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THERMODYNAMICS OF SOLUTIONS CONTAINING ACETONITRILE AND 1-BUTANOL

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ABSTRACT

Vapor–liquid and liquid–liquid equilibria and excess enthalpies for binary and ternary mixtures including acetonitrile and 1-butanol are well correlated with the association model of Nagata and Tamura, which contains four association constants for the alcohol and two association constants for acetonitrile, and solvation constants between unlike molecules, with allowance for a non-polar interaction term between components.

INTRODUCTION

Thermodynamic studies for solutions of acetonitrile with methanol, ethanol and propanols have been carried out using the association model based on mole fraction statistics [1,2]. The model includes four association constants for alcohol molecules, two association constants for acetonitrile molecules and two solvation constants between alcohol open-chains and acetonitrile complexes, with allowance for the physical interaction term expressed by the NRTL equation [3]. The spectroscopic and thermodynamic properties of solutions of 1-butanol in hydrocarbons are well reproduced with the alcohol association model [4]. This paper shows the correlation of vapor–liquid equilibrium and excess enthalpy data of binary acetonitrile + 1-butanol solution, and the prediction of vapor–liquid and liquid–liquid equilibrium and excess enthalpy data of ternary mixtures containing acetonitrile, 1-butanol and a hydrocarbon, by use of the association model with binary parameters alone.

ASSOCIATION MODEL

In a ternary mixture formed by 1-butanol, acetonitrile and benzene, A stands for the alcohol, B for acetonitrile and C for benzene. We assume that:

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(1) The alcohol exists in the mixture in the forms of linear, hydrogen-bonded polymers as well as cyclic ones formed by chemical reactions and the association constants for these reactions are defined in terms of mole fractions

$$K_2 = x_{A_2}/x_{A_1}^2 \qquad \text{for } A_1 + A_1 = A_2 \tag{1}$$

$$K_3 = x_{A_3}/x_{A_2}x_{A_1} \qquad \text{for } A_2 + A_1 = A_3 \tag{2}$$

$$K = x_{A_{i+1}}/x_{A_i}x_{A_1} \qquad \text{for } A_i + A_1 = A_{i+1} \; i \geqslant 3 \tag{3}$$

$$K_{cy} = \theta/i = x_{A_i}(\text{cyclic})/x_{A_i}(\text{linear}) \qquad \text{for } x_{A_i}(\text{linear}) = x_{A_i}(\text{cyclic}) \qquad i > 4 \tag{4}$$

(2) Acetonitrile forms cyclic dimer and linear polymers

$$K_B' = x_{B_2}/x_{B_1}^2 \qquad \text{for } B_1 + B_1 = B_2 \tag{5}$$

$$K_B = x_{B_{i+1}}/x_{B_i}x_{B_1} \qquad \text{for } B_i + B_1 = B_{i+1} \qquad i \geqslant 1 \tag{6}$$

(3) All of the components solvate each other

$$K_{A_iB} = x_{A_iB}/x_{A_i}x_{B_1} \qquad \text{for } A_i + B_1 = A_iB \quad i \geqslant 1 \tag{7}$$

$$K_{A_iB_j} = x_{A_iB_j}/x_{A_i}x_{B_j} \qquad \text{for } A_i + B_j = A_iB_j \quad i \geqslant 1 \text{ and } j \geqslant 2 \tag{8}$$

$$K_{A_iC} = x_{A_iC}/x_{A_i}x_{C_1} \qquad \text{for } A_i + C_1 = A_iC \quad i \geqslant 1 \tag{9}$$

$$K_{BC} = x_{BC}/x_{B_1}x_{C_1} \qquad \text{for } B_1 + C_1 = BC \tag{10}$$

(4) The temperature-dependence of the equilibrium constants is given by the van't Hoff relation and the enthalpies of hydrogen-bond formation and complex formation are independent of the temperature and the degree of association

$$\begin{aligned}
&\partial \ln K_2/\partial(1/T) = -h_2/R && \partial \ln K_3/\partial(1/T) = -(2h_A - h_2)/R \\
&\partial \ln K/\partial(1/T) = -h_A/R && \partial \ln \theta/\partial(1/T) = -h_A/R \\
&\partial \ln K_{A_iB}/\partial(1/T) = -h_{A_iB}/R && \partial \ln K_{A_iB_j}/\partial(1/T) = -h_{A_iB_j}/R \\
&\partial \ln K_{A_iC}/\partial(1/T) = -h_{A_iC}/R && \partial \ln K_{BC}/\partial(1/T) = -h_{BC}/R
\end{aligned} \tag{11}$$

