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Vapor−Liquid Equilibrium for Binary Systems of 2-Propanol + 1,1-Diethoxyethane at 353 K, Ethyl Ethanoate + 1,1-Diethoxyethane at 348 K, and 2-Propanone + 1,1-Diethoxyethane at 328 K

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The isothermal vapor−liquid equilibrium was measured for 2-propanol + 1,1-diethoxyethane at 353 K, ethyl ethanoate + 1,1-diethoxyethane at 348 K, and 2-propanone + 1,1-diethoxyethane at 328 K with a circulation still. The liquid and condensed vapor phase samples were analyzed by gas chromatography. The experimental data was correlated with the Wilson and NRTL activity coefficient models. Experimental results and the fitted model were also compared with the predictive coefficient model original UNIFAC.

Introduction

Ethanol is produced in vast amounts by fermentation. It is commonly separated from the fermentation broth using distillation to nearly azeotropic ethanol + water mixtures, which are processed by azeotropic distillation or by absorption masses, such as zeolites, to obtain almost pure ethanol. There are small amounts of side products of fermentation left in ethanol and also some components formed as a result of oxidation during processing. One of these trace components is acetaldehyde, which readily reacts with ethanol to form 1,1-diethoxyethane (acetal). Other trace components present in ethanol are 2-propanol, ethyl ethanoate, and 2-propanone. These trace components can cause problems in processes using ethanol and containing recycles. Our aim was to study the behavior of 1,1-diethoxyethane with some other components mentioned above. Thus, in this work isothermal vapor−liquid equilibrium data for binary system of 2-propanol + 1,1-diethoxyethane at 353 K, ethyl ethanoate + 1,1-diethoxyethane at 348 K, and 2-propanone + 1,1-diethoxyethane at 328 K were measured. No other VLE measurements of these binary systems have been published in the open literature.

Experimental Section

Materials. 1,1-Diethoxyethane (99.1% purity by gas chromatography (GC) peak area) was provided by Aldrich, and 2-propanol (99.8%, GC) was provided by Fluka. 2-Propanone (99.5%, GC) and ethyl ethanoate (99.5%, GC) were supplied by Merck. These materials were used without further purification except for drying over molecular sieves (Merck 3A).

Apparatus for VLE Measurements. The VLE runs were conducted with a circulation still of the Yerazunis type[1] built at the glass workshop of the Helsinki University of Technology with minor modifications to the original design.[2] Approximately 80 mL of reagents was needed to run the apparatus. Temperature was measured with a Thermolyzer S2541 (Frontec) temperature meter with a Pt-100 probe calibrated at Inspecta Oy. The Pt-100 probe was located at the bottom of the packed section of the equilibrium chamber. The resolution of the temperature measurement system was 0.005 K, and the calibration uncertainty was ±0.015 K. The uncertainty of the whole temperature measurement system is estimated to be ±0.05 K. Pressure was measured with a Druck pressure transducer (0 to 100 kPa) and a Red Lion panel meter. The inaccuracy of the instruments was reported to be ±0.07 kPa by the manufacturer. The pressure measurement system was calibrated against a DHPPC-2 pressure calibrator. The inaccuracy of the whole pressure measurement system including the calibration uncertainty is expected to be less than ±0.15 kPa. The accuracy of the analysis of the compositions of the phases is estimated to be 0.005 mole fraction. To improve mixing in the sampling chambers and mixing chamber of the condensed vapor phase and liquid phase, the DC electric motors (Graupner speed 400 motor) were equipped with magnetic stir bars, which deliver smooth and consecutive stirring action in the chambers. The experimental setup was described previously.[3]

Analysis of VLE and GC Calibration. The equilibrated liquid phase was cooled and withdrawn from the sample chamber. The equilibrated vapor phase was first condensed and then sampled in the liquid phase from the sample chamber. The liquid and vapor samples were analyzed with an HP 6850A gas chromatograph with an autosampler and flame ionization detector (FID). The GC column used was an HP-1 (methyl siloxane, length 60 m, nominal diameter 250 μm, nominal film thickness 1.0 μm). The following parameters were used: oven temperature, 90 °C; run time, 15 min; inlet split ratio; 50:1; carrier gas, He (1.0 $\mathrm{mL \cdot min^{-1}}$); FID temperature, 250 °C. Toluene was used as a solvent for the samples to reduce the volume of the sample.

