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A Study of Correlative Modeling on Viscosity for Organic Binary Liquid Systems

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Abstract

In order to systematically investigate correlating capability of viscosity models, six well-known correlative models of Grunberg-Nissan (GN), Hind (HND), Heric (HRC), Ausländer (AUS) for dynamic viscosity, η, and McAllister 3-body (MAC3) and McAllister 4-body (MAC4) for kinematic viscosity, ν, were employed and tested for viscosity data of 83 organic binary liquid systems consisting of 33 different aromatic hydrocarbons (ArH), alkanes (RH), cycloalkanes (CyRH) and alkanols (ROH). Keeping ArH as a common component and increasing the chain length of other components, the systems were categorized as Category 1: ArH + RH, Category 2: ArH + CyRH, Category 3: ArH + ArH and Category 4: ArH + ROH. For all the models fitting parameters along with the statistical parameters such as SPD σ(%), ASPD σ(%), OASPD σ(%) and GOASPD σ(%) were computed by the Nonlinear Least Squares Minimization (NLSM) technique with the 'Solver' add-in package. Among the four categories, Category 3, OASPD, σ(%) values lie between 0.16 and 0.21, indicating that all the models fitted extremely well. However, for dynamic viscosities, the η AUS model demonstrated the best correlating capabilities with GOASPD σ(%) of 0.48, while the HND model performed the poorest with GOASPD σ(%) at 3.8. For the kinematic viscosities, ν in both the MAC3 and MAC4 models yielded satisfactory results with GOASPD σ(%) as < 1.0 %.

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Keywords: *Binary Liquid System, Viscosity, Correlative Model, Ausländer Model, McAllister Model.*

1. Introduction

Studies on thermodynamic and transport properties of multi-component liquid mixtures are essential, and the knowledge obtained from their theoretical and experimental study is directly applied in chemical engineering designs, calculations and modeling [1]–[3]. Among the various properties, viscosity is considered as one of the fundamental transport properties of liquid systems. Viscosity is increasingly used to understand internal organizations and the nature of the intermolecular force of interactions of pure components and constituents of liquid mixtures [4]–[6]. In addition, a comprehensive knowledge of viscosity is required for the determination of flow in pipelines and capillaries, heat-transfer and mass-transfer operations, developing separation methods and surface facilities, chemical engineering designs and disciplines, as well as energy transference calculations

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[7]–[12]. From the theoretical point of view, by employing different correlative and predictive models, the data obtained for viscosity of binary liquid mixtures are directly utilized to calculate excess thermodynamic properties and to test their relative correlative abilities [13],[14]. However, correlative models contain (one or more) adjustable parameters of which, their values are determined by fitting those models to experimental mixture data. The use of some optimization techniques as well as computer simulation correlative models usually leads to better results [15]–[17]. From the work of Arrhenius in 1877 and by the appearance of at least 50 empirical or semi-theoretical equations, a large number of researchers carried out investigations to describe the viscosity of such binary liquid mixtures and their continued theoretical and practical interests may also play a significant role in determining the method for prediction of this property [18]. So, for calculating the viscosity of binary liquid mixtures, there is a need for a compatible, solid, authentic and accurate analytical predictive method. Its popularity would increase due to process and reservoir simulators [19].

Various studies have explored methods for correlating Newtonian viscosity in liquid hydrocarbon and petroleum fluid mixtures, offering practical engineering applications. Monnery et al. [19] reviewed these methods, while Qunfang et al. [18] correlated viscosities of 47 binary liquid mixtures with a 1.05% overall average absolute deviation. Mirjana et al. [3] utilized Teja-Rice and McAllister models for 31 binary liquid mixtures, analyzing results based on the mixture's structure, component nature, and alcohol molecule alkyl chain length. Qiao et al. [10] determined viscosities for acetic acid and water, fitting data with a correlation equation, resulting in a 2.48% average absolute deviation. Knežević et al. [2] tested 42 correlation models, with Heric I, Heric–Brewer II, and Krishnan–Laddha models showing the best correlative characteristics (overall absolute average deviation < 2%). Bajić et al. [15] calculated viscosities for ionic liquid and organic compound mixtures using various models, comparing results with literature data. Dey et al. [4] evaluated benzene and cyclohexane binary system viscosities using 16 correlative approaches, assessing predictive capabilities through the absolute average percentage deviations (AAPDs). Recently, Ramachandran et al. [12] measured viscosities of binary mixtures and employed multiple correlation models, including Grunberg-Nissan, Katti and Chaudhri, Hind, Tamara Kurata, Frenkel, McAllister's three body, four body models, Heric, Auslander, and Jouyban-Acree relations. The Kendall-Monroe, Bingham, Arrhenius, and Kendall viscosity relations were used to calculate and compare standard deviation percentages (σ%) between experimental and calculated viscosity data. Rocky et al. [32] investigated correlative models like Grunberg-Nissan, Hind, Heric, Ausländer, McAllister 3-body, and 4-body equations. They found that models with more parameters yielded satisfactory results, and the quality of correlations depended on the symmetry of model equations, molecular interactions, and the number of available data points. The study concluded that specific intermolecular interactions influenced the deviations, with higher-parameter models showing potential for accurate predictions.

In the extensive literature on binary liquid mixtures, numerous viscosity models have been proposed, encompassing theoretical, semi-theoretical, empirical, and semi-empirical approaches. Despite the many models, only a few models exhibit reliable predictive accuracy for mixture viscosity. Furthermore, previous studies often lack a systematic approach in selecting mixtures for viscosity calculations, leading to a deficiency in systematically investigating models. To address this gap, there is a need for a more systematic approach which involves organizing models based on increasing interaction parameters and arranging mixtures in a sequential manner according to the ascending chain length of their components. By adopting this systematic approach, with one component of the mixture serving as a common factor, the correlating capability and versatility of the models can be accurately determined.

