

Correlative and predictive models of viscosity study on Propiophenone with isomeric xylenes binary mixtures at $T=(303.15 \text{ to } 318.15) \text{ K}$

Dittakavi Ramachandran^{*,†}, Nanduri Gayatri Devi^{*}, and Nanduri Veera Naga Bala Srinivasa Rao^{**}

^{*}Acharya Nagarjuna University, Guntur, India

^{**}DRG Government Degree College, Tadepalligudem, India

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Abstract—Densities and viscosities of binary mixtures of Propiophenone with *o*-xylene, *m*-xylene and *p*-xylene were measured over the entire composition range at $T=(303.15 \text{ to } 318.15) \text{ K}$ and at 0.1 Mpa atmospheric pressure. Experimental data used to calculate excess molar volume (V^E), deviation of viscosity ($\Delta\eta$), excess Gibbs free energy (G^E) activation of viscous flow for each binary system and the results were fitted to the Redlich-Kister polynomial equation to obtain the fitting coefficients and standard deviations. Viscosity values used to compute single adjustable interaction parameters from Grunberg and Nissan, Katti and Chaudhri, Hind et al., Tamara Kurata and Frenkel relations. Deviations in thermodynamic properties of the binary mixtures were discussed in terms of their molecular interactions between the components. Viscosity data correlated with the McAllister's three body/four body models, Heric, Auslander, and Jouyban-Acree relations having two and three adjustable parameters for the studied binary mixtures. Viscosity relations like Kendall-Monroe, Bingham, Arrhenius, and Kendall were used to calculate and compare the standard deviation percentage, ($\sigma \%$), between the experimental and calculated viscosity data. The studied systems showed specific intermolecular interactions and the percentage deviations were in good agreement with the experimental values. Obtained results are useful in various chemical and industrial processes.

Keywords: Viscosity, Excess Molar Volume, McAllister's Models, Jouyban-Acree Relation, Auslander Equation, Viscosity Relations

INTRODUCTION

Physical properties of liquid mixtures such as density (ρ) and viscosity (η) are important in designing, engineering and chemical processes. Thermodynamic properties of binary mixtures are essential to understand the intermolecular interactions of the liquid mixtures. Of these, viscosity of binary mixtures is vital for hydraulic calculations of fluid transport and for energy transference calculations in the chemical, foodstuff, cosmetic and pharmaceutical industries. Current study is on viscosity analysis on the binary mixtures of propiophenone with isomeric xylenes. Propiophenone is used as a component in some perfumes and as an intermediate in the synthesis of pharmaceutical and other organic compounds [1]. Xylenes with other components are used as chemical intermediates, and xylenes are also used as solvent in leather, rubber, dyes, pesticides, printing industries etc. [2]. Hence, the thermodynamic properties of the studied pure liquids and liquid mixtures are quite essential for the industrial processes.

Literature survey reveals studies on the measurement of densities, viscosities, ultrasonic speeds, relative permittivity etc., of binary mixtures containing xylenes with other components at different temperatures [3-9]. Apparently no study has been reported on the viscosity model studies on propiophenone with *o*-xylene, *m*-xylene and *p*-xylene binary mixtures. The studied liquid mixtures are very

much useful in designing and chemical processes.

Densities ρ and viscosities η , of binary liquid mixtures of propiophenone with *o*-xylene, *m*-xylene and *p*-xylene were measured over the entire composition range at $T=(303.15\text{-}318.15) \text{ K}$. Experimental data were used to compute excess molar volume (V^E), deviation of viscosity ($\Delta\eta$), one interaction parameters of Grunberg and Nissan (d_{12}), excess Gibbs free energy of activation (G^E), Katti and Chaudhary (W_{vis}/RT), Hind et al., (H_{12}), Tamara-Kurata (T_{12}) and Frenkel (F_{12}) for the studied binary mixtures and also correlated with McAllister's three/four body models, Heric, Auslander and Jouyban-Acree relations for two and three adjustable parameters to understand more of the intermolecular interactions between the component molecules of the binary mixtures. Zero adjustable parameter relations such as Kendall and Monroe, Bingham, Arrhenius, and Kendall were used to correlate the experimental viscosity data and to calculate the standard deviation percentages. Obtained results were in good agreement with the experimental data. Deviations in excess properties of these binary mixtures at all four temperatures reflect the specific interactions between the unlike molecules. The values of V^E , $\Delta\eta$ and G^E of binary mixtures were fitted to the Redlich-Kister polynomial equation to estimate the coefficients and standard deviations between the experimental and calculated values.

MATERIALS AND METHODS

1. Materials

Propiophenone as a main component and *o*-xylene, *m*-xylene and *p*-xylene are taken as secondary components. These chemicals were

[†]To whom correspondence should be addressed.

E-mail: dittakavirc17@gmail.com

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Table 1. Provenance and purity of the materials used

Chemicals	CAS number	Source	Mass fraction purity (final)
Propiophenone	93-55-0	S.D fine Chemicals, India	>99%
<i>o</i> -Xylene	95-47-6	S.D fine Chemicals, India	0.994
<i>m</i> -Xylene	108-38-3	S.D fine Chemicals, India	0.995
<i>p</i> -Xylene	106-42-3	S.D fine Chemicals, India	0.995

Table 2. Comparison of experimental and literature values of density and viscosity of pure liquids at $T=303.15$ – 318.15 K and 0.1 MPa

Compound	T/K	Density (ρ)		Ref.	Viscosity (η)		Ref.
		Experimental	Literature		Experimental	Literature	
		gm/cm ⁻³	gm/cm ⁻³		mPa·s	mPa·s	
Propiophenone	303.15	1.0045	1.0044	a	1.5150	1.5100	a
	308.15	1.0015	1.0006	a	1.4690	1.4690	b
	313.15	0.9985			1.4230		
	318.15	0.9955			1.3770		
<i>o</i> -Xylene	303.15	0.8671	0.8605	c	0.7197	0.7272	c
	308.15	0.8648	0.8667	d	0.6799	0.674, 0.676	g
	313.15	0.8625	0.8633	e	0.6401	0.621, 0.627	e
	318.15	0.8601	0.8659	f	0.6003	0.591, 0.589	d
<i>m</i> -Xylene	303.15	0.8526	0.8553	e	0.5540	0.557, 0.558	e
	308.15	0.8491	0.8513	d	0.5268	0.531, 0.533	g
	313.15	0.8457	0.8471	e	0.4996	0.508, 0.497	e
	318.15	0.8423	0.8427	d	0.4724	0.470, 0.471	d
<i>p</i> -Xylene	303.15	0.8488	0.8521	e	0.5703	0.597, 0.590	e
	308.15	0.8456	0.8482	d	0.5411	0.548, 0.547	g
	313.15	0.8425	0.8435	e	0.5119	0.538, 0.537	e
	318.15	0.8395	0.8395	d	0.4827	0.486, 0.484	d

purchased from S.D. Fine chemicals Ltd., India and used in the present investigation. Purification of the solvents was by the methods as described earlier [10,11]. The mass fraction purities of the liquids were as propiophenone (>99%), *o*-xylene (99.7%), *m*-xylene (99.5%), and *p*-xylene (99.5%) and water content found less than 0.003 mass%. Purities of the solvents are shown in Table 1.

Binary mixtures were prepared by mixing weighed amounts of the pure liquids, adopting the method of closed system by using Mettler Toledo (ME204) balance with the precision of ± 0.1 mg. The purity of the liquids was checked by comparing the measured values of densities and viscosities with literature data at temperatures $T=(303.15$ – $318.15)$ K and are given in Table 2. References are a[12], b[13], c[14], d[7], e[15], f[16], g[17].

2. Apparatus and Procedure

Anton Paar (DSA 5000 M) oscillating u-tube densimeter, automatically thermostatic within ± 0.01 K, was used to calculate the density of the pure components and the binary mixtures over the whole composition range $T=(303.15$ to $318.15)$ K. The accuracy for density was $\pm 1 \times 10^{-2}$ kg·m⁻³. At regular intervals calibration was done at the required temperature using dry air and ultrapure water (Millipore). Averages of ten consecutive measurements were taken between the temperatures from (303.15 to 318.15) K with an increment of 5 K under atmospheric pressure. The standard uncertainty associated with the measurement of density is ± 0.01 kg·m⁻³.

The viscosity of the binary liquid mixtures was measured by using a suspended level Ubbelohde viscometer with proper calibration and the method as described earlier [18] that the viscometer was calibrated at 303.15 ± 0.01 K using distilled water. The viscometer constant k was calculated from viscosity (η_w), density (ρ_w), and flow time (t_w) of water using the relation $k=(\eta_w/\rho_w t_w)$. An average of 10 measurements was taken to determine the k value. An electronic stopwatch capable of measuring time to ± 0.01 s was used for flow time measurement. Capillary-end kinetic energy corrections were applied and found to be negligible. The estimated uncertainty in viscosity was ± 0.0010 mPa·s. The mole fractions of each mixture were measured with the masses of the components. The uncertainty in measured mole fractions was ± 0.0001 .

The calculated values of densities and viscosities with respective mole fractions of the studied binary mixtures at $T=(303.15$ to $318.15)$ K are tabulated in Table 3.

