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Measurement and Prediction of Thermo Physical Properties to study Intermolecular Interactions of Binary Liquid Mixtures at Various Temperatures by Using McAllister Model

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Abstract*— The important of fundamental data for the design and optimization of chemical process are densities and viscosities. The study conducted in this research are viscosities, η, and densities, ρ, of 1,4 Dioxane with two hydrocarbons Benzene and Chloro Benzene have been measured over the entire range of composition at (303.15, 308.15, and 313.15) K. From experimentations excess molar volumes, V^E, and deviations in viscosities, Δη, calculated. The calculated excess molar volumes, V^E and deviations in viscosities, Δη exhibited positive and negative values respectively over the whole range of composition in both binary systems. The McAllister model is used to deviations in viscosities, Δη, to derive the binary coefficients and standard deviations of these binary systems. The fitted outcomes and the calculated data clearly indicated that weak interactions present in two mixtures. This is due to number and position of methyl groups exist in aromatic hydrocarbons. It can conclude that the data found with the values fitted by the corresponding McAllister model with high degree of precision.

Index Terms: Density, Excess molar volume, Oswald-Sprengel Pyknometer, Oswald viscometer, Viscosity and Viscosity deviation.

I. INTRODUCTION

Chemical design and effective chemical process optimization, the molecular interactions existing in process fluids and thermo physical properties like densities and viscosities, are playing key role. These two are essential for engineering designs involving chemical separations, heat transfer, mass transfer, and fluid flow and important from practical and theoretical points of view for understanding liquid theory[1].The quantitative viscosity, excess molar volume and density data of liquid mixtures are required to solve many engineering problems, involve in chemical separations, heat transfer, mass transfer, and fluid flow are important from practical and theoretical points of view, for understanding liquid theory. The low polarity of 1, 4 Dioxane is interesting to study with hydrocarbon mixtures, for the type of interaction between the components of binary systems. 1,4 Dioxane commonly known as excellent aprotic solvent, it has a zero dipole movement and cyclic ether, that has an electron donor ability towards aromatic rings, it acts like weak electron acceptors. 1, 4 Dioxane is used as a stabilizer in aluminium containers and solvent in inks and adhesives. There are few reports on density and viscosity data of 1, 4 Dioxane with hydrocarbon mixtures [1, 2, 12].In earlier papers, we had studied thermo physical properties of binary systems [2, 12]. In the present paper, it has been reported density (ρ) and viscosity (η) of pure 1, 4 Dioxane, Benzene and Chloro Benzene for the binary system constituted by these two chemicals at entire range of composition and temperature T= (303.15 to 313.15) K. With this data, the excess molar volume and deviation in viscosity have been computed.These results fitted to the McAllister's three-body model, which used to correlate the kinematic viscosities of these binary mixtures [1]. This analyzed technique to derive the binary coefficients and to estimate the standard deviation (σ) between experimental and calculated data.**The effect of the number and position of the methyl groups in these aromatic hydrocarbons on molecular interactions in these mixtures also being discussed. A literature search showed that no measurements have been previously reported by using McAllister's three-body model for the mixtures studied in this paper.**

II. EXPERIMENTAL PROCEDURE

A. Materials:

1, 4-Dioxane, Benzene, ChloroBenzene, this were all supplied by M/s E.Merck Ltd. With the stated purities better than 99 %, were stored over molecular sieves (0.3 nm). 1, 4 Dioxane with purity of 99 % provided by Sigma-Aldrich Chemicals and it was used without further purification. To minimize the contact of these reagents with moist air, the products were kept in sealed bottles in a desiccator. The purity of the substances were determined by GLC. Densities and

viscosities of pure substances and experimental values comparison with literature values are listed in Table 1[1, 2, 3, and 4].

