

Prediction of vapor-liquid equilibria for multicomponent systems by a modified concentration dependent surface area parameter model

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Abstract

The following modifications have been made to use the previous proposed model (concentration dependent surface area parameter (CDSAP) model: Y. Iwai, Fluid Phase Equilibria 465(2018) 24-33) for estimating the activity coefficients of multicomponent systems.

- (1) It is removed the restriction condition for the surface area parameters of pure components.
- (2) A limiting condition is added to the surface area parameters of pure components for binary systems to avoid taking extreme values.
- (3) The surface area parameters of pure components determined by binary systems are used only for the corresponding term in the excess Gibbs free energy.

As a result, it is possible to estimate the activity coefficients of multicomponent systems using the parameters determined from binary systems. This is called p-CDSAP model. The vapor-liquid equilibria for acetone (1) + acetonitrile (2) + benzene (3) + ethanol (4) system are chosen to evaluate the applicability of p-CDSAP model. The estimated results by p-CDSAP model are compared with those by NRTL and UNIQUAC models. The estimated results by p-CDSAP model are comparable or slightly better than those of NRTL and UNIQUAC models.

Keywords

CDSAP model, activity coefficient, vapor liquid equilibrium

1. Introduction

NRTL model [1] and UNIQUAC model [2] are widely used to correlate phase equilibria, however, it is difficult to correlate ternary liquid-liquid equilibria and constituent binary vapor-liquid equilibria with the same set of parameters by these models. It is also difficult to correlate vapor-liquid equilibria with the same set of parameters over the entire concentration range in some systems by these models. Therefore, the author proposed CDSAP (Concentration dependent surface area parameter) model [3], assuming that the surface area (number of interacting pairs) of the quasi-chemical equilibrium depends on the composition. However, the model requires iterative calculations to obtain the activity coefficients for multicomponent (three or more components) systems, which makes the calculations complicated. Therefore, the author proposed a new model for the activity coefficients that solves the problem [4,5]. The model is called r-CDSAP model. However, r-CDSAP model has a problem that it is inconvenient to use in multicomponent systems, because there are restriction conditions for the parameters of pure substances. r-CDSAP model is excellent as a correlation model, but there are various problems in using it as a model for estimating multicomponent systems using parameters obtained from binary systems. In this study, these problems of r-CDSAP model are improved so that it can be applied to estimate the vapor-liquid equilibria of multicomponent systems. The model is called p-CDSAP model. The group contribution models such as UNIFAC model [6] and ASOG model [7], and COSMO-RS model [8] are widely used as estimation methods for multicomponent systems. It is also known that NRTL and UNIQUAC models can also be used to estimate multicomponent systems using parameters obtained from binary systems. p-CDSAP model in this study is positioned as an option in the utilization of a series of CDSAP models.

2. CDSAP model [3]

The excess Gibbs free energy G^E of CDSAP model is expressed as

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

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

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

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

where R is the gas constant, T is the temperature, q is the surface area (number of interacting pairs), n is the amount of component, θ_i is the average surface area, θ_{ji} is the local surface area of component j around component i , x is the mole fraction, q_i^0 is the molecular surface area parameter when component i is pure component, q_{ji}^∞ is the molecular surface area parameter when component i is infinitely dilute in component j , and ΔE_{ij} is the exchange energy. The activity coefficient γ_i of component i is obtained by the following equation.

$$RT \ln \gamma_i = (\partial G^E / \partial n_i)_{T,p,n_{j \neq i}} \quad (5)$$

3. r-CDSAP model [4,5]

The excess Gibbs free energy G^E of binary system for r-CDSAP model was obtained by Taylor expanding G^E of CDSAP around $\Delta E_{12}=0$ and approximating it to the second term, as shown in the following equation.

$$G^E(\Delta E_{12}) \cong G^E(0) + \frac{dG^E(0)}{d\Delta E_{12}} \Delta E_{12} = -RT \Delta E_{12} \frac{q_1 n_1 q_2 n_2}{q_1 n_1 + q_2 n_2} \quad (6)$$

The general equation is expressed as follows.

$$\frac{G^E}{RT} = \sum_i \sum_{j \neq i} (-\Delta E_{ij}) (q_i n_i) \theta_j \quad (7)$$

Here, q_i is given by Eq. (3) and θ_i is given by Eq. (2).

