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LIQUID–LIQUID EQUILIBRIA FOR TERNARY ACETONITRILE–ETHANOL–SATURATED HYDROCARBON AND ACETONITRILE–1-PROPANOL–SATURATED HYDROCARBON MIXTURES

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ABSTRACT

Isothermal vapour–liquid equilibrium data for acetonitrile–1-propanol at 45 °C were measured by use of a recirculating still. The liquid–liquid equilibria of (acetonitrile–ethanol)–(*n*-hexane or *n*-heptane or *n*-octane) and those of (acetonitrile–1-propanol)–(cyclohexane or *n*-hexane or *n*-heptane) were obtained from measurements of tie-lines. The experimental results were compared with those calculated from the UNIQUAC associated-solution model.

INTRODUCTION

Ternary liquid–liquid equilibrium (LLE) data are essential for the testing of solution models. The UNIQUAC associated-solution model has been shown to be useful in the calculations of phase equilibria for binary and ternary alcohol mixtures [1–4]. My preliminary study indicated that the ternary experimental LLE data of Kikic et al. [5] for acetonitrile–ethanol–saturated hydrocarbon mixtures at 25 °C have much larger solubility envelopes than those calculated from the UNIQUAC associated-solution model. This suggests that further experimental work on these mixtures will be necessary.

In this paper, I report the vapour–liquid equilibrium (VLE) data of acetonitrile–1-propanol at 45 °C and the results of LLE studies of ternary mixtures of ethanol or 1-propanol with acetonitrile and saturated hydrocarbons and compare the experimental values with those estimated by means of the UNIQUAC associated-solution model [2]. Binary isothermal VLE, used to evaluate the binary parameters of the UNIQUAC associated-solution model, have been taken from the literature: for ethanol–acetonitrile at 20 °C [6]; for ethanol–*n*-hexane at 25 °C [6]; for ethanol–*n*-heptane at 30 °C [7]; for ethanol–*n*-octane at 45 °C [6]; for 1-propanol–benzene at 45 °C [6]; for 1-propanol–cyclohexane at 25 °C [6]; for 1-propanol–*n*-hexane

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at 25 °C [6]; for 1-propanol–*n*-heptane at 25.01 °C [6]; for acetonitrile–benzene at 45 °C [9]. Mutual solubility data for acetonitrile–(cyclohexane or *n*-hexane or *n*-heptane or *n*-octane) have been previously measured [10,11].

EXPERIMENTAL

Materials

C.P. ethanol and 1-propanol were subjected to fractional distillation twice after drying over calcium oxide. Guaranteed reagent acetonitrile, cyclohexane, *n*-hexane, *n*-heptane and *n*-octane were used without further purification. The densities of the chemicals used for experimental work, measured with an Anton Paar densimeter (DMA40) at 25 °C, were very closely comparable with literature values [12].

Apparatus

VLE data of acetonitrile–1-propanol were determined at 45 °C by the use of a Boublik recirculation still described previously [13]. The liquid and condensed vapour compositions were determined by refractive index measurements using a Shimadzu Pulfrich refractometer at 25 °C and a calibration chart prepared from mixtures of known composition. The experimental errors involved in the observed VLE data were estimated to be 0.002 mole fraction for each phase composition, 0.02 kPa for pressure and 0.05 °C for temperature. LLE measurements were the same as those described previously [1]. The composition of each conjugate liquid phase was determined using a Shimadzu gas chromatograph (GC-8C) and a Shimadzu chromatopac (C-E1B). The observed liquid mole fractions were believed to have an error of ±0.002.

RESULTS AND DISCUSSION

Table 1 presents isothermal VLE data together with the values of the liquid-phase activity coefficients γ and the vapour-phase fugacity coefficients ϕ calculated according to the following relations:

$$\gamma_I = P\phi_I y_I / \left\{ x_I P_I^{\mathrm{s}} \phi_I^{\mathrm{s}} \exp\left[v_I^{\mathrm{L}} (P - P_I^{\mathrm{s}}) / RT \right] \right\} \tag{1}$$

$$\ln \phi_I = \left(2 \sum_J y_J B_{IJ} - \sum_I \sum_J y_I y_J B_{IJ} \right) \frac{P}{RT} \tag{2}$$

where P, x_I and y_I are the experimental values of pressure, liquid-phase mole fraction and vapour-phase mole fraction respectively. The second virial

