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Representation of ternary liquid–liquid equilibria by means of a modified form of the Wilson equation

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Abstract

Experimental tie-line results are reported for acetic acid + benzene + triethylamine and acetic acid + chloroform + triethylamine at 25°C. A three-parameter Wilson equation, proposed by Novák et al. for binary partially miscible mixtures, is extended to reproduce liquid–liquid equilibria for many ternary mixtures. The proposed form of the modified Wilson equation is not identical to that used by Novák et al. for ternary mixtures, although both forms include three additional ternary parameters. Calculated results show a good performance comparable to that of the modified Hiranuma–Wilson equation, which contains four parameters for binary partially miscible mixtures.

Keywords: Binary system; LLE; Model; Modified Wilson equation; Ternary system

List of symbols

a_{ij}	molar energy parameter of Wilson equation for i–j pair
b_{ij}	binary parameter
C_{123i}	ternary constant of Eq. (4)
F	objective function given by Eq. (10)
g^{E}	molar excess Gibbs free energy
n_i	number of moles of component i

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n_T total number of moles of components
P total pressure
R universal gas constant
T absolute temperature
x_i liquid-phase mole fraction of component i
V_i liquid molar volume of pure component i

Greek letters

γ_i activity coefficient of component i
Λ_{ij}, Λ_{jki} binary and ternary parameters of Wilson equation

1. Introduction

The original Wilson equation with two binary parameters is well known for its good performance for completely miscible mixtures and is not applicable to partially miscible mixtures. To overcome this shortcoming, several investigators have presented modified forms of the Wilson equation [1]. These modified Wilson equations, having only binary parameters, fail usually in the precise representation of ternary liquid–liquid equilibria (LLE) for many mixtures. Therefore, additional ternary parameters will be necessary to correlate well ternary LLE. This paper presents the application of the Wilson equation modified by Novák et al. [2] for binary partially miscible mixtures to ternary LLE. The modified binary Wilson equation is expressed by

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

where the binary parameter Λ_{ij} is related to the molar energy parameter α_{ij}

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

For completely miscible mixtures, the empirical parameter b_{12} should be zero and Eq. (1) reduces to the original Wilson equation.

Novák et al. [3] correlated the ternary LLE of the water + ethanol + toluene system using the superposition of the Wilson and Redlich–Kister equations with a ternary term, which is given by

$$g^E/RT = (g^E/RT)_{\text{Redl-Kist}} + (g^E/RT)_{\text{Wilson}} + (g^E/RT)_{\text{tern}} \quad (3)$$

where

$$(g^E/RT)_{\text{tern}} = x_1x_2x_3(c_{1231}x_1 + c_{1232}x_2 + c_{1233}x_3) \quad (4)$$

The Redlich–Kister equation was used for water + ethanol, the Wilson equation for ethanol + toluene, and the modified Wilson equation for water + toluene.

This paper reports tie-line results for acetic acid + benzene + triethylamine and acetic acid + chloroform + triethylamine and shows that the proposed form of the Wilson equation for ternary partially miscible mixtures works better than Eq. (3)

and compares with the results based on an extension of the Hiranuma–Wilson equation with four binary parameters per binary [4].

2. Experimental

Acetic acid (Wako Pure Chemical Industries Ltd., analytical reagent grade) and triethylamine (Nakarai Tesque Inc., guaranteed reagent grade) were used as received. Benzene (Kanto Chemical Co., first grade) was purified by repeated recrystallization. Chloroform (Wako Pure Chemical Industries Ltd., analytical reagent grade) was washed with concentrated sulfuric acid, then with dilute sodium hydride and with distilled water, and then dried over potassium carbonate and fractionally distilled. The densities of the chemicals used for experimental work, measured with an Anton-Paar densimeter (DMA40) at 25°C, agreed well with literature values [5] as shown in Table 1.

Tie-lines were measured as described previously [6]. The compositions of two equilibrated liquids were analyzed using a Shimadzu gas chromatograph (GC-8C)

Table 1
Densities of pure components at 25°C

Component	Density/(g mol^{-1})		Component	Density/(g mol^{-1})	
	Obs.	Lit. [4]		Obs.	Lit. [4]
Acetic acid	1.04365	1.04392	Benzene	0.87365	0.87360
Triethylamine	0.72296	0.72305	Chloroform	1.47913	1.47970

Table 2
Experimental tie-line values for two ternary systems at 20°C

Phase I			Phase II		
x_1	x_2	x_3	x_1	x_2	x_3
Acetic acid(1) + benzene(2) + triethylamine(3)					
0.5786	0.0000	0.4214	0.1770	0.0000	0.8230
0.5523	0.0173	0.4304	0.1964	0.0306	0.7730
0.5323	0.0311	0.4366	0.1979	0.0527	0.7494
0.5053	0.0506	0.4441	0.2041	0.0807	0.7152
0.4980	0.0611	0.4409	0.2165	0.0908	0.6927
Acetic acid(1) + chloroform(2) + triethylamine(3)					
0.5786	0.0000	0.4214	0.1770	0.0000	0.8230
0.5717	0.0081	0.4202	0.1767	0.0164	0.8069
0.5474	0.0199	0.4327	0.1997	0.0275	0.7728
0.5358	0.0275	0.4367	0.2245	0.0365	0.7390
0.5130	0.0419	0.4450	0.2205	0.0560	0.7235
0.4957	0.0492	0.4551	0.2420	0.0646	0.6934

and a Shimadzu Chromatopac (C-R3A). The experimental error of observed mole fractions was at most 0.002.