Substituting Eq. (7) into Eq. (5), the activity coefficient equation is derived. For example, in a binary system, the activity coefficient becomes

$$\ln \gamma_1 = \{c_{21}^0 + (c_{21}^\infty - c_{21}^0)x_2^2\}\theta_2^2 + (c_{12}^\infty - c_{12}^0)x_2^2\theta_1^2 \quad (8)$$

$$\ln \gamma_2 = \{c_{12}^0 + (c_{12}^\infty - c_{12}^0)x_1^2\}\theta_1^2 + (c_{21}^\infty - c_{21}^0)x_1^2\theta_2^2 \quad (9)$$

where

$$c_{21}^0 = -q_1^0 \Delta E_{12}, \quad c_{12}^0 = -q_2^0 \Delta E_{12}, \quad c_{21}^\infty = -q_{21}^\infty \Delta E_{12}, \quad c_{12}^\infty = -q_{12}^\infty \Delta E_{12} \quad (10)$$

$$c_1 = c_{21}^0 x_1 + c_{21}^\infty x_2 \quad (11)$$

$$c_2 = c_{12}^0 x_2 + c_{12}^\infty x_1 \quad (12)$$

$$\theta_1 = \frac{c_1 x_1}{c_1 x_1 + c_2 x_2}, \quad \theta_2 = 1 - \theta_1 \quad (13)$$

In a binary system, the parameters are c^0 and c^∞ as shown in Eq. (10), and q and $-\Delta E_{ij}$ are not separated and do not appear directly. On the other hand, in a multicomponent system with three or more components, q and $-\Delta E_{ij}$ become independent parameters. This model can correlate phase equilibria of binary systems very well, and it is also shown that ternary liquid-liquid equilibria and constituent binary vapor-liquid equilibria can be correlated well with the same

set of parameters [4,5]. However, since Eq. (3) is used as the surface area (number of interaction pairs) q_i , the same value of q_i^0 is assumed to be used for component i in a multicomponent systems, which reduces the degrees of freedom.

4. Model proposed in this study (p-CDSAP model)

The p-CDSAP model proposed in this study is based on r-CDSAP model with the following modifications.

4.1 Examination of q_i^0

r-CDSAP model is obtained by Taylor expansion of CDSAP model and approximating it to the second term. Therefore, when one apply it to the real systems and determine the parameters, one finds that q_i^0 changes for systems with large $-\Delta E_{ij}$. The parameter values of are examined with the literature data [9,10] of the vapor-liquid equilibria for ethanol (1) + acetone (2) + hexane (3) at 55°C. The parameters of CDSAP model to fit the activity coefficients of the ternary system and its binary systems are shown in Table 1. As can be seen in Table 1, the values of $-\Delta E_{ij}$ increase in the order of $-\Delta E_{12} < -\Delta E_{23} < -\Delta E_{13}$ due to the polarity difference between the components in each system, and the calculated results by CDSAP are shown in green lines in Fig. 1. As shown by the blue lines in Fig. 1(a), the deviation is negligible when the activity coefficients are calculated by r-CDSAP model using the parameter values obtained by CDSAP for ethanol(1) + acetone(2) system. However, as shown by the blue lines in Fig. 1(b), the deviation in acetone (2) + hexane (3) system becomes large when the activity coefficients are calculated by r-CDSAP model using the parameter values obtained by CDSAP, and as shown by the blue lines in Fig. 1(c), the deviation becomes extremely large in ethanol (1) + hexane (3) system. Since $\ln \gamma_i^\infty = c_{ji}^\infty = q_{ji}^\infty \times -\Delta E_{ij}$ for both CDSAP and r-CDSAP models, the values of q_{ji}^∞ , q_{ij}^∞ , and $-\Delta E_{ij}$ are fixed. Then, the parameters of r-CDSAP model q_i^0 are obtained to fit the experimental values of each binary system. Table 2 shows the parameters of r-CDSAP model obtained to fit the experimental data of each binary system. The calculated results of the activity coefficients using these values are shown in Fig. 1 as red lines. The values of q_i^0 are different for each binary system, although the experimental values are reproduced well. This means that the value of q_i^0 needs to be changed according to the partner component of binary system. Therefore, the author decided that the value of q_i^0 can be different for each binary system. The parameters c_{21}^0 , c_{12}^0 , c_{21}^∞ , and c_{12}^∞ which are obtained for binary system are also used for multicomponent systems. The restriction condition for c^0 in multicomponent system ($c_{21}^0 c_{32}^0$

$c_{13}^0 = c_{12}^0 c_{23}^0 c_{31}^0$), which is used in r-CDSAP model, is no used in p-CDSAP model.

