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Isobaric Vapor–Liquid Equilibria for Binary Systems of Acetone + Isopropenyl Acetate, 2-Butanone + Isopropenyl Acetate, and Isopropenyl Acetate + Acetylacetone at 101.3 kPa

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Isobaric vapor–liquid equilibrium (VLE) data have been determined with a circulation still at 101.3 kPa for binary systems of acetone + isopropenyl acetate, 2-butanone + isopropenyl acetate, and isopropenyl acetate + acetylacetone. No azeotropic behavior was found in any of the systems studied. Thermodynamic consistency has been tested for all VLE data. The experimental data have been correlated satisfactorily by the NRTL, UNIQUAC, and Wilson models. Predictions by using group-contribution methods, such as the original UNIFAC, UNIFAC−Dortmund, and UNIFAC−Lyngby, have been compared with the experimental results.

Introduction

Acetylacetone (2,4-pentanedione) is an important reagent in analytical and coordination chemistry.[1] It could be produced by thermal rearrangement of isopropenyl acetate presently.[2] Isopropenyl acetate is known to be synthesized by the addition of ketene to acetone in the presence of a strong acid catalyst.[3]

Purifying isopropenyl acetate could improve the yield of acetylacetone and the quality of the product when the isopropenyl acetate is subject to thermal rearrangement.[2] In the isomerization process, isopropenyl acetate and acetylacetone are partially pyrolyzed into 2-butanone.[4,5] To obtain high quality acetylacetone, it is important to removal acetone, isopropenyl acetate, and 2-butanone from the crude reaction solution, respectively. Distillation is a feasible process for such separations, and experimental vapor–liquid equilibrium data are indispensable for process design and optimization. In a previous paper, Kozempel et al.[6] reported experimental isothermal VLE data for acetone + isopropenyl acetate at 328.15 K; however, isobaric VLE data are not available in the literature for this system.

In this work, isobaric vapor–liquid equilibrium data for the binary systems of acetone + isopropenyl acetate, 2-butanone + isopropenyl acetate, and isopropenyl acetate + acetylacetone were investigated at 101.3 kPa. The nonideality of the vapor phase has been accounted for in terms of the molar second virial coefficients, estimated by the method of Tsonopoulos.[7] The thermodynamic consistency according to the Van Ness method,[8] modified by Fredenslund et al.,[9] was being used to check for these three systems. The experimental results were correlated by use of the Wilson,[10] NRTL,[11] and UNIQUAC[12] equations. Group-contribution methods such as the original UNIFAC,[9] UNIFAC−Dortmund,[13,14] and UNIFAC−Lyngby[15] were applied to predict VLE data.

Experimental Section

Materials. Acetylacetone was supplied by Huzhou Xin’aote Pharmaceutical & Chemical Co., Ltd. with a minimum mass fraction purity of 99.8 %. Isopropenyl acetate was provided by Shanghai Lingfeng Chemical Reagent Co., Ltd. and had a minimum mass fraction purity of 99.8 %. Acetone was supplied by Shanghai Lingfeng Chemical Reagent Co., Ltd. with a minimum mass fraction purity of 99.5 %. 2-Butanone was provided by Shanghai Feida Chemical Reagent Co., Ltd. with a minimum mass fraction purity of 99.5 %. The purity of the chemicals was checked by a gas chromatograph (GC). All chemicals were used without further purification in this paper. The density and refractive index of pure components were measured and compared with literature data (Table 1). Densities were measured by a DMA-4100 densimeter (Anton Paar GmbH, Germany), with an accuracy of ± 0.0001 $g\cdot cm^{-3}$. Refractive indices were measured by a WZS-I Abbe refractometer (Shanghai Optical Instruments Factory, China), with an accuracy of ± 0.0001.

Table 1. Densities (ρ) and Refractive Indexes (n_D) of Pure Compounds Compared with Literature Data[16]

	$\rho/g\cdot cm^{-3}$ (298.15 K)		n_D (293.15 K)	
compound	exp	lit	exp	lit
acetylacetone	0.9719	0.9721	1.4491	1.4494
isopropenyl acetate	0.9278	0.9281[a]	1.4029	1.4033
acetone	0.7849	0.7845	1.3582	1.3588
2-butanone	0.7994	0.7999	1.3793	1.3788

[a] Taken from ref 3.

