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Densities and Viscosities of the Binary Mixtures of Phenylmethanol with 2-Butanone

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ABSTRACT: Densities and viscosities of the binary systems of phenylmethanol with 2-butanone were measured for the entire composition range at T = (303.15, 308.15, 313.15, 318.15, and 323.15) K and at the atmospheric pressure. The excess molar volumes were derived from the experimental data and were fitted with the Redlich−Kister equation to obtain their coefficients and standard deviations. The Grunberg−Nissan equation was used to correlate the viscosity data. Furthermore, kinematic viscosities were compared with those predicted by the UNIFAC-VISCO model. The cause of relatively large deviations between the experimental and predicted kinematic viscosities was explored by comparing the combinatorial and residual terms of the UNIFAC-VISCO model.

INTRODUCTION

Fluid property data are essential to explore their usability in the analytical applications that work with mixed solvents, process engineering design applications, and other related areas and have drawn considerable interest in recent years. We are interested in the accumulation of the physical property data of organic liquid mixtures,[1−5] and as a continuation of our effort, we are reporting here the density and viscosity data of the binary mixtures of phenylmethanol with 2-butanone at T = (303.15, 308.15, 313.15, 318.15, and 323.15) K.

Phenylmethanol is a polar solvent with a strong self-associating nature via hydrogen bonding, while 2-butanone is an aprotic and protophilic solvent, and both the liquids are widely used as a solvent in the laboratory and industrial processes. In this study, density and viscosity of the binary liquid mixtures of different compositions covering the entire range were measured. The experimental density data were used to derive the excess molar volume, V_m^E, and the Redlich−Kister type polynomial equation[6] was used to estimate the binary coefficients and the standard deviations. The Grunberg−Nissan interaction parameter which indicates the strength of interactions of the components was calculated using the correlation equation.[7] Kinematic viscosities for the binary system predicted by the UNIFAC-VISCO model[8,9] were compared with the experimental values. The experimental data and the derived quantities were used to understand the likely interactions of the >C═O group of 2-butanone either with the −OH group of phenylmethanol or with the delocalized π-electrons of the benzene ring in phenylmethanol.

Our survey of the literature shows that there have not been any density or viscosity data reported for the binary system of phenylmethanol with 2-butanone.

Table 1. Comparison of Experimental Densities, ρ_{exp}, and Viscosities, η_{exp}, of Pure Solvents with Literature Values at T = (303.15, 308.15, 313.15, 318.15, and 323.15) K

		ρ/g·cm^{-3}			η/mPa·s		
component	T/K	exp.	lit.	ref	exp.	lit.	ref
phenylmethanol	303.15	1.0376	1.0376	16	4.689	4.670	17
	308.15	1.0338	1.0337	17	4.058	4.004	17
						4.093	18
	313.15	1.0301	1.0294	17	3.531	3.530	19
	318.15	1.0263	1.02572	20	3.103	3.120	20
	323.15	1.0225	1.0221	16	2.747		
2-butanone	303.15	0.7940	0.7944	21	0.362	0.3653	22
						0.366	23
	308.15	0.7888	0.7888	24	0.345	0.3441	24
			0.7885	22		0.3440	22
	313.15	0.7835	0.7831	25	0.330		
	318.15	0.7783	0.7785	26	0.316	0.311	26
	323.15	0.7730	0.7733	1	0.303		

EXPERIMENTAL SECTION

Phenylmethanol (Aldrich, 0.99 mass fraction purity) and 2-butanone (Aldrich, > 0.99 mass fraction purity) were used without additional treatment. Density and viscosity data from literature have been used to ascertain the solvent purity (Table 1).

An analytical balance (model: B 204-S, Mettler Toledo, Switzerland) with an uncertainty of ± 0.0001 g was used to prepare the binary mixtures. Mixtures were prepared by mass just before use and were completely miscible over the entire composition range. The uncertainty in the mole fraction was less than $\pm 1 \cdot 10^{-4}$. A 25 mL specific gravity bottle and A-type Ostwald viscometer, previously calibrated with redistilled water, were

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Table 2. Composition, Experimental Densities, ρ_{exp}, and Excess Molar Volumes, V_m^E, for the Binary Mixtures of Phenylmethanol (1) with 2-Butanone (2) at T = (303.15, 308.15, 313.15, 318.15, and 323.15) K

