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Vapor−Liquid Equilibria of Binary and Ternary Mixtures of 2-Propanol, 1-Chlorobutane, and Acetonitrile at 101.3 kPa

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Vapor−liquid equilibria were measured at 101.3 kPa for three binary mixtures and one ternary mixture of 2-propanol, 1-chlorobutane, and acetonitrile. Thermodynamic consistency of data for the three binary mixtures was tested using the Herington method. The Soave−Redlich−Kwong equation of state was used to calculate the vapor-phase fugacity coefficients. The experimental vapor−liquid equilibrium data were reduced, and binary parameters for four activity coefficient models, such as van Laar, Wilson, NRTL, and UNIQUAC, were fitted. The four models with their best-fitted parameters were used to predict the ternary vapor−liquid equilibria. A comparison of model performances was made by using the criterion of absolute average deviations in boiling point and in vapor-phase composition.

Introduction

The binary systems in this work have interesting characteristics with respect to the solubility of polymers. At 25 °C the pure liquids 2-propanol, 1-chlorobutane, and acetonitrile are poor solvents of poly(methyl methacrylate), while mixtures of these liquids act as a good solvent for such a polymer (Fernández-Piérola and Horta, 1981). To adequately interpret the thermodynamic behavior of polymers in mixed solvents, it is necessary to know the vapor−liquid equilibria (VLE) of the mixtures acting as solvents. On the other hand, accurate VLE is of theoretical as well as practical importance from the standpoint of the technology of separation processes. A survey of the literature disclosed isobaric VLE data for the 1-chlorobutane + 2-propanol system at 94.4 kPa (Wisniak and Akunis, 1995) and for the acetonitrile + 2-propanol system at 101.33 kPa (Mato and Gonzalez-Benito, 1985). The literature only contains isothermal values for the mixture acetonitrile + 1-chlorobutane at 298.69, 348.18, and 398.89 K (Khurma et al., 1983). Despite these earlier measurements, it was decided to make additional experimental measurements with the goal of completing an updated study that would enhance our understanding of their behavior.

In the present paper VLE were measured for the ternary system 2-propanol + 1-chlorobutane + acetonitrile and three constituent binary systems 2-propanol + 1-chlorobutane, 2-propanol + acetonitrile, and 1-chlorobutane + acetonitrile at 101.3 kPa using an all-glass recirculation still. The results of the three binary mixtures have been treated thermodynamically, considering the nonideality of both phases and verifying their thermodynamic consistency. Various activity coefficient models with their best-fitted binary parameters were used to predict the vapor−liquid equilibria of the ternary system.

Experimental Section

Materials. All organic chemicals used in this study were reagent grade. 2-Propanol (99.9 mass %), 1-chlorobutane (99.5 mass %), and acetonitrile (99.9 mass %) were supplied by Fisher Scientific (U.S.A.). All chemicals were dried with 3-Å molecular sieves. In all cases water contents in the liquids used were not greater than 0.05% (v/v). No further purification was performed for these chemicals, and no detectable impurities were found with the gas chromatography (GC). The pure component properties were measured in this work, and the comparisons with published values are shown in Table 1. Densities, ρ, of pure compounds were measured at (298.15 ± 0.01) K using the Anton Paar DMA 58 densimeter with an accuracy of $\pm 1 \times 10^{-5}$ g/cm^3. Refractive indices, n_D, of pure chemicals were measured at (298.15 ± 0.1) K by an Abbe refractometer, Atago RX-5000, with an accuracy of ±0.000 01 unit.

Apparatus and Procedure. The apparatus used was an all-glass equilibrium still (NGW, Germany) with a provision for both vapor and liquid recirculation as described by Hunsmann (1967). The still has a total capacity of about 100 cm^3. The equilibrium temperature was measured by a calibrated Pt-100 resistance thermometer with an accuracy of ±0.01 K. The pressure was maintained at 101.3 kPa through a piezoresistive absolute-pressure transducer. The accuracy of pressure control was within ±0.1 kPa.

The charge for experimental runs was prepared by mixing known masses of the three components. The steady conditions were reached in about 1 h as indicated by the constant boiling temperatures of the liquid and vapor phases within the equilibrium cell, and their difference was within 0.02 K. The system was allowed to maintain this equilibrium state for about 30 min, before samples were taken.

