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Vapor−Liquid Equilibria for the Binary Systems of 1-Butanol with Some Halohydrocarbons at 40.0 and 101.3 kPa

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Isobaric vapor−liquid equilibrium measurements at 40.0 and 101.3 kPa are reported for 1-butanol + chlorocyclohexane, + chlorobenzene, + bromocyclohexane, + bromobenzene. Some of the studied systems show minimum temperature azeotropes. The experimental data were tested for thermodynamic consistency and satisfactorily correlated with the Margules, Van Laar, Wilson, NRTL, and UNIQUAC equations. Predictions with the UNIFAC method and ASOG method were also obtained.

Introduction

In this paper, isobaric vapor−liquid equilibrium measurements for 1-butanol with some halohydrocarbons at 40.0 and 101.3 kPa are reported. This study is a continuation of our research project on vapor−liquid equilibrium for systems containing butanols and halogenated compounds (Lafuente et al., 1995; Rodríguez et al., 1995). The experimental results have been checked for thermodynamic consistency and the activity coefficients have been correlated with the following models: Margules (1895), Van Laar (1910), Wilson (1964), NRTL (Renon and Prausnitz, 1968), and UNIQUAC (Abrams and Prausnitz, 1975).

Predictions of vapor−liquid equilibrium with two group contribution methods, namely, the UNIFAC method (Gmehling et al., 1993) and the ASOG method (Tochigi et al., 1990), have been also done. The results obtained were satisfactory.

Experimental Section

Chemicals. The liquids used were 1-butanol (better than 99.8 mol %) and chlorocyclohexane (better than 99 mol %) obtained from Aldrich together with chlorobenzene and bromobenzene (better than 99.5 mol %) and bromocyclohexane (better than 99 mol %) provided by Fluka. The purity of the materials was checked by GLC, and this analysis showed that the impurities did not exceed 0.3 mol %. The liquids were used without further purification, 1-butanol was dried over activated molecular sieves type 0.3 nm from Merck. The comparison of measured physical properties of the chemicals (densities and normal boiling points) with literature values is shown in Table 1.

Apparatus and Procedure. The still used to measure VLE was an all-glass dynamic recirculating one, equipped with a Cottrell pump. It is a commercial unit (Labodest model) built in Germany by Fischer, capable of handling pressures from 0.25 to 400 kPa, and temperatures up to 523.15 K. This still allows good mixing of the vapor and liquid phases and good separation of the phases once they reach equilibrium, and it prevents entrainment of liquid drops and partial condensation in the vapor phase. The equilibrium temperatures were measured to an accuracy of ±0.01 K by means of a thermometer (model F25) from Automatic Systems Laboratories, and the pressure in the still was measured with a pressure transducer Druck PDCR 110/W (pressure indicator DPI201) with an accuracy of ±0.1 kPa. Compositions of both phases vapor and liquid were determined by measuring their densities at 298.15 K with an Anton Paar DMA-58 vibrating tube densimeter that was previously calibrated at atmospheric pressure with doubly distilled water and dry air. Prior to this, density-calibration curves for these systems were obtained (Artigas et al., 1996), and the estimated uncertainty in

