

Thermodynamic properties of binary liquid mixtures formed by cyclic ether (1,4-Dioxane) with aromatic hydrocarbons at 298.15K.

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ABSTRACT: The present review aims to explore and analyze the thermodynamic and acoustic behavior of binary liquid mixtures through the study of excess sound velocity (u^E) and excess viscosity (ηE). The primary objective is to investigate how molecular interactions, temperature variation, and compositional changes influence these excess properties, which serve as sensitive indicators of non-ideal mixing behavior. Each of the experimental methods specific gravity bottle, Ostwald viscometer and ultrasonic interferometer offers special benefits in terms of precision, throughput, and operational simplicity. These approaches enable a strong knowledge of the thermodynamic behaviour of liquid mixes over a large spectrum of situations. Ultrasonic velocity (u), Density (ρ) and viscosity (η) of the binary liquid mixture of 1,4 -Dioxane (Cyclic ether) and Benzene, Ethyl benzene, Toluene were measured over entire composition range. The measurements were done at constant temperature 298.15 K and at ultrasonic frequency 3Mh.Z. From these data excess values are evaluated. From the properties of these excess values, the nature and strength of molecular

interactions in these binary systems are discussed. The non-linear variation of these parameters shows the presence of interaction and their excess values is indicative of interaction between the components molecule in all systems. Results across several binary systems showed consistent correlations between V^E , H^E , and speed of sound, therefore exposing the link between volumetric and energetic changes with molecular packing, hydrogen bonding, and structural fit. Whereas positive values indicate weaker, repulsive, or sterically inhibited interactions. Temperature was found to have a major impact on these characteristics, usually reducing favourable interactions and moving surplus features either towards zero or positive values. Understanding molecular interactions depends on excess thermodynamic and acoustic features, so this study emphasises their relevance and provides essential information for solvent selection, formulation design, and process optimisation in chemical and pharmaceutical sectors.

Keywords: Binary mixtures, thermodynamic properties, Density, Viscosity, 1,4-Dioxane, Ethyl benzene, Molecular interaction.

Introduction: In the recent years, mixed solvents rather than pure solvents find practical application in most chemical processes as their properties are less known [1-2]. Ultrasonic technique has become a powerful tool for studying the molecular behaviour of liquid mixtures [3]. This is because of its ability of characterizing physico - chemical behaviour of liquid medium [4-6]. Binary liquid mixtures due to their unusual behaviour have attracted considerable attention [7]. Data on some of the properties associated with the liquids and liquid mixtures like density, viscosity and ultrasonic velocity find extensive applications in chemical engineering process simulation, textile industries solution theory and molecular dynamics [8] . These measurements are used to study the molecular interactions in pure liquids, liquid mixtures and ionic interactions in solution comprising either single or mixed solutes [9-10]. The studies on solution properties of liquid mixtures consisting of polar as well as non-polar components find applications in industrial and technological process. 1,4-Dioxane finds wide application in various industries as an organic solvent, its miscibility with numerous other solvents permits its use with them, thereby increasing their individual efficiency. Therefore in order to have a clear understanding of the intermolecular interaction between 1,4 –Dioxane (Cyclic ether) - Benzene, 1,4 –Dioxane - Ethyl benzene, 1,4 –Dioxane

- Toluene molecules, an attempt has been made to measure density, viscosity and ultrasonic velocity for the mixtures over the entire composition range. The present investigation aims at understanding the molecular interactions based on thermodynamically parameters and their excess functions in the binary mixture at 298.15 K.

