

COMPARATIVE ANALYSIS OF EXPERIMENTAL AND NUMERICAL INVESTIGATION ON THERMOPHYSICAL PROPERTIES IN HYDROCARBON MIXTURES USING JOUYBAN-ACREE MODEL AT VARIOUS TEMPERATURES

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ABSTRACT

The thermophysical properties of liquid mixtures provide additional information regarding molecular interactions. A perusal of the literature revealed that the predictions of thermophysical properties of liquid mixtures are scarce. With an aim, the thermophysical properties of viscosity, excess molar volume V^E and viscosity deviations $\Delta \eta$ of liquid mixtures are predicted by using various nonlinear models. Inthis research Jouyban-Acree viscosity models have been used for predicting viscosity of Acetophenone with P-xylene and 1, 4 Dioxane with Benzene at different mole fractions measured at various temperatures in the atmospheric pressure condition. From experimentation excess volumes, V^E , and deviations in viscosities, $\Delta \eta$, of mixtures at infinite dilutions have been obtained. The measured systems show positive V^E and negative $\Delta \eta$ with increasing temperatures.From the positive excess molar volume, when aromatics, which exist in a highly associated form in the pure state, are mixed with polar solvents (ketones), the monomerization occurs and new specific interactions appear in the solution. The negative viscosity deviation depends on the size and shape of the molecules and molecular interactions. These measured data tailored to the Jouyban-Acree nonlinear models to derive the binary coefficients Jouyban-Acree model is more adequate for the thermo physical and the standard deviation was found to be <2.06 %. The molecular interactions existing between the components and comparison of liquid mixtures were also discussed.

Keywords: acetophenone, 1, 4 dioxane, viscosity measurement, Jouyban- Acree model.

INTRODUCTION

The quantitative viscosity 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 (Martin, 2001 and Ramesh et al., 2014). In our earlier paper, we had studied thermo physical properties of binary systems (Ramesh et al., 2014). In the present paper, it has been reported density (ρ) and viscosity (η) of pure 1, 4 Dioxane, Acetophenone, P-xylene and Benzenefor the binary system constituted by these two chemicals at entire range of composition and temperature 303.15K to 313.15K. With this data, the excess molar volume and deviation in viscosity have been computed. These results have been fitted to the Jouyban-Acree models and polynomial equations. The Jouyban-Acree model was used to correlate the viscosity and deviations in viscosities, $\Delta \eta$, this analysis

technique was used to derive the binary coefficients, estimated the standard deviation (σ) between the experimental and calculated data (Jouyban *et al.*, 2005). The variation of these 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 Jouyban-Acree models for the mixtures studied in this paper.

EXPERIMENTAL SECTION

Materials

1, 4-Dioxane, Acetophenone, P-xylene and Benzene this were all supplied by Merck Ltd. With the stated purities better than 99%, were stored over molecular sieves (0.3 nm). To minimize the contact of these reagents with moist air, the products were kept in sealed bottles in a desiccator. The densities and viscosities of pure substances and experimental values comparison with literature values are listed in Table-1 (Martin, 2001; Ramesh *et al.*, 2014; Jouyban *et al.*, 2005; Baskaran, 2012; Laura, 2012 and Redlich, 1948).

	ρ (g·	cm ⁻³)	η (m Pa. s)		
Component	Literature	Experimental	Literature	Experimental	
1, 4-Dioxane	1.0222	1.0271	1.0985	1.0958	
Benzene	0.8660	0.8649	0.6255	0.6251	
Acetophenone	1.0199	1.0087	1.518	1.514	
P-xylene	0.85225	0.9540	0.1670	0.1667	

Table-1. Experimental Densities and viscosities of Pure Liquids with Literature Values at 303.15K.

Apparatus and procedure

In 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 to 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 0.002 kg·m ³. 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 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, n, after multiplication by the density. The reproducibility of dynamic viscosity was found to be within 0.003 mPa•s. A thermostatically controlled, wellstirred 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 maintained within ± 0.01 K, was used for all the measurements. The kinematic viscosity of solution γ is given by

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$$\gamma = \mathbf{a}(\mathbf{t}) - \left(\frac{b}{t}\right) \tag{1}$$

- = kinematic viscosity, γ
- t = flow time,
- = constant а
- constant b =

The uncertainty for the dynamic viscosity determination is estimated to be ± 0.5 %.