Procedure. Pure component 1 was introduced into the circulation still, and its vapor pressure was measured. Then, component 2 was introduced into the equilibrium still. It took approximately 15 to 30 min to achieve a constant boiling temperature when the difference in the boiling points of the pure components was large. The temperature was held constant for approximately 35 min to guarantee the steady-state condition before sampling. Approximately 1 mL of toluene was added to the 2-mL autosampler vials before sampling was carried out. The

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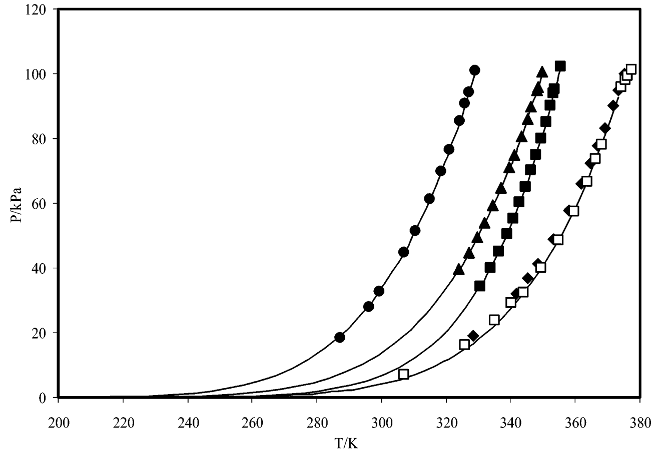

Figure 1. Vapor pressure of pure substances: ◆, 1,1-diethoxyethane, measured, this work; □, 1,1-diethoxyethane, Adkins and Nissen;[11] ■, 2-propanol, measured, this work; ●, 2-propanone, measured, this work; ▲, ethyl ethanoate, measured, this work; –, calculated from Antoine constants in the literature.[12]

Table 1. Experimental Vapor Pressure of 1,1-Diethoxyethane

T/K	P/kPa	T/K	P/kPa
375.27	100.0	357.95	57.9
373.41	94.8	353.38	49.2
371.70	90.1	348.33	41.1
369.09	83.1	345.13	36.7
366.99	77.9	341.62	32.1
364.68	72.3	328.35	19.0
361.96	66.1		

Table 2. Isothermal VLE Data, Liquid-Phase x_1 and Vapor-Phase y_1 Mole Fractions, Pressure P, Temperature T, and Activity Coefficients γ_i for the 2-Propanol (1) + 1,1-Diethoxyethane (2) System at 353 K

x_1	y_1	T/K	P/kPa	$\gamma_{1,\text{calcd}}$	$\gamma_{2,\text{calcd}}$	$\Delta\gamma_1{}^a$	$\Delta\gamma_2{}^b$
0.0000	0.0000	353.38	48.9		1.00		0.00
0.0151	0.0525	353.36	50.9	1.91	1.00	−0.04	0.00
0.0450	0.1479	353.36	54.9	1.94	1.00	−0.14	0.00
0.0852	0.2378	353.37	59.6	1.79	1.01	−0.06	−0.01
0.1354	0.3352	353.34	64.6	1.72	1.01	−0.07	0.00
0.1767	0.3934	353.35	68.3	1.63	1.02	−0.05	0.00
0.2370	0.4652	353.35	72.7	1.53	1.03	−0.04	0.00
0.2845	0.5086	353.33	75.8	1.45	1.05	−0.02	0.00
0.3321	0.5468	353.28	78.3	1.38	1.07	−0.01	−0.01
0.3687	0.5736	353.28	80.1	1.34	1.09	0.00	−0.01
0.4163	0.6068	353.29	82.3	1.28	1.12	0.00	−0.01
0.4580	0.6329	353.32	84.0	1.24	1.14	0.01	−0.01
0.4991	0.6543	353.32	85.5	1.20	1.19	0.01	−0.02
0.5315	0.6758	353.31	86.6	1.18	1.20	0.01	−0.01
0.5715	0.7015	353.33	88.0	1.15	1.23	0.00	0.00
0.6298	0.7330	353.34	89.7	1.11	1.30	0.00	0.00
0.6870	0.7624	353.33	91.2	1.08	1.39	0.01	−0.01
0.7350	0.7913	353.34	92.6	1.06	1.46	0.00	−0.01
0.7790	0.8202	353.34	93.5	1.05	1.52	0.00	0.01
0.8152	0.8427	353.32	93.9	1.03	1.60	0.00	0.01
0.9162	0.9158	353.31	94.2	1.00	1.90	0.00	−0.02
0.9452	0.9446	353.32	94.0	1.00	1.90	0.00	0.07
0.9638	0.9624	353.31	94.0	1.00	1.96	0.00	0.08
0.9826	0.9812	353.31	93.9	1.00	2.03	0.00	0.09
0.9944	0.9940	353.35	93.9	1.00	2.01	0.00	0.16
1.0000	1.0000	353.34	93.9	1.00		0.00	

