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Articles

Isobaric Vapor−Liquid Equilibria of Diethyl Carbonate with Four Alkanes at 101.3 kPa

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Isobaric vapor−liquid equilibria have been measured for the four binary systems hexane + diethyl carbonate (DEC), DEC + heptane, DEC + octane, and cyclohexane + DEC at 101.3 kPa. The activity coefficients were found to be thermodynamically consistent and were satisfactorily correlated with the Wilson and UNIQUAC equations. They were also compared with results obtained from the application of the ASOG and UNIFAC group contribution methods.

1. Introduction

Currently, papers[1,2] have been published related to dialkyl carbonates because of the possible use of oxygenated volatile organic compounds as fuel additives. This paper is a continuation of our work on the determination of the thermodynamic properties of dialkyl carbonates.[3−8] To ascertain dialkyl carbonate properties with other components, we present experimental isobaric vapor−liquid equilibria (VLE) data for the binary systems hexane + diethyl carbonate (DEC), DEC + heptane, DEC + octane, and cyclohexane + DEC at 101.3 kPa. No VLE data have been previously published at this pressure.

The experimental results have been compared with those correlated through the Wilson[9] and UNIQUAC[10] equations. The interaction parameters of the functional group −OCOO− with CH_3− and $-CH_{2cy}$− in the ASOG[11] model and of the group −OCOO− with CH_3− in the UNIFAC[12] model have been determined previously.[13] ASOG and UNIFAC group contribution methods have been employed to predict the binary systems.

2. Experimental Section

Chemicals. Except for diethyl carbonate, which was supplied by Fluka, the pure components were supplied by Merck. They were degassed by ultrasound and dried over molecular sieves Type 4 Å (supplied by Aldrich) and kept in an inert argon (with a maximum content of water of 2×10^{-6} by mass fraction) atmosphere. Their mass fraction purities were >99.5 mass % for diethyl carbonate, >99.5 mass % for hexane, >99.5 mass % for heptane, >99 mass % for octane, and >99.5 mass % for cyclohexane. Close agreement with recent published density and refractive index values, shown in Table 1, confirms their purity.

Table 1. Comparison of Density ρ and Refractive Index n_D with Literature Data for Pure Components at 298.15 K

	ρ/(g·cm^{-3})		n_D	
component	expt	lit.	expt	lit.
diethyl carbonate	0.9691	0.969 26[a] 0.969 24[b]	1.382 40	1.382 87[a] 1.382 84[c]
hexane	0.6548	0.654 84[a,d]	1.372 26	1.372 26[a,d]
heptane	0.6794	0.679 46[a,d]	1.385 12	1.385 11[a,d]
octane	0.6985	0.698 62[a,d]	1.395 05	1.395 05[a,d]
cyclohexane	0.7738	0.773 89[a]	1.423 63	1.423 54[a]

[a] Riddick et al.[23] [b] Francesconi et al.[24] [c] Cocero et al.[22] [d] Das et al.[25]

Apparatus and Procedure. A glass Fischer LABODEST VLE apparatus model 602/D, manufactured by Fischer Labor und Verfahrenstechnik (Germany), was used in the equilibrium determinations. The equilibrium vessel was a dynamic recirculating still, equipped with a Cottrell circulation pump. This pump ensures that both liquid and vapor phases are in intimate contact during boiling and also in contact with the temperature sensing element. The equilibrium temperature was measured with a digital Yokogawa model 7563 thermometer with an accuracy of ±0.01 K. The apparatus is equipped with a glass temperature probe PT 100 with an accuracy of ±0.01 K. For the pressure measurement, a digital pressure controller Ruska model 7218 with an accuracy of ±0.001 kPa was used. Equilibrium conditions were assumed when constant temperature and pressure were obtained for 30 min or longer. Then, samples of liquid and condensate were taken for analysis. The composition analysis of both samples was determined using an Anton-Paar DSA-48 digital vibrating tube densimeter. Densimetry was used to establish standard curves for each binary system after the density−composition curves for the mixtures had been plotted. The root-mean-square deviation in the mole fraction was usually <0.001.