Standard deviation

It is calculated using the relationship.

$$\sigma = \sum \left(\frac{\left(V_{ex} \cdot V_{ca}\right)^2}{N \cdot P}\right)^{\frac{1}{2}}$$
(2)

N = Number of data points

Experimental Excess molar volume $V_{ex} =$

Calculated Excess molar volume $V_{ca} =$

Р = Number of coefficients

The calculated values of coefficients along with the standard deviation (σ) are given in Table 10 and 11. Interaction parameters and Predicted kinematic viscosities and Excess molar volume of Benzene (Ramesh et al., 2014, Jouyban et al., 2005, Senthil, 2004, Fabio, 2006, Francesconi, 2005 and Redlich, 1948) and P-xylene (Ramesh et al., 2014, Jouyban et al., 2005 and Baskaran and Kubendran, 2007) and 1, 4 Dioxaneand at 303.15K. Acetophenonemixtures 308.15Kand 313.15Kare presented in Tables 2, 3, 4, 5, 6, 7, 8 and 9.

Nonlinear Model

The binary mixtures Acetophenone with P-xylene and 1, 4 Dioxane with Benzene. The calculated data of Viscosity, $\Delta \eta$ were correlated with the composition data by the Jouyban Acree equation. The size ratio of the two molecules should be less than 1.5.

$$\ln v_{\rm m} = x_1 \, \ln v_1 + \, x_2 \ln v_2 + \left(\frac{x_1 x_2}{T}\right) \sum a_i \left(x_1 - x_2\right)^i \quad (3)$$

 v_m = viscosity of mixture,

= viscosity of component one, v_1

= viscosity of component two, v_2

= constant ai

The values of coefficients, ai, were evaluated by using the method of least-squares with all points weighted equally.



<i>x</i> ₁	$\gamma_{exp}(c.s)$	ρ (g. cm ⁻³)	η (mpa.s)	$V^E(cc.gmol^{-1})$	$\Delta \eta$ (mpa.s)
0.0000	0.6251	0.8649	0.5407	0.0000	0.0000
0.1047	0.6607	0.8788	0.5807	0.3452	-0.0125
0.2083	0.7022	0.8943	0.6280	0.7459	-0.0284
0.3109	0.7248	0.9196	0.6665	1.3899	-0.0569
0.4124	0.7464	0.9306	0.6946	2.0456	-0.0840
0.5128	0.7877	0.9428	0.7426	2.7869	-0.1043
0.6122	0.8230	0.9640	0.7934	2.5869	-0.1030
0.7106	0.8761	0.9740	0.8534	2.2154	-0.0942
0.8080	0.9390	0.9971	0.9363	1.7458	-0.0717
0.9045	1.0043	1.0090	1.0133	1.1911	-0.0414
1.0000	1.0669	1.0271	1.0958	0.0000	0.0000

Table-2. Experimental densities and viscosities of 1, 4 Dioxane + Benzene at 303.15K.

Table-3. Experimental Densities and viscosities of1, 4 Dioxane + Benzene at 308.15K.

<i>x</i> ₁	$\gamma_{exp}(c.s)$	$\rho(g.cm^{-3})$	$\boldsymbol{\eta}$ (mpa.s)	$V^E(cc.gmol^{-1})$	$\Delta \eta$ (mpa.s)
0.0000	0.6151	0.8488	0.5221	0.0000	0.0000
0.1047	0.6522	0.8599	0.5608	0.5928	-0.0203
0.2083	0.6820	0.8744	0.5963	1.1237	-0.0436
0.3109	0.7022	0.8807	0.6185	1.8691	-0.0689
0.4124	0.7208	0.8907	0.6420	2.6123	-0.0987
0.5128	0.7420	0.9026	0.6698	3.2546	-0.1176
0.6122	0.7724	0.9155	0.7072	3.0799	-0.1133
0.7106	0.8126	0.9357	0.7604	2.6899	-0.1080
0.8080	0.8699	0.9520	0.8282	2.2451	-0.0877
0.9045	0.9341	0.9793	0.9147	1.5369	-0.0562
1.0000	0.9927	1.0169	1.0094	0.0000	0.0000

Table-4. Experimental densities and viscosities of1, 4 Dioxane + Benzene at 313.15K.

