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Correlation of quaternary liquid–liquid equilibria using a modified form of the Wilson equation

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Abstract

A modified three-parameter Wilson equation, proposed for binary partially miscible mixtures, is extended to correlate quaternary liquid–liquid equilibria. The proposed form of the modified Wilson equation involves binary, ternary and quaternary parameters and gives good calculated results comparable to those derived from the modified Hiranuma–Wilson equation, which has four parameters for binary partially miscible mixtures.

Keywords: Binary system; LLE; Modified Wilson equation; Quaternary system; Ternary system; VLE

List of symbols

a_{ij}	molar energy parameter of Wilson equation for i–j interaction
b_{ij}	binary parameter of modified Wilson equation for i–j pair
F	objective function given by Eq. (8)
g^{E}	molar excess Gibbs free energy
n_i	number of moles of component i
n_{T}	total number of components
R	universal gas constant
T	absolute temperature
x_i	liquid mole fraction of component i
V_i	liquid molar volume of pure component i

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Greek letters

γ_i	activity coefficient of component *i*
Λ_{ij}, Λ_{jki}, Λ_{jkli}	binary, ternary and quaternary parameters

1. Introduction

Among several forms of the Wilson equation modified for binary partially miscible mixtures [1], Novák et al. [2] proposed the following form

$$g^{E}/R = -x_1\ln(x_1 + \Lambda_{12}x_2) - x_2\ln(x_2 + \Lambda_{21}x_1) + b_{12}x_1x_2 \tag{1}$$

where b_{12} is the empirical parameter, and the binary parameter Λ_{ij} is expressed in terms of molar energy parameter a_{ij} and the molar volumes of pure components, V_i and V_j.

$$\Lambda_{ij} = (V_j/V_i)\exp(-a_{ij}/T) \tag{2}$$

A new ternary extended form of Eq. (1), which is not identical with the ternary modification proposed by Novák et al. [3], has shown good applicability in the correlation of ternary liquid–liquid equilibria (LLE) [4].

In this paper, the ternary modified form of Eq. (1) is further extended for the correlation of quaternary LLE.

2. Proposed model

The excess molar Gibbs free energy for a quaternary mixture is expressed by

$$\begin{aligned}
g^{E}/RT = {} & -\sum_{i=1}^{4} x_i \ln\left(\sum_{j=1}^{4}\Lambda_{ij}x_j + \frac{1}{2}\sum_{j=1}^{4}\sum_{k=1,j\neq k\neq i}^{4}\Lambda_{jki}x_jx_k\right.\\
& \left. + \frac{1}{6}\sum_{j=1}^{4}\sum_{k=1}^{4}\sum_{l=1,j\neq k\neq l\neq i}^{4}\Lambda_{jkli}x_jx_kx_l\right) + \frac{1}{2}\sum_{i=1}^{4}\sum_{j=1}^{4} b_{ij}x_ix_j\\
= {} & -x_1\ln(x_1 + \Lambda_{12}x_2 + \Lambda_{13}x_3 + \Lambda_{14}x_4 + \Lambda_{231}x_2x_3\\
& + \Lambda_{241}x_2x_4 + \Lambda_{341}x_3x_4 + \Lambda_{2341}x_2x_3x_4) - x_2\ln(\Lambda_{21}x_1 + x_2 + \Lambda_{23}x_3\\
& + \Lambda_{24}x_4 + \Lambda_{132}x_1x_3 + \Lambda_{142}x_1x_4 + \Lambda_{342}x_3x_4 + \Lambda_{1342}x_1x_3x_4)\\
& - x_3\ln(\Lambda_{31}x_1 + \Lambda_{32}x_2 + x_3 + \Lambda_{34}x_4 + \Lambda_{123}x_1x_2 + \Lambda_{143}x_1x_4\\
& + \Lambda_{243}x_2x_4 + \Lambda_{1243}x_1x_2x_4) - x_4\ln(\Lambda_{41}x_1 + \Lambda_{42}x_2 + \Lambda_{43}x_3 + x_4\\
& + \Lambda_{124}x_1x_2 + \Lambda_{134}x_1x_3 + \Lambda_{234}x_2x_3 + \Lambda_{1234}x_1x_2x_3) + b_{12}x_1x_2\\
& + b_{13}x_1x_3 + b_{14}x_1x_4 + b_{23}x_2x_3 + b_{24}x_2x_4 + b_{34}x_3x_4
\end{aligned} \tag{3}$$

where $\Lambda_{ii} = 1$, $\Lambda_{ij} \neq \Lambda_{ji}$, and $\Lambda_{jki}(=\Lambda_{kji})$ and $\Lambda_{jkli}(=\Lambda_{jlki} = \Lambda_{kjli} = \Lambda_{klji} = \Lambda_{ljki} = \Lambda_{lkji})$ are the ternary and quaternary parameters. Eq. (3) can be derived in the same manner as described previously [5].