3. Results and Discussion

Experimental vapor pressures of DEC were measured, as shown Table 2. Antoine coefficients were obtained by applying the equation

$$\ln P_i°/\mathrm{mmHg} = A - \frac{B}{t/°\mathrm{C} - C} \tag{1}$$

Vapor pressures, $P_i°$, were fitted with the Antoine equation, whose parameters A, B, and C of the pure

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Table 2. Experimental Vapor Pressure and Boiling Point for Diethyl Carbonate

t/°C	P_i°/mmHg	t/°C	P_i°/mmHg	t/°C	P_i°/mmHg
79.36	160	113.04	520	131.31	880
82.22	180	114.31	540	132.07	900
85.03	200	115.54	560	132.87	920
87.60	220	116.61	580	133.63	940
89.94	240	117.78	600	134.39	960
92.08	260	118.92	620	135.14	980
94.20	280	119.98	640	135.88	1000
96.20	300	121.07	660	136.57	1020
98.05	320	122.11	680	137.30	1040
99.83	340	123.33	700	138.01	1060
101.63	360	124.17	720	138.68	1080
103.16	380	125.29	740	139.87	1100
104.83	400	126.17	760	140.03	1120
106.26	420	127.05	780	140.60	1140
107.68	440	127.94	800	141.24	1160
109.14	460	128.79	820	141.88	1180
110.45	480	129.63	840	142.50	1200
111.74	500	130.47	860		

Table 3. Antoine Coefficients

component	A	B	C
hexane[a]	6.876 01	1171.17	224.408
heptane[a]	6.896 77	1264.90	216.540
octane[a]	6.918 68	1351.99	209.155
cyclohexane[a]	6.839 17	1200.31	222.500
DEC (this work)	6.751 35	1208.59	186.349

[a] Daubert and Danner.[14]

components are reported in Table 3. The experimental results and published saturated vapor pressures[14] of DEC are represented in Figure 1.

The temperature, t, and the liquid-phase and vapor-phase mole fractions, x_i and y_i, for the binary systems hexane + DEC, DEC + heptane, DEC + octane, and cyclohexane + DEC at 101.3 kPa are presented in Tables 4−7, and they are plotted in Figures 2 and 3. The activity coefficients, γ_i, were calculated from the equation

$$\ln \gamma_i = \ln \frac{y_i P}{x_i P_i^\circ} + \frac{(B_{ii} - V_i^{\mathrm{L}})(P - P_i^\circ)}{RT} + \frac{P}{2RT}\sum_{j=1}^{n}\sum_{k=1}^{n} y_j y_k (2\delta_{ji} - \delta_{jk}) \quad (2)$$

$$\delta_{ji} = 2B_{ji} - B_{jj} - B_{ii} \quad (3)$$

$$\delta_{jk} = 2B_{jk} - B_{jj} - B_{kk} \quad (4)$$

where t and P are the boiling point and the total pressure, respectively, V_i^{L} is the molar liquid volume of component i, B_{ii} and B_{jj} are the second virial coefficients of the pure gases, P_i° is the vapor pressure, and δ_{ij} is related to the cross second virial coefficient. The molar virial coefficients B_{ii}, B_{jj}, B_{ij}, and B_{jk} were estimated by the method of Hayden and O'Connell[15] by using the molecular parameters suggested by Prausnitz et al.[16] The critical properties of all components were taken from the DIPPR database.[14] These data for the binary systems are presented in Tables 4−7.

The results reported in these tables indicate that all the systems exhibit a positive deviation from ideal behavior and a minimum boiling azeotrope for the binary system octane + DEC. Azeotropic compositions have been obtained by determining the x_1 values that make the function $(x_1 - y_1) = f(x_1)$ equal to zero. The corresponding azeotropic temperatures were computed from a cubic polynomial equation $t = f(x_1)$, whose coefficients were obtained by fitting the experimental results around the azeotropic point and using the x_1 values previously determined. The composition and boiling temperature of the azeotrope for the binary mixture octane + DEC are 0.5111 and 391.58 K, respectively. The VLE data, reported in Table 8, were found to be thermodynamically consistent by the point-to-point method of Van Ness et al.[17] and modified by Fredeslund et al.[18]