[a] $\Delta\gamma_1 = \gamma_{1,\text{model}} - \gamma_{1,\text{calcd}}$. [b] $\Delta\gamma_2 = \gamma_{2,\text{model}} - \gamma_{2,\text{calcd}}$.

Table 3. Isothermal VLE Data, Liquid-Phase x_1 and Vapor-Phase y_1 Mole Fractions, Pressure P, Temperature T, and Activity Coefficients γ_i for the 2-Propanone (1) + 1,1-Diethoxyethane (2) System at 328 K

x_1	y_1	T/K	P/kPa	$\gamma_{1,\text{calcd}}$	$\gamma_{2,\text{calcd}}$	$\Delta\gamma_1{}^a$	$\Delta\gamma_2{}^b$
0.0000	0.0000	328.35	19.0		1.00		0.00
0.0746	0.3906	328.24	28.6	1.58	0.99	−0.01	0.01
0.1169	0.4960	328.30	33.6	1.49	1.01	−0.01	0.00
0.2374	0.6686	328.24	44.9	1.32	1.02	0.00	0.01
0.3066	0.7339	328.31	51.1	1.27	1.02	−0.02	0.04
0.3277	0.7467	328.31	52.7	1.25	1.03	−0.02	0.03
0.3450	0.7558	328.31	54.0	1.23	1.04	−0.01	0.03
0.3875	0.7815	328.30	57.1	1.20	1.06	−0.01	0.03
0.4302	0.8033	328.32	60.1	1.17	1.07	−0.01	0.03
0.4690	0.8240	328.31	63.2	1.15	1.08	−0.02	0.05
0.5240	0.8443	328.29	66.3	1.11	1.12	−0.01	0.04
0.5961	0.8688	328.34	71.0	1.07	1.18	0.00	0.02
0.6749	0.8965	328.27	75.7	1.04	1.24	0.00	0.02
0.7272	0.9139	328.31	79.1	1.03	1.28	0.00	0.02
0.7637	0.9261	328.24	81.4	1.02	1.30	0.00	0.02
0.8509	0.9521	328.29	87.3	1.01	1.43	0.00	−0.03
0.9019	0.9680	328.30	90.6	1.00	1.50	0.00	−0.05
0.9478	0.9825	328.31	93.7	1.00	1.59	0.00	−0.09
0.9777	0.9925	328.33	95.8	1.00	1.63	0.00	−0.10
0.9923	0.9973	328.32	96.8	1.00	1.68	0.00	−0.14
1.0000	1.0000	328.32	97.3	1.00		0.00	

[a] $\Delta\gamma_1 = \gamma_{1,\text{model}} - \gamma_{1,\text{calcd}}$. [b] $\Delta\gamma_2 = \gamma_{2,\text{model}} - \gamma_{2,\text{calcd}}$.

Table 4. Isothermal VLE Data, Liquid-Phase x_1 and Vapor-Phase y_1 Mole Fractions, Pressure P, Temperature T, and Activity Coefficients γ_i for the Ethyl Ethanoate (1) + 1,1-Diethoxyethane (2) System at 348 K