	T/K = 303.15		T/K = 308.15		T/K = 313.15		T/K = 318.15		T/K = 323.15	
	ρ_{exp}	V_m^E	ρ_{exp}	V_m^E	ρ_{exp}	V_m^E	ρ_{exp}	V_m^E	ρ_{exp}	V_m^E
x_1	$g \cdot cm^{-3}$	$cm^3 \cdot mol^{-1}$	$g \cdot cm^{-3}$	$cm^3 \cdot mol^{-1}$	$g \cdot cm^{-3}$	$cm^3 \cdot mol^{-1}$	$g \cdot cm^{-3}$	$cm^3 \cdot mol^{-1}$	$g \cdot cm^{-3}$	$cm^3 \cdot mol^{-1}$
0.0000	0.7940	0.0000	0.7888	0.0000	0.7835	0.0000	0.7783	0.0000	0.7730	0.0000
0.1003	0.8254	−0.4210	0.8204	−0.4389	0.8154	−0.4670	0.8104	−0.4862	0.8053	−0.5053
0.2000	0.8547	−0.6994	0.8499	−0.7308	0.8451	−0.7706	0.8403	−0.8042	0.8355	−0.8484
0.3010	0.8825	−0.8528	0.8778	−0.8833	0.8730	−0.9090	0.8683	−0.9415	0.8636	−0.9835
0.3991	0.9075	−0.8623	0.9030	−0.9000	0.8984	−0.9312	0.8939	−0.9712	0.8893	−1.0087
0.5001	0.9326	−0.8790	0.9282	−0.9118	0.9238	−0.9463	0.9194	−0.9812	0.9150	−1.0232
0.6000	0.9563	−0.8491	0.9520	−0.8758	0.9478	−0.9123	0.9436	−0.9513	0.9393	−0.9856
0.7008	0.9782	−0.6816	0.9741	−0.7103	0.9699	−0.7258	0.9658	−0.7561	0.9617	−0.7908
0.8001	0.9985	−0.4529	0.9946	−0.4827	0.9905	−0.4868	0.9856	−0.4141	0.9827	−0.5522
0.8997	1.0181	−0.2091	1.0143	−0.2292	1.0104	−0.2315	1.0065	−0.2420	1.0027	−0.2645
1.0000	1.0376	0.0000	1.0338	0.0000	1.0301	0.0000	1.0263	0.0000	1.0225	0.0000

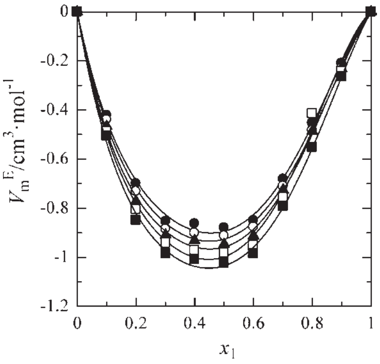

Figure 1. Excess molar volumes for the system phenylmethanol (1) + 2-butanone (2): ●, 303.15 K; ○, 308.15 K; ▲, 313.15 K; □, 318.15 K; ■, 323.15 K.

Table 3. Coefficients, A_i, of the Redlich−Kister Equation (eq 2), Expressing Excess Molar Volumes, V_m^E, and Standard Deviation, σ, for the Binary Mixtures of Phenylmethanol (1) with 2-Butanone (2) at T = (303.15, 308.15, 313.15, 318.15, and 323.15) K

T/K	A_0	A_1	A_2	A_3	σ
303.15	−3.5764	−0.6174	−0.0394	−1.5488	0.0204
308.15	−3.7035	−0.6727	−0.1846	−1.4255	0.0186
313.15	−3.8312	−0.6856	−0.2004	−1.7601	0.0199
318.15	−3.9907	−0.8351	0.1331	−2.0097	0.0385
323.15	−4.1379	−0.7466	−0.4488	−1.7385	0.0222

used for the measurement of density and viscosity correspondingly. The mean uncertainties in densities and viscosities, respectively, were estimated to be 0.0004 $g \cdot cm^{-3}$ and ± 0.5 %. A thermostatic water bath equipped with the Thermo Haake DC10 controller (Thermo Fisher Scientific, MA, USA) was used to maintain the temperatures with an average uncertainty of ± 0.05 K.

All of the measurements were conducted in triplicate and averaged for calculations. LAB Fit[10] and LSM[11] curve-fitting programs were used for least-square regression analyses.