Table 1
Binary parameters

System (1 + 2)	Temp./°C	a_{12}/K	a_{21}/K	b_{12}	Ref.
Acetic acid + 1-butanol	115.7–120.3	58.80	−68.03		[6]
Acetic acid + *n*-butyl acetate	117.7–125.2	591.12	−343.42		[7]
Acetic acid + chloroform	61.4–118.1	472.71	−34.81		[7]
Acetone + acetic acid	35.0	−242.83	242.83		[8]
Acetone + 1-butanol	58.1–115.0	311.54	−54.97		[6]
Acetone + 2-butanone	56.9–78.5	−117.21	239.97		[8]
Acetone + chloroform	35.0	−31.10	−217.38		[8]
Acetone + 1-propanol	62.9–86.6	106.61	90.01		[9]
Acetonitrile + benzene	45.0	392.49	−7.60		[10]
Acetonitrile + ethanol	20.0	306.47	221.38		[11]
Acetonitrile + methanol	30.0	21.43	335.80		[12]
Acetonitrile + 1-propanol	55.0	407.39	84.78		[13]
Acetonitrile + 2-propanol	50.0	945.69	−26.29		[14]
Benzene + cyclohexane	40.0	85.77	67.31		[15]
1-Butanol + 2-butanone	80.7–111.2	31.52	139.17		[16]
1-Butanol + *n*-butyl acetate	116.8–121.2	96.72	109.66		[6]
1-Butanol + chloroform	62.6–115.1	426.69	−76.58		[6]
Chloroform + toluene	61.6–108.1	−814.25	277.85		[10]
Ethanol + 1-butanol	80.0–115.0	−4.22	61.83		[12]
Ethanol + 2-butanone	55.0	289.87	−8.59		[12]
Ethanol + chloroform	35.0	878.70	−195.51		[12]
Ethanol + cyclohexane	25.0	1017.2	262.62		[11]
Methanol + benzene	35.0	899.85	92.17		[12]
Methanol + ethanol	25.0	41.72	−45.18		[11]
Methanol + 1-propanol	40.0	75.32	−28.00		[13]
Methanol + 2-propanol	55.0	627.74	−451.28		[12]
1-Propanol + 1-butanol	40.0	71.19	−60.73		[13]
1-Propanol + 2-butanone	79.9–94.8	150.65	36.33		[11]
1-Propanol + cyclohexane	25.0	866.91	194.53		[11]
2-Propanol + cyclohexane	60.0	797.96	142.66		[6]
Water + acetic acid	100.3–115.2	403.09	3.57		[17]
Water + acetone	25.0	796.10	−54.01		[18]
Water + ethanol	25.0	440.56	27.73		[17]
Water + 1-propanol	94.8–89.3	639.20	814.60		[17]
Acetonitrile + cyclohexane	25.0	−12.37	−74.20	3.7668	[19]
Methanol + cyclohexane	25.0	1085.1	358.56	0.1498	[20]
Water + 1-butanol	25.0	571.39	889.65	0.5618	[21]
Water + 2-butanone	25.0	343.33	354.39	1.5798	[22]
Water + *n*-butyl acetate	25.0	878.25	907.12	1.4855	[21]
Water + chloroform	25.0	269.45	365.59	4.8870	[23]
Water + toluene	25.0	819.78	862.37	3.9156	[24]

The activity coefficient of component *i* in a quaternary mixture is derived from

$$\ln \gamma_i = \frac{1}{RT}\left(\frac{\partial n_T g^E}{\partial n_i}\right)_{P,T,n_{j \neq i}} \tag{4}$$

Table 2
Calculated results of ternary liquid–liquid equilibria at 25°C

No.	System (1 + 2 + 3)	Number of tie-lines	Ternary parameters	Root-mean squared deviations/(mol%)		Ref.
				I[a]	II[b]	
1	Acetonitrile + benzene + cyclohexane	6	$\Lambda_{231} = -0.4087$ $\Lambda_{132} = 6.2405$ $\Lambda_{123} = -1.1275$	3.83	1.01	[26]
2	Acetonitrile + ethanol + cyclohexane	13	$\Lambda_{231} = -2.9940$ $\Lambda_{132} = 0.3476$ $\Lambda_{123} = 1.7226$	4.85	0.75	[27]
3	Acetonitrile + methanol + cyclohexane	7	$\Lambda_{231} = -0.4771$ $\Lambda_{132} = -0.2378$ $\Lambda_{123} = -0.0524$	1.57	0.43	[28]
4	Acetonitrile + 1-propanol + cyclohexane	8	$\Lambda_{231} = 0.5785$ $\Lambda_{132} = 0.1551$ $\Lambda_{123} = -0.0925$	6.09	0.37	[29]
5	Acetonitrile + 2-propanol + cyclohexane	6	$\Lambda_{231} = -0.1216$ $\Lambda_{132} = 0.6615$ $\Lambda_{123} = 0.2473$	0.89	0.53	[30]
6	Methanol + benzene + cyclohexane	6	$\Lambda_{231} = 0.1847$ $\Lambda_{132} = 0.5128$ $\Lambda_{123} = -0.0502$	3.07	0.27	[26]
7	Methanol + ethanol + cyclohexane	7	$\Lambda_{231} = 0.2847$ $\Lambda_{132} = -0.0058$ $\Lambda_{123} = -0.2095$	6.63	0.23	[26]
8	Methanol + 1-propanol + cyclohexane	4	$\Lambda_{231} = 4.1791$ $\Lambda_{132} = 1.8617$ $\Lambda_{123} = -1.9982$	5.07	0.52	[31]