Compositions of sampled liquid and condensed vapor phases were analyzed with a Perkin-Elmer Autosystem gas chromatograph (GC), after calibration with gravimetrically prepared standard solutions. A flame ionization detector was used along with a 3m × 3 mm SS column packed by Porapak Q. The GC response peaks were integrated by using a Perkin-Elmer 1020 integrator. At least two analyses were made of each liquid and each vapor composition. The accuracy of equilibrium composition measurements was estimated as ±0.001 mole fraction.

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Table 1. Comparison of the Measured Normal Boiling Points, Densities, and Refractive Indices of Pure Components in This Work with Literature Values

	T_b/K		ρ(298.15 K)/g cm^{-3}		n_D	
components	this work	lit.	this work	lit.	this work	lit.
2-propanol	355.40	355.41[a] 355.39[b]	0.781 05	0.780 9[a] 0.781 0[c]	1.375 79	1.3752[a]
1-chlorobutane	351.58	351.58[a] 351.73[d]	0.880 94	0.880 97[e] 0.880 95[f] 0.880 93[g]	1.400 01	1.4000[a]
acetonitrile	354.70	354.95[h] 354.75[i]	0.776 68	0.776 49[f] 0.776 76[j] 0.776 0[k]	1.342 05	1.342[l]

[a] TRC, 1994. [b] Ambrose and Sprake, 1970. [c] Proust and Stein, 1979. [d] Kemme and Kreps, 1969. [e] Artigas et al., 1993. [f] Riddick et al., 1986. [g] Prolong et al., 1984. [h] Dojcansky and Heinrich, 1974. [i] Lide and Kehiaian, 1974. [j] Stein and Proust, 1971. [k] Mollerup and Fredenslund, 1976. [l] Moumouzias et al., 1991.

Table 2. Experimental VLE Data for 2-Propanol (1) + 1-Chlorobutane (2) at 101.3 KPa

T/K	x_1	y_1	γ_1	γ_2
351.58	0.000	0.000		1.000
348.39	0.046	0.130	3.757	1.005
345.67	0.139	0.251	2.682	1.043
345.49	0.151	0.256	2.537	1.056
345.02	0.193	0.288	2.277	1.079
344.71	0.237	0.312	2.035	1.114
344.50	0.296	0.338	1.780	1.169
344.41	0.360	0.364	1.582	1.239
344.42	0.395	0.379	1.501	1.279
344.53	0.462	0.401	1.352	1.383
344.67	0.504	0.418	1.284	1.451
344.99	0.567	0.448	1.207	1.561
345.22	0.600	0.460	1.160	1.641
345.89	0.670	0.504	1.107	1.789
346.26	0.699	0.526	1.091	1.853
347.57	0.778	0.597	1.054	2.051
348.65	0.824	0.646	1.031	2.198
350.17	0.877	0.728	1.027	2.308
350.95	0.899	0.756	1.009	2.463
353.95	0.969	0.908	0.999	2.767
355.40	1.000	1.000	1.000	

Table 3. Experimental VLE Data for 2-Propanol (1) + Acetonitrile (3) at 101.3 KPa

T/K	x_1	y_1	γ_1	γ_3
354.70	0.000	0.000		1.000
352.86	0.053	0.098	2.057	1.005
351.78	0.093	0.158	1.972	1.013
350.86	0.146	0.227	1.872	1.016
350.17	0.195	0.276	1.751	1.031
349.81	0.228	0.306	1.685	1.043
349.45	0.270	0.339	1.599	1.062
349.01	0.350	0.400	1.481	1.098
348.90	0.384	0.419	1.421	1.125
348.79	0.453	0.455	1.313	1.193
348.80	0.504	0.485	1.258	1.243
348.87	0.549	0.518	1.230	1.276
349.06	0.613	0.548	1.156	1.387
349.24	0.651	0.581	1.146	1.417
349.37	0.682	0.592	1.109	1.509
349.87	0.740	0.634	1.073	1.629
350.47	0.796	0.682	1.046	1.771
351.37	0.850	0.743	1.031	1.893
352.05	0.889	0.786	1.015	2.086
353.38	0.940	0.867	1.006	2.303
355.40	1.000	1.000	1.000	