$x_1 \qquad \gamma_{exp}(c.s)$		ρ (g. cm ⁻³)	η (mpa.s)	$V^{E}(cc.gmol^{-1})$	$\Delta \eta$ (mpa.s)
0.0000	0.5987	0.8418	0.5040	0.0000	0.0000
0.1047	0.6357	0.8594	0.5463	0.9541	-0.0256
0.2083	0.6512	0.8668	0.5644	1.6854	-0.0610
0.3109	0.6769	0.8750	0.5923	2.4962	-0.0855
0.4124	0.7002	0.8830	0.6182	3.1244	-0.1152
0.5128	0.7154	0.8917	0.6379	3.7015	-0.1325
0.6122	0.7448	0.8985	0.6692	3.6013	-0.1302
0.7106	0.7658	0.9072	0.6947	3.2570	-0.1224
0.8080	0.8121	0.9273	0.7531	2.7245	-0.1069
0.9045	0.8666	0.9512	0.8243	1.7895	-0.0782
1.0000	0.9327	1.0128	0.9446	0.0000	0.0000

	303.15K		308.	308.15K		.15K
<i>x</i> ₁	$\gamma_{exp}(c.s)$	$\boldsymbol{\gamma_{pred}}(c.s)$	$\gamma_{exp}(c.s)$	$\gamma_{pred}(c.s)$	$\gamma_{exp}(c.s)$	$\gamma_{pred}(c.s)$
0.0000	0.6251	0.6251	0.6151	0.6151	0.5987	0.5987
0.1047	0.6607	0.3475	0.6522	0.6521	0.6357	0.9449
0.2083	0.7022	0.3411	0.6820	0.6822	0.6512	1.0066
0.3109	0.7248 0.	0.4466	0.7022	0.7023	0.6769	0.8497
0.4124	0.7464	0.4602	0.7208	0.7203	0.7002	0.8488
0.5128	0.7877	0.4459	0.7420	0.7422	0.7154	0.9374
0.6122	0.8230	0.5082	0.7724	0.7721	0.7448	0.9487
0.7106	0.8761	0.5995	0.8126	0.8136	0.7658	0.8908
0.8080	0.9390	0.6146	0.8699	0.8689	0.8121	0.8946
0.9045	1.0043	0.7100	0.9341	0.9344	0.8666	0.9480
1.0000	1.0669	1.0669	0.9927	0.9927	0.9327	0.9327

Table-5. Prediction of viscosities Jouyban- Acree nonlinear model for 1, 4 Dioxane + Benzeneat 303.15K, 308.15K and 313.15K.

Table-6. Experimental Densities and viscosities of Acetophenone + P-xylene at 303.15K.

<i>x</i> ₁	$\gamma_{exp}(c.s)$	$\rho(g. cm^{-3})$	η (mpa.s)	$V^E(cc.gmol^{-1})$	$\Delta \eta$ (mpa.s)
0.0000	0.1696	1.0087	0.1711	0.0000	0.0000
0.0991	0.1694	1.0073	0.1706	0.2104	-0.0255
0.1896	0.1689	1.0072	0.1701	0.2873	-0.0785
0.2946	0.1685	1.0064	0.1696	0.4402	-0.0785
0.3642	0.1700	0.9962	0.1694	0.4413	-1.0979
0.4424	0.1698	0.9942	0.1688	0.4491	-1.2097
0.5956	0.1707	0.9868	0.1684	0.4911	-1.0478
0.6896	0.1710	0.9823	0.1680	0.3790	-0.0460
0.7956	0.1717	0.9752	0.1674	0.1381	-0.0576
0.8976	0.1730	0.9650	0.1669	0.0924	-0.0421
1.0000	0.1747	0.9539	0.1666	0.0000	0.0000

Table-7. Experimental densities and viscosities of Acetophenone + P-xylene at 308.15K.