(5) There are non-polar interactions described by the NRTL eqn. (3) between all of the components.

The activity coefficient of any component in the ternary mixture is expressed by

$$\ln \gamma_I = \ln\left(\frac{x_{I_1}}{x_{I_1}^{\star} x_I}\right) + \frac{\sum_J \tau_{JI} G_{JI} x_J}{\sum_K G_{KI} x_K} + \sum_J \frac{x_J G_{IJ}}{\sum_K G_{KJ} x_K}\left(\tau_{IJ} - \frac{\sum_R x_R \tau_{RJ} G_{RJ}}{\sum_K G_{KJ} x_J}\right) \tag{12}$$

where

$$\tau_{JI} = a_{JI}/T \tag{13}$$

$$G_{JI} = \exp(-\alpha_{JI}\tau_{JI}) \tag{14}$$

α_{JI} $(=\alpha_{IJ})$ is the nonrandomness parameter set as 0.3 and $x^{\star}_{C_1} = 1$ for benzene.

The monomer mole fractions of the components are simultaneously solved from mass balance equations relating the nominal mole fractions to the monomer mole fractions and the equilibrium constants and eqn. (19)

$$x_A = \left\{\left[1 + K_{A_iB}x_{B_1} + K_{A_iC}x_{C_1} + \frac{K_{A_iB_j}x_{B_1}w}{(1-w)}\right]\right.$$
$$\left.\times\left[x_{A_1} + 2K_2x^2_{A_1} + \frac{K_2K_3x^3_{A_1}(3-2z)}{(1-z)^2}\right] + \frac{K_2K_3K^2\theta x^5_{A_1}}{(1-z)}\right\}/S \tag{15}$$

$$x_B = \left\{\left[K_{A_iB}x_{B_1} + \frac{K_{A_iB_j}x_{B_1}w(2-w)}{(1-w)^2}\right]\left[x_{A_1} + K_2x^2_{A_1} + \frac{K_2K_3x^3_{A_1}}{(1-z)}\right]\right.$$
$$\left. + \frac{x_{B_1}}{(1-w)} + 2K'_Bx^2_{B_1} + K_{BC}x_{B_1}x_{C_1}\right\}/S \tag{16}$$

$$x_C = \left\{K_{A_iC}x_{C_1}\left[x_{A_1} + K_2x^2_{A_1} + \frac{K_2K_3x^3_{A_1}}{(1-z)}\right] + K_{BC}x_{B_1}x_{C_1} + x_{C_1}\right\}/S \tag{17}$$

where $w = K_Bx_{B_1}$, $z = Kx_{A_1}$ and S is the stoichiometric sum given by

$$S = \left[1 + K_{A_iB}x_{B_1} + K_{A_iC}x_{C_1} + \frac{K_{A_iB_j}x_{B_1}w}{(1-w)}\right]$$
$$\times\left[x_{A_1} + 2K_2x^2_{A_1} + \frac{K_2K_3x^3_{A_1}(3-2z)}{(1-z)^2}\right]$$
$$+ \frac{K_2K_3K^2\theta x^5_{A_1}}{(1-z)} + \left[K_{A_iB}x_{B_1} + K_{A_iC}x_{C_1} + \frac{K_{A_iB_j}x_{B_1}w(2-w)}{(1-w)^2}\right]$$
$$\times\left[x_{A_1} + K_2x^2_{A_1} + \frac{K_2K_3x^3_{A_1}}{(1-z)}\right] + \frac{x_{B_1}}{(1-w)^2} + 2K'_Bx^2_{B_1} + 2K_{BC}x_{B_1}x_{C_1} + x_{C_1} \tag{18}$$