■ RESULTS AND DISCUSSION

Experimental densities, ρ_{exp}, and excess molar volume, V_m^E, for different binary compositions of phenylmethanol with 2-butanone at T = (303.15, 308.15, 313.15, 318.15, and 323.15) K are listed in Table 2. Excess molar volumes were calculated using the following relation:

$$V_m^E = [(x_1M_1 + x_2M_2)/\rho_{exp} - \{(x_1M_1)/\rho_1 + (x_2M_2)/\rho_2\}] \qquad (1)$$

where ρ_{exp} is for the density of the binary mixtures. The mole fraction, molar mass, and density of phenylmethanol are represented with x_1, M_1, and ρ_1, and the corresponding quantities of 2-butanone are denoted with x_2, M_2, and ρ_2, respectively. The excess molar volume, V_m^E, of the (phenylmethanol + 2-butanone) system as a function of the binary compositions at T = (303.15, 308.15, 313.15, 318.15, and 323.15) K is shown in Figure 1. The V_m^E values are negative throughout the composition range, decrease with an increase in phenylmethanol concentration up to the mole fraction, $x_1 \approx 0.5$, and then increase further with the change in x_1. Such behavior may be accounted to the change in orientation of the molecules within the mixtures due to the interaction between the components with the addition of H-bonding containing liquid into the system and partial interstitial accommodation of ketone molecules into the network of alcohol at the phenylmethanol-rich conditions. It is assumed that the latter effect not only compensates the former effect but also predominates largely, resulting in a net negative V_m^E. There are two probable pathways, which may result from the change in orientation of the molecules within the mixtures: (a) interaction between the −OH group of phenylmethanol and the >C═O group of 2-butanone through H-bonding (i.e., −O−H···O═C<) and (b) interaction between the delocalized π-electrons of the benzene ring in phenylmethanol with the >C═O group of 2-butanone.[12] The decreasing trend in the variations of V_m^E values is observed with the rise of temperatures, that is, $\partial V_m^E/\partial T < 0$. A greater population of intermolecular complex within the system, which is attributable to the increasing proportion

Table 4. Composition and Experimental Viscosities, η_{exp}, for the Binary Mixtures of Phenylmethanol (1) with 2-Butanone (2) at T = (303.15, 308.15, 313.15, 318.15, and 323.15) K

	η_{exp}/mPa·s				
x_1	T/K = 303.15	T/K = 308.15	T/K = 313.15	T/K = 318.15	T/K = 323.15
0.0000	0.362	0.345	0.330	0.316	0.303
0.1003	0.446	0.424	0.403	0.385	0.368
0.2000	0.554	0.524	0.497	0.472	0.451
0.3010	0.697	0.655	0.616	0.582	0.551
0.3991	0.876	0.818	0.765	0.718	0.676
0.5001	1.131	1.046	0.970	0.904	0.845
0.6000	1.476	1.351	1.242	1.146	1.063
0.7008	1.947	1.761	1.599	1.461	1.341
0.8001	2.586	2.307	2.063	1.866	1.694
0.8997	3.452	3.033	2.682	2.391	2.144
1.0000	4.689	4.058	3.531	3.103	2.747

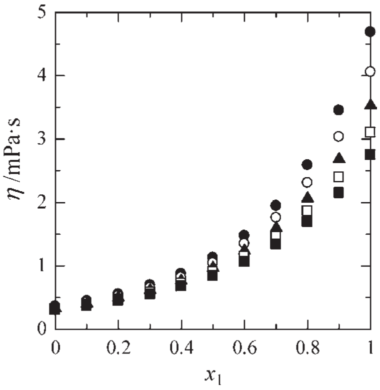

Figure 2. Experimental viscosities for the system phenylmethanol (1) + 2-butanone (2): ●, 303.15 K; ○, 308.15 K; ▲, 313.15 K; □, 318.15 K; ■, 323.15 K.

of smaller multimers as well as the monomers of phenylmethanol and 2-butanone with the increase in temperature, is thus supposed to explain such behavior.

The composition dependence of $V_m^{\,E}$ can be represented by a Redlich–Kister type equation:[6]

$$Y = x_1 x_2 \sum_{i=0}^{n} A_i (1 - 2x_1)^i \tag{2}$$

where Y refers to $V_m^{\,E}$, and the mole fractions of phenylmethanol and 2-butanone are represented with x_1 and x_2, respectively. The coefficients A_i in eq 2 were obtained by fitting the equation with the experimental values (Table 4). The standard deviation values, as listed in Table 3, were calculated from the following equation:

$$\sigma(Y) = [\sum (Y_{\text{expt}} - Y_{\text{calc}})^2/(n - p)]^{(1/2)} \tag{3}$$

where n is the number of experimental points, p is the number of coefficients of eq 2, and Y_{expt} and Y_{calc} are the experimental and calculated values of the properties.