Table 1

Parameter values of CDSAP model for ethanol (1) + acetone (2) + hexane (3) at 55°C.

i	j	q_i^0	q_{ji}^∞	q_{ij}^∞	$-\Delta E_{ij}$
1	2	1.000	1.136	1.269	0.507
2	3	1.356	1.093	1.291	1.364
3	1	1.738	1.210	0.988	2.576

Table 2

Parameter values of r-CDSAP model determined by sub-binary systems of ethanol (1) + acetone (2) + hexane (3) at 55°C.

i	j	q_i^0	q_j^0	$-\Delta E_{ij}$
1	2	1.263	0.614	0.507
2	3	0.414	3.398	1.364
3	1	0.679	0.503	2.576

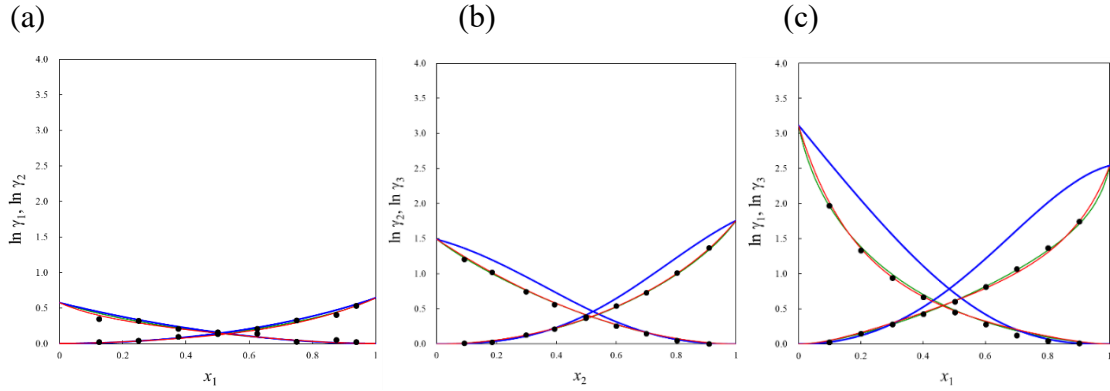


Fig. 1. Activity coefficients for sub-binary systems for ethanol(1) + acetone(2) + hexane(3) at 55°C. (a): ethanol(1) + acetone(2), (b): acetone(2) + hexane(3), (c): ethanol(1) + hexane(3). Black circles: Experimental data, (a): [9], (b): [10], (c): [10]. Green lines: CDSAP calculated with parameters in Table 1, blue lines: r-CDSAP calculated with parameters in Table 1, red lines: r-CDSAP calculated with parameters in Table 2.

4.2 Values of c_{ji}^0 and c_{ij}^0

As mentioned in the previous paper [4], when trying to find the values of c_{ji}^0 and c_{ij}^0 to fit

the experimental data of $(i) + (j)$ binary system, the deviation with respect to the experimental data may be almost the same even if the values of c_{ji}^0 and c_{ij}^0 change over a wide range. This is advantageous when r-CDSAP model is used for correlations, because it allows a large degree of freedom in the selection of parameter values, but it is disadvantageous when it is used for the estimations of multicomponent systems. Therefore, when calculating c_{ji}^0 and c_{ij}^0 for each binary system, one can consider that q is large for a component with a large number of interaction pairs, whether infinite dilution or pure component. Similarly, q is small for a component with a small number of interaction pairs, whether infinite dilution or pure component. Since $c = q \times -\Delta E_{ij}$, the following limiting condition is assumed.

$$\frac{c_{ji}^0}{c_{ji}^\infty} = \frac{c_{ij}^0}{c_{ij}^\infty} \quad (\because \frac{q_i^0}{q_{ji}^\infty} = \frac{q_j^0}{q_{ij}^\infty}) \quad (14)$$

Table 3 lists the parameters of ethanol (1) + acetone (2) + hexane (3) at 55°C when the limiting condition is applied. The correlated results are good as shown in Figure 2.