THEORY

The following relations were used to calculate the viscosity deviations and excess thermodynamic functions, dependence of viscosity on temperatures of binary liquid mixtures over the entire composition range to understand further on the molecular interactions of the binary mixtures.

Table 3. Experimental data of density (ρ) and viscosity (η) for the Propiophenone with *o*-xylene, *m*-xylene and *p*-xylene binary mixtures at T=(303.15-318.15) K

X_1	Density (ρ)/g·cm ⁻³				Viscosity (η)/mPa·s			
	303.15 K	308.15 K	313.15 K	318.15 K	303.15 K	308.15 K	313.15 K	318.15 K
PPH+ <i>o</i> -xylene								
0.0000	0.8671	0.8648	0.8625	0.8601	0.7197	0.6799	0.6401	0.6003
0.0918	0.8813	0.8791	0.8768	0.8745	0.7803	0.7412	0.7028	0.6655
0.1852	0.8954	0.8932	0.8909	0.8886	0.8462	0.8075	0.7686	0.7306
0.2804	0.9094	0.9071	0.9048	0.9025	0.9151	0.8758	0.8362	0.7977
0.3774	0.9233	0.9210	0.9186	0.9161	0.9876	0.9475	0.9074	0.8682
0.4762	0.9371	0.9347	0.9322	0.9297	1.0642	1.0234	0.9827	0.9429
0.5769	0.9508	0.9483	0.9457	0.9431	1.1455	1.1039	1.0625	1.0221
0.6796	0.9643	0.9617	0.9591	0.9565	1.2306	1.1888	1.1467	1.1057
0.7843	0.9778	0.9751	0.9724	0.9697	1.3205	1.2781	1.2354	1.1938
0.8911	0.9911	0.9883	0.9856	0.9827	1.4143	1.3703	1.3270	1.2847
1.0000	1.0045	1.0015	0.9985	0.9955	1.5150	1.4690	1.4230	1.3770
PPH+ <i>m</i> -xylene								
0.0000	0.8526	0.8491	0.8457	0.8423	0.5540	0.5268	0.4996	0.4724
0.0931	0.8685	0.8652	0.8620	0.8587	0.6331	0.6054	0.5780	0.5515
0.1877	0.8841	0.8809	0.8778	0.8746	0.7155	0.6871	0.6578	0.6296
0.2837	0.8996	0.8964	0.8933	0.8902	0.8010	0.7707	0.7396	0.7096
0.3812	0.9149	0.9118	0.9087	0.9056	0.8901	0.8578	0.8249	0.7930
0.4803	0.9301	0.9271	0.9241	0.9210	0.9833	0.9490	0.9142	0.8805
0.5809	0.9453	0.9423	0.9393	0.9363	1.0811	1.0448	1.0082	0.9726
0.6831	0.9603	0.9574	0.9545	0.9515	1.1830	1.1451	1.1065	1.0690
0.7871	0.9753	0.9724	0.9695	0.9666	1.2893	1.2496	1.2091	1.1696
0.8927	0.9900	0.9871	0.9842	0.9813	1.3997	1.3571	1.3146	1.2731
1.0000	1.0045	1.0015	0.9985	0.9955	1.5150	1.4690	1.4230	1.3770
PPH+ <i>p</i> -xylene								
0.0000	0.8488	0.8456	0.8424	0.8395	0.5703	0.5411	0.5119	0.4827
0.0934	0.8651	0.8621	0.8590	0.8562	0.6511	0.6214	0.5926	0.5639
0.1882	0.8811	0.8782	0.8752	0.8725	0.7322	0.7035	0.6731	0.6428
0.2844	0.8970	0.8941	0.8911	0.8884	0.8164	0.7868	0.7548	0.7228
0.3820	0.9127	0.9098	0.9069	0.9041	0.9040	0.8727	0.8391	0.8055
0.4811	0.9284	0.9255	0.9225	0.9198	0.9955	0.9623	0.9270	0.8918
0.5817	0.9439	0.9410	0.9381	0.9353	1.0918	1.0562	1.0193	0.9823
0.6839	0.9593	0.9565	0.9536	0.9508	1.1918	1.1552	1.1165	1.0778
0.7876	0.9746	0.9718	0.9689	0.9662	1.2963	1.2579	1.2175	1.1771
0.8930	0.9897	0.9868	0.9840	0.9811	1.4048	1.3632	1.3210	1.2788
1.0000	1.0045	1.0015	0.9985	0.9955	1.5150	1.4690	1.4230	1.3770

The standard uncertainties for density u (ρ)=0.001 kg·cm⁻³, viscosity u (η)=0.0010 mPa·s, mole fraction u (x_1)=0.0001, temperature u (T)=0.01 K and for time u (t)=0.01 sec

a) Excess molar volume is calculated by using the relation

$$V^E/(\text{cm}^3 \cdot \text{mol}^{-1}) = \frac{x_1 M_1 + x_2 M_2}{\rho_m} - \left\{ \frac{x_1 M_1}{\rho_1} + \frac{x_2 M_2}{\rho_2} \right\} \quad (1)$$

where ρ_m , ρ_1 and ρ_2 , are the densities, M_1 and M_2 are molar masses of components 1 and 2 of the binary mixtures and x_1 and x_2 are the mole fractions.

b) Viscosity deviation is calculated by using the relation

$$\Delta\eta = \eta - (x_1 \eta_1 + x_2 \eta_2) \quad (2)$$

where η_1 and η_2 are the viscosities of components 1 and 2 of the binary mixtures and x_1 and x_2 are the mole fractions.

c) The excess Gibbs free energy of activation of viscous flow (G^{*E}) is obtained by the equation

$$G^{*E} = RT[\ln \eta_m V_m - (x_1 \ln V_1 \eta_1 + x_2 \ln V_2 \eta_2)] \quad (3)$$

where V_m is the molar volume of the mixture, V_1 and V_2 , x_1 , x_2 and η_1 , η_2 are the molar volumes, mole fractions and viscosity values of component 1 and component 2, respectively, R and T have their usual meanings.

1. Single Adjustable Interaction Parameters

d) Grunberg and Nissan [19] proposed the following logarithmic equation for the measurement of viscosity of liquid mixtures;

$$\ln \eta = x_1 \ln \eta_1 + x_2 \ln \eta_2 + x_1 x_2 d_{12} \quad (4)$$

where d_{12} is a parameter proportional to interchange energy; it may be considered as an approximate measure to know the strength of molecular interactions between the mixing components.

e) Katti and Chaudhri [20] proposed the following equation:

$$\ln \eta V = x_1 \ln V_1 \eta_1 + x_2 \ln V_2 \eta_2 + x_1 x_2 W_{vis}/RT \quad (5)$$

where W_{vis}/RT is an interaction energy parameter, and x_1 and x_2 are the mole fractions.

f) Hind et al. [21] suggested an equation for the viscosity of binary liquid mixtures as

$$\eta = x_1^2 \eta_1 + x_2^2 \eta_2 + 2x_1 x_2 H_{12} \quad (6)$$

where H_{12} is Hind interaction constant which is a function of composition and temperature, η is the predicted value of dynamic viscosity.

g) Tamara-Kurata's equation [22] for the viscosity of binary liquid mixtures:

$$\eta = x_1 \phi_1 \eta_1 + x_2 \phi_2 \eta_2 + 2(x_1 \phi_1 x_2 \phi_2)^{0.5} T_{12} \quad (7)$$

where ϕ_i is the ideal state volume fraction and is defined by the relation:

$$\phi_i = \frac{x_i V_i}{\sum_{i=1}^2 x_i V_i} \quad (8)$$

where ϕ_i is the volume fraction of the pure components, x_i is the mole fraction and V_i is the molar volume

h) Frenkel [23] equation

$$\ln \eta = x_1^2 \ln \eta_1 + x_2^2 \ln \eta_2 + 2x_1 x_2 F_{12} \quad (9)$$

where F_{12} is Frenkel interaction constant which is a function of composition and temperature, η is the predicted value of dynamic viscosity.

2. Two Adjustable Interaction Parameters

Experimental viscosity data were incorporated in the McAllister's three body model equation to compute the adjustable interaction parameters and standard deviation of the studied binary mixtures for the entire composition range at $T=(303.15$ to $318.15)$ K.

a) The McAllister three body relation is

$$\begin{aligned} \ln \eta = & x_1^3 \ln \eta_1 + 3x_1^2 x_2 \ln a + 3x_1 x_2^2 \ln b + x_2^3 \ln \eta_2 \\ & - \ln \left[\left(x_1 + \frac{x_2 M_2}{M_1} \right) \right] + 3x_1^2 x_2 \ln \left[\left(2 + \frac{M_2}{M_1} \right) / 3 \right] \\ & + 3x_1 x_2^2 \ln \left[\left(1 + 2 \frac{M_2}{M_1} \right) / 3 \right] + x_2^3 \ln \left[\left(\frac{M_2}{M_1} \right) \right] \end{aligned} \quad (10)$$

where η is the viscosity of the binary liquid mixture and, x_1 , η_1 , M_1 , x_2 , η_2 , M_2 are the mole fractions, viscosities and molecular weights of component 1 and component 2, respectively. 'a' and 'b' are the adjustable interaction parameters.

b) Heric equation:

$$\ln(\eta_{mix}) = x_1 \ln(\eta_1) + x_2 \ln(\eta_2) + x_1 \ln(M_1) + x_2 \ln(M_2) \quad (11)$$

$$- \ln(x_1 M_1 + x_2 M_2) + x_1 x_2 [\alpha_{12} + \alpha_{21}(x_1 - x_2)]$$

where η_{mix} is the viscosity of the binary liquid mixture and, x_1 , η_1 , M_1 , x_2 , η_2 , M_2 are the mole fractions, viscosities and molecular weights of component 1 and component 2, respectively. ' α_{12} ' and ' α_{21} ' are the adjustable interaction parameters.