Apparatus and Procedure:

Measurements of the density, ρ, and the kinematic viscosity, γ, of pure liquids and their solutions were carried out using a Pycknometer and Oswald Viscometer and two integrated Pt 100 thermometers. The temperature in the cell was regulated (0.001K with a proportional temperature controller). The apparatus was first calibrated with triple distilled water. The uncertainties in density measurements were estimated to be (2·10-3 kg·m-3 and.? Further information about the experimental techniques has been the kinematic viscosities of the pure liquids and their mixtures were measured at (303.15, 308.15, and 313.15) K .The viscometer was filled with liquid or liquid mixtures, and its limbs were closed with Teflon caps taking due precaution to reduce evaporation losses. An electronic digital stopwatch with a readability of 0.01 s was used for flow time measurements. Experiments were repeated a minimum of four times for all compositions, and the results were averaged. The caps of the limbs were removed during the measurement of flow times. The measured values of kinematic viscosity, γ, were converted to dynamic viscosity,η, after multiplication by the density. The reproducibility of dynamic viscosity was found to be within (0.003 mPa·s). A thermostatically controlled, well-stirred water bath whose temperature was controlled to (0.01 K) was used for all the measurements. Conductivity measurements were carried out in a jacket containing a conductivity cell of cell constant 1.0 cm-1. Water was circulated in the jacket from thermostat, and the temperature was maintained within ± 0.01 K was used for all the measurements. The kinematic viscosity of solution γ is given by

$$\gamma = (at - (b/t)) \quad (1)$$

Where γ is the kinematic viscosity, t is the flow time, the two constants a, and b are the kinematic viscosities, γ, and densities. The uncertainty for the dynamic viscosity determination is estimated to be ± 0.5 %.

B. Standard Deviation

Standard deviation has calculated using the relationship.

$$\sigma = \left[\sum (V^{E}_{exp} - V^{E}_{cal})^2 / (N - M)\right]^{1/2} \quad (2)$$

Where, N-Number of data points
V^{E}_{exp}–Experimental Excess molar volume
V^{E}_{cal}– Calculated Excess molar volume
M – Number of coefficients

The calculated values of coefficients along with the standard deviation (σ) are given in Table 6 and Table 7. Interaction parameters and Predicted kinematic viscosities and Excess molar volume of Benzene [2, 4] and ChloroBenzene [3, 5] and 1, 4 Dioxane mixture at (303.15, 308.15 and 313.15) K are presented in Tables 2,3,4,5,6 and Table 7 respectively.

C. McAllister Model

The binary mixtures 1, 4 Dioxane + Benzene, 1, 4 Dioxane + Chloro Benzene. The calculated data of $\Delta\eta$ was correlated with the composition data by the McAllister model equation. The size ratio of the two molecules should be less than 1.5.Eyring's theory of absolute reaction rates was used to develop a model to predict the viscosity of liquid mixtures. This model, which assumes the free energy of activation for viscosity are additive on mole fraction basis.

$$\ln \nu = x_1^3 \ln \nu_1 + 3x_1^2 x_2 \ln \nu_{12} + 3x_1 x_2^2 \ln \nu_{21} + x_2^3 \ln \nu_2 - \ln(x_1 + x_2 M_2/M_1) + 3x_1^2 x_2 \ln(((2 + M_2/M_1))/3) + 3x_1 x_2^2 \ln((1 + 2M_2/M_1)/3) + x_2^3 \ln(M_2 M_1) \quad (3)$$

This McAllister equation is based on three - body model. It contains two constants namely υ_{12} and υ_{21}. The constants can be evaluated using least square method. The coefficients υ_{12} and υ_{21} of $\Delta\eta$ for all temperatures of binary liquid mixtures as shown in Table 6 and Table 7, variations of $\Delta\eta$with mole fraction $x_1$of 1,4dioxane along with the smoothed $\Delta\eta$ values calculated by using equation 3 at 303.15K to 313.15K.

III. Results and Discussion

The experimental values of excess molar volume, dynamic viscosities, η, of pure liquids 1, 4-Dioxane, Benzene, and ChloroBenzene at the investigated temperature 303.15K and compared with literature values [2, 3, 4] are shown in Table 1. The values of V^E are positive and Δη are negative for all (1, 4-Dioxane +Benzene), (1, 4-Dioxane +Chlorol Benzene) systems. The sign of excess volume, of a system depends on the relative magnitude of expansion/contraction on mixing of two liquids. For this systems expansion dominate the contraction factors, the V^E becomes positive. The viscosity deviation, Δη is negative, because of weak molecular interactions between mixtures. Excess molar volume, V^E, and viscosity deviation, Δη, Predicted Excess molar volume, kinematic *v*iscosities by Redlitch-Kister non-linear model at various temperatures and atmospheric pressure are reported in Tables 2,3,4, and 5, for the 1, 4-Dioxane +Benzene and ChloroBenzene +1, 4 Dioxane mixtures. The obtained thermo physical data shows high degree precision and given Redlitch-Kister constants and minimum standard deviations, these show in Table 6 and Table 7.