Ultrasonic velocity is one such property which is very important for the study of molecular interactions in solution, its gives information about the extent of interaction and some other parameters such as adiabatic compressibility and free length, velocity determination of ultrasound waves enables us to study the properties of the medium, in which it travels. Owing to the importance of ultrasound velocity in the investigation of molecular interactions in solution. Its measurements have been carried out in the different binary liquid mixtures studied in the present work. Recently, ultrasonic method has become a powerful tool to provide information regarding the physical and chemical properties of liquid system. The ultrasonic study in organic liquid mixture is interesting to discuss non-linear behaviour with respect to concentration and frequency. The ultrasonic study of pure liquids and liquid mixtures is essential in understanding the nature of molecular interactions. The thermodynamic and acoustical properties of binary liquid systems have been studied for several reasons. The most important of these is which gives us information about the molecular interactions. The viscosity of binary liquid systems is one of the properties widely used in various chemical-engineering fields regarding the fluid flow, mass and heat transfer calculations. This property is also used for characterization of pure components. Viscometric investigations of binary liquid systems enable determination of some useful thermodynamic and other parameters which are highly sensitive to molecular interactions. *The quantitative and qualitative analyses of excess functions provide information about the nature of molecular interactions in the binary mixtures. The literature survey reveals that there has been practically no study of the binary mixtures of these systems from the point of view of their ultrasonic behaviour.* In an attempt to explore the nature of interactions occurring between the mixing components, the densities, ρ , speeds of sound, u , viscosities, η , of binary mixtures of 1,4-dioxane with benzene, ethyl benzene and toluene have been measured over the entire range of composition at 298.15 K. From the experimental values of the density,

speed of sound and viscosity, the values of the excess sound velocity, u^E , and excess viscosity, η^E have been calculated. The calculated excess and deviation functions have been analyzed in terms of molecular interactions and structural effects. Binary mixture of 1,4 – Dioxane (Cyclic ether) with aromatic hydrocarbons is of great interest due to their extremely non-ideal behaviour and practical importance to the chemical industry.

Experimental:

Chemicals / Materials.

The chemicals used in the present work were high purity laboratory reagent grade samples of 1,4-dioxane, Benzene Ethyl benzene and Toluene, were supplied by CDH Ltd. New Delhi, India with purity 99.5%. All chemicals was stored over sodium hydroxide pellets for several days and fractionally distilled twice. All the chemicals were stored in dark bottles over freshly activated molecular sieve to minimize adsorption of moisture. The purity of the solvent was ascertained by comparing the measured density, dynamic viscosities and sound velocity of the pure component at 298.15K with the available literature [11-15] as shown in Table 2. The reported experimental values of density (ρ), viscosity (η) and sound velocity (u) conform closely to their corresponding literature values, with an average of the absolute value of deviation $3.6 \times 10^{-3} \text{ kg m}^{-3}$ and $3.3 \times 10^{-3} \text{ m. Pa. s.}$

Table 1. CAS Registry Number, Mass Fraction Purity of the chemicals.

Component	Formula	CAS Reg. No.	Supplier	Mass Fraction Purity (%)	Water Content	Method analysis	Purity method
1,4-Dioxane	$\text{C}_4\text{H}_8\text{O}_2$	123-91-1	CDH, (P) Ltd. New Delhi, India	99.9%	0.1%	Double distillation	
Benzene	C_6H_6	108-67-8	CDH, (P) Ltd. New Delhi, India	98.0%	0.01%	Double distillation	
Ethyl benzene	C_8H_{10}	100-41-4	CDH, (P) Ltd. New Delhi, India	98.0%	0.1%	Double distillation	
Toluene	C_7H_8	108-88-3	CDH, (P) Ltd. New Delhi, India	99.0%	0.1%	Double distillation	

Table-2: Comparison of Experimental and Literature density (ρ), speed of sound (u) and refractive index (n) of pure Components with Available Literature Values at T = 298.15K.

Name of Liquid	Density (ρ) g.cm ⁻³		Viscosity (η) m.Pa.s		Sound Velocity (u) m.s ⁻¹	
	Experimental	Literature	Experimental	Literature	Experimental	Literature
1, 4 Dioxane	1.0212	1.0246 ¹¹	1.1285	1.1944 ¹⁴	1346	1344 ¹²
Benzene	0.8672	0.8736 ¹²	0.5851	0.5961 ¹²	1292	1299 ¹²
Ethyl Benzene	0.8674	0.8620 ¹³	0.6299	0.628 ¹³	1324	1312 ¹³
Toluene	0.8576	0.8623 ¹²	0.6026	0.5525 ¹⁵	1306	1307 ¹²