x_1	y_1	T/K	P/kPa	$\gamma_{1,\text{calcd}}$	$\gamma_{2,\text{calcd}}$	$\Delta\gamma_1{}^a$	$\Delta\gamma_2{}^b$
0.0000	0.0000	348.33	41.1		1.00		0.00
0.0839	0.1891	348.31	46.2	1.11	1.00	−0.013	0.00
0.1336	0.2793	348.33	49.4	1.09	1.00	−0.009	0.00
0.2080	0.3942	348.33	53.7	1.08	1.00	−0.002	0.01
0.2746	0.4802	348.30	57.6	1.07	1.00	−0.001	0.01
0.3274	0.5386	348.32	60.7	1.05	1.01	0.004	0.00
0.3793	0.5907	348.30	63.6	1.04	1.01	0.007	0.00
0.4245	0.6336	348.31	66.2	1.04	1.02	0.004	0.00
0.5088	0.7020	348.32	71.2	1.03	1.04	0.002	−0.01
0.5695	0.7487	348.30	74.6	1.03	1.04	−0.003	−0.01
0.6382	0.7951	348.31	78.2	1.02	1.06	−0.001	−0.02
0.7023	0.8352	348.32	81.5	1.02	1.08	−0.001	−0.02
0.7570	0.8686	348.33	84.5	1.01	1.09	−0.004	−0.02
0.8055	0.8952	348.31	86.8	1.01	1.12	−0.002	−0.03
0.8554	0.9239	348.30	89.3	1.01	1.12	−0.004	−0.02
0.9059	0.9507	348.31	91.7	1.00	1.14	−0.002	−0.03
0.9625	0.9807	348.34	94.3	1.00	1.15	0.000	−0.01
0.9897	0.9948	348.32	95.5	1.00	1.15	0.000	0.00
1.0000	1.0000	348.31	95.9	1.00		0.000	1.16

[a] $\Delta\gamma_1 = \gamma_{1,\text{model}} - \gamma_{1,\text{calcd}}$. [b] $\Delta\gamma_2 = \gamma_{2,\text{model}} - \gamma_{2,\text{calcd}}$.

samples from the liquid phase and from the vapor condensate were taken with a 1-mL Hamilton sample lock syringe. The syringe was flushed with 0.1 to 0.2 mL of sample before a (0.4 to 0.5)-mL sample was taken and injected into the cooled 2-mL autosampler vial. The temperatures are selected so that the system maximum pressure does not exceed atmospheric pressure and maximum accuracy is maintained.

Results and Discussion

The temperature and vapor pressure of pure components are presented in Figure 1, and 1,1-diethoxyethane vapor pressure data measured in this work are presented in Table 1. A Bancroft point occurred at about 230 K for the 2-propanol (1) + 1,1-diethoxyethane (2) system when each vapor pressure was extrapolated to lower temperature by the Antoine parameters.[12] The measured isothermal equilibrium data and calculated activity coefficients are reported in Tables 2 to 4 and shown in Figures 2 to 5. The three systems exhibit positive deviations from Raoult's law. Azeotropic behavior with a minimum boiling temperature was observed for the 2-propanol (1) + 1,1-diethoxyethane (2) system ($T = 353$ K, $P = 94.2$ kPa, $x_1 = 0.916 \pm 0.005$). Other systems that were measured did not show azeotropic

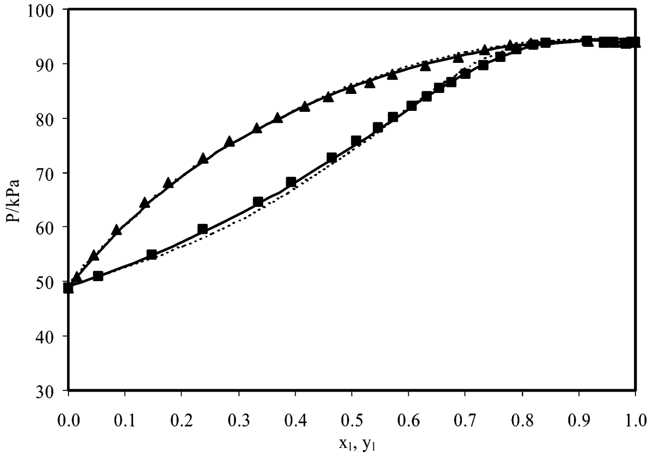

Figure 2. Pressure−composition diagram for the 2-propanol (1) + 1,1-diethoxyethane (2) system at 353 K: ▲, x_1 from the data; ■, y_1 from the data; −, x_1, y_1 from the Wilson model; · · ·, x_1, y_1 from the original UNIFAC model.