The excess molar volumes, V^E, dynamic viscosity, η, and molar refraction changes of mixing were calculated from experimental values using the following expressions.

$$V^E = V_M - \sum_{i=1}^{n} X_i V_i \quad (4)$$

Where V_M is the molar volume of the mixture, η is the dynamic viscosity, and V_i is Molar volume.

The variation of excess volumes with the mole fraction of Benzene and ChloroBenzene with 1, 4 Dioxane at (303 .15, 308.15 and 313.15) K are represented in Figure 1 and Figure 2.

Table 1 Experimental Densities and viscosities of Pure Liquids with Literature Values at 298.15K

		ρ/g·cm^{-3}		η/(m Pa s)	
Component	**T(K)**	**Lit**	**Exp**	**Lit**	**Exp**
1, 4-Dioxane	**298.15**	**1.0280**	**1.0278**	**1.1664**	**1.1662**
Benzene	**298.15**	**0.8683**	**0.8684**	**0.6300**	**0.6289**
Chloro benzene	**298.15**	**1.1000**	**1.0998**	**0.7150**	**0.7098**

Table 2 Experimental Densities and viscosities of Benzene (1) + 1,4Dioxane (2) at 303.15, 308.15 and 313.15 K

	T/K=303.15			**T/K=308.15**			**T/K=313.15**		
X1	ρ/g·cm^{-3}	η/mPa·s	V^E /cm^3·mol^{-1}	ρ/g·cm^{-3}	η/mPa·s	V^E /cm^3·mol^{-1}	ρ/g·cm^{-3}	η/mPa·s	V^E /cm^3·mol^{-1}
0.0000	**0.8649**	**0.5407**	**0.0000**	**0.8488**	**0.5221**	**0.0000**	**0.8418**	**0.5040**	**0.0000**
0.1047	**0.8788**	**0.5807**	**0.3452**	**0.8599**	**0.5608**	**0.5928**	**0.8594**	**0.5463**	**0.9541**
0.2083	**0.8943**	**0.6280**	**0.7459**	**0.8744**	**0.5963**	**1.1237**	**0.8668**	**0.5644**	**1.6854**
0.3109	**0.9196**	**0.6665**	**1.3899**	**0.8807**	**0.6185**	**1.8691**	**0.8750**	**0.5923**	**2.4962**
0.4124	**0.9306**	**0.6946**	**2.0456**	**0.8907**	**0.6420**	**2.6123**	**0.8830**	**0.6182**	**3.1244**
0.5128	**0.9428**	**0.7426**	**2.7869**	**0.9026**	**0.6698**	**3.2546**	**0.8917**	**0.6379**	**3.7015**
0.6122	**0.9640**	**0.7934**	**2.5869**	**0.9155**	**0.7072**	**3.0799**	**0.8985**	**0.6692**	**3.6013**
0.7106	**0.9740**	**0.8534**	**2.2154**	**0.9357**	**0.7604**	**2.6899**	**0.9072**	**0.6947**	**3.2570**
0.8080	**0.9971**	**0.9363**	**1.7458**	**0.9520**	**0.8282**	**2.2451**	**0.9273**	**0.7531**	**2.7245**
0.9045	**1.0090**	**1.0133**	**1.1911**	**0.9793**	**0.9147**	**1.5369**	**0.9512**	**0.8243**	**1.7895**
1.0000	**1.0271**	**1.0958**	**0.0000**	**1.0169**	**1.0094**	**0.0000**	**1.0128**	**0.9446**	**0.0000**

Table 3Experimental Densities and viscosities of Chloro Benzene (1) + 1,4Dioxane (2)at 303.15, 308.15 and 313.15 K