In the present study, we are interested to investigate some of those existing correlating models for the viscosity of binary liquid systems. Utilizing experimental viscosity data from previous literatures, the applicability of some well-known correlating models, viz., one parameter-based Grunberg-Nissan (GN) [20] and Hind (HND) [21], two parameter-based Heric (HRC) [22], three parameter-based Auslander (AUS) for dynamic viscosities (η), and the two parameter-based McAllister 3-body (MAC3) and three parameter-based McAllister 4-body (MAC4) [23] models for kinematic viscosities (ν), were tested. In this regard, we have considered a very large number of liquid–liquid binary systems mainly consisting of a variety of hydrocarbons of different chemical structures and molecular sizes, *viz.*, aromatic hydrocarbons (ArH), alkanes (RH), cycloalkanes (CyRH) and some alkanols (ROH), where the systems were chosen such that in all cases an ArH would always be a common component. In view of pursuing a systematic analysis, the systems under consideration were classified into the following categories: ArH + RH, ArH + ArH, ArH + CyRH and ArH + ROH. Employing the dynamic viscosities, η and kinematic viscosities, ν data in the whole range of composition, $0 \leq x_2 \leq 1$, at temperatures between T = 298.15 and 323.15 K under 1 atm pressure (collected from literatures), they were correlated with six standard models. The relevant coefficients/interaction parameters and standard percentage deviations, $\sigma(\%)$ were calculated. Thorough the statistical data analysis finally the correlating capability and versatility of the models are ascertained.

2. Methodology

Various equations or laws of mixing can be defined according to the number of adjustable parameters/interactions in use to compute the extent of deviations of binary liquid systems. In our present investigation, the following correlative models were used.

2.1. Dynamic Viscosity Models

Grunberg and Nissan (GN) model equation:

$$\mathrm{Log}\eta_s = N_1\log\eta_1 + N_2\log\eta_2 + N_1N_2G_{12} \quad (1)$$

where, G_{12} is an interaction parameter.

Hind (HND) model equation:

$$\eta_{mix} = x^2\eta_{11} + 2x(1-x)\eta_{12} + (1-x)^2\eta_{22} \quad (2)$$

where, η_{12} is an interaction parameter.

Heric (HRC) model equation:

$$\ln\eta = x_1\ln\eta_1 + x_2\ln\eta_2 + x_1\ln M_1 + x_2\ln M_2 - \ln[x_1M_1 + x_2M_2] + x_1x_2\{H_{12} + H_{21}(x_1 - x_2)\} \quad (3)$$

where, H_{12} and H_{21} are the requisite adjustable parameters.

Ausländer (AUS) model equation:

$$\eta = \{x_1\eta_1(x_1 + B_{12}x_2) + x_2\eta_2A_{21}(B_{21}x_1 + x_2)\}/\{x_1(x_1 + B_{12}x_2) + x_2A_{21}(B_{21}x_1 + x_2)\} \quad (4)$$

where B_{12}, B_{21} and A_{21} are the adjustable parameters.

2.2. Kinematic Viscosity Models

McAllister 3-body (MAC3) model equation:

$$\ln\eta = x_1^3\ln\eta_1 + 3x_1^2x_2\ln Z_{12} + 3x_1x_2^2\ln Z_{21} + x_2^3\ln\eta_2 - \ln[x_1 + x_2(M_2/M_1)] + 3x_1^2x_2\ln[(2 + M_2/M_1)/3] + 3x_1x_2^2\ln[(1 + 2M_2/M_1)/3] + x_2^3\ln(M_2/M_1) \quad (5)$$

McAllister 4-body (MAC4) model equation:

$$\ln\eta=x_1^4\ln\eta_1+4x_1^3x_2\ln Z_{1112}+6x_1^2x_2^2\ln Z_{1122}+4x_1x_2^3\ln Z_{2221}+x_2^4\ln\eta_2\ln[x_1+x_2(M_2/M_1)]+4x_1^3x_2\ln[(3+M_2/M_1)/4]+6x_1^2x_2^2\ln[(1+M_2/M_1)/2]+4x_1x_2^3\ln[(1+3M_2/M_1)/4]+x_2^4\ln(M_2/M_1) \quad (6)$$

Here, Z_{12}, Z_{21}, Z_{1112}, Z_{1122}, and Z_{2221} represent the interaction parameters and M_i is the molar mass of pure component i.

2.3. Data optimization

A Microsoft Excel Macro Based Spreadsheet was made especially with the 'Solver' add-in package for calculating all the selected binary systems. By setting the 'Solver' in the spreadsheet, prior to applying the Nonlinear Least Squares Minimization (NLSM) technique, all of the coefficients/interaction parameters of the correlative models [Equations (1) to (6] were obtained [25]–[27]. Comparisons were then made with available literature values for those binary systems in which any one of the correlative models was applied by previous investigation [28]–[33] using similar optimization techniques.

3. Results and discussion

In the present investigation, all the Alkanes (RH), Cycloalkanes (CyRH), Aromatic Hydrocarbons (ArH), and Alcohols (ROH) were of simple molecular forms with different sizes and shapes. Table 1 lists the organic liquids with their short name and CAS number. Altogether, there are 83 binary systems; an Aromatic Hydrocarbon is taken as the common component in each system. The systems were chosen according to the increasing chain length and categorised into four types. The first category is ArH + RH systems and there are 22 systems within the temperature range of 298.15 – 323.15 K. There are 11 systems of ArH + CyRH in the second category and their data are at temperatures also within the same temperature range. The third category belongs to ArH + ArH type with a total of 9 systems and their data are at temperatures between 298.15 – 313.15 K. In the fourth category, 41 ArH + ROH type systems within the temperature range 298.15 – 323.15 K are considered. All the experimental data of η and ν were obtained from previous publications [29], [30], [34]–[56].