Apparatus and Procedure

Air tight stopper bottles were used for the preparation of the mixtures and were placed in the dark place. The losses in the mixtures were kept to minimum, as evidenced by repeated measurements of physical properties over an interval of 2-3 days during in which before use time no change in physical properties was observed. The mixtures were well mixed by shaking before use. Binary mixtures were prepared by mass, using an electronic analytical balance (Model K-15 Deluxe, K Roy Instruments Pvt. Ltd.) with an accuracy of $\pm 0.00001 \times 10^{-3}$ kg as described elsewhere. The possible error in the mole fraction was estimated to be less than 1×10^{-4} . Five samples were prepared for one system, and their density and sound velocity were measured on the same day.

Density:

Densities of pure liquids and their binary mixtures were determined by using a R. D. Bottle with a 25 cm³ is used to measure the densities (ρ) of pure liquids and binary mixtures. The R. D. Bottle is calibrated by using conductivity water (having specific conductance less than 1×10^6 ohm⁻¹) with 0.9970 and 0.9940 gcm⁻³ as its densities at T = 298.15 K, respectively. The R. D. Bottle filled with air bubbles free liquids is kept in a thermostat water bath (MSI Goyal

Scientific, Meerut, India) controlled with a thermal equilibrium. The precision of the density measurements was estimated to be ± 0.0002 g cm $^{-3}$.

Sound velocity: The ultrasonic velocity were measured using a multi-frequency ultrasonic interferometer (Model F80D, Mittal Enterprise, New Delhi, India) working at 3 M.Hz. The meter was calibrated with water and benzene. Measurement of sound velocity through medium was based on the accurate determination of the wavelength of ultrasonic waves of known frequency produced by quartz crystal in the measuring cell. The interferometer cell was filled with the test liquid, and water was circulated around the measuring cell from a water bath. The uncertainty was estimated to be 0.1 ms $^{-1}$. The measured values of ultrasonic velocities of pure 1,4-dioxane, benzene, ethyl benzene, and toluene at 298.15K were 1346, 1292, 1324, and 1306 m.s $^{-1}$ respectively, which compare well with the corresponding literature values.

Viscosity:

The viscosity of pure liquids and their binary mixture were measured using suspended Ostwald viscometer having a capacity of about 15 ml and the capillary having a length of about 90 mm and 0.5 mm internal diameter has been used to measure the flow time of pure liquids and liquid mixtures and it was calibrated with triply distilled water, methanol and benzene at 298.15 K. The details of the methods and techniques have been described by researchers [16-17]. The efflux time was measured with an electronic stop watch (Racer) with a time resolution (± 0.015), and an average of at least four flow time readings was taken. Glass stopper was placed at the opening of the viscometer to prevent the loss due to evaporation during measurements. The two bulbs reservoir, one at the top and other at the bottom of the viscometer linked to each other by U type facilitate the free full of liquid at atmospheric pressure. Viscosity values (η) of pure liquids and their binary mixtures are calculated using the solution.

$$\frac{\eta}{\rho} = at - \frac{b}{t} \quad (1)$$

Where t is the efflux time and a and b are viscometric constants.

The measured viscosities have reproducibility within ± 0.002 m.Pa.s. The measured values of viscosities of pure 1,4-dioxane, benzene, ethyl benzene, and toluene at 298.15 K were 1.1285, 0.5851, 0.6299, 0.6026 mPas . which compare well with the corresponding literature values.

THEORETICAL:

The excess sound velocity (u^E) is evaluated from the experimental values of ultrasound velocities for component liquid and their binary mixtures by

$$u^E = u_{1,2} - u_1 X_1 + u_2 X_2 \quad (1)$$

Where $u_{1,2}$ is ultrasound velocity in the mixture and u_1, u_2, X_1, X_2 are the sound velocities and mole fractions respectively of the component liquid 1 and 2.

The ultrasonic velocity (u), density (ρ) and viscosity (η) in pure liquids and liquid mixtures of various concentrations have been measured at 298.15 K.

