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THERMODYNAMICS OF SOLUTIONS OF ACETONITRILE WITH PROPANOLS

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ABSTRACT

The thermodynamic properties of solutions containing acetonitrile and one of the propanols have been correlated with an extended model which assumes the self-association of acetonitrile and the propanol and complex formation between acetonitrile and the propanol, with an allowance for non-polar interactions. Vapor–liquid and liquid–liquid equilibrium and excess enthalpy data for ternary mixtures including acetonitrile, the propanol, and a non-associating component have been successfully predicted by use of the model with only the parameters obtained from constituent binary mixtures.

INTRODUCTION

The association of acetonitrile and simple aliphatic alcohols in non-associating components has been studied in the accurate reproduction of experimental isothermal vapor–liquid equilibrium and excess enthalpy results over the whole range of mole fraction by use of the chemical theory of associated solutions [1–4]. The infrared spectroscopic changes in the dilute region of the alcohol are well reproduced with the association model [3,4].

We [5] extended the model to correlate the isothermal vapor–liquid equilibrium and excess enthalpy data of acetonitrile–methanol and acetonitrile–ethanol mixtures and to predict phase equilibria and excess enthalpies for ternary mixtures containing acetonitrile, methanol or ethanol, and a non-associating component. In this paper the extended model is applied to the phase equilibrium and excess enthalpy data of binary and ternary mixtures of acetonitrile with 1-propanol or 2-propanol and a non-associating component.

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ASSOCIATION MODEL

A stands for the alcohol, B for acetonitrile, and C for a non-associating component. The alcohol association model [2–4] assumes that open chains of any length are present according to three chemical equilibria, K_2 for $A_1 + A_1 = A_2$, K_3 for $A_2 + A_1 = A_3$, and K for $A_i + A_1 = A_{i+1}$ $(i \geqslant 3)$, and that cyclic groups are in equilibrium with the open chains by $K_{cy} = \theta/i$ $(i > 4)$ for A_i(linear) = A_i(cyclic), and includes a solvation constant K_{AC} for $A_i + C_1 = A_iC$. Similarly the acetonitrile association model uses the two self-association constants of acetonitrile molecules, K'_B for $B_1 + B_1 = B_2$ and K_B for $B_i + B_1 = B_{i+1}$, and one solvation constant K_{BC} for $B_1 + C_1 = BC$ [1]. The extended model [5] includes the equilibrium constants described above plus two solvation constants between the propanol and acetonitrile: K_{A_iB} for $A_i + B_1 = A_iB$ and $K_{A_iB_j}$ for $A_i + B_j = A_iB_j (i \geqslant 1,\ j \geqslant 2)$.

The activity coefficient of any component I in a ternary mixture containing the propanol, acetonitrile, and a non-associating component is given by

$$\ln \gamma_I = \ln\left(\frac{x_{I_1}}{x^*_{I_1} x_I}\right) + \frac{\sum_J \tau_{JI} G_{JI} x_J}{\sum_K G_{KI} x_K} + \sum_J \frac{x_J G_{IJ}}{\sum_K G_{KJ} x_K}\left(\tau_{IJ} - \frac{\sum_R x_R \tau_{RJ} G_{RJ}}{\sum_K G_{KJ} x_K}\right) \tag{1}$$

where $x^*_{C_1} = 1$ for the non-associating component, τ_{JI} and G_{JI} are defined by

$$\tau_{JI} = a_{JI}/T \tag{2}$$

$$G_{JI} = \exp[-\alpha_{JI}\tau_{JI}] \tag{3}$$

and the nonrandomness parameter α_{JI} $(= \alpha_{IJ})$ is taken as 0.3.