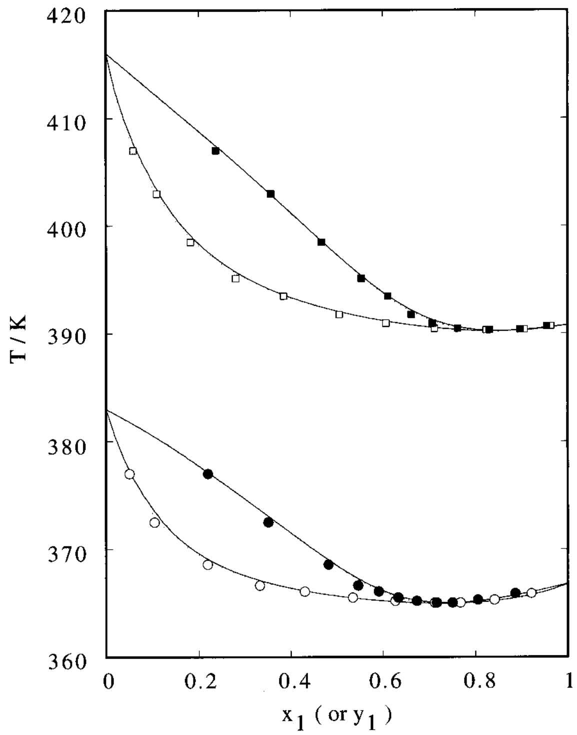

Figure 1. $T-x_1-y_1$ diagram for 1-butanol (1) + chlorocyclohexane (2): (○, ●) experimental data at 40.0 kPa; (□, ■) experimental data at 101.3 kPa; (—) Wilson equation.

Table 1. Physical Properties (Densities at 298.15 K and Normal Boiling Points) of the Pure Compounds

	ρ/kg m^{-3}		T_b/K	
compound	exptl	lit.	exptl	lit.
1-butanol	805.85	806.0[a]	390.84	390.881[b]
chlorocyclohexane	993.23		415.76	
chlorobenzene	1101.00	1101.1[c]	404.86	404.91[d]
bromocyclohexane	1326.16		438.88	
bromobenzene	1488.18	1488.20[e]	429.01	429.058[e]

[a] TRC (1966). [b] TRC (1976). [c] TRC (1990). [d] TRC (1989). [e] Riddick et al. (1986).

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Table 2. Experimental VLE Data at the Indicated Pressure

