

Simultaneous Correlation of Liquid-liquid Equilibria for Ternary Systems and Phase Equilibria for Constitutive Binary Systems by Modified New Activity Coefficient Model

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Abstract

A modified concentration dependent surface area parameter model is proposed to avoid the restriction conditions for the parameters of pure components. The model is applied to correlate the ternary liquid-liquid equilibria and the constitutive binary phase equilibria with the same parameter sets. The correlated results for both vapor-liquid equilibria and liquid-liquid equilibria agree very well with the experimental data and are better than NRTL and UNIQUAC which are widely used. Temperature dependence parameters are introduced to the model. Correlations are performed for multiple experimental data at different temperatures, and it shows that the model represents experimental data at multiple temperatures with the same parameter sets simultaneously.

Keywords

Activity coefficient, Vapor-liquid equilibrium, Liquid-liquid equilibrium

1. Introduction

NRTL [1] and UNIQUAC [2] models, which are widely used to correlate phase equilibria, have difficulty in correlating the ternary liquid-liquid equilibria and the constitutive binary vapor-liquid equilibria with the same parameter set. In addition, some systems are difficult to correlate the entire concentration range of the vapor-liquid equilibrium with the same parameter set. Iwai et al. [3] proposed CDSAP (Concentration Dependent Surface Area Parameter) model, which assumes that the surface area of the quasi-chemical equation depends on the composition. CDSAP model has a problem that it requires iterative calculations to obtain activity coefficients for ternary systems or more, which makes the calculations complicated. Then, a new activity

coefficient model, r-CDSAP model [4, 5], was proposed to solve this problem. However, r-CDSAP model is slightly inconvenient to use for ternary systems or more, because there are restriction conditions for the parameters of pure components. In this study, the authors propose a new improved activity coefficient model, f-CDSAP model, which solves the problem of r-CDSAP model. The ternary liquid-liquid equilibria and the constitutive binary vapor-liquid and liquid-liquid equilibria are correlated simultaneously with the same parameter sets by f-CDSAP model. In addition, temperature dependent parameters are introduced into f-CDSAP model, and simultaneous correlations for the ternary liquid-liquid equilibria and the constitutive binary phase equilibria with different temperatures are performed.

2. Activity Coefficient models

2.1. CDSAP model [3]

In CDSAP model, the excess Gibbs free energy G^E is expressed as follows.

$$\frac{G^E}{RT} = \sum_i q_i n_i \ln \frac{\theta_{ii}}{\theta_i} \quad (1)$$

$$\theta_j = \frac{q_j n_j}{\sum_m (q_m n_m)} \quad (2)$$

$$q_i = q_i^0 x_i + \sum_{j \neq i} q_{ji}^\infty x_j \quad (3)$$

$$\frac{\theta_{ij} \theta_{ji}}{\theta_{ii} \theta_{jj}} = \exp (\Delta E_{ij}) \quad (4)$$

where R is the gas constant, T is the temperature, q is the surface area (interaction number), n is the amount of substance, θ_j is the average surface are fraction of component j , θ_{ji} is the local surface area fraction of component j around component i , x is the mole fraction, q_i^0 is the molecular surface area parameter of pure component i , q_{ji}^∞ is the molecular surface area parameter of component i in component j at infinite dilution, and ΔE_{ij} is the exchange interaction energies.

2.2. r-CDSAP model [4, 5]

The excess Gibbs free energy for binary system of r-CDSAP model is obtained by the sum of the first 2 terms of Taylor series at $\Delta E_{12} = 0$ for G^E of CDSAP model as follows.

$$G^E(\Delta E_{12}) \cong G^E(0) + \frac{dG^E(0)}{d\Delta E_{12}} \Delta E_{12} = -RT \Delta E_{12} \frac{q_1 n_1 q_2 n_2}{q_1 n_1 + q_2 n_2} \quad (5)$$

The general form is expressed as follows.