<i>x</i> ₁	$\gamma_{exp}(c.s)$	$\boldsymbol{\rho}(g.cm^{-3})$	η (mpa.s)	$V^E(cc.gmol^{-1})$	$\Delta \eta$ (<i>mpa.s</i>)
0.0000	0.1676	1.0086	0.1690	0.0000	0.0000
0.0991	0.1682	1.0072	0.1694	0.3238	-0.3245
0.1896	0.1677	1.0071	0.1689	0.4010	-0.4065
0.2946	46 0.1675 1.0 42 0.1689 0.9	1.0063	0.1686	0.4194	-0.4170
0.3642		0.9960	0.1682	0.4266	-0.9532
0.4424	0.1687	0.9941	0.1677	0.3490	-1.0175
0.5956	0.1695	0.9862	0.1672	0.3539	-0.8685
0.6896	0.1699	0.9820	0.1668	0.3436	-0.2199
0.7956	0.1706	0.9751	0.1664	0.2914	-0.2225
0.8976	0.1720	0.9650	0.1660	0.2088	-0.2066
1.0000	0.1738	0.9530	0.1656	0.0000	0.0000



<i>x</i> ₁	$\gamma_{exp}(c.s)$	$\boldsymbol{\rho}(g.cm^{-3})$	η (mpa.s)	$V^E(cc.gmol^{-1})$	$\Delta \eta$ (mpa.s)
0.0000	0.1667	1.0075	0.1679	0.0000	0.0000
0.0991	0.1675	1.0061	0.1685	0.3118	-0.4456
0.1896	0.1670	1.0060	0.1680	0.4180	-0.4356
0.2946	0.1667	1.0052	0.1676	0.4154	-0.4223
0.3642	0.1680	0.9950	0.1672	0.4356	-0.8321
0.4424	0.1679	0.9930	0.1667	0.3190	-0.9196
0.5956	0.1687	0.9857	0.1663	0.3229	-0.8185
0.6896	0.1691	0.9811	0.1659	0.2946	-0.2145
0.7956	0.1698	0.9741	0.1654	0.2644	-0.2178
0.8976	0.1713	0.9639	0.1651	0.2008	-0.2115
1.0000	0.1726	0.9528	0.1645	0.0000	0.0141

Table-8. Experimental Densities and viscosities of Acetophenone + P-xylene at 313.15K.

Table-9. Prediction of viscosities Jouyban- Acree nonlinear model for Acetophenone + P-xyleneat 303.15K, 308.15K and 313.15K.

	303.15K		308.	308.15K		313.15K	
<i>x</i> ₁	$\gamma_{exp}(c.s)$	$\gamma_{pred}(c.s)$	$\gamma_{exp}(c.s)$	$\gamma_{pred}(c.s)$	$\gamma_{exp}(c.s)$	$\gamma_{pred}(c.s)$	
0.0000	0.1696	0.1696	0.1676	0.1676	0.1667	0.1667	
0.0991	0.1694	0.1684	0.1682	0.1687	0.1675	0.1573	
0.1896	0.1689	0.1669	0.1677	0.1667	0.1670	0.1547	
0.2946	0.1685	0.1575	0.1675	0.1585	0.1667	0.1555	
0.3642	42 0.1700	0.1490	0.1689	0.1493	0.1680	0.1463	
0.4424	0.1698	0.14881	0.1687	0.1498	0.1679	0.1498	
0.5956	0.1707	0.1566	0.1695	0.1502	0.1687	0.1576	
0.6896	0.1710	0.1700	0.1699	0.1653	0.1691	0.1595	
0.7956	0.1717	0.1707	0.1706	0.1709	0.1698	0.1607	
0.8976	0.1730	0.1720	0.1720	0.1718	0.1713	0.1652	
1.0000	0.1747	0.1747	0.1738	0.1737	0.1726	0.1726	

The sign of excess volume, of a system depends on the relative magnitude of expansion/contraction on mixing of two liquids. The variation of viscosity deviation, $\Delta\eta$,with the molfraction of Benzene +1,4Dioxane and Acetophenone + P-xylinewas studied at 303.15K, 308.15Kand 313.15K. The obtained thermo physical data shows high degree precision and it is given Jouyban-Acreeconstants and minimum standard deviations, these are shown in Table-10 and Table-11.