Apparatus and Procedure. The apparatus used for the VLE data measurements was a modified Ellis equilibrium still described by Walas.[17] In this still, both liquid and condensed vapor phases (cooled into liquid) are continuously recirculated to provide intimate contact of the phases and ensure that equilibrium can be established rapidly. In each experiment, equilibria conditions were assumed when a constant vapor and liquid temperature was obtained for at least 30 min and the samples of the liquid and condensed vapor were withdrawn for analysis.

Temperature was measured by using a calibrated precision mercury thermometer with an accuracy of ± 0.05 K. Pressure was maintained with the help of a pressure control system as detailed in the literature.[18,19] It consisted of a vacuum pump, two $CaCl_2$ drying bottles, a mercury pressure gauge, a constant

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Table 2. Experimental Vapor–Liquid Equilibrium Data: Temperature (T), Liquid-Phase and Vapor-Phase Mole Fractions (x_1,y_1), and Activity Coefficients (γ_i) for the Acetone (1) + Isopropenyl Acetate (2) System at 101.3 kPa

T/K	x_1	y_1	γ_1	γ_2
330.52	0.953	0.987	0.997	1.119
332.30	0.875	0.962	1.001	1.124
335.88	0.732	0.908	1.013	1.101
338.07	0.669	0.878	1.005	1.080
340.99	0.568	0.825	1.021	1.069
342.84	0.524	0.797	1.015	1.049
345.10	0.462	0.754	1.022	1.035
347.51	0.399	0.702	1.031	1.031
348.36	0.377	0.683	1.039	1.024
350.69	0.328	0.630	1.034	1.024
352.54	0.290	0.589	1.041	1.010
356.15	0.218	0.496	1.062	0.997
358.23	0.178	0.429	1.071	1.003
361.12	0.131	0.341	1.077	0.997
363.04	0.100	0.277	1.091	0.995
366.71	0.046	0.142	1.114	0.994

Table 3. Experimental Vapor–Liquid Equilibrium Data: Temperature (T), Liquid-Phase and Vapor-Phase Mole Fractions (x_1,y_1), and Activity Coefficients (γ_i) for the 2-Butanone (1) + Isopropenyl Acetate (2) System at 101.3 kPa

T/K	x_1	y_1	γ_1	γ_2
354.06	0.892	0.937	0.996	0.958
355.15	0.821	0.890	0.996	0.967
355.87	0.777	0.859	0.994	0.973
356.61	0.725	0.822	0.997	0.976
357.47	0.673	0.781	0.996	0.981
358.83	0.590	0.712	0.996	0.985
359.57	0.547	0.673	0.995	0.987
360.17	0.512	0.640	0.994	0.990
361.36	0.439	0.569	0.995	0.996
362.12	0.396	0.524	0.994	0.998
363.74	0.314	0.431	0.988	0.998
364.39	0.280	0.391	0.988	0.998
365.5	0.223	0.320	0.985	0.999
366.35	0.184	0.270	0.979	0.997
366.94	0.152	0.226	0.977	0.999
368.03	0.104	0.157	0.966	0.997
369.23	0.044	0.068	0.960	0.997

pressure tank (0.75 m^3), a magnetic valve, an electromagnetic delay, and a NaCl solution U tube. The pressure at the top part of the tank was maintained at 101.3 kPa by adjusting the magnetic valve. If the pressure in the tank was higher than 101.3 kPa, the plug in the NaCl solution U-type tube will touch the liquid surface, the magnetic valve will open, and the pressure in the tank will decrease. The air was passed through a filter and a $CaCl_2$ drying bottle before entering the system. In this system, a TJ-800 Mercury U-type pressure gauge was used, whose precision was within ± 0.01 kPa. Atmospheric pressure was measured by a Fortin-type mercury barometer located adjacent to the experimental apparatus with an accuracy of ± 0.04 kPa. The uncertainty of the whole pressure measurement system was estimated to be less than ± 0.10 kPa.

The liquid and vapor samples were analyzed by a GC112A gas chromatograph (GC) equipped with a flame ionization detector (FID) produced by Shanghai Hengping Scientific Instrument Co., Ltd. The GC column was an FFAP capillary column (25 m × Φ 0.32 mm × 0.5 μm). High-purity nitrogen (99.9999 % purity) was used as the carrier gas at a constant flow rate of 5.26 mL·min^{-1}. The injector, detectors, and oven temperature were kept at (423.15, 453.15, and 363.15) K, respectively. The gas chromatograph was calibrated with gas mixtures of known compositions that were prepared gravimetrically by an electronic balance (uncertainty of ± 0.0001 g). The uncertainty of the measured mole fraction was ± 0.001.