Viscosity Deviations:

The viscosity deviations ($\Delta\eta$) with mole fraction were calculated by the following:

$$\Delta\eta = \eta_{12} - \sum_{i=1}^2 i\eta_i, \quad (2)$$

Where, X_i , η_i , and η_{12} refer, respectively, to the mole fraction and viscosities of i^{th} pure components and of the binary mixtures.

The excess value of A^E of these thermodynamic parameters have been obtained by subtracting the ideal value from the experimental value

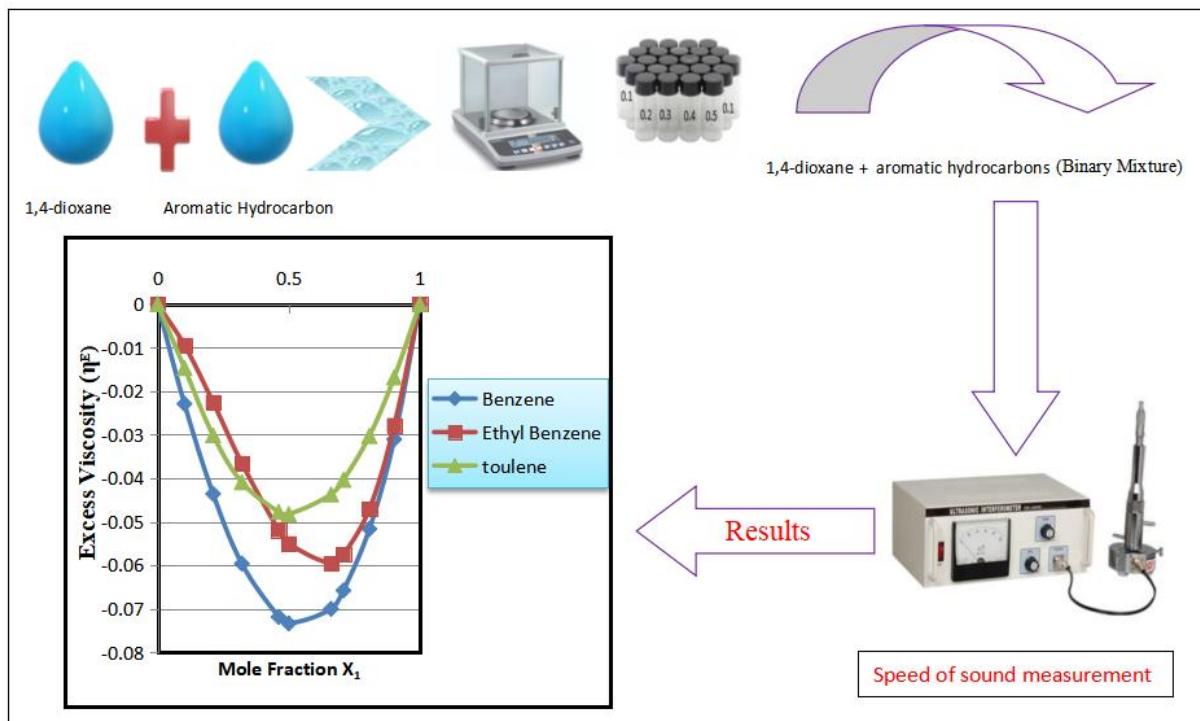
$$A^E = A_{\text{exp.}} - (X_1 A_1 + X_2 A_2)$$

Where A represents the parameter such as intermolecular free length, molar volume, available volume, free volume and isentropic compressibility and X_1 and X_2 are the mole fractions of components whose parameters.

Results and Discussion

The experimental values of densities, sound velocity and viscosities of the hydrocarbons are compared with the literature values and are presented in Table 2. It was found that the experimental values are in proximity with the literature values. Insufficient data on densities, viscosities and sound velocity of pure 1,4-dioxane, Benzene, Ethyl benzene, Toluene, is available. The densities, ρ , viscosities, η , and sound velocity, u , of binary mixtures were measured at 298.15 ± 0.01 K as a function of the composition of the corresponding binary

mixtures. The results of the study are presented in Tables 3. A perusal of table 3 shows that the sound velocity increase with mole fraction of 1,4-dioxane increases for all the binary mixtures. Ultrasonic wave are high frequency mechanical waves. Their velocities in a medium depend inversely on density and the compressibility of the medium.