Table 3

Parameter values of p-CDSAP model determined by sub-binary systems of ethanol (1) + acetone (2) + hexane (3) at 55°C.

i	j	c_{ji}^0	c_{ij}^0	c_{ji}^∞	c_{ij}^∞
1	2	0.477	0.564	0.519	0.614
2	3	1.454	1.645	1.472	1.666
3	1	1.648	2.195	2.151	2.867

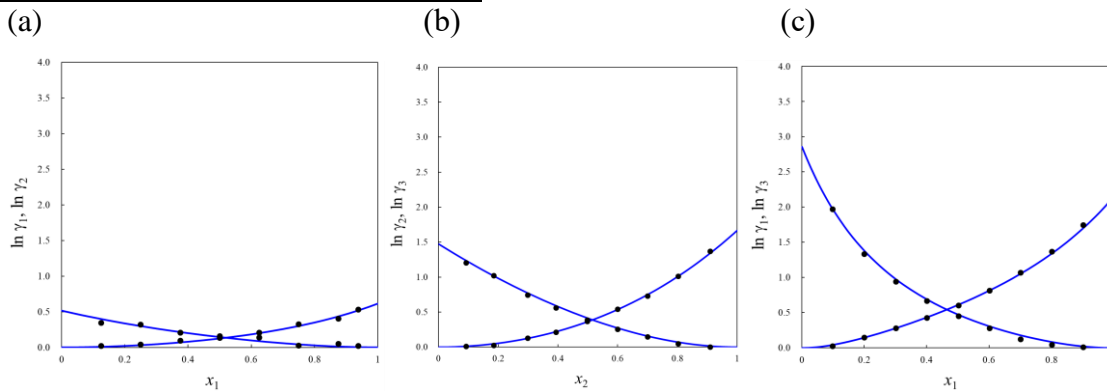


Fig. 2. Activity coefficients for sub-binary systems for ethanol(1) + acetone(2) + hexane(3) at 55°C. (a): Systems and data sources are same in Fig. 1. Blue lines: p-CDSAP calculated with parameters in Table 3.

4.3 Calculations of $-\Delta E_{ij}$

In a binary system, the parameters are c as mentioned in section 3, and q and $-\Delta E_{ij}$ are not separated and do not appear directly. On the other hand, q and $-\Delta E_{ij}$ need to be separated and calculated for multicomponent systems. The value of $-\Delta E_{ij}$ becomes larger as the polarity difference between the components increases in each system, and the value of the infinite dilution activity coefficient also increases accordingly. In a binary system, c are the essential parameters, and $c = q \times -\Delta E_{ij}$. Therefore, once the value of $-\Delta E_{ij}$ is determined, the values of q can be obtained from $q = -\Delta E_{ij} / c$. Also, since q is the number of interaction pairs per molecule, it does not seem to depend much on size of molecule. Therefore, $-\Delta E_{ij}$ is assumed to be obtained by the following equation.

$$-\Delta E_{ij} = (c_{ji}^{\infty} + c_{ij}^{\infty})/2 \quad (15)$$

The values of q are calculated by the following equations.

$$q_i^0 = -\Delta E_{ij} / c_{ji}^0 \quad (16)$$

$$q_{ji}^{\infty} = -\Delta E_{ij} / c_{ji}^{\infty} \quad (17)$$

According to this assumption, $q_{ji}^{\infty} + q_{ij}^{\infty} = 2$ for all binary systems, and the average value of q is 1. Also, the values of q^0 are close to 1.