Table 4. Experimental Vapor–Liquid Equilibrium Data: Temperature (T), Liquid-Phase and Vapor-Phase Mole Fractions (x_1,y_1), and Activity Coefficients (γ_i) for Isopropenyl Acetate (1) + Acetylacetone (2) System at 101.3 kPa

T/K	x_1	y_1	γ_1	γ_2
371.23	0.940	0.977	1.002	1.330
371.96	0.890	0.958	1.017	1.282
373.27	0.837	0.937	1.017	1.237
374.66	0.779	0.913	1.024	1.204
376.63	0.681	0.870	1.054	1.176
376.73	0.670	0.866	1.064	1.166
379.15	0.582	0.824	1.090	1.118
379.81	0.570	0.816	1.081	1.118
382.03	0.499	0.774	1.103	1.098
383.19	0.453	0.750	1.139	1.078
385.35	0.389	0.708	1.181	1.057
387.53	0.341	0.665	1.195	1.056
389.66	0.295	0.619	1.217	1.056
395.51	0.204	0.510	1.251	1.022
396.19	0.194	0.492	1.248	1.026
399.13	0.157	0.435	1.271	1.008
402.58	0.113	0.350	1.310	1.005
406.09	0.073	0.251	1.334	1.013
410.02	0.039	0.152	1.365	1.002

Table 5. Physical Properties of the Pure Components[a]

	acetone	2-butanone	acetylacetone	isopropenyl acetate
MW/g·mol^{-1}	58.080	72.107	100.117	100.117
T_b/K	329.44	352.79	413.55	370.35[b]
T_c/K	508.20	535.50	602.00	552.79[c]
P_c/bar	47.02	41.54	39.60	36.91[c]
V_c/m·kmol^{-1}	0.209	0.267	0.323	0.324[c]
Z_c	0.233	0.249	0.256	0.260[c]
RD/Å	2.746	3.135	4.017	3.490[d]
DM/D	2.88	2.76	2.81	2.21[e]
r	2.574	3.248	4.019[d]	3.922[d]
q	2.34	2.88	3.52[d]	3.56[d]
ω	0.306	0.324	0.496	0.344[f]
	coefficients for vapor pressure[g]			
A	70.72	114.74	129.02	76.599
B	−5685	−7130	−8624	−7049.1
C	−7.351	−15.184	−17.212	−7.7919
D	6.30E−06	0.01723	0.01697	2.15E−17
E	2	1	1	6

[a] Taken from ref 21, unless noted. [b] Taken from ref 22. [c] Calculated by using the Joback method.[23] [d] Calculated by using the group-contribution method.[23] [e] Taken from ref 24. [f] Calculated by using the definition method.[23] [g] The coefficients of acetylacetone and isopropenyl acetate were taken from ref 25.

Results and Discussion

The isobaric VLE data and the calculated activity coefficients are listed in Tables 2 to 4 for the binary systems of acetone + isopropenyl acetate, 2-butanone + isopropenyl acetate, and isopropenyl acetate + acetylacetone at 101.3 kPa, respectively.

The activity coefficients of the components in the liquid phase were calculated as follows:

$$y_i\widehat{\varphi}_i^{V}P = x_i\gamma_i P_i^{S}\varphi_i^{S}\exp\left\{\frac{V_i^{L}(P-P_i^{S})}{RT}\right\} \quad (1)$$

where P is the total pressure, y_i is the mole fraction of component i in the vapor-phase, x_i is the mole fraction of component i in the liquid-phase, R is the gas constant, T is the temperature, P_i^S is the vapor pressure of pure component i, V_i^L is the liquid molar volume of pure component i, calculated from the modified Rackett equation,[20] $\widehat{\varphi}_i^V$ and $\widehat{\varphi}_i^S$ are the fugacity coefficients of component i in the mixture vapor phase and in the pure state, respectively, γ_i is the activity coefficient of component i.