The sum of the mole fractions of all chemical species present in the mixture must be unity

$$\left[1 + K_{A_iB}x_{B_1} + K_{A_iC}x_{C_1} + \frac{K_{A_iB_j}x_{B_1}w}{(1-w)}\right]\left[x_{A_1} + K_2x_{A_1}^2 + \frac{K_2K_3x_{A_1}^3}{(1-z)}\right]$$
$$- \frac{K_2K_3\theta}{K^3}\left[\ln(1-z) + z + \frac{z^2}{2} + \frac{z^3}{3} + \frac{z^4}{4}\right]$$
$$+ \frac{x_{B_1}}{(1-w)} + K'_Bx_{B_1}^2 + K_{BC}x_{B_1}x_{C_1} + x_{C_1} = 1 \tag{19}$$

The monomer mole fractions in pure liquid states, $x_{A_1}^\star$ and $x_{B_1}^\star$, are obtained from eqns. (20) and (21), respectively

$$x_{A_1}^\star + K_2x_{A_1}^{\star 2} + K_2K_3x_{A_1}^{\star 3}/(1-z^\star)$$
$$- \frac{K_2K_3\theta}{K^3}\left[\ln(1-z^\star) + z^\star + \frac{z^{\star 2}}{2} + \frac{z^{\star 3}}{3} + \frac{z^{\star 4}}{4}\right] = 1 \tag{20}$$

$$x_{B_1}^\star/(1-w^\star) + K'_Bx_{B_1}^{\star 2} = 1 \tag{21}$$

The ternary excess molar enthalpy of the mixture is given by the sum of chemical and physical contributions

$$H^{\mathrm{E}} = H_{\mathrm{chem}}^{\mathrm{E}} + H_{\mathrm{phys}}^{\mathrm{E}} \tag{22}$$

$$H_{\mathrm{chem}}^{\mathrm{E}} = \left\{\left[1 + K_{A_iB}x_{B_1} + K_{A_iC}x_{C_1} + \frac{K_{A_iB_j}x_{B_1}w}{(1-w)}\right]\right.$$
$$\times\left[h_2K_2x_{A_1}^2 + \frac{h_AK_2K_3x_{A_1}^3(2-z)}{(1-z)^2}\right] + \frac{h_AK_2K_3K^2\theta x_{A_1}^5}{(1-z)}$$
$$+\left(1 + K_{A_iB_j}\left[x_{A_1} + K_2x_{A_1}^2 + \frac{K_2K_3x_{A_1}^3}{(1-z)}\right]\right)\frac{h_Bx_{B_1}w}{(1-w)^2}$$
$$+h'_BK'_Bx_{B_1}^2 + \left[x_{A_1} + K_2x_{A_1}^2 + \frac{K_2K_3x_{A_1}^3}{(1-z)}\right]$$
$$\times\left[h_{A_iB}K_{A_iB}x_{B_1} + h_{A_iC}K_{A_iC}x_{C_1} + \frac{h_{A_iB_j}K_{A_iB_j}x_{B_1}w}{(1-w)}\right]$$
$$\left.+h_{BC}K_{BC}x_{B_1}x_{C_1}\right\}/S$$
$$-x_A\left[h_2K_2x_{A_1}^{\star 2} + \frac{h_AK_2K_3x_{A_1}^{\star 3}(2-z^\star)}{(1-z^\star)^2} + \frac{h_AK_2K_3K^2\theta x_{A_1}^{\star 5}}{(1-z^\star)}\right]/S_A^\star$$
$$-x_B\left[h'_BK'_Bx_{B_1}^{\star 2} + \frac{h_Bx_{B_1}^\star w^\star}{(1-w^\star)^2}\right]/S_B^\star \tag{23}$$

where $w^\star = K_B x^\star_{B_1}$, $z^\star = K x^\star_{A_1}$, and $S^\star_A$ and $S^\star_B$ are the stoichiometric sums given by