Table 4 lists the experimental viscosities, η_{exp}, for different binary compositions at T = (303.15, 308.15, 313.15, 318.15, and 323.15) K. Figure 2 represents the variation of η_{exp} against the mole fraction of phenylmethanol (x_1) at different temperatures. The viscosity of a mixture strongly depends on the structure of liquids and bond enthalpy and, consequently, on the molecular interactions between the components of the mixture.[13] A gradual increase in the viscosity of the mixtures can be observed with the increasing concentration of phenylmethanol in the binary mixtures with 2-butanone which indicates the formation of more and more flow-resistant species within the system as the proportion of x_1 increases. The magnitude of η_{exp} decreases as the temperature is higher, and such a temperature effect on viscosities is a common phenomenon.[14]

Table 5. Grunberg–Nissan Interaction Parameters (d) for the Binary Mixtures of Phenylmethanol (1) with 2-Butanone (2) at T = (303.15, 308.15, 313.15, 318.15, and 323.15) K

x_1	T/K = 303.15	T/K = 308.15	T/K = 313.15	T/K = 318.15	T/K = 323.15
0.1003	−0.540	−0.474	−0.413	−0.358	−0.317
0.2000	−0.535	−0.470	−0.406	−0.352	−0.279
0.3010	−0.550	−0.483	−0.422	−0.368	−0.314
0.3991	−0.575	−0.507	−0.439	−0.380	−0.324
0.5001	−0.564	−0.497	−0.429	−0.370	−0.312
0.6000	−0.545	−0.476	−0.405	−0.344	−0.285
0.7008	−0.535	−0.466	−0.396	−0.334	−0.277
0.8001	−0.518	−0.453	−0.399	−0.325	−0.269
0.8997	−0.547	−0.487	−0.413	−0.350	−0.298

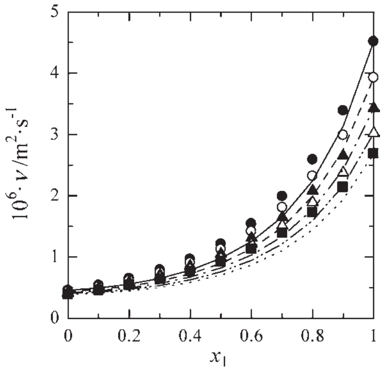

Figure 3. Kinematic viscosities for the system phenylmethanol (1) + 2-butanone (2): ●, ν_{exp} at 303.15 K; ○, ν_{exp} at 308.15 K; ▲, ν_{exp} at 313.15 K; □, ν_{exp} at 318.15 K; ■, ν_{exp} at 323.15 K ; —, ν_{cal} at 303.15 K; — —, ν_{cal} at 308.15 K; — · —, ν_{cal} at 313.15 K; — · · —, ν_{cal} at 318.15 K; · · ·, ν_{cal} at 323.15 K.

Viscosity data were analyzed based on the Grunberg–Nissan treatment,[7] whose parameter gives qualitative information about molecular interactions:

$$\eta_{\text{mix}} = \exp(x_1 \ln \eta_1 + x_2 \ln \eta_2 + x_1 x_2 d) \tag{4}$$

where d is an interaction parameter that is a function of the nature of the components and temperature and has been regarded as a measure of the strength of molecular interactions between the mixture components. The Grunberg–Nissan interaction parameter (d) values are found to be negative and quite large in magnitude for the whole range of composition and decrease with the increase of temperature (Table 5). According to Fort and Moore,[15] such a pattern in d values indicates the tendency of the

Table 6. Comparison of the Experimental and Calculated Values of the Kinematic Viscosity

T/K	AAD/%	$\lvert E_\nu \rvert_{\max}$/%	$\lvert\Delta\nu/\nu\rvert_{\max}\cdot 100$
303.15	12.27	19.3	15.9
308.15	13.01	20.5	14.0
313.15	13.73	21.7	12.2
318.15	14.37	22.7	10.6
323.15	14.95	23.7	9.0