The stoichiometric mole fractions of components are expressed in terms of the monomer mole fractions and the equilibrium constants.

$$x_A = \left\{\left[1 + K_{A_iB} x_{B_1} + K_{A_iC} x_{C_1} + \frac{K_{A_iB_j} x_{B_1} w}{(1-w)}\right] \times \left[x_{A_1} + 2K_2 x_{A_1}^2 + \frac{K_2 K_3 x_{A_1}^3 (3-2z)}{(1-z)^2}\right] + \frac{K_2 K_3 K^2 \theta x_{A_1}^5}{(1-z)}\right\}/S \tag{4}$$

$$x_B = \left\{\left[K_{A_iB} x_{B_1} + \frac{K_{A_iB_j} x_{B_1} w(2-w)}{(1-w)^2}\right]\left[x_{A_1} + K_2 x_{A_1}^2 + \frac{K_2 K_3 x_{A_1}^3}{(1-z)}\right] + \frac{x_{B_1}}{(1-w)^2} + 2K'_B x_{B_1}^2 + K_{BC} x_{B_1} x_{C_1}\right\}/S \tag{5}$$

$$x_C = \left\{K_{A_iC} x_{C_1}\left[x_{A_1} + K_2 x_{A_1}^2 + \frac{K_2 K_3 x_{A_1}^3}{(1-z)}\right] + K_{BC} x_{B_1} x_{C_1} + x_{C_1}\right\}/S \tag{6}$$

where $w = K_B x_{B_1}$, $z = K x_{A_1}$, and the stoichiometric sum S is given by

$$S = \left[1 + K_{A_iB} x_{B_1} + K_{A_iC} x_{C_1} + \frac{K_{A_iB_j} x_{B_1} w}{(1-w)}\right]$$
$$\times \left[x_{A_1} + 2K_2 x_{A_1}^2 + \frac{K_2 K_3 x_{A_1}^3 (3-2z)}{(1-z)^2}\right]$$
$$+ \frac{K_2 K_3 K^2 \theta x_{A_1}^5}{(1-z)} + \left[K_{A_iB} x_{B_1} + K_{A_iC} x_{C_1} + \frac{K_{A_iB_j} x_{B_1} w(2-w)}{(1-w)^2}\right]$$
$$\times \left[x_{A_1} + K_2 x_{A_1}^2 + \frac{K_2 K_3 x_{A_1}^3}{(1-z)}\right] + \frac{x_{B_1}}{(1-w)^2} + 2K'_B x_{B_1}^2 + 2K_{BC} x_{B_1} x_{C_1} + x_{C_1} \tag{7}$$

The sum of the mole fractions of all species present in the mixture is unity.

$$\left[1 + K_{A_iB} x_{B_1} + K_{A_iC} x_{C_1} + \frac{K_{A_iB_j} x_{B_1} w}{(1-w)}\right]\left[x_{A_1} + K_2 x_{A_1}^2 + \frac{K_2 K_3 x_{A_1}^3}{(1-z)}\right]$$
$$- \frac{K_2 K_3 \theta}{K^3}\left[\ln(1-z) + z + \frac{z^2}{2} + \frac{z^3}{3} + \frac{z^4}{4}\right]$$
$$+ \frac{x_{B_1}}{(1-w)} + K'_B x_{B_1}^2 + K_{BC} x_{B_1} x_{C_1} + x_{C_1} = 1 \tag{8}$$

Ternary excess enthalpy is given by

$$H^E = H^E_{\text{chem}} + H^E_{\text{phys}} \tag{9}$$

where H^E_{chem} and H^E_{phys} are expressed by eqns. (10) and (13).