Table 4. Experimental VLE Data for 1-Chlorobutane (2) + Acetonitrile (3) at 101.3 KPa

T/K	x_2	y_2	γ_2	γ_3
354.70	0.000	0.000		1.000
351.88	0.038	0.111	2.896	1.005
349.07	0.095	0.224	2.544	1.018
347.77	0.134	0.280	2.346	1.028
346.57	0.186	0.333	2.086	1.053
345.88	0.229	0.370	1.924	1.073
345.32	0.274	0.395	1.747	1.114
344.89	0.322	0.428	1.632	1.144
344.51	0.384	0.462	1.495	1.199
344.33	0.428	0.475	1.387	1.267
344.22	0.480	0.500	1.307	1.332
344.18	0.532	0.526	1.242	1.405
344.22	0.579	0.546	1.183	1.494
344.41	0.643	0.581	1.127	1.616
344.64	0.687	0.612	1.103	1.694
345.11	0.743	0.654	1.074	1.812
345.67	0.793	0.689	1.041	1.987
346.62	0.850	0.741	1.014	2.215
348.01	0.908	0.825	1.013	2.335
349.10	0.942	0.877	1.004	2.515
351.58	1.000	1.000	1.000	

Results and Discussion

VLE had been measured at 101.3 kPa for three binary mixtures of 2-propanol + 1-chlorobutane, 2-propanol + acetonitrile, and 1-chlorobutane + acetonitrile. The results are presented in Tables 2−4, respectively. The activity coefficient γ of pure liquid i in a nonideal mixture at temperature T and pressure P was calculated from

$$\varphi_i P y_i = \gamma_i P_i^\circ x_i \varphi_i^{\mathrm{s}} \exp[v_i^{\mathrm{L}}(P - P_i^\circ)/RT] \tag{1}$$

where x and y are the liquid and vapor mole fractions in equilibrium, φ is the vapor-phase fugacity coefficient, P° is the vapor pressure of pure component, v^{L} is the liquid molar volume, and superscript s stands for saturation. The vapor-phase fugacity coefficients were calculated from the Soave−Redlich−Kwong (SRK) equation of state (Soave, 1972), where the binary interaction parameter, k_{ij}, was set to be 0. The vapor pressures of the pure components were calculated from

$$\ln(P^\circ/\mathrm{Pa}) = A + B(T/\mathrm{K}) + C\ln(T/\mathrm{K}) + D(T/\mathrm{K})^E \tag{2}$$

where A, B, C, D, and E are coefficients. The liquid molar volumes were evaluated from the Rackett equation as modified by Spencer and Danner (1972). The calculated activity coefficients for the binary systems were also presented in Tables 2−4.

Table 5 presents the parameters of the pure component pertaining to this investigation. This table includes the vapor pressure constants (A, B, C, D, and E), critical properties, acentric factors, Rackett constants, and UNIQUAC parameters for pure components. The vapor-pressure constants of 2-propanol, 1-chlorobutane, and acetonitrile were obtained form a regression of the data sources reported by Boublik et al. (1984). The critical properties and acentric factors were obtained from the compilation of Reid et al. (1987). The Rackett constant of 1-chlorobutane was estimated from the method proposed

Table 5. Pure Component Properties Used in This Work[a]

parameters	2-propanol	1-chlorobutane	acetonitrile
A	92.935	85.569	58.302
B	−8177.1	−6530.0	−5385.6
C	−10.031	−9.6276	−5.4954
D	3.9988×10^{-6}	7.8596×10^{-6}	5.3634×10^{-6}
E	2	2	2
T_c/K	508.3	537.0	545.5
P_c/bar	47.6	38.2	48.3
ω	0.665	0.274	0.327
Z_{RA}	0.2499	0.2550	0.1987
UNIQUAC q	2.5078	3.1919	1.7237
UNIQUAC r	2.7790	3.7153	1.8701

[a] A, B, C, D, and E are parameters in the vapor pressure equation: $\ln(P°/\mathrm{Pa}) = A + B/(T/\mathrm{K}) + C \ln(T/\mathrm{K}) + D(T/\mathrm{K})^E$.