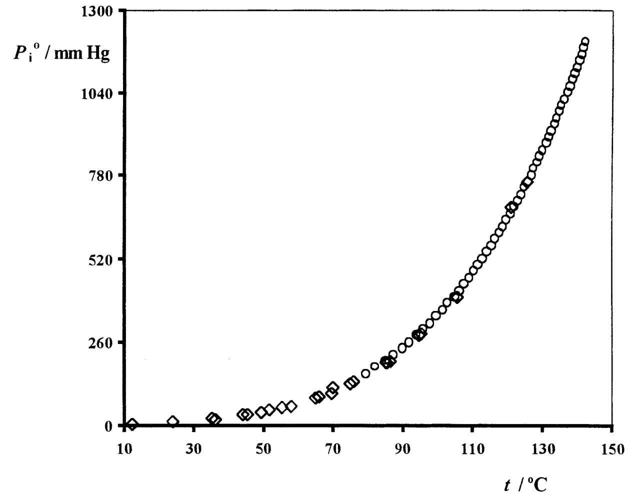

Figure 1. Experimental and comparative vapor pressure values of DEC as a function of the temperature: (O) this work; (Δ) Daubert and Danner.[14]

Table 4. Vapor−Liquid Equilibrium Data for the Hexane (1) + DEC (2) System at 101.3 kPa

T/K	x	y	γ_1	γ_2
397.09	0.0131	0.0764	1.524	0.988
394.90	0.0252	0.1397	1.508	0.990
390.22	0.0519	0.2664	1.536	0.992
386.34	0.0759	0.3587	1.535	0.998
383.29	0.0960	0.4254	1.538	1.003
379.06	0.1268	0.5103	1.534	1.009
373.71	0.1701	0.6007	1.521	1.029
369.83	0.2057	0.6639	1.525	1.030
366.26	0.2439	0.7164	1.514	1.032
363.46	0.2785	0.7517	1.492	1.045
361.47	0.3036	0.7758	1.486	1.050
359.89	0.3295	0.7956	1.464	1.053
357.67	0.3671	0.8193	1.434	1.070
356.00	0.3997	0.8382	1.408	1.075
354.87	0.4252	0.8502	1.384	1.085
353.85	0.4501	0.8605	1.360	1.098
352.67	0.4818	0.8723	1.330	1.116
351.43	0.5246	0.8845	1.282	1.154
350.25	0.5639	0.8964	1.249	1.182
349.42	0.6050	0.9047	1.203	1.241
348.82	0.6317	0.9117	1.181	1.263
347.64	0.6927	0.9242	1.129	1.362
347.32	0.7107	0.9279	1.115	1.393
346.74	0.7429	0.9346	1.092	1.456
345.87	0.7945	0.9459	1.059	1.562
345.44	0.8192	0.9511	1.046	1.633
344.55	0.8663	0.9608	1.025	1.837
343.85	0.9092	0.9687	1.005	2.223
343.29	0.9375	0.9762	0.999	2.510
342.58	0.9701	0.9878	0.997	2.778
342.28	0.9892	0.9954	0.995	2.946

Correlation and Prediction. The parameters of the Wilson and UNIQUAC equations were optimized by minimizing the objective function

$$\mathrm{OF} = \sum_{j=1}^{np}\sum_{i=1}^{nc}\left[\frac{\gamma_{ij}^{\mathrm{exp}} - \gamma_{ij}^{\mathrm{cal}}}{\gamma_{ij}^{\mathrm{exp}}}\right]^2 \quad (5)$$

Table 5. Vapor–Liquid Equilibrium Data for the Heptane (1) + DEC (2) System at 101.3 kPa