3. Three Adjustable Interaction Parameters

Experimental viscosity data were incorporated in several relations to compute the adjustable interactions parameters and standard deviations of the studied binary mixtures for the entire compositions range at $T=(303.15$ to $318.15)$ K.

a) The McAllister four body model relation is

$$\begin{aligned} \ln \eta = & x_1^4 \ln \eta_1 + 4x_1^3 x_2 \ln A + 6x_1^2 x_2^2 \ln B + 4x_1 x_2^3 \ln C + x_2^4 \ln \eta_2 \\ & - \ln \left[\left(x_1 + \frac{x_2 M_2}{M_1} \right) \right] + 4x_1^3 x_2 \ln \left[\left(3 + \frac{M_2}{M_1} \right) / 4 \right] \\ & + 6x_1^2 x_2^2 \ln \left[\left(1 + \frac{M_2}{M_1} \right) / 2 \right] \\ & + 4x_1 x_2^3 \ln \left[\left(1 + 3 \frac{M_2}{M_1} \right) / 4 \right] + x_2^4 \ln \left[\left(\frac{M_2}{M_1} \right) \right] \end{aligned} \quad (12)$$

b) Auslander model is

$$\eta = (\eta_1 x_1 (x_1 + B_{12} x_2) + \eta_2 (A_{21} x_2 (B_{21} x_1 + x_2))) / (x_1 (x_1 + B_{12} x_2) + (A_{21} x_2) (B_{21} x_1 + x_2)) \quad (13)$$

c) Jouyban-Acree equation is

$$\begin{aligned} \ln \eta = & x_1 \ln \eta_1 + x_2 \ln \eta_2 + A_0 \left[\frac{x_1 x_2}{T} \right] + A_1 \left[\frac{x_1 x_2 (x_1 - x_2)}{T} \right] \\ & + A_2 \left[\frac{x_1 x_2 (x_1 - x_2)^2}{T} \right] \end{aligned} \quad (14)$$

where η is the viscosity of the binary liquid mixture and, x_1 , η_1 , M_1 , x_2 , η_2 , M_2 are the mole fractions, viscosities and molecular weights of component 1 and component 2 respectively. A , B , C , B_{12} , A_{21} , B_{21} , A_0 , A_1 , and A_2 are the adjustable interaction parameters.

4. Viscosity Relations with No Adjustable Parameters

Viscosities of binary liquid mixtures are calculated by using the following relations having without any adjustable parameters and also computed the Standard deviation percentages.

a) Kendall and Monroe equation [24]

$$\eta = (x_1 \eta_1^{1/3} + x_2 \eta_2^{1/3})^3 \quad (15)$$

where η is the viscosity of the binary liquid mixture and, x_1 , η_1 , x_2 , η_2 , are the mole fractions and viscosities of component 1 and component 2, respectively.

b) Bingham equation [25]

$$\eta = \sum x_i \eta_i \quad (16)$$

where x_i , η_i are the mole fraction and viscosity of i th component, respectively.

c) Arrhenius equation [26] for the viscosity of pure liquid and mixtures can be modified as

$$\ln \eta V = \sum x_i \ln \eta_i V_i \quad (17)$$

d) Kendall equation-viscosity of binary liquid mixture is given as

$$\ln \eta = \sum x_i \ln \eta_i \quad (18)$$

Table 4. (a) Excess molar volume (V^E), deviation in viscosity ($\Delta\eta$), Gibbs free energy activation (G^E), Grunberg-Nissan (d_{12}), Katti-chaudhari (W_{vis}/RT), Hind et al. (H_{12}), Frenkel (F_{12}) and Tamara-Kurata (T_{12}) constants for the binary mixtures of Propiophenone with *o*-xylene over the entire composition range at $T=(303.15$ - $318.15)$ K

X_1	V^E	$\Delta\eta$	G^E	d_{12}	W_{vis}/RT	H_{12}	T_{12}	F_{12}	V^E	$\Delta\eta$	G^E	d_{12}	W_{vis}/RT	H_{12}	T_{12}	F_{12}
PPH+<i>o</i>-xylene																
303.15 K																
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.0918	-0.0775	-0.0125	0.3023	0.1493	0.1457	1.0426	1.0301	0.1179	-0.1073	-0.0091	0.4880	0.2416	0.2352	0.9767	0.9636	-0.5088
0.1852	-0.1358	-0.0208	0.5853	0.1592	0.1558	1.0483	1.0364	0.1229	-0.1877	-0.0165	0.8475	0.2317	0.2256	0.9768	0.9645	0.0691
0.2804	-0.1782	-0.0276	0.7680	0.1561	0.1529	1.0490	1.0379	0.1213	-0.2399	-0.0234	1.0491	0.2144	0.2089	0.9736	0.9623	0.0605
0.3774	-0.2066	-0.0322	0.8665	0.1513	0.1482	1.0488	1.0387	0.1189	-0.2679	-0.0282	1.1505	0.2018	0.1967	0.9715	0.9614	0.0542
0.4762	-0.2177	-0.0342	0.8950	0.1472	0.1442	1.0488	1.0399	0.1168	-0.2721	-0.0302	1.1712	0.1933	0.1886	0.9709	0.9620	0.0500
0.5769	-0.2071	-0.0330	0.8623	0.1447	0.1419	1.0497	1.0422	0.1156	-0.2599	-0.0292	1.1155	0.1880	0.1836	0.9717	0.9642	0.0473
0.6796	-0.1750	-0.0296	0.7483	0.1405	0.1381	1.0495	1.0436	0.1135	-0.2266	-0.0254	0.9761	0.1843	0.1801	0.9732	0.9673	0.0455
0.7843	-0.1232	-0.0229	0.5688	0.1369	0.1351	1.0496	1.0455	0.1117	-0.1756	-0.0187	0.7540	0.1832	0.1791	0.9763	0.9722	0.0449
0.8911	-0.0559	-0.0141	0.3042	0.1266	0.1259	1.0448	1.0426	0.1065	-0.1032	-0.0108	0.4165	0.1767	0.1724	0.9761	0.9740	0.0417
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
308.15 K																
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.0918	-0.0979	-0.0111	0.3765	0.1870	0.1814	1.0076	0.9948	-0.1901	-0.1339	-0.0061	0.6491	0.3218	0.3128	0.9518	0.9381	-0.8365
0.1852	-0.1674	-0.0185	0.7117	0.1945	0.1895	1.0131	1.0009	0.0966	-0.2228	-0.0135	1.0337	0.2830	0.2752	0.9439	0.9314	0.0463
0.2804	-0.2156	-0.0254	0.9010	0.1840	0.1794	1.0115	1.0003	0.0914	-0.2719	-0.0204	1.2478	0.2552	0.2484	0.9381	0.9268	0.0324
0.3774	-0.2413	-0.0302	0.9992	0.1751	0.1709	1.0102	1.0001	0.0869	-0.2930	-0.0252	1.3519	0.2370	0.2312	0.9350	0.9249	0.0233
0.4762	-0.2481	-0.0322	1.0234	0.1688	0.1648	1.0098	1.0009	0.0838	-0.2949	-0.0272	1.3656	0.2253	0.2200	0.9340	0.9252	0.0174
0.5769	-0.2353	-0.0312	0.9787	0.1648	0.1611	1.0105	1.0030	0.0818	-0.2811	-0.0262	1.2952	0.2182	0.2132	0.9349	0.9275	0.0139
0.6796	-0.1996	-0.0274	0.8574	0.1615	0.1582	1.0115	1.0056	0.0801	-0.2470	-0.0224	1.1338	0.2140	0.2092	0.9372	0.9313	0.0118
0.7843	-0.1471	-0.0207	0.6593	0.1594	0.1566	1.0133	1.0092	0.0791	-0.1967	-0.0157	0.8825	0.2146	0.2096	0.9423	0.9382	0.0121
0.8911	-0.0760	-0.0128	0.3520	0.1479	0.1457	1.0087	1.0066	0.0733	-0.1164	-0.0078	0.5102	0.2164	0.2112	0.9487	0.9466	0.0130
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
318.15 K																
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.0918	-0.0979	-0.0111	0.3765	0.1870	0.1814	1.0076	0.9948	-0.1901	-0.1339	-0.0061	0.6491	0.3218	0.3128	0.9518	0.9381	-0.8365
0.1852	-0.1674	-0.0185	0.7117	0.1945	0.1895	1.0131	1.0009	0.0966	-0.2228	-0.0135	1.0337	0.2830	0.2752	0.9439	0.9314	0.0463
0.2804	-0.2156	-0.0254	0.9010	0.1840	0.1794	1.0115	1.0003	0.0914	-0.2719	-0.0204	1.2478	0.2552	0.2484	0.9381	0.9268	0.0324
0.3774	-0.2413	-0.0302	0.9992	0.1751	0.1709	1.0102	1.0001	0.0869	-0.2930	-0.0252	1.3519	0.2370	0.2312	0.9350	0.9249	0.0233
0.4762	-0.2481	-0.0322	1.0234	0.1688	0.1648	1.0098	1.0009	0.0838	-0.2949	-0.0272	1.3656	0.2253	0.2200	0.9340	0.9252	0.0174
0.5769	-0.2353	-0.0312	0.9787	0.1648	0.1611	1.0105	1.0030	0.0818	-0.2811	-0.0262	1.2952	0.2182	0.2132	0.9349	0.9275	0.0139
0.6796	-0.1996	-0.0274	0.8574	0.1615	0.1582	1.0115	1.0056	0.0801	-0.2470	-0.0224	1.1338	0.2140	0.2092	0.9372	0.9313	0.0118
0.7843	-0.1471	-0.0207	0.6593	0.1594	0.1566	1.0133	1.0092	0.0791	-0.1967	-0.0157	0.8825	0.2146	0.2096	0.9423	0.9382	0.0121
0.8911	-0.0760	-0.0128	0.3520	0.1479	0.1457	1.0087	1.0066	0.0733	-0.1164	-0.0078	0.5102	0.2164	0.2112	0.9487	0.9466	0.0130
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000

