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Application of Redlich-Kister Modelto Measure and Predict Thermo Physical Properties of Binary Liquid Mixtures at Various Temperatures

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Abstract*— Study conducts in this research are Viscosities,η, and densities, of 1, 4-dioxane with Benzene at different mole fractions and various temperatures in the atmospheric pressure condition. From experimentations excess volumes, V^E,and deviation sin viscosities,Δη, of mixtures at infinite dilutions have been obtained. The measured systems exhibited positive values of V_m^E and negative values of Δη. The binary mixture 1, 4 dioxane + Benzene show positive V^E and negative Δη with increasing temperatures. The outcomes clearly indicate that weak interactions present in mixture. It is mainly because of number and position of methyl groups exist in these aromatic hydrocarbons. These measured data tailored to the non-linear models to derive the binary coefficients. Standard deviations areconsidered between the fitted outcomes and the calculated data is helpful deliberate mixing behavior of the binary mixtures. It can conclude that in our cases, the data found with the values correlated by the corresponding models very well. The molecular interactions existing between the components and comparison of liquid mixtures also discussed.

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

I. INTRODUCTION

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 act 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 our 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 have been fitted to the Redlich-Kister and polynomial equations. The Redlich-Kister was used to correlate the excess volumes, V^E, and deviations in viscosities, Δη, this analysis technique was used to derive the binary coefficients, estimated the standard deviation (σ) between the experimental and calculated data [4].The variation ofthese parameters with the composition and temperature of the mixtures has been discussed in terms of the molecular interactions in these mixtures. The effect of the number and position of the methyl groups in these aromatic hydrocarbons on molecular interactions in these mixtures has also been discussed. A literature search showed that no measurements have been previously reported by using Redlich-Kister 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].

B. 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 ($2\cdot10^{-3}$ 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 to flow time measurements. Experiments 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

$$\gamma = (at - (b/t)) \qquad \textbf{(1)}$$

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 %.

C. Standard Deviation

Standard deviation has calculated using the relationship.

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

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 Chloro Benzene [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.

D. Non-linear Model

The binary mixtures are 1, 4 Dioxane + Benzene, 1, 4 Dioxane + Chloro Benzene. The calculated data of V^E, $\Delta\eta$ were correlated with the composition data by the Redlitch-Kister equation. The size ratio of the two molecules should be less than 1.5.

$$V^E = x_1 x_2 \sum a_{i-1} (x_1 - x_2)^{i-1} \qquad (3)$$

In case of V^E, excess molar volume is replaced by $\Delta\eta$in equation 3. The values of coefficients, A_i, were evaluated by using the method of least-squares with all points weighted equally. The coefficients A_1 to A_7 of $\Delta\eta$ and V^E for all temperatures of binary liquid mixtures as shown in Table 6 and Table 7, variations of $\Delta\eta$ and V^E with mole fraction $x_1$of 1,4dioxane along with the smoothed $\Delta\eta$ and V^E 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 Table 5,for the 1, 4-Dioxane +Benzene and ChloroBenzene +1, 4 Dioxane mixtures. The obtained thermo physical data shows high degree precision and is given Redlitch-Kister constants and minimum standard of deviations, as shown in Table 6 and Table 7.

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 3 Experimental 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 Excess molar volume, kinematic viscosities by Redlitch-Kister nonlinear model for 1, 4 Dioxane + Benzene at 303.15, 308.15 and 313.15 K

X_1	V^E (pred)(cc/gmole) (R-Kister) T/K=303.15	Δη (pred) (RKister) T/K=303.15	V^E (pred)(cc/gmole) (R-Kister) T/K=308.15	Δη(pred) (RKister) T/K=308.15	V^E (pred)(cc/gmole) (R-Kister) T/K=313.15	Δη(pred) (RKister) T/K=313.15
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.1047	0.5928	-0.0152	0.5003	-0.0123	-0.0022	-0.0067
0.2083	0.8083	-0.0304	0.9202	-0.0280	0.5167	-0.0223
0.3109	1.8691	-0.0469	1.6707	-0.0525	1.8909	-0.0502
0.4124	2.5191	-0.0689	2.5929	-0.0830	3.1593	-0.0776
0.5128	2.9542	-0.0854	3.0484	-0.1044	3.6501	-0.0910
0.6122	3.2751	-0.0876	2.9991	-0.1106	3.9576	-0.0986
0.7106	2.8697	-0.0768	2.9580	-0.1067	4.9098	-0.1142
0.8080	2.8574	-0.0566	2.9038	-0.0902	5.6370	-0.1208
0.9045	1.8379	-0.0265	1.7546	-0.0474	3.5359	-0.0737
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000

