

# Physicochemical study of intermolecular interactions in 1,4-dioxane+ aromatic hydrocarbons binary mixtures at 298.15K by using ultrasonic and viscometric methods

Dhirendra Kumar Sharma<sup>1\*</sup> and Awadhesh Gaur<sup>2</sup>

<sup>1</sup>Department of Chemistry, Institute of Basic Science, Bundelkhand University, Jhansi (U.P), India.

<sup>2</sup>Department of Mechanical Engineering, IET, Bundelkhand University Jhansi

\*Corresponding author

**Abstract:** The speeds of sound, ( $u$ ) density ( $\rho$ ) and viscosities, ( $\eta$ ) of the binary mixtures of 1,4-dioxane with benzene, ethyl benzene, toluene, o-xylene, m-xylene and p-xylene, over the entire composition, including those of pure liquids, were measured at temperature 298.15K and at atmospheric pressure. From the experimental data, the excess speed of sound,  $u^E$ , and excess viscosity, ( $\eta^E$ ) have been calculated. The results indicated the presence of weak interactions between 1,4-dioxane and aromatic hydrocarbon molecules, which follows the order:

benzene > ethyl benzene > toluene > o-xylene > m-xylene > p-xylene. It is observed that the interactions depend on the number and position of the methyl groups in these aromatic hydrocarbons. Further, the viscosities of these binary mixtures were correlated theoretically by using various empirical and semi empirical models and the results were compared with the experimental findings.

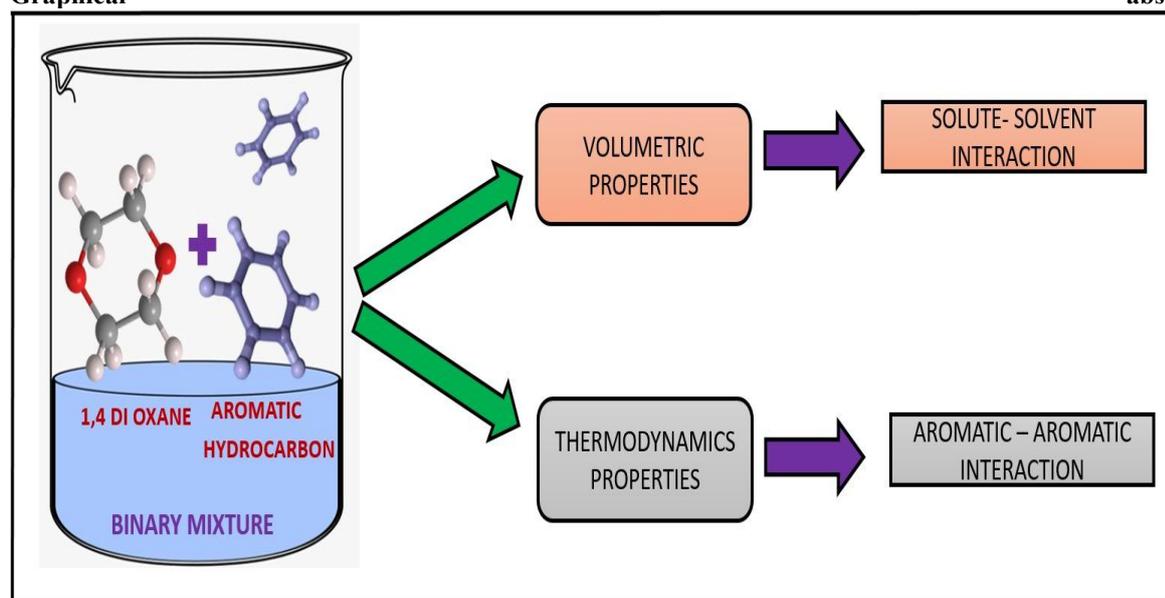
**Keywords:** Speed of sound, Viscosity, 1,4-Dioxane, Aromatic hydrocarbon, Excess properties, Intermolecular interactions.

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abstract



## I. Introduction:

Liquid mixtures consisting of polar and non-polar components are of considerable importance in industries such as petrochemical, pharmaceutical and dye. The formation and destruction of azeotropes in petrochemical industries, the biological activity of drug molecules and the activation energy of the metabolic process basically depend on the type and strength of the intermolecular interactions. Various methods are available to identify these interactions and ultrasonic study is one such more reliable and commonly used study. Thermodynamic and transport properties of liquid mixtures have been extensively used to study the departure of