Table 4. (b) Excess molar volume (V^E), deviation in viscosity ($\Delta\eta$), Gibbs free energy activation (G^E), Grunberg-Nissan (d_{12}), Katti-chaudhari (W_{vis}/RT), Hind et al. (H_{12}), Frenkel (F_{12}) and Tamara-Kurata (T_{12}) constants for the binary mixtures of Propiophenone with *m*-xylene over the entire composition range at $T=(303.15-318.15)$ K

X_1	V^E	$\Delta\eta$	G^E	d_{12}	W_{vis}/RT	H_{12}	T_{12}	F_{12}	V^E	$\Delta\eta$	G^E	d_{12}	W_{vis}/RT	H_{12}	T_{12}	F_{12}
PPH+<i>m</i>-xylene																
303.15 K																
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.0931	-0.1161	-0.0104	0.9716	0.4708	0.4623	0.9729	0.9595	0.1478	-0.1698	-0.0076	1.1730	0.5717	0.5582	0.9163	0.9034	-0.3914
0.1877	-0.1916	-0.0189	1.6383	0.4392	0.4317	0.9725	0.9601	0.1320	-0.2584	-0.0151	1.9165	0.5160	0.5050	0.9118	0.8999	0.0874
0.2837	-0.2380	-0.0256	2.0396	0.4100	0.4033	0.9715	0.9601	0.1174	-0.3025	-0.0219	2.3276	0.4695	0.4602	0.9073	0.8965	0.0641
0.3812	-0.2590	-0.0302	2.2209	0.3844	0.3783	0.9704	0.9602	0.1046	-0.3271	-0.0267	2.5001	0.4343	0.4258	0.9047	0.8950	0.0465
0.4803	-0.2648	-0.0323	2.2171	0.3626	0.3569	0.9698	0.9609	0.0937	-0.3359	-0.0289	2.4776	0.4069	0.3988	0.9035	0.8951	0.0328
0.5809	-0.2546	-0.0311	2.0617	0.3459	0.3402	0.9706	0.9631	0.0853	-0.3281	-0.0278	2.2925	0.3863	0.3783	0.9042	0.8971	0.0226
0.6831	-0.2285	-0.0275	1.7470	0.3299	0.3242	0.9710	0.9652	0.0774	-0.3022	-0.0239	1.9502	0.3703	0.3620	0.9062	0.9006	0.0145
0.7871	-0.1846	-0.0211	1.2914	0.3156	0.3096	0.9716	0.9675	0.0702	-0.2500	-0.0173	1.4544	0.3577	0.3487	0.9096	0.9057	0.0083
0.8927	-0.1123	-0.0122	0.7018	0.3008	0.2944	0.9711	0.9689	0.0628	-0.1534	-0.0093	0.8001	0.3453	0.3356	0.9126	0.9106	0.0021
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
308.15 K																
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.0931	-0.1406	-0.0091	1.0633	0.5168	0.5060	0.9440	0.9309	-0.1166	-0.1803	-0.0051	1.3447	0.6545	0.6399	0.8945	0.8817	-0.6688
0.1877	-0.2218	-0.0166	1.7854	0.4795	0.4705	0.9435	0.9313	0.1116	-0.2815	-0.0126	2.1057	0.5671	0.5549	0.8834	0.8718	0.0685
0.2837	-0.2715	-0.0234	2.1870	0.4405	0.4324	0.9403	0.9292	0.0921	-0.3321	-0.0194	2.5201	0.5087	0.4982	0.8769	0.8664	0.0393
0.3812	-0.2948	-0.0282	2.3606	0.4094	0.4021	0.9381	0.9282	0.0765	-0.3566	-0.0242	2.6876	0.4672	0.4578	0.8734	0.8640	0.0186
0.4803	-0.3011	-0.0304	2.3464	0.3846	0.3777	0.9371	0.9284	0.0641	-0.3653	-0.0264	2.6531	0.4360	0.4270	0.8719	0.8637	0.0030
0.5809	-0.2949	-0.0293	2.1741	0.3657	0.3588	0.9377	0.9304	0.0547	-0.3564	-0.0253	2.4507	0.4134	0.4044	0.8727	0.8658	-0.0083
0.6831	-0.2670	-0.0254	1.8502	0.3504	0.3434	0.9393	0.9336	0.0470	-0.3306	-0.0214	2.0858	0.3965	0.3871	0.8754	0.8700	-0.0168
0.7871	-0.2175	-0.0188	1.3765	0.3375	0.3300	0.9417	0.9377	0.0406	-0.2764	-0.0148	1.5631	0.3850	0.3748	0.8805	0.8767	-0.0225
0.8927	-0.1316	-0.0108	0.7474	0.3215	0.3135	0.9414	0.9393	0.0326	-0.1754	-0.0068	0.8775	0.3795	0.3681	0.8891	0.8871	-0.0252
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
318.15 K																
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.0931	-0.1406	-0.0091	1.0633	0.5168	0.5060	0.9440	0.9309	-0.1166	-0.1803	-0.0051	1.3447	0.6545	0.6399	0.8945	0.8817	-0.6688
0.1877	-0.2218	-0.0166	1.7854	0.4795	0.4705	0.9435	0.9313	0.1116	-0.2815	-0.0126	2.1057	0.5671	0.5549	0.8834	0.8718	0.0685
0.2837	-0.2715	-0.0234	2.1870	0.4405	0.4324	0.9403	0.9292	0.0921	-0.3321	-0.0194	2.5201	0.5087	0.4982	0.8769	0.8664	0.0393
0.3812	-0.2948	-0.0282	2.3606	0.4094	0.4021	0.9381	0.9282	0.0765	-0.3566	-0.0242	2.6876	0.4672	0.4578	0.8734	0.8640	0.0186
0.4803	-0.3011	-0.0304	2.3464	0.3846	0.3777	0.9371	0.9284	0.0641	-0.3653	-0.0264	2.6531	0.4360	0.4270	0.8719	0.8637	0.0030
0.5809	-0.2949	-0.0293	2.1741	0.3657	0.3588	0.9377	0.9304	0.0547	-0.3564	-0.0253	2.4507	0.4134	0.4044	0.8727	0.8658	-0.0083
0.6831	-0.2670	-0.0254	1.8502	0.3504	0.3434	0.9393	0.9336	0.0470	-0.3306	-0.0214	2.0858	0.3965	0.3871	0.8754	0.8700	-0.0168
0.7871	-0.2175	-0.0188	1.3765	0.3375	0.3300	0.9417	0.9377	0.0406	-0.2764	-0.0148	1.5631	0.3850	0.3748	0.8805	0.8767	-0.0225
0.8927	-0.1316	-0.0108	0.7474	0.3215	0.3135	0.9414	0.9393	0.0326	-0.1754	-0.0068	0.8775	0.3795	0.3681	0.8891	0.8871	-0.0252
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000

Table 4. (c) Excess molar volume (V^E), deviation in viscosity ($\Delta\eta$), Gibbs free energy activation (G^E), Grunberg-Nissan (d_{12}), Katti-chaudhari (W_{vis}/RT), Hind et al. (H_{12}), Frenkel (F_{12}) and Tamara-Kurata (T_{12}) constants for the binary mixtures of Propiophenone with *p*-xylene over the entire composition range at $T=(303.15\text{-}318.15) \text{ K}$