T/K	x	y	γ_1	γ_2
398.17	0.0133	0.0614	2.366	0.975
397.09	0.0213	0.0935	2.309	0.978
394.61	0.0438	0.1677	2.133	0.985
392.20	0.0695	0.2326	1.974	0.999
390.80	0.0858	0.2746	1.951	1.001
389.21	0.1064	0.3160	1.881	1.011
387.08	0.1381	0.3711	1.794	1.027
384.73	0.1782	0.4324	1.718	1.043
383.04	0.2175	0.4775	1.621	1.062
381.48	0.2595	0.5204	1.541	1.081
379.95	0.3083	0.5643	1.463	1.103
379.31	0.3390	0.5833	1.399	1.127
378.21	0.3805	0.6173	1.357	1.143
377.72	0.4039	0.6330	1.328	1.158
376.97	0.4448	0.6580	1.279	1.187
375.97	0.4930	0.6933	1.248	1.204
375.40	0.5322	0.7149	1.211	1.236
374.87	0.5660	0.7336	1.185	1.267
374.48	0.5934	0.7502	1.168	1.284
373.79	0.6514	0.7766	1.122	1.371
373.33	0.6882	0.7980	1.105	1.407
373.05	0.7235	0.8172	1.085	1.450
372.65	0.7658	0.8380	1.063	1.538
372.32	0.8175	0.8649	1.037	1.665
372.14	0.8526	0.8872	1.025	1.732
371.96	0.8833	0.9042	1.013	1.870
371.77	0.9262	0.9352	1.004	2.015
371.62	0.9580	0.9605	1.001	2.169
371.49	0.9774	0.9786	1.004	2.197

Table 6. Vapor–Liquid Equilibrium Data for the Octane (1) + DEC (2) System at 101.3 kPa

T/K	x	y	γ_1	γ_2
398.43	0.0250	0.0541	2.432	1.005
397.29	0.0521	0.1061	2.277	1.001
396.11	0.0830	0.1582	2.098	1.005
395.21	0.1116	0.2011	2.030	1.007
394.38	0.1507	0.2443	1.942	1.013
393.74	0.1816	0.2811	1.884	1.017
393.19	0.2192	0.3167	1.802	1.026
392.76	0.2578	0.3485	1.767	1.032
392.46	0.2901	0.3741	1.632	1.050
392.27	0.3146	0.3926	1.506	1.078
392.11	0.3434	0.4103	1.447	1.096
391.96	0.3672	0.4297	1.394	1.113
391.88	0.3852	0.4407	1.330	1.147
391.75	0.4280	0.4704	1.269	1.191
391.66	0.4615	0.4842	1.219	1.235
391.59	0.4977	0.5045	1.181	1.275
391.57	0.5345	0.5225	1.139	1.333
391.62	0.5743	0.5415	1.114	1.383
391.74	0.6169	0.5663	1.101	1.412
391.85	0.6578	0.5968	1.092	1.436
392.21	0.7084	0.6389	1.073	1.494
392.72	0.7673	0.6834	1.061	1.536
393.51	0.8278	0.7408	1.047	1.598
394.04	0.8514	0.7686	1.031	1.689
394.60	0.8773	0.7971	1.019	1.783
395.21	0.9015	0.8293	1.013	1.837
395.90	0.9257	0.8629	1.000	1.998
396.51	0.9450	0.8934	0.996	2.065
397.22	0.9661	0.9273	0.990	2.141
397.92	0.9861	0.9675	0.987	2.211