a real liquid mixture behavior from ideality. Further, these properties have been widely used to study the intermolecular interactions between the various species present in the mixture. The knowledge of physicochemical properties of non-aqueous binary liquid mixtures has relevance in theoretical and applied areas of research and such results are frequently used in designing process (flow, mass transfer or heat transfer calculations) in many chemical and industrial processes [1,2]. The speeds of sound, viscosity of binary liquid mixtures and parameters derived from these properties can be used as important tools for investigating intermolecular interactions between component molecules in liquid mixtures [3–9]. The speed of sound can be considered as a thermodynamic property, provided that a negligible amount of ultrasonic absorption of the acoustic waves of low frequency and of low amplitude is observed; in that case, the ultrasonic absorption of the acoustic waves is negligible [10]. In continuation to our ongoing research on experimental and theoretical studies of physicochemical properties of non-aqueous binary liquid mixtures [11–13], here we report the results of ultrasonic and viscometric studies on the binary mixtures of 1,4-dioxane with six aromatic hydrocarbons (ethyl benzene, benzene, toluene, o-xylene, m-xylene and p-xylene) over the entire composition range at six different systems. 1,4-Dioxane is an excellent aprotic solvent, having zero dipole moment [14], and is commercially used in polymerization and other chemical reactions, in the cleaning of polymer surfaces and electronic materials. On the other hand, the aromatic hydrocarbons possess large quadrupole moments [15], causing an orientation order between molecules of these liquids. The orientation order is due to the partial alignment of neighbouring segments or possibly of whole molecules [15]. Also, the binary mixtures containing aromatic hydrocarbons are interesting because they find applications in the studies of polymer phase diagrams and preferential interaction of polymers in mixed solvents [16,17]. 1,4-dioxane is cyclic ether, having electron-donor ability [18] towards the aromatic rings which act like electron-acceptors [19]. Therefore, the 1,4-dioxane + aromatic hydrocarbon mixtures will be interesting as these may involve charge-transfer interactions which may be influenced by the presence of alkyl groups on the ring. A survey of literature indicates that there have been some studies [3–9, 20] on 1,4-dioxane + benzene / toluene / o-xylene mixtures from the point of view of their ultrasonic and viscometric behaviour.

In the present paper, we report speed of sounds, ( $u$ ), density ( $\rho$ ) and viscosities, ( $\eta$ ) of 1,4-dioxane + ethyl benzene +, benzene, + toluene + o-xylene, + m-xylene and + p-xylene, binary mixtures, including those of pure liquids, at temperature 298.15 K and atmospheric pressure, covering the entire composition range, expressed by the mole fraction,  $x_1$  of 1,4-dioxane. From the experimental data, excess speed of sound, ( $u^E$ ) and excess viscosity, ( $\eta^E$ ) have been calculated. The variation of these parameters with composition and temperature of the mixtures have been discussed in terms of intermolecular interaction in these mixtures.

In the present work, the measurement of ultrasonic velocity, density, viscosity and computation of related parameters at 298.15K in six non-ideal binary mixtures of 1,4-dioxane + o-xylene, 1,4-dioxane + m-xylene, 1,4-dioxane + p-xylene, 1,4-dioxane + benzene, 1,4-dioxane + ethyl benzene and 1,4-dioxane + toluene have been studied.

## II. Materials and Methods

**2.1 Materials** 1, 4 Dioxane, Merk (Emplura) Mumbai, India was supplied with purity  $\geq 99.0\%$ , Benzene, Merk (Emplura) Mumbai, India with  $\geq 99.0\%$ , Ethyl Benzene, CDH, Gujrat, India with  $\geq 99.0\%$ , Toluene, Merk (Emplura) Mumbai, India with  $\geq 99.0\%$ , o- Xylene, CDH, Gujrat, India with  $\geq 99.0\%$ , m-Xylene, CDH, Gujrat, India with  $\geq 99.0\%$ , p-Xylene, CDH, Gujrat, India with  $\geq 99.0\%$ , respectively with corresponding literature values [21-30]. Since the agreement with the literature values is very good. The reported experimental values of density ( $\rho$ ), viscosity ( $\eta$ ) 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. Specification of Different Chemicals.**

| Name of Liquid | CAS No.  | Source                        | Mass Fraction Purity | Water Content | Purification Method |
|----------------|----------|-------------------------------|----------------------|---------------|---------------------|
| 1, 4 Dioxane   | 123-91-1 | Merk ( Emplura) Mumbai, India | 99.0%                | 0.1%          | Double Distillation |
| Benzene        | 71-43-2  | Merk( Emplura) Mumbai, India  | 99.0%                | 0.001%        | Double Distillation |
| Ethyl Benzene  | 100-41-4 | CDH, Gujrat, India            | 99.0%                | 0.1%          | Double Distillation |
| Toluene        | 108-88-3 | Merk( Emplura) Mumbai, India  | 99.0%                | 0.1%          | Double Distillation |
| o- Xylene      | 95-47-6  | CDH, Gujrat, India            | 99.0%                | 0.01%         | Double Distillation |
| m-Xylene       | 108-38-3 | CDH, Gujrat, India            | 99.0%                | 0.01%         | Double Distillation |
| p-Xylene       | 106-42-3 | CDH, Gujrat, India            | 99.0%                | 0.05%         | Double Distillation |