Table 1. List of different types of organic liquids under investigation.

Compound	**Short Name**	**CAS Number**
Aromatic Hydrocarbons (ArH)		
benzene	Bn	71-43-2
toluene	Tn	108-88-3
ethylbenzene	EBn	100-41-4
o-xylene	*o*-Xy	95-47-6
m-xylene	*m*-Xy	108-38-3
p-xylene	*p*-Xy	106-42-3
mesitylene	MST	108-67-8
Alkanes (RH)		
pentane	C_5	109-66-0
hexane	C_6	110-54-3
heptane	C_7	142-82-5
octane	C_8	111-65-9
nonane	C_9	111-84-2
decane	C_{10}	124-18-5
dodecane	C_{12}	112-40-3

Compound	Short Name	CAS Number
Cycloalkanes(CyRH)		
cyclohexane	CyC_6	110-82-7
cyclooctane	CyC_8	292-64-8
Alkanols (ROH)		
methanol	MeOH	67-56-1
ethanol	EtOH	64-17-5
2-methylpropanol-1	2-MePrOH-1	78-83-1
2-methylpropanol-2	2-MePrOH-2	75-65-0
propan-1-ol	PrOH	71-23-8
propan-2-ol	2-PrOH	67-63-0
butan-1-ol	BuOH	71-36-3
butan-2-ol	2-BuOH	78-92-2
pentan-1-ol	PnOH	71-36-3
pentan-2-ol	2-PnOH	6032-29-7
hexan-1-ol	HxOH	111-27-3
hexan-2-ol	2-HxOH	626-93-7
heptan-1-ol	HpOH	111-70-6
heptan-2-ol	2-HpOH	543-49-7
octan-1-ol	OcOH	111-87-5
octan-2-ol	2-OcOH	123-96-6
decan-1-ol	DcOH	112- 30-1

In order to examine the correlating capability of the models for viscosity, the one parameter-based Grunberg and Nissan (GN) and Hind (HND) equations, two parameter-based Heric (HRC), and the three parameter-based Ausländer (AUS) models were used fitting the experimental data of dynamic viscosities, η. On the other hand, the two parameter-based McAllister 3-body (MAC3) and three-parameter McAllister 4-body (MAC4) equations, were correlated with the kinematic viscosities, ν, where, $\nu = \eta/\rho$.

To achieve the main objectives, all systems fitting parameters of each viscosity model were calculated. Also, the correlating ability of each model was tested by calculating the standard percentage deviation (SPD), σ%, between the experimental and calculated values.

$$\sigma(\%) = \left[\left(\frac{1}{n-p} \right) \sum \left(100 \times \frac{\eta_{\text{exp}} - \eta_{\text{cal}}}{\eta_{\text{exp}}} \right)^2 \right]^{1/2} \tag{7}$$

Here, η_{exp} and η_{cal} refer to experimental and calculated viscosity (η and ν), n represents the number of data points and p is the number of coefficients considered. All the coefficients of equations (1) to (6) were obtained using the Non-linear Regression Analysis. For all systems, the values of σ% at different temperature ranges and available literature fitting coefficients are detailed in Supporting Information (Tables S1-S8). Table 2 to Table 5 list all the systems under the 4 different categories along with their data sources, data points, temperature ranges, temperature points, and Average Standard Percentage Deviations, ASPD σ (%) as well as Overall Average Standard Percentage Deviations, OASPD σ (%) of the models. Table 6 summarizes the overall correlating abilities of all six models and calculates their Grand Overall Average Standard Percentage Deviations, GOASPD σ (%). The category-wise results of the fitting parameters and σ (%) for all 83 systems are discussed in section 3.1 to 3.4.

Table 2. Average standard percentage deviations (ASPD), σ(%) for Category 1: ArH + RH.

No.	**Systems**	**Data Points**	**Temp. Range (K)**	**Temp. Points**	**ASPD σ(%)**					
					GN	HND	HRC	AUS	MAC3	MAC4
1	benzene + hexane [44]	16	298.15	1	1.09	2.88	0.57	0.28	0.57	0.44
2	benzene + heptane [51]	11	308.15-313.15	2	1.68	1.94	0.55	0.51	0.55	0.52
3	benzene + octane [52]	11	298.15	1	1.63	1.97	0.50	0.33	0.48	0.36
4	benzene + decane [44]	16	298.15	1	1.80	1.37	0.74	0.31	0.72	0.23
5	benzene + dodecane [52]	12	298.15	1	1.57	0.99	0.34	0.15	0.24	0.22
6	toluene + pentane [53]	12	298.15	1	4.29	2.52	2.73	0.32	2.66	1.93
7	toluene + n-heptane [54]	11	298.15-323.15	6	0.44	0.82	0.07	0.08	0.08	0.08
8	toluene + octane [55]	11	308.15-313.15	2	0.54	0.45	0.16	0.15	0.15	0.15
9	toluene + decane [56]	7	298.15	1	0.55	0.26	0.13	0.03	0.10	0.05
10	ethylbenzene + hexane [57]	19	298.15	1	1.16	2.57	0.71	0.65	0.71	0.68
11	ethylbenzene + heptane [55]	11	308.15-313.15	2	0.35	0.51	0.27	0.27	0.18	0.17
12	ethylbenzene + octane [55]	11	308.15-313.15	2	0.31	0.31	0.24	4.13	0.24	0.26
13	o-xylene + hexane [57]	16	298.15	1	2.07	4.51	0.58	0.52	0.57	0.52
14	o-xylene + n-heptane [54]	11	298.15-323.15	6	0.32	1.14	0.09	0.08	0.09	0.08
15	o-xylene + decane [56]	7	298.15	1	0.36	0.24	0.10	0.02	0.10	0.04
16	m-xylene + hexane [57]	15	298.15	1	1.64	2.66	0.54	0.50	0.53	0.53
17	m-xylene + decane [34]	12	308.15-313.15	2	0.63	0.54	0.56	0.59	0.57	0.60
18	p-xylene + hexane [57]	15	298.15	1	1.30	2.53	0.25	0.06	0.25	0.10
19	p-xylene + n-heptane [35]	11	313.15-323.15	2	0.55	0.46	0.48	0.51	0.25	0.26
20	p-xylene + n-octane [35]	11	298.15-323.15	6	0.53	0.68	0.20	0.20	0.20	0.20
21	p-xylene + decane [56]	7	298.15	1	0.59	0.28	0.13	0.09	0.14	0.12
22	mesitylene + n-heptane [54]	11	298.15-323.15	6	0.09	0.40	0.08	0.08	0.07	0.08
Overall Average Standard Percentage Deviations, OASPD σ(%)					**1.07**	**1.37**	**0.46**	**0.45**	**0.43**	**0.35**