Table 6. Correlation Parameters and the Absolute Average Deviations in Boiling Points and Vapor-Phase Mole Fractions for the Binary Systems[a]

models	A_{12}	A_{21}	α_{12}	$(\Delta T)_{av}$/K	$(\Delta y)_{av}$
	2-Propanol + 1-Chlorobutane				
van Laar	1.4875	1.0337		0.13	0.002
Wilson	555.61	15.523		0.06	0.002
NRTL	73.817	451.11	0.333 85	0.06	0.002
UNIQUAC	−97.705	32.597		0.08	0.002
	2-Propanol + Acetonitrile				
van Laar	0.81226	0.96777		0.06	0.002
Wilson	−24.480	385.80		0.05	0.002
NRTL	274.35	65.305	0.295 91	0.10	0.002
UNIQUAC	287.55	−72.728		0.16	0.003
	1-Chlorobutane + Acetonitrile				
van Laar	1.1979	0.99572		0.10	0.002
Wilson	59.512	398.70		0.04	0.003
NRTL	108.16	320.93	0.316 40	0.07	0.002
UNIQUAC	265.71	−27.367		0.07	0.003

[a] The binary adjustable parameters for various models are: Wilson, $A_{ij} = (\lambda_{ij} - \lambda_{ii})/R$; NRTL, $A_{ij} = (g_{ij} - g_{jj})/R$; UNIQUAC, $A_{ij} = (U_{ij} - U_{jj})/R$.

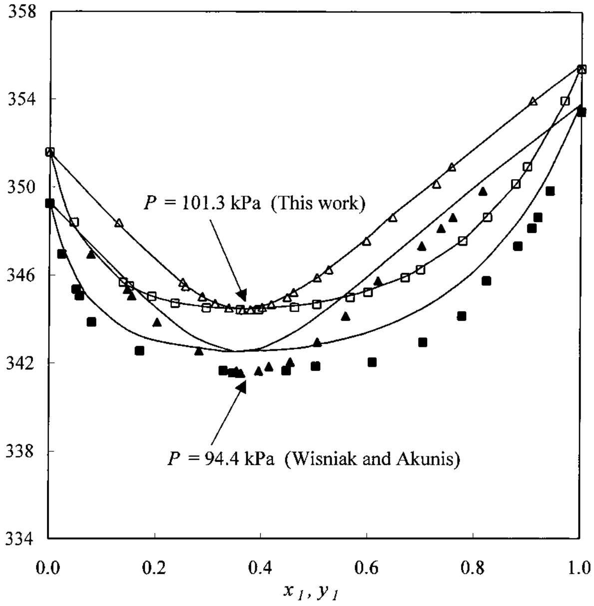

Figure 1. $T-x-y$ dragram for the 2-propanol (1) + 1-chlorobutane (2) system: present work at 101.3 kPa, (□) liquid phase; (△) vapor phase; Wisniak and Akunis (1995) at 94.4 kPa, (■) liquid phase, (▲) vapor phase; (—) Wilson model with parameters from Table 6.

by Vetere (1992). The UNIQUAC parameters depend only on the molecular structure of the pure components and have been obtained from Fredenslund et al. (1977).

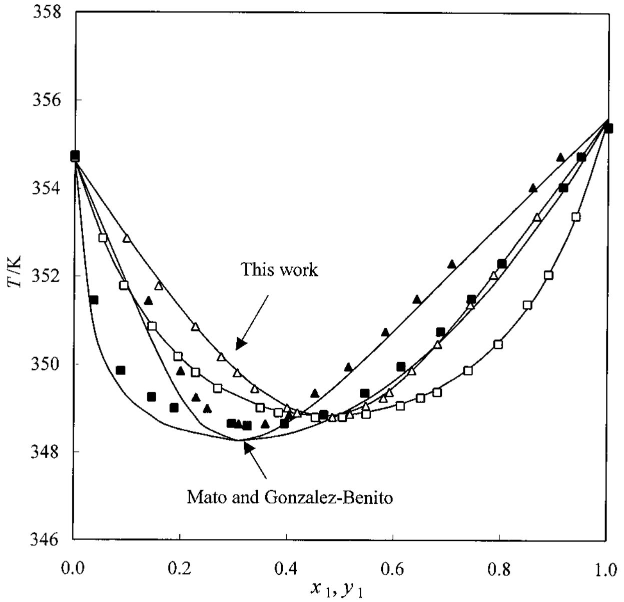

Figure 2. $T-x-y$ dragram for the 2-propanol (1) + acetonitrile (3) system at 101.3 kPa: present work, (□) liquid phase, (△) vapor phase; Mato and Gonzalez-Benito (1985), (■) liquid phase, (▲) vapor phase; (—) fitted by Wilson model.