Table 6. Interaction Parameters A_{ij}, Root Mean Squared Deviations between Calculated and Experimental Vapor-Phase Mole Fractions σy_1, and Temperature σT/K for the Binary Systems Using the Wilson, NRTL, and UNIQUAC Models

model	A_{12}[a] / J·mol^{-1}	A_{21}[a] / J·mol^{-1}	α	σT[b] / K	σy_1[b]
acetone (1) + isopropenyl acetate (2)					
Wilson	−193.38	185.66		0.38	0.011
NRTL	498.51	−342.94	0.3	0.42	0.010
UNIQUAC	247.63	−183.97		0.31	0.008
2-butanone (1) + isopropenyl acetate (2)					
Wilson	181.95	−132.23		0.17	0.006
NRTL	−133.69	147.46	0.3	0.24	0.007
UNIQUAC	−87.35	94.25		0.15	0.005
isopropenyl acetate (1) + acetylacetone (2)					
Wilson	198.74	−47.41		0.27	0.007
NRTL	−147.76	314.51	0.3	0.34	0.008
UNIQUAC	−98.61	151.74		0.28	0.007

[a] The interaction parameters for various models are as follows: Wilson, $A_{ij} = (\lambda_{ij} - \lambda_{ii})/R$; NRTL, $A_{ij} = (g_{ij} - g_{ii})/R$; UNIQUAC, $A_{ij} = (U_{ij} - U_{ii})/R$. [b] $\sigma T = \sqrt{\sum_{i=1}^{n} (T_i^{\text{calc}} - T_i^{\text{expt}})^2/n}$; $\sigma y_1 = \sqrt{\sum_{i=1}^{n} (y_{1,i}^{\text{calc}} - y_{1,i}^{\text{expt}})^2/n}$

The vapor-phase fugacity coefficients were calculated from the virial equation of state truncated at the second virial term, which is given by

$$\ln \varphi_i = \left(2\sum_j y_j B_{ij} - \mathrm{B_m}\right) P/RT \tag{2}$$

with

$$\mathrm{B_m} = \sum_i \sum_j y_i y_j B_{ij} \tag{3}$$

B_{ii} and B_{ij} are the pure and cross second virial coefficients estimated by the method of Tsonopoulos[7] to characterize the vapor-phase deviation from ideal behavior.

The vapor pressure of pure component was calculated from the following equation:

$$\ln P_i^{\mathrm{S}}(\mathrm{Pa}) = A + B/T + C \ln T + DT^E \tag{4}$$

where A, B, C, D, and E are component specific coefficients for vapor pressure. The coefficients for vapor pressure, and the properties of the pure components are given in Table 5. The values of the activity coefficients calculated are listed in Tables 2 to 4.

The experimental results of the binary systems were tested for thermodynamic consistency by means of the point-to-point test of Van Ness,[8] modified by Fredenslund et al.[9] According to this test, the experimental data are consistent if the mean absolute deviation between calculated and measured vapor phase compositions, Δy, is less than 0.01. The results of this test for the binary systems were 0.0061, 0.0027, and 0.0036 for acetone + isopropenyl acetate, 2-butanone + isopropenyl acetate, and isopropenyl acetate + acetylacetone, respectively, indicating that the VLE results for all the three systems are thermodynamically consistent.

The VLE experimental data were correlated with Wilson, NRTL, and UNIQUAC equations, by minimizing the objective function F:

$$F = \sum_{k=1}^{n} \sum_{i=1}^{2} \left[\left(\frac{\gamma_i^{\text{expt}} - \gamma_i^{\text{calc}}}{\gamma_{\mathrm{i}}^{\text{expt}}}\right)^2\right]_k \tag{5}$$

Where, n is the number of experimental VLE data. As recommended by Renon and Prausnitz,[11] the mixture nonrandomness parameter in the NRTL equation was set as 0.3. The interaction parameters for the Wilson, NRTL, and UNIQUAC equations, A_{12} and A_{21}, and the root mean squared deviations (rmsd) between the experimental and calculated values are listed in Table 6.