Table 5:Prediction of Excess molar volume, kinematic viscosities by Redlitch-Kister nonlinear model for 1, 4 Dioxane + Chloro Benzene at 303.15, 308.15 and 313.15 K

X_1	V^E (pred)(cc/gmole) (R-Kister) T/K=303.15	Δη (pred) (RKister) T/K=303.15	V^E (pred)(cc/gmole) (R-Kister) T/K=308.15	Δη(pred) (RKister) T/K=308.15	V^E (pred)(cc/gmole) (R-Kister) T/K=313.15	Δη(pred) (RKister) T/K=313.15
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.1047	0.6155	-0.0275	0.5939	-0.0280	0.0916	-0.0151
0.2083	1.8173	-0.0445	1.4300	-0.0385	0.9841	-0.0172
0.3109	2.4007	-0.0705	1.7156	-0.0516	1.2546	-0.0292
0.4124	2.1594	-0.0999	1.5077	-0.0736	1.1503	-0.0472
0.5128	1.6546	-0.1003	1.1811	-0.0777	1.0232	-0.0453
0.6122	1.3692	-0.0813	0.9418	-0.0656	0.7853	-0.0308
0.7106	1.2613	-0.0869	0.7190	-0.0697	0.6366	-0.0390
0.8080	0.9633	-0.1168	0.3926	-0.0923	0.5709	-0.0710
0.9045	0.3693	-0.0897	0.0745	-0.0710	0.1701	-0.0618
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000

Table 6Parameters of studiedRedlitch-Kister nonlinear model, constants and standard deviations (σ) for Excess Volume **of 1, 4 Dioxane + Benzene and 1, 4 Dioxane + Chloro Benzene, at 303.15, 308.15 and 313.15 K**

Temperature (T/K) T/K	A_1	A_2	A_3	A_4	A_5	A_6	A_7	σ
Benzene (1) + 1,4Dioxane (2)								
303.15	-56.509	-27.653	66.619	26.913	-15.736	0.7609	5.6556	0.2113
308.15	-67.92	-34.501	74.331	30.247	- 18.49	4.2666	12.104	0.2120
313.15	-87.369	- 102.92	76.667	98.355	- 3.6776	5.0706	14.481	0.8709
Chloro Benzene (1) + 1,4Dioxane (2)								
303.15	19.75	-9.324	-44.93	18.79	18.32	-9.362	6.85	0.2217
308.15	11.15	4.818	- 26.09	1.381	10.04	6.102	4.877	0.1487
313.15	-87.369	- 102.9	76.667	98.355	-3.677	5.0706	15.481	0.1391

Table 7Parameters of studiedRedlitch-Kister nonlinear model, constants and standard deviations (σ) for Viscosity Deviation**of 1, 4 Dioxane + Benzene and 1, 4 Dioxane + Chloro Benzene, at 303.15, 308.15 and 313.15 K**

Temperature (T/K)	A_1	A_2	A_3	A_4	A_5	A_6	A_7	σ
Benzene (1) + 1,4Dioxane (2)								
303.15	1.328	0.0171	- 1.5746	0.2169	0.5935	- 0.2341	-0.3483	0.005
308.15	1.3354	0.6588	- 1.3364	- 0.3553	0.4108	-0.3047	- 0.4107	0.002
313.15	1.842	1.8177	- 1.5655	- 1.6702	0.0816	- 0.1671	-0.3602	0.011
Chloro Benzene (1) + 1,4Dioxane (2)								
303.15	4.434	2.343	- 5.013	2.612	0.981	0.255	-0.408	0.020
308.15	3.841	1.527	-4.342	- 1.654	0.811	0.116	-0.314	0.015
313.15	3.451	1.8	- 4.045	- 2.032	0.778	0.224	- 0.187	0.012

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.