$$H^E_{\text{chem}} = \Bigg\{\left[1 + K_{A_iB} x_{B_1} + K_{A_iC} x_{C_1} + \frac{K_{A_iB_j} x_{B_1} w}{(1-w)}\right]$$
$$\times \left[h_2 K_2 x_{A_1}^2 + \frac{h_A K_2 K_3 x_{A_1}^3 (2-z)}{(1-z)^2}\right]$$
$$+ \frac{h_A K_2 K_3 K^2 \theta x_{A_1}^5}{(1-z)} + \left(1 + K_{A_iB_j}\left[x_{A_1} + K_2 x_{A_1}^2 + \frac{K_2 K_3 x_{A_1}^3}{(1-z)}\right]\right)\frac{h_B x_{B_1} w}{(1-w)^2}$$
$$+ h'_B K'_B x_{B_1}^2 + \left[x_{A_1} + K_2 x_{A_1}^2 + \frac{K_2 K_3 x_{A_1}^3}{(1-z)}\right]$$

$$\times\left[h_{A_iB}K_{A_iB}x_{B_1}+h_{A_iC}K_{A_iC}x_{C_1}+\frac{h_{A_iB_j}K_{A_iB_j}x_{B_1}w}{(1-w)}\right]$$

$$\left.+h_{BC}K_{BC}x_{B_1}x_{C_1}\right\}/S$$

$$-x_A\left[h_2K_2x_{A_1}^{*2}+\frac{h_AK_2K_3x_{A_1}^{*3}(2-z^*)}{(1-z^*)^2}+\frac{h_AK_2K_3K^2\theta x_A^{*5}}{(1-z^*)}\right]/S_A^*$$

$$-x_B\left[h'_BK'_Bx_{B_1}^{*2}+\frac{h_Bx_{B_1}^*w^*}{(1-w^*)^2}\right]/S_B^* \tag{10}$$

where $w^*=K_Bx_{B_1}^*$, $z^*=Kx_{A_1}^*$, the superscript * denotes pure liquid state and the stoichiometric sums S_A^* and S_B^* are defined by

$$S_A^*=x_{A_1}^*+2K_2x_{A_1}^{*2}+\frac{K_2K_3x_{A_1}^{*3}(3-2z^*)}{(1-z^*)^2}+\frac{K_2K_3K^2\theta x_{A_1}^{*5}}{(1-z^*)} \tag{11}$$

$$S_B^*=2K'_Bx_{B_1}^{*2}+\frac{x_{B_1}^*}{(1-w^*)^2} \tag{12}$$

$$H_{\text{phys}}^{E}=R\sum_I x_I\left[\frac{\sum_J x_J\frac{\partial(\tau_{JI}G_{JI})}{\partial(1/T)}}{\sum_K G_{KI}x_K}-\frac{\sum_J \tau_{JI}G_{JI}x_J\sum_K x_K\frac{\partial G_{KI}}{\partial(1/T)}}{\left(\sum_K G_{KI}x_K\right)^2}\right] \tag{13}$$

We assume the linear temperature dependence of a_{JI} given by

$$a_{JI}=C_I+D_I(T-273.15) \tag{14}$$

We can obtain x_{A_1}, x_{B_1}, and x_{C_1} by simultaneous solution of two equations out of eqns. (4)–(6) and eqn. (8), $x_{A_1}^*$ from eqn. (8) at pure alcohol state, and $x_{B_1}^*$ from eqn. (8) at pure acetonitrile state.

RESULTS

Binary data reduction

Vapor–liquid and liquid–liquid equilibrium and excess enthalpy data for binaries constituting ternary mixtures containing the propanol and acetonitrile were reduced with the association model.

The vapor–liquid equilibrium relation in common use is

$$\phi_Iy_IP=x_I\gamma_I\phi_I^*P_I^*\exp[v_I^{*L}(P-P_I^*)/RT] \tag{15}$$

where y is the vapor-phase mole fraction, P is the total pressure, P^* is the pure-component vapor pressure, and $v^{*\mathrm{L}}$ is the pure-liquid molar volume calculated from a quadratic equation in terms of temperature [5]. The fugacity coefficient ϕ is calculated by

$$\ln \phi_I = \frac{P}{RT}\left(2\sum_J y_J B_{IJ} - \sum_I \sum_J y_I y_J B_{IJ}\right) \tag{16}$$

where B_{IJ} is the second virial coefficient estimated by the correlation of Hayden and O'Connell [6]. The vapor pressures of pure components were obtained from the Antoine equation with parameters from the literature [7–9]. Parameter estimation was performed by minimizing the sum-of-squares of relative deviation in pressure plus the sum-of-squares of deviations in vapor-phase mole fraction using the simplex method [10].