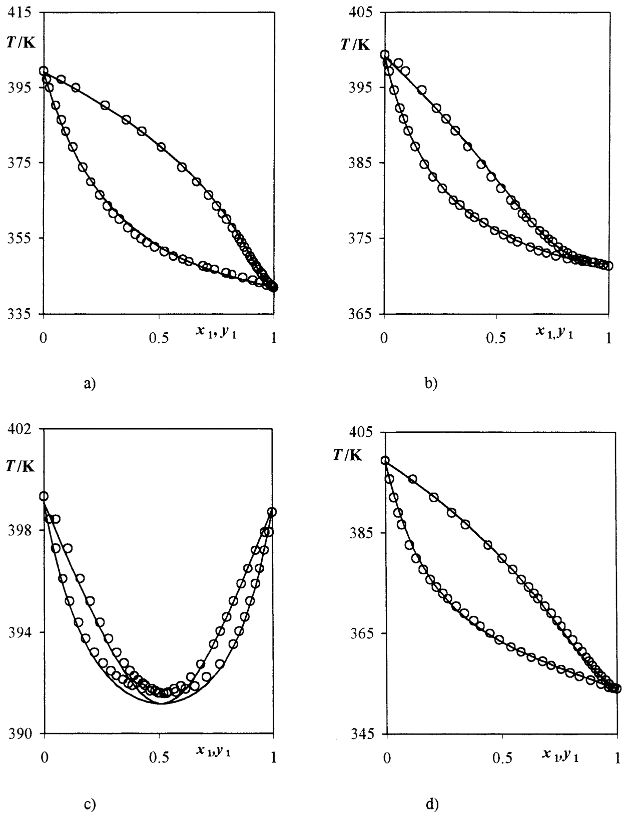

Figure 2. Boiling temperature diagram (t) vs (x_1, y_1) for the binary systems at 101.3 kPa: (○) this work; (—) correlated model; (a) hexane (1) + DEC (2), Wilson; (b) DEC (1) + heptane (2), UNIQUAC; (c) DEC (1) + octane (2), UNIQUAC; (d) cyclohexane (1) + DEC (2), Wilson.

Table 7. Vapor–Liquid Equilibrium Data for the Cyclohexane (1) + DEC (2) System at 101.3 kPa

T/K	x	y	γ_1	γ_2
395.56	0.0181	0.1192	2.347	0.988
391.89	0.0372	0.2108	2.182	1.002
388.83	0.0552	0.2890	2.155	1.005
386.55	0.0704	0.3453	2.125	1.007
382.50	0.1047	0.4438	2.017	1.004
379.88	0.1329	0.5042	1.920	1.003
377.66	0.1619	0.5506	1.815	1.009
375.69	0.1923	0.5883	1.713	1.022
374.20	0.2197	0.6182	1.635	1.030
372.87	0.2475	0.6423	1.559	1.046
371.93	0.2683	0.6603	1.513	1.054
370.40	0.3046	0.6892	1.446	1.068
368.96	0.3410	0.7160	1.393	1.082
367.51	0.3804	0.7417	1.343	1.100
366.47	0.4093	0.7605	1.315	1.109
365.05	0.4504	0.7846	1.280	1.127
363.68	0.4908	0.8082	1.255	1.137
362.27	0.5416	0.8309	1.214	1.171
361.25	0.5850	0.8479	1.179	1.207
360.19	0.6287	0.8663	1.154	1.233
359.37	0.6778	0.8789	1.111	1.326
358.58	0.7149	0.8930	1.094	1.363
357.79	0.7576	0.9068	1.071	1.438
357.09	0.7975	0.9205	1.054	1.508
356.39	0.8374	0.9331	1.037	1.621
355.59	0.8862	0.9511	1.022	1.747
354.80	0.9304	0.9660	1.011	2.046
354.26	0.9655	0.9811	1.005	2.338
354.05	0.9800	0.9885	1.004	2.485

The correlating parameters for the binary systems were calculated using the Thermath program, which was extended by Castier[19] to determine model parameters for the Wilson and UNIQUAC excess Gibbs free energy models from vapor–liquid equilibrium data. This computer program is capable of generating a code that can be readily interfaced with the INTBIS/INTLIB package modified by Schnepper and Stadherr[20] and guaranteeing that the global minimum of the objective function was achieved. In Table 9, the correlating parameters and the root-mean-square deviation with respect to boiling point and vapor-phase mole fraction are reported. Inspection of the results shows that all models are suitable to correlate the binary data.