Table 3. Average standard percentage deviations (ASPD), σ(%) for Category 2: ArH + CyRH.

No.	**Systems**	**Data Points**	**Temp. Range (K)**	**Temp. Points**	**ASPD σ(%)**					
					GN	HND	HRC	AUS	MAC3	MAC4
1	benzene +cyclohexane [36]	7	298.15-313.15	2	0.31	1.20	0.19	0.05	0.17	0.05
2	benzene + cyclooctane [51]	11	308.15-313.15	2	0.30	4.32	0.31	0.33	0.31	0.33
3	toluene + cyclohexane [37]	11	298.15-323.15	5	1.01	1.86	0.13	0.12	0.13	0.13
4	toluene + cyclooctane [51]	11	308.15-313.15	2	1.87	7.32	0.92	0.94	0.92	0.91
5	ethylbenzene + cyclohexane [55]	11	308.15-313.15	2	1.39	2.20	0.37	0.11	0.37	0.13
6	ethylbenzene + cyclooctane [51]	11	298.15-313.15	2	1.84	6.64	0.45	0.40	0.45	0.37
7	o-xylene+cyclohexane [37]	11	298.15-323.15	5	0.94	1.14	0.14	0.05	0.13	0.05
8	m-xylene+cyclohexane [34]	12	308.15-313.15	2	1.55	2.44	0.68	0.33	0.68	0.38
9	m-xylene+cyclooctane [34]	11	308.15-313.15	2	2.63	8.59	0.65	0.55	0.63	0.57
10	p-xylene+cyclohexane [35]	11	298.15-313.15	3	1.62	2.39	0.47	0.39	0.47	0.41
11	mesitylene+cyclohexane [37]	11	303.15-323.15	5	2.13	2.63	0.54	0.09	0.52	0.13
Overall Average Standard Percentage Deviations, OASPD σ(%)					**1.39**	**3.45**	**0.41**	**0.28**	**0.41**	**0.29**

Table 4. Average standard percentage deviations (ASPD), σ(%) for Category 3: ArH + ArH.

No.	**Systems**	**Data Points**	**Temp. Range (K)**	**Temp. Points**	**Average Standard Percentage Deviations, ASPD σ(%)**					
					GN	HND	HRC	AUS	MAC3	MAC4
1	benzene + toluene [51]	11	313.15	1	0.13	0.13	0.12	0.13	0.13	0.13
2	benzene + ethylbenzene [51]	11	313.15	1	0.39	0.39	0.21	0.22	0.21	0.21
3	benzene + o-xylene [56]	7	298.15	1	0.21	0.17	0.16	0.16	0.16	0.17
4	toluene + ethylbenzene [51]	11	308.15-313.15	2	0.74	0.73	0.63	0.63	0.63	0.64
5	toluene + o-xylene [56]	7	298.15	1	0.01	0.06	0.01	0.01	0.01	0.01
6	toluene + p-xylene [56]	7	298.15	1	0.02	0.02	0.02	0.02	0.02	0.01
7	o-xylene + p-xylene [38]	7	298.15-303.15	2	0.01	0.03	0.01	0.01	0.01	0.01
8	o-xylene + m-xylene [38]	11	298.15-303.15	2	0.19	0.19	0.20	0.18	0.18	0.12
9	p-xylene + m-xylene [38]	11	298.15-303.15	2	0.09	0.09	0.08	0.09	0.08	0.08
Overall Average Standard Percentage Deviations, OASPD σ(%)					**0.21**	**0.21**	**0.17**	**0.17**	**0.17**	**0.16**

Table 5. Average standard percentage deviations (ASPD), σ(%) for Category 4: ArH + ROH.