The equation of liquid–liquid equilibrium, for any component I, is

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

where the superscripts I and II refer to the two equilibrium liquid phases. A set of the energy parameters was obtained by solution of eqn. (17).

Fitting the model to excess enthalpy data was made by use of the computer program which minimizes the sum-of-squares of deviation between the experimental value and that calculated with the simplex method.

The thermodynamic parameters of the pure associated components are as follows: for acetonitrile [1], $K'_\mathrm{B} = 8.35$ and $K_\mathrm{B} = 2.1$ at 45°C, $h'_\mathrm{B} = -8.9$ kJ mol^{-1} and $h_\mathrm{B} = -6.7$ kJ mol^{-1}; for 1-propanol [4], $K_2 = 35$, $K_3 = 90$, $K = 40$, and $\theta = 75$ at 25°C; for 2-propanol [4], $K_2 = 35$, $K_3 = 85$, $K = 30$, and $\theta = 70$ at 25°C; $h_2 = -21.2$ kJ mol^{-1} and $h_\mathrm{A} = -23.5$ kJ mol^{-1} for the propanol [4]. The values of the solvation constants and enthalpies of complex formation between unlike molecules are listed in Table 1. All the h values were assumed to be independent of temperature and fix the temperature dependence of the equilibrium constants by the van't Hoff relation. Binary calculated results are presented in Table 2 for phase equilibria and in

TABLE 1

Solvation equilibrium constants and enthalpies of complex formation

System (A–B)	Temp. (°C)	$K_{\mathrm{A}_i\mathrm{B}}$	$K_{\mathrm{A}_i\mathrm{B}_j}$	$-h_{\mathrm{A}_i\mathrm{B}}$ (kJ mol^{-1})	$-h_{\mathrm{A}_i\mathrm{B}_j}$ (kJ mol^{-1})
1-Propanol–acetonitrile	55	40	35	22.0	16.8
2-Propanol–acetonitrile	50	45	40	22.0	16.8
1-Propanol–benzene	25	3.2		8.2	
1-Propanol–chloroform	55	12		24.5	
2-Propanol–benzene	25	2.8		8.2	
Acetonitrile–benzene	45	0.2 [a]		5.2	

[a] 1:1 complex formation is assumed.

TABLE 2

Binary parameters and absolute arithmetic mean deviations as obtained from vapor–liquid equilibrium data reduction

System (A–B)	Temp. (°C)	No. of data points	Parameters		Abs. arith. mean dev.		Ref.
			a_{BA} (K)	a_{AB} (K)	Vapor mole fraction ($\times 10^3$)	Press. (mm Hg)	
1-Propanol–acetonitrile	55	10	−108.35	346.49	5.4	1.2	11
1-Propanol–benzene	45	11	44.57	−17.01	8.5	3.6	12
1-Propanol–chloroform	55	9	734.99	−352.78	5.8	1.4	13
1-Propanol–*n*-heptane	25	11	85.48	3.64		0.4	14
1-Propanol–*n*-hexane	25	25	11.93	78.34		1.4	15
2-Propanol–acetonitrile	50	15	−69.77	360.18	5.2	0.9	16
2-Propanol–benzene	50	15	−103.60	196.91	5.4	1.8	17
2-Propanol–cyclohexane	50	11	270.53	−180.82	3.1	2.4	17
Acetonitrile–benzene	45	11	−187.77	368.93	2.9	1.2	18
	55	12	−193.98	406.74	5.8	1.6	19
Acetonitrile–chloroform	55	11	110.69	−81.71	3.8	3.4	20
Acetonitrile–cyclohexane	50	MS[a]	287.49	434.76			16
Acetonitrile–*n*-heptane	25	MS	279.67	647.27			21
Acetonitrile–*n*-hexane	25	MS	236.38	592.00			21

[a] MS = mutual solubility data.