T/K	x_1	y_1	γ_1	γ_2	T/K	x_1	y_1	γ_1	γ_2
1-Butanol (1) + Chlorocyclohexane (2) at 40.0 kPa									
377.17	0.0514	0.2198	2.839	1.005	365.22	0.6267	0.6735	1.149	1.611
372.53	0.1040	0.3506	2.679	1.034	365.05	0.7118	0.7168	1.084	1.821
368.62	0.2189	0.4815	2.044	1.084	365.05	0.7675	0.7505	1.053	1.988
366.66	0.3322	0.5458	1.654	1.190	365.33	0.8407	0.8054	1.019	2.241
366.09	0.4296	0.5908	1.418	1.281	365.93	0.9208	0.8862	0.999	2.580
365.54	0.5342	0.6334	1.251	1.433					
1-Butanol (1) + Chlorocyclohexane (2) at 101.3 kPa									
407.35	0.0588	0.2368	2.368	1.008	390.95	0.6070	0.7078	1.163	1.468
402.96	0.1097	0.3568	2.190	1.012	390.47	0.7118	0.7622	1.086	1.652
398.49	0.1824	0.4667	1.987	1.035	390.33	0.8239	0.8300	1.027	1.941
395.16	0.2803	0.5535	1.710	1.082	390.40	0.9056	0.8971	1.007	2.187
393.52	0.3843	0.6107	1.453	1.157	390.70	0.9636	0.9546	0.997	2.481
391.77	0.5053	0.6619	1.271	1.317					
1-Butanol (1) + Chlorobenzene (2) at 40.0 kPa									
367.17	0.0902	0.2635	2.882	1.009	362.45	0.6029	0.5754	1.146	1.567
364.55	0.1925	0.3688	2.107	1.065	362.62	0.6558	0.6045	1.099	1.674
363.29	0.3087	0.4366	1.640	1.160	362.95	0.7248	0.6500	1.055	1.832
362.62	0.4317	0.4940	1.365	1.297	363.92	0.8359	0.7460	1.007	2.156
362.51	0.5407	0.5443	1.206	1.451	365.47	0.9331	0.8762	0.993	2.445
1-Butanol (1) + Chlorobenzene (2) at 101.3 kPa									
396.57	0.1065	0.2859	2.220	1.002	388.57	0.7171	0.7070	1.068	1.635
393.94	0.1700	0.3691	1.958	1.027	388.84	0.8030	0.7690	1.027	1.837
391.60	0.2646	0.4505	1.662	1.080	389.16	0.8547	0.8166	1.014	1.959
390.15	0.3687	0.5138	1.429	1.161	389.54	0.8989	0.8619	1.004	2.096
389.03	0.5017	0.5840	1.241	1.300	389.96	0.9379	0.9094	1.001	2.212
388.66	0.6091	0.6409	1.136	1.446	390.39	0.9627	0.9435	0.997	2.268
1-Butanol (1) + Bromocyclohexane (2) at 40.0 kPa									
397.87	0.0252	0.2185	2.769	0.998	369.87	0.5789	0.8127	1.240	1.431
392.10	0.0543	0.3696	2.637	0.995	368.91	0.6808	0.8413	1.135	1.657
386.61	0.0924	0.4878	2.476	1.008	368.02	0.7926	0.8777	1.054	2.032
380.83	0.1554	0.5987	2.229	1.033	367.67	0.8620	0.9031	1.012	2.452
375.90	0.2658	0.6996	1.834	1.057	367.44	0.9095	0.9293	0.996	2.752
373.04	0.3576	0.7418	1.616	1.151	367.23	0.9213	0.9435	1.007	2.549
371.23	0.4623	0.7794	1.411	1.255					
1-Butanol (1) + Bromocyclohexane (2) at 101.3 kPa									
432.87	0.0234	0.1582	1.950	1.002	395.42	0.5679	0.8311	1.256	1.302
428.05	0.0425	0.2595	1.996	1.017	393.92	0.6803	0.8629	1.145	1.498
417.63	0.1109	0.4834	1.895	1.008	392.52	0.8006	0.8928	1.055	1.963
411.52	0.1771	0.5940	1.740	1.016	392.05	0.8545	0.9152	1.029	2.161
405.17	0.2677	0.6938	1.629	1.035	391.65	0.9046	0.9404	1.013	2.346
400.57	0.3594	0.7531	1.522	1.095	391.23	0.9548	0.9681	1.002	2.686
398.15	0.4665	0.7815	1.315	1.253					
1-Butanol (1) + Bromobenzene (2) at 40.0 kPa									
388.38	0.0342	0.2374	3.059	1.023	368.75	0.5741	0.7506	1.209	1.487
384.80	0.0587	0.3654	3.115	0.982	368.16	0.6581	0.7804	1.123	1.667
380.70	0.1005	0.4749	2.747	0.975	367.77	0.7199	0.8036	1.074	1.846
375.10	0.1832	0.5840	2.291	1.031	367.26	0.8129	0.8487	1.026	2.169
372.15	0.2892	0.6521	1.819	1.100	367.14	0.8541	0.8749	1.011	2.310
370.55	0.3939	0.6948	1.516	1.198	367.06	0.9026	0.9082	0.997	2.547
369.49	0.4845	0.7235	1.340	1.326	367.17	0.9497	0.9479	0.984	2.788
1-Butanol (1) + Bromobenzene (2) at 101.3 kPa									
420.18	0.0405	0.2577	2.575	0.979	394.40	0.5865	0.7845	1.188	1.386
412.97	0.0941	0.4136	2.185	0.997	393.15	0.6865	0.8246	1.112	1.547
407.28	0.1620	0.5471	1.990	0.978	392.47	0.7521	0.8510	1.072	1.698
403.65	0.2179	0.6137	1.856	0.994	391.93	0.8197	0.8814	1.038	1.890
399.91	0.2993	0.6765	1.677	1.038	391.45	0.8894	0.9181	1.012	2.160
397.48	0.4035	0.7197	1.431	1.137	391.21	0.9322	0.9447	1.002	2.398
395.75	0.4925	0.7525	1.298	1.244	391.08	0.9657	0.9705	0.998	2.539

Table 3. Composition ($x_{1(\mathrm{az})}$) and Boiling Temperature ($T_{(\mathrm{az})}$) of the Azeotropic Mixtures

system	P/kPa	$x_{1(\mathrm{az})}$	$T_{(\mathrm{az})}$/K
1-butanol + chlorocyclohexane	40.0	0.733	365.0
	101.3	0.846	390.0
chlorobenzene	40.0	0.549	362.3
	101.3	0.687	388.5
bromobenzene	40.0	0.935	366.7

the determination of both liquid and vapor phase mol fractions is ±0.0001.