$$S^\star_A = x^\star_{A_1} + 2K_2 x^{\star 2}_{A_1} + \frac{K_2 K_3 x^{\star 3}_{A_1}(3 - 2z^\star)}{(1 - z^\star)^2} + \frac{K_2 K_3 K^2 \theta x^{\star 5}_{A_1}}{(1 - z^\star)} \tag{24}$$

$$S^\star_B = 2K'_B x^{\star 2}_{B_1} + x^\star_{B_1}/(1 - w^\star)^2 \tag{25}$$

$$H^{\mathrm{E}}_{\mathrm{phys}} = R\sum_I x_I \left[\frac{\sum_J x_J \dfrac{\partial(\tau_{JI} G_{JI})}{\partial(1/T)}}{\sum_K G_{KI} x_K} - \frac{\sum_J \tau_{JI} G_{JI} x_J \sum_K x_K \dfrac{\partial G_{KI}}{\partial(1/T)}}{\left(\sum_K G_{KI} x_K\right)^2} \right] \tag{26}$$

The temperature-dependence of the energy parameters is assumed to be given by

$$a_{JI} = C_I + D_I(T - 273.15) \qquad a_{IJ} = C_J + D_J(T - 273.15) \tag{27}$$

CALCULATED RESULTS

Ternary experimental phase equilibrium data are available: vapor–liquid equilibrium data for the 1-butanol + acetonitrile + benzene at 60 °C [5]; liquid–liquid equilibrium data for the acetonitrile + 1-butanol + cyclohexane, acetonitrile + 1-butanol + n-hexane and acetonitrile + 1-butanol + n-heptane systems at 25 °C [6]. Excess molar enthalpies were measured at 25 °C for the acetonitrile + 1-butanol + benzene system [7].

Vapor–liquid equilibrium calculations were performed using the following thermodynamic relations for component I

$$\phi_I y_I P = x_I \gamma_I \phi^{\mathrm{s}}_I P^{\mathrm{s}}_I \exp\left[v^{\mathrm{L}}_I (P - P^{\mathrm{s}}_I)/RT\right] \tag{28}$$

$$\ln \phi_I = \left(2\sum_J y_J B_{IJ} - \sum_I \sum_J y_I y_J B_{IJ}\right)\frac{P}{RT} \tag{29}$$

where ϕ, y and P are the vapor-phase fugacity coefficient, vapor-phase mole fraction and total pressure, respectively, v^{L} is the pure-liquid molar volume estimated from a quadratic equation in terms of temperature [1]. The pure-component vapor pressures P^{s} were obtained from the Antoine equation [8,9] and the original ref. 5. The second virial coefficients B_{IJ} were estimated from the general correlation of Hayden and O'Connell [10]. The energy parameters were calculated using a program which minimizes the sum-of-squares of relative deviation in pressure plus the sum-of-squares of deviations in vapor-phase mole fraction by means of the simplex method [11].

TABLE 1

Solvation equilibrium constants and enthalpies of complex formation

System (A + B)	Temp. (°C)	K_{A_iB}	$K_{A_iB_j}$	$-h_{A_iB}$ (kJ mol^{-1})	$-h_{A_iB_j}$ (kJ mol^{-1})
1-Butanol + acetonitrile	60	30	25	22.0	16.8
1-Butanol + benzene	25	2.8		8.2	
Acetonitrile + benzene	45	0.2 [a]		5.2	

[a] 1 : 1 Complex formation is assumed.

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

$$(\gamma_I x_I)^{\mathrm{I}} = (\gamma_I x_I)^{\mathrm{II}} \tag{30}$$

where the superscripts I and II denote the two equilibrium liquid phases.