	T/K=303.15			**T/K=308.15**			**T/K=313.15**		
X1	ρ/g·cm^{-3}	η/mPa·s	V^E /cm^3·mol^{-1}	ρ/g·cm^{-3}	η/mPa·s	V^E /cm^3·mol^{-1}	ρ/g·cm^{-3}	η/mPa·s	V^E /cm^3·mol^{-1}
0.0000	**1.1366**	**0.7558**	**0.0000**	**1.1086**	**0.7206**	**0.0000**	**1.0822**	**0.6809**	**0.0000**
0.1047	**1.1201**	**0.7673**	**0.5604**	**1.0944**	**0.7252**	**0.5371**	**1.0710**	**0.6952**	**0.4887**
0.2083	**1.0937**	**0.7748**	**1.9872**	**1.0741**	**0.7371**	**1.6208**	**1.0618**	**0.7152**	**1.0183**
0.3109	**1.0796**	**0.7878**	**2.3061**	**1.0660**	**0.7545**	**1.5553**	**1.0551**	**0.7289**	**0.7859**
0.4124	**1.0721**	**0.8043**	**2.0160**	**1.0630**	**0.7744**	**1.4469**	**1.0495**	**0.7498**	**0.6886**
0.5128	**1.0642**	**0.8436**	**1.7489**	**1.0513**	**0.8026**	**1.2423**	**1.0449**	**0.7810**	**0.4868**
0.6122	**1.0562**	**0.8672**	**1.4765**	**1.0442**	**0.8150**	**1.0416**	**1.0393**	**0.7943**	**0.3647**
0.7106	**1.0479**	**0.8929**	**1.2159**	**1.0389**	**0.8484**	**0.6675**	**1.0339**	**0.8273**	**0.2116**
0.8080	**1.0401**	**0.9395**	**0.8978**	**1.0333**	**0.8786**	**0.3442**	**1.0268**	**0.8341**	**0.1953**
0.9045	**1.0342**	**0.9652**	**0.4037**	**1.0258**	**0.9050**	**0.1027**	**1.0210**	**0.8534**	**0.0488**
1.0000	**1.0271**	**1.0958**	**0.0000**	**1.0169**	**1.0094**	**0.0000**	**1.0100**	**0.9420**	**0.0000**

Table 4 Prediction of kinematic viscosities by Mc-Allister nonlinear model for 1, 4 Dioxane + Benzene at 303.15, 308.15 and 313.15 K

X_1	$\gamma_{expt\ (C.S)}$ T/K=303.15	γ_{pred} (c.s) (Mc-Allister) T/K=303.15	$\gamma_{expt\ (C.S)}$ T/K=308.15	γ_{pred} (c.s) (Mc-Allister) T/K=308.15	$\gamma_{expt\ (C.S)}$ T/K=313.15	γ_{pred} (c.s) (Mc-Allister) T/K=313.15
0.0000	**0.6251**	**0.6251**	**0.6151**	**0.6151**	**0.5987**	**0.5987**
0.1047	**0.6607**	**0.6623**	**0.6522**	**0.6473**	**0.6357**	**0.6303**
0.2083	**0.7022**	**0.6951**	**0.6820**	**0.6750**	**0.6512**	**0.6558**
0.3109	**0.7248**	**0.7254**	**0.7022**	**0.7000**	**0.6769**	**0.6774**
0.4124	**0.7464**	**0.7550**	**0.7208**	**0.7242**	**0.7002**	**0.6972**
0.5128	**0.7877**	**0.7861**	**0.7420**	**0.7498**	**0.7154**	**0.7177**
0.6122	**0.8230**	**0.8214**	**0.7724**	**0.7792**	**0.7448**	**0.7415**
0.7106	**0.8761**	**0.8635**	**0.8126**	**0.8148**	**0.7658**	**0.7714**
0.8080	**0.9390**	**0.9156**	**0.8699**	**0.8598**	**0.8121**	**0.8105**
0.9045	**1.0043**	**0.9818**	**0.9341**	**0.9176**	**0.8666**	**0.8628**
1.0000	**1.0669**	**1.0669**	**0.9927**	**0.9927**	**0.9327**	**0.9327**

Table 5Prediction of kinematic viscosities by Mc-Allister nonlinear model for 1, 4 Dioxane + Chloro Benzene at 303.15, 308.15 and 313.15 K