Scheme 1. Interactions between 1,4-dioxane with aromatic hydrocarbons at 298.15K

Table 3. Values of density (ρ), sound velocity (u) and viscosity (η) for Various 1,4-Dioxane Mole Fractions x_1 of the Binary Mixture (1,4-Dioxane (1) + Aromatic hydrocarbon (2)) at Temperatures $T = 298.15$ K.

Mole fraction of 1,4-dioxane (1)	Density (ρ) g.cm ⁻³	Sound velocity (u) m.s ⁻¹	Viscosity (η) m.Pa.s
1,4-dioxane + Benzene			
0.0000	0.8672	1292	0.5851
0.1024	0.8684	1296	0.6301
0.2117	0.8752	1302	0.6618
0.3214	0.8894	1307	0.7012
0.4617	0.9026	1311	0.7526

0.5001	0.9326	1317	0.7915
0.6616	0.9424	1324	0.8125
0.7088	0.9621	1328	0.8498
0.8079	0.9860	1334	0.9750
0.9026	1.0084	1340	1.0441
1.0000	1.0212	1346	1.1285

1,4-dioxane + Ethyl Benzene

0.0000	0.8674	1324	0.6299
0.1024	0.8692	1326	0.6471
0.2117	0.8780	1328	0.6741
0.3214	0.9080	1330	0.7288
0.4617	0.9208	1332	0.7927
0.5001	0.9404	1334	0.8314
0.6616	0.9548	1336	0.8886
0.7088	0.9764	1338	0.9246
0.8079	0.9924	1340	0.9597
0.9026	0.9992	1342	0.9880
1.0000	1.0212	1346	1.1285

1,4-dioxane + Toluene

0.0000	0.8576	1306	0.6026
0.1024	0.8708	1312	0.6564
0.2117	0.8816	1319	0.7101
0.3214	0.8976	1325	0.7365
0.4617	0.9026	1331	0.7789
0.5001	0.9336	1337	0.8181
0.6616	0.9676	1340	0.8667
0.7088	0.9732	1342	0.9150
0.8079	0.9837	1344	0.9590

0.9026	0.9932	1345	0.9980
1.0000	1.0212	1346	1.1285

Table-3 shows that the density of the binary mixtures increase with increasing mole fraction of the 1,4-dioxane. This trend may suggest that the increasing concentration of the 1,4-dioxane leads to increase the number of particles in a given region[18]. The increasing trend of viscosity with increasing mole fraction of 1,4-dioxane is due to that the intermolecular interaction between the 1,4-dioxane aromatic hydrocarbons. The increasing in density and viscosity with increase in concentration of 1,4-dioxane indicating loosening of intermolecular forces due to thermal agitation of molecules in the mixture. In this present study, ultrasonic velocity increasing with increasing concentration of 1,4-dioxane (Table-3). Moreover acoustic impedance shows the same trend of ultrasonic velocity. It may be due to the structural changes occurring in the mixtures resulting in weakening of intermolecular forces.

For the binary liquid mixture 1,4-dioxane (1) + Benzene (2), 1,4-dioxane (1) + Ethyl Benzene (2) and 1,4-dioxane (1) + Toluene (2), the obtained excess sound velocity (u^E) values are negative over the whole composition range at 298.15 K as depicted in figure-1. Various type of interaction which are possible and which can operate in the binary liquid mixtures containing 1,4- dioxane and aromatic hydrocarbon, that can produce negative deviation in excess sound velocity and excess viscosity.

The excess sound velocity (u^E) for the binary systems of 1,4-dioxane (1) + Benzene (2), 1,4-dioxane (1) + Ethyl Benzene (2) and 1,4-dioxane (1) + Toluene (2) are graphically presented in Fig. 1, together with the Redlich-Kister correlation. It is evident that all three mixtures exhibit significant negative u^E values, across the whole composition range and at working temperature 298.15K.