9	Methanol + 2-propanol + cyclohexane	7	$\Lambda_{231} = 0.3108$ $\Lambda_{132} = 0.3926$ $\Lambda_{123} = -0.2054$	4.54	0.51	[26]
10	Water + acetic acid + 1-butanol	15	$\Lambda_{231} = 0.6308$ $\Lambda_{132} = 0.7506$ $\Lambda_{123} = -0.5672$	2.29	0.44	[26]
11	Water + acetic acid + *n*-butyl acetate	15	$\Lambda_{231} = 1.6322$ $\Lambda_{132} = 0.7901$ $\Lambda_{123} = -0.5663$	9.15	1.29	[26]
12	Water + acetic acid + chloroform	8	$\Lambda_{231} = 0.8565$ $\Lambda_{132} = 2.8658$ $\Lambda_{123} = -0.5166$	2.45	0.37	[26]
13	Water + acetone + 1-butanol	5	$\Lambda_{231} = -1.4062$ $\Lambda_{132} = 2.7254$ $\Lambda_{123} = -1.1677$	4.87	0.12	[26]
14	Water + acetone + 2-butanone	7	$\Lambda_{231} = -0.1329$ $\Lambda_{132} = 1.3673$ $\Lambda_{123} = -0.0504$	2.46	0.31	[26]
15	Water + acetone + chloroform	8	$\Lambda_{231} = 0.1246$ $\Lambda_{132} = 2.6798$ $\Lambda_{123} = -1.6377$	1.41	0.24	[26]
16	Water + 1-butanol + *n*-butyl acetate	4	$\Lambda_{231} = 0.8350$ $\Lambda_{132} = 0.0917$ $\Lambda_{123} = 1.1953$	6.11	0.66	[26]
17	Water + 1-butanol + chloroform	4	$\Lambda_{231} = -0.2990$ $\Lambda_{132} = 0.9789$ $\Lambda_{123} = 3.9884$	5.02	0.41	[26]
18	Water + 2-butanone + 1-butanol	4	$\Lambda_{231} = 0.1012$ $\Lambda_{132} = 1.2869$ $\Lambda_{123} = -0.4125$	8.07	1.62	[32]
19	Water + chloroform + toluene	4	$\Lambda_{231} = -1.5527$ $\Lambda_{132} = 0.9985$ $\Lambda_{123} = -0.2304$	0.08	0.01	[24]

Table 2 (*Continued*)

No.	System (1+2+3)	Number of tie-lines	Ternary parameters	Root-mean squared deviations/(mol%)		Ref.
				I[a]	II[b]	
20	Water+ethanol+1-butanol	10	$\Lambda_{231} = -0.4890$ $\Lambda_{132} = 1.0116$ $\Lambda_{123} = -0.6118$	2.02	0.12	[26]
21	Water+ethanol+2-butanone	3	$\Lambda_{231} = 3.1496$ $\Lambda_{132} = 0.2609$ $\Lambda_{123} = 0.0103$	2.95	0.22	[32]
22	Water+ethanol+chloroform	15	$\Lambda_{231} = 2.1510$ $\Lambda_{132} = 2.1746$ $\Lambda_{123} = -0.2054$	10.04	0.63	[26]
23	Water+1-propanol+1-butanol	9	$\Lambda_{231} = 0.1010$ $\Lambda_{132} = 0.1299$ $\Lambda_{123} = -1.1837$	1.02	0.30	[26]
24	Water+1-propanol+2-butanone	3	$\Lambda_{231} = 1.0041$ $\Lambda_{132} = 3.0677$ $\Lambda_{123} = -0.2791$	3.21	0.15	[26]

[a] Predicted results based on binary parameters. [b] Correlated results based on binary and ternary parameters.