The thermodynamic association parameters for 1-butanol and acetonitrile [4,12] were set as: for 1-butanol, $K_2 = 30$, $K_3 = 90$, $K = 35$ and $\theta = 75$ at 25°C, $h_2 = -21.2$ kJ mol^{-1} and $h_A = -23.5$ kJ mol^{-1}; for acetonitrile, $K_B' = 8.35$ and $K_B = 2.1$ at 45°C, $h_B' = -8.9$ kJ mol^{-1} and $h_B = -6.7$ kJ mol^{-1}. The solvation constants and enthalpies of complex formation are listed in Table 1. Table 2 gives the calculated results obtained in fitting the model to vapor–liquid equilibrium data for binary systems. Figure 1 compares the experimental vapor–liquid equilibria of 1-butanol + acetonitrile at 60°C and 1-butanol + benzene at 45°C with the calculated results. The

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 (K)		Abs. arith. mean dev.		Ref.
			a_{AB}	a_{BA}	Δy [a] ($\times 10^3$)	ΔP (kPa)	
1-Butanol + acetonitrile	60	8	−233.28	612.79	5.5	0.120	5
1-Butanol + benzene	45	9	−251.13	320.78	3.7	0.493	13
1-Butanol + cyclohexane	50	14	681.27	−410.17	9.1	0.280	14
1-Butanol + n-heptane	60	19	245.96	−175.48	3.8	0.187	15
1-Butanol + n-hexane	59.38	24	499.26	−308.22	3.2	0.613	16
Acetonitrile + benzene	55	12	−193.98	406.75	5.8	0.213	17
Acetonitrile + cyclohexane	25	MS [b]	389.34	478.84			18
Acetonitrile + n-heptane	25	MS	282.64	646.79			6
Acetonitrile + n-hexane	25	MS	322.69	527.46			6

[a] $\Delta y = \Sigma_1^N \mid y_1(\text{exptl}) - y_1(\text{calc}) \mid / N$, where N is the total experimental points.
[b] MS = mutual solubility.

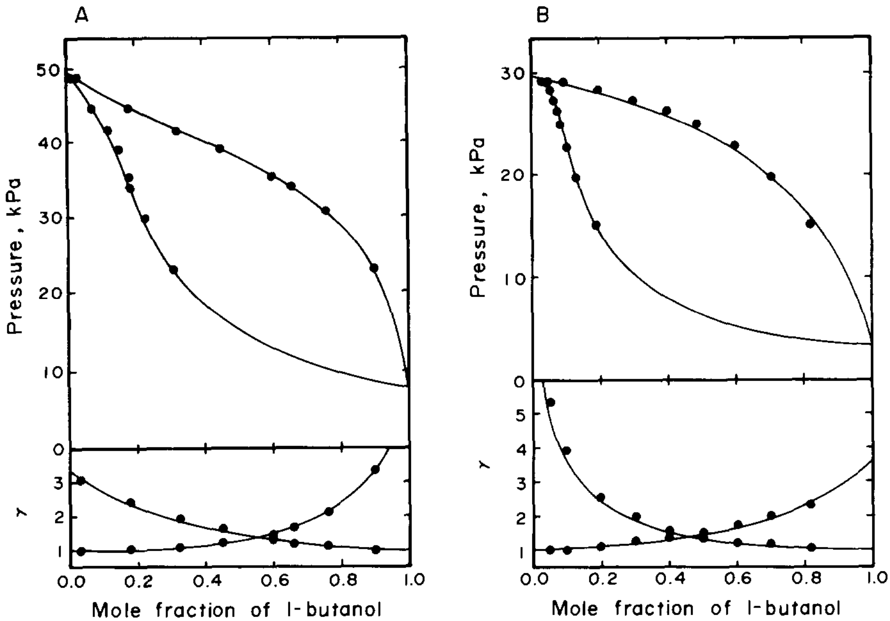

Fig. 1. Vapor–liquid equilibria for (a) 1-butanol(1)+acetonitrile(2) at 60 °C and (b) 1-butanol(1)+benzene(2) at 45 °C. Calculated (———). Experimental (●): (A) data of Nagata [5]; (B) data of Brown and Smith [13].

ternary prediction of vapor–liquid equilibrium for 1-butanol(1) + acetonitrile(2) + benzene(3) at 60 °C was made for the fifteen ternary points. The average deviations calculated from $\Delta y_i = \Sigma_j^N \mid y_i(\text{exptl}) - y_i(\text{calc}) \mid_j / N$, where N is the total experimental points, were $\Delta y_1 = 0.0036$, $\Delta y_2 = 0.0062$ and $\Delta y_3 = 0.0045$. The average pressure deviation was 0.693 kPa and the average relative pressure deviation was 1.4%. Figure 2 shows the ternary predicted solubility envelopes of the three systems at 25 °C.