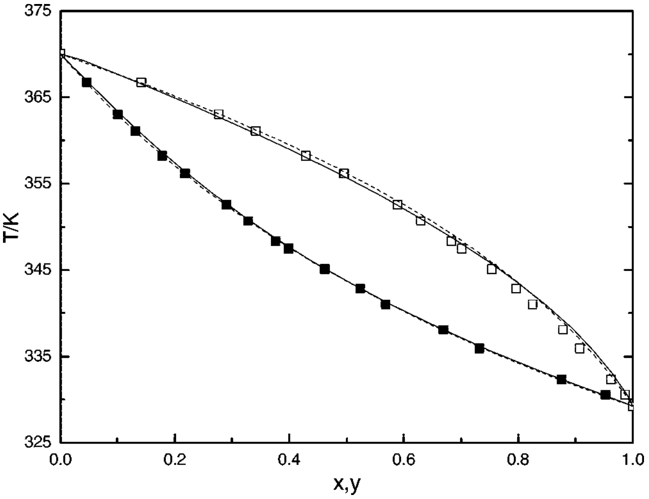

Figure 1. $T-x-y$ diagram for acetone (1) + isopropenyl acetate (2) at 101.3 kPa: ■, experimental liquid-phase mole fractions, x_1; □, experimental vapor-phase mole fractions, y_1; —, UNIQUAC equation; - - -, UNIFAC−Dortmund method.

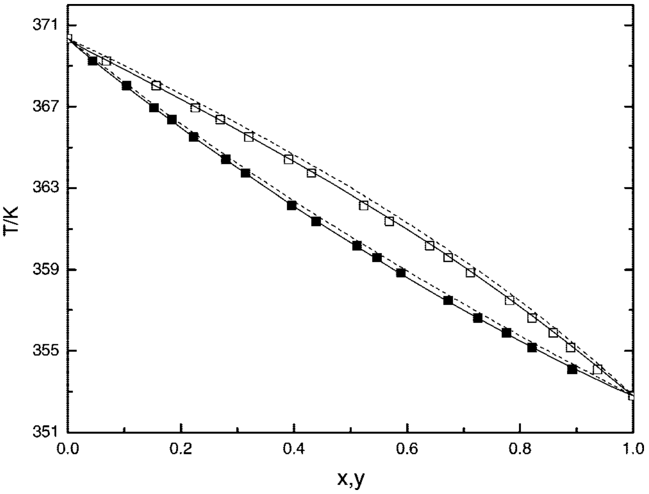

Figure 2. $T-x-y$ diagram for 2-butanone (1) + isopropenyl acetate (2) at 101.3 kPa: ■, experimental liquid-phase mole fractions, x_1; □, experimental vapor-phase mole fractions, y_1; —, UNIQUAC equation; - - -, UNIFAC−Dortmund method.

As seen from Table 6, the results of 2-butanone + isopropenyl acetate and isopropenyl acetate + acetylacetone mixtures can be best represented by the UNIQUAC equation, and the data of acetone + isopropenyl acetate mixture can be best represented by the Wilson equation. However, all systems yield quite small deviations in T and y, which could mean that all the three equations show good performance on correlating the activity coefficients. The experimental VLE data are graphically represented in Figures 1 to 3. The solid lines in these diagrams are obtained from the UNIQUAC equation. It can be seen that azeotropic behaviors are not observed for any of the three systems. Figures 4 to 6 show comparisons between the experimental and calculated activity coefficients for the three systems, respectively.

VLE Predictions

The group-contribution methods play an important role in the prediction of phase equilibria, and equations of state based

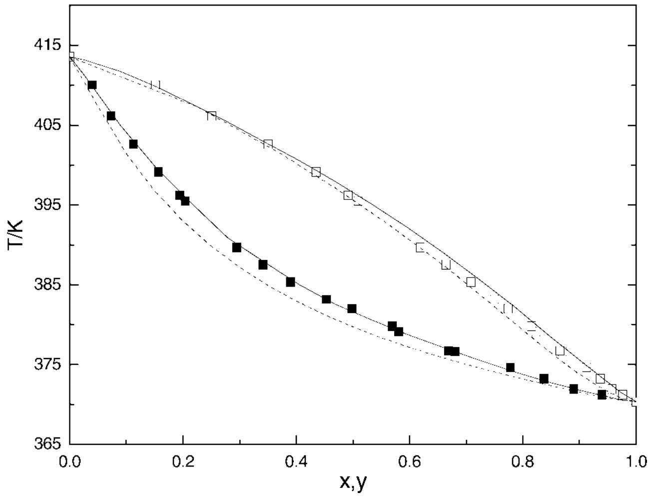

Figure 3. $T-x-y$ diagram for isopropenyl acetate (1) + acetylacetone (2) at 101.3 kPa: ■, experimental liquid-phase mole fractions, x_1; □, experimental vapor-phase mole fractions, y_1; —, UNIQUAC equation; - - -, UNIFAC−Dortmund method.