X_1	V^E	$\Delta\eta$	G^E	d_{12}	W_{vis}/RT	H_{12}	T_{12}	F_{12}	V^E	$\Delta\eta$	G^E	d_{12}	W_{vis}/RT	H_{12}	T_{12}	F_{12}
303.15 K																
PPH+<i>p</i>-xylene																
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.0934	-0.1219	-0.0074	1.0081	0.4876	0.4783	0.9989	0.9862	0.1707	-0.1708	-0.0043	1.2397	0.6020	0.5882	0.9418	0.9293	-0.3850
0.1882	-0.2033	-0.0159	1.6123	0.4323	0.4240	0.9907	0.9791	0.1431	-0.2692	-0.0102	1.9812	0.5327	0.5210	0.9339	0.9226	0.1079
0.2844	-0.2558	-0.0226	1.9730	0.3972	0.3895	0.9871	0.9765	0.1255	-0.3252	-0.0162	2.3758	0.4794	0.4690	0.9276	0.9174	0.0813
0.3820	-0.2820	-0.0272	2.1334	0.3702	0.3631	0.9850	0.9755	0.1120	-0.3482	-0.0208	2.5248	0.4390	0.4297	0.9233	0.9142	0.0611
0.4811	-0.2926	-0.0293	2.1246	0.3489	0.3419	0.9841	0.9758	0.1013	-0.3535	-0.0232	2.4837	0.4086	0.3997	0.9210	0.9131	0.0459
0.5817	-0.2819	-0.0281	1.9764	0.3331	0.3263	0.9849	0.9780	0.0935	-0.3412	-0.0226	2.2869	0.3863	0.3776	0.9210	0.9144	0.0347
0.6839	-0.2540	-0.0245	1.6785	0.3188	0.3119	0.9859	0.9805	0.0863	-0.3136	-0.0185	1.9572	0.3727	0.3637	0.9246	0.9194	0.0279
0.7876	-0.2051	-0.0180	1.2557	0.3088	0.3016	0.9888	0.9851	0.0813	-0.2641	-0.0120	1.4815	0.3657	0.3558	0.9316	0.9280	0.0244
0.8930	-0.1217	-0.0091	0.7044	0.3037	0.2962	0.9949	0.9930	0.0788	-0.1692	-0.0045	0.8447	0.3664	0.3552	0.9438	0.9419	0.0248
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
308.15 K																
PPH+<i>p</i>-xylene																
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.0934	-0.1550	-0.0063	1.0969	0.5328	0.5204	0.9676	0.9550	-0.1051	-0.1920	-0.0023	1.3988	0.6795	0.6637	0.9160	0.9035	-0.6788
0.1882	-0.2421	-0.0122	1.8147	0.4876	0.4772	0.9650	0.9534	0.1290	-0.3001	-0.0082	2.1672	0.5832	0.5699	0.9029	0.8916	0.0874
0.2844	-0.2941	-0.0182	2.2013	0.4438	0.4346	0.9603	0.9498	0.1071	-0.3587	-0.0142	2.5706	0.5191	0.5075	0.8949	0.8848	0.0553
0.3820	-0.3195	-0.0228	2.3523	0.4087	0.4003	0.9567	0.9474	0.0896	-0.3824	-0.0188	2.7164	0.4728	0.4623	0.8899	0.8810	0.0322
0.4811	-0.3275	-0.0252	2.3210	0.3816	0.3735	0.9546	0.9465	0.0760	-0.3901	-0.0212	2.6641	0.4387	0.4288	0.8875	0.8797	0.0151
0.5817	-0.3158	-0.0246	2.1405	0.3613	0.3534	0.9545	0.9477	0.0659	-0.3782	-0.0206	2.4476	0.4140	0.4041	0.8875	0.8810	0.0028
0.6839	-0.2876	-0.0205	1.8321	0.3485	0.3405	0.9575	0.9522	0.0595	-0.3512	-0.0165	2.0937	0.3994	0.3891	0.8916	0.8865	-0.0045
0.7876	-0.2374	-0.0140	1.3828	0.3408	0.3321	0.9632	0.9595	0.0556	-0.2994	-0.0100	1.5889	0.3931	0.3816	0.9000	0.8964	-0.0077
0.8930	-0.1447	-0.0065	0.7772	0.3361	0.3268	0.9709	0.9690	0.0533	-0.1894	-0.0025	0.9196	0.3995	0.3867	0.9167	0.9148	-0.0045
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
318.15 K																
PPH+<i>p</i>-xylene																
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.0934	-0.1550	-0.0063	1.0969	0.5328	0.5204	0.9676	0.9550	-0.1051	-0.1920	-0.0023	1.3988	0.6795	0.6637	0.9160	0.9035	-0.6788
0.1882	-0.2421	-0.0122	1.8147	0.4876	0.4772	0.9650	0.9534	0.1290	-0.3001	-0.0082	2.1672	0.5832	0.5699	0.9029	0.8916	0.0874
0.2844	-0.2941	-0.0182	2.2013	0.4438	0.4346	0.9603	0.9498	0.1071	-0.3587	-0.0142	2.5706	0.5191	0.5075	0.8949	0.8848	0.0553
0.3820	-0.3195	-0.0228	2.3523	0.4087	0.4003	0.9567	0.9474	0.0896	-0.3824	-0.0188	2.7164	0.4728	0.4623	0.8899	0.8810	0.0322
0.4811	-0.3275	-0.0252	2.3210	0.3816	0.3735	0.9546	0.9465	0.0760	-0.3901	-0.0212	2.6641	0.4387	0.4288	0.8875	0.8797	0.0151
0.5817	-0.3158	-0.0246	2.1405	0.3613	0.3534	0.9545	0.9477	0.0659	-0.3782	-0.0206	2.4476	0.4140	0.4041	0.8875	0.8810	0.0028
0.6839	-0.2876	-0.0205	1.8321	0.3485	0.3405	0.9575	0.9522	0.0595	-0.3512	-0.0165	2.0937	0.3994	0.3891	0.8916	0.8865	-0.0045
0.7876	-0.2374	-0.0140	1.3828	0.3408	0.3321	0.9632	0.9595	0.0556	-0.2994	-0.0100	1.5889	0.3931	0.3816	0.9000	0.8964	-0.0077
0.8930	-0.1447	-0.0065	0.7772	0.3361	0.3268	0.9709	0.9690	0.0533	-0.1894	-0.0025	0.9196	0.3995	0.3867	0.9167	0.9148	-0.0045
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000

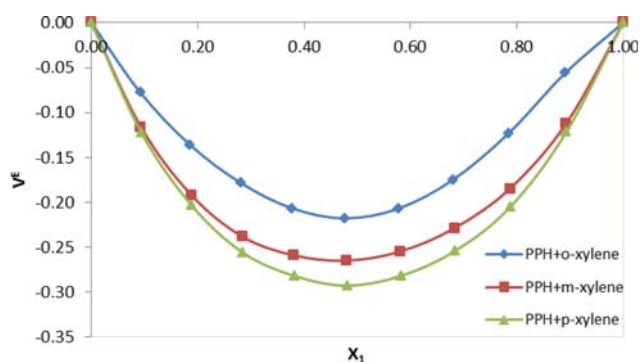


Fig. 1. Plot of excess molar volume (V^E) against mole fraction of (X_1) for PPH+*o*-xylene (◆), PPH+*m*-xylene (■), and PPH+*p*-xylene (▲) binary systems at $T=303.15$ K.

5. Standard Deviation Percentage ($\sigma\%$)

Standard deviation percentage ($\sigma\%$) between the experimental and calculated viscosity values is computed by using the relation as

$$\sigma(\%) = \left(\frac{1}{n-k} \sum \left\{ \frac{100(\eta_{exp} - \eta_{cal})}{\eta_{exp}} \right\}^2 \right)^{1/2} \quad (19)$$

where $\sigma(\%)$ is the standard deviation percentage, n is the number of experimental points and ' k ' is the number of coefficient. η_{exp} and η_{cal} are the experimental and calculated viscosity values of the binary liquid mixtures.

RESULTS AND DISCUSSION

Computed parameter values (V^E , $\Delta\eta$, G^{*E} , d_{12} , W_{vis}/RT , H_{12} , T_{12} , F_{12}) are given in Table 4(a)-4(c) at temperatures from $T=(303.15-318.15)$ K for the binary mixtures of propiophenone with *o*-xylene, *m*-xylene and *p*-xylene.

1. Excess Molar Volume (V^E)

Fig. 1 shows that the values of excess molar volume are negative for the binary mixtures of propiophenone with *o*-xylene, *m*-xylene and *p*-xylene over the entire composition range at $T=(303.15)$ K. V^E values are decreasing with increase in temperature from 303.15 K to 318.15 K. V^E values are high at mole fractions 0.4762, 0.4803 and 0.4811 for the binary liquids PPH+*o*-xylene, PPH+*m*-xylene and PPH+*p*-xylene, respectively, at all four temperatures.

More negative V^E values for the PPH+*p*-xylene are obtained than the other two binary liquids. Negative values of V^E suggest specific intermolecular interactions between the components. Fewer negative values are observed for the *o*-xylene, due to the presence of steric hindrance. It suggests the presence of significant charge transfer (donor-acceptor) interactions between PPH and isomeric xylenes. This is also supported by Nain et al. [27,28]. The order of the studied systems was PPH+*p*-xylene > PPH+*m*-xylene > PPH+*o*-xylene.

2. Deviation in Viscosity ($\Delta\eta$)

The deviation in viscosity reveals the strength of intermolecular interactions. Under the current study, deviations in viscosity ($\Delta\eta$) values (Fig. 2 to 5) are negative for all the studied binary liquid mixtures and the negative values are decreasing with increase in temperature.