	$\gamma_{expt\ (C.S)}$ T/K=303.15	γ_{pred} (c.s) (Mc-Allister) T/K=303.15	$\gamma_{expt\ (C.S)}$ T/K=308.15	γ_{pred} (c.s) (Mc-Allister) T/K=308.15	$\gamma_{expt\ (C.S)}$ T/K=313.15	γ_{pred} (c.s) (Mc-Allister) T/K=313.15
0.0000	0.6650	0.6650	0.6500	0.6500	0.6292	0.6292
0.1047	0.6850	0.6915	0.6627	0.6726	0.6491	0.6303
0.2083	0.7084	0.7129	0.6863	0.6918	0.6736	0.6558
0.3109	0.7298	0.7333	0.7078	0.7097	0.6908	0.6774
0.4124	0.7502	0.7545	0.7285	0.7283	0.7144	0.6972
0.5128	0.7928	0.7791	0.7635	0.7498	0.7474	0.7177
0.6122	0.8211	0.8099	0.7805	0.7764	0.7643	0.7415
0.7106	0.8521	0.8501	0.8166	0.8106	0.8002	0.7714
0.8080	0.9033	0.9033	0.8503	0.8555	0.8124	0.8105
0.9045	0.9333	0.9742	0.8822	0.9147	0.8358	0.8628
1.0000	1.0669	1.0669	0.9927	0.9927	0.9327	0.9327

Table 6Prediction of viscosities by Mc-Allister nonlinear model for 1, 4 Dioxane + Benzene and1, 4 Dioxane + Chloro Benzene at 303.15, 308.15 and 313.15 K

X_1	1, 4 Dioxane + Benzene			1, 4 Dioxane + Chloro Benzene		
	Viscosity Deviation Δη (cP) T/K=303.15	Viscosity DeviationΔη (cP) T/K=308.15	Viscosity Deviation Δη (cP) T/K=313.15	Viscosity Deviation Δη (cP)T/K=303.15	Viscosity Deviation Δη (cP) T/K=308.15	Viscosity DeviationΔη (cP) T/K=313.15
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.1047	-0.0125	-0.0203	-0.0256	-0.0241	-0.0256	-0.0133
0.2083	-0.0284	-0.0436	-0.0610	-0.0519	-0.0436	-0.0206
0.3109	-0.0569	-0.0689	-0.0855	-0.0737	-0.0559	-0.0340
0.4124	-0.0840	-0.0987	-0.1152	-0.0917	-0.0653	-0.0399
0.5128	-0.1043	-0.1176	-0.1325	-0.0865	-0.0661	-0.0352
0.6122	-0.1030	-0.1133	-0.1302	-0.0967	-0.0824	-0.0480
0.7106	-0.0942	-0.1080	-0.1224	-0.1045	-0.0774	-0.0410
0.8080	-0.0717	-0.0877	-0.1069	-0.0910	-0.0753	-0.0599
0.9045	-0.0414	-0.0562	-0.0782	-0.0981	-0.0768	-0.0660
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000

Table 7:Parameters of studiedMc-Allister nonlinear model, constants and standard deviations (σ) for Viscosity Deviationof 1, 4 Dioxane + Benzene and 1, 4 Dioxane + Chloro Benzene, at 303.15, 308.15 and 313.15 K

Temperature (T/K)	υ_{12}	υ_{21}	σ
Benzene (1) + 1,4Dioxane (2)			
303.15K	0.7773	0.7655	0.0127
308.15K	0.7343	0.7370	0.0080
313.15K	0.6889	0.7215	0.0037
Chloro Benzene (1) + 1,4Dioxane (2)			
303.15K	0.7499	0.7640	0.0152
308.15K	0.7269	0.7357	0.0127
313.15K	0.7175	0.7304	0.0037

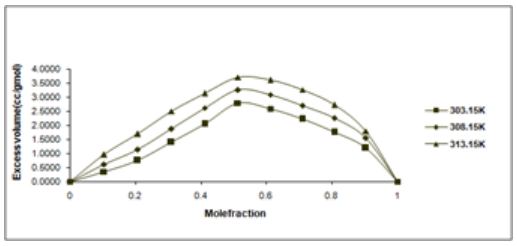

Fig. 1 Excess molar volume (V^E) for 1, 4 Dioxane + Benzene at 303.15, 308.15 and 313.15 K

The sign of excess volume of a system depends on the relative magnitude of expansion/contraction on mixing of two liquids. If the factors causing expansion dominate the contraction factors, the (V^E) becomes positive.