Table 2 gives the experimental tie-line results for the ternary acetic acid + benzene + triethylamine and acetic acid + chloroform + triethylamine systems at 25°C.

3. Proposed model

We use the following g^E expression for ternary mixtures

$$\begin{aligned} g^E/RT = & -x_1 \ln(x_1 + \Lambda_{12}x_2 + \Lambda_{13}x_3 + \Lambda_{231}x_2x_3) \\ & - x_2 \ln(\Lambda_{21}x_1 + x_2 + \Lambda_{32}x_3 + \Lambda_{132}x_1x_3) \\ & - x_3 \ln(\Lambda_{31}x_1 + \Lambda_{32}x_2 + x_3 + \Lambda_{123}x_1x_2) \\ & + b_{12}x_1x_2 + b_{13}x_1x_3 + b_{23}x_2x_3 \end{aligned} \tag{5}$$

where Λ_{231}, Λ_{132}, and Λ_{123} are the ternary parameters. Eq. (5) is easily derived in the same way as described previously [4].

The activity coefficient of any component i is derived from

$$\ln \gamma_i = \frac{1}{RT}\left[\frac{\partial n_T g^E}{\partial n_i}\right]_{P,T,n_{j(j\neq i)}} \tag{6}$$

Substitution of Eq. (5) into Eq. (6) gives

$$\begin{aligned} \ln \gamma_1 = & -\ln(x_1 + \Lambda_{12}x_2 + \Lambda_{13}x_3 + \Lambda_{231}x_2x_3) + 1 \\ & - x_1\left[\frac{1 - \Lambda_{231}x_2x_3}{x_1 + \Lambda_{12}x_2 + \Lambda_{13}x_3 + \Lambda_{231}x_2x_3}\right] \\ & - x_2\left[\frac{\Lambda_{21} - \Lambda_{132}(1 - x_1)x_3}{\Lambda_{21}x_1 + x_2 + \Lambda_{23}x_3 + \Lambda_{132}x_1x_2}\right] \\ & - x_3\left[\frac{\Lambda_{31} - \Lambda_{123}(1 - x_1)x_2}{\Lambda_{31}x_1 + \Lambda_{32}x_3 + \Lambda_{123}x_1x_2}\right] \\ & + b_{12}(1 - x_1)x_2 + b_{13}(1 - x_1)x_3 - b_{23}x_2x_3 \end{aligned} \tag{7}$$

Ln γ_2 and ln γ_3 are similarly obtained.

4. Calculated results

4.1. Binary systems

Each set of the three binary parameters were obtained by solving the thermodynamic equation for each component at two equilibrium liquid phases

$$(x_i\gamma_i)^{\mathrm{I}} = (x_i\gamma_i)^{\mathrm{II}} \tag{8}$$

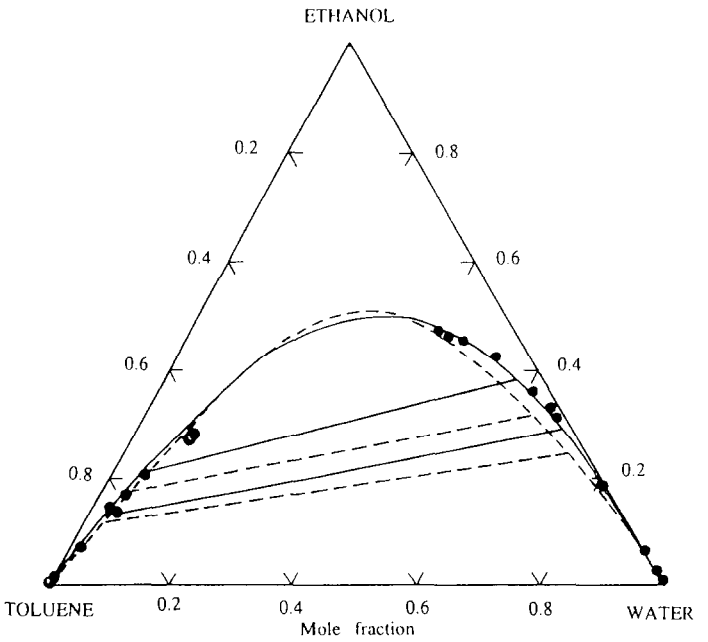

Fig. 1. Calculated ternary liquid–liquid equilibria for water + ethanol + toluene at 25°C [35]. Experimental, ●. Calculated: – – –, with the method of Novák et al. [3]; ——, with the present method.