Table 3 for excess enthalpies. Figures 1–3 illustrate some representative examples to demonstrate the accuracy of fit of the model to the experimental data.

Ternary predictions based on binary parameters

Table 4 lists predicted vapor–liquid equilibria for three ternary mixtures. Figure 4 compares calculated liquid–liquid equilibria with experimental data for three ternary mixtures where two binaries are completely miscible and

TABLE 3

Binary parameters and absolute arithmetic mean deviations as obtained from excess enthalpy data reduction at 25°C

System (A–B)	No. of data points	Parameters				Abs. arith. mean dev. (J mol^{-1})	Ref.
		C_A (K)	C_B (K)	D_A	D_B		
1-Propanol–acetonitrile	22	−222.32	65.49	−0.9056	−0.8345	8.2	22
1-Propanol–benzene	10	−1135.43	594.67	−3.8897	2.0141	7.9	23
2-Propanol–acetonitrile	19	−627.06	−258.28	−2.7189	−1.5730	8.0	22
2-Propanol–benzene	17	−1097.98	485.01	−3.6946	0.7566	7.0	24
Acetonitrile–benzene	15	399.37	−21.88	2.6005	−0.9393	1.3	25

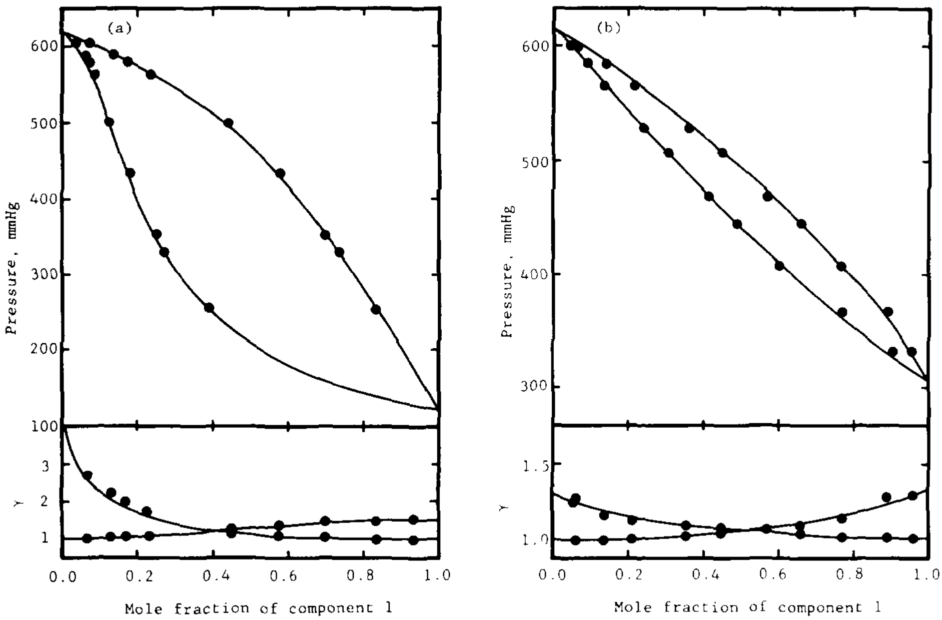

Fig. 1. Vapor–liquid equilibria for (a) 1-propanol(1)–chloroform(2) and (b) acetonitrile(1)–chloroform(2) at 55°C. Calculated (———). Experimental (●): (a), data of Nagata [13]; (b), data of Nagata and Kawamura [20].