All three binary systems satisfactorily passed the Redlich–Kister thermodynamic consistency test using the Herington analysis (van Winkle, 1967). According to the Herington method, the experimental data are thermodynamically consistent if (D − J) is less than 0. The Herington's $D - J$ for the Redlich–Kister test was found to be −19.61, −7.61, and −14.91 for 2-propanol + 1-chlorobutane, 2-propanol + acetonitrile, and 1-chlorobutane + acetonitrile, respectively. The VLE experimental data of the three binary systems were then used to obtain the binary parameters of various activity coefficient models. The van Laar, Wilson, NRTL, and UNIQUAC models were employed in our regressions. The expressions of the activity coefficient by these models were listed in the literature (Prausnitz et al., 1986). Estimation of energy parameters of all the models studied was based on minimization of the objective function F in terms of predicted and experimental γ values. The function F for a binary system can be stated as

$$F = \sum_{k=1}^{N}\sum_{i=1}^{2}\left[\left(\frac{\gamma_i^{e} - \gamma_i^{c}}{\gamma_i^{e}}\right)^2\right]_k \tag{3}$$

where N is the number of data points and the superscripts "e" and "c" indicate experimental and calculated values, respectively. The fitted parameters together with the mean values of the absolute deviations in boiling point, ΔT, and in vapor-phase mole fraction, Δy are shown in Table 6. The deviations of the regressions are reasonably small, and it indicates that all activity coefficient models are adequate to represent the binary experimental data. Generally, the Wilson equation yielded the best overall results.

All three binary systems form a minimum boiling azeotrope. The azeotropic temperatures at 101.3 kPa are 344.42 K with $x_1 = 0.371$ for 2-propanol (1) + 1-chlorobutane (2), 348.80 K with $x_1 = 0.475$ for 2-propanol (1) + acetonitrile (3), and 344.19 K with $x_2 = 0.533$ K for 1-chlorobutane (2) + acetonitrile (3). Figure 1 plots the experimental values at 101.3 kPa presented herein and the values at 94.4 kPa published by Wisniak and Akunis (1995)

