

Predictive Modeling of Thermo Physical Properties in Deep Eutectic Solvent Systems Using Jouyban–Acree and Mcallister Correlations

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ABSTRACT

The present study investigates the excess viscosity behavior of binary mixtures of 2-propenol and benzyl alcohol over the entire composition range at temperatures 298.15, 308.15, and 318.15 K. Experimental viscosity values were measured and correlated using the Redlich–Kister polynomial equation to estimate interaction parameters and to assess molecular interactions in the system. The theoretical viscosities were computed using several predictive models, including the Eyring, Kendall–Monroe, Frenkel, Dey (modified), Hind, and Wijk mixing laws. Correlation coefficients and standard deviations were further examined using the Jouyban–Acree and McAllister four-body models. The comparison between experimental and calculated excess viscosity values demonstrates good agreement, confirming the validity of the selected models. The observed negative excess viscosity throughout the composition range indicates specific dipole–dipole and hydrogen bonding interactions between the unlike molecules. This study provides a comprehensive insight into the molecular association and non-ideal behavior of alcohol-based binary mixtures, relevant for industrial solvent design and thermo physical modeling.

Keywords: Excess viscosity, Hind, Jouyban–Acree, McAllister model, Hydrogen bonding

INTRODUCTION

Liquids and liquid mixtures are central to many chemical, pharmaceutical, and industrial processes and understanding their transport properties especially viscosity is essential for design, optimization, and safety considerations. In particular, binary mixtures of alcohols often exhibit non-ideal behavior due to molecular interactions such as hydrogen bonding, dipole–dipole forces, and steric effects, making them a useful tested for studying mixing behavior and correlation modeling. Binary systems involving alcohols are of particular interest because of their strong tendency for hydrogen bonding and association–dissociation equilibria. 2-Propenol (allyl alcohol) and benzyl alcohol differ significantly in molecular size, polarity, and hydrogen-bonding capabilities. The interaction

between these components often leads to complex thermodynamic behavior, reflected in deviations from ideal mixing. Both 2-propenol and benzyl alcohol possess wide industrial and pharmaceutical importance due to their physicochemical versatility. 2-propenol is commonly used as a solvent, antiseptic, and cleaning agent in pharmaceutical formulations and laboratory applications because of its volatility and bactericidal properties [1]. Benzyl alcohol, on the other hand, acts as a preservative, local anesthetic, and solvent in drug preparations, cosmetics, and perfumery products due to its low toxicity and aromatic characteristics [2]. In industrial contexts, 2-propenol serves as a raw material in paints, coatings, and fuel additives, while benzyl alcohol is utilized in resins, dye carriers, and photographic materials [3]. The binary mixture of 2-propenol and benzyl alcohol provides an excellent system for investigating non-

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ideal molecular interactions and thermo physical behavior, which are crucial for designing efficient solvent systems in chemical and pharmaceutical industries [4]. Such studies also aid in understanding molecular association, hydrogen bonding, and deviation from ideality, enabling optimized process control and enhanced formulation design. In continuation of our previous investigations [5-10] on binary liquid mixtures, the present study aims to analyze the thermo physical and viscous behavior of the 2-Propenol + Benzyl alcohol system at three different temperatures 298.15 K, 308.15 K, and 318.15 K and range of mole fractions. The experimental data were analyzed to evaluate the excess viscosity (η^E) by six mixing predictive models, such as, Eyring [11-12], Kendall–Monroe [13], Frenkel [14], Dey [15] (Modified), Hind [16], and Wijk[17]. Excess viscosity (η^E) and excess molar volume (V^E), are very sensitive parameters for understanding molecular interactions and deviations from ideal behavior. The Redlich–Kister [18] polynomial was employed to correlate the excess

properties with composition and to obtain polynomial coefficients that describe the strength and type of interactions between unlike molecules. Additionally, the McAllister [19] four-body and Jouyban–Acree [20-21] correlation models were applied to theoretically interpret the viscosity–composition relationship and assess their predictive accuracy through comparison with literature [22] values. The main objectives of the present work are to (i) analyze molecular interactions using excess viscosity and excess molar volume, (ii) correlate the data using the Redlich–Kister polynomial and suitable theoretical models, (iii) examine the temperature dependence of these interactions, and (iv) Identify the most accurate correlation model for predicting viscosity in hydrogen-bonded binary systems. This study provides valuable information regarding the molecular association and thermodynamic behavior of alcohol mixtures, contributing to their efficient use in industrial and pharmaceutical formulations.