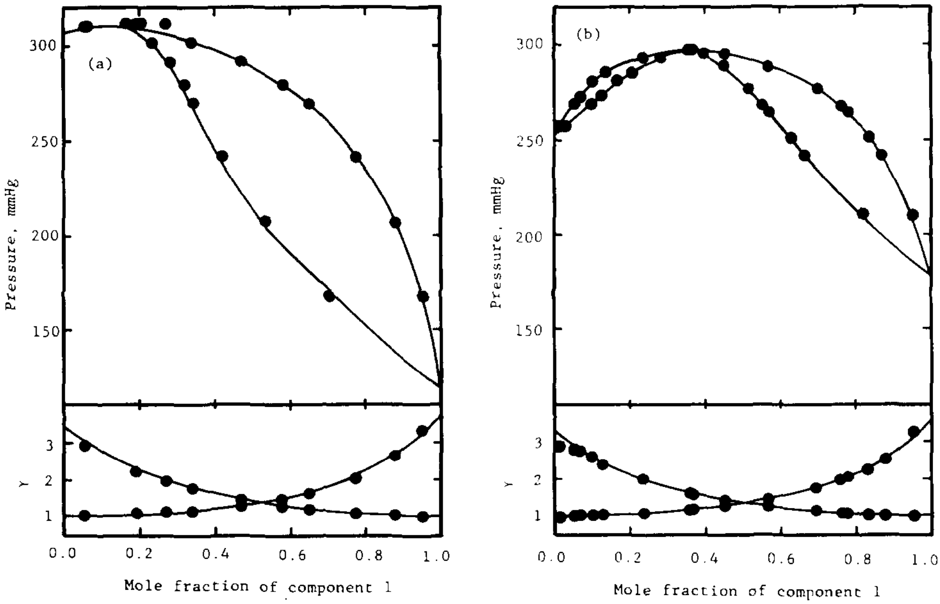

Fig. 2. Vapor–liquid equilibria for (a) 1-propanol(1)–acetonitrile(2) at 55°C and (b) 2-propanol(1)–acetonitrile(2) at 50°C. Calculated (———). Experimental (●): (a), data of Ohta et al. [11]; (b), data of Nagata and Katoh [16].

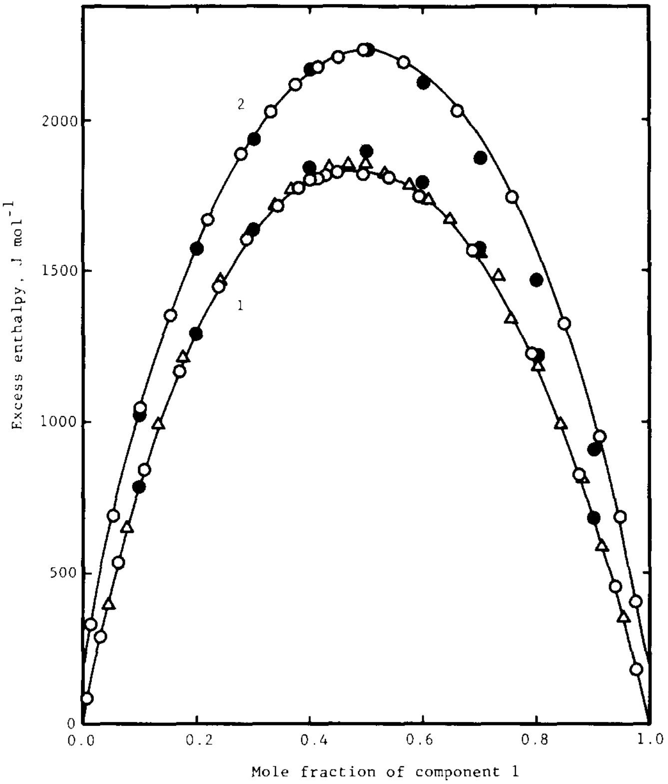

Fig. 3. Molar excess enthalpies for two binary systems at 25°C. Calculated (———). Experimental: (1) 1-propanol(1)–acetonitrile(2), data of Nagata and Tamura [22] (○), data of Dohnal et al. [28] (△), data of Mato and Coca [29] (●); (2) 2-propanol(1)–acetonitrile(2), data of Nagata and Tamura [22] (○), data of Mato and Coca [29] (●). Note that the ordinate for 2-propanol–acetonitrile is replaced upwards by 200 J mol^{-1} to avoid overlap.