Experimental Results

Vapor–liquid equilibrium data (T, x_1, and y_1), along with calculated activity coefficients at 40.0 and 101.3 kPa, are presented in Table 2 and Figures 1-4. The systems show positive deviations from ideality. Some of the systems show minimum temperature azeotropes. Information about composition and boiling temperature of the azeotropes is summarized in Table 3.

The activity coefficients γ_i were calculated, taking into account the non-ideality of the vapor phase, from the following equations:

$$\gamma_i = \frac{y_i P}{x_i P_i^{\mathrm{o}}} \exp\left[\frac{(B_{ii} - V_i^{\mathrm{o}})(P - P_i^{\mathrm{o}}) + (1 - y_i)^2 P \delta_{ij}}{RT}\right] \qquad (1)$$

$$\delta_{ij} = 2B_{ij} - B_{ii} - B_{jj} \qquad (2)$$

where x_i and y_i are the liquid and vapor phase compositions, P is the total pressure, P_i^{o} are the vapor pressures of the pure compounds calculated by the Antoine equation, where the constants are given in Table 4, B_{ii} are the second virial coefficients, B_{ij} are the cross second virial coefficients calculated using a suitable mixing rule (Amdur and Mason, 1958), and V_i^{o} are the molar volumes of the saturated liquids. The correction for the nonideality of the vapor

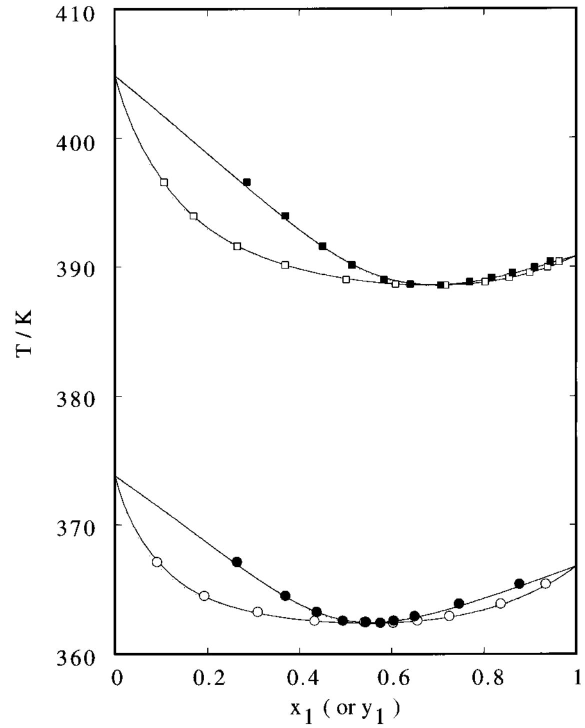

Figure 2. $T-x_1-y_1$ diagram for 1-butanol (1) + chlorobenzene (2): (○, ●) experimental data at 40.0 kPa; (□, ■) experimental data at 101.3 kPa; (—) Wilson equation.