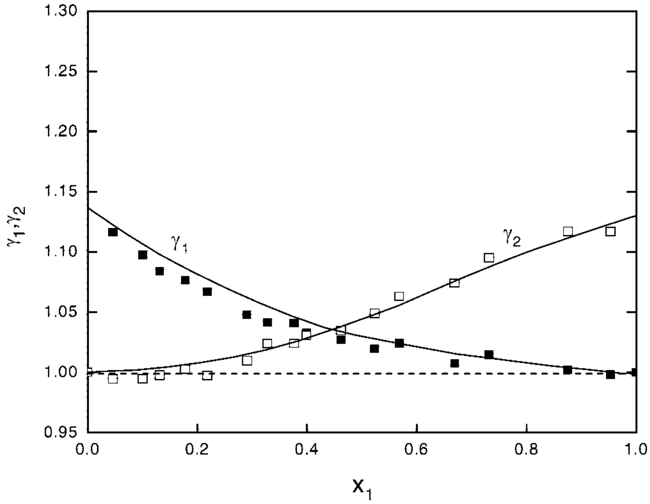

Figure 4. Activity coefficient diagram for acetone (1) + isopropenyl acetate (2) at 101.3 kPa: ■, experimental data, γ_1; □, experimental data, γ_2; —, calculated data using the UNIQUAC equation.

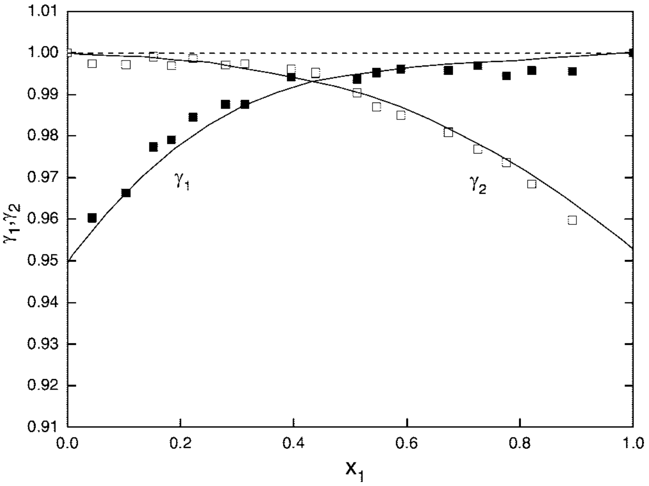

Figure 5. Activity coefficient diagram for 2-butanone (1) + isopropenyl acetate (2) at 101.3 kPa: ■, experimental data, γ_1; □, experimental data, γ_2; —, calculated data using the UNIQUAC equation.

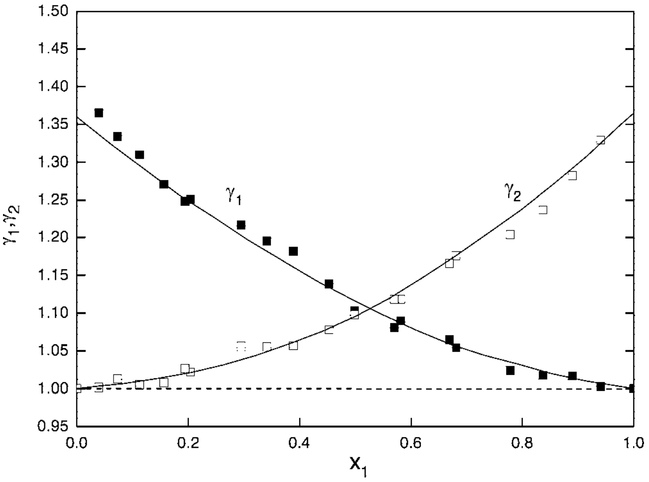

Figure 6. Activity coefficient diagram for isopropenyl acetate (1) + acetylacetone (2) at 101.3 kPa: ■, experimental data, γ_1; □, experimental data, γ_2; —, calculated data using the UNIQUAC equation.