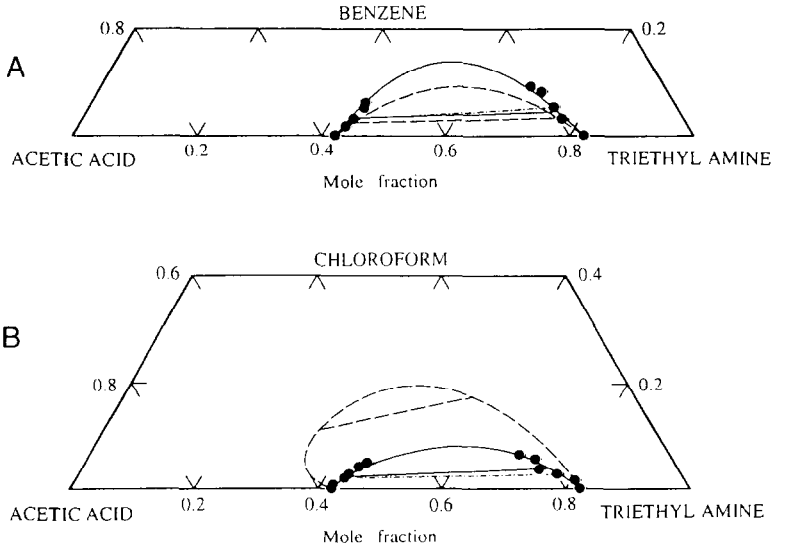

Fig. 2. Calculated liquid–liquid equilibria for two ternary systems at 25°C. Experimental tie-line (●— – —●), this work. Calculated: – – –, with only binary parameters; ——, with binary and ternary parameters. A, acetic acid + benzene + triethylamine; B, acetic acid + chloroform + triethylamine.

Table 3
Binary parameters

System (1 + 2)	Temp./°C	a_{12}/K	a_{12}/K	b_{12}	Ref.
Water + methanol	39.9	246.77	−17.56		[7]
Water + ethanol	25.0	440.56	27.73		[8]
Water + 1-propanol	94.75–89.30	639.20	814.60		[8]
Water + acetone	25.0	796.10	−54.01		[7]
Water + acetic acid	100.3–115.2	403.09	3.57		[8]
Methanol + methyl acetate	30.0	429.30	−11.18		[9]
Methanol + ethyl ether	30.0	614.00	−121.09		[10]
Methanol + tetrahydrofuran	25.0	410.49	−105.91		[10]
Methanol + benzene	35.0	899.85	92.17		[9]
Methanol + toluene	63.8–83.1	1003.1	110.68		[10]
Methanol + acetone	25.0	249.41	−30.80		[9]
Methanol + 2-propanol	55.0	627.74	−451.28		[9]
Aniline + methanol	20.0	−52.29	338.35		[10]
Ethanol + ethyl acetate	70.0	316.15	20.88		[9]
Ethanol + cyclohexane	25.0	1017.2	262.62		[10]
Ethanol + benzene	45.0	802.21	71.52		[9]
Ethanol + toluene	35.0	844.28	85.04		[9]
Ethanol + chloroform	35.0	878.70	−195.51		[9]
Acetonitrile + ethanol	20.0	306.47	221.38		[10]
1-Propanol + benzene	60.0	582.77	141.96		[9]
2-Propanol + cyclohexane	60.0	797.96	142.66		[11]
Acetonitrile + benzene	45.0	392.49	−7.60		[12]
Aniline + acetronitrile	20.0	390.78	−105.43		[13]
Acetic acid + chloroform	61.4–118.1	472.71	−34.81		[14]
Acetic acid + tetrachloromethane	20.0	669.94	9.44		[14]
Acetic acid + benzene	20.0	610.73	0.52		[14]
Tetrachloromethane + triethylamine	20.0	−193.23	366.63		[13]
Chloroform + triethylamine	9.99	−12.31	−416.73		[13]
Acetone + cyclohexane	25.0	560.93	225.79		[15]
Benzene + triethylamine	60.0	101.61	−29.92		[12]
Nitromethane + benzene	45.0	366.76	73.90		[12]
Tetrahydrofuran + cyclohexane	25.0	−382.04	1457.82		[16]
Acetone + methyl acetate	40.0	41.40	−8.15		[15]
Ethyl ether + cyclohexane	25.0	206.42	295.52		[17]
Methyl acetate + cyclohexane	35.0	386.97	213.91		[14]
Benzene + *n*-heptane	45.0	55.20	216.55		[18]
Benzene + cyclohexane	25.0	85.77	67.31		[19]
Benzene + *n*-octane	65.0	143.26	8.41		[18]
Toluene + *n*-hexane	71.15–102.1	124.04	10.58		[19]
Toluene + cyclohexane	25.0	98.73	53.45		[19]
Toluene + *n*-heptane	25.0	79.24	82.40		[18]
Toluene + *n*-octane	110.86–124.14	115.42	−1.38		[18]
n-Octane + cyclohexane	25.0	258.69	−91.33		[20]
Water + benzene[a]	25.0	1753.30	1376.60	0.9341	[21]
Water + benzene[a]	37.8	1332.40	1077.40	1.0503	[21]
Water + benzene[a]	45.0	955.48	785.82	2.8929	[22]
Water + methyl acetate[a]	30.0	291.45	446.90	1.6710	[23]
Water + ethyl acetate[a]	70.0	910.97	840.20	0.4851	[23]
Water + chloroform[a]	25.0	269.45	365.59	4.8870	[21]