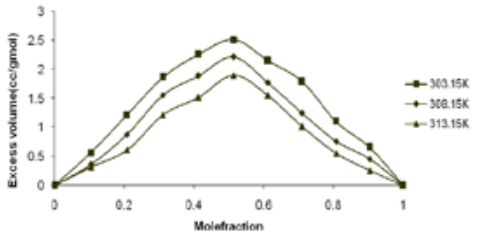

Fig. 2 Excess molar volume (V^E) for 1, 4 Dioxane + Chloro Benzene at 303.15, 308.15 and 313.15 K

On the other hand, if the contraction factors dominate the expansion factors, then V^E become negative. The factors that are responsible for expansion in volume are as follows, i. Loss of dipolar association, ii. The geometry of molecular structure, which does not allow fitting of one component into other component, iii. Steric hindrance opposes proximity of the constituent molecules. The negative V^E values arise due to dominance of the following factors. i. Chemical interaction between constituent chemicals. ii. Accommodation of molecules of one component into the interstitials of the molecules of the other component [8, 9]. iii. Geometry of the molecular structure that favors fitting of the component molecules with each other [9, 10, 12]. The negative V^E values in the mixtures under study indicate that interactions between molecules of the mixtures are stronger than interactions between molecules in the pure liquids and that associative force dominate the behavior of the solution.

$$\Delta\eta = \eta - \sum_{i=1}^{n} x_i \eta_i \qquad (5)$$

Where (Δη) is the viscosity deviation of the mixture, η is the dynamic viscosity.

The results of variation in viscosity deviations of binary systems consisting of Benzene and ChloroBenzene with 1, 4 Dioxane at temperatures of 303.15K, 308.15K, and 313.15K are represented in figure 3 and 4.

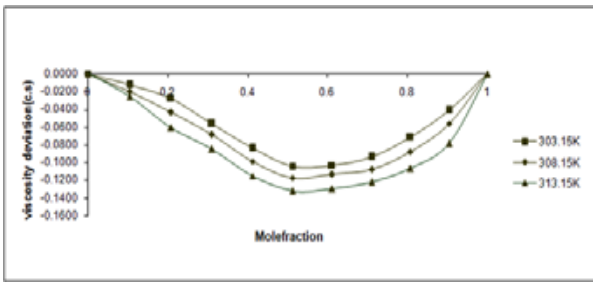

Fig. 3 Deviation in viscosity (Δη) for 1,4 Dioxane + Benzene at 303.15, 308.15 and 313.15 K

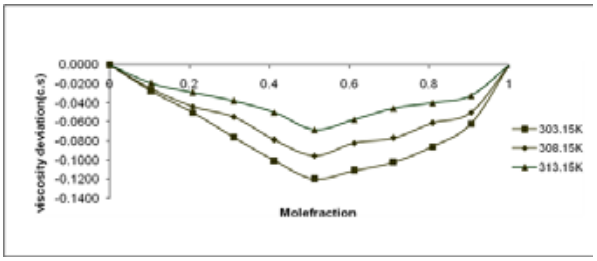

Fig. 4 Deviation in viscosity (Δη) for 1,4Dioxane + Chloro Benzene at 303.15, 308.15 and 313.15 K

This result shows negative deviations [6, 7, 11, 12] over the entire range of mole fraction. The viscosity of the mixture strongly depends on the entropy of mixture, which is related with liquid's structure and enthalpy. It will consequently with molecular interactions between the components of the mixtures. Therefore, the viscosity deviation depends on molecular interactions as well as on the size and shape of the molecules.

IV. CONCLUSION

From the study, 1, 4 Dioxane is repulse towards Benzene and it forms dipole-dipole bond. Inductive effect of Chlorine group in Chlorobenzene is donating electron, due to this Chlorine group becomes slightly positive and at the same time phenyl group becomes negative, this makes the compound to feebly dipolar. In this case, the force between unlike molecules is lesser than the force between like molecules in mixtures. The structural contributions are mostly negative and arise from interstitial accommodation and changes in free volume. The actual volume changed therefore depends on the relative strength of these effects. It can be concluded that the positive Excess molar volumes and negative deviations viscosity due to weak molecular interactions existing between the binary mixtures of 1, 4 Dioxane + Benzene + Chloro Benzene. Viscosity of the binary mixture and the McAllister three-body model is very well suited for correlating Kinematic viscosity of the binary mixture with minimum standard deviation in present study.

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