**Table 1. Density ( $\rho$ ), sound velocity ( $u$ ) and viscosity ( $\eta$ ) of pure Components at T = 298.15K.**

| Compound      | $\rho$ (g.cm <sup>-3</sup> ) |                      | $u$ (m.s <sup>-1</sup> ) |                    | $\eta$ (mPa s) |                      |
|---------------|------------------------------|----------------------|--------------------------|--------------------|----------------|----------------------|
|               | Observed                     | Literature           | Observed                 | Literature         | Observed       | Literature           |
| 1, 4 Dioxane  | 1.0212                       | 1.0246 <sup>21</sup> | 1346                     | 1344 <sup>23</sup> | 1.1285         | 1.1944 <sup>22</sup> |
|               |                              | 1.0268 <sup>22</sup> |                          | 1346 <sup>22</sup> |                | 1.1960 <sup>22</sup> |
| Benzene       | 0.8672                       | 0.8736 <sup>23</sup> | 1292                     | 1299 <sup>23</sup> | 0.5851         | 0.5961 <sup>27</sup> |
|               |                              | 0.8738 <sup>24</sup> |                          | 1296 <sup>27</sup> |                | 0.6040 <sup>30</sup> |
| Ethyl Benzene | 0.8674                       | 0.8620 <sup>25</sup> | 1324                     | 1312 <sup>25</sup> | 0.6299         | 0.6280 <sup>25</sup> |
|               |                              | 0.8626 <sup>25</sup> |                          | 1319 <sup>28</sup> |                | 0.6373 <sup>25</sup> |
| Toluene       | 0.8576                       | 0.8623 <sup>23</sup> | 1306                     | 1307 <sup>23</sup> | 0.6026         | 0.5540 <sup>30</sup> |
|               |                              | 0.8624 <sup>24</sup> |                          | 1309 <sup>29</sup> |                | 0.5531 <sup>29</sup> |
| o- Xylene     | 0.8730                       | 0.8756 <sup>26</sup> | 1332                     | 1347 <sup>26</sup> | 0.7983         | 0.7550 <sup>26</sup> |
|               |                              | 0.8755 <sup>23</sup> |                          | 1347 <sup>23</sup> |                | 0.7576 <sup>29</sup> |
| m-Xylene      | 0.8592                       | 0.8600 <sup>23</sup> | 1322                     | 1323 <sup>23</sup> | 0.5797         | 0.5850 <sup>26</sup> |
|               |                              | 0.8601 <sup>26</sup> |                          | 1323 <sup>26</sup> |                | 0.5880 <sup>29</sup> |
| p-Xylene      | 0.8498                       | 0.8568 <sup>23</sup> | 1314                     | 1314 <sup>26</sup> | 0.5811         | 0.6020 <sup>26</sup> |
|               |                              | 0.8562 <sup>26</sup> |                          | 1315 <sup>26</sup> |                | 0.6050 <sup>26</sup> |

## 2.2 Measurements:

### 2.2.1 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  $1 \times 10^{-4}$ . Five samples were prepared for one system, and their density and sound velocity were measured on the same day.

### 2.2.2 Density:

Densities of pure liquids and their binary mixtures were determined by using a R. D. Bottle with a 25 cm<sup>3</sup> is used to measure the densities ( $\rho$ ) of pure liquids and binary mixtures. The R. D. Bottle is calibrated by using conductivity water (having specific conductance less than  $1 \times 10^6$  ohm<sup>-1</sup>) with 0.9970 and 0.9940 gcm<sup>-3</sup> as its densities at T = 298.15 K, respectively. The R. D. Bottle filled with air bubbles free liquids is kept in a thermostate water bath (MSI Goyal Scientific, Meerut, India) controlled with a thermal equilibrium. The precision of the density measurements was estimated to be  $\pm 0.0002$  g cm<sup>-3</sup>.

**2.2.3 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<sup>-1</sup>. The measured values of ultrasonic velocities of pure 1,4-dioxane, o-xylene, m-xylene, p-xylene, benzene, ethyl benzene, and toluene at 298.15K were 1346, 1332, 1322, 1314, 1292, 1324, and 1306 m.s<sup>-1</sup> respectively, which compare well with the corresponding literature values.

### 2.2.4 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 [19-20]. The efflux time was measured with an electronic stop watch (Racer) with a time resolution ( $\pm 0.015$ ), 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 ( $\eta$ ) 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  $\pm 0.002$  m.Pa.s. The measured values of viscosities of pure 1,4-dioxane, o-xylene, m-xylene, p-xylene, benzene, ethyl benzene, and toluene at 298.15 K were 1.1285, 0.7983, 0.5797, 0.5811, 0.5851, 0.6299, 0.6026 mPas . which compare well with the corresponding literature values.

### III. THEORETICAL:

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

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

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 ( $\rho$ ) and viscosity ( $\eta$ ) in pure liquids and liquid mixtures of various concentrations have been measured at 298.15 K.

#### 3.1 Viscosity Deviations:

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

$$\Delta\eta = \eta_{12} - \sum_{i=1}^2 \eta_i \quad (3)$$

Where,  $x_i, \eta_i$ , and  $\eta_{12}$  refer, respectively, to the mole fraction and viscosities of  $i^{\text{th}}$  pure components and of the binary mixtures.