Theoretical Modeling

$$\text{Eyring relation: } \ln \eta = x_1^2 + x_2^2 + 2x_1x_2 \ln \sum_{i=1}^2 (\eta_i/2) \quad (1)$$

$$\text{Kendall-Munroe: } \ln \eta = x_1 \ln \eta_1 + x_2 \ln \eta_2 \quad (2)$$

$$\text{Hind et al: } \eta = x_1^2 \eta_1 + x_2^2 \eta_2 + 2x_1x_2 H_{12} \quad (3)$$

$$\text{Frenkel relation: } \ln \eta = x_1^2 \ln \eta_1 + x_2^2 \ln \eta_2 + 2x_1x_2 \ln (\eta_1 + \eta_2)/2 \quad (4)$$

$$\text{Dey et al (modified Frenkel relation): } \eta_{12} = \frac{2(\eta_1 \eta_2)}{|\eta_1 + \eta_2|} \quad (5)$$

$$\text{Wijk (WK) relation: } \ln \eta = x_1^2 \ln \eta_1 + x_2^2 \ln \eta_2 + 2x_1x_2 \ln 2 (\eta_1 \eta_2) / (\eta_1 + \eta_2) \quad (6)$$

Where η_1 and η_2 are viscosity of pure liquids and η is the viscosity of binary liquid mixture.

RESULTS AND DISCUSSION

The Redlich–Kister polynomial equation (7) is widely used to correlate the excess properties of binary liquid mixtures and to elucidate the nature of molecular interactions between the components. The obtained coefficients (A_0 – A_3) for the binary mixture of 2-propenol + benzyl alcohol at three temperatures 298.15, 308.15, and 318.15 K along with the

corresponding standard deviations (δ) calculated by equation (8) are presented in Table 1. A close observation reveals that at 298.15 K, the first coefficient $A_0 = -1.909$ exhibits a relatively large negative value, indicating strong specific interactions such as hydrogen bonding and dipole–dipole forces between the component molecules. The magnitude of A_0 decreases progressively with increasing temperature from -1.909 to -0.736 , suggesting a weakening of molecular association as thermal agitation becomes more significant at higher temperatures.

Table 1. Standard deviation (δ) and Coefficient of Redlich-Kister polynomial

2-Propenol+Benzyl alcohol					
T/K	A_0	A_1	A_2	A_3	δ
298.15	-1.909	0.014	-0.040	-0.337	0.005
308.15	-1.262	-0.011	-0.086	-0.233	0.007
318.15	-0.736	-0.159	-0.115	-0.005	0.006

The subsequent coefficients (A_1 – A_3) show both positive and negative deviations, revealing the non-ideal nature of the mixture and the presence of composition-dependent interactions. The sign changes of these coefficients imply a complex interplay between self-association of alcohol molecules and cross-interactions between 2-propenol and benzyl alcohol. The standard deviation (δ) values remain very low (0.005–0.007) across all temperatures, confirming that the Redlich–Kister polynomial provides an excellent fit to the experimental data and accurately represents the variation of excess viscosity with mole fraction. Deviation in viscosity ($\Delta\eta$) were represented in form of polynomial by redlich kister for correlating measured data as;

$$\Delta\eta = X_1(1 - X_1) \sum_{i=0}^n A_i(1 - X_1)^i \quad (7)$$

Where A_i represent the polynomial coefficient computed by least square multiple regression method and compared in term of standard deviation (δ) by the equation given below;