Table 3 (continued)

System (1 + 2)	Temp./°C	a_{12}/K	a_{12}/K	b_{12}	Ref.
Water + toluene[a]	25.0	819.78	862.37	3.9156	[24]
Methanol + *n*-hexane[a]	25.0	75.85	133.46	2.4600	[21]
Methanol + cyclohexane[a]	25.0	1085.10	358.56	0.1498	[25]
Methanol + *n*-heptane[a]	25.0	177.58	94.87	2.4065	[26]
Methanol + *n*-octane[a]	25.0	175.59	218.50	2.6096	[26]
Acetonitrile + cyclohexane[a]	25.0	−12.37	−74.20	3.7668	[27]
Acetonitrile + *n*-heptane[a]	45.0	127.75	153.55	2.5941	[28]
Acetic acid + triethylamine[a]	20.0	1240.60	−21.29	0.3224	[23]
Acetic acid + triethylamine[a]	25.0	850.91	−150.51	0.6809	[29]
Aniline + cyclohexane[a]	25.0	450.21	140.94	0.7971	[30]
Nitromethane + cyclohexane[a]	25.0	118.85	−133.38	3.5957	[21]

[a] Mutual solubility data.

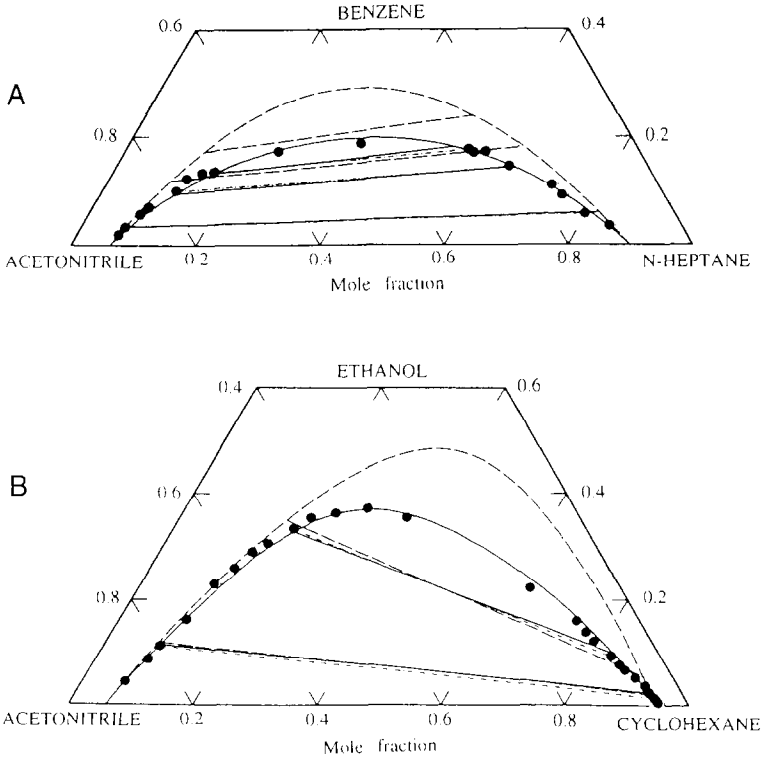

Fig. 3. Calculated liquid–liquid equilibria for two ternary systems. Experimental tie-line (●— – – –●). Calculated: – – –, with only binary parameters; —, with binary and ternary parameters. A, acetonitrile + benzene + *n*-heptane at 45°C [23]; B, acetonitrile + ethanol + cyclohexane at 25°C [31].