The excess molar volumes, V^E , dynamic viscosity, η , and molar refraction changes of mixing were

calculated from experimental values using the following expressions.

$$\mathbf{v}^{\mathrm{E}} = \mathbf{v}_{\mathrm{M}} \sum_{i=1}^{n} \mathbf{X}_{i} \mathbf{v}_{i}(4)$$

 $v_M = \text{molar volume of the mixture},$

 v_i = Molar volume of components,

The variation of excess volumes with the mole fraction of Benzene + 1, 4 Dioxane ,Acetophenone + P-xylene with at 303.15K, 308.15K and 313.15K are represented in Figure-1 and Figure-2.

(σ) of 1, 4 Dioxane + Benzene at 303.15K, 308.15K and 313.15K.									
Temperature	a ₁	\mathbf{a}_2	a ₃	\mathbf{a}_4	\mathbf{a}_5	a ₆	\mathbf{a}_7	σ	
303.15K	11694	-2606.2	- 13119	2844.8	2156	-229.06	-738.22	0.4590	
308.15K	-65.449	78.811	58.256	13.025	75.979	-91.992	- 68.85	0.0008	
313.15K	-6087.9	3534.4	6983.1	3818.1	-1164.5	266.98	271.41	0.3138	

Table-10. Parameters of studiedJouyban-Acree nonlinear model, constants and standard deviations (σ) of 1, 4 Dioxane + Benzene at 303.15K, 308.15K and 313.15K.

Table-11. Parameters of studiedJouyban- Acree nonlinear model, constants and standard deviations (σ) of Acetophenone + P-xylene at 303.15K, 308.15K and 313.15K

Temperature	a ₁	\mathbf{a}_2	a ₃	\mathbf{a}_4	σ
303.15K	220.66	-201.16	69.492	205.61	0.0235
308.15K	225.34	-200.09	68.558	206.33	0.0263
313.15K	227.34	-201.18	69.602	207.235	0.0274



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



Figure-2. Excess molar volume (V^E) for Acetophenone + P-xyleneat 303K.15, 308.15K and 313.15K.

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. 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 (Fabio, 2006, Francesconi, 2005 and Senthil Raja *et al.*, 2004). iii. Geometry of the molecular structure that favors fitting of the component molecules with each other (Francesconi, 2005 and Hiannie, 2005). 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 behaviour of the solution.

$$\Delta \eta = \eta - \sum_{i=1}^{n} X_i \eta_i \tag{5}$$



Where, $(\Delta \eta)$ 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+1, 4 Dioxane and Acetophenone+P-xylene at temperatures of 303.15K, 308.15K, and 313.15K are represented in Figure 3 and 4.



Figure-3. Deviation in viscosity $(\Delta \eta)$ for 1, 4 Dioxane + Benzene at 303.15K, 308.15K and 313.15 K.



Figure-4. Deviation in viscosity ($\Delta\eta$) for Acetophenone + P-xyleneat 303.15K, 308.15K and 313.15K.

This result shows negative deviations (Senthil Raja *et al.*, 2004, Baskaran, 2007) 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.

CONCLUSIONS

In this research, Viscosities (η) and densities (ρ) for the binary liquid mixture of 1, 4 Dioxane + Benzene + Acetophenone + P-xylenesystems were found out 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. From the positive excess molar volume, when aromatics, which exist in a highly associated form in the pure state, are mixed with polar solvents (ketones), the monomerization occurs and new specific interactions appear in the solution. 1, 4 Dioxane is repulse towards the Benzene group 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. The deviation in viscosity depends on the size and shape of the molecules and molecular interactions. Without strong specific interactions, the viscosity deviations are negative. The viscosity deviations become less negative due to the increase of thermal energy. 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 Jouyban- Acree Modelis very well suited for correlating Kinematic viscosity of the binary mixture with minimum standard deviation of < 2.06% in this study and Acetophenone + P-xylene system shows high accuracy in viscosity prediction with Jouyban-Acreemodel. Itis far superior to the other predictive 1, 4 Dioxane + benzene binary mixture.

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