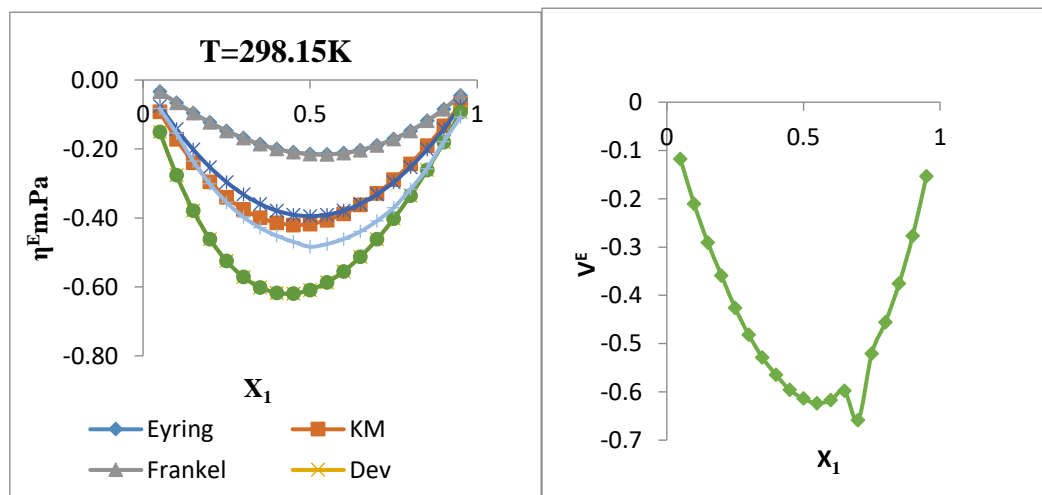
$$\delta = \left[\left(\sum_{i=1}^n \frac{(\eta_{\text{exp}} - \eta_{\text{tho}})^2}{(n - p)} \right)^{1/2} \right] \quad (8)$$

Excess viscosity was computed by the following relation;

$$\eta^E = \eta - (\eta_1 X_1 + \eta_2 X_2) \quad (9)$$

The experimental and theoretically computed values of excess viscosity (η^E) for the binary mixture of 2-

propenol + benzyl alcohol at temperatures 298.15, 308.15, and 318.15 K are presented in table2. The theoretical estimations were made using six predictive mixing models such as Eyring, Kendall–Monroe (KM), Frenkel (Fre), Dey (modified) relation, Hind–Ubbelhode (H–U), and Wijk (WK) equations were used to calculate the theoretical values of viscosity over the entire range of mole fractions at different temperature. At all studied temperatures, the experimental η^E values are negative throughout the composition range, reaching a minimum near the equimolar region ($X_1 \approx 0.45$ – 0.55). This negative deviation indicates structure-breaking interactions between unlike molecules, where the dipole–dipole and hydrogen-bonding forces between 2-propenol and benzyl alcohol molecules are weaker than the self-associative interactions present in the pure components. Such behavior is characteristic of systems where dispersion forces dominate over specific interactions, leading to a decrease in the flow resistance of the mixture compared to the ideal behavior. Clear temperature dependence is observed in the data. As the temperature increases from 298.15 K to 318.15 K, the magnitude of negative η^E decreases from -0.48 to -0.20 , signifying a weakening of associative forces with rising temperature. This trend can be attributed to thermal agitation disrupting intermolecular hydrogen bonds, resulting in less ordered molecular structures and a reduction in non-ideality. Among the six theoretical models, the Eyring and KM relations show closer agreement with the experimental values across the composition range, particularly at higher temperatures, suggesting that these models better represent the viscous behavior of the system under study.