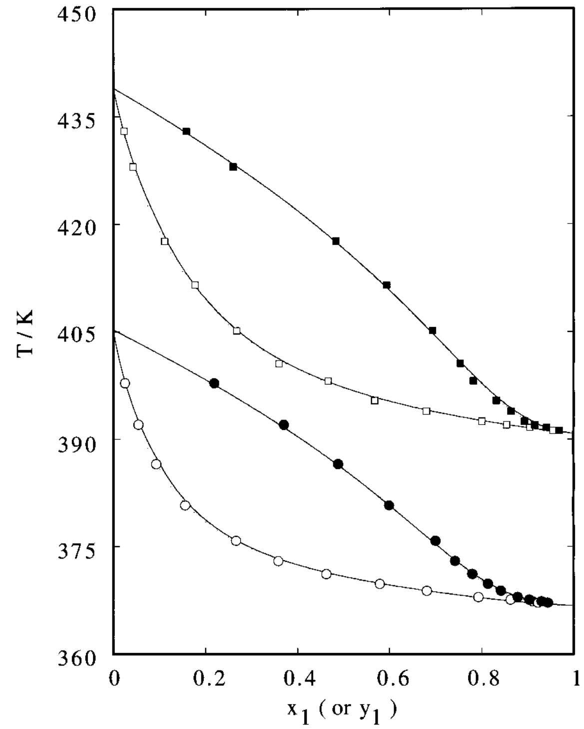

Figure 3. $T-x_1-y_1$ diagram for 1-butanol (1) + bromocyclohexane (2): (○, ●) experimental data at 40.0 kPa; (□, ■) experimental data at 101.3 kPa; (—) Wilson equation.

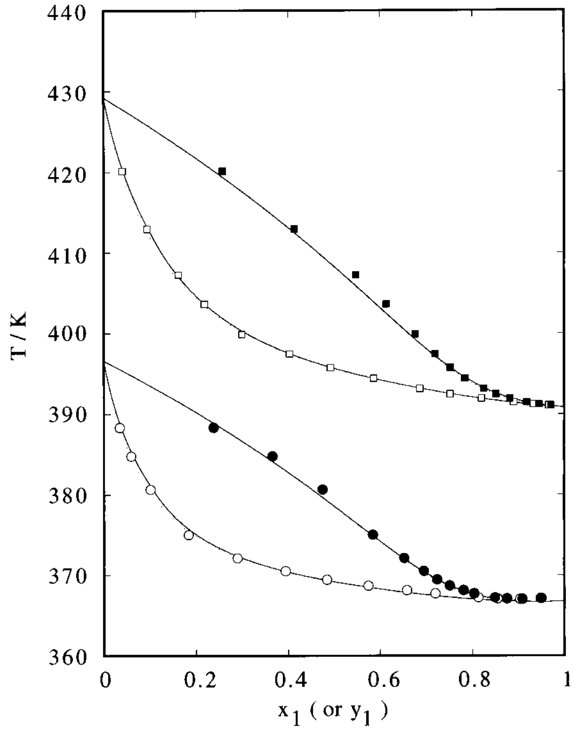

Figure 4. $T-x_1-y_1$ diagram for 1-butanol (1) + bromobenzene (2): (○, ●) experimental data at 40.0 kPa; (□, ■) experimental data at 101.3 kPa; (—) Wilson equation.

Table 4. Constants of the Antoine Equation, $\log(P/\text{kPa}) = A - B/(C + t/°\text{C})$ for Vapor Pressures of the Pure Compounds

compound	*A*	*B*	*C*
1-butanol	6.547 43	1338.769	177.042
chlorocyclohexane	5.735 40	1235.480	188.647
chlorobenzene	6.309 63	1556.6	230
bromocyclohexane	6.12 93	1568.129	214.459
bromobenzene	6.379 12	1688.4	230

Table 5. Experimental Vapor Pressures of Bromocyclohexane

P_i^o/kPa	*T*/K	P_i^o/kPa	*T*/K	P_i^o/kPa	*T*/K	P_i^o/kPa	*T*/K
5.0	346.70	30.0	396.07	60.0	419.09	90.2	434.41
7.5	356.75	35.0	400.32	65.0	422.09	95.0	436.34
10.2	365.27	40.0	405.30	69.8	424.69	101.3	438.88
15.0	374.89	45.0	408.74	75.0	427.34		
20.0	383.83	50.0	412.83	80.2	429.91		
25.0	389.84	55.0	415.79	85.0	432.05		

Table 6. Results of the Thermodynamic Consistency Test, Average Deviation ΔP and Δy

system	*P*/kPa	ΔP/kPa	Δy
1-butanol + chlorocyclohexane	40.0	0.2	0.0042
	101.3	0.5	0.0010
chlorobenzene	40.0	0.2	0.0023
	101.3	0.3	0.0010
bromocyclohexane	40.0	0.2	0.0021
	101.3	0.8	0.0039
bromobenzene	40.0	0.4	0.0044
	101.3	0.5	0.0063

phase, represented by the exponential term in eq 1, was important only at low mole fractions.