No.	**Systems**	**Data Points**	**Temp. Range (K)**	**Temp. Points**	**Average Standard Percentage Deviations, ASPD σ(%)**					
					GN	HND	HRC	AUS	MAC3	MAC4
1	benzene + ethanol[39]	11	298.15	1	1.63	0.78	0.68	0.25	0.68	0.37
2	benzene +2-methylpropanol-1 [40]	11	303.15	1	2.02	6.05	0.34	0.15	0.34	0.19
3	benzene +2-methylpropanol-2 [40]	11	303.15	1	2.77	17.00	1.57	0.63	1.63	0.63
4	benzene + 1-butanol [40]	11	303.15	1	2.05	3.13	0.26	0.26	0.27	0.24
5	benzene + 2-butanol [40]	11	303.15	1	3.17	10.1	1.09	0.67	1.10	0.89
6	benzene + 1-pentanol [41]	14	293.15	1	5.12	8.04	0.44	0.67	0.44	0.52
7	benzene + 1-hexanol [42]	11	303.15-308.15	2	3.53	7.63	1.07	1.21	1.42	1.15
8	toluene + ethanol [43]	11	303.15-308.15	2	1.40	0.49	0.48	0.50	0.46	0.49
9	toluene + 1-propanol [43]	11	303.15-313.15	3	1.14	4.82	1.12	0.78	1.13	0.98
10	toluene + butanol-1[43]	11	303.15-313.15	3	2.09	6.16	0.86	1.08	0.86	0.98
11	toluene + 2- methylpropanol-1[43]	11	303.15-313.15	3	9.33	30.2	3.15	1.46	3.17	0.71
12	toluene + 1-pentanol [43]	11	303.15-313.15	3	3.73	6.56	0.51	0.62	0.51	0.52
13	toluene + 1-heptanol [30]	21	303.15-313.15	2	3.41	9.95	1.86	1.06	1.88	1.10
14	toluene + 1-octanol [30]	21	303.15-313.15	2	2.07	14.0	1.50	1.12	1.51	1.13
15	toluene + 1-decanol [30]	21	303.15-313.15	2	3.33	18.1	3.36	2.90	3.38	2.51
16	toluene + 2-hexanol [29]	21	298.15-308.15	2	2.94	15.8	3.28	2.48	3.31	2.02
17	toluene + 2-heptanol [29]	21	298.15-308.15	2	4.79	16.3	2.53	2.42	2.52	2.47
18	toluene + 2-octanol [29]	21	298.15-308.15	2	6.49	18.6	2.17	1.98	2.18	1.96
19	ethylbenzene + ethanol [45]	10	298.15-308.15	2	0.58	1.29	0.47	0.50	0.38	0.40
20	ethylbenzene + 1-propanol [45]	9	298.15-308.15	2	1.67	6.62	1.91	0.51	1.95	0.61
21	ethylbenzene + 1-butanol [45]	10	298.15-308.15	2	1.89	8.70	1.88	1.86	1.86	1.87
22	ethylbenzene + 1-decanol [46]	16	298.15-308.15	2	6.66	34.6	0.43	0.42	0.43	0.42
23	o-xylene + methanol [47]	11	303.15-323.15	3	1.61	1.63	1.26	1.04	1.16	0.86
24	o-xylene + ethanol [47]	11	303.15-323.15	3	0.04	0.03	0.11	0.01	0.11	0.02
25	o-xylene + 1-hexanol [48]	11	308.15-318.15	2	2.80	4.87	0.56	0.72	0.57	0.64
26	o-xylene + 1-decanol [46]	16	298.15-308.15	2	6.26	20.3	1.05	0.54	1.05	0.49
27	m-xylene + ethanol [47]	9	313.15-323.15	2	0.34	0.36	0.34	0.35	0.36	0.32

28	m-xylene+1-hexanol [48]	11	308.15-318.15	2	2.79	8.53	0.61	0.70	0.62	0.67
29	m-xylene + 1-decanol [46]	16	298.15-308.15	2	4.32	26.1	0.17	0.23	0.17	0.12
30	p-xylene + ethanol [47]	8	303.15-323.15	3	0.32	0.29	0.24	0.24	0.26	0.26
31	p-xylene + n-propanol [49]	11	303.15-323.15	3	1.28	0.01	0.25	0.01	0.24	0.13
32	p-xylene + n-butanol [49]	11	303.15-323.15	3	2.97	0.40	0.77	0.30	0.84	0.38
33	p-xylene + 1-pentanol [41]	14	293.15	1	2.92	11.92	0.87	0.22	0.88	0.22
34	p-xylene + 1-hexanol [48]	11	308.15-318.15	2	2.58	9.24	0.59	0.65	0.60	0.60
35	p-xylene + 1-decanol [46]	16	298.15-308.15	2	7.30	25.9	0.68	0.50	0.67	0.47
36	mesitylene + ethanol [50]	11	298.15-308.15	3	1.47	1.81	1.06	0.93	1.07	0.81
37	mesitylene + 1-propanol [50]	11	298.15-308.15	3	1.96	5.28	1.45	1.27	1.46	1.29
38	mesitylene + propan-2-ol [50]	11	298.15-308.15	3	4.76	11.58	1.57	0.63	1.60	0.84
39	mesitylene + 1-butanol [50]	11	298.15-308.15	3	5.49	11.65	4.54	4.87	4.83	5.54
40	mesitylene + pentan-1-ol [50]	11	298.15-308.15	3	3.68	11.15	3.83	4.09	3.84	4.03
41	mesitylene + 1-decanol [46]	16	298.15-308.15	2	6.86	28.3	1.73	0.22	1.74	0.29
Overall Average Standard Percentage Deviations, OASPD σ(%)					**3.21**	**10.4**	**1.28**	**1.00**	**1.30**	**0.98**

Table 6. Overall correlating abilities of the models expressed as overall average standard percentage deviations **(**OASPD**)**, σ(%) for differentl categories of binary systems.