Table 4
Calculated results of ternary liquid–liquid equilibria

System (1 + 2 + 3)	Temp./°C	Number of tie-lines	Ternary parameters	Root-mean-squared deviations/mol%			Ref.
				I[a]	II[b]	III[c]	
Acetonitril + benzene + *n*-heptane	45.0	9	$\Lambda_{231} = 1.4837$ $\Lambda_{132} = 0.7032$ $\Lambda_{123} = -0.1604$	0.59	0.56	4.08	[23]
Acetonitrile + ethanol + cyclohexane	25.0	13	$\Lambda_{231} = -2.9940$ $\Lambda_{132} = 0.3476$ $\Lambda_{123} = 1.7226$	0.75			[31]
Methanol + 2-propanol + cyclohexane	25.0	7	$\Lambda_{231} = 0.5151$ $\Lambda_{132} = -0.1112$ $\Lambda_{123} = -0.2241$	0.50	0.51	5.97	[32]
Methanol + ethyl ether + cyclohexane	25.0	4	$\Lambda_{231} = -1.1367$ $\Lambda_{132} = 7.0167$ $\Lambda_{123} = -1.6846$	0.29	0.38	2.21	[23]
Methanol + methyl acetate + cyclohexane	25.0	7	$\Lambda_{231} = -0.0669$ $\Lambda_{132} = 0.5162$ $\Lambda_{123} = -0.2382$	0.28	0.27	0.52	[23]
Methanol + tetrahydrofuran + cyclohexane	25.0	5	$\Lambda_{231} = 0.3946$ $\Lambda_{132} = 6.5981$ $\Lambda_{123} = 0.0936$	0.30	0.32	0.73	[23]
Methanol + acetone + cyclohexane	25.0	7	$\Lambda_{231} = -0.2007$ $\Lambda_{132} = 0.3848$ $\Lambda_{123} = -0.3470$	0.35	0.38	2.56	[32]
Methanol + cyclohexane + *n*-octane	25.0	9	$\Lambda_{231} = -2.0425$ $\Lambda_{132} = 0.7566$ $\Lambda_{123} = -0.2551$	0.41			[33]
Methanol + benzene + cyclohexane	25.0	6	$\Lambda_{231} = 0.2611$ $\Lambda_{132} = 0.2620$ $\Lambda_{123} = 0.1530$	0.29	0.30	2.01	[32]
Methanol + benzene + *n*-heptane	25.0	8	$\Lambda_{231} = -1.1740$ $\Lambda_{132} = -0.0789$ $\Lambda_{123} = 1.7042$	0.34	0.44	2.01	[26]
Methanol + benzene + *n*-octane	25.0	6	$\Lambda_{231} = -2.1595$ $\Lambda_{132} = 2.3748$ $\Lambda_{123} = 0.2077$	0.66	0.66	4.46	[26]
Methanol + toluene + *n*-hexane	25.0	4	$\Lambda_{231} = 21.4730$ $\Lambda_{132} = 162.890$ $\Lambda_{123} = -4.4844$	0.93	0.91	2.36	[34]
Methanol + toluene + cyclohexane	25.0	7	$\Lambda_{231} = 0.3244$ $\Lambda_{132} = 0.3381$ $\Lambda_{123} = 0.0915$	0.21	0.23	1.04	[32]

Table 4 (continued)

System (1 + 2 + 3)	Temp./°C	Number of tie-lines	Ternary parameters	Root-mean-squared deviations/mol%			Ref.
				I[a]	II[b]	III[c]	
Methanol + toluene + *n*-heptane	25.0	7	$\Lambda_{231} = -2.3249$ $\Lambda_{132} = 0.7627$ $\Lambda_{123} = 1.4744$	0.52	0.38	1.71	[26]
Methanol + toluene + *n*-octane	25.0	7	$\Lambda_{231} = -3.0890$ $\Lambda_{132} = 1.3579$ $\Lambda_{123} = 0.7338$	0.40	0.34	2.92	[26]
Water + methanol + benzene	45.0	13	$\Lambda_{231} = -0.2513$ $\Lambda_{132} = 3.0204$ $\Lambda_{123} = -0.4921$	0.80	0.82	8.97	[23]
Water + ethanol + benzene	25.0	12	$\Lambda_{231} = 1.5063$ $\Lambda_{132} = -0.6060$ $\Lambda_{123} = -0.2644$	0.85	0.91	5.53	[23]
Water + 1-propanol + benzene	37.8	6	$\Lambda_{231} = 0.3036$ $\Lambda_{132} = -0.1698$ $\Lambda_{123} = 0.8922$	0.72	0.72	10.85	[23]
Water + ethanol + toluene	25.0	11	$\Lambda_{231} = 0.8510$ $\Lambda_{132} = 3.4811$ $\Lambda_{123} = -0.3599$	1.10			[35]
Water + ethanol + toluene	25.0	14	$\Lambda_{231} = 0.5376$ $\Lambda_{132} = 3.7199$ $\Lambda_{123} = -0.3320$	0.42			[24]
Water + ethanol + chloroform	25.0	15	$\Lambda_{231} = 2.1510$ $\Lambda_{132} = 2.1746$ $\Lambda_{123} = -0.2054$	0.63			[32]
Water + ethanol + ethyl acetate	70.0	6	$\Lambda_{231} = 0.3310$ $\Lambda_{132} = 1.0258$ $\Lambda_{123} = -0.5177$	0.19	0.31	9.72	[23]
Water + acetone + methyl acetate	30.0	4	$\Lambda_{231} = -0.1902$ $\Lambda_{132} = 1.2772$ $\Lambda_{123} = -0.4789$	0.32	0.35	6.63	[23]
Water + acetic acid + chloroform	25.0	8	$\Lambda_{231} = 0.8565$ $\Lambda_{132} = 2.8658$ $\Lambda_{123} = -0.5166$	0.37			[32]
Acetic acid + tetrachloromethane + triethylamine	20.0	11	$\Lambda_{231} = -0.1577$ $\Lambda_{132} = -1.6568$ $\Lambda_{123} = 0.3816$	0.91			[23]
Acetic acid + benzene + triethylamine	25.0	5	$\Lambda_{231} = -0.0546$ $\Lambda_{132} = 0.0761$ $\Lambda_{123} = -0.3081$	0.42			This work