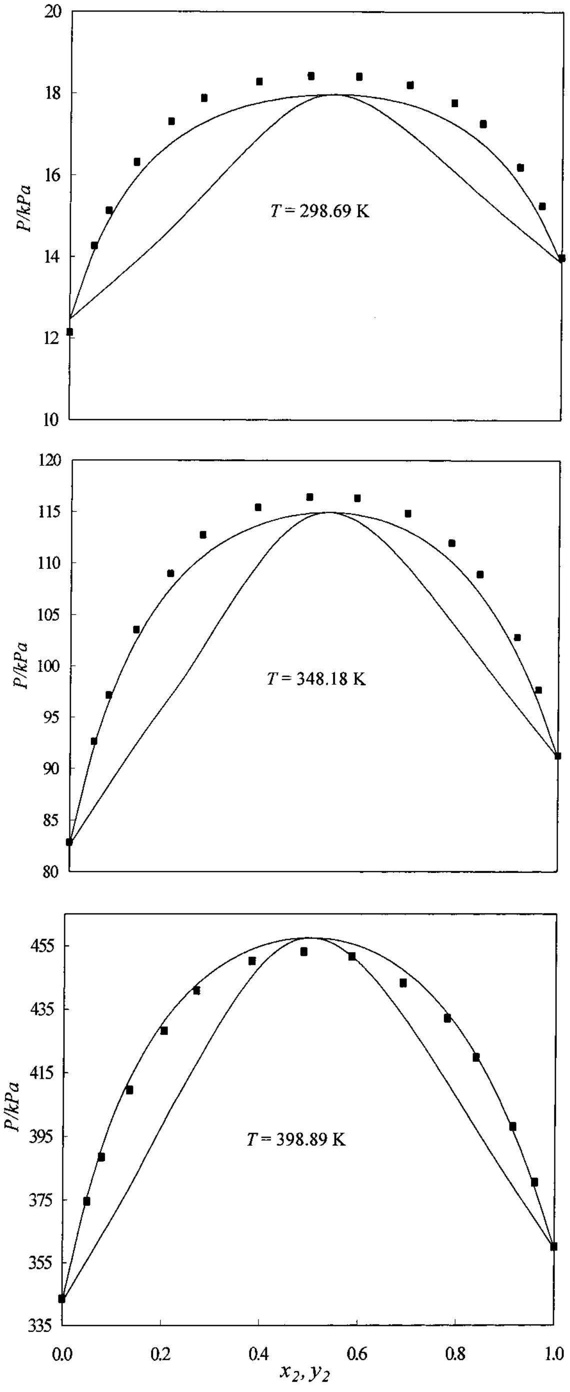

Figure 3. $P-x-y$ diagram for the 1-chlorobutane (2) + acetonitrile (3) system at 298.69, 348.18, and 398.89 K: (■) experimental $P-x$ data from Khurma et al. (1983); (—) Wilson model with parameters from Table 6.

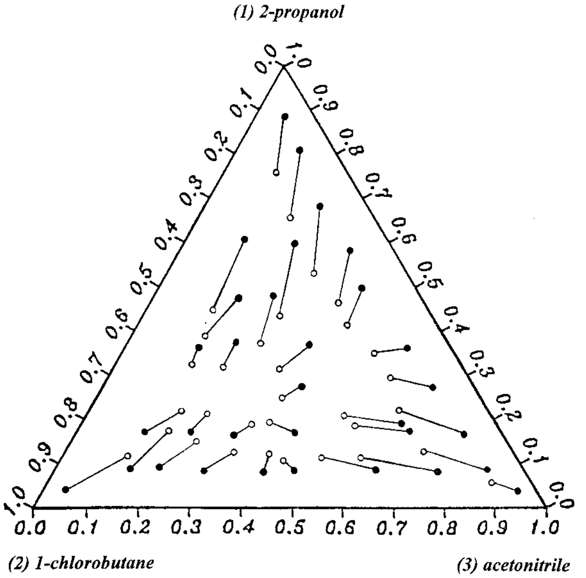

Figure 4. Composition chart for the 2-propanol (1) + 1-chlorobutane (2) + acetonitrile (3) system at 101.3 kPa: (●) liquid phase, (○) vapor phase.

Table 7. Experimental VLE Data for 2-Propanol (1) + 1-Chlorobutane (2) + Acetonitrile (3) at 101.3 KPa

T/K	x_1	x_2	y_1	y_2	γ_1	γ_2	γ_3
346.95	0.047	0.912	0.118	0.803	3.538	1.014	2.446
351.17	0.895	0.051	0.761	0.129	1.011	2.561	2.265
345.03	0.084	0.824	0.159	0.696	2.887	1.033	2.127
344.01	0.156	0.744	0.215	0.642	2.193	1.090	1.995
343.37	0.370	0.518	0.323	0.540	1.427	1.343	1.742
343.79	0.482	0.404	0.383	0.479	1.276	1.508	1.701
344.78	0.605	0.284	0.440	0.416	1.121	1.805	1.765
349.37	0.828	0.074	0.665	0.175	1.026	2.529	1.920
343.70	0.099	0.711	0.151	0.604	2.459	1.083	1.817
343.09	0.166	0.632	0.195	0.568	1.943	1.168	1.686
343.06	0.367	0.414	0.307	0.462	1.385	1.452	1.517
343.84	0.485	0.295	0.370	0.389	1.223	1.674	1.536
345.17	0.592	0.191	0.437	0.305	1.119	1.944	1.597
347.29	0.693	0.093	0.533	0.184	1.069	2.255	1.660
343.31	0.089	0.622	0.126	0.555	2.320	1.152	1.575
342.82	0.166	0.523	0.187	0.497	1.885	1.246	1.473
343.41	0.382	0.297	0.312	0.379	1.333	1.642	1.369
346.71	0.596	0.087	0.476	0.167	1.137	2.227	1.440
343.14	0.095	0.505	0.116	0.493	2.015	1.267	1.403
342.93	0.175	0.409	0.180	0.439	1.713	1.403	1.323
343.07	0.281	0.339	0.252	0.398	1.484	1.527	1.325
345.99	0.500	0.103	0.415	0.188	1.217	2.166	1.309
343.19	0.099	0.454	0.121	0.469	2.013	1.339	1.314
344.61	0.192	0.190	0.205	0.297	1.657	1.937	1.102
346.05	0.374	0.090	0.352	0.162	1.376	2.132	1.184
344.00	0.094	0.293	0.114	0.380	1.931	1.638	1.152
344.84	0.179	0.179	0.196	0.289	1.683	1.986	1.089
346.51	0.282	0.081	0.299	0.162	1.521	2.335	1.089
345.40	0.095	0.182	0.120	0.305	1.897	2.025	1.061
346.85	0.184	0.087	0.222	0.179	1.707	2.377	1.046
347.76	0.090	0.088	0.128	0.194	1.939	2.476	1.020
350.77	0.040	0.037	0.070	0.104	2.114	2.881	1.007