Antoine constants for bromocyclohexane were obtained from our own vapor pressure measurements over the pressure range 5.0−101.3 kPa using the same still as for the binary systems; the results are shown in Table 5. The constants were calculated by a nonlinear regression procedure (Nelder and Mead, 1965), the standard deviation is 0.2 kPa. The Antoine constants for 1-butanol, chlorobenzene, and bromobenzene were obtained from Riddick et al. (1986) and for chlorocyclohexane were taken from Gmehling et al. (1984). The second virial coefficients for 1-butanol were obtained from TRC tables (TRC, 1972) and for the halogenated compounds were estimated by the Redlich−Kwong equation (Redlich and Kwong, 1949). The molar volumes of the saturated liquids were calculated using the Yen and Woods method (Yen and Woods, 1968).

The thermodynamic consistency of the experimental results was checked using the Van Ness method (Van Ness et al., 1973), described by Fredenslund et al. (1977) using

Table 7. Correlation Parameters, Average Deviations ΔT and Δy, and Activity Coefficients at Infinite Dilution

equation	A_{12}	A_{21}	ΔT/K	Δy	γ_1^∞	γ_2^∞	equation	A_{12}	A_{21}	ΔT/K	Δy	γ_1^∞	γ_2^∞
						1-Butanol (1) + Chlorocyclohexane (2) at 40.0 kPa							
Margules	1.2239[a]	1.0912[a]	0.18	0.0043	3.40	2.98	NRTL (α_{12} = 0.3)	1246.7912[b]	2736.4287[b]	0.18	0.0034	3.34	2.99
Van Laar	1.2252[a]	1.0962[a]	0.18	0.0043	3.40	2.99	UNIQUAC	−417.6592[b]	1650.5740[b]	0.18	0.0034	3.33	2.98
Wilson	3427.7302[b]	871.1244[b]	0.21	0.0039	3.41	3.08							
						1-Butanol (1) + Chlorocyclohexane (2) at 101.3 kPa							
Margules	1.0014	0.9733	0.16	0.0026	2.72	2.65	NRTL	1384.8161	2163.5508	0.15	0.0026	2.67	2.64
Van Laar	1.0013	0.9736	0.16	0.0026	2.72	2.65	UNIQUAC	−337.2761	1405.8982	0.15	0.0026	2.66	2.64
Wilson	3012.1004	767.2333	0.18	0.0030	2.70	2.67							
						1-Butanol (1) + Chlorobenzene (2) at 40.0 kPa							
Margules	1.3233	0.9538	0.12	0.0034	3.76	2.60	NRTL	316.0979	3858.9302	0.14	0.0028	3.82	2.64
Van Laar	1.3660	0.9716	0.12	0.0028	3.92	2.64	UNIQUAC	−489.4778	1708.7576	0.14	0.0026	3.90	2.65
Wilson	3919.9337	592.5934	0.15	0.0022	4.11	2.70							
						1-Butanol (1) + Chlorobenzene (2) at 101.3 kPa							
Margules	1.0296	0.8769	0.08	0.0012	2.80	2.40	NRTL	720.4241	2750.9572	0.09	0.0010	2.77	2.41
Van Laar	1.0379	0.8800	0.09	0.0010	2.82	2.41	UNIQUAC	−109.4337	1016.8000	0.09	0.0010	2.78	2.41
Wilson	2931.9283	754.1510	0.10	0.0011	2.85	2.43							
						1-Butanol (1) + Bromocyclohexane (2) at 40.0 kPa							
Margules	1.0720	1.1895	0.12	0.0017	2.92	3.29	NRTL	2131.9181	1764.2394	0.11	0.0029	2.85	3.27
Van Laar	1.0739	1.1919	0.12	0.0017	2.92	3.29	UNIQUAC	−96.9553	1192.6702	0.11	0.0029	2.83	3.25
Wilson	2848.6626	1365.0296	0.14	0.0029	2.87	3.36							
						1-Butanol (1) + Bromocyclohexane (2) at 101.3 kPa							
Margules	0.7125	1.1331	0.23	0.0037	2.04	3.11	NRTL	3942.7229	−199.3744	0.21	0.0035	2.06	3.16
Van Laar	0.7425	1.1670	0.20	0.0041	2.10	3.21	UNIQUAC	969.2684	−45.8593	0.22	0.0035	2.06	3.15
Wilson	1503.5594	2352.7723	0.21	0.0037	2.07	3.23							
						1-Butanol (1) + Bromobenzene (2) at 40.0 kPa							
Margules	1.2507	1.1207	0.23	0.0046	3.49	3.07	NRTL	1220.1742	2940.6525	0.23	0.0055	3.40	3.07
Van Laar	1.2523	1.1242	0.23	0.0047	3.50	3.08	UNIQUAC	−37.7463	1134.2357	0.23	0.0053	3.41	3.07
Wilson	3229.8407	1251.2938	0.24	0.0059	3.48	3.16							
						1-Butanol (1) + Bromobenzene (2) at 101.3 kPa							
Margules	0.9636	1.0004	0.16	0.0063	2.62	2.72	NRTL	1647.3052	1901.3544	0.13	0.0077	2.55	2.71
Van Laar	0.9636	1.0008	0.16	0.0063	2.62	2.72	UNIQUAC	378.1179	490.5635	0.14	0.0074	2.55	2.71
Wilson	2442.6726	1342.6783	0.14	0.0076	2.58	2.74							