Table 4 (continued)

System (1 + 2 + 3)	Temp./°C	Number of tie-lines	Ternary parameters	Root-mean-squared deviations/mol%			Ref.
				I[a]	II[b]	III[c]	
Acetic acid + chloroform + triethylamine	25.0	6	$\Lambda_{231} = -0.2959$ $\Lambda_{132} = 0.7487$ $\Lambda_{123} = 0.5813$	0.39			This work
Aniline + methanol + cyclohexane	25.0	10	$\Lambda_{231} = 0.2237$ $\Lambda_{132} = -0.0343$ $\Lambda_{123} = -0.2813$	0.57			[30]
Aniline + acetonitrile + cyclohexane	25.0	6	$\Lambda_{231} = -1.4282$ $\Lambda_{132} = 12.3710$ $\Lambda_{123} = 1.7063$	1.10			[36]
Nitromethane + benzene + cyclohexane	25.0	3	$\Lambda_{231} = 0.7477$ $\Lambda_{132} = 0.1815$ $\Lambda_{123} = 0.8282$	0.85	0.85	3.46	[23]

[a] This work. [b] Modified form of Hiranuma–Wilson equation ($a_{13} = 1.5$ for aqueous systems) [6]. [c] Higashiuchi et al. [37].

with

$$\sum_i x_i^{\mathrm{I}} = 1 \quad \text{and} \quad \sum_i x_i^{\mathrm{II}} = 1 \tag{9}$$

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

Table 3 shows the values of the binary parameters collected from the literature or calculated from published phase equilibrium data [7–30].

4.2. Ternary systems

The ternary parameters, given in Table 4, were sought by minimizing the objective function

$$F = \left[\sum_k \min \sum_i \sum_j (x_{ijk,\mathrm{calc}} - x_{ijk,\mathrm{exptl}})^2 / 6M \right]^{1/2} \tag{10}$$

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 this purpose [38]. Figs. 1–3 illustrate typical calculated results. As shown in Fig. 1, for the water + ethanol + toluene system, the present approach gives a better correlation than that of Novák et al. [3]. Figs. 2–5 compare the calculated results with the experimental values for some typical systems. Table 4 also lists the results for 19 systems from two modifications of the Wilson equation: Hiranuma–Wilson [4] and that of Higashiuchi et al. [37]. The method of Higashiuchi et al. gave poor results for many

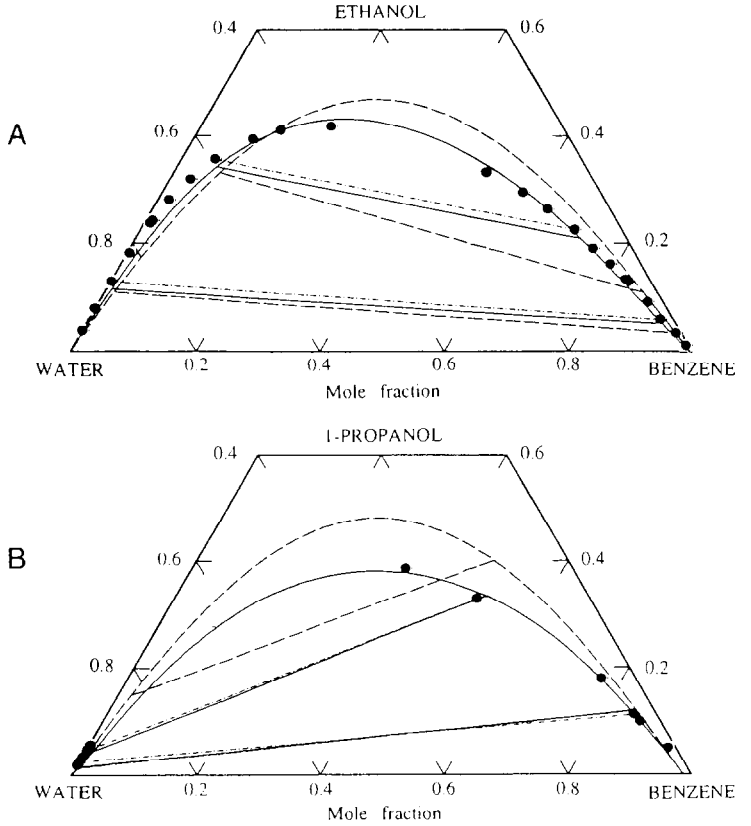

Fig. 4. Calculated liquid–liquid equilibria for two ternary systems [23]. Experimental tie-line (●— – – —●). Calculated: – – –, with only binary parameters; ——, with binary and ternary parameters. A, water + ethanol + benzene at 25°C; B, water + 1-propanol + benzene at 37.8°C.