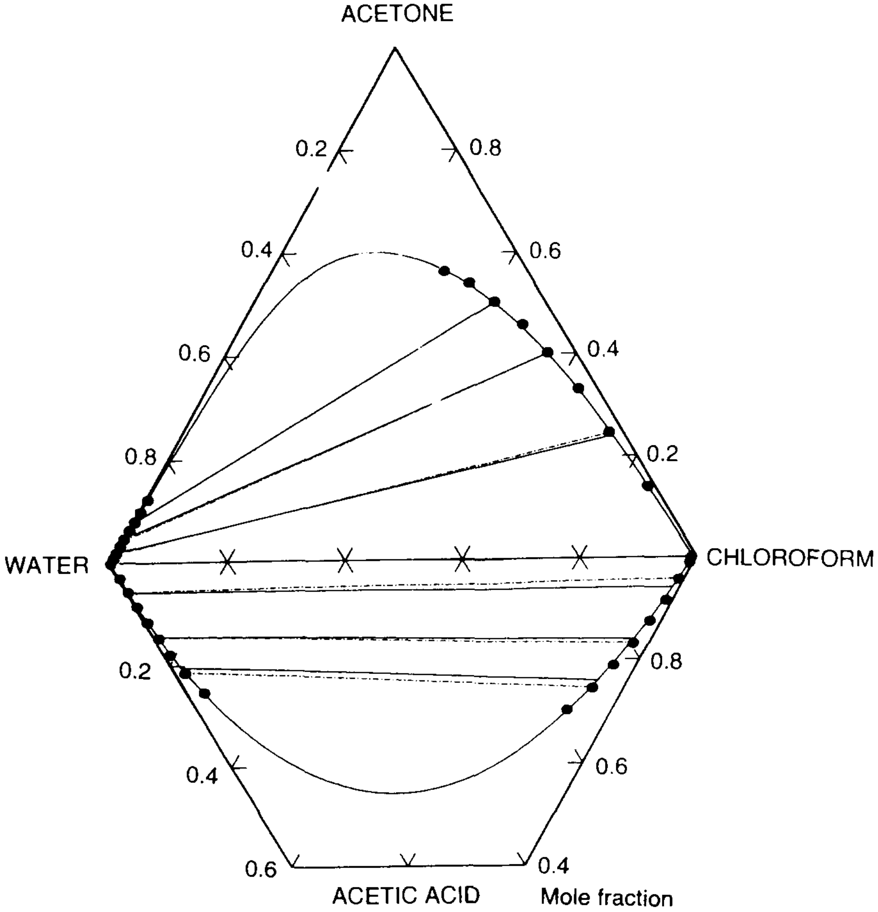

Fig. 1. Representation of ternary liquid–liquid equilibria using the modified Wilson model as obtained by incorporating ternary parameters: —●—, experimental: water + acetone + chloroform [26]; water + acetic acid + chloroform [26]. ——, calculated with binary and ternary parameters.

Then, $\ln \gamma_1$ is given by

$$\ln \gamma_1 = -\ln\left(\sum_{i=1}^{4} \Lambda_{1i}x_i + \Lambda_{231}x_2x_3 + \Lambda_{241}x_2x_4 + \Lambda_{341}x_3x_4 + \Lambda_{2341}x_2x_3x_4\right)$$

$$+ 1 - x_1\left(\frac{1 - \Lambda_{231}x_2x_3 - \Lambda_{241}x_2x_4 - \Lambda_{341}x_3x_4 - 2\Lambda_{2341}x_2x_3x_4}{\sum_{i=1}^{4} \Lambda_{1i}x_i + \Lambda_{231}x_2x_3 + \Lambda_{241}x_2x_4 + \Lambda_{341}x_3x_4 + \Lambda_{2341}x_2x_3x_4}\right)$$

$$- x_2\left(\frac{\Lambda_{21} + \Lambda_{132}(1 - x_1)x_3 + \Lambda_{142}(1 - x_1)x_4 - \Lambda_{342}x_3x_4 + \Lambda_{1342}(1 - x_1)x_3x_4}{\sum_{i=1}^{4} \Lambda_{2i}x_i + \Lambda_{132}x_1x_3 + \Lambda_{142}x_1x_4 + \Lambda_{342}x_3x_4 + \Lambda_{1342}x_1x_3x_4}\right)$$

$$-x_3\left(\frac{\Lambda_{31}+\Lambda_{123}(1-x_1)x_2+\Lambda_{143}(1-x_1)x_4-\Lambda_{243}x_2x_4+\Lambda_{1243}(1-x_1)x_2x_4}{\sum_{i=1}^{4}\Lambda_{3i}x_i+\Lambda_{123}x_1x_2+\Lambda_{143}x_1x_4+\Lambda_{243}x_2x_4+\Lambda_{1243}x_1x_2x_4}\right)$$

$$-x_4\left(\frac{\Lambda_{41}+\Lambda_{124}(1-x_1)x_2+\Lambda_{134}(1-x_1)x_3-\Lambda_{234}x_2x_3+\Lambda_{1234}(1-x_1)x_2x_3}{\sum_{i=1}^{4}\Lambda_{4i}x_i+\Lambda_{124}x_1x_2+\Lambda_{134}x_1x_3+\Lambda_{234}x_2x_3+\Lambda_{1234}x_1x_2x_3}\right)$$

$$+b_{12}(1-x_1)x_2+b_{13}(1-x_1)x_3+b_{14}(1-x_1)x_4-b_{23}x_2x_3-b_{24}x_2x_4-b_{34}x_3x_4 \tag{5}$$

3. Calculated results

3.1. Binary systems

Table 1 gives the binary parameters taken from vapor–liquid equilibrium data [6–18] and calculated from mutual solubility data [19–24]. Each set of the three binary parameters for partially miscible mixtures was solved by use of the following thermodynamic equation for each component of two equilibrium liquid phases

$$(x_i\gamma_i)^{\mathrm{I}}=(x_i\gamma_i)^{\mathrm{II}} \tag{6}$$

with

$$\sum_i x_i^{\mathrm{I}}=1 \quad \text{and} \quad \sum_i x_i^{\mathrm{II}}=1 \tag{7}$$

The binary parameters, which predicted LLE did not deviate largely from the experimental ternary tie-lines, were selected.