Category	**No. of Binary Systems**	**Total Temp. Points**	**Total Data Points**	**OASPD σ(%)**					
				GN	HND	HRC	AUS	MAC3	MAC4
ArH + RH	22	54	264	1.07	1.37	0.46	0.45	0.43	0.35
ArH + CyRH	11	43	136	1.39	3.45	0.41	0.28	0.41	0.29
ArH + ArH	9	22	94	0.21	0.21	0.17	0.17	0.17	0.16
ArH + ROH	41	87	533	3.21	10.4	1.28	1.00	1.30	0.98
Grand Overall Average Standard Percentage Deviations, GOASPD σ(%)				**1.47**	**3.86**	**0.58**	**0.48**	**0.58**	**0.45**

3.1. Category 1: ArH + RH

In Category 1 (ArH + RH), there are 22 binary liquid systems consisting of 7 aromatic hydrocarbons and 5 aliphatic hydrocarbons. Tables S1 and S2 provide the fitting parameters of dynamic and kinematic viscosities, η and υ, respectively within the temperature range, 298.15–323.15 K for the relevant models along with the computed standard percentage deviations, σ(%). The results for η & υ for ArH + RH exhibit that both patterns of deviations and their extents are different for the systems as well as for different models. As Tables 2 shows, ASPD σ(%) of η for the GN, HND, HRC and AUS models lie within the ranges of 0.09-4.29, 0.24-2.88, 0.07-2.73 and 0.02-4.13, respectively. It has been found that for the systems starting from Bn + C_6 to EBn + C_{10} as well as from *o*-Xy + C_6 to MST + C_7, i.e., where the hydrocarbons change from C_6 to C_{12}, the observed deviations decreased linearly. That means, for a particular ArH, the relevant deviations would decrease as the chain length of RH increases.

However, most systems deviations are the maximum for the HND and GN models, where both models contain a single interaction parameter. In the case of GN, despite the irregularity, deviations in chain length initially increase and then decrease until reaching the last the system in this category. The deviations for HND decrease as the chain length of RH increases. However, a comparison shows that the deviations for GN are lower than those of HND. On the other hand, deviations are lowered significantly for the three parameter-based AUS and the two parameter-based HRC equations. Results for both models are very close to each other, except for Ebn + C10, the AUS model yields the lowest deviation. Up to Tn + C_{10} system, deviations also decreased linearly with the increase in the chain length, continuing this trend until the last system, MST + C_7. Overall, deviations for dynamic viscosity η for all the systems under Category 1 decreased in the following order: AUS < HRC < GN < HND.

On the other hand, results for kinematic viscosities, ν, in Table 2 clearly show that compared to AUS, HRC, GN and HND models the deviations for the two parameter-based McAllister (MAC3) and three parameters-based McAllister (MAC4) models are relatively small as the ASPD σ(%) values for the latter are within the range of 0.07-2.66. Except for Tn + C_5, they are the least for Tn + C_7 and MST + C_7 for both the MAC3 and MAC4 models. Furthermore, deviations decrease linearly with the increasing chain length for different systems under this category and they continue to decrease till the last system, i.e., MST + C_7.

Again, it has been observed that the decrease is affected more or less while the ArH changes from Bn to Tn. With the increase of chain length of RH, deviations are found to decrease linearly. Correlations are slightly poor for the EBn system, but MST is consistently found to be excellent for both dynamic and kinematic viscosities as in the last system, MST + C_7. Again, deviations also seem to decrease with the increasing number of CH_3 as substituents (from Tn to Xys and MST). In view of the isomeric effect, observed deviations for *o*-xylene and *p*-xylene are lower than that of *m*-xylene. That means, other than the effect of counter RH chain length, fittings or correlations may improve when the ArH becomes bulkier with more substituents.

Table 2 exhibits the OASPD σ(%) values for HRC, AUS, MAC3 and MAC4, which are very close to one another and their correlating abilities appear to be satisfactory lying between 0.35–0.46. Meanwhile, those relevant values for the GN and HND models are all larger than 1.00, 1.07 and 1.37, respectively. The McAllister 4-body model thus is suggested to be the best with its OASPD of 0.35, and this is closely followed by the McAllister 3-body model. The AUS, HRC and HND models appear to be the poorest with the maximum ASPD value of 1.37. Thus, for Category 1, the order of increasing applicability of the studied models follows MAC4 > MAC3 > AUS > HRC > GN > HND.

3.2. Category 2: ArH + CyRH

In Category 2, there are ArH + CyRH type systems from 7 different ArH and 2 CyRH. Tables S3 and S4 provide the fitting parameters of the concerned models along with the detailed results of σ(%) for η and ν, respectively, and Table 3 summarizes ASPD and OASPD σ(%) for all the models under consideration. The ASPD σ(%) values are ranged between 0.30-2.63, 1.14-8.59, 0.13-0.92 and 0.05-0.94 for the GN, HND, HRC and AUS models respectively. It has also been found that, except for the GN and HND models, the ASPD σ(%) values for the HRC and AUS models for η as well as MAC3 and MAC4 for ν are very much close to one another, lying within the range of 0.05 – 0.94. So, for Category 2, the correlating abilities for all these four models are said to be quite satisfactory.

In view of the models compatibility, some deviations are quite random and significant for the HND and GN models. Interestingly, the observed deviations are the lowest for the three parameter-based AUS and two parameter-based HRC. The results for both the AUS and HRC models overlap and ASPD σ(%) increases with the increment of the alkyl chain length. On the other hand, the deviations for ν are even lower and they follow the trend: MAC4 < MAC3. Moreover, their results further reveal that deviations in ν also increase as the alkyl chain length attached to ArH increases.