The excess molar enthalpies of the three binaries constituting the 1-

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				Deviation ($J\ mol^{-1}$)	Ref.
		C_A (K)	C_B (K)	D_A	D_B		
1-Butanol + acetonitrile	17	−476.65	−392.48	−2.2380	−2.0039	6.6	7
1-Butanol + benzene	10	701.44	462.37	−1.1130	−1.9384	7.7	19
Acetonitrile + benzene	15	399.37	−21.88	2.6004	−0.9393	1.3	20

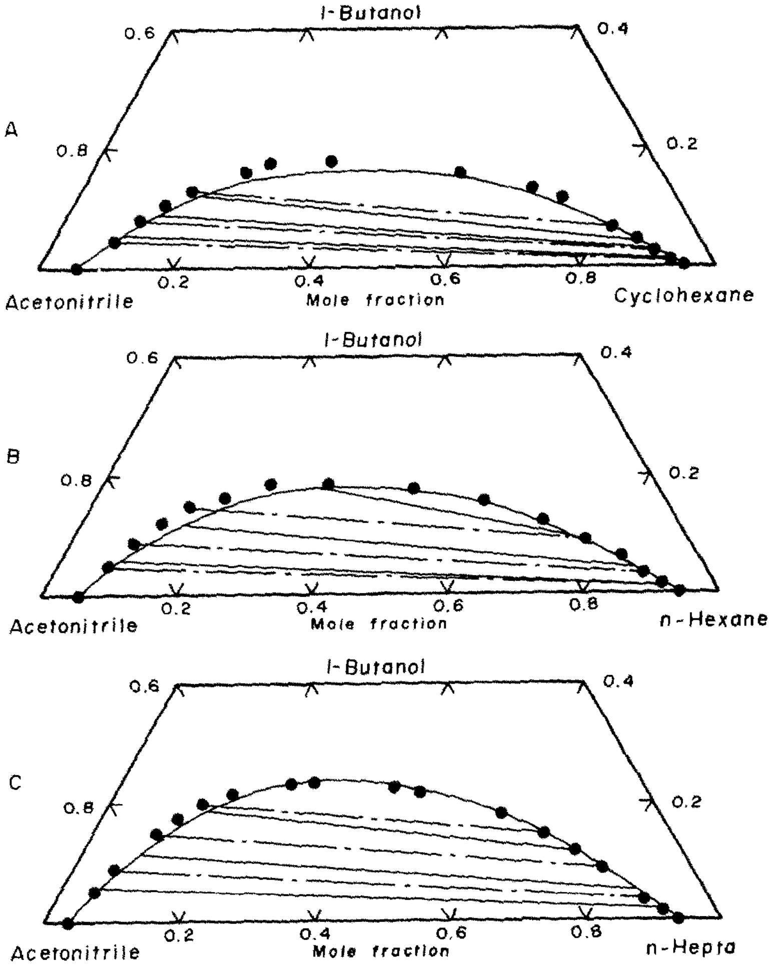

Fig. 2. Ternary liquid–liquid equilibria at 25 °C. Calculated (———). Experimental tie line (●—-—-—●) data of Nagata [6]: (A) acetonitrile + 1-butanol + cyclohexane; (B) acetonitrile + 1-butanol + n-hexane; (C) acetonitrile + 1-butanol + n-heptane.

butanol + acetonitrile + benzene system were well reduced with the association model using the simplex method and the calculated results are summarized in Table 3. Figure 3 shows the calculated results and the experimental data points for the 1-butanol + acetonitrile and 1-butanol + benzene systems at 25 °C. The ternary prediction of excess molar enthalpy was 21.6 J mol^{-1} for the sixty-six data points of the 1-butanol + acetonitrile + benzene system at 25 °C using only the binary parameters listed in Table 3.