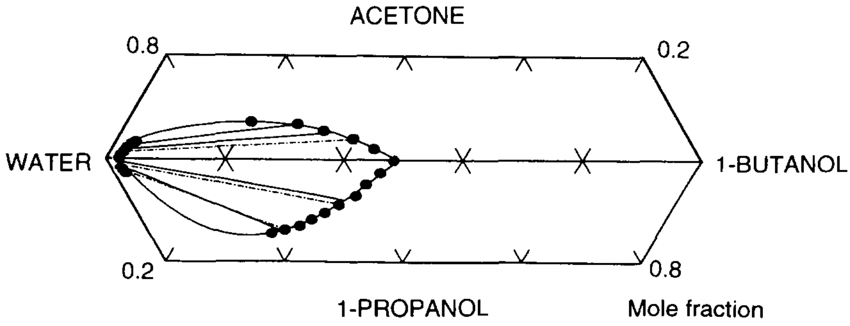

Fig. 2. Representation of ternary liquid–liquid equilibria using the modified Wilson model as obtained by incorporating ternary parameters: —●—, experimental: water + acetone + 1-butanol [26]; water + 1-propanol + 1-butanol [26]. ——, calculated with binary and ternary parameters.

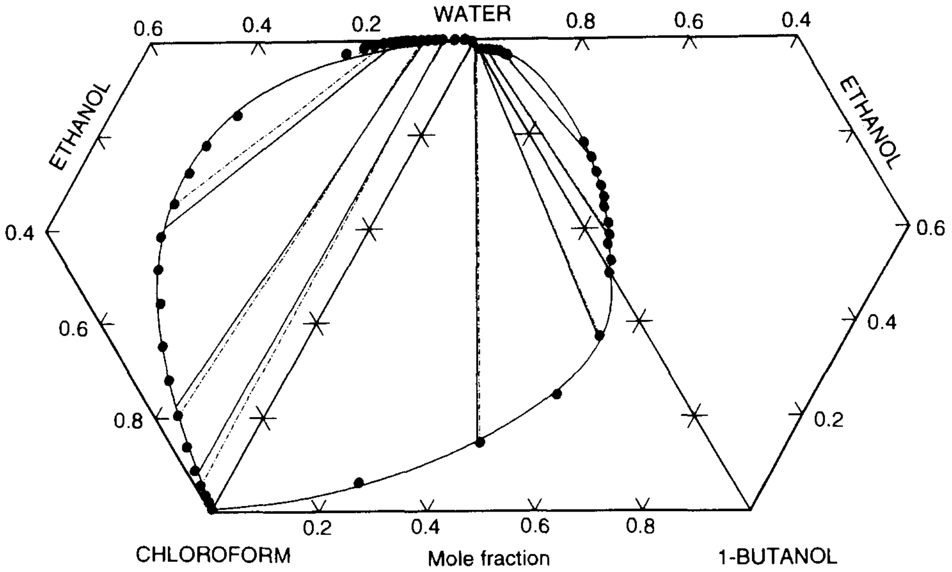

Fig. 3. Representation of ternary liquid–liquid equilibria using the modified Wilson model as obtained by incorporating ternary parameters: —●—, experimental: water + ethanol + 1-butanol [26]; water + ethanol + chloroform [26]; water + 1-butanol + chloroform [26]. ——, calculated with binary and ternary parameters.

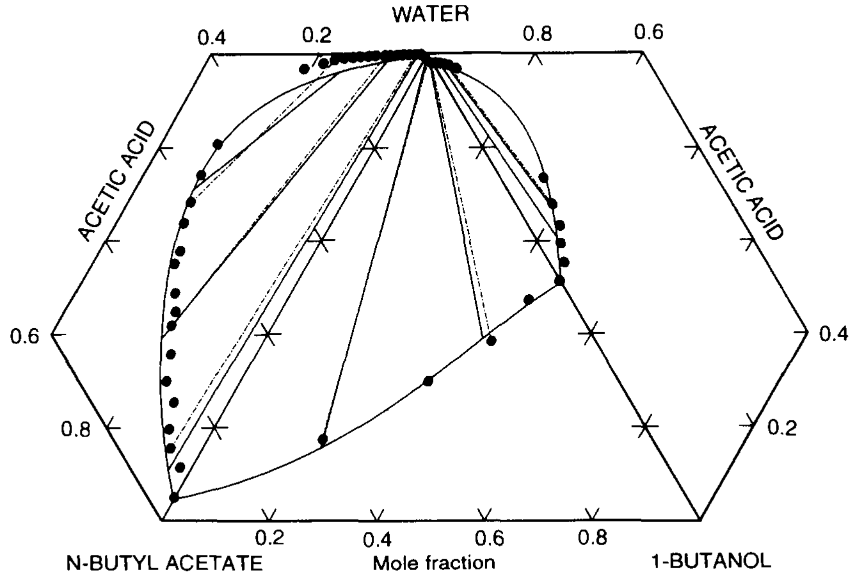

Fig. 4. Representation of ternary liquid–liquid equilibria using the modified Wilson model as obtained by incorporating ternary parameters: —●—, experimental: water + acetic acid + 1-butanol [26]; water + acetic acid + n-butyl acetate [26]; water + 1-butanol + n-butyl acetate [26]. ——, calculated with binary and ternary parameters.