$$\frac{G^E}{RT} = \frac{1}{2} \sum_i \sum_j (-\Delta E_{ij}) (q_i n_i) \theta_j, \quad \Delta E_{ij} = \Delta E_{ji}, \quad \Delta E_{ii} = 0 \quad (6)$$

For ternary systems or more, restriction conditions of q^0 are necessary. The form for ternary

system is expressed by the next equation.

$$\frac{q_1^0 q_2^0 q_3^0}{q_2^0 q_3^0 q_1^0} = 1 \quad (7)$$

2.3. *f*-CDSAP model

In *f*-CDSAP model, q_i^0 of the binary system (*i*) + (*j*) is determined for each binary system.

$$q_i^0 = q_{ji}^* \quad (8)$$

For ternary systems or more, q_i^0 is expressed as follows.

$$q_i^0 = \frac{\sum_{j \neq i} q_{ji}^* x_j}{\sum_{j \neq i} x_j} \quad (9)$$

The surface area q_i is obtained by the following equation.

$$q_i = \frac{\sum_j q_{ji}^* x_j}{\sum_j x_j} x_i + \sum_j q_{ij}^\infty x_j \quad (10)$$

This eliminates the requirement to fix the value of q_i^0 for each substance in ternary systems or more. *f*-CDSAP model is the same as *r*-CDSAP model for binary systems, and also the same for ternary systems or more if all q_{ji}^* for each component are the same value. The fitting parameters are c_{ji}^* and c_{ji}^∞ represented by the following equations for binary systems, and c_{ji}^* , c_{ji}^∞ , and $-\Delta E_{ij}$ for ternary systems or more.

$$c_{ji}^* = -\Delta E_{ij} q_{ji}^* \quad (11)$$

$$c_{ji}^\infty = -\Delta E_{ij} q_{ji}^\infty \quad (12)$$

The values of $-\Delta E_{ij}$ are relative, and if one $-\Delta E_{ij}$ is fixed (e.g., $\Delta E_{12} = 1$), the other values can be determined.

3. Results and discussion

3.1. Correlation for isothermal systems

Ternary liquid-liquid equilibria and constitutive binary vapor-liquid and liquid-liquid equilibria were correlated by *f*-CDSAP model with the same parameter sets. The correlated results by *f*-CDSAP model are compared with the those by *r*-CDSAP, NRTL, and UNIQUAC models which were reported by Iwai [4]. Table 1 shows the calculated systems and data sources. The data of vapor-liquid and liquid-liquid equilibria for constitutive binary systems were taken from the same or similar temperatures as those for the ternary liquid-liquid equilibria. The parameters were determined by Marquardt method. The objective function F is as follows.

$$F = \frac{\sum^{N_{VLE}} \sum_i^2 (|\gamma_i^{\text{exp}} - \gamma_i^{\text{calc}}| / \gamma_i^{\text{exp}})}{2N_{VLE}} + \sum_{p=1}^2 \frac{|x_3^{p,\text{exp}} - x_3^{p,\text{calc}}|}{2} + \frac{1}{N_t} \sum_{t=1}^{N_t} \sqrt{\sum_{p=1}^2 \sum_{i=1}^3 \frac{(x_i^{p,t,\text{exp}} - x_i^{p,t,\text{calc}})^2}{5}} \quad (13)$$

The first, second, and third terms on the right side of Eq. (13) correspond to the vapor-liquid equilibria of the binary systems (1) + (2) and (2) + (3), the liquid-liquid equilibria of the binary system (1) + (3), and the ternary system (1) + (2) + (3), respectively. N_{VLE} is the number of activity coefficient data for binary vapor-liquid equilibria, subscript p means the liquid phase, t means the tie line, and N_t is the number of tie lines.