The negative values may be due to dispersion or weak dipole-

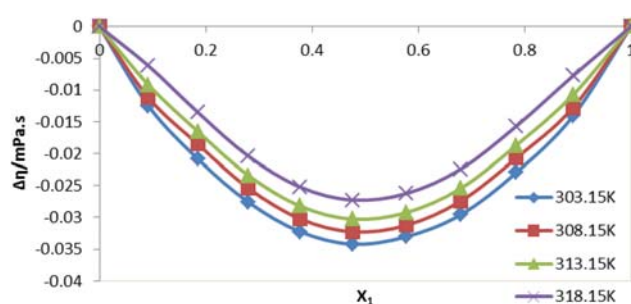


Fig. 2. Plot of deviation in viscosity against mole fraction of (X_1) at $T=303.15$ K (◆), 308.15 K (■), 313.15 K (▲) and 318.15 K (×) for the PPH+*o*-xylene system.

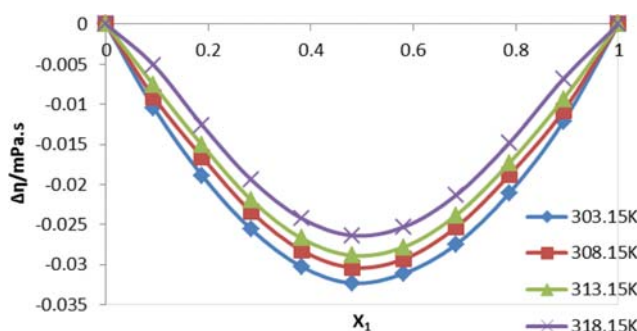


Fig. 3. Plot of deviation in viscosity against mole fraction of (X_1) at $T=303.15$ K (◆), 308.15 K (■), 313.15 K (▲) and 318.15 K (×) for the PPH+*m*-xylene system.

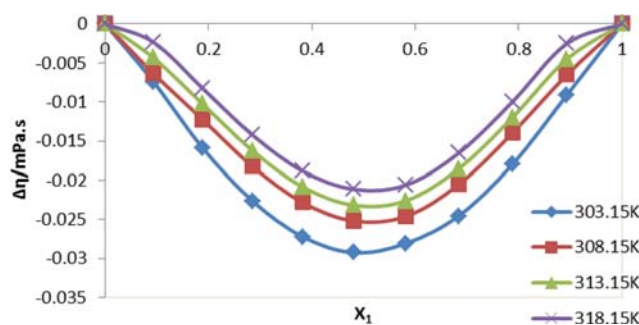


Fig. 4. Plot of deviation in viscosity against mole fraction of (X_1) at $T=303.15$ K (◆), 308.15 K (■), 313.15 K (▲) and 318.15 K (×) for the PPH+*p*-xylene system.

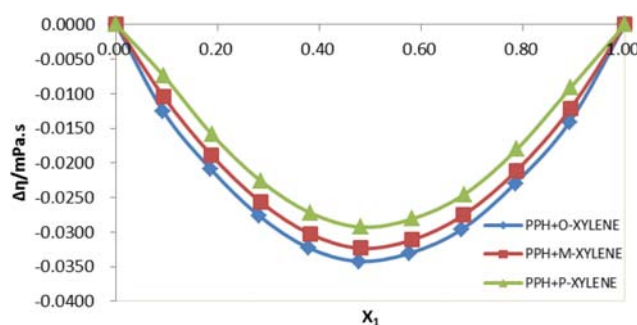


Fig. 5. Plot of deviation in viscosity against mole fraction of (X_1) for PPH+*o*-xylene (◆), PPH+*m*-xylene (■), and PPH+*p*-xylene (▲) binary systems at $T=303.15$ K.

dipole intermolecular interactions on the addition of one component to the other [15,29], $\square-\square^*$ interactions between the \square -electrons of the isomeric xylenes and the \square^* electrons of the phenyl group, deviations in viscosity is less negative to more negative from *p*-xylene to *o*-xylene, it is supported by Singh et al. [4,9].

This is due to the difference in size and shape of the component molecules and the loss of dipolar association in pure component, which is in agreement with Fort and Moore [30] and Pikkarainen [31]. It is generally explained that deviation in viscosity tends to become more negative where dispersion and dipolar interactions are operating and positive deviation in viscosity ($\Delta\eta$) tends to increase as the specific interactions and charge transfer complexes increases. The deviations in viscosity values at the equimolar composition were peak and their values were -0.0342 , -0.0323 and -0.0293 for PPH+*o*-xylene, PPH+*m*-xylene and PPH+*p*-xylene respectively at $T=303.15$ K. The interactions are in order of PPH+*p*-xylene > PPH+*m*-xylene > PPH+*o*-xylene.

3. Gibbs Free Energy (G^E)

Positive G^E values were observed for all the three studied binary mixtures, indicating specific interactions (like dipole-dipole inter-

actions and hydrogen bonding) between unlike molecules, which is in accordance with Reed and Taylor [32], where it is explained that the positive deviations in Gibbs free energy activation G^{*E} may be due to specific interactions like hydrogen bonding and charge transfer, whereas the negative deviations may be due to dispersion forces within the systems. Fig. 6-8, represents the graph for the Gibbs free energy (G^{*E}) values for each binary system at $T=(303.15-318.15)$ K. Fig. 9 represents the graph for Gibbs free energy values of all the three systems at $T=303.15$ K.

4. Grunberg Nissan (d_{12})

Under the current study, the values of $\Delta\eta$ and d_{12} are negative and positive, respectively, over the entire composition range for all the three binary mixtures, which indicates weak specific interactions between unlike molecules; it is in agreement with the Nigam and Mahal [33] where it is explained that negative $\Delta\eta$ and positive d_{12} indicates weak specific interactions, positive values of both deviation of viscosity $\Delta\eta$ and Grunberg-Nissan parameter d_{12} indicate the presence of strong specific interactions, and negative values of both $\Delta\eta$ and d_{12} indicate absence of specific interactions.

5. Katti-Chaudhary (W_{vis}/RT) and Frenkel (F_{12})

The values W_{vis}/RT and F_{12} of all the three studied binary liquid

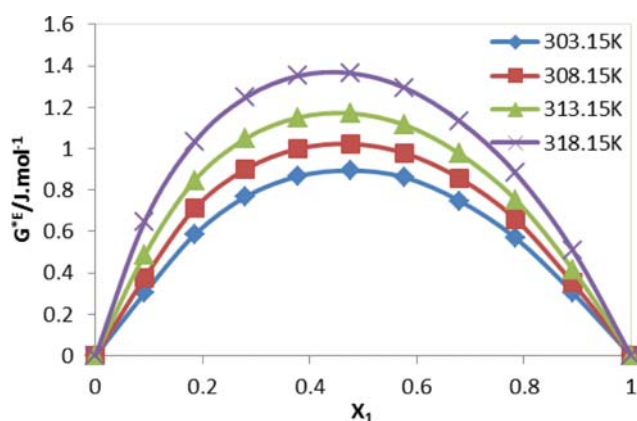


Fig. 6. Plot of deviations in Gibbs free energy (G^E) against mole fraction of (X_1) at $T=303.15$ K (◆), 308.15 K (■), 313.15 K (▲) and 318.15 K (×) for the PPH+*o*-xylene system.

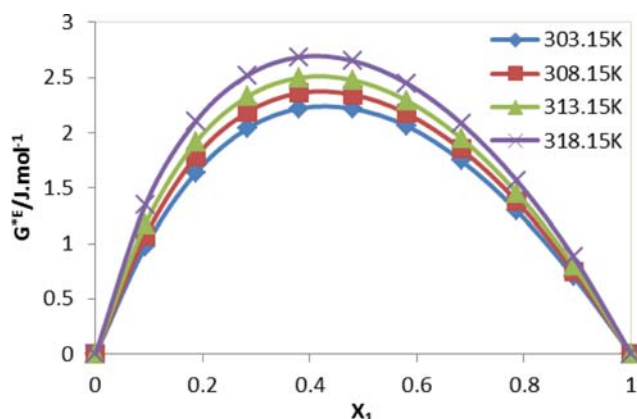


Fig. 7. Plot of deviations in Gibbs free energy (G^E) against mole fraction of (X_1) at $T=303.15$ K (◆), 308.15 K (■), 313.15 K (▲) and 318.15 K (×) for the PPH+*m*-xylene system.

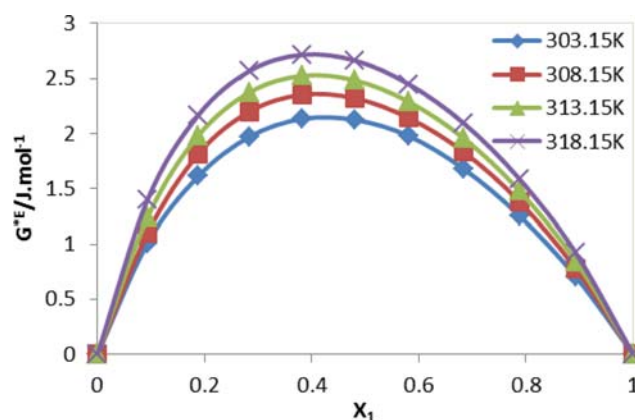


Fig. 8. Plot of deviations in Gibbs free energy (G^E) against mole fraction of (X_1) at $T=303.15$ K (◆), 308.15 K (■), 313.15 K (▲) and 318.15 K (×) for the PPH+*p*-xylene system.