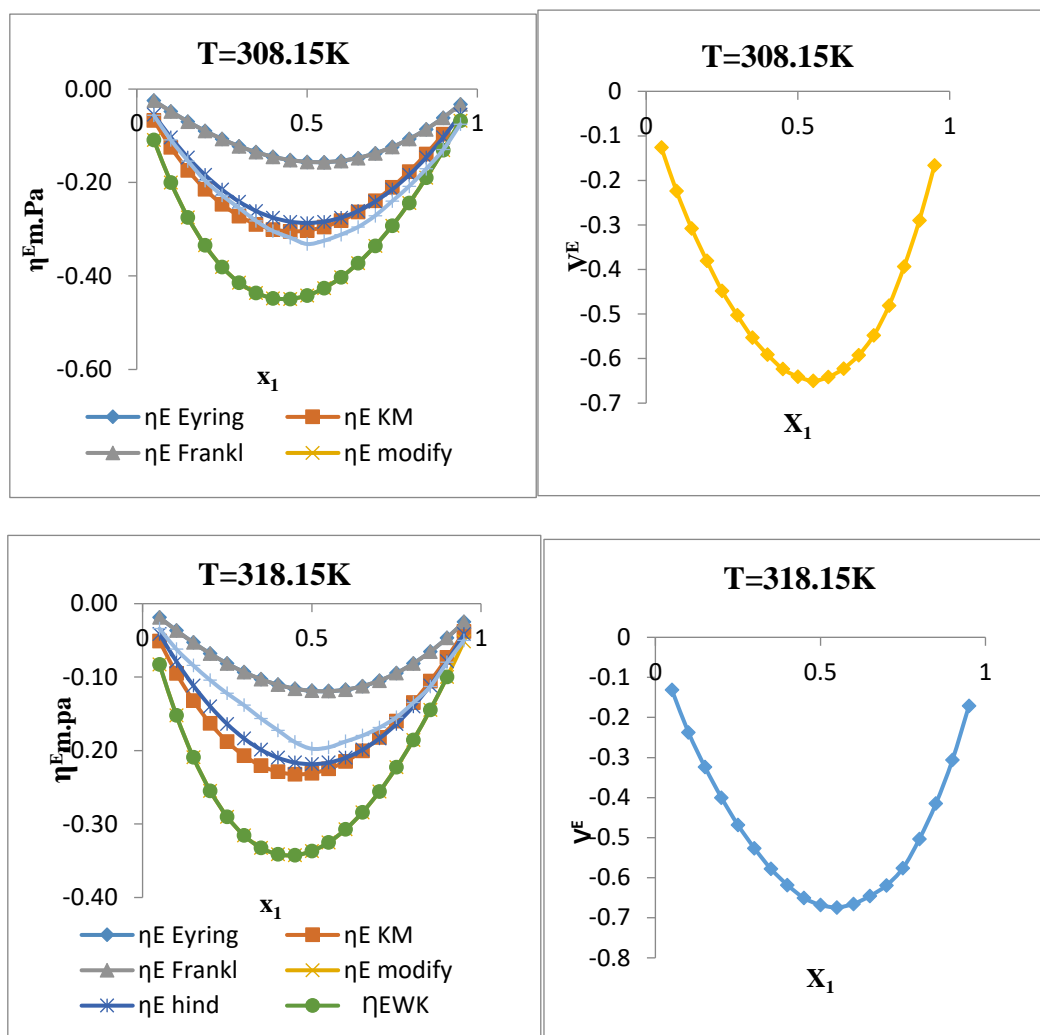


Figure (1) plot of excess viscosity versus X_1
 Figure (2) plot of excess Volume versus X_1

In contrast, the Dey (modified) and Wijk (WK) equations show larger deviations, predicting more negative η^E values than observed experimentally. This indicates that these models may overestimate the effect of molecular asymmetry and interaction strength in this particular binary mixture. The η^E - X_1 plots as shown in figure-1 for the binary mixture of 2-propenol + benzyl alcohol at 298.15 K, 308.15 K, and 318.15 K show negative excess viscosity over the entire composition range, indicating structure-breaking interactions due to the disruption of hydrogen bonding between unlike molecules. The magnitude of η^E decreases with increasing temperature, confirming that intermolecular association weakens at higher thermal energy. The

minimum η^E occurs near the equimolar region ($X_1 \approx 0.5$) at all temperatures, suggesting maximum non-ideality at this composition. The variation of excess molar volume (V^E) with mole fraction (X_1) for the binary mixture at 298.15 K shows a negative deviation throughout the composition range, with a minimum near $X_1 \approx 0.5$. This behavior indicates strong specific interactions between unlike molecules and a resultant volume contraction upon mixing. The negative V^E values as shown in figure -2 suggest the presence of dipole-dipole and hydrogen-bond interactions, leading to a closer molecular packing between 2-propenol and benzyl alcohol. The minimum near equimolar composition implies maximum structural reorganization and strong intermolecular attraction at this ratio. Similar trends were also observed for 308.15 and 318.15K.