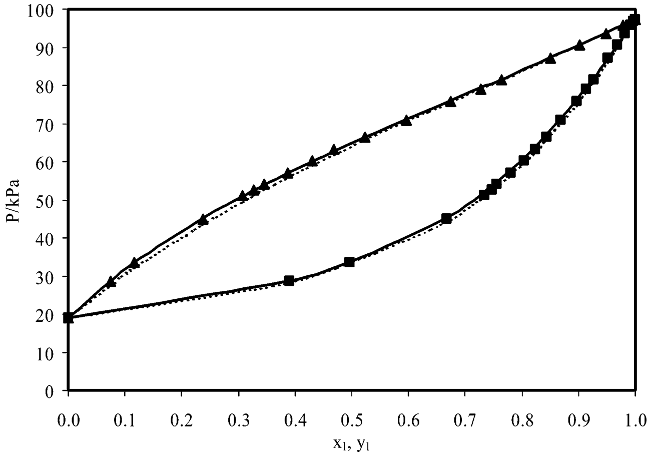

Figure 3. Pressure−composition diagram for the 2-propanone (1) + 1,1-diethoxyethane (2) system at 328 K: ▲, x_1 from the data; ■, y_1 from the data; −, x_1, y_1 from the Wilson model; · · ·, x_1, y_1 from the original UNIFAC model.

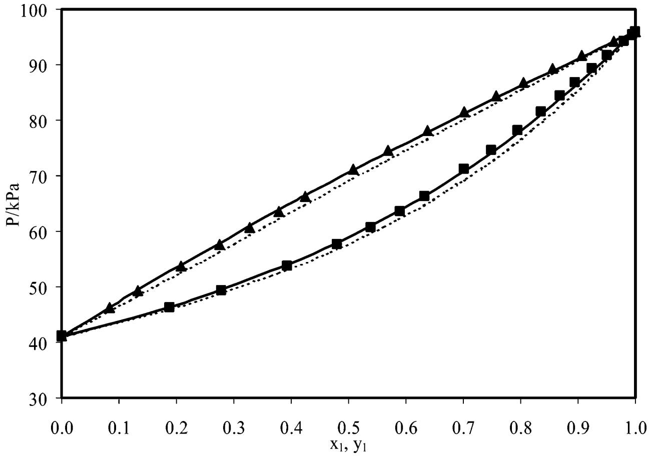

Figure 4. Pressure−composition diagram for the ethyl ethanoate (1) + 1,1-diethoxyethane (2) system at 348 K: ▲, x_1 from the data; ■, y_1 from the data; −, x_1, y_1 from the Wilson model; · · ·, x_1, y_1 from the original UNIFAC model.

behavior. Activity coefficients γ_i were calculated from the equation

$$y_i P \phi_i = \gamma_i x_i P_{\mathrm{vp}i} \phi_i^{\mathrm{s}} \exp \int_{P_{\mathrm{vp}i}}^{P} \frac{v_{\mathrm{L},i}}{RT}\,\mathrm{d}P \tag{1}$$

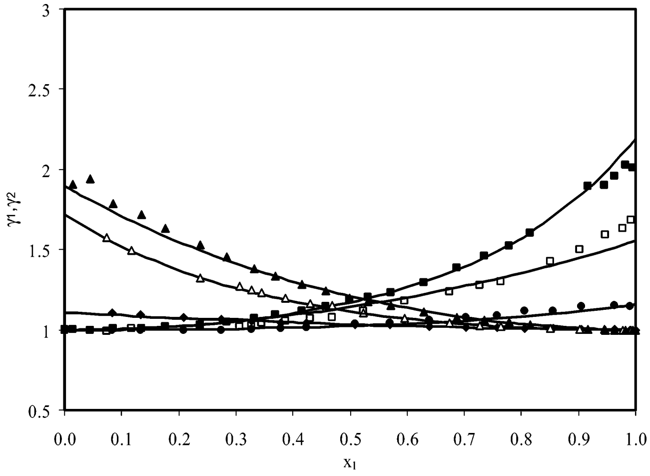

Figure 5. Activity coefficient−composition diagram for the 2-propanol (1) + 1,1-diethoxyethane (2) system at 353 K: ▲, γ_1 from the data; ■, γ_2 from the data; −, γ_1, γ_2 from the Wilson model for the 2-propanone (1) + 1,1-diethoxyethane (2) system at 328 K; △, γ_1 from the data; □, γ_2 from the data; −, γ_1, γ_2 from the Wilson model. Activity coefficient−composition diagram for the ethyl ethanoate (1) + 1,1-diethoxyethane (2) system at 348 K: ◆, γ_1 from the data; ●, γ_2 from the data; −, γ_1, γ_2 from the Wilson model.