Table 7. Root Mean Squared Deviations between Calculated and Experimental Vapor-Phase Mole Fractions σy_1 and Temperature σT/K for the Binary Systems Using UNIFAC Models

model	σT/K	σy_1
acetone (1) + isopropenyl acetate (2)		
UNIFAC	0.99	0.021
UNIFAC−Lyngby	0.67	0.019
UNIFAC−Dortmund	0.13	0.011
2-butanone (1) + isopropenyl acetate (2)		
UNIFAC	0.72	0.028
UNIFAC−Lyngby	0.17	0.012
UNIFAC−Dortmund	0.22	0.020
isopropenyl acetate (1) + acetylacetone (2)		
UNIFAC	1.84	0.030
UNIFAC−Lyngby	1.19	0.024
UNIFAC−Dortmund	1.80	0.030

on the group contribution concept are available.[26–29] In this work, the original UNIFAC method, with the structural and group-interaction parameters recommended by Gmehling et al.,[30] the UNIFAC−Dortmund method, and the UNIFAC−Lyngby method have been used to predict the vapor–liquid equilibrium of the systems studied. The temperature and vapor-phase composition obtained experimentally were compared with the theoretical predictions using those methods. The root mean squared deviations in temperature and vapor-phase composition are listed in Table 7. The comparisons between the experimental VLE data and the prediction of the UNIFAC−Dortmund model are presented in Figures 1 to 3.

Conclusions

Isobaric vapor–liquid equilibrium (VLE) data were determined at 101.3 kPa for binary systems of acetone + isopropenyl acetate, 2-butanone + isopropenyl acetate, and isopropenyl acetate + acetylacetone. The thermodynamic consistency was tested for all the binary VLE data by Van Ness's method. The experimental data were correlated by using the Wilson, NRTL, and UNIQUAC equations. It was shown that the deviations of all the models were, in general, reasonably small. For the 2-butanone + isopropenyl acetate and isopropenyl acetate + acetylacetone systems, the UNIQUAC equation gives the best results, whereas the Wilson equation gives the smallest deviations for the acetone + isopropenyl acetate system.

The original UNIFAC and Modified-UNIFAC group-contribution methods have been used to predict the vapor–liquid equilibrium. According to the root mean squared deviations shown in Table 7, the UNIFAC−Dortmund method gives satisfactory predictions for the acetone + isopropenyl acetate system, while the UNIFAC−Lyngby method gives better agreement for the 2-butanone + isopropenyl acetate system. However, the predictions for the isopropenyl acetate + acetylacetone system do not show good agreement with the experimental results.