Table2. Experimental and theoretical excess viscosity of binary mixture from 298.15-318.15K

X_1	$\eta E_{(EXP)}$	$\eta E_{(Eyring)}$	$\eta E_{(KM)}$	$\eta E_{(Fra)}$	$\eta E_{(Dey)}$	$\eta E_{(H-U)}$	ηE_{WK}
T=298.15K							
0.05	-0.08	-0.03	-0.09	-0.03	-0.15	-0.08	-0.15
0.1	-0.15	-0.07	-0.17	-0.07	-0.28	-0.14	-0.28
0.15	-0.24	-0.10	-0.24	-0.10	-0.38	-0.20	-0.38
0.2	-0.30	-0.12	-0.30	-0.12	-0.46	-0.25	-0.46
0.25	-0.35	-0.15	-0.34	-0.15	-0.53	-0.30	-0.53
0.3	-0.40	-0.17	-0.37	-0.17	-0.57	-0.33	-0.57
0.35	-0.43	-0.19	-0.40	-0.19	-0.60	-0.36	-0.60
0.4	-0.45	-0.20	-0.41	-0.20	-0.62	-0.38	-0.62
0.45	-0.47	-0.21	-0.42	-0.21	-0.62	-0.39	-0.62
0.5	-0.48	-0.21	-0.42	-0.21	-0.61	-0.39	-0.61
0.55	-0.48	-0.22	-0.41	-0.22	-0.59	-0.39	-0.59
0.6	-0.46	-0.21	-0.39	-0.21	-0.56	-0.38	-0.56
0.65	-0.44	-0.20	-0.36	-0.20	-0.51	-0.36	-0.51
0.7	-0.41	-0.19	-0.33	-0.19	-0.46	-0.33	-0.46
0.75	-0.37	-0.17	-0.29	-0.17	-0.40	-0.30	-0.40
0.8	-0.32	-0.15	-0.24	-0.15	-0.34	-0.25	-0.34
0.85	-0.26	-0.12	-0.19	-0.12	-0.26	-0.20	-0.26
0.9	-0.18	-0.08	-0.13	-0.08	-0.18	-0.14	-0.18
0.95	-0.11	-0.04	-0.07	-0.04	-0.09	-0.08	-0.09
T=308.15K							
0.05	-0.06	-0.02	-0.07	-0.02	-0.11	-0.05	-0.11
0.1	-0.11	-0.05	-0.12	-0.05	-0.20	-0.10	-0.20
0.15	-0.15	-0.07	-0.17	-0.07	-0.27	-0.15	-0.27
0.2	-0.20	-0.09	-0.21	-0.09	-0.33	-0.18	-0.33
0.25	-0.23	-0.11	-0.25	-0.11	-0.38	-0.21	-0.38
0.3	-0.25	-0.12	-0.27	-0.12	-0.41	-0.24	-0.41
0.35	-0.28	-0.13	-0.29	-0.13	-0.44	-0.26	-0.44
0.4	-0.30	-0.14	-0.30	-0.14	-0.45	-0.28	-0.45
0.45	-0.32	-0.15	-0.30	-0.15	-0.45	-0.28	-0.45
0.5	-0.33	-0.16	-0.30	-0.16	-0.44	-0.29	-0.44
0.55	-0.32	-0.16	-0.29	-0.16	-0.43	-0.28	-0.43
0.6	-0.31	-0.15	-0.28	-0.15	-0.40	-0.28	-0.40
0.65	-0.30	-0.15	-0.26	-0.15	-0.37	-0.26	-0.37
0.7	-0.27	-0.14	-0.24	-0.14	-0.34	-0.24	-0.34
0.75	-0.24	-0.12	-0.21	-0.12	-0.29	-0.21	-0.29
0.8	-0.21	-0.11	-0.18	-0.11	-0.24	-0.18	-0.24
0.85	-0.17	-0.09	-0.14	-0.09	-0.19	-0.15	-0.19
0.9	-0.13	-0.06	-0.10	-0.06	-0.13	-0.10	-0.13
0.95	-0.08	-0.03	-0.05	-0.03	-0.07	-0.05	-0.07
T=318.15K							
0.05	-0.03	-0.02	-0.05	-0.02	-0.08	-0.04	-0.08
0.1	-0.06	-0.04	-0.09	-0.04	-0.15	-0.08	-0.15
0.15	-0.08	-0.05	-0.13	-0.05	-0.21	-0.11	-0.21
0.2	-0.10	-0.07	-0.16	-0.07	-0.25	-0.14	-0.25
0.25	-0.12	-0.08	-0.19	-0.08	-0.29	-0.16	-0.29
0.3	-0.14	-0.09	-0.21	-0.09	-0.32	-0.18	-0.32
0.35	-0.16	-0.10	-0.22	-0.10	-0.33	-0.20	-0.33
0.4	-0.17	-0.11	-0.23	-0.11	-0.34	-0.21	-0.34
0.45	-0.19	-0.12	-0.23	-0.12	-0.34	-0.22	-0.34
0.5	-0.20	-0.12	-0.23	-0.12	-0.34	-0.22	-0.34
0.55	-0.20	-0.12	-0.22	-0.12	-0.32	-0.22	-0.32