Table 5. Physical Property Values Used in Calculations of the Pure Components[a]

component	2-propanol	2-propanone	ethyl ethanoate	1,1-diethoxyethane
T_c/K	508.31[b]	508.20[b]	523.30[b]	541.00[b]
P_c/MPa	4.764[b]	4.7015[b]	3.8800[b]	2.980[b]
ω	0.6689[b]	0.3064[b]	0.3664[b]	0.4321[b]
v_i/cm^3 × mol^{-1}	76.784[b]	73.931[b]	98.594[b]	143.889[b]
A	9.80738[c]	6.70561[c]	8.20025[c]	6.174980[c]
B	3631.488[c]	2405.608[c]	3308.903[c]	2456.685[c]
C	−55.0183[c]	−62.0458[c]	−34.4944[c]	−86.1209[c]
T_{min}/K	330.64[c]	287.34[c]	324.06[c]	328.35[c]
T_{max}/K	355.49[c]	328.98[c]	349.76[c]	375.27[c]

[a] Critical temperature T_c; critical pressure P_c; acentric factor ω; liquid molar volume at 298.15 K (saturation pressure) v_i; pure-component vapor pressure equation parameters (A, B, C) for the Antoine equation, see eq 2 for dimensions and details. (Vapor pressure data measured), recommended temperature range of the vapor pressure correlation T_{min}, T_{max} [b] Daubert and Danner.[13] [c] Measured data.

where y_i is the mole fraction of component i in the vapor phase, P is the total pressure of the system, ϕ_i is the fugacity coefficient of component i in the vapor phase, x_i is the mole fraction of component i in the liquid phase, $P_{\mathrm{vp}i}$ is the vapor pressure of pure component i at the system temperature, ϕ_i^{s} is the pure-component saturated liquid fugacity coefficient at the system temperature, $v_{\mathrm{L},i}$ is the molar volume of pure component i in the liquid phase at the system temperature, T is the temperature in Kelvin, and R is the universal gas constant (8.31441 J·K^{-1}·mol^{-1}).

The Soave−Redlich−Kwong equation of state with quadratic mixing rules in the attractive parameter and linear in covolume was used for the vapor-phase calculation.[4] The liquid phase was modeled with the Wilson equation.[5] The critical temperature, critical pressure, acentric factor, and liquid molar volume for each component used in the calculation are presented in Table 5. The vapor pressure of the pure substances was calculated from an Antoine-type equation

$$P/\mathrm{MPa} = \exp\left(A - \frac{B}{(T/\mathrm{K} + C)}\right) \tag{2}$$

Table 6. Results of the Consistency Tests and Fitted Wilson and NRTL Equation Parameters for 2-Propanol + 1,1-Diethoxyethane (System 1) at 353 K, 2-Propanone + 1,1-Diethoxyethane (System 2) at 328 K, and Ethyl Ethanoate + 1,1-Diethoxyethane (System 3) at 348 K, Averages of Absolute Vapor Molar Fraction Residuals Δy_{ave} and Pressure Residuals ΔP_{ave} for the Wilson and NRTL ($\alpha_{12} = \alpha_{21} = 0.4$) Fits and Original UNIFAC Prediction

	integral test	infinite dilution test	point test $\lvert\Delta y_{aver}\rvert$ $\lvert\Delta P_{aver}\rvert$/kPa			Wilson		NRTL	
binary pair	D%	%	Wilson	NRTL	UNIFAC	$\lambda_{12} - \lambda_{11}$ J·mol^{-1}	$\lambda_{21} - \lambda_{22}$ J·mol^{-1}	$g_{12} - g_{11}$ K	$g_{21} - g_{22}$ K
system 1	2.8	−1.7 ($x_1 = 0$) −5.9 ($x_1 = 1$)	0.003 0.27	0.002 0.29	0.006 0.74	2365.518	−9.984	229.84	53.354
system 2	1.2	−4.3 ($x_1 = 0$) −13.9 ($x_1 = 1$)	0.003 0.15	0.003 0.14	0.007 0.66	3140.713	−1660.002	8.4812	167.07
system 3	7.5	12.0 ($x_1 = 0$) 6.8 ($x_1 = 1$)	0.001 0.23	0.002 0.33	0.007 0.47	399.301	111.522	28.229	14.082

The parameters of this equation were optimized from the data measured with our apparatus. These parameters with the recommended temperature range of the vapor pressure equations are also presented in Table 5.