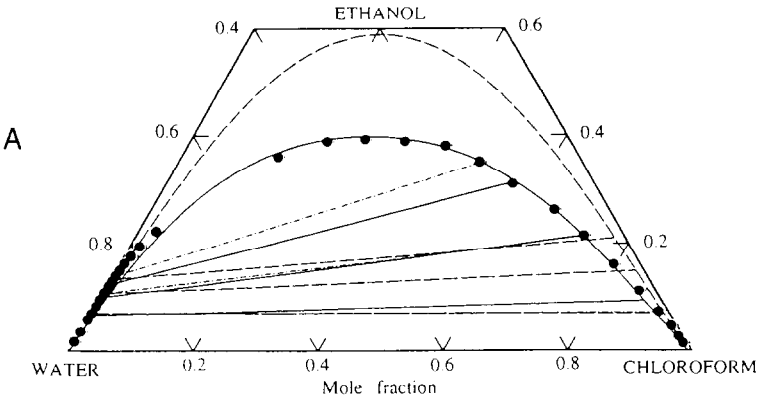

Fig. 5 A.

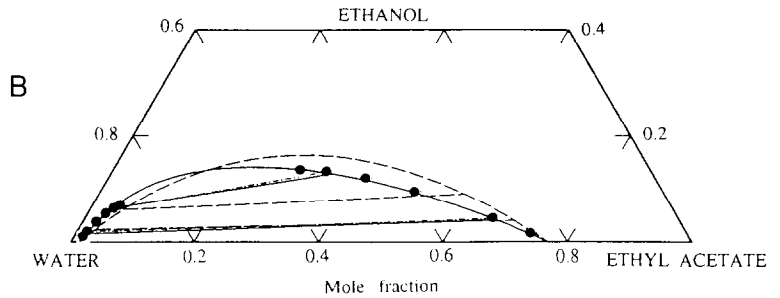

Fig. 5. Calculated liquid–liquid equilibria for two ternary systems. Experimental tie-line (●— – —●). Calculated: – – –, with only binary parameters; ——, with binary and ternary parameters. A, water + ethanol + chloroform at 25°C [32]; B, water + ethanol + ethyl acetate at 70°C [23].

systems. The average deviation for 19 systems is 0.473 for this work and 0.494 for that based on the Hiranuma–Wilson equation [6]. We may conclude that the present ternary modification of the binary modified Wilson equation proposed by Novák et al. is better than that suggested by Novák et al. for ternary systems and has good results comparable to those based on the binary Hiranuma–Wilson equation.

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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. Říčná and V. Kubíček, Collect. Czech. Chem. Commun., 54 (1989) 586–601.
[4] I. Nagata and T. Watanabe, Fluid Phase Equilibria, 72 (1992) 1–14.
[5] J.A. Riddick, W.B. Bunger and T.K. Sakano, Organic Solvents, 4th edn., Wiley-Interscience, New York, 1986.
[6] I. Nagata, Thermochim. Acta, 210 (1992) 281–292.
[7] J. Gmehling and U. Onken, Vapor–Liquid Equilibrium Data Collection, Aqueous Organic Systems, Vol. I, Part 1, DECHEMA Chemistry Data Ser., DECHEMA, Frankfurt am Main, Germany, 1977.
[8] J. Gmehling, U. Onken and W. Arlt, Vapor–Liquid Equilibrium Data Collection, Aqueous Organic Systems (Supplement 1), Vol. I, Part 1a, DECHEMA Chemistry Data Ser., DECHEMA, Frankfurt am Main, Germany, 1981.
[9] J. Gmehling and U. Onken, Vapor–Liquid Equilibrium Data Collection, Organic Hydroxy Compounds: Alcohols, Vol. I, Part 2a, DECHEMA Chemistry Data Ser., DECHEMA, Frankfurt am Main, Germany, 1977.