TABLE 1

Vapour–liquid equilibrium data for acetonitrile(1)–1-propanol(2) at 45 °C [a]

x_1	y_1	P (kPa)	γ_1	γ_2	ϕ_1	ϕ_2
0	0	9.266				
0.065	0.416	15.225	3.597	1.023	0.977	0.991
0.109	0.521	17.825	3.129	1.031	0.972	0.990
0.205	0.629	21.345	2.389	1.071	0.966	0.990
0.320	0.681	23.278	1.800	1.174	0.962	0.990
0.537	0.750	25.758	1.301	1.495	0.958	0.990
0.619	0.765	26.744	1.193	1.773	0.956	0.990
0.749	0.814	27.851	1.090	2.220	0.954	0.991
0.802	0.838	27.851	1.048	2.453	0.954	0.991
0.970	0.958	28.158	1.001	4.257	0.953	0.995
1	1	27.771				

[a] $B_{11} = -4550$ cm^3 mol^{-1}, $B_{22} = -1858$ cm^3 mol^{-1}, $B_{12} = -2462$ cm^3 mol^{-1}, $v_1^L = 53.75$ cm^3 mol^{-1}, $v_2^L = 73.05$ cm^3 mol^{-1}.

coefficients B_{IJ} were calculated by the Hayden–O'Connell correlation [14]. P_I^s and v_I^L are the measured vapour pressure of the pure Ith component and the pure liquid molar volume calculated from the modified Rackett equation [15], respectively. Tables 2 and 3 give experimental tie-line results.

The activity coefficients of components in a ternary mixture of alcohol A with two nonassociating components B and C are given according to the UNIQUAC associated-solution model:

$$\ln \gamma_A = \ln\left(\frac{\Phi_{A_1}}{\Phi_{A_1}^0 x_A}\right) + r_A\left(\frac{1}{V_A^0} - \frac{1}{V}\right) - \left(\frac{Z}{2}\right) q_A\left[\ln\left(\frac{\Phi_A}{\theta_A}\right) + 1 - \frac{\Phi_A}{\theta_A}\right]$$

$$+ q_A\left[1 - \ln\left(\sum_J \theta_J \tau_{JA}\right) - \sum_J \frac{\theta_J \tau_{AJ}}{\sum_K \theta_K \tau_{KJ}}\right] \qquad (3)$$

$$\ln \gamma_B = \ln\left(\frac{\Phi_{B_1}}{x_B}\right) + 1 - \frac{r_B}{V} - \left(\frac{Z}{2}\right) q_B\left[\ln\left(\frac{\Phi_B}{\theta_B}\right) + 1 - \frac{\Phi_B}{\theta_B}\right]$$

$$+ q_B\left[1 - \ln\left(\sum_J \theta_J \tau_{JB}\right) - \sum_J \frac{\theta_J \tau_{BJ}}{\sum_K \theta_K \tau_{KJ}}\right] \qquad (4)$$

$\ln \gamma_C$ is given by changing the subscript B in eqn. (4) to C. The model assumes the linear polymerization of alcohol molecules ($A_i + A_1 = A_{i+1}$) and solvation equilibria between the terminal hydroxyl groups of alcohol chains and solvating molecules ($A_i + B_1 = A_iB$ and $A_i + C_1 = A_iC$). The