In conclusion, the association model is able to reproduce the vapor–liquid equilibrium and excess molar enthalpy data of the 1-butanol + acetonitrile system and to predict vapor–liquid and liquid–liquid equilibria and excess

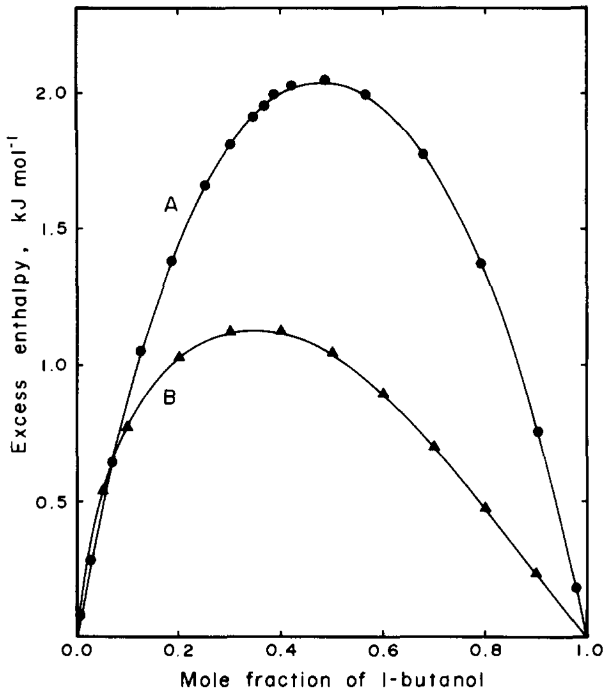

Fig. 3. Excess molar enthalpies for two binary systems at 25 °C. Calculated (———). Experimental: (A) 1-butanol + acetonitrile, data of Nagata and Tamura [7] (●); (B) 1-butanol + benzene, data of Mrazek and Van Ness [19] (▲).

molar enthalpies for the ternary 1-butanol + acetonitrile + hydrocarbon systems with good accuracy.

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. (27)
G_{IJ}	coefficient as defined by $\exp(-\alpha_{IJ}\tau_{IJ})$
H^E	excess molar enthalpy
h_2	enthalpy of hydrogen-bond formation of alcohol dimer
h_A	enthalpy of hydrogen-bond formation of alcohol higher polymer including cyclic case
h_{A_iB}	enthalpy of formation of chemical complex A_iB between alcohol i-mer and acetonitrile monomer
$h_{A_iB_j}$	enthalpy of formation of chemical complex A_iB_j between alcohol i-mer and acetonitrile j-mer
h_{A_iC}	enthalpy of formation of chemical complex A_iC 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_{A_iB}	solvation constant of formation of chemical complex A_iB between alcohol i-mer and acetonitrile monomer, $i \geqslant 1$
$K_{A_iB_j}$	solvation constant of formation of chemical complex A_iB_j between alcohol i-mer and acetonitrile j-mer, $i \geqslant 1$, $j \geqslant 2$
K_{A_iC}	solvation constant of formation of chemical complex A_iC 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^s	saturated vapor pressure of pure component I
R	universal gas constant
S	stoichiometric sum
T	absolute temperature
v_I^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 parameter of NRTL equation ($= \alpha_{JI}$)
γ_I	activity coefficient of component I
θ	constant related to K_{cy}
τ_{IJ}	coefficient as defined by a_{IJ}/T
ϕ_I	vapor-phase fugacity coefficient of component I
ϕ_I^s	vapor-phase fugacity coefficient of pure component I at system temperature T and pressure P_I^s

Subscripts

A, B, C	alcohol, acetonitrile and nonassociating component, respectively
A_1, A_i	alcohol monomer and i-mer

A_iB complex formation between alcohol i-mer and acetonitrile monomer

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

I, J, K components

phys physical

Superscripts

E excess

L liquid

s saturation

⋆ pure-liquid reference state

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