Table 2 shows the parameter values of f-CDSAP model determined. Fig. 1 shows the correlated results of liquid-liquid equilibria for methanol (1) + benzene (2) + cyclohexane (3) system at 298.15 K (Fig. 1 (a)), and activity coefficients for constitutive binary systems (Fig. 1 (b) and (c)). f-CDSAP model can correlate both the ternary liquid-liquid equilibria and the binary vapor-liquid equilibria with high accuracy. Table 3 shows the deviations by f-CDSAP, r-CDSAP, NRTL, and UNIQUAC models for each system. In all systems, f-CDSAP model shows more accurate correlation than NRTL and UNIQUAC models. In addition, the correlation accuracy of f-CDSAP model is improved compared with r-CDSAP model due to increasing the number of degrees of freedom.

Table 1

Calculated system and data sources.

System	Component(1)	Component(2)	Component(3)	Vapor-liquid equilibria				Liquid-liquid equilibria			
				(1) + (2)		(2) + (3)		(1) + (3)		(1) + (2) + (3)	
				$T(K)$	Ref.	$T(K)$	Ref.	$T(K)$	Ref.	$T(K)$	Ref.
a	Acetonitrile	Benzene	Heptane	318.15	[6]	318.15	[7]	318.15	[7]	318.15	[7]
b	1-Butanol	1-Propanol	Water	(0.1MPa)	[8]	303.15	[9]	310.95	[10]	310.95	[10]
c	Benzene	1-Propanol	Water	313.15	[11]	303.15	[7]	310.95	[12]	310.95	[10]
d	Methanol	Benzene	Cyclohexane	298.15	[13]	298.15	[14]	298.15	[15]	298.15	[15]
e	Methanol	Toluene	Cyclohexane	318.15	[16]	318.15	[17]	298.15	[15]	298.15	[15]
f	Methanol	Acetone	Cyclohexane	298.15	[18]	298.15	[19]	298.15	[15]	298.15	[15]
g	Cyclohexane	Benzene	Nitromethane	298.15	[14]	298.15	[20]	298.15	[21]	298.15	[21]
h	Hexane	Ethanol	Acetonitrile	313.15	[22]	313.15	[22]	313.15	[22]	313.15	[22]
i	Water	Acetonitrile	Ethyl acetate	333.15	[22]	333.15	[22]	333.15	[22]	333.15	[22]

Table 2

Parameter values of f-CDSAP model.

System ^a	c_{21}^*	c_{12}^*	c_{21}^∞	c_{12}^∞	$-\Delta E_{12}$
	c_{31}^*	c_{13}^*	c_{31}^∞	c_{13}^∞	$-\Delta E_{13}$
	c_{32}^*	c_{23}^*	c_{32}^∞	c_{23}^∞	$-\Delta E_{23}$
a	1.890	0.457	1.182	1.055	1.000
	1.721	2.909	3.004	3.489	0.969
	0.209	1.379	0.414	0.875	0.475
b	0.499	0.017	0.081	0.003	0.009
	7.552	0.894	4.018	0.904	1.000
	25.661	0.369	2.555	1.656	0.764
c	0.541	7.795	1.425	2.577	0.042
	6.283	0.542	7.130	5.257	1.000
	38.591	0.285	2.423	1.698	0.527
d	1.865	1.161	3.314	2.200	1.000
	2.745	1.618	3.144	2.735	0.817
	0.239	1.042	0.431	0.494	0.625
e	2.666	1.156	2.848	2.216	1.000
	2.050	1.932	3.134	2.958	0.422
	0.288	0.473	0.406	0.378	0.205
f	0.627	0.864	0.911	0.983	1.000
	2.154	1.864	3.146	2.923	1.734
	2.080	1.068	2.539	1.976	0.950
g	0.121	1.324	0.504	0.471	1.000
	1.931	2.805	3.718	3.984	3.146
	0.885	1.402	1.317	1.092	0.506
h	0.735	5.069	2.225	3.766	1.000
	2.314	2.865	2.969	2.885	1.088
	0.845	1.817	1.259	1.378	0.641
i	0.336	18.184	1.814	2.527	1.000
	1.204	14.073	2.010	4.715	1.191
	0.820	0.182	0.423	0.431	0.090

^aSee Table 1 for system symbols.