TABLE 2

Tie-line data for acetonitrile–ethanol–saturated hydrocarbon at 25 ° C

Phase I			Phase II		
x_1	x_2	x_3	x_1	x_2	x_3
Acetonitrile(1)–ethanol(2)–*n*-hexane(3)					
0.9433	0	0.0567	0.0584	0	0.9416 [a]
0.8948	0.0453	0.0599	0.0603	0.0055	0.9342
0.8503	0.0848	0.0649	0.0576	0.0102	0.9322
0.8059	0.1247	0.0694	0.0606	0.0174	0.9220
0.7594	0.1632	0.0774	0.0638	0.0270	0.9092
0.7125	0.2015	0.0860	0.0641	0.0364	0.8995
0.6706	0.2316	0.0978	0.0717	0.0484	0.8799
0.6278	0.2653	0.1069	0.0762	0.0629	0.8609
0.5835	0.2936	0.1229	0.0867	0.0798	0.8335
0.5446	0.3192	0.1362	0.0979	0.1008	0.8013
0.4969	0.3447	0.1584	0.1017	0.1199	0.7784
0.4702	0.3555	0.1743	0.1066	0.1314	0.7620
0.4225	0.3703	0.2072	0.1203	0.1603	0.7194
0.3389	0.3738	0.2873	0.1665	0.2349	0.5986
Acetonitrile(1)–ethanol(2)–*n*-heptane(3)					
0.9622	0	0.0378	0.0621	0	0.9379 [a]
0.8066	0.1411	0.0523	0.0656	0.0213	0.9131
0.6896	0.2442	0.0662	0.0743	0.0455	0.8802
0.5950	0.3192	0.0858	0.0798	0.0758	0.8444
0.5236	0.3742	0.1022	0.0846	0.1052	0.8102
0.4431	0.4218	0.1351	0.0966	0.1477	0.7557
0.3539	0.4592	0.1869	0.1131	0.2085	0.6784
0.2628	0.4295	0.3077	0.1724	0.3212	0.5064
0.2415	0.4083	0.3502	0.1831	0.3405	0.4764
Acetonitrile(1)–ethanol(2)–*n*-octane(3)					
0.9801	0	0.0199	0.0568	0	0.9432 [a]
0.7796	0.1832	0.0372	0.0644	0.0274	0.9082
0.6450	0.3039	0.0511	0.0711	0.0623	0.8666
0.5428	0.3894	0.0678	0.0767	0.0958	0.8275
0.4715	0.4438	0.0847	0.0791	0.1239	0.7970
0.3720	0.5061	0.1219	0.0874	0.1783	0.7343
0.2723	0.5343	0.1934	0.1060	0.2721	0.6219
0.2010	0.4994	0.2996	0.1472	0.3968	0.4560

[a] Taken from ref. 11.

self-association constant K_A and the solvation constants K_{AB} and K_{AC} are defined by

$$K_A = \left(\Phi_{A_{i+1}}/\Phi_{A_i}\Phi_{A_1}\right)\left[i/(i+1)\right] \quad (5)$$

$$K_{AB} = \left(\Phi_{A_iB}/\Phi_{A_i}\Phi_{B_1}\right)\left[i/(ir_A + r_B)\right] \quad (6)$$

$$K_{AC} = \left(\Phi_{A_iC}/\Phi_{A_i}\Phi_{C_1}\right)\left[i/(ir_A + r_C)\right] \quad (7)$$

TABLE 3

Tie-line data for acetonitrile–1-propanol–saturated hydrocarbon at 25 °C

Phase I			Phase II		
x_1	x_2	x_3	x_1	x_2	x_3
Acetonitrile(1)–1-propanol(2)–cyclohexane(3)					
0.9399	0	0.0601	0.0440	0	0.9560 [a]
0.8519	0.0556	0.0925	0.0546	0.0103	0.9351
0.7859	0.1007	0.1134	0.0705	0.0233	0.9062
0.7521	0.1221	0.1258	0.0717	0.0322	0.8961
0.7100	0.1490	0.1410	0.0845	0.0477	0.8678
0.6633	0.1732	0.1635	0.0960	0.0652	0.8388
0.6189	0.1963	0.1848	0.1084	0.0852	0.8064
0.5684	0.2092	0.2224	0.1332	0.1078	0.7590
0.5215	0.2238	0.2547	0.1562	0.1316	0.7122
Acetonitrile(1)–1-propanol(2)–*n*-hexane(3)					
0.9433	0	0.0567	0.0584	0	0.9416 [b]
0.8411	0.0805	0.0784	0.0797	0.0192	0.9011
0.8046	0.1081	0.0873	0.0878	0.0315	0.8807
0.7530	0.1446	0.1024	0.0956	0.0492	0.8552
0.6540	0.2058	0.1402	0.1171	0.0915	0.7914
0.6019	0.2244	0.1737	0.1541	0.1214	0.7245
0.5398	0.2439	0.2163	0.1719	0.1485	0.6796
0.5045	0.2482	0.2473	0.2081	0.1734	0.6185
0.4684	0.2537	0.2779	0.2488	0.1918	0.5594
Acetonitrile(1)–1-propanol(2)–*n*-heptane(3)					
0.9622	0	0.0378	0.0621	0	0.9379 [b]
0.8872	0.0640	0.0488	0.0706	0.0137	0.9157
0.7789	0.1509	0.0702	0.0910	0.0474	0.8616
0.7222	0.1902	0.0876	0.0945	0.0738	0.8317
0.6597	0.2318	0.1085	0.1171	0.1037	0.7792
0.6154	0.2564	0.1282	0.1377	0.1349	0.7274
0.5767	0.2706	0.1527	0.1624	0.1595	0.6781
0.5256	0.2897	0.1847	0.1922	0.1931	0.6147
0.4424	0.2975	0.2601	0.2576	0.2405	0.5019