0.6	-0.19	-0.12	-0.21	-0.12	-0.31	-0.21	-0.31
0.65	-0.18	-0.11	-0.20	-0.11	-0.28	-0.20	-0.28
0.7	-0.17	-0.10	-0.18	-0.10	-0.26	-0.18	-0.26
0.75	-0.15	-0.09	-0.16	-0.09	-0.22	-0.16	-0.22
0.8	-0.13	-0.08	-0.13	-0.08	-0.19	-0.14	-0.19
0.85	-0.11	-0.07	-0.11	-0.07	-0.14	-0.11	-0.14
0.9	-0.08	-0.05	-0.07	-0.05	-0.10	-0.08	-0.10
0.95	-0.05	-0.02	-0.04	-0.02	-0.05	-0.04	-0.05

From the Absolute average % deviation results presented in Table3 clearly indicate that, the accuracy of the six viscosity models for the 2-Propenol + Benzyl alcohol) system follows the order: Kendall–Monroe (Kndm) > Hind > Dey ≈ Wijk > Eyring ≈ Frenkel. This sequence indicates that the Kendall–Monroe model provides the best agreement with experimental data, showing the lowest deviation values at all studied temperatures. The Hind model also demonstrates strong predictive reliability, with deviations close to those of the Kndm model. Moderate deviations in the Dey and Wijk models suggest partial consistency with experimental data, while the Eyring and Frenkel models show

comparatively larger deviations, reflecting their limited applicability for this binary system. The calculated parameters of the McAllister four-body and Jouyban–Acree viscosity models for the binary mixture of 2-propenol + benzyl alcohol at 298.15, 308.15, and 318.15 K are presented in Table 4. These empirical models are widely applied to correlate the viscosity–composition data of liquid mixtures and to assess molecular interactions through fitted coefficients. For the McAllister model, the coefficients A_0 , A_1 , and A_2 calculated by equation (10) show a systematic decrease with increasing temperature, reflecting the temperature-dependent weakening of molecular associations between 2-propenol and benzyl alcohol molecules.

Table3. AAPD of six mixing models at different temperatures

Absolute Average % deviation			
T/K	298.15	308.15	318.15
η_{Eyring}	5.719	4.766	2.743
η_{Kndm}	1.566	1.011	1.475
$\eta_{Frenkel}$	5.719	4.766	2.743
$\eta_{Dey (Modified)}$	2.598	3.287	5.089
η_{Hind}	1.881	1.035	1.002
η_{wijk}	2.598	3.287	5.089

The higher coefficient values at 298.15 K ($A_0 = 2.87$, $A_1 = 3.62$, $A_2 = 4.43$) indicate stronger intermolecular hydrogen bonding and associative forces, while their decline at 318.15 K ($A_0 = 1.78$, $A_1 = 2.15$, $A_2 = 2.69$) signifies thermal disruption of these interactions. The low standard deviation ($\delta = 0.01–0.03$) suggests excellent correlation between experimental and calculated viscosities, confirming that the McAllister four-body model effectively describes the viscosity behavior of the system across all studied temperatures.