[10] J. Gmehling, U. Onken and W. Arlt, Vapor–Liquid Equilibrium Data Collection, Organic Hydroxy Compounds (Supplement 1), Vol. I, Part 2c, DECHEMA Chemistry Data Ser., DECHEMA, Frankfurt am Main, Germany, 1982.
[11] J. Gmehling, U. Onken and W. Arlt, Vapor–Liquid Equilibrium Data Collection, Organic Hydroxy Compounds: Alcohols and Phenols, Vol. I, Part 2b, DECHEMA Chemistry Data Ser., DECHEMA, Frankfurt am Main, Germany, 1978.
[12] J. Gmehling, U. Onken and W. Arlt, Vapor–Liquid Equilibrium Data Collection, Aromatic Hydrocarbons, Vol. I, Part 7, DECHEMA Chemistry Data Ser., DECHEMA, Frankfurt am Main, Germany, 1980.
[13] J. Gmehling, U. Onken and W. Arlt, Vapor–Liquid Equilibrium Data Collection, Halogen, Nitrogen, Sulfur and other Compounds, Vol. I, Part 7, DECHEMA Chemistry Data Ser., DECHEMA, Frankfurt am Main, Germany, 1984.
[14] J. Gmehling, U. Onken and P. Grenzheuser, Vapor–Liquid Equilibrium Data Collection, Carboxylic Acids, Anhydrides, Esters, Vol. I, Part 5, DECHEMA Chemistry Data Ser., DECHEMA, Frankfurt am Main, Germany, 1982.
[15] J. Gmehling, U. Onken and W. Arlt, Vapor–Liquid Equilibrium Data Collection, Aldehydes, Ketones, Ethers, Vol. I, Parts 3 and 4, DECHEMA Chemistry Data Ser., DECHEMA, Frankfurt am Main, Germany, 1979.
[16] H. Arm and D. Bankey, Helv. Chim. Acta, 52 (1969) 279–281.
[17] H. Arm, D. Bankey, K. Strub and M. Walti, Helv. Chim. Acta, 50 (1967) 1013–1016.
[18] J. Gmehling, U. Onken and W. Arlt, Vapor–Liquid Equilibrium Data Collection, Aliphatic Hydrocarbons C_7–C_{18}, Vol. I, Part 6b, DECHEMA Chemistry Data Ser., DECHEMA, Frankfurt am Main, Germany, 1980.
[19] J. Gmehling, U. Onken and W. Arlt, Vapor–Liquid Equilibrium Data Collection, Aliphatic Hydrocarbons C_4–C_6, Vol. I, Part 6a, DECHEMA Chemistry Data Ser., DECHEMA, Frankfurt am Main, Germany, 1980.
[20] J. Gmehling, U. Onken and B. Kolbe and B. Kolbe, Vapor–Liquid Equilibrium Data Collection, Aliphatic Hydrocarbons (Supplement 1), Vol. I, Part 6c, DECHEMA Chemistry Data Ser., DECHEMA, Frankfurt am Main, Germany, 1983.
[21] J.M. Sørensen and W. Arlt, Liquid–Liquid Equilibrium Data Collection, Binary Systems, Vol. V, Part 1, DECHEMA Chemistry Data Ser., DECHEMA, Frankfurt am Main, Germany, 1979.
[22] J.F. McCants, J.H. Jones and W.H. Hopson, Ind. Eng. Chem., 45 (1953) 454–456.
[23] J.M. Sørensen and W. Arlt, Liquid–Liquid Equilibrium Data Collection, Ternary Systems, Vol. V, Part 2, DECHEMA Chemistry Data Ser., DECHEMA, Frankfurt am Main, Germany, 1980.
[24] R. Ruiz, D. Prats and V. Gomis, J. Chem. Eng. Data, 30 (1985) 412–416.
[25] I. Nagata, Fluid Phase Equilibria, 18 (1984) 83–92.
[26] H. Higashiuchi, Y. Sakurai, Y. Iwai, Y. Arai and M. Nagatani, Fluid Phase Equilibria, 36 (1987) 35–47.
[27] I. Nagata and T. Ohta, J. Chem. Eng. Data, 28 (1983) 256–259.
[28] D.A. Palmer and B.D. Smith, J. Chem. Eng. Data, 17 (1972) 71–76.
[29] R.L. Scott, J. Chem. Phys., 25 (1956) 193–205.
[30] I. Nagata, Thermochim. Acta, 186 (1991) 123–130.
[31] I. Nagata, Fluid Phase Equilibria, 26 (1986) 59–68.
[32] E.A. Macedo and P. Rasmussen, Liquid–Liquid Equilibrium Data Collection, Supplement 1, Vol. V, Part 4, DECHEMA Chemistry Data Ser., DECHEMA, Frankfurt am Main, Germany, 1987.
[33] I. Nagata, Thermochim. Acta, 210 (1992) 281–292.
[34] Y. Iwai, Y. Nishimura, M. Watanabe and Y. Arai, Fluid Phase Equilibria, 25 (1986) 107–112.
[35] K. Řična, J. Matouš, J.P. Novák and V. Kubiček, Collect. Czech. Chem. Commun., 54 (1989) 581–585.
[36] I. Nagata, J. Chem. Thermodyn., in press.
[37] H. Higashiuchi, Y. Sakurai, M. Nagatani and Y. Arai, Sekiyu Gakkaishi, 33 (1990) 62–66.
[38] J.A. Nedler and R. Mead, Comput. J., 7 (1965) 308–313.