The strength of interaction between the component molecules of binary mixtures is well reflected in the deviation of the excess functions from ideality.

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

$$A^E = A_{exp} - (X_1 A_1 + X_2 A_2) \quad (4)$$

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.

### IV. Results and Discussion

The experimental values of ultrasonic velocity (u), density ( $\rho$ ) and viscosity ( $\eta$ ) of 1,4-Dioxane with + aromatic hydrocarbons mixtures at 298.15K are listed in Table 3. From these values, we have computed excess Sound velocity ( $u^E$ ) and excess viscosity ( $\eta^E$ ) and are presented in table 3.

**Table 3. Density ( $\rho$ ), ultrasonic velocity (u), viscosity ( $\eta$ ), excess sound velocity ( $u^E$ ) and excess viscosity ( $\eta^E$ ) of binary mixture of 1,4-Dioxane (1) + aromatic hydrocarbons (2) at 298.15 K**

| Mole fraction 1,4-Dioxane ( $x_1$ ) | Density ( $\rho$ ) / g.cm <sup>3</sup> | Sound velocity (u) / ms <sup>-1</sup> | Viscosity ( $\eta$ ) / mPas. | Excess Sound velocity ( $u^E$ ) / ms <sup>-1</sup> | Excess viscosity ( $\eta^E$ ) / mPas. |
|-------------------------------------|--|---------------------------------------|------------------------------|--|---------------------------------------|
| <b>1,4-Dioxane + o-xylene</b>       |  |                                       |                              |  |                                       |
| 0.0000                              | 0.8730                                 | 1332                                  | 0.7983                       | 0.0000   | 0.0000                                |
| 0.1260                              | 0.8784                                 | 1334                                  | 0.8073                       | -0.6286  | -0.0274                               |
| 0.2289                              | 0.8916                                 | 1336                                  | 0.8402                       | -1.2277  | -0.0560                               |
| 0.3214                              | 0.9040                                 | 1338                                  | 0.8739                       | -1.7171  | -0.0814                               |
| 0.4460                              | 0.9236                                 | 1340                                  | 0.9133                       | -2.1187  | -0.1052                               |
| 0.5089                              | 0.9336                                 | 1341                                  | 0.9340                       | -2.1752  | -0.1093                               |
| 0.6046                              | 0.9456                                 | 1342                                  | 0.9515                       | -2.1080  | -0.1122                               |
| 0.7060                              | 0.9644                                 | 1343                                  | 0.9761                       | -1.9836  | -0.1078                               |
| 0.8072                              | 0.9824                                 | 1344                                  | 1.0005                       | -1.5468  | -0.0894                               |
| 0.9198                              | 1.0044                                 | 1345                                  | 1.0516                       | -0.8856  | -0.0588                               |
| 1.0000                              | 1.0212                                 | 1346                                  | 1.1285                       | 0.0000   | 0.0000                                |
| <b>1,4-Dioxane + m-xylene</b>       |  |                                       |                              |  |                                       |
| 0.0000                              | 0.8592                                 | 1322                                  | 0.5797                       | 0.0000   | 0.0000                                |
| 0.1265                              | 0.8600                                 | 1324                                  | 0.6103                       | -0.5376  | -0.0457                               |
| 0.2278                              | 0.8732                                 | 1326                                  | 0.6501                       | -1.1166  | -0.0962                               |
| 0.3133                              | 0.8804                                 | 1328                                  | 0.6760                       | -1.6414  | -0.1433                               |
| 0.4151                              | 0.8996                                 | 1330                                  | 0.7012                       | -2.1297  | -0.1897                               |
| 0.5030                              | 0.9144                                 | 1334                                  | 0.7446                       | -2.2118  | -0.1984                               |