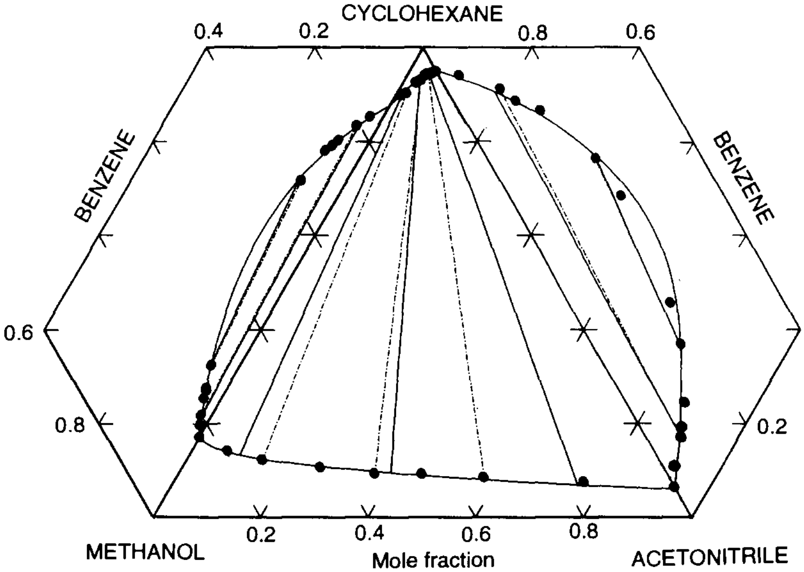

Fig. 5. Representation of ternary liquid–liquid equilibria using the modified Wilson model as obtained by incorporating ternary parameters: —●—, experimental: cyclohexane + benzene + acetonitrile [26]; cyclohexane + benzene + methanol [26]; cyclohexane + acetonitrile + methanol [28]. ——, calculated with binary and ternary parameters.

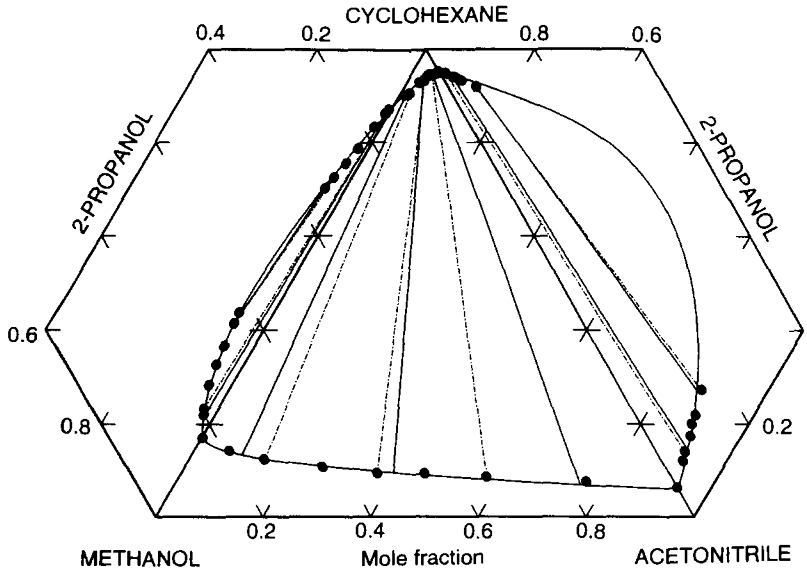

Fig. 6. Representation of ternary liquid–liquid equilibria using the modified Wilson model as obtained by incorporating ternary parameters: —●—, experimental: cyclohexane + 2-propanol + acetonitrile [30]; cyclohexane + 2-propanol + methanol [26]; cyclohexane + acetonitrile + methanol [28]. ——, calculated with binary and ternary parameters.