Table 3

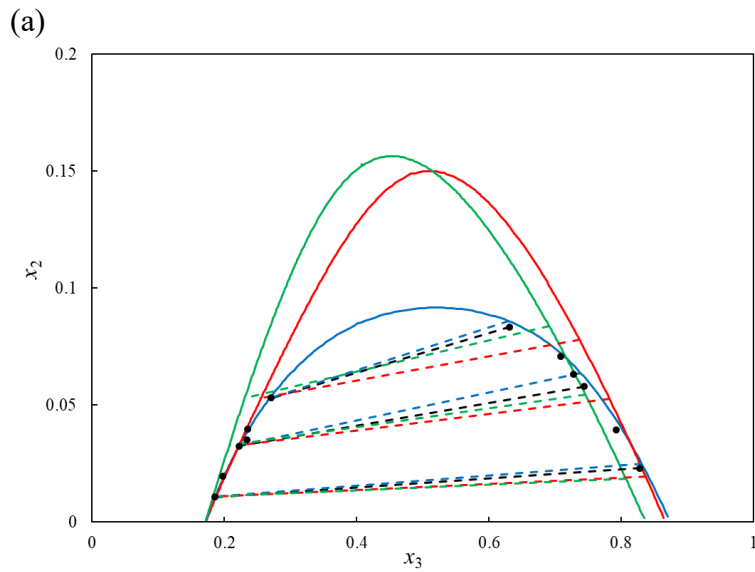
Deviations by several models. Deviations by r-CDSAP, NRTL, and UNIQUAC models are cited from previous work [4].

System ^a	f-CDSAP		r-CDSAP		NRTL		UNIQUAC	
	VLE(1)+(2) ^b VLE(2)+(3) ^b	ternary LLE ^c	VLE(1)+(2) ^b VLE(2)+(3) ^b	ternary LLE ^c	VLE(1)+(2) ^b VLE(2)+(3) ^b	ternary LLE ^c	VLE(1)+(2) ^b VLE(2)+(3) ^b	ternary LLE ^c
a	0.6	0.8	0.6	0.9	0.6	0.9	5.0	1.9
	0.9		0.9		1.3		1.4	
b	1.2	0.6	1.7	1.4	1.5	4.3	2.4	3.4
	3.0		4.1		5.2		5.8	
c	3.6	0.4	3.6	0.7	3.6	1.0	4.3	4.1
	2.2		2.3		4.9		5.6	
d	0.7	0.3	0.8	0.7	1.2	3.2	3.2	1.8
	0.2		0.6		1.4		4.5	
e	1.1	0.2	1.2	0.9	2.4	4.8	3.2	2.6
	0.1		0.7		3.3		3.0	
f	3.8	0.6	3.9	0.7	3.8	1.6	7.3	2.1
	0.5		0.9		0.7		3.5	
g	0.2	0.9	0.3	0.9	0.5	1.1	1.3	1.4
	2.3		2.3		2.3		3.3	
h	1.3	0.8	1.5	1.5	3.4	1.7	6.7	4.3
	1.1		1.1		1.4		1.5	
i	0.9	0.8	1.1	0.9	1.4	1.3	4.4	3.0
	0.5		0.8		0.6		1.0	
Average	1.4	0.6	1.6	1.0	2.2	2.2	3.7	2.7

^aSee Table 1 for system symbols.

$$^b\text{Deviation} = \frac{\sum^{N_{\text{VLE}}} \sum_i^2 (|\gamma_i^{\text{exp}} - \gamma_i^{\text{calc}}| / \gamma_i^{\text{exp}})}{2N_{\text{VLE}}} \times 100.$$

$$^c\text{Deviation} = \frac{100}{N_t} \sum_{t=1}^{N_t} \sqrt{\sum_{p=1}^2 \sum_{i=1}^3 \frac{(x_i^{p,t,\text{exp}} - x_i^{p,t,\text{calc}})^2}{5}}.$$