|                                  |        |      |        |         |         |
|----------------------------------|--------|------|--------|---------|---------|
| 0.6080                           | 0.9324 | 1336 | 0.8027 | -2.2266 | -0.2084 |
| 0.7108                           | 0.9608 | 1338 | 0.8607 | -2.1107 | -0.2015 |
| 0.8125                           | 0.9740 | 1340 | 0.9065 | -1.6547 | -0.1698 |
| 0.9993                           | 0.9940 | 1342 | 0.9713 | -0.9145 | -0.1144 |
| 1.0000                           | 1.0212 | 1346 | 1.1285 | 0.0000  | 0.0000  |
| <b>1,4-Dioxane + p-xylene</b>    |        |      |        |         |         |
| 0.0000                           | 0.8498 | 1314 | 0.5811 | 0.0000  | 0.0000  |
| 0.1807                           | 0.8668 | 1318 | 0.6252 | -1.5838 | -0.0567 |
| 0.2356                           | 0.8780 | 1324 | 0.6537 | -2.9144 | -0.1069 |
| 0.3049                           | 0.8936 | 1328 | 0.6965 | -3.8592 | -0.1450 |
| 0.4153                           | 0.9032 | 1330 | 0.7355 | -4.4697 | -0.1733 |
| 0.5027                           | 0.9144 | 1334 | 0.7872 | -4.5155 | -0.1766 |
| 0.6078                           | 0.9388 | 1338 | 0.8519 | -4.1134 | -0.1674 |
| 0.7108                           | 0.9684 | 1340 | 0.9013 | -3.8091 | -0.1571 |
| 0.8152                           | 0.9828 | 1342 | 0.9375 | -2.8854 | -0.1236 |
| 0.9993                           | 0.9948 | 1344 | 0.9721 | -1.6326 | -0.0753 |
| 1.0000                           | 1.0212 | 1346 | 1.1285 | 0.0000  | 0.0000  |
| <b>1,4-Dioxane + benzene</b>     |        |      |        |         |         |
| 0.0000                           | 0.8672 | 1292 | 0.5851 | 0.0000  | 0.0000  |
| 0.1024                           | 0.8684 | 1296 | 0.6301 | -1.2690 | -0.0455 |
| 0.2117                           | 0.8752 | 1302 | 0.6618 | -2.5497 | -0.0941 |
| 0.3214                           | 0.8894 | 1307 | 0.7012 | -3.6565 | -0.1381 |
| 0.4617                           | 0.9026 | 1311 | 0.7526 | -4.6450 | -0.1791 |
| 0.5001                           | 0.9326 | 1317 | 0.7915 | -4.8058 | -0.1861 |
| 0.6616                           | 0.9424 | 1324 | 0.8125 | -4.8213 | -0.1885 |
| 0.7088                           | 0.9621 | 1328 | 0.8498 | -4.5920 | -0.1794 |
| 0.8079                           | 0.9860 | 1334 | 0.9750 | -3.7075 | -0.1431 |
| 0.9026                           | 1.0084 | 1340 | 1.0441 | -2.2937 | -0.0837 |
| 1.0000                           | 1.0212 | 1346 | 1.1285 | 0.0000  | 0.0000  |
| <b>4-Dioxane + ethyl benzene</b> |        |      |        |         |         |
| 0.0000                           | 0.8674 | 1324 | 0.6299 | 0.0000  | 0.0000  |
| 0.1364                           | 0.8692 | 1326 | 0.6471 | -0.6815 | -0.0550 |
| 0.1891                           | 0.8780 | 1328 | 0.6741 | -1.4977 | -0.1034 |
| 0.4029                           | 0.9080 | 1330 | 0.7288 | -2.2998 | -0.1430 |
| 0.5135                           | 0.9208 | 1332 | 0.7927 | -3.1224 | -0.1760 |
| 0.6270                           | 0.9404 | 1334 | 0.8314 | -3.2797 | -0.1809 |
| 0.7043                           | 0.9548 | 1336 | 0.8886 | -3.4670 | -0.1783 |
| 0.7567                           | 0.9764 | 1338 | 0.9246 | -3.3425 | -0.1696 |
| 0.8021                           | 0.9924 | 1340 | 0.9597 | -2.7543 | -0.1380 |
| 0.9080                           | 0.9992 | 1342 | 0.9880 | -1.7157 | -0.0895 |
| 1.0000                           | 1.0212 | 1346 | 1.1285 | 0.0000  | 0.0000  |
| <b>1,4-Dioxane + toluene</b>     |        |      |        |         |         |
| 0.0000                           | 0.8576 | 1306 | 0.6026 | 0.0000  | 0.0000  |
| 0.1139                           | 0.8708 | 1312 | 0.6564 | -2.0692 | -0.0444 |
| 0.2186                           | 0.8816 | 1319 | 0.7101 | -3.6376 | -0.0874 |
| 0.3184                           | 0.8976 | 1325 | 0.7365 | -4.5996 | -0.1233 |
| 0.4206                           | 0.9026 | 1331 | 0.7789 | -5.0214 | -0.1542 |
| 0.5276                           | 0.9336 | 1337 | 0.8181 | -4.9921 | -0.1590 |
| 0.5787                           | 0.9676 | 1340 | 0.8667 | -4.2626 | -0.1589 |
| 0.6261                           | 0.9732 | 1342 | 0.9150 | -3.8809 | -0.1518 |
| 0.7875                           | 0.9837 | 1344 | 0.9590 | -2.8616 | -0.1248 |
| 0.8846                           | 0.9932 | 1345 | 0.9980 | -1.6420 | -0.0824 |
| 1.0000                           | 1.0212 | 1346 | 1.1285 | 0.0000  | 0.0000  |

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, o-xylene, m-xylene, p-xylene, Benzene, Ethyl benzene, Toluene, is available. The densities,  $\rho$ , viscosities,  $\eta$ , and sound velocity,  $u$ , of binary mixtures were measured at  $298.15 \pm 0.01$  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. 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 [31]. 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.