TABLE 4

Predicted results for ternary vapor–liquid equilibria

System	Temp. (°C)	No. of data points	Abs. arith. mean dev.		Ref.
			Vapor mole fraction ($\times10^3$)	Press. (mm Hg)	
1-Propanol–acetonitrile–chloroform	55	19	4.7 8.1 9.9	3.1	13
1-Propanol–acetonitrile–benzene	45	27	6.7 5.2 9.1	3.6	26
2-Propanol–acetonitrile–benzene	50	23	6.4 7.9 6.4	7.6	27

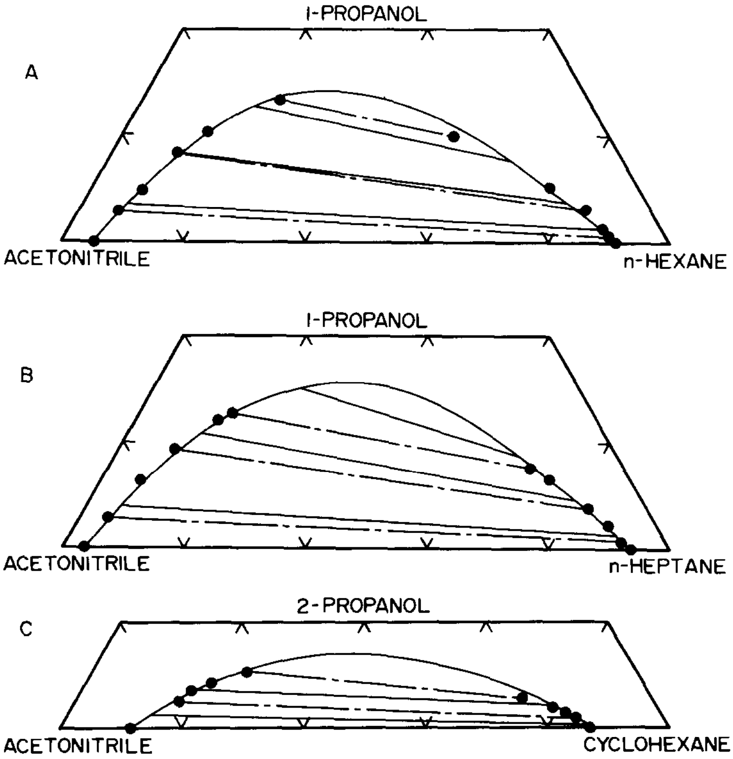

Fig. 4. Ternary liquid–liquid equilibria. Calculated (———). Experimental tie-line (●– · – ●): (A) acetonitrile–1-propanol–*n*-hexane at 25°C, data of Kikic et al. [30]; (B) acetonitrile–1-propanol–*n*-heptane at 25°C, data of Kikic et al. [30]; (C) acetonitrile–2-propanol–cyclohexane at 50°C, data of Nagata and Katoh [16].

only one binary is partially miscible, indicating that the agreement is good except near the plait point. Predicted results for ternary excess enthalpies are given in Table 5. These results show that the model is able to predict with good accuracy the thermodynamic properties of acetonitrile, one of the propanols, and a non-associating component by use of only binary parameters.