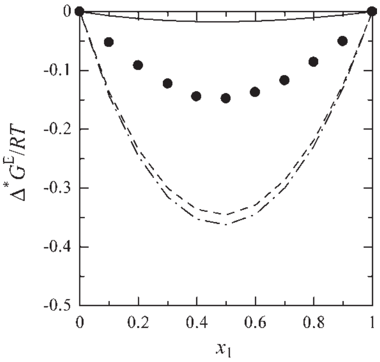

Figure 4. Excess Gibbs energy of activation for the system phenylmethanol (1) + 2-butanone (2) at T = 303.15 K: ●, $\Delta^*G^E{}_{exp}/RT$; —, $\Delta^*G^{EC}{}_{cal}/RT$; — —, $\Delta^*G^{ER}{}_{cal}/RT$; — · —, $\Delta^*G^E{}_{cal}/RT$.

systems to move toward more ideal conditions, which are due to the absence of specific interaction and presence of dominant dispersive force between the components.

In this study, kinematic viscosities were calculated with the experimental density and viscosity data and compared with those predicted by the UNIFAC-VISCO model.[8,9] In the UNIFAC-VISCO model, the relationship between the kinematic viscosity and the excess Gibbs energy of activation, Δ^*G^E, is expressed in the following form:

$$\ln(\nu M) = \sum_{i}^{NC} x_i \ln(\nu_i M_i) + \frac{\Delta^*G^E}{RT} \qquad (5)$$

$$\Delta^*G^E = \Delta^*G^{EC} + \Delta^*G^{ER} \qquad (6)$$

where M is the molecular weight, and Δ^*G^E, Δ^*G^{EC}, and Δ^*G^{ER} are the excess Gibbs energy of activation and its combinatorial and the residual parts, respectively. In the UNIFAC-VISCO model, the combinatorial part expresses the contribution of shape differences among molecules to the Δ^*G^E, while the residual part accounts for the enthalpy effect caused by mixing. In Figure 3, kinematic viscosities predicted by the UNIFAC-VISCO model are compared with the experimental ones. For the prediction, the interaction parameters reported by Gaston-Bonhomme et al.[9] are used. Experimental kinematic viscosities, ν_{exp}, experimental viscosities, η_{exp}, and experimental densities, ρ_{exp}, in Tables 2 and 4 have the following relationship:

$$\nu_{exp} = \frac{\eta_{exp}}{\rho_{exp}} \qquad (7)$$

As can be seen from the figure, the kinematic viscosities obtained by the UNIFAC-VISCO model are smaller than the experimental ones at all temperatures. The relative errors, $E_{\nu,i}$, and the absolute average differences, AAD, were expressed by the following equations and are shown in Table 6:

$$E_{\nu,i} = \frac{\nu_{exp,i} - \nu_{cal,i}}{\nu_{exp,i}} \cdot 100 \qquad (8)$$

$$AAD = \frac{1}{n}\sum_{i}^{N_p} \lvert E_{\nu,i} \rvert \qquad (9)$$

where i denotes the i-th experimental data and n is the number of the experimental data at each temperature. To explore the cause of the deviations, Δ^*G^E, Δ^*G^{EC}, Δ^*G^{ER}, and Δ^*G^E from the experimental data were also compared with each other. The experimental Δ^*G^E was obtained by the following equation:

$$\left.\frac{\Delta^*G^E}{RT}\right|_{exp} = \ln(\nu_{exp}M) - \sum_{i}^{NC} x_i \ln(\nu_i M_i) \qquad (10)$$

where NC is the number of components. Since the results of comparisons were similar at all temperatures, that at 303.15 K is shown in Figure 4 as an example. As can be seen from the figure, the absolute values for $\Delta^*G^E{}_{exp}$ are much smaller than the predicted ones. Since absolute values for the combinatorial part are smaller than those for the residual part, the residual part seems to be responsible for the large deviation. The greater absolute values of the residual term mean the individual contributions of constituent groups in the system are overestimated by the UNIFAC-VISCO model.

■ CONCLUSIONS

Densities and viscosities for the binary mixtures of phenylmethanol with 2-butanone have been determined experimentally as a function of temperature and at the atmospheric pressure. Excess molar volumes have been derived from the experimental density data, and the patterns are negative throughout the composition range. An analysis of viscosity data based on the Grunberg–Nissan treatment shows that no specific interaction exists among the component molecules. Furthermore, experimental kinematic viscosities were obtained by densities and viscosities and compared with those predicted by the UNIFAC-VISCO model. The deviations between experimental and predicted kinematic viscosities are large and the larger the temperature higher. By the comparisons among excess Gibbs energies of activation, the residual term in the UNIFAC-VISCO model seems to be responsible for those large deviations.

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