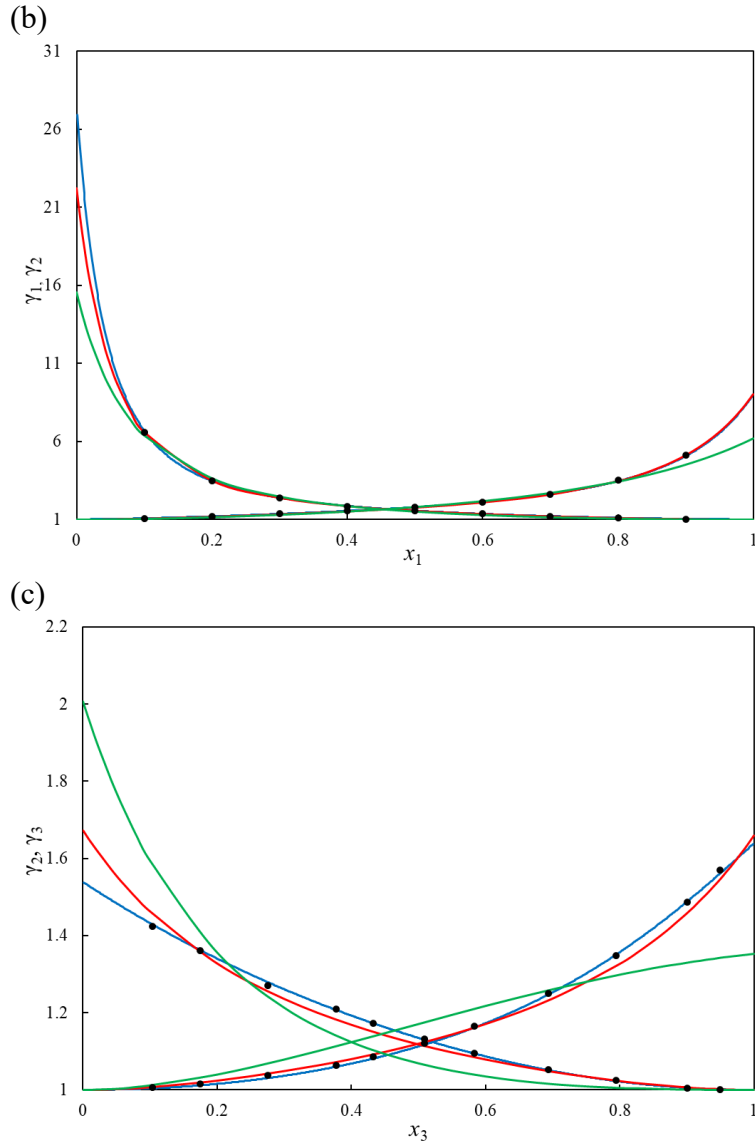


Fig. 1. (a) Liquid-liquid equilibria for methanol (1) + benzene (2) + cyclohexane (3) system at 298.15 K. (b) Activity coefficients for methanol (1) + benzene (2) system at 298.15 K. (c) Activity coefficients for benzene (2) + cyclohexane (3) system at 298.15 K. ●: Experimental data, —; —; —; Correlated results by f-CDSAP; NRTL; UNIQUAC models.

3.2. Correlations with temperature dependence parameters

The temperature dependence expressed by the following equations was assumed for the parameters of f-CDSAP model.

$$c_{ji}^* = A_{ji}^* + \frac{B_{ji}^*}{T} \quad (14)$$

$$c_{ji}^\infty = A_{ji}^\infty + \frac{B_{ji}^\infty}{T} \quad (15)$$

where T is the temperature, and A_{ji}^* , B_{ji}^* , A_{ji}^∞ , and B_{ji}^∞ are new fitting parameters.

Table 4 listed the systems and data sources for the ternary liquid-liquid equilibria and constitutive binary vapor-liquid and liquid-liquid equilibria calculated in this work.

Table 5 shows the parameter values and correlated results for each system. Fig. 2 shows the correlated results of ternary liquid-liquid equilibria for benzene (1) + ethanol (2) + water (3) system at 303.15 K and 328.15 K. Both the ternary liquid-liquid equilibria and binary vapor-liquid equilibria are correlated with high accuracy at each temperature with the same parameter set.