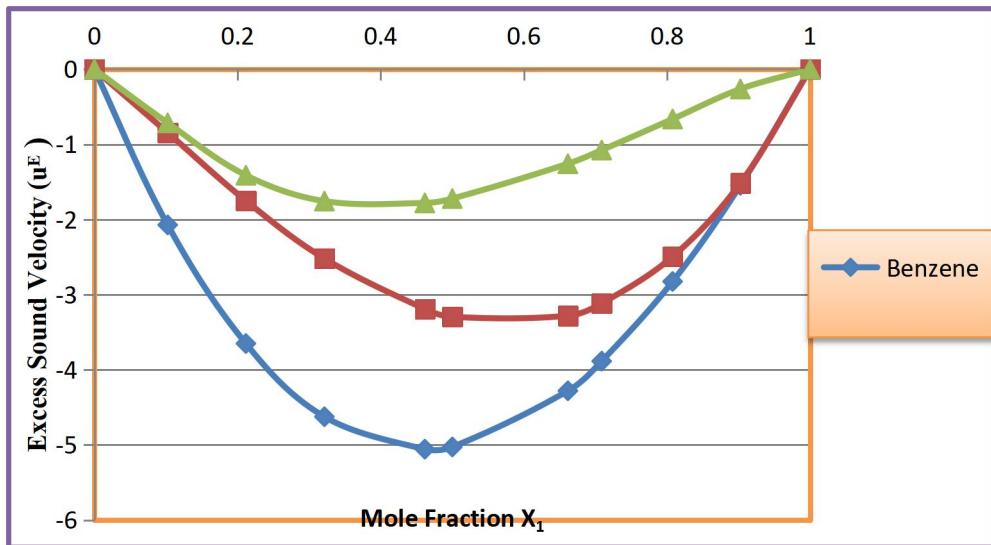


Figure-1: Plots of excess sound velocity (u^E) versus mole fraction of 1,4-Dioxane (x_1) at 298.15K for binary mixtures of 1,4-Dioxane with Benzene (diamond), Ethyl benzene (square) and Toluene (triangle).

The nature of molecular interactions between the components of the liquid mixtures, it is of interest to discuss the same in terms of excess parameters rather than actual values. Non-ideal liquid mixtures show considerable deviation from linearity in their concentrations and this can be interpreted as the presence of strong or weak interactions. The extent of deviation depends upon the nature of the constituents and composition of the mixtures. The thermodynamic excess properties are found to be more sensitive towards intermolecular interaction among the component molecules of liquid mixtures. The sign and extent of deviation of excess parameters depend upon on the strength of interaction between unlike molecules (19). So various excess acoustic and thermodynamic parameters have been evaluated and corresponding graphs are also given.

The sign and magnitude of excess ultrasonic velocity (u^E) play an important role in describing molecular rearrangement as a result of the molecular interaction between the component molecules in the mixtures. The excess ultrasonic velocity (u^E) curves at 298.15 K varying with mole fraction of 1,4-dioxane are represented in Figure-1 for the three binary systems. The excess ultrasonic velocity values exhibiting negative in all three binary systems. Generally, the value of the excess function (u^E) depend upon several physical and chemical contributions (20,21). The physical contribution depends mainly on two factors, namely:

1. The dispersion forces or weak dipole-dipole interaction that leads to positive values.
2. The geometrical effect allowing the fitting of molecules of two different sizes in to each other's structure resulting in negative values

The chemical contributions include breaking up of the associates present in pure liquids, resulting in negative u^E . The ultrasonic velocity in a mixture is mainly influenced by its molecular property. The results for the excess sound velocity (u^E) plotted in figure-1 are negative for all the binary system studied. The observed negative trends in excess sound velocity indicate that the effect due to the breaking up of self-associated structure of the components of the mixtures is dominant over the effect of dipole-dipole interaction between unlike molecule. The negative values of excess sound velocity (u^E) increase with the increase in mole fraction which indicates the increase in strength of interaction with all three binary mixtures. The negative excess sound velocity (u^E) clearly suggests that there exist strong molecular interaction between the molecules of all the three binary liquid mixtures.