The overall standard percentage deviation OASPD σ(%) are close to one another and are all < 1.00 except for those of the GN and HND models. Hence, all these models correlating abilities are considered entirely satisfactory. From Table 3, AUS for η and MAC4 model for υ represent excellent results with OASPD σ(%) as 0.28 and 0.29, respectively followed by MAC3 and HRC with the same OASPD σ(%) value of 0.41. On the other hand, the HND model appeared to be the poorest, with an OASPD σ(%) value maximum of 3.45 followed by GN with OASPD σ(%) of 1.39. For the systems in Category 2, the observed correlating abilities of the models ultimately follow the order of MAC4 ≈ AUS > MAC3 ≈ HRC > GN > HND.

3.3. Category 3: ArH + ArH

In Category 3, there are 9 ArH + ArH type systems consisting of 7 different aromatic hydrocarbons, ArH. Tables S5 and S6 provide the fitting parameters for all six models and their detailed results of relevant σ(%). Table 4 shows all the σ(%) values are in relatively low ranges for both η (0.01-0.74) and ν (0.01-0.64). Table 4 shows that the ASPD σ(%) values are minima, even for HND and GN. However, the maximum deviations are found from HND followed by the GN model, whereas the HRC and AUS models yielded highly satisfactory results with almost similar ASPD σ(%). Again, deviations are the highest for Tn + EBn even when applying the HRC and AUS models. For Bn + Tn to Tn + EBn, deviations increased as the alkyl chain length of ArH increased. As the deviations increase from Tn + o-Xy to Tn + m-Xy, increasing number of CH_3 show more significant deviations.

On the other hand, as Table 4 shows, the MAC3 & MAC3 models exhibit almost equal deviations and their ASPD σ(%) are of similar trend for all the systems in Category 3. Here, the system of Tn + EBn shows the largest deviations for both η and ν. However, the Tn + o-Xy system as well as the o-Xy + p-Xy system shows the best results with the ASPD σ(%) value as 0.01 for HRC & AUS (for η) and MAC3 & MAC4 (for ν).

As Table 4 depicts, all OASPD σ(%) values lie within the range of 0.16 – 0.21. Here also, the HND model appears to have the maximum OASPD value, 0.21, but it is still better with Category 3 compared to Categories I & 2. However, their OASPD σ(%) values are small and very close; hence, correlating abilities of all these six models are considered highly satisfactory for Category 3. The MAC4 with OASPD σ(%) 0.16% appears to fit the experimental ν the best, whereas MAC3, AUS and HRC also closely follow as their OASPD σ(%) is the same, and it is 0.17.

3.4. Category 4: ArH + ROH

In Category 4, 41 ArH + ROH type systems are formed from 7 ArH and 13 ROH Tables S7 and S8 summarize the fitting parameters along with the detailed results of σ(%) values for all of the models, while the corresponding ASPD σ(%) and OASPD σ(%) values are listed in Table 5. For the models, the ASPD σ(%) values range from 0.04 to 9.33 for GN, from 0.01 to 34.6 for HND, from 0.11 to 4.54 for HRC, and from 0.01 to 4.87 for AUS.

The detailed results of η also exhibit that for the systems in Category 4, the maximum deviations arise from HND followed by GN. For the systems starting from Bn + EtOH to MST + DcOH, the ASPD σ(%) of HND yielded abnormally high values especially for the systems containing DcOH. Otherwise, with the increasing chain length of ArH the increment in deviation continues till the last system. A similar trend is also observed for the GN model. The deviations are lowered down significantly for AUS followed by HRC. For the systems from Bn + EtOH to MST + DcOH, i.e., where the alkanol changes from C_2 to C_{10}, the deviations of both the models first tend to increase up to Tn + OcOH and then decrease. From Bn(1) + EtOH system to MST + DcOH, i.e., where the ArH changes from C_6 to C_9, the observed deviations are pretty satisfactory for C_8.

On the other hand, as Table 5 shows, ASPD σ(%) lies within the range of 0.11-4.83 and 0.02-5.54 for MAC3 and MAC4 models, respectively. Except for MST + BuOH, for most of the systems under Category 4, the least deviations are clearly due to the MAC4 model. Interestingly, deviations are affected more or less while the ArH changes from Bn to MST, i.e., with the increasing number of CH_3 as a substituent. Also, due to its isomeric effect, p-Xylene is found to yield slightly better results than *o*- and *m*-Xylenes.

As Table 5 exhibits, in Category 4, the OASPD σ(%) values of η for different systems are within a wide range of 1.00-10.40. Here, the HND also yielded the highest value and the deviations are within satisfactory range for AUS. On the other hand, the OASPD σ(%) values for MAC3 and MAC4 are 1.30 and 0.98, respectively. For the systems under Category 4, the correlating capability is found to follow the order: MAC4 > AUS > MAC3 ≈ HRC > GN >> HND.

Finally, Table 6 summarizes the OASPD σ(%) values of all the correlative models for all the systems under different categories. Table 6 and Figure 1 show that, among all of the investigated systems, viscosity data fit the models best for the systems under Category 3 (ArH + ArH type), where OASPD σ(%) values are 0.21 for GN & HND, 0.17 for HRC, AUS & MAC3 and 0.16 for MAC4. This may be attributed to the structural similarity between the component liquids under Category 3, which tend to behave ideally better. The results are reasonably suitable for the ArH + CyRH and ArH + RH type of systems; the former appears to be slightly better due to their cyclic structures compared to the linear structures of RH molecules. On the other hand, the systems in Category 4 have remarkably high OASPD σ(%) values for all the models. This may be attributed to the basic essential dissimilarity between component liquids. Here, one of the components is a hydrocarbon, RH, while the other is an alkanol, ROH. Among all these models for dynamic viscosity η, the correlating ability of AUS is the best and poorest for HND, whereas, for ν, the best fit is due to MAC4. The two parameter-based HRC and MAC3 yielded almost similar results. A comparison of their OASPD σ(%) further reveals that, as σ(%) values of HND are exceptionally high, especially for the ArH + ROH systems, this one parameter-based HND may not be suitable to correlate viscosities for binary solutions.