[a] Taken from ref. 10.
[b] Taken from ref. 11.

In eqns. (3) and (4), Z is the coordination number set as 10, Φ_I and θ_I are the nominal segment fraction and the nominal area fraction respectively expressed in terms of molecular geometric structural parameters r and q, and τ_{JI} is related to the energy parameter a_{JI}:

$$\Phi_I = r_I x_I / \sum_J r_J x_J \tag{8}$$

$$\theta_I = q_I x_I / \sum_J q_J x_J \tag{9}$$

$$\tau_{JI} = \exp(-a_{JI}/T) \tag{10}$$

The monomer segment fractions, Φ_{A_1}, Φ_{B_1} and Φ_{C_1}, are simultaneously solved from the following mass balance equations:

$$\Phi_A = \frac{\Phi_{A_1}}{\left(1 - K_A\Phi_{A_1}\right)^2}\left[1 + r_A\left(K_{AB}\Phi_{B_1} + K_{AC}\Phi_{C_1}\right)\right] \tag{11}$$

$$\Phi_B = \Phi_{B_1}\left[1 + \frac{r_B K_{AB}\Phi_{A_1}}{\left(1 - K_A\phi_{A_1}\right)}\right] \tag{12}$$

$$\Phi_C = \Phi_{C_1}\left[1 + \frac{r_C K_{AC}\Phi_{A_1}}{\left(1 - K_A\Phi_{A_1}\right)}\right] \tag{13}$$

The true molar volume of the ternary mixture V, the true molar volume of pure alcohol solution V_A° and the monomer segment fraction of pure alcohol $\Phi_{A_1}^\circ$ are expressed by

$$\frac{1}{V} = \frac{\Phi_{A_1}}{r_A\left(1 - K_A\Phi_{A_1}\right)} + \frac{\Phi_{B_1}}{r_B}\left[1 + \frac{K_{AB}r_B\Phi_{A_1}}{\left(1 - K_A\Phi_{A_1}\right)}\right] + \frac{\Phi_{C_1}}{r_C}\left[1 + \frac{K_{AC}r_C\Phi_{A_1}}{\left(1 - K_A\Phi_{A_1}\right)}\right] \tag{14}$$

$$\frac{1}{V_A^0} = \frac{\left(1 - K_A\Phi_{A_1}^0\right)}{r_A} \tag{15}$$

$$\Phi_{A_1}^0 = \left[2K_A + 1 - \left(1 + 4K_A\right)^{1/2}\right]/2K_A^2 \tag{16}$$

An optimum set of the energy parameters was determined from the VLE data using a computer program, based on the maximum likelihood principle, as described by Prausnitz et al. [16]. The standard deviations in the measured variables were taken as $\sigma_P = 0.133$ kPa, $\sigma_T = 0.05$ K, $\sigma_x = 0.001$ and $\sigma_y = 0.003$. The molecular geometric structural parameters of the pure components were calculated according to the method of Vera et al. [17] and are

TABLE 4

Molecular structural parameters of pure components

Component	r	q
Ethanol	1.69	1.55
1-Propanol	2.23	1.98
Acetonitrile	1.50	1.40
Benzene	2.56	2.05
Cyclohexane	3.18	2.55
n-Heptane	4.15	3.52
n-Hexane	3.61	3.09
n-Octane	4.69	3.95