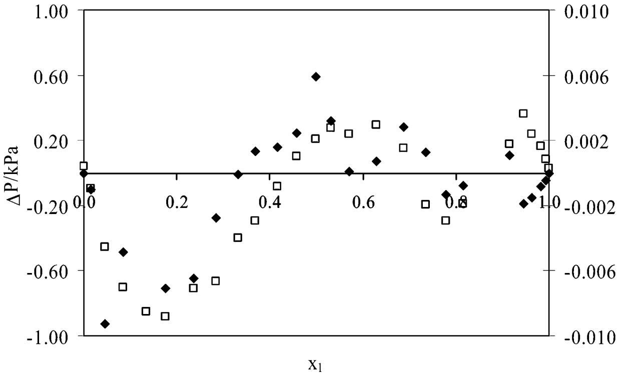

Figure 6. Point test for the 2-propanol (1) + 1,1-diethoxyethane (2) system at 353 K: ◆, Δy; □, ΔP.

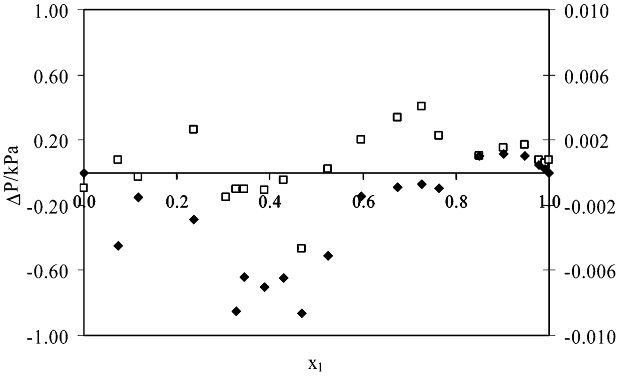

Figure 7. Point test for the 2-propanone (1) + 1,1-diethoxyethane (2) system at 328 K: ◆, Δy; □, ΔP.

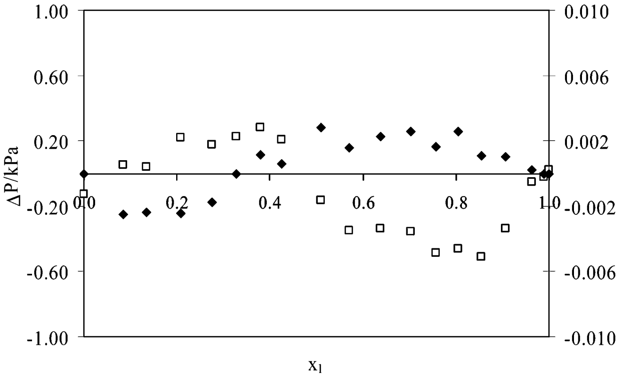

Figure 8. Point test for the ethyl ethanoate (1) + 1,1-diethoxyethane (2) system at 348 K: ◆, Δy; □, ΔP.

The objective function,[6] OF, used for fitting the Wilson equation parameters is given by eq 3, where N is the number of points used in the fit.

$$\mathrm{OF} = \frac{1}{N}\sum_{i=1}^{N}\left(\frac{P_{\mathrm{model}} - P_{\mathrm{meas}}}{P_{\mathrm{meas}}}\right)^2 \tag{3}$$

The parameters of the Wilson[5] and NRTL[7] models are given in Table 6. Comparisons of the experimental points and the fitted Wilson model are given in Figures 2 to 5. Good agreement between the experimental data and model is achieved. Data were also predicted using an original UNIFAC[8] model. The experimental data and UNIFAC predictions match well but not better than the Wilson and NRTL models.

The results of the integral test,[9] the point test[9] (Figures 6, 7, and 8), and the infinite dilution test[10] (Figures 9, 10, and 11) are presented in Table 5. Those data sets satisfy the consistency criterion of all three tests.