4.4 Calculations of G^E

The excess Gibbs free energy G^E is given by Eq. (7). Here, the problem is how to obtain q^0 . The values of q_i^0 and q_j^0 can be obtained from $(i) + (j)$ system as shown in section 4.1. When calculating i, j term on the right-hand side of Eq. (7), one can use the values of q_i^0 and q_j^0 obtained from $(i) + (j)$ system. However, q_k^0 ($k \neq i, j$), which is not related to i and j , cannot be obtained from $(i) + (j)$ system. Therefore, the average value of 1.0 is assumed to use for q_k^0 ($k \neq i, j$). For example, in a ternary system, $q_1 = q_1^0 x_1 + q_{21}^{\infty} x_2 + q_{31}^{\infty} x_3$, $q_2 = q_2^0 x_2 + q_{12}^{\infty} x_1 + q_{32}^{\infty} x_3$, and $q_3 = q_3^0 x_3 + q_{13}^{\infty} x_1 + q_{23}^{\infty} x_2$ from Eq. (3). When calculating the term of $i=1, j=2$ on the right-hand side of Eq. (7), the values of q_1^0 and q_2^0 which are obtained from $(1) + (2)$ system are used, and $q_3^0 = 1.0$ since the value cannot be obtained from $(1) + (2)$ system. Similarly, when calculating the term of $i=1$ and $j=3$ on the right-hand side of Eq. (7), the values of q_1^0 and q_3^0 which are obtained from $(1) + (3)$ binary system are used, and $q_2^0 = 1.0$.

4.5 General expression of activity coefficients

The activity coefficient γ_m of component m is expressed by the following equations.

$$\begin{aligned} \ln \gamma_m &= \left(\partial \frac{G^E}{RT} / \partial n_m \right)_{T,p,n_{n \neq m}} \\ &= \sum_i \sum_{j \neq i} \left[-\Delta E_{ij} \{ Q_{i,m} \theta_j + \theta_i Q_{j,m} - (\theta_i \theta_j) \sum_k Q_{k,m} \} \right] \quad (18) \end{aligned}$$

$$Q_{i,i} = q_i + x_i(q_i^0 - q_i) \quad (19)$$

$$Q_{i,m} = x_i(q_{mi}^\infty - q_i) \quad \text{where } i \neq m \quad (20)$$

The role for q^0 mentioned in section 4.4 should be applied to calculate the activity coefficients.

5. Application to real systems

5.1 Applied systems and data sources

The vapor-liquid equilibria for acetone (1) + acetonitrile (2) + benzene (3) + ethanol (4) system were chosen to evaluate the applicability of p-CDSAP model. Because the vapor-liquid equilibrium data for the sub-binary and sub-ternary systems are available in the literature. Also, the size of each component is almost the same, and the polarity of each component varies. Table 4 shows the temperatures of the sub-binary, sub-ternary, and quaternary systems and the data sources. The non-ideality in vapor phases were evaluated with the second virial coefficients, when the activity coefficients were obtained from the literature data of vapor-liquid equilibria. The second virial coefficients were calculated by Hayden and O'Connell method [20].

Table 4

Data sources of acetone (1) + acetonitrile (2) + benzene (3) + ethanol (4) and sub-binary and ternary systems.

System	T(K)	Ref.
(1)+(2)	318	[11]
(1)+(3)	318	[12]
(1)+(4)	328	[9]
(2)+(3)	318	[13]
(2)+(4)	313	[14]
(3)+(4)	318	[15]
(1)+(2)+(3)	318	[16]
(1)+(2)+(4)	318	[17]
(2)+(3)+(4)	318	[18]
(1)+(2)+(3)+(4)	318	[19]

5.2 Correlations of binary systems

5.2.1 p-CDSAP model

The parameters of c_{ji}^0 , c_{ij}^0 , c_{ji}^∞ , and c_{ij}^∞ in p-CDSAP model were optimized by Marquardt method with the conditions shown in Eq. (14) for each binary system. The objective function F is as follow.

$$F = \frac{\sum_{i=1}^{N_{\text{VLE}}} \sum_{j=1}^2 \left(\left| \gamma_i^{\text{exp}} - \gamma_i^{\text{calc}} \right| / \gamma_i^{\text{exp}} \right)}{2N_{\text{VLE}}} \times 100 \quad (21)$$

where N_{VLE} is the number of data. Table 5 lists the values of the binary parameters determined. The values of $-\Delta E_{ij}$ which are calculated by Eq.(15), and q^0 and q^∞ which are calculated by Eqs. (16) and (17) are also listed in Table 5. These values are not used for the calculations for the binary systems, however, are used for the calculations for multicomponent systems.