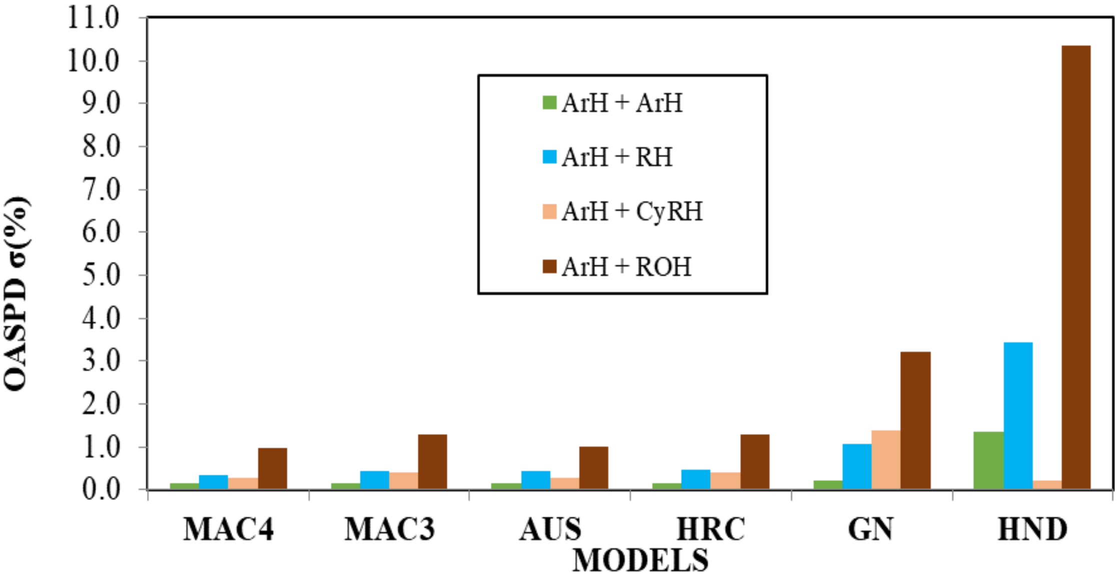

Figure 1: Representation of overall average standard percentage deviation **(**OASPD**)**, σ(%) for different categories of binary liquid systems.

Table 6 shows the results of the models as Grand Overall Average Standard Percentage Deviations, GOASPD σ(%), as: GN (1.47 %), HND (3.86%), HRC (0.58 %), AUS (0.48 %) , MAC3 (0.58 %) and MAC4 (0.45 %) for all systems. The results reveal that where the highest value belongs to the one-parameter HND model, the three parameters based MAC4 yielded the lowest value. The GOASPD σ(%) values are almost similar for the two-parameter MAC3 and HRC models. These results show that the greater the number of interaction parameters of the models, the better their fitting with experimental viscosity data would be.

The Hind model is the simplest known empirical equation, which is linear and known to work well only with ideal solutions. It employs an interaction parameter and simply taking the average of the individual viscosity values. From experimental data, viscosity does not follow a linear trend with a change in mole fraction where linear equations do not perform well [4]. An examination of Table 6 and Figure 1 reveals that the HND approach results in the highest deviations. The results align with expectations since the viscosity relation considers the simple additivity rule of the component liquids. The GN model is a modified Kendall-Munroe [57] model and involves one changeable parameter for binary systems having a linear relationship with mole fraction and logarithmic viscosity. As Tables 2 to 5 show, correlation results from GN are better than HND but not as good as the other models. Therefore, it can be suggested that, models following the rule of logarithmic additivity can correlate far better than the simple additive relations.

The HRC model is modified from an equation developed by Katti and Chaudhri [58] containing two interaction parameters. It has shown deviations less than those of the HND and GN models. Similarly, AUS is also an empirical equation with three interaction parameters and therefore, has shown deviations that are less than HND and GN and sometimes better than the HRC model.

On the other hand, McAllister's correlative approaches (MAC3 & MAC4) work much better than all others due to more significant number of interaction parameters. The parameters have their merits in ascertaining the strength of molecular interactions in liquid mixtures. The MAC3 approach is seen to work for like molecules with similar sizes. However, as this model considers only two-dimensional interactions of three molecules at a time, it may not give a correct picture considering the motion of a molecule in a mixture from one equilibrium position to another. The four-body interaction in MAC4 thus provides a more accurate representation involving a three-dimensional treatment. All 83 systems under the four categories thus fit much better with the MAC4 as evident from the relevant σ(%) values. The main reason may be attributed to the fact that the MAC4 approach considers that most of the possible interactions are taking place within the component liquids.

4. Conclusion

For all categories of systems, the correlative models under the present investigation yielded viscosity values, which are found to be of similar variational patterns to those shown by the experimental data. The statistical parameters such as SPD σ(%), ASPD σ(%), OASPD σ(% and GOASPD σ(%) exhibited their correlating capability as well as versatility. It has been established that, due to its purely additive nature, the HND model shows the highest deviations, GOASPD σ(%) of 3.86 and with very high σ(%), indicating that its fitting capability is the minimum. The correlating capabilities of 2 parameter-based AUS and 3 parameter-based MAC3 & MAC4 models are designated as the best ones having relatively low σ(%) values of 0.48, 0.58 and 0.45 respectively, and especially in case of liquid mixtures of higher order where the number of interactions is large and they are more or less of complex behavior.

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Conflict of Interest

We declare no conflict regarding the publication of the study.

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