given in Table 4. The association constant of the alcohol at 50 °C was taken from Brandani [18]: for ethanol, $K_A = 110.4$ and for 1-propanol, $K_A = 87.0$. The enthalpy of hydrogen-bond formation h_A is set as -23.2 kJ mol^{-1}, which is the enthalpy of dilution of ethanol in *n*-hexane at 25 °C [19]. The following values of the solvation constant and the enthalpy of complex formation are used: for ethanol–acetonitrile, $K_{AB} = 50$ at 40 °C and $h_{AB} = -17$ kJ mol^{-1} [2]; for 1-propanol–acetonitrile, $K_{AB} = 35$ at 50 °C and $h_{AB} = -17$ kJ mol^{-1}; for 1-propanol–benzene, $K_{AB} = 2.5$ at 50 °C and $h_{AB} = -8.3$ kJ mol^{-1} [2]. h_A and h_{AB} were assumed to be independent of temperature and the temperature dependence of the equilibrium constants

TABLE 5

Results obtained in fitting the UNIQUAC associated-solution model to vapour–liquid equilibria and liquid–liquid equilibria for binary mixtures

Mixture	Temp. (°C)	Number of data points	Root-mean-square deviations				Parameters	
			δP (kPa)	δT (K)	δx ($\times 10^3$)	δy ($\times 10^3$)	a_{AB} (K)	a_{BA} (K)
Ethanol(A)–acetonitrile(B)	20	13	0.048	0.00	0.0		544.89	65.26
Ethanol(A)–*n*-hexane(B)	25	25	0.112	0.00	0.2		165.83	−74.30
Ethanol(A)–*n*-heptane(B)	30	31	0.077	0.00	0.2		66.97	−2.41
Ethanol(A)–*n*-octane(B)	45	17	0.092	0.01	1.8	2.3	38.15	23.03
1-Propanol(A)–acetonitrile(B)	45	9	0.200	0.00	0.9	6.8	636.27	72.99
1-Propanol(A)–benzene(B)	45	11	0.019	0.00	0.1	1.4	114.53	−8.23
1-Propanol(A)–cyclohexane(B)	25	27	0.096	0.00	0.1		250.39	−130.94
1-Propanol(A)–*n*-hexane(B)	25	25	0.147	0.00	0.1		249.75	−125.72
1-Propanol(A)–*n*-heptane(B)	25.01	14	0.025	0.00	0.0		102.72	−46.89
Acetonitrile(A)–benzene(B)	45	12	0.104	0.02	0.5	3.6	−10.54	258.38
Acetonitrile(A)–cyclohexane(B)	25	MS [a]					142.38	689.58
Acetonitrile(A)–*n*-hexane(B)	25	MS					100.74	692.54
Acetonitrile(A)–*n*-heptane(B)	25	MS					99.25	692.34
Acetonitrile(A)–*n*-octane(B)	25	MS					113.20	704.95

[a] MS, mutual solubilities.