TABLE 5

Predicted results for ternary excess enthalpies at 25°C

System	No. of data points	Abs. arith. mean dev. (J mol^{-1})	Ref.
1-Propanol–acetonitrile–benzene	56	21.5	22
2-Propanol–acetonitrile–benzene	61	18.4	22

LIST OF SYMBOLS

A, B, C	alcohol, acetonitrile, and non-associating component, respectively
a_{IJ}	binary interaction parameter
C_I, D_I	constants of eqn. (14)
G_{IJ}	coefficient as defined by exp $(-\alpha_{IJ}\tau_{IJ})$
H^{E}	molar excess enthalpy
h_2	enthalpy of hydrogen-bond formation of alcohol dimer
h_{A}	enthalpy of hydrogen-bond formation of alcohol higher polymer including cyclic polymer
$h_{\mathrm{A}_i\mathrm{B}}$	enthalpy of formation of chemical complex $\mathrm{A}_i\mathrm{B}$ between alcohol i-mer and acetonitrile
$h_{\mathrm{A}_i\mathrm{B}_j}$	enthalpy of formation of chemical complex $\mathrm{A}_i\mathrm{B}_j$ between alcohol i-mer and acetonitrile j-mer
$h_{\mathrm{A}_i\mathrm{C}}$	enthalpy of formation of chemical complex $\mathrm{A}_i\mathrm{C}$ between alcohol i-mer and non-associating component
h'_{B}	enthalpy of formation for head-to-head dimerization of acetonitrile
h_{B}	enthalpy of formation for head-to-tail chain association of acetonitrile
h_{BC}	enthalpy of formation of chemical complex BC between acetonitrile and non-associating component
K_2	association constant of dimer formation of alcohol
K_3	association constant of open chain trimer formation of alcohol
K	association constant of open chain i-mer formation of alcohol, $i > 3$
K_{cy}	association constant for cyclization of open chain i-mer as defined by θ/i, $i > 4$
$K_{\mathrm{A}_i\mathrm{B}}$	solvation constant of formation of chemical complex $\mathrm{A}_i\mathrm{B}$ between alcohol i-mer and acetonitrile, $i \geqslant 1$
$K_{\mathrm{A}_i\mathrm{B}_j}$	solvation constant of formation of chemical complex $\mathrm{A}_i\mathrm{B}_j$ between alcohol i-mer and acetonitrile j-mer, $i \geqslant 1$, $j \geqslant 2$
$K_{\mathrm{A}_i\mathrm{C}}$	solvation constant of formation of chemical complex $\mathrm{A}_i\mathrm{C}$ between alcohol i-mer and non-associating component, $i \geqslant 1$
K'_{B}	association constant of head-to-head dimerization of acetonitrile
K_{B}	association constant of head-to-tail chain association of acetonitrile
K_{BC}	solvation constant of formation of chemical complex BC between acetonitrile and non-associating component
P	total pressure
P_I^*	vapor pressure of pure component I
R	gas constant
S	stoichiometric sum
T	absolute temperature
$v_I^{*\mathrm{L}}$	liquid molar volume of pure component I
x_I	liquid-phase mole fraction of component I

y_I	vapor-phase mole fraction of component I
w	coefficient as defined by $K_B x_{B_1}$
z	coefficient as defined by $K x_{A_1}$

Greek letters

α_{IJ}	nonrandomness parameters of NRTL equation
γ_I	activity coefficient of component I
θ	constant related to K_{cy}
τ_{JI}	coefficient as defined by a_{JI}/T
ϕ_I	vapor-phase fugacity coefficient of component I
ϕ_I^*	vapor-phase fugacity coefficient of pure component I at system temperature T and pressure P_I^*

Subscripts

A, B, C	alcohol, acetonitrile, and non-associating component, respectively
A_1, A_i	alcohol monomer and i-mer
A_iB	complex formation between alcohol i-mer and acetonitrile
A_iB_j	complex formation between alcohol i-mer and acetonitrile j-mer
A_iC	complex formation between alcohol i-mer and non-associating component
BC	complex formation between acetonitrile and non-associating component
chem	chemical
phys	physical
I, J, K	components

Superscripts

E	excess
L	liquid
*	pure liquid

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