Table 5

Parameter values of p-CDSAP model determined by sub-binary systems for acetone (1) + acetonitrile (2) + benzene (3) + ethanol (4) system.

i	j	c_{ji}^0	c_{ij}^0	c_{ji}^∞	c_{ij}^∞	$-\Delta E_{ij}$ $=(c_{ji}^\infty + c_{ij}^\infty)/2$	q_{ji}^0	q_{ij}^0	q_{ji}^∞	q_{ij}^∞
1	2	0.054	0.029	0.073	0.040	0.0565	0.956	0.519	1.295	0.703
1	3	0.452	0.362	0.510	0.408	0.459	0.985	0.789	1.111	0.889
1	4	0.564	0.477	0.617	0.522	0.5695	0.990	0.838	1.083	0.917
2	3	0.858	0.782	1.244	1.135	1.1895	0.721	0.658	1.046	0.954
2	4	1.242	1.118	1.360	1.224	1.292	0.961	0.865	1.053	0.947
3	4	1.220	1.943	1.549	2.467	2.008	0.607	0.967	0.771	1.229

5.2.2 NRTL model

NRTL model [1] was also used to correlate the binary systems for comparison. The values of α were fixed to 0.2, 0.3, or 0.4. The value of α was also optimized for each binary system. The optimized method and the objective function are the same as for p-CDSAP model. The parameter values for the sub-binary systems are listed in Table 6.

Table 6

Parameter values of NRTL model determined by sub-binary systems for acetone (1) + acetonitrile (2) + benzene (3) + ethanol (4) system.

		$\alpha=0.2$		$\alpha=0.3$		$\alpha=0.4$		$\alpha=\text{optimized}$		
i	j	τ_{ji}	τ_{ij}	τ_{ji}	τ_{ij}	τ_{ji}	τ_{ij}	τ_{ji}	τ_{ij}	α
1	2	0.8201	-0.6634	0.6188	-0.4813	0.5065	-0.3809	0.0596	0.0222	20.1791
1	3	0.7943	-0.2866	0.6164	-0.1209	0.5356	-0.0433	0.3622	0.2090	1.6803
1	4	0.7540	-0.1519	0.6062	-0.0082	0.5333	0.0673	0.4227	0.2695	1.2226
2	3	0.7674	0.3579	0.7195	0.4399	0.7075	0.4923	0.8860	0.7679	1.0031
2	4	0.9042	0.4435	0.8566	0.5413	0.8545	0.6055	0.8943	0.6745	0.5446
3	4	-0.0133	2.2595	0.3612	1.9574	0.6000	1.8753	0.7919	0.4495	-1.1727

5.2.3 UNIQUAC model

The original UNIQUAC model [2] and a modified UNIQUAC model [21] were also used to correlate the binary systems. The parameter values of r and q are cited from Gmehling and Onken [22]. Those are listed in Table 7. The parameter value of q' for ethanol in the modified UNIQUAC model is cited from Anderson and Prausnitz [21]. The optimized method and the objective function are the same as for p-CDSAP model. The parameter values for the sub-binary systems are listed in Table 8.

Table 7

Parameter values of UNIQUAC models [22].

component	r	q
acetone	2.574	2.336
acetonitrile	1.870	1.724
benzene	3.188	2.400
ethanol	2.106	1.972

q' for ethanol is 0.92 in modified UNIQUAC [21].

Table 8

Parameter values of UNIQUAC models determined by sub-binary systems for acetone (1) + acetonitrile (2) + benzene (3) + ethanol (4) system.

		Original		Modified	
<i>i</i>	<i>j</i>	a_{ij} (J mol ⁻¹)	a_{ji} (J mol ⁻¹)	a_{ij} (J mol ⁻¹)	a_{ji} (J mol ⁻¹)
1	2	-243.09	390.65	-243.09	390.65
1	3	-885.98	1424.86	-885.98	1424.86
1	4	329.03	408.13	2739.98	-1011.75
2	3	-438.95	2125.33	-438.95	2125.33
2	4	373.64	1653.95	3314.47	166.50
3	4	3610.55	-588.70	8386.88	-1087.98

5.2.4 Correlated results

Figure 3 shows the deviations. The deviations are almost the same by all models for acetone (1) + acetonitrile (2), acetone (1) + benzene (3), acetone (1) + ethanol (4), and acetonitrile (2) + ethanol (4) systems. For acetonitrile (2) + benzene (3) system, the deviations by p-CDSAP model and NRTL model with optimized parameter α are almost the same, and those by NRTL model with fixed values of α and by UNIQUAC models are larger. For benzene (3) + ethanol (4) system, the deviations by p-CDSAP model and NRTL model with optimized parameter α are almost the same, and those by NRTL model with fixed values of α and by the original UNIQUAC model are larger. On the other hand, the modified UNIQUAC has a smaller deviation in the system. Because the parameter values of q' in the modified UNIQUAC were optimized for the systems containing alcohols.