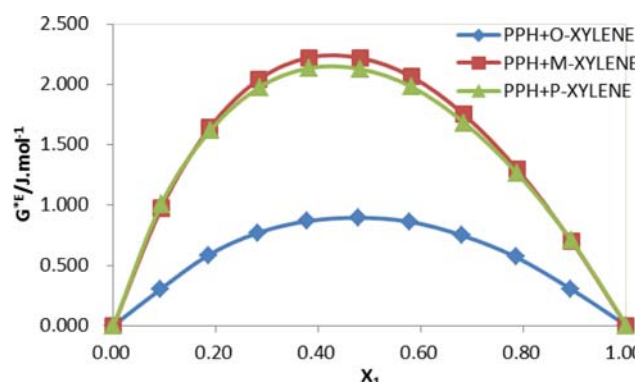


Fig. 9. Plot of deviation in Gibbs free energy (G^E) against mole fraction of (X_1) for PPH+*o*-xylene (◆), PPH+*m*-xylene (■), and PPH+*p*-xylene (▲) binary systems at $T=303.15$ K.

Table 5. Standard deviation percentage values against viscosity relations of the binary mixtures of Propiophenone with *o*-xylene, *m*-xylene and *p*-xylene at $T=(303.15-318.15)$

Binary mixture	T/K	Kendall-Munroe	Bingham	Arrhenius	Kendall
		σ %	σ %	σ %	σ %
PPH+ <i>o</i> -xylene	303.15	0.95	2.27	1.42	2.52
	308.15	1.25	2.19	1.79	2.92
	313.15	1.58	2.10	2.21	3.37
	318.15	2.06	1.92	2.78	3.97
PPH+ <i>m</i> -xylene	303.15	3.42	2.32	5.04	6.17
	308.15	3.73	2.22	5.46	6.57
	313.15	4.03	2.15	5.87	6.97
	318.15	4.45	1.98	6.42	7.51
PPH+ <i>p</i> -xylene	303.15	3.40	2.01	4.97	6.00
	308.15	3.89	1.74	5.57	6.58
	313.15	4.27	1.61	6.07	7.08
	318.15	4.69	1.49	6.61	7.62

mixtures are positive for the entire composition range except some stray points of F_{12} where some negative values were observed, which supports the existing specific interactions for all the three binary liquid mixtures.

6. Tamara-Kurata (T_{12}) and Hind et al. (H_{12})

The values of single interaction parameters of Tamara-Kurata (T_{12}), and Hind et al. (H_{12}) do not differ appreciably from each other. This is in agreement with Fort and Moore [30] in regard to the nature of parameters T_{12} and H_{12} . Positive values are observed for all the three binary liquid mixtures over the entire composition range, and the values are decreasing with increase in temperatures from 303.15 to 318.15 K.

7. Viscosity Relations

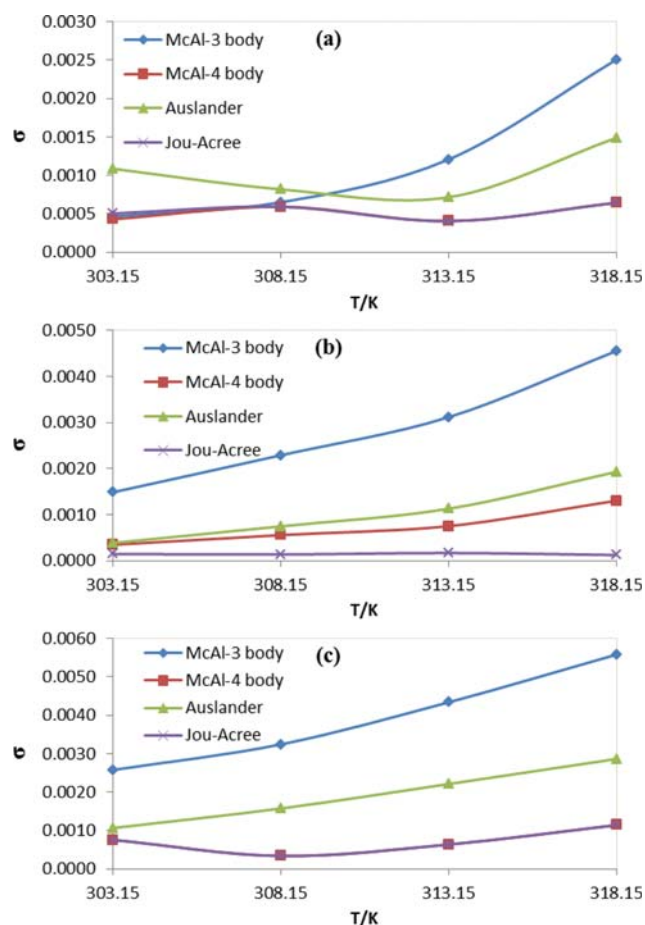
Viscosity values were calculated by using the relations of Kendall-Monroe, Bingham, Arrhenius-Eyring's, and Kendall. Results were compared with the experimental data. Average standard deviation percentage (σ %) values were calculated and tabulated in Table 5. Bingham values (σ % = 1.92 to 2.27 for PPH+*o*-xylene, 1.98 to 2.32 for PPH+*m*-xylene and 1.49 to 2.01 for PPH+*p*-xylene) were consistent at all the three binary mixtures, whereas other three relation values were comparatively higher.

8. Viscosity Relations with Two and Three Interaction Parameters

Viscosity data were used to calculate the two/three interaction parameters by correlating with the McAllister's three/four body equations, Heric relation (two parameters), Auslander (three adjustable parameters) and Jouyban-Acree (three interaction parameters) relations and respective standard deviations with the method of least squares. Interaction coefficients, standard deviation values and standard deviation percentages from these relations are tabulated in Table 6 and Table 7.

Interaction parameters of the respective model equations have theoretical significance and are functions of interaction energies. Depending on the intermolecular interactions the parameter values may be positive or negative.

In the McAllister's three body model equation (two interaction parameters), the standard deviation percentage values (σ %) are ranging from 0.22 to 2.63 for PPH+*o*-xylene, 2.01 to 7.43 for PPH+*m*-

**Fig. 10.** Standard deviation values of McAllister three (◆)/four body (■) models, Auslander (▲) relation and Jouyban-Acree (×) relation for the binary mixtures of PPH+[(a) *o*-xylene, (b) *m*-xylene and (c) *p*-xylene] at $T=(303.15-318.15)$ K.

xylene, 3.35 to 17.79 for PPH+*p*-xylene at temperatures from (303.15 to 318.15) K. Heric equation's σ % values are higher than the McAl-

Table 6. Calculated values of interaction parameters for the binary mixtures of Propiophenone with *o*-xylene, *m*-xylene and *p*-xylene at $T=(303.15\text{-}318.15) \text{ K}$

Binary mixture	T/K	McAllister three body			Heric equation			McAllister four body			Auslander equation			Jouyban-Acree equation		
		a	b	σ	α_{12}	α_{21}	σ	A	B	C	σ	B12	A21	B21	σ	σ
PPH + <i>o</i> -xylene	303.15	1.2370	0.9780	0.0005	0.3819	-0.1029	0.0407	1.2987	0.7374	0.9039	0.0004	50.0588	59.7201	0.0172	0.0011	44.4842
	308.15	1.1948	0.9422	0.0006	0.4177	-0.1202	0.0431	1.2602	0.7068	0.8696	0.0006	49.0363	57.8785	0.0173	0.0008	51.2157
	313.15	1.1526	0.9066	0.0012	0.4475	-0.1365	0.0438	1.2229	0.6752	0.8361	0.0004	48.2238	56.3519	0.0167	0.0007	58.2883
PPH + <i>m</i> -xylene	303.15	1.1110	0.8739	0.0025	0.4807	-0.1553	0.0435	1.1893	0.6420	0.8065	0.0006	98.5317	113.2969	0.0050	0.0015	67.4164
	308.15	1.1862	0.9082	0.0015	0.3819	-0.1029	0.0025	1.2716	0.6834	0.8090	0.0003	52.8221	60.4910	0.0169	0.0004	109.0201
	313.15	1.1469	0.8804	0.0023	0.4177	-0.1202	0.0020	1.2363	0.6561	0.7840	0.0006	52.2788	59.4369	0.0167	0.0007	115.5263
PPH + <i>p</i> -xylene	303.15	1.1073	0.8514	0.0031	0.4475	-0.1365	0.0030	1.2005	0.6286	0.7581	0.0008	51.5205	58.3137	0.0163	0.0011	121.9184
	308.15	1.0681	0.8254	0.0046	0.4807	-0.1553	0.0042	1.1685	0.5992	0.7363	0.0013	50.2331	56.3169	0.0156	0.0019	130.1910
	313.15	1.1934	0.9226	0.0026	0.3819	-0.1029	0.0023	1.2855	0.6833	0.8296	0.0008	51.8533	58.6845	0.0160	0.0011	104.4918
PPH + <i>p</i> -xylene	303.15	1.1560	0.8975	0.0032	0.4177	-0.1202	0.0029	1.2512	0.6580	0.8055	0.0003	49.4304	55.0054	0.0162	0.0016	114.4322
	308.15	1.1160	0.8690	0.0043	0.4475	-0.1365	0.0038	1.2168	0.6290	0.7804	0.0006	41.9448	46.3246	0.0187	0.0022	122.1693
	318.15	1.0758	0.8409	0.0056	0.4807	-0.1553	0.0050	1.1826	0.5999	0.7555	0.0011	48.8059	53.4739	0.0144	0.0029	130.7757