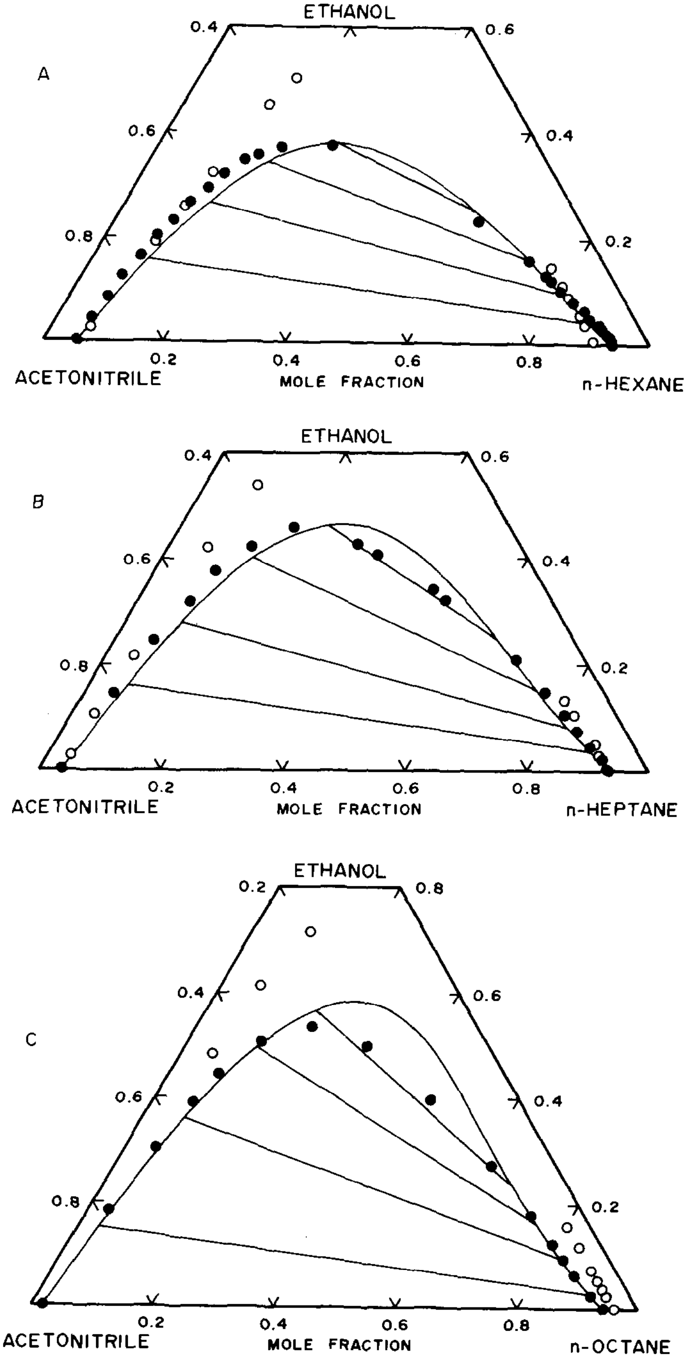

Fig. 1. Experimental tie-lines and binodal curves predicted from the UNIQUAC associated-solution model at 25 °C (A, acetonitrile–ethanol–*n*-hexane; B, acetonitrile–ethanol–*n*-heptane; C, acetonitrile–ethanol–*n*-octane): ○, experimental results of Kikic et al. [5]; ●, experimental results of this work; ——, calculated values.

are defined by the van't Hoff relation. The pure component vapour pressures used in VLE data reduction were obtained from the Antoine equation [20,21], except for 1-propanol–acetonitrile. Table 5 shows calculated results for binary mixtures. The results of VLE data reduction for 1-propanol–hydrocarbon and acetonitrile–benzene were taken from a previous paper [2] and those of LLE data reduction for acetonitrile–saturated hydrocarbon were also cited from a previous paper [11].

The LLE calculations are based on the thermodynamic criterion of equilibrium that the activity of each component is the same in two phases:

$$(x_I\gamma_I)^{\mathrm{I}} = (x_I\gamma_I)^{\mathrm{II}} \tag{17}$$

where I and II indicate equilibrium phases.