$$\begin{aligned} \ln \eta &= x_1^4 \ln \eta_1 + 4x_1^3 x_2 \ln A_0 + 6x_1^2 x_2^2 \ln A_1 + 4x_1 x_2^3 \ln A_2 \\ &+ x_2^4 \ln \eta_2 - \ln[(x_1 + x_2 M_2/M_1)] \\ &+ 4x_1^3 x_2 \ln[(3 + M_2/M_1)/4] \\ &+ 6x_1^2 x_2^2 \ln[(1 + M_2/M_1)/2] \\ &+ 4x_1 x_2^3 \ln[(1 + 3 M_2/M_1)/4] \\ &+ x_2^4 \ln[(M_2/M_1)] \end{aligned} \tag{10}$$

Table4. Coefficients of correlation models and standard deviation from 298.15-318.15K

Mc Allister -4- body model				Joubyan Acree model			
T/K	298.15	308.15	318.15	T/K	298.15	308.15	318.15
A ₀	2.87	2.27	1.78	J ₀	-21.19	-8.59	27.70
A ₁	3.62	2.67	2.15	J ₁	-45.51	-40.29	-39.23
A ₂	4.43	3.43	2.69	J ₂	-20.01	-13.12	-20.01
δ	0.01	0.03	0.02	δ	0.01	0.01	0.07

The Jouyban–Acree model coefficients (J_0, J_1, J_2) also exhibit notable temperature dependence. Coefficients were calculated by the equation (11);

$$\ln \eta = X_1 \ln \eta_1 + X_2 \ln \eta_2 + J_0 \left[\frac{X_1 X_2}{T} \right] + J_1 \left[\frac{X_1 X_2 (X_1 - X_2)}{T} \right] + J_2 \left[\frac{X_1 X_2 (X_1 - X_2)^2}{T} \right] \quad (11)$$

At lower temperatures, the large negative J_0 (−21.19) and J_1 (−45.51) values indicate strong non-ideal behavior and significant deviation from additivity, mainly due to hydrogen bonding between unlike molecules. With rising temperature, these values tend toward less negative or even positive magnitudes (for instance, $J_0 = 27.70$ at 318.15 K), reflecting reduced molecular interactions and increased random mixing. The model's standard deviation ($\delta = 0.01$ – 0.07) lies within acceptable limits, further supporting the adequacy of this correlation for the present binary system. On the basis of above discussion, it is observed that both models provide good agreement with experimental viscosity data. However, the McAllister four-body model yields better predictive accuracy with consistently lower δ values, indicating that it more precisely accounts for the size and shape differences of the associating alcohol molecules in the mixture.

CONCLUSION

The experimental and theoretical analysis of the binary liquid mixture of 2-Propenol and Benzyl alcohol at temperatures 298.15 K, 308.15 K, and 318.15 K reveals a consistent trend of negative excess viscosity (η^E) and negative excess molar volume (V^E) across the entire composition range. These negative deviations clearly indicate strong molecular interactions between unlike components, primarily

governed by dipole–dipole and hydrogen-bonding forces. The observed volume contraction (negative V^E) signifies efficient molecular packing, while the decrease in viscosity (negative η^E) suggests partial breakdown of the self-associated structures of the pure components upon mixing. The temperature dependence of both η^E and V^E shows that the magnitude of their negative values decreases with increasing temperature, reflecting the thermal disruption of hydrogen bonding and weakening of specific interactions. Thus, at elevated temperatures, the mixture tends toward ideal behavior due to the reduction in association effects. The viscosity–composition relationship for 2-Propenol + Benzyl alcohol mixtures is most accurately described by the Kendall–Monroe model, followed by the Hind model, particularly at elevated temperatures where molecular interactions weaken and deviations from ideality diminish. The combined analysis of experimental data, excess properties, and theoretical correlations confirms that the binary system exhibits non-ideal mixing behavior dominated by specific associative interactions and molecular rearrangements. The Redlich–Kister polynomial show excellent agreement with experimental data, indicating good fitting. Among the correlation models used, the McAllister four-body model provides more accurate and consistent results than the Jouyban–Acree model, as evidenced by its lower standard deviation values across temperatures. The study enhances the understanding of thermo physical and transport properties of hydrogen-bonded binary systems and provides useful insight for industrial formulations and solvent design, where viscosity control and molecular compatibility are essential.

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CONFLICT OF INTEREST

Authors declare that there is no conflict of interest regarding publication of the article.

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