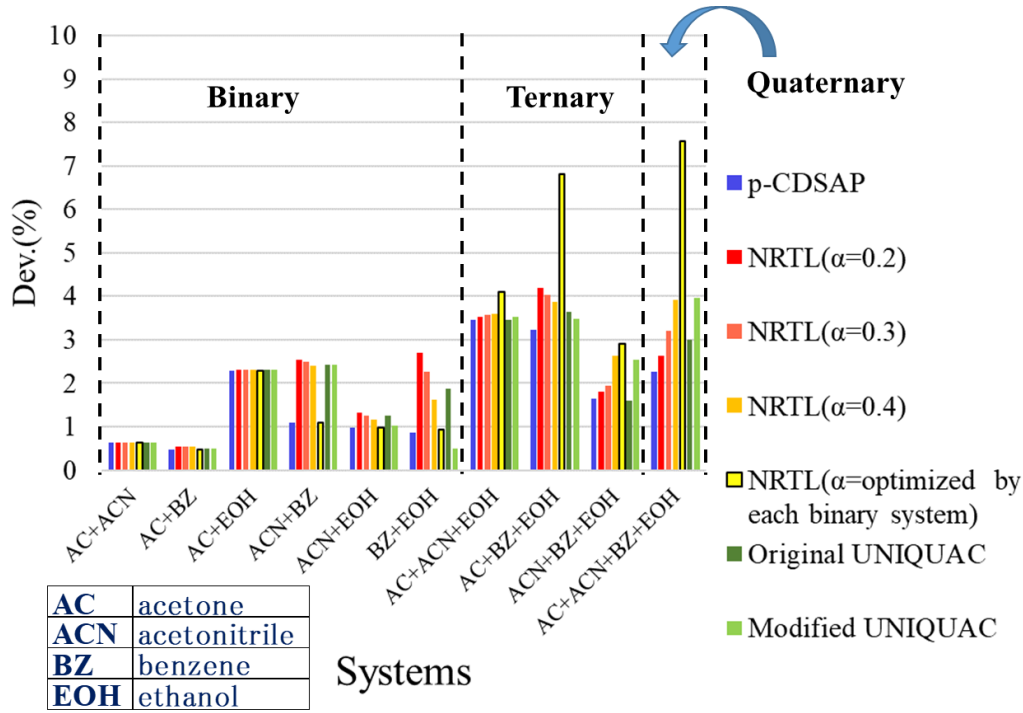


Fig.3. Deviations for acetone (1) + acetonitrile (2) + benzene (3) + ethanol (4) and sub-binary and ternary systems calculated by p-CDSAP, NRTL and UNIQUAC models.

$$\text{Dev. (\%)} = \frac{\sum |\gamma^{\text{exp}} - \gamma^{\text{calc}}| / \gamma^{\text{exp}}}{\text{No.data of } \gamma} \times 100$$

5.3 Estimations for multicomponent systems

The parameters determined by the binary systems are used for the estimations of the ternary and quaternary systems. The estimated results by p-CDSAP model are comparable or slightly better than those by NRTL and UNIQUAC models. The estimated results by NRTL model with optimized α by binary systems do not give better results for the ternary and quaternary systems. Similar trend is observed for the modified UNIQUAC. The parameter values of q' were modified to fit well for the systems containing alcohols, however, the estimated results for the multicomponent systems are not necessarily improved.

6. Conclusion

p-CDSAP model is proposed in this work. The activity coefficients of multicomponent systems can be estimated using the parameters obtained from the activity coefficients of binary systems by p-CDSAP model. The estimated results by p-CDSAP model are comparable or slightly better than those of NRTL and UNIQUAC models. However, various assumptions are used in deriving the model, but it is not clear whether the assumptions are optimal or not. In addition, it is necessary to apply p-CDSAP model for many systems in the future.

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