Table 7. Standard deviation percentage ($\sigma\%$) values calculated with the viscosity models for the three binary mixtures at $T=(303.15\text{-}318.15) \text{ K}$

Binary mixture	T/K	McAlister three body		McAlister four body		Auslander		Jouy-Acree	
		$\sigma\%$	$\sigma\%$	$\sigma\%$	$\sigma\%$	$\sigma\%$	$\sigma\%$	$\sigma\%$	$\sigma\%$
PPH+ <i>o</i> -xylene	303.15	0.22	0.42	0.10	0.23				
	308.15	1.08	0.61	0.07	0.61				
	313.15	2.63	0.46	0.07	0.46				
	318.15	2.62	0.91	0.17	0.91				
PPH+ <i>m</i> -xylene	303.15	3.38	0.56	0.03	0.56				
	308.15	2.01	0.39	0.08	0.39				
	313.15	7.43	2.51	0.14	2.51				
	318.15	3.34	1.37	0.27	1.37				
PPH+ <i>p</i> -xylene	303.15	17.79	0.70	0.13	0.70				
	308.15	3.35	0.19	0.18	0.19				
	313.15	5.51	0.82	0.27	0.81				
	318.15	6.84	1.55	0.37	1.55				

lister three body model.

Among McAllister three/four body models and Auslander, the values of a , b , A , B , C , B_{12} , A_{21} and B_{21} adjustable parameters are positive, supporting weak type of molecular interactions for all the three binary liquid mixtures. Parameter values of Jouyban-Acree and Heric relations are positive and negative; it indicates non-linear molecular interactions. The standard deviation values are ranging from (0.0003-0.0013) for the McAllister's four body model and Jouyban-Acree models followed by Auslander ranging from (0.0004-0.0029) for the three interaction parameter relations of the studied binary

mixtures. Fig. 10 shows the standard deviation values for the McAllister's 3 body and 4 body, Auslander and Jouyban-Acree relations.

McAllister's three and four body model parameter values are decreasing with increase in temperature, and σ values are increasing with increase in temperatures for all the three binary mixtures. In the two adjustable parameter relations, McAllister's three body model standard deviation values are having more satisfactory results than the Heric equation, and in the three adjustable parameter relations, Jouyban-Acree relation followed by McAllister's four body models and Auslander relations are in good agreement with

Table 8. Coefficients of Redlich-Kister Polynomial Equation & standard deviation values of binary systems PPH+*o*-xylene, PPH+*m*-xylene and PPH+*p*-xylene at $T=(303.15-318.15)$ K

Property	Temp. (K)	A0	A1	A2	A3	A4	σ
<i>PPH+o-xylene</i>							
V^E ($\text{cm}^3 \cdot \text{mol}^{-1}$)	303.15	-0.8670	0.0753	0.1743	0.2326	0.0239	0.0006
	308.15	-0.9874	0.1634	0.0159	0.1234	0.0129	0.0006
	313.15	-1.0855	0.1856	-0.1532	-0.0724	0.0214	0.0006
	318.15	-1.1711	0.2226	-0.4018	0.0403	0.0642	0.0010
$\Delta\eta$ (mPa.s)	303.15	-0.1371	-0.0001	0.0186	0.0040	-0.0532	0.0002
	308.15	-0.1297	0.0027	0.0413	-0.0057	-0.0691	0.0002
	313.15	-0.1208	0.0012	0.0290	-0.0022	-0.0117	0.0003
	318.15	-0.1093	0.0039	0.0458	-0.0095	0.0080	0.0001
G^{*E} ($\text{J} \cdot \text{mol}^{-1}$)	303.15	3.5637	-0.4594	0.5014	0.1204	-1.1909	0.0046
	308.15	4.0591	-0.6076	1.1561	-0.0624	-1.7203	0.0065
	313.15	4.6441	-0.8486	1.2737	-0.1121	-0.9524	0.0066
	318.15	5.4238	-0.9560	1.3102	-0.8633	0.5449	0.0016
<i>PPPH+m-xylene</i>							
V^E ($\text{cm}^3 \cdot \text{mol}^{-1}$)	303.15	-1.0546	0.1157	-0.3391	0.0005	0.0003	0.0005
	308.15	-1.2069	0.0619	-0.4436	0.1543	-0.0513	0.0010
	313.15	-1.3406	0.0224	-0.5897	0.3193	-0.2006	0.0010
	318.15	-1.4561	0.0558	-0.7375	0.1754	-0.1290	0.0007
$\Delta\eta$ (mPa.s)	303.15	-0.1290	-0.0002	0.0189	-0.0028	-0.0203	0.0001
	308.15	-0.1218	0.0001	0.0433	-0.0056	-0.0390	0.0001
	313.15	-0.1155	-0.0001	0.0361	-0.0070	0.0001	0.0002
	318.15	-0.1054	0.0012	0.0470	-0.0087	0.0225	0.0000
G^{*E} ($\text{J} \cdot \text{mol}^{-1}$)	303.15	8.8062	-2.3546	0.8256	-0.3326	0.1816	0.0057
	308.15	9.2914	-2.5668	1.8766	-0.6871	-0.8154	0.0036
	313.15	9.8274	-2.7662	1.7258	-1.0290	0.4227	0.0039
	318.15	10.5137	-3.0182	2.1063	-1.6684	1.4205	0.0033
<i>PPH+p-xylene</i>							
V^E ($\text{cm}^3 \cdot \text{mol}^{-1}$)	303.15	-1.1640	0.0698	-0.3132	0.0310	0.0210	0.0010
	308.15	-1.3039	0.0813	-0.4817	0.1302	-0.1362	0.0015
	313.15	-1.4072	0.1320	-0.6817	-0.0138	-0.1179	0.0009
	318.15	-1.5508	0.0930	-0.8723	0.0736	-0.0360	0.0016
$\Delta\eta$ (mPa.s)	303.15	-0.1169	0.0003	0.0234	0.0006	0.0237	0.0002
	308.15	-0.1016	-0.0001	0.0644	0.0029	-0.0264	0.0003
	313.15	-0.0934	-0.0067	0.0658	0.0176	0.0054	0.0000
	318.15	-0.0851	-0.0006	0.0669	0.0034	0.0370	0.0003
G^{*E} ($\text{J} \cdot \text{mol}^{-1}$)	303.15	8.4468	-2.1655	0.9909	-0.8847	1.2265	0.0036
	308.15	9.1832	-2.7434	2.4338	-0.3888	-0.5695	0.0026
	313.15	9.8300	-2.9787	2.5678	-0.8661	0.4963	0.0013
	318.15	10.5453	-3.2660	2.7074	-1.3630	1.7089	0.0041

the experimental data of the studied three binary mixtures. Among all the correlative approaches, McAllister three/four body models have a good number of correction terms. In the overall analysis, as the number of interaction adjustable parameters increases, the predicting ability of the correlating relations gives better results.

The values of excess molar volume (V^E), deviations in viscosity ($\Delta\eta$) and excess Gibbs Energy (G^E) with respect to the mole fraction at temperatures from 303.15 K to 318.15 K were fitted to the Redlich-Kister Equation [34] of the type:

$$Y^E = x_1 x_2 \{ a_0 + a_1(x_1 - x_2) + a_2(x_1 - x_2)^2 + \dots + a_4(x_1 - x_2)^4 \} \dots \quad (20)$$

where, Y^E is for V^E , $\Delta\eta$ and G^E in the polynomial degree. The values of a_0 , a_1 , a_2 , a_3 , and a_4 are the coefficients of the polynomial equation and were obtained by the method of least-squares and are given in Table 8 along with standard deviation values. The standard deviations are calculated by using the equation:

$$\sigma(Y^E) = \left\{ \frac{\sum (Y_{obs}^E - Y_{cal}^E)^2}{n - m} \right\}^{1/2} \quad (21)$$

where n is the total number of experimental points and m is the number of coefficients.

CONCLUSIONS

Densities and viscosities of Propiophenone with *o*-xylene, *m*-xylene and *p*-xylene were measured over the entire composition range at $T=(303.15$ to $318.15)$ K. Excess molar volume (V^E), deviation of viscosities ($\Delta\eta$) and excess Gibbs free energy (G^E) of activation for viscous flow were calculated by using the experimental data, and the results were correlated with Redlich-Kister polynomial equation to compute the coefficients and standard deviations. The G^E , W_{vis}/RT , H_{12} , d_{12} , F_{12} and T_{12} values were positive for all the three binary mixtures, which suggests weak intermolecular interactions. Viscosity data were compared with several viscosity relations like Kendall-Monroe, Bingham, Arrhenius, and Kendall. Experimental viscosity data correlated with the McAllister three/four body models, Heric, Auslander and Jouyban-Acree relations. Based on the standard deviation percentage values, Auslander relation, Jouyban-Acree relation and McAllister's four body model are in good agreement with the experimental data. It is concluded that the increase in the number of parameters gives better predicting ability. These findings would be helpful to control the process parameters and manufacture equipment where accurate design is required for various chemical and industrial processes, such as separation of chemicals, fluid flow, heat flow or chemical reactions.

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