[a] Dimensionless. [b] Joules per mole.

Table 8. VLE Predictions, Average Deviations ΔT and Δy

		UNIFAC		ASOG	
system	P/kPa	ΔT/K	Δy	ΔT/K	Δy
1-butanol + chlorocyclohexane	40.0	0.58	0.0099	0.06	0.0024
	101.3	0.82	0.0096	0.30	0.0041
chlorobenzene	40.0	0.71	0.0123	0.11	0.0050
	101.3	0.46	0.0083	0.60	0.0130
bromocyclohexane	40.0	1.54	0.0173	4.26	0.0431
	101.3	1.63	0.0152	4.95	0.0405
bromobenzene	40.0	0.42	0.0079	1.33	0.0142
	101.3	0.41	0.0050	1.10	0.0089

a third-order Legendre polynomial for the excess free energies. According to this test, experimental data are considered consistent if the average deviation in y (Δy) is smaller than 0.01. All the experimental data are consistent, as one can see in Table 6.

The activity coefficients were correlated with the Margules, Van Laar, Wilson, NRTL, and UNIQUAC equations. Estimation of the parameters for all the equations was based on minimization, using the mentioned nonlinear regression procedure, of an objective function F in terms of experimental and calculated γ_i values. The function F (Silverman and Tassios, 1984) for a binary system can be stated as

$$F = \sum_{i=1}^{i=N}\left[\left(\frac{\gamma_1^{\mathrm{exp}} - \gamma_1^{\mathrm{cal}}}{\gamma_1^{\mathrm{exp}}}\right)^2 + \left(\frac{\gamma_2^{\mathrm{exp}} - \gamma_2^{\mathrm{cal}}}{\gamma_2^{\mathrm{exp}}}\right)^2\right]_i \tag{3}$$

where N is the number of experimental data. The parameters A_{12} and A_{21}, see definitions in Gmehling et al. (1984), along with the average deviation in T (ΔT), the average deviation in y (Δy), and the activity coefficients at infinite dilution are listed in Table 7. The average deviation in temperature is less than 0.3 K and in vapor composition is less than 0.01, so all the equations correlated the activity coefficients quite well.

VLE Predictions

The UNIFAC method and the ASOG method were employed to predict the vapor−liquid equilibrium. For all systems the temperature and vapor-phase composition obtained experimentally were compared with the theoretical predictions and the average deviations in temperature and vapor-phase composition are given in Table 8. These values indicate that the two methods give satisfactory predictions, except for the system 1-butanol + bromocyclohexane when the ASOG method is used. It can be also observed that both methods yield similar results although the UNIFAC method predicts somewhat better the experimental behavior of the systems containing bromobenzene and bromocyclohexane.

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