Table 4

Calculated systems and data sources.

System	Component(1)	Component(2)	Component(3)	Vapor-liquid equilibria				Liquid-liquid equilibria			
				(1) + (2)		(2) + (3)		(1) + (3)		(1) + (2) + (3)	
				<i>T</i> (K)	Ref.	<i>T</i> (K)	Ref.	<i>T</i> (K)	Ref.	<i>T</i> (K)	Ref.
i	Benzene	Ethanol	Water	303.15	[23]	298.15	[25]	303.15-373.15	[27]	303.15	[28]
				313.15	[23]	323.15	[26]			313.15	[28]
				323.15	[24]	333.15	[26]			328.15	[28]
ii	Ethyl acetate	Ethanol	Water	313.15	[29]	313.15	[31]	(101.3kPa)	[32]	313.15	[33]
				323.15	[30]	323.15	[26]			323.15	[34]
				343.15	[29]	333.15	[26]			333.15	[34]
iii	Benzene	1-Propanol	Water			343.15	[29]			343.15	[35]
				298.15	[13]	298.15	[38]	303.15	[41]	303.15	[43]
				313.15	[11]	303.15	[7]	310.95	[12]	310.95	[10]
				318.15	[36]	323.15	[39]	313.15	[42]	318.15	[43]
				333.15	[37]	333.15	[40]	333.15	[42]	333.15	[43]

Table 5

Values of parameters and deviations of f-CDSAP model with temperature dependency.

System ^a	Parameters								Deviation		
	A_{21}^*	$B_{21}^*(K)$	A_{12}^*	$B_{12}^*(K)$	A_{21}^∞	$B_{21}^\infty(K)$	A_{12}^∞	$B_{12}^\infty(K)$	$-\Delta E_{12}$	VLE	LLE
	A_{31}^*	$B_{31}^*(K)$	A_{13}^*	$B_{13}^*(K)$	A_{31}^∞	$B_{31}^\infty(K)$	A_{13}^∞	$B_{13}^\infty(K)$	$-\Delta E_{13}$	(1)+(2) ^b VLE	(1)+(3) ^c LLE
	A_{32}^*	$B_{32}^*(K)$	A_{23}^*	$B_{23}^*(K)$	A_{32}^∞	$B_{32}^\infty(K)$	A_{23}^∞	$B_{23}^\infty(K)$	$-\Delta E_{23}$	(2)+(3) ^b	(1)+(2)+(3) ^d
i	1.232	-37.457	-4.022	1941.899	0.831	236.681	-2.580	1601.750	1.000	1.05	0.34
	1.709	21.721	-1.058	505.287	5.504	330.005	3.100	564.421	1.791	0.87	1.35
	1.015	188.140	0.882	52.670	3.625	-662.381	1.122	-61.111	0.874		
ii	0.806	208.667	-1.632	729.439	-0.231	400.784	-1.068	657.194	1.000	0.95	0.50
	3.556	-175.292	0.929	182.890	5.106	-227.072	0.517	699.916	1.442	0.97	1.91
	1.766	-98.579	-0.122	386.811	3.037	-434.091	0.771	52.430	1.867		
iii	1.360	-275.816	0.180	1822.889	-2.344	1186.424	-5.013	2400.307	0.041	2.43	0.12
	-58.587	22755.890	-1.898	729.387	58.404	-12714.682	3.759	498.523	1.000	4.43	2.99
	17.388	2102.218	0.380	-9.782	5.223	-808.900	0.793	222.435	0.265		

^aSee Table 4 for system symbols.