Literature Cited

(1) Kato, M. Activity Coefficients of Pseudo-Ternary Mixtures Containing Acetylacetone, Its Tautomers, and Organic Solvents. *Ind. Eng. Chem. Fundam.* **1980**, *19*, 253–259.
(2) Hagemeyer, H. J.; Hull, D. C. Reactions of Isopropenyl Acetate. *Ind. Eng. Chem.* **1949**, *41*, 2920–2924.
(3) Gwynn, B. H.; Degering, E. F. Condensation Product of Ketene with Ketones. *J. Am. Chem. Soc.* **1942**, *64*, 2216–2218.
(4) Allan, R. J. P.; McGee, J.; Ritchie, P. D. Studies in Pyrolysis. Part XII. Reversible Thermal Rearrangement as a Competitive Route in the Pyrolysis of Enol Carboxylates. *J. Chem. Soc.* **1957**, 4700–4705.
(5) Charles, R. G.; Hickam, W. M.; Hoene, J. V. The Pyrolysis of Acetylacetone. *J. Phys. Chem.* **1959**, *63*, 2084–2085.
(6) Kozempel, M. F.; Kelly, C. M. Vapor-liquid Equilibriums of Acetone, Isopropenyl Acetate, and Acetic Anhydride. *J. Chem. Eng. Data* **1975**, *20*, 367–370.
(7) Tsonopoulos, C. An Empirical Correlation of Second virial Coefficients. *AIChE J.* **1974**, *20*, 263–272.
(8) Van Ness, H. C.; Byer, S. M.; Gibbs, R. E. Vapor-liquid Equilibrium. I: An Appraisal of Data Reduction Methods. *AIChE J.* **1973**, *19*, 238–244.
(9) Fredenslund, A.; Gmehling, J.; Rasmussen, P. *Vapor-Liquid Equilibria Using UNIFAC: A Group-Contribution Method*; Elsevier: Amsterdam, 1977.
(10) Wilson, G. M. Vapor-liquid equilibrium. XI. A New Expression for the Excess Free Energy of Mixing. *J. Am. Chem. Soc.* **1964**, *86*, 127–130.
(11) Renon, H.; Prausnitz, J. M. Local Compositions in Thermodynamic Excess Functions for Liquid Mixtures. *AIChE J.* **1968**, *14*, 135–144.
(12) Abrams, D. S.; Prausnitz, J. M. Statistical Thermodynamics of Liquid Mixtures: a New Expression for the Excess Gibbs Energy of Partially or Completely Miscible Systems. *AIChE J.* **1975**, *21*, 116–128.
(13) Weidlich, U.; Gmehling, J. A Modified UNIFAC Model. 1. Prediction of VLE, h^E, and γ^∞. *Ind. Eng. Chem. Res.* **1987**, *26*, 1372–1381.
(14) Gmehling, J.; Li, J.; Schiller, M. A Modified UNIFAC Model. 2. Present Parameter Matrix and Results for Different Thermodynamic Properties. *Ind. Eng. Chem. Res.* **1993**, *32*, 178–193.
(15) Larsen, B. L.; Rasmussen, P.; Fredenslund, A. A Modified UNIFAC Group-Contribution Model for Prediction of Phase Equilibria and Heats of Mixing. *Ind. Eng. Chem. Res.* **1987**, *26*, 2274–2286.
(16) Lide, D. R. *CRC Handbook of Chemistry and Physics*, 87th ed.; Taylor and Francis: Boca Raton, FL, 2007.
(17) Walas, S. M. *Phase Equilibria in Chemical Engineering*; Butterworth-Heinemann: London, 1985.
(18) Chang, W. X.; Wan, H.; Guan, G. F.; Yao, H. Q. Isobaric Vapor-Liquid Equilibria for Water + Acetic Acid + (*N*-Methyl Pyrrolidone or *N*-Methyl Acetamide). *Fluid Phase Equilib.* **2006**, *242*, 204–209.
(19) Zhang, G. X.; Weeks, B. L.; Wei, J. H. Vapor-Liquid Equilibria Data for Methanol + 2-Propanol+ 2-Methyl-2-butanol and Constituent Binary Systems at 101.3 kPa. *J. Chem. Eng. Data* **2007**, *52*, 878–883.
(20) Rackett, H. G. Equation of State for Saturated Liquids. *J. Chem. Eng. Data* **1970**, *15*, 514–517.
(21) Daubert, T. E.; Danner, R. P. *Physical and Thermodynamic Properties of Pure Chemicals (Data Compilation)*; Hemisphere Publishing Corporation: New York, 1989.
(22) Hart, R.; Smets, G. Vitesse de polymérisation et de copolymérisation de l'acétate d'isopropényle. *J. Polym. Sci.* **1950**, *5*, 55–67.
(23) Reid, R. C.; Prausnitz, J. M.; Poiling, B. E. *The Properties of Gases and Liquids,* 4th ed.; McGraw-Hill: New York, 1987.
(24) Krishna, B.; Saksena, A. R.; Singh, B. An Exact Approach for the Evaluation of Dipole Moments of Esters. *Indian J. Chem.* **1973**, *11*, 391–392.
(25) Askonas, C. F.; Daubert, T. E. Vapor Pressure Determination of Eight Oxygenated Compounds. *J. Chem. Eng. Data* **1988**, *33*, 225–229.
(26) Valderrama, J. O.; Alvarez, V. H. A new group contribution method based on equation of state parameters to evaluate the critical properties of simple and complex molecules. *Can. J. Chem. Eng.* **2006**, *84*, 431–446.
(27) Jaubert, J. N.; Mutelet, F. VLE predictions with the Peng-Robinson equation of state and temperature dependent k_{ij} calculated through a group contribution method. *Fluid Phase Equilib.* **2004**, *224*, 285–304.
(28) Jaubert, J. N.; Vitu, S.; Mutelet, F.; Corriou, J. P. Extension of the PPR78 model (Predictive 1978, Peng Robinson EOS with temperature dependent k_{ij} calculated through a group contribution method) to systems containing aromatic compounds. *Fluid Phase Equilib.* **2005**, *237*, 193–211.
(29) Vitu, S.; Jaubert, J. N.; Mutelet, F. Extension of the PPR78 model (Predictive 1978, Peng Robinson EOS with temperature dependent k_{ij} calculated through a group contribution method) to systems containing naphtenic compounds. *Fluid Phase Equilib.* **2006**, *243*, 9–28.
(30) Gmehling, J.; Rasmussen, P.; Fredenslund, A. Vapor-Liquid Equilibria by UNIFAC Group Contribution. Revision and Extension 2. *Ind. Eng. Chem., Process. Des. Dev.* **1982**, *21*, 118–127.

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