Table 3
Calculated results of quaternary liquid–liquid equilibria at 25°C

No.	System (1 + 2 + 3 + 4)	Number of tie-lines	Quaternary parameters	Root-mean squared deviations/(mol%)			Ref.
				I[a]	II[b]	III[c]	
1	Acetonitrile + methanol + benzene + cyclohexane	14	$\Lambda_{2341} = -0.2833$ $\Lambda_{1342} = -0.6980$ $\Lambda_{1243} = -13.6033$ $\Lambda_{1234} = 8.0985$	0.77	0.59		[30]
2	Acetonitrile + ethanol + methanol + cyclohexane	17	$\Lambda_{2341} = 0.1576$ $\Lambda_{1342} = 5.2490$ $\Lambda_{1243} = -2.5283$ $\Lambda_{1234} = -4.2013$	0.90	0.45		[33]
3	Acetonitrile + methanol + 1-propanol + cyclohexane	16	$\Lambda_{2341} = -10.8682$ $\Lambda_{1342} = -3.6025$ $\Lambda_{1243} = 6.1605$ $\Lambda_{1234} = 2.9527$	0.57	0.50		[33]
4	Acetonitrile + methanol + 2-propanol + cyclohexane	15	$\Lambda_{2341} = -4.6335$ $\Lambda_{1342} = 10.6945$ $\Lambda_{1243} = -11.9251$ $\Lambda_{1234} = -1.5668$	0.69	0.48		[30]
5	Water + acetone + acetic acid + chloroform	32	$\Lambda_{2341} = 26.6474$ $\Lambda_{1342} = -30.5533$ $\Lambda_{1243} = 8.5486$ $\Lambda_{1234} = 34.4903$	1.06	0.68	0.49	[26]
6	Water + acetone + 1-propanol + 1-butanol	26 24[d]	$\Lambda_{2341} = 0.4751$ $\Lambda_{1342} = 0.0930$ $\Lambda_{1243} = -0.1042$ $\Lambda_{1234} = 0.4513$	1.72 0.27	0.27	0.35	[26]

Table 3 (*Continued*)

No.	System (1 + 2 + 3 + 4)	Number of tie-lines	Quaternary parameters	Root-mean squared deviations/(mol%)			Ref.
				I[a]	II[b]	III[c]	
7	Water + acetone +	19	$\Lambda_{2341} = -25.3082$	2.13	1.23	0.77	[26]
	1-propanol + 2-butanone	16[d]	$\Lambda_{1342} = 21.1367$	0.47			
		18[d]	$\Lambda_{1243} = 22.2692$		0.60		
			$\Lambda_{1234} = 0.2288$				
8	Water + ethanol +	44	$\Lambda_{2341} = -8.6345$	2.41	0.95	1.19	[34]
	1-butanol + chloroform	41	$\Lambda_{1342} = 0.6061$	1.02			
		43	$\Lambda_{1243} = -3.1470$		0.66		
			$\Lambda_{1234} = 15.0766$				
9	Water + acetic acid +	40	$\Lambda_{2341} = -0.9124$	1.59	0.79	0.79	[21]
	1-butanol + *n*-butyl acetate	38	$\Lambda_{1342} = 2.6966$	0.74			
			$\Lambda_{1243} = -1.5271$				
			$\Lambda_{1234} = 0.1436$				
10	Water + ethanol +	11	$\Lambda_{2341} = 1.0161$	2.81	0.57	0.99	[32]
	2-butanone +	10	$\Lambda_{1342} = -19.6757$	1.76			
	1-butanol		$\Lambda_{1243} = 20.0962$				
			$\Lambda_{1234} = -1.4775$				

[a] Predicted results based on binary and ternary parameters. [b] Correlated results based on binary, ternary and quaternary parameters. [c] Correlated results based on the modified Hiranuma–Wilson equation ($\alpha_w = 1.5$) [5]. [d] Rejected tie-lines for which the calculations did not show phase separation.

3.2. *Ternary systems*

Table 2 shows the ternary calculated results. The ternary parameters were obtained by minimizing the objective function

$$F = \left[\sum_k \min \sum_i \sum_j (x_{ijk,\text{calc.}} - x_{ijk,\text{exptl.}})^2 / 6M \right]^{1/2} \tag{8}$$

where $i = 1,2$ (phases), $j = 1,2,3$ (components), and $k = 1,2,3 \ldots M$ (tie-lines) and min means minimum values. A simplex method was used for the parameter determination procedure [25]. Figs. 1–6 show the calculated and experimental LLE for the ternary mixtures making up the six quaternary mixtures.

3.3. *Quaternary systems*

Table 3 gives the calculated results for ten quaternary systems. The quaternary parameters were sought in the way similar to that used in ternary parameter determination. The calculated results demonstrate the proposed model has a good performance in the correlation of LLE, which is nearly comparable with that of the previous approach based on the binary four-parameter Hiranuma–Wilson Eq.[15].

4. Conclusion

The presented model has a correlation ability comparable with that of the previous four-parameter Hiranuma–Wilson equation for binary partially miscible mixtures, especially for the six quaternary aqueous mixtures studied.

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References

[1] J.P. Novák, J. Matouš and J. Pick, Liquid–Liquid Equilibria, Elsevier, Amsterdam, 1987.

[2] J.P. Novák, P. Voňka, J. Suška, J. Matouš and J. Pick, Collect. Czech. Chem. Commun., 39 (1974) 3580–3592, 3593–3598.

[3] J. Novák, J. Matouš, K. Říčna and V. Kubíček, Collect. Czech. Chem. Commun., 54 (1989) 586–601.

[4] I. Nagata, Thermochim. Acta, 249 (1994) 75–87.

[5] I. Nagata and T. Watanabe, Thermochim. Acta, 208 (1992) 43–59.

[6] J. Gmehling, U. Onken and W. Arlt, Vapor–Liquid Equilibrium Data Collection, Organic Hydroxy Compounds: Alcohols and Phenols, DECHEMA Chemistry Data Ser., Vol. I. Part 2b, DECHEMA, Frankfurt am Main, Germany, 1978.