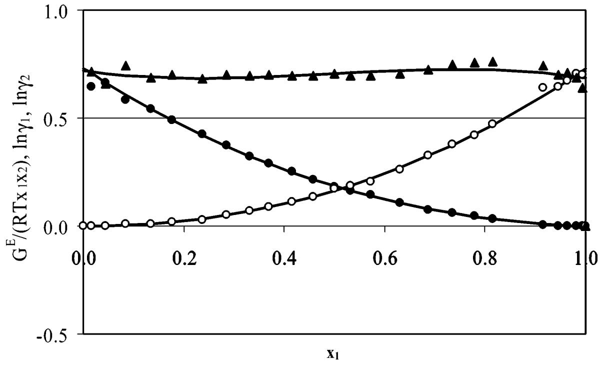

Figure 9. Infinite dilution test for the 2-propanol (1) + 1,1-diethoxyethane (2) system at 353K: ▲, $G^E/(RTx_1x_2)$; ●, ln γ_1; ○, ln γ_2.

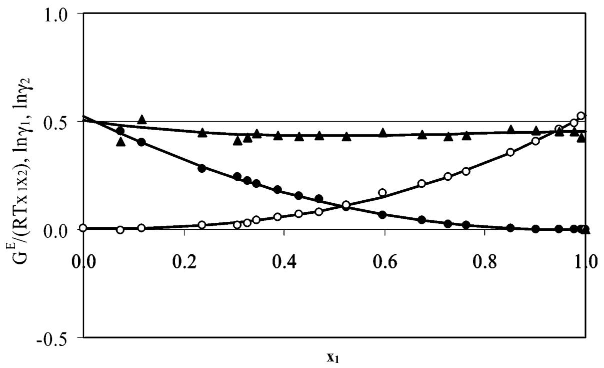

Figure 10. Infinite dilution test for the 2-propanone (1) + 1,1-diethoxyethane diethoxyethane (2) system at 328 K: ▲, $G^E/(RTx_1x_2)$; ●, ln γ_1; ○, ln γ_2.

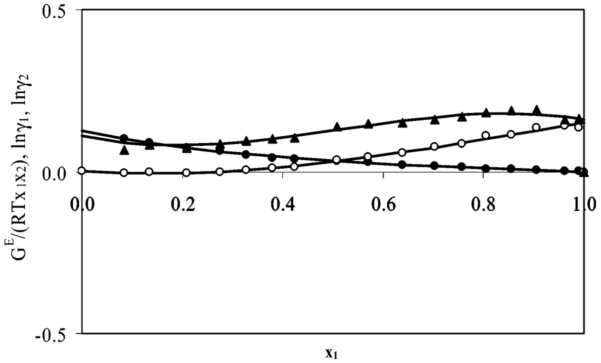

Figure 11. Infinite dilution test for the ethyl ethanoate (1) + 1,1-diethoxyethane (2) system at 348 K: ▲, $G^E/(RTx_1x_2)$; ●, ln γ_1; ○, ln γ_2.

The infinite dilution test is carried out by the following function based on the equation

$$I_1 = 100|I_1^*| < 30 \text{ and } I_2 = 100|I_2^*| < 30 \tag{4}$$

where

$$I_1^* = \left[\frac{\left(\dfrac{G^E}{RTx_1x_2}\right)_{x_1=0} - \ln\left(\dfrac{\gamma_1}{\gamma_2}\right)_{x_1=0}}{\ln\left(\dfrac{\gamma_1}{\gamma_2}\right)_{x_1=0}}\right]$$

and

$$I_2^* = \left[\frac{\left(\dfrac{G^E}{RTx_1x_2}\right)_{x_2=0} - \ln\left(\dfrac{\gamma_2}{\gamma_1}\right)_{x_2=0}}{\ln\left(\dfrac{\gamma_2}{\gamma_1}\right)_{x_2=0}}\right] \tag{5}$$

In the point test, the data set is considered to be consistent if the average deviations between the measured and calculated mole fractions of the vapor phase are smaller than 0.01. This is shown in Table 6 and Figures 6 to 8.

Conclusions

The VLE of isothermal temperature data was measured for 2-propanol + 1,1-diethoxyethane at 353 K, ethyl ethanoate + 1,1-diethoxyethane at 348 K, and 2-propanone + 1,1-diethoxyethane at 328 K with a circulation still. An azeotropic point was observed for the 2-propanol (1) + 1,1-diethoxyethane (2) system. The VLE data of the binary systems was correlated using local composition models. The parameters of the Wilson and NRTL equations were obtained by minimizing the objective function. Both of these equations gave a good fit when compared to the group contribution method UNIFAC.

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