parveen , D. Shukla , M. Yasmin, M. Gupta, and J.P. Shukla, ultrasonic and volumetric study of N–H bond complexes in binary mixtures. *J. Sol. Chem.* 40,889-899 (2011).
16. B. Thanuja, G. Nithya and C. C.Kanagam, Ultrasonic studies of intermolecular interactions in binary mixtures of 4-methoxy benzoin with various solvents: Excess molar functions of ultrasonic parameters at different concentrations and in different solvents. *Ultrasonics Sonochem.*19, 1213-1220 (2012).

17. V. Vanathi, S. Mullainathan and S. Nithyanantham, Acoustical parameters of toluene + chloroform + cyclohexane mixtures at 303.15, 308.15 and 313.15K. *Russian. J. Phys. Chem.* 86, 1204-1207 (2012).
18. A. M. Zaichikov and M. A. Krest'yaninov, Structural and thermodynamic properties and intermolecular interactions in aqueous and acetonitrile solutions of aprotic amides. *J. Struct Chem*. 54, 336–344 (2013).
19. M.K.M.Z. Hyder, S. Akhtar, S.H. Mir and A. Khosla, Density, excess molar volume and some of their derived properties of the binary systems of methyl acetate with methyl derivatives of monoethanolamine between 293.15 and 313.15 K. *Microsyst Technol.* 24, 4357–4371 (2018).
20. S. Elangovan and D.W. Amente, Intermolecular interaction of methyl formate with 1-butanol, 1-pentanol and 1-hexanol at 303K. *Material. Sci. Res. India.*14, 212-214 (2017).
21. H. Iloukhani and M.Soleimani, Measurement and modeling the excess molar volumes and refractive index deviations of binary mixtures of 2-propanol, 2-butanol and 2- pentanol with n-propylamine. *J. Sol. Chem.* 46, 2135–2158 (2017).
22. O.E.A Adam and A.M. Awwad, Estimation of excess molar volumes and theoretical viscosities of binary mixtures of benzene + n-alkanes at 298.15 K. *Int J. Ind Chem.* 7, 391–400 (2016).
23. O. Redlich and A. T. Kister, Algebraic representation of thermodynamic properties and the classification of solutions. *Ind. Eng Chem.* 40, 345-348 (1948).
24. R.J. Fort and W. R. Moore, viscosities of binary liquid mixtures. *Trans. Faraday Soc*. 62, 1112-1119 (1966).