for the 2-propanol + 1-chlorobutane system. The minimum azeotrope at 94.4 kPa was reported to contain 37.2 mol % 2-propanol and to boil at 341.60 K. As shown in Figure 1, VLE were also calculated for 94.4 kPa using the Wilson model with the parameters from Table 6, but the boiling temperatures of Wisniak and Akunis are about 0.92 K less than ours. Figure 2 contains a comparison of our experimental values with the values presented by Mato and Gonzalez-Benito (1985) for the mixture of 2-propanol + acetonitrile at 101.3 kPa. The discrepancy between the present values and the literature values was substantial. The composition value for the azeotrope presented here was 47.5 mol % 2-propanol as compared to the literature value of 32.5 mol % 2-propanol. However, our investigation may be well supported by the fact that the composition at maximum excess volume is shifted from 40 to 45 mol % 2-propanol as temperature increases from 293.15 to 323.15 K (Paez and Contreras, 1995). Figure 3 presents comparisons of $p-x-y$ curves calculated using the Wilson model with the parameters from Table 6 and the corresponding experimental $p-x$ values obtained from Khurma et al. (1983) for the mixture of 1-chlorobutane + acetonitrile at 298.69, 348.18, and 398.89 K. In all cases a reasonable agreement between experimental and calculated results is obtained.

Table 8. Absolute Average Deviations and Maximum Deviations for the Prediction of the 2-Propanol (1) + 1-Chlorobutane (2) + Acetonitrile (3) Ternary from Their Constituent Binaries

deviations	van Laar	Wilson	NRTL	UNIQUAC
$(\Delta T)_{\mathrm{av}}$/K	0.21	0.13	0.07	0.27
$(\Delta T)_{\max}$/K	0.34	0.21	0.24	0.41
$(\Delta y_1)_{\mathrm{av}}$	0.0035	0.0019	0.0034	0.0046
$(\Delta y_1)_{\max}$	0.0107	0.0079	0.0098	0.0107
$(\Delta y_2)_{\mathrm{av}}$	0.0033	0.0026	0.0026	0.0043
$(\Delta y_2)_{\max}$	0.0111	0.0104	0.0085	0.0129

Altogether 32 experimental vapor−liquid equilibrium measurements for the ternary system 2-propanol + 1-chlorobutane + acetonitrile were obtained at 101.3 kPa, and the results are presented in Table 7. The equilibrium composition diagram of the ternary system is shown in Figure 4. The experimental data of the ternary system were used to examine the capabilities of the four liquid models to predict ternary behavior from the constituent binary parameters given in Table 6. The calculated deviations from all these methods were summarized in Table 8. Of these predictive methods, the NRTL equation predicted the ternary data as well as the Wilson equation, followed by the van Laar and UNIQUAC equations.

Conclusion

Analysis of experimental data for the binary systems using the van Laar, Wilson, NRTL, and UNIQUAC equations shows that all four models were generally satisfactory for all the mixtures. Furthermore, the experimental measurements of the three binaries adequately fulfilled the thermodynamic requirements using the Herington test.

Inspection of various predictions for the ternary equilibrium based on the information of the constituent binaries shows that although four methods used (van Laar, Wilson, NRTL, and UNIQUAC) may be deemed satisfactory, the Wilson and NRTL methods exhibit somewhat smaller deviations from the experimental results than do the others.

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