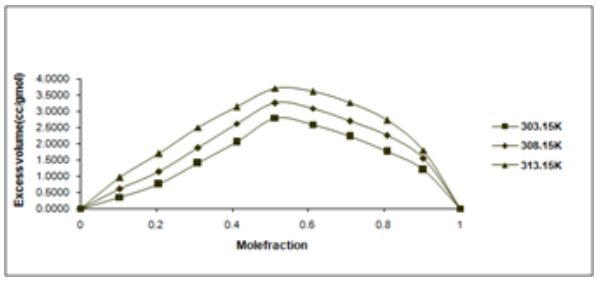

Fig.1. Excess molar volume (V^E) for 1, 4 Dioxane + Benzene at 303.15, 308.15 and 313.15 K

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.

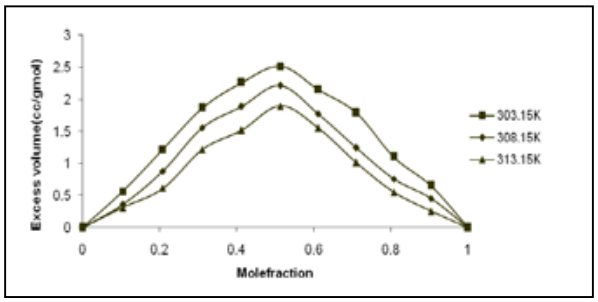

Fig.2. Excess molar volume (V^E) for 1, 4 Dioxane + Chloro Benzene at 303.15, 308.15 and 313.15 K

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 indicates 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.

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

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 Chloro Benzene with 1, 4 Dioxane at temperatures of 303.15K, 308.15K, and 313.15K are represented in figure 3 and 4.

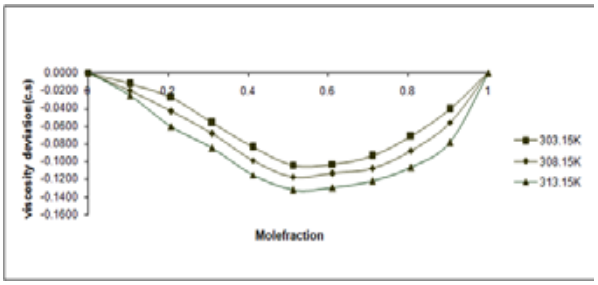

Fig.3. Deviation in viscosity (Δη) for 1,4 Dioxane + Benzene at 303.15, 308.15 and 313.15 K

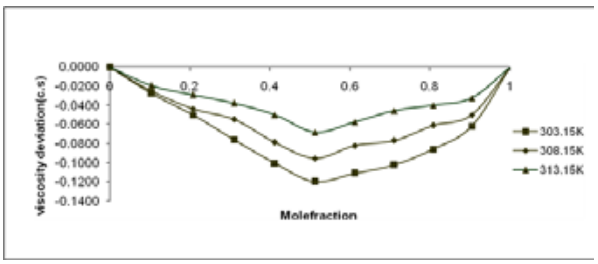

Figure 4. Deviation in viscosity (Δη) for 1,4Dioxane + Chloro Benzene at 303.15, 308.15 and 313.15 K

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 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

In the present study of Viscosities (η) and Excess molar volume (V^E), for the binary liquid mixture of 1, 4 Dioxane + Benzene + ChloroBenzene system was found as a function of mole fraction at atmospheric pressure and at temperatures of 303.15K, 308.15K, and 313.15K. The excess values of the mixtures show a systematic change with increasing temperature. With an increase in temperature, the intermolecular interactions between molecules become weak. 1, 4 Dioxane is repulse towards the Benzenering in Benzene and it forms dipole-dipole bond .In this case, the force between unlike molecules is lesser than the force between like molecules

in mixtures. It can be concluded that the positive excess molar volumes and negative deviations viscosity due to weak molecular interactions in mixtures. Viscosity and density of the binary mixture and the Redlitch-Kister model is very well suited for correlating Kinematic viscosity and Excess molar volumes of the binary mixture with minimum standard deviation in present study and it is far superior to the other predictive models

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