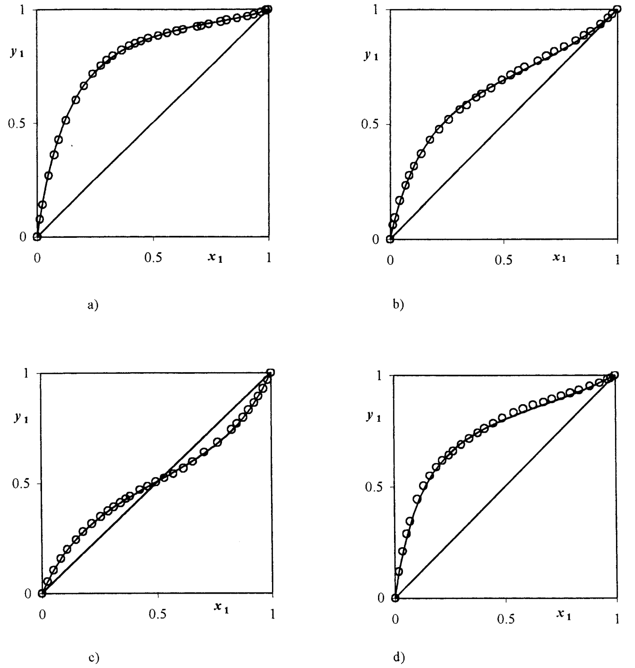

Figure 3. Liquid-phase (x_1) vs vapor-phase (y_1) composition for the binary systems at 101.3 kPa: (○) this work; (—) correlated model; (a) hexane (1) + DEC (2), Wilson; (b) DEC (1) + heptane (2), UNIQUAC; (c) DEC (1) + octane (2), UNIQUAC; (d) cyclohexane (1) + DEC (2), Wilson.

Table 8. Consistency Tests for the Binary Mixtures

system	ΔP/mmHg	Δy
hexane + DEC	0.005	0.003
heptane + DEC	0.021	0.007
octane + DEC	0.002	0.004
cyclohexane + DEC	0.010	0.007

Table 9. Parameters and Deviations between Experimental and Calculated Values for the Binary Systems at 101.3 kPa

model	parameters				ΔT/K	Δy
		Hexane + DEC				
Wilson	$\lambda_{11} - \lambda_{12}$[a]	−275.51	$\lambda_{21} - \lambda_{22}$	1172.93	0.71	0.0067
UNIQUAC	$u_{11} - u_{12}$[b]	627.34	$u_{21} - u_{22}$	−333.19	0.80	0.0042
		Heptane + DEC				
Wilson	$\lambda_{11} - \lambda_{12}$	189.57	$\lambda_{21} - \lambda_{22}$	497.79	0.24	0.0063
UNIQUAC	$u_{11} - u_{12}$	118.60	$u_{21} - u_{22}$	27.55	0.20	0.0069
		Octane + DEC				
Wilson	$\lambda_{11} - \lambda_{12}$	134.83	$\lambda_{21} - \lambda_{22}$	626.36	0.31	0.0041
UNIQUAC	$u_{11} - u_{12}$	229.17	$u_{21} - u_{22}$	−63.28	0.31	0.0038
		Cyclohexane + DEC				
Wilson	$\lambda_{11} - \lambda_{12}$	366.05	$\lambda_{21} - \lambda_{22}$	307.14	0.49	0.0112
UNIQUAC	$u_{11} - u_{12}$	−65.69	$u_{21} - u_{22}$	232.81	0.56	0.0120

[a] $\{\lambda_{ij} - \lambda_{ii}\}/\{(\text{cal mol}^{-1})\}$. [b] $\{u_{ij} - u_{jj}\}/\{(\text{cal mol}^{-1})\}$.

Table 10. ASOG Interaction Parameters

	CH_3		CH_{2cyc}		OCOO	
	m_{kl}	n_{kl}	m_{kl}	n_{kl}	m_{kl}	n_{kl}
CH_3	0	0	0.1530	2.1	**0.0840**	**−197.47**
CH_{2cyc}	−0.1842	0.3	0	0	**−1.5562**	**454.77**
OCOO	**0.3526**	**−174.23**	**1.0414**	**−474.98**	0	0

Figures 2 and 3 show the correlated line corresponding to the best model for each binary system.