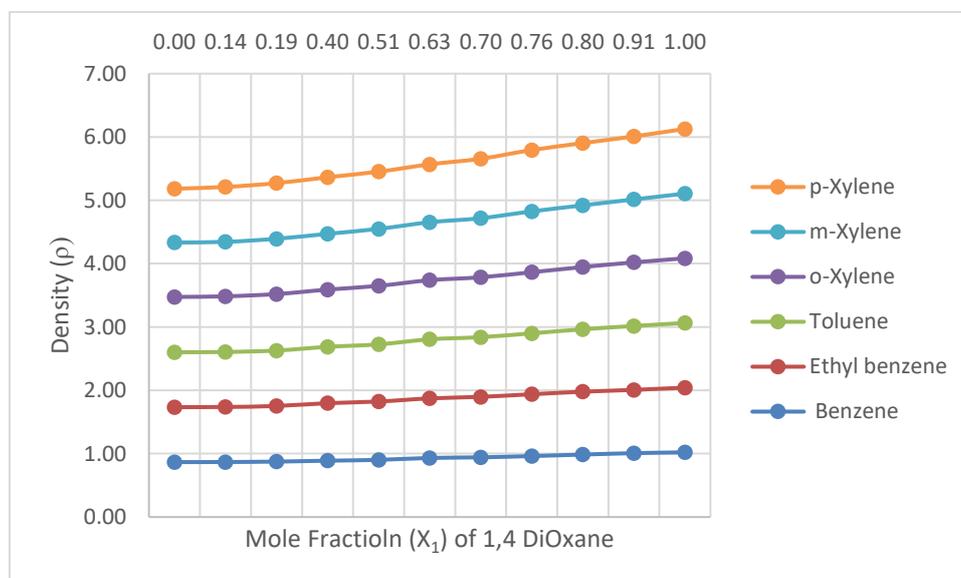


Fig. 1. Variation of density ( $\rho$ ) vs. mole fraction,  $x_1$  of 1,4-dioxane for the binary mixtures at 298.15 K. Points show experimental values and curves show smoothed values using Eq. (4).

The variations ( $u^E$ ) and ( $\eta^E$ ) and with composition of the mixtures, along with smoothed values using equation (5) at 298.15 K are presented graphically in Figs. 2- 3.

The results presented in Fig. 2 indicates that excess sound velocity ( $u^E$ ) values are negative for 1,4-dioxane + benzene/ ethyl benzene, toluene, o-xylene/m-xylene/p-xylene mixtures over entire mole fraction range at investigated temperature 298.15K. The magnitude of deviations in excess sound velocity ( $u^E$ ) (Fig. 2) follows the sequence: ethyl benzene < benzene < toluene < p-xylene < m-xylene < o-xylene. This suggests that there is an increase in the compressibility of the mixtures as we move from benzene to o-xylene.

The behaviour of excess sound velocity ( $u^E$ ) with composition of the mixture has been qualitatively examined by taking into consideration the nature of the component molecules in the pure state and in the mixture.

The molecules of 1,4-dioxane are non polar and those of the aromatic hydrocarbons (benzene, ethyl benzene, toluene, o-xylene, m-xylene and p-xylene,) possess large quadrupole moment [15], which causes molecular order in the pure state. The mixing of 1,4-dioxane with the aromatic hydrocarbons, would induce a reduction in the molecular order in the latter, resulting in an expansion in volume. Also, there is a possibility of the electron donor-acceptor type interactions between electronegative oxygen atoms of 1,4-dioxane (as donor) and the p-electrons of ring of aromatic hydrocarbon molecules (as acceptor), leading to a decrease in volume, resulting in negative excess sound velocity ( $u^E$ ) values. The observed negative values of excess sound velocity ( $u^E$ ) for 1,4-dioxane + benzene, ethyl benzene /toluene o-xylene / m-xylene /p-xylene, mixtures indicates specific interactions between 1,4-dioxane and benzene/toluene molecules, leading to a increase in density of the mixture, which results in negative excess sound velocity ( $u^E$ ) values. The negative values of excess sound velocity ( $u^E$ ) for 1,4-dioxane + benzene, ethyl benzene / toluene, o-xylene / m-xylene / p-xylene mixtures indicates weak interactions between 1,4-dioxane and o-xylene /m-xylene / p-xylene/ molecules, leading to a increase in compressibility of the mixture, which results in negative excess sound velocity ( $u^E$ ) values. The observed trends in excess sound velocity ( $u^E$ ) values suggest the donor-acceptor (charge transfer) interactions between 1,4-dioxane and aromatic hydrocarbon

molecules in these mixtures follows the order: benzene > ethyl benzene >toluene > p-xylene > m-xylene > o-xylene > mesitylene. Similar type of donor-acceptor interactions have also reported by Ma et al., between oxygen atom of sulpholane and p-electrons of the aromatic hydrocarbons and by Ali et al. [11,12], between oxygen atom of dimethyl sulfoxide and p-electrons on the ring of the aromatic hydrocarbons binary mixtures.