[7] J. Gmehling, U. Onken and P. Grenzhauser, Vapor–Liquid Equilibrium Data Collection, Carboxylic Acids, Anhydrides, Esters, DECHEMA Chemistry Data Ser., Vol. I, Part 5, DECHEMA, Frankfurt am Main, Germany, 1982.

[8] J. Gmehling, U. Onken and W. Arlt, Vapor–Liquid Equilibrium Data Collection, Aldehydes and Ketones, DECHEMA Chemistry Data Ser., Vol. I, Parts 3 + 4, DECHEMA, Framkfurt am Main, Germany, 1979.

[9] N. Gültekin, J. Chem. Eng. Data, 34 (1989) 168–171.

[10] J. Gmehling, U. Onken and W. Arlt, Vapor–Liquid Equilibrium Data Collection, Aromatic Hydrocarbons, DECHEMA Chemistry Data Ser., Vol. I, Part 7, DECHEMA, Frankfurt am Main, Germany, 1980.

[11] J. Gmehling, U. Onken and W. Arlt, Vapor–Liquid Equilibrium Data Collection, Organic Hydroxy Compounds: Alcohols (Suppl. 1), DECHEMA Chemistry Data Ser., Vol. I, Part 2c, DECHEMA, Frankfurt am Main, Germany, 1982.

[12] J. Gmehling and U. Onken, Vapor–Liquid Equilibrium Data Collection, Organic Hydroxy Compounds: Alcohols, DECHEMA Chemistry Data Ser., Vol. I, Part 2a, DECHEMA, Frankfurt am Main, Germany, 1977.

[13] J. Gmehling, U. Onken and J.R. Rarey-Nies, Vapor–Liquid Equilibrium Data Collection, Organic Hydroxy Compounds: Alcohols (Suppl. 3), DECHEMA Chemistry Data Ser., Vol. I, Part 2e, DECHEMA, Frankfurt am Main, Germany, 1988.

[14] I. Nagata and K. Katoh, Thermochim. Acta, 39 (1980) 45–62.

[15] J. Gmehling, U. Onken and W. Arlt, Vapor–Liquid Equilibrium Data Collection, Aliphatic Hydrocarbons C_4–C_6, DECHEMA Chemistry Data Ser., Vol. I, Part 6a, DECHEMA, Frankfurt am Main, Germany, 1980.

[16] J. Gmehling, U. Onken and U. Weidlich, Vapor–Liquid Equilibrium Data Collection, Organic Hydroxy Compounds: Alcohols and Phenols (Suppl. 2), DECHEMA Chemistry Data Ser., Vol. I, Part 2c, DECHEMA, Frankfurt am Main, Germany, 1988.

[17] J. Gmehling, U. Onken and W. Arlt, Vapor–Liquid Equilibrium Data Collection, Aqueous–Organic Systems (Suppl. 1), DECHEMA Chemistry Data Ser., Vol. I, Part 1a, DECHEMA, Frankfurt am Main, Germany, 1981.

[18] J. Gmehling and U. Onken, Vapor–Liquid Equilibrium Data Collection, Aqueous–Organic Systems, DECHEMA Chemistry Data Ser., Vol. I, Part 1, DECHEMA, Frankfurt am Main, Germany, 1977.

[19] I. Nagata and T. Ohta, J. Chem. Eng. Data, 28 (1983) 256–259.

[20] I. Nagata, Fluid Phase Equilibria, 18 (1984) 83–92.

[21] F. Ruiz, D. Prats, V. Gomis and P. Varo, Fluid Phase Equilibria, 18 (1984) 171–183.

[22] F. Ruiz and D. Prats, Fluid Phase Equilibria, 10 (1983) 95–114.

[23] J.M. Sørense and W. Arlt, Liquid–Liquid Equilibrium Data Collection, Binary Systems, DECHEMA Chemistry Data Ser., Vol. V, Part 1, DECHEMA, Frankfurt am Main, Germany, 1979.

[24] F. Ruiz and V. Gomis, J. Chem. Eng. Data, 30 (1985) 412–416.

[25] J.A. Nelder and R. Mead, Comput. J., 7 (1965) 308–313.

[26] E.A. Macedo and P. Rasmussen, Liquid–Liquid Equilibrium Data Collection, Suppl. 1, DECHEMA Chemistry Data Ser., Vol. V, Part 4, DECHEMA, frankfurt am Main, Germany, 1987.

[27] I. Nagata, Fluid Phase Equilibria, 26 (1986) 59–68.

[28] I. Nagata, Thermochim. Acta, 114 (1987) 227–238.

[29] I. Nagata, Thermochim. Acta, 119 (1987) 357–368.

[30] I. Nagata, Thermochim. Acta, 127 (1988) 109–118.

[31] I. Nagata, Thermochim. Acta, 127 (1988) 337–346.

[32] F. Ruiz, D. Prats and V. Gomis, An. Quim., 82 (1986) 393–398.

[33] I. Nagata, Thermochim. Acta, 214 (1993) 339–348.

[34] F. Ruiz, D. Prats and V. Gomis, J. Chem. Eng. Data, 29 (1984) 147–151.