Isobaric vapor−liquid data for the binary mixtures were predicted by the ASOG and UNIFAC group contribution methods. The volume and area parameters for the combinatorial term involved in the UNIFAC model, R_k and Q_k, are obtained from Luo et al.[21]

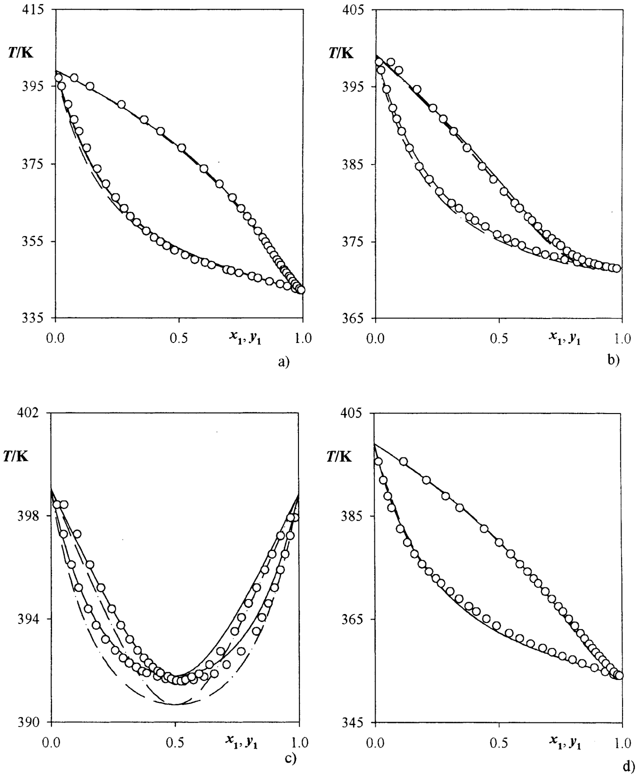

Figure 4. Experimental results (○) of boiling temperature (t) vs (x_1, y_1) and the corresponding fitting curves using (—) the ASOG model and (-.-) the UNIFAC model for the binary systems: (a) hexane (1) + DEC (2); (b) DEC (1) + heptane (2); (c) DEC (1) + octane (2); (d) cyclohexane (1) + DEC (2) at 101.3 kPa.

Table 11. UNIFAC Interaction Parameters

	CH_3	OCOO
CH_3	0	**394.18**
OCOO	**1266.87**	0

Table 12. Root-Mean-Square Deviations of Boiling Points, σ(*t*), and Vapor-Phase Mole Fraction, σ(*y*ᵢ), Resulting from the Prediction of VLE Using ASOG and UNIFAC Models

model	$\sigma(y)$	$\sigma(t)$/K	model	$\sigma(y)$	$\sigma(t)$/K
Hexane + DEC			Octane + DEC		
ASOG	0.0145	0.9510	ASOG	0.0070	0.2913
UNIFAC	0.0265	1.6888	UNIFAC	0.0069	0.7421
Heptane + DEC			Cyclohexane + DEC		
ASOG	0.0079	0.2289	ASOG	0.0098	0.7712
UNIFAC	0.0129	0.6812	UNIFAC	0.0140	0.9430

For both models the residual term is related to the interaction parameters, which are the parameters to be fitted with the help of experimental data. The calculated pairs of interaction parameters for both models are listed in Tables 10 and 11 as well as those of the other group pairs. Figure 4 displays comparisons of the calculated and the experimental VLE results for the binary systems. A very good success was achieved. Table 12 presents the root-mean-square deviation between experimental and predicted values in boiling points and vapor-phase mole fractions for the binary systems by applying both predictive models. The binary systems cyclohexane + DEC at 298.65 K (Cocero et al.[22]) have been used to verify the new parameters for both models. The resulting root-mean-square deviations are shown in Table 13.

Group contribution methods have been employed to predict the VLE of the binary systems under study. The root-mean-square deviations of both the boiling point and

Table 13. Root-Mean-Square Deviations of Pressure, $\sigma(P)$, and Vapor-Phase Mole Fraction, $\sigma(y_i)$, Resulting from the Prediction of VLE Using ASOG and UNIFAC for Published Data

system	ssothermal VLE/model	$\sigma(P)$/mmHg	$\sigma(y)$
cyclohexane + DEC[a]	$t = 298.65$K/ASOG	5.51	0.0476
cyclohexane + DEC[a]	$t = 298.65$K/UNIFAC	6.33	0.0431

[a] Cocero et al.[22]

the composition of the vapor phase indicate that ASOG is more accurate; however, for the published data of Cocero et al.,[22] ASOG and UNIFAC models give similar predictions.

Acknowledgment

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