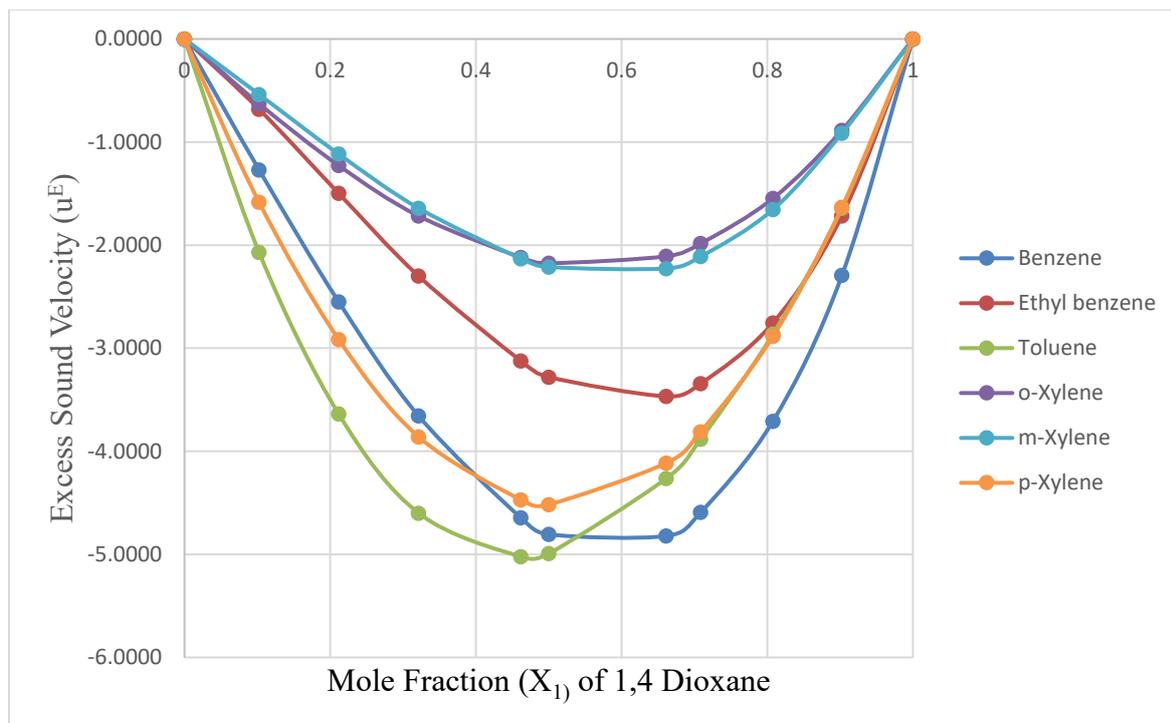


Fig. 2. Variation of excess sound velocity, ( $u^E$ ) vs. mole fraction,  $x_1$  of 1,4-dioxane for the binary mixtures at 298.15 K. Points show experimental values and curves show smoothed values using Eq. (4).

The results presented in Fig. 4 indicate that excess viscosity ( $\eta^E$ ) values are negative for 1,4-dioxane + benzene, ethyl benzene, toluene /o-xylene / m-xylene / p-xylene, mixtures over entire composition range at investigated temperature 298.15K. The observed negative excess viscosity ( $\eta^E$ ) values for 1,4-dioxane + benzene, ethyl benzene, toluene /o-xylene/ m-xylene / p-xylene mixtures indicate specific interactions between the component molecules and negative values of excess viscosity ( $\eta^E$ ) for 1,4-dioxane +benzene, ethyl benzene, toluene/o-xylene/m-xylene/p-xylene/ mixtures indicate the presence of weak interactions between the component molecules [32,33], possibility the electron donor-acceptor type interactions [34] between electronegative oxygen atoms of 1,4-dioxane (as donor) and the p-electrons of ring of aromatic hydrocarbon molecules (as acceptor), resulting in negative excess viscosity ( $\eta^E$ ) values. The magnitudes of excess viscosity ( $\eta^E$ ) values (Fig. 3) at equimolar composition of these mixtures follow the order: benzene > ethyl benzene > toluene > p-xylene > m-xylene > o-xylene > mesitylene, which indicates that the order of the interactions between unlike molecules in these mixtures follows the sequence: benzene > ethyl benzene > toluene > p-xylene > m-xylene > o-xylene > mesitylene. Thus, these trends of excess viscosity ( $\eta^E$ ) further support the behaviors of excess viscosity ( $\eta^E$ ), excess sound velocity ( $u^E$ ), [13] regarding interactions prevailing in these mixtures [35-36].

Amongst the xylenes the magnitude of negative ( $\eta^E$ ) values follow the order: o-xylene < m-xylene < p-xylene; which suggest that the position of  $-\text{CH}_3$  groups on the aromatic ring plays a significant role in deciding the magnitude of ( $\eta^E$ ) and, hence, the order of interaction between the component molecules of the mixtures. The more negative ( $\eta^E$ ) values for 1,4-dioxane + p-xylene are found due to the fact that 1,4-dioxane molecules could approach more closely to the ring of p-xylene from two directions as compared to o- and m-xylenes, leading to maximum interaction between 1,4-dioxane molecule with the former xylene than with the latter two xylenes. These trends are in good agreement with the excess molar volume,  $V_m^E$  values reported [37] for dimethyl sulfoxide + xylene binary mixtures.