$${}^b\text{Deviation} = \frac{\sum^{N_{\text{VLE}}} \sum_i^2 \left(\left| \gamma_i^{\text{exp}} - \gamma_i^{\text{calc}} \right| / \gamma_i^{\text{exp}} \right)}{2N_{\text{VLE}}} \times 100.$$

$${}^c\text{Deviation} = \sum_{p=1}^2 \frac{\left| x_3^{p,\text{exp}} - x_3^{p,\text{calc}} \right|}{2} \times 100.$$

$${}^d\text{Deviation} = \frac{100}{N_t} \sum_{t=1}^{N_t} \sqrt{\sum_{p=1}^2 \sum_{i=1}^3 \frac{\left(x_i^{p,t,\text{exp}} - x_i^{p,t,\text{calc}} \right)^2}{5}}.$$

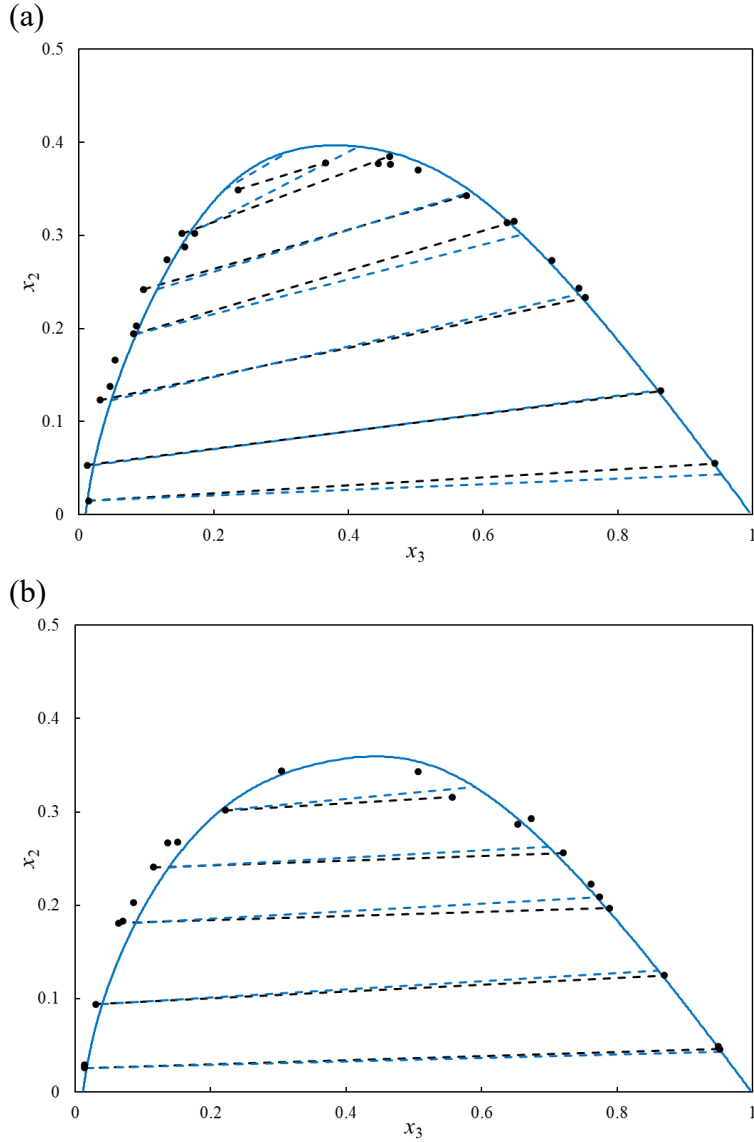


Fig. 2. Liquid-liquid equilibria for benzene (1) + ethanol (2) + water (3) system. (a) at 303.15 K. (b) at 328.15 K. •: Experimental data, — : Correlated results by f-CDSAP model.

4. Conclusion

f-CDSAP model is proposed to avoid the restriction conditions for the parameters of pure

components in r-CDSAP model. f-CDSAP model is applied to correlate the ternary liquid-liquid equilibria and the constitutive binary phase equilibria. The correlated results represent the experimental data with high accuracy and are better than NRTL and UNIQUAC models. In addition, temperature dependence parameters were introduced to correlate ternary liquid-liquid equilibria and constitutive binary phase equilibria at multiple temperatures. The correlated results are good, indicating that it is possible to express the temperature dependence.

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