The measurement of viscosity in binary liquid mixture gives some reliable information in the study of intermolecular interaction. The molecules of one or more components forming the temarise are either polar, associating or accordingly show non-ideal behavior's in mixtures. Negative values of η^E in most of the cases are the consequence of lower viscosity contributions of similar non-specific interaction and hydrogen bonding effect of molecular species in real mixtures rather than those in the corresponding ideal mixtures. In the present study, it is observed that, for the three binary systems the η^E values gradually decrease up to the mole fraction around 0.5 and then begins to increase [Figure 2](#) more over it is observed that the η^E values decrease as the concentration of x_1 increase. The negative values imply the presence of dispersion forces between the mixing components in the mixtures.

The excess transport properties of the mixtures are influenced by three main types of contributions, namely,

- (i) due to non specific Van der Waals type forces,
- (ii) due to hydrogen bonding, dipole-dipole, and donor-acceptor interaction between unlike molecules, and
- (iii) due to the fitting of smaller molecules into the voids created by the bigger molecules.

The experimental values of viscosities as a function of mole fraction of cumene for three systems are shown in [Fig 2](#). The three systems exhibit a negative deviation of excess viscosity over entire mole fraction range with a maximum corresponding to a mole fraction of about 0.5 at the temperature studied. These deviations indicate specific molecular interactions between different molecules. According to Fort and Moore, the excess viscosity gives the strength of the molecular interaction between the interacting molecules. The excess value of viscosity at

the three binary mixtures 1,4-dioxane (1) + Benzene (2), 1,4-dioxane (1) + Ethyl Benzene (2) and 1,4-dioxane (1) + Toluene (2) at the 298.15 K are shown in figure - 2. The Figure -2 represents the variation of excess viscosity (η^E) is found to be negative for all three binary liquid mixtures over the entire composition range at the 298.15 K. Which suggest the presence of weak intermolecular interactions. For systems where dispersion, induction and dipolar forces are operating, the values of excess viscosity are found to be negative, whereas the existence of specific interaction leading to the formation of complexes in mixtures tends to make negative.