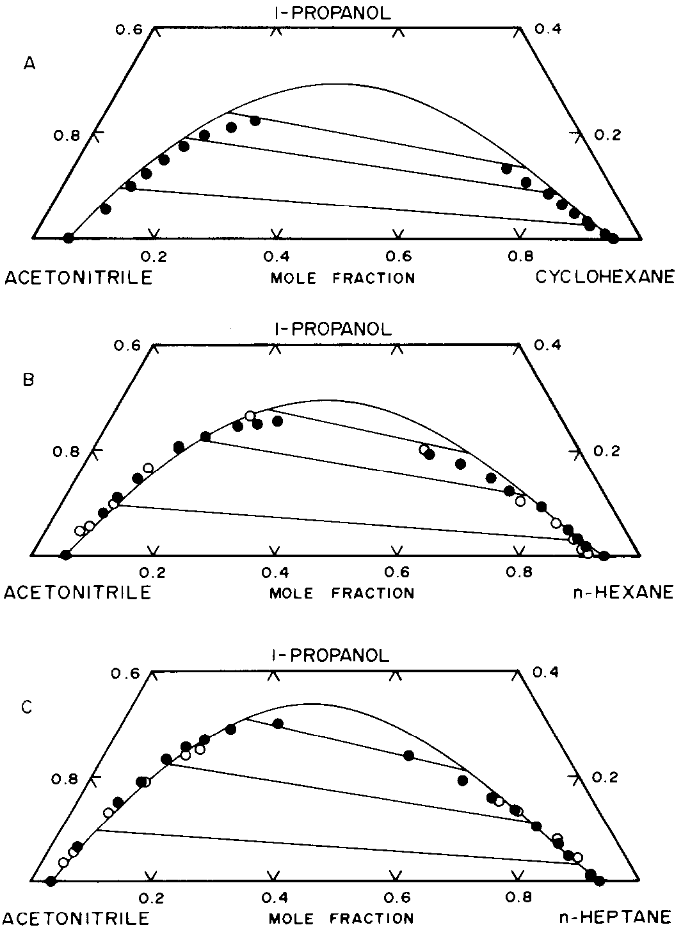

Fig. 2. Experimental tie-lines and binodal curves predicted from the UNIQUAC associated-solution model at 25 °C (A, acetonitrile–1-propanol–cyclohexane; B, acetonitrile–1-propanol–*n*-hexane; C, acetonitrile–1-propanol–*n*-heptane): ○, experimental results of Kikic et al. [22]; ●, experimental results of this work; ———, calculated values.

TABLE 6

Ternary calculated results of vapour–liquid equilibrium for 1-propanol(1)–acetonitrile(2)–benzene(3) at 45 °C

	Vapour mole fractions ($\times 10^3$)			Pressure	
	δy_1	δy_2	δy_3	δP (kPa)	$\delta P/P$ (%)
Mean	3.6	4.7	5.5	0.3805	1.19 [a]
	2.8	4.8	4.6	0.3452	1.11 [b]
Root-mean-square deviation	4.6	5.7	6.7	0.4490	1.43 [a]
	3.6	6.1	5.5	0.4179	1.37 [b]

[a] Taken from ref. 23; $K_{12} = 30$ at 50 °C for 1-propanol–acetonitrile.

[b] This work; $K_{12} = 35$ at 50 °C for 1-propanol–acetonitrile.

Figures 1 and 2 compare the experimental results for the ternary acetonitrile–ethanol–saturated hydrocarbon mixtures and for the ternary acetonitrile–1-propanol–saturated hydrocarbon mixtures with calculated values obtained by use of the binary parameters alone. For acetonitrile–ethanol–saturated hydrocarbon the experimental data of Kikic et al. [5] show much larger solubility envelopes than the present data. For acetonitrile–1-propanol–saturated hydrocarbon the present data agree rather well with the data of Kikic et al. [22]. The calculated values predict the present experimental data with reasonable accuracy, although perfect agreement is not obtained. For 1-propanol–acetonitrile, $K_{AB} = 35$ is used at 50 °C instead of $K_{AB} = 30$ at 50 °C [23] in order to obtain better calculated results of ternary LLE. Moreover, this change in K_{AB} gives slightly improved results of ternary VLE prediction as shown in Table 6.

LIST OF SYMBOLS

A, B, C	alcohols and active nonassociating components
a_{IJ}	binary interaction parameter
B_{IJ}	second virial coefficient
h_A	enthalpy of hydrogen-bond formation
h_{AB}	enthalpy of complex formation between unlike molecules
K_A	self-association constant of alcohol
K_{AB}, K_{AC}	solvation constants between unlike molecules
P	total pressure
P_I^S	vapour pressure of pure component I
q_I	molecular geometric area parameter of pure component I
R	universal gas constant
r_I	molecular geometric size parameter of pure component I
T	absolute temperature

V	true molar volume of alcohol mixture
V_A^0	true molar volume of pure alcohol liquid
v_I^L	pure liquid molar volume of component I
x_I	liquid-phase mole fraction of component I
y_I	vapour-phase mole fraction of component I
Z	coordination number, equal to 10

Greek letters

γ_I	activity coefficient of component I
θ_I	area fraction of component I
σ_P, σ_T	standard deviations in pressure and temperature
σ_x, σ_y	standard deviations in liquid-phase and vapour-phase mole fractions
τ_{IJ}	coefficient as defined by $\exp(-a_{IJ}/T)$
Φ_I	segment fraction of component I
ϕ_I	fugacity coefficient of component I at P and T
ϕ_I^s	fugacity coefficient of pure component I at P_I^s and T

Superscripts

0	pure liquid reference state
L	liquid
s	saturation

Subscripts

A, B, C	alcohol and active nonassociating components
A_1, A_i	monomer and *i*-mer of alcohol
A_iB, A_iC	complex formation between unlike molecules
AB, AC	complexes formed by A and B or C
B_1, C_1	monomers of active nonassociating components
I, *J*, *K*	components

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