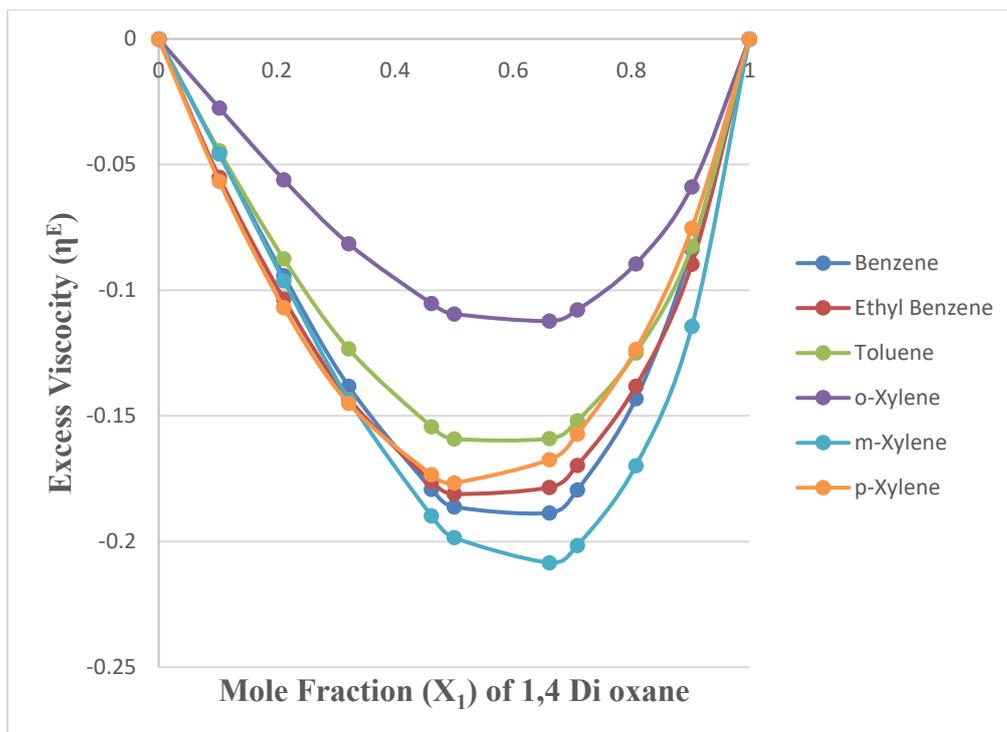


Fig. 3. Variation of excess viscosity, ( $\eta^E$ ) vs. mole fraction,  $x_1$  of 1,4-dioxane for the binary mixtures at 298.15 K. Points show experimental values and curves show smoothed values using Eq. (4).

## V. Conclusions

The speed of sounds and viscosities of binary mixtures of 1,4-dioxane + benzene, ethyl benzene, + toluene, + o-xylene, + m-xylene, and p-xylene, including those of pure liquids have been measured at temperature 298.15K. The values of excess sound velocity ( $u^E$ ), and excess viscosity, ( $\eta^E$ ) have been calculated. The variation of these parameters with composition indicated the presence of weak interactions of electron donor acceptor type between electronegative oxygen atoms of 1,4- dioxane (acting as donor) and the p-electrons of ring of aromatic hydrocarbon molecules (acting as acceptor) in these mixtures and this interaction decrease with increase in temperature. The strength of interactions in these mixtures follows the order: benzene > ethyl benzene > toluene > p-xylene > m-xylene > o-xylene > mesitylene. It is also observed that the interactions depend upon the number and position of methyl groups in these aromatic hydrocarbon molecules. It has been observed that all the investigated models correlate the viscosity results well for all the systems, and the predicting ability of these correlating relations increases as the number of adjustable parameters in the relation increases.

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## Credit authorship contribution statement

**Dhirendra Kumar Sharma:** Writing – review & editing, Writing – original draft, Visualization, Validation, Supervision, Methodology, Investigation, Formal analysis, Data curation, Conceptualization.

**Awadhesh Gaur:** Visualization, Validation, Supervision, Investigation, Formal analysis, Conceptualization.

## Declaration of competing interest

The authors state that none of the work described in this study could have been influenced by any known competing financial interests or personal relationships.

## Data availability

The data that has been used is confidential.

## Nomenclature

$\rho$ - Density of the mixture ( $\text{g.cm}^{-3}$ )

$u$  -Sound speed of the mixture ( $\text{m.s}^{-1}$ )

$u^E$ -Excess Sound Velocity ( $\text{m.s}^{-1}$ )

$\eta$ -Viscosity ( $\text{m.Pas}$ )

M- Molar mass  
T-Temperature  
P-Pressure  
V-Volume  
 $\eta^E$ -Excess Viscosity (m.Pas)  
T- Temperature (Kelvin)

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