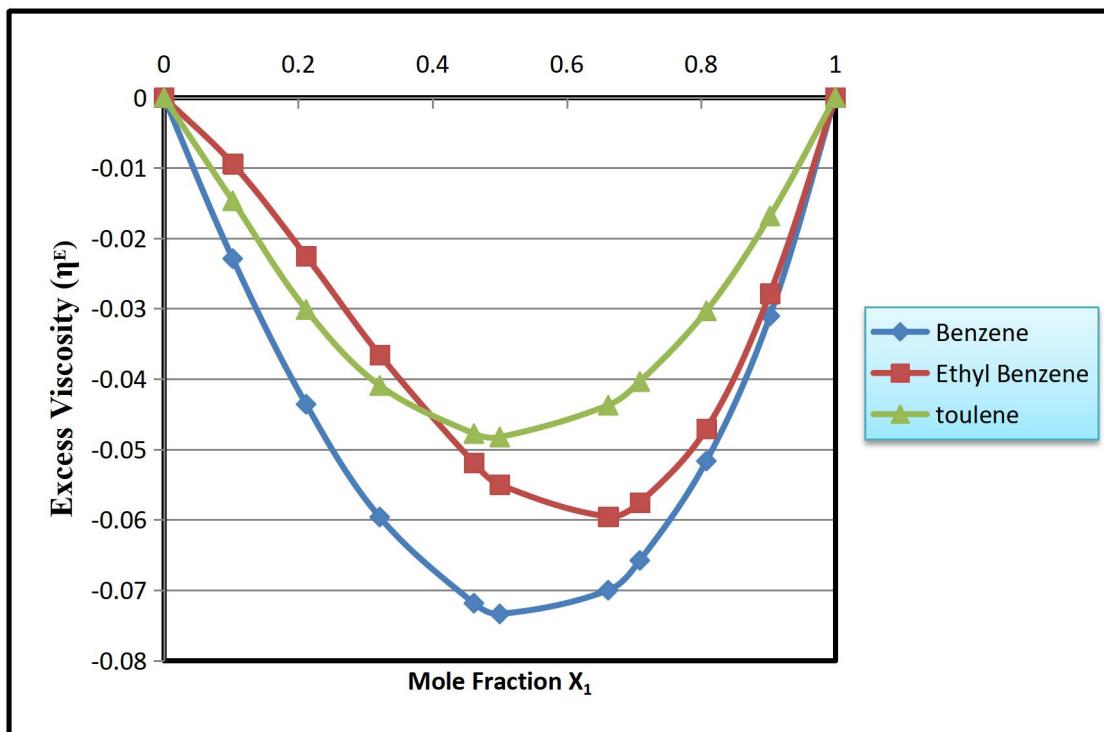


Figure-2: Plots of excess viscosity (η^E) versus mole fraction of 1,4-Dioxane (x1) at 298.15K for binary mixtures of 1,4-Dioxane with Benzene (◇), Ethyl benzene (□) and Toluene (△).

The viscosities of binary liquid mixture 1,4-dioxane (1) + Benzene (2), 1,4-dioxane (1) + Ethyl Benzene (2) and 1,4-dioxane (1) + Toluene (2), at temperature T = 298.15 K decrease linearly with increase in mole fraction of 1,4-Dioxane. Excess viscosity (η^E) values are negative for the all three binary system over the whole mole fraction range (Figure-2).

A reduction in viscosity with increase in mole fraction of 1,4-dioxane suggests that the existing intermolecular interactions are weakening in magnitude. However, the increasing sound velocity and density with increasing mole fraction of 1,4-dioxane leads to a notion that the system is getting more and more compact, which is not true as the interactions due to 1,4-dioxane are dispersive in nature.

The excess viscosity values, which represent the deviation from rectilinear dependence of η_{exp} of binary mixture on mole fraction, have been used to explain the mixture component's intermolecular interaction. The negative η^E values might indicate that the average degree of cross-association between aromatic hydrocarbon and 1,4-dioxane gradually decreased as the chain length of hydrocarbon increased. Thus, the larger negative deviation for the system containing longer chain hydrocarbon confirmed strong dispersion forces in this system. It can be seen from figure-2 that in the mixture, absolute value of (η^E) decrease at mole fraction is raised. An increment of temperature diminishes the self association of the pure component and also the hetero association between unlike molecule, because of the increase of the thermal energy. This lead to less negative values of $\Delta\eta$ as temperature is raised as observed in the present binary mixture [22-23]. Many workers have reported similar behaviour where negative value of (η^E) indicates dispersive interaction. The negative values of excess viscosity (η^E) observed in 1,4-dioxane (1) + Benzene (2) mixture indicate the presence of strong intermolecular interaction amongst the mixing components. The values of excess viscosity (η^E) for all the three system studied are indicative of the predominance of dispersion forces.

Conclusion: In this paper the ultrasonic velocity (u), density (ρ) and viscosity (η) have been measured over the whole composition range at temperature $T = 298.15$ K for the binary mixture 1,4-dioxane (1) + Benzene (2), 1,4-dioxane (1) + Ethyl Benzene (2) and 1,4-dioxane (1) + Toluene (2). Excess sound velocity and deviations in viscosity, for binary mixtures have been calculated and fitted to a Redlich–Kister equation. It is obvious that, there exist a molecular interaction between the components of the mixture. In specific weak molecular interaction like dipole-dipole, dipole-induced dipole and dispersive forces are found to exist between the components of the individual mixtures.

Declarations Conflict of interest

The authors have no competing interests to declare that are relevant to the content of this article.

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Data availability statement

The authors confirm that the data supporting the findings of this study are available within the article.

Author Contribution Statement

Dhirendra Kumar Sharma, Research design, Investigation, Writing-Original draft preparation and Manuscript correction.

Suneel Kumar, Data Analysis and Mathematical Calculation.

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