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PREDICTION OF TERNARY PHASE EQUILIBRIA FROM BINARY DATA

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ABSTRACT

The two-parameter UNIQUAC equation is modified to give better results of vapor–liquid and liquid–liquid equilibria for a variety of binary systems. The proposed equation is easily extended to a multicomponent system without including any ternary (or multicomponent) parameters. The good capability of the equation in data reduction is shown by many illustrative examples for various kinds of strongly nonideal binary and ternary mixtures.

NOTATION

a_{ij}	Extended UNIQUAC binary interaction parameter related to Δu_{ji} and τ_{ij}
B_{ij}	Second virial coefficient for i–j interaction
g^E	Excess molar Gibbs energy
P	Total pressure
P_i^s	Saturation pressure of pure component i
q_i	Molecular-geometric area parameter for component i
q_i'	Molecular-interaction area parameter for component i
r_i	Molecular volume parameter for component i
R	Gas constant
T	Absolute temperature
Δu_{ji}	Extended UNIQUAC binary interaction parameter
u^E	Molar energy of mixing
v_i^L	Molar liquid volume of pure component i
x_i	Liquid-phase mole fraction of component i
x_i^I, x_i^{II}	Liquid-phase mole fractions of component i in phase I and II
y_i	Vapor-phase mole fraction of component i
Z	Lattice coordination number, here equal to 10

Greek letters

γ_i	Activity coefficient of component i
θ_i	Area fraction of component i
θ_{ij}	Local area fraction of a molecule i about a central molecule j
τ_{ij}	Extended UNIQUAC binary parameter related to Δu_{ji}

ϕ_i	Fugacity coefficient of component i
ϕ_i^s	Fugacity coefficient of pure component i at its saturation pressure
ϕ_i	Segment fraction of component i

INTRODUCTION

The UNIQUAC equation, which was derived by Abrams and Prausnitz [1], is particularly useful since it is applicable to a number of strongly nonideal liquid mixtures. This equation has two adjustable parameters per binary. To obtain better results with water or alcohols, Anderson and Prausnitz [2] have used the pure component molecular structural parameter q', which is different from q for these components, in the residual term of the UNIQUAC equation. Nagata and Katoh [3] have presented a modification of the UNIQUAC equation, called effective UNIQUAC. The effective UNIQUAC equation, like the UNIQUAC equation, usually requires an additional third parameter to correlate successfully ternary liquid–liquid equilibria. In this work another modification of the UNIQUAC equation to predict simultaneously ternary vapor–liquid and liquid–liquid equilibria using binary parameters alone is proposed.

BASIC THERMODYNAMIC RELATIONS

I present the following modified form of the two-parameter UNIQUAC equation, called extended UNIQUAC.

$$g^E = g^E\,(\text{combinatorial}) + g^E\,(\text{residual}) \tag{1}$$

For a binary system

$$\frac{g^E\,(\text{combinatorial})}{RT} = x_1 \ln\frac{\phi_1}{x_1} + x_2 \ln\frac{\phi_2}{x_2} \left(\frac{Z}{2}\right)\left(q_1 x_1 \ln\frac{\theta_1}{\phi_1} + q_2 x_2 \ln\frac{\theta_2}{\phi_2}\right) \tag{2}$$

$$\frac{g^E\,(\text{residual})}{RT} = -q_1' x_1 \ln(\theta_1 + \theta_2\tau_{21}) - q_2' x_2 \ln(\theta_2 + \theta_1\tau_{12}) \tag{3}$$

where Z is the coordination number equal to 10, and segment fraction, ϕ, and area fraction, θ, are given by

$$\phi_1 = \frac{x_1 r_1}{x_1 r_1 + x_2 r_2} \qquad \phi_2 = \frac{x_2 r_2}{x_1 r_1 + x_2 r_2} \tag{4}$$

$$\theta_1 = \frac{x_1 q_1}{x_1 q_1 + x_2 q_2} \qquad \theta_2 = \frac{x_2 q_2}{x_1 q_1 + x_2 q_2} \tag{5}$$

r, q and q' are pure component molecular structural parameters. In the original UNIQUAC equation $q' = q$ and in the effective UNIQUAC equation $q' = 1$.

The extended UNIQUAC equation is easily derived if we assume that the energy of mixing, u^E, for a binary mixture is given by

$$u^E = q_1'x_1\theta_{21}\Delta u_{21} + q_2'x_2\theta_{12}\Delta u_{12} \tag{6}$$

and if we follow the procedure of Maurer and Prausnitz [4] used in the derivation of the three-parameter UNIQUAC equation. q' is a correction factor of interaction. The local area fraction is defined in terms of the area fraction, θ, and binary interaction parameter, Δu.

$$\theta_{21} = \theta_2\exp(-\Delta u_{21}/RT)/[\theta_1 + \theta_2\exp(-\Delta u_{21}/RT)] \tag{7}$$

$$\theta_{12} = \theta_1\exp(-\Delta u_{12}/RT)/[\theta_2 + \theta_1\exp(-\Delta u_{12}/RT)] \tag{8}$$

Values of q' for water, nitromethane and alcohols have been empirically obtained to give a good fit to various systems containing these components: water, 0,96; nitromethane, 1.29; methanol, 0.99; ethanol, 0.92; propanols, 0.89; butanols, 0.88. For other components studied in this work I suggest $q' = q^{0.2}$.

The two adjustable parameters, τ_{12} and τ_{21}, are related to binary interaction energies, Δu_{12} and Δu_{21}.

$$\tau_{12} = \exp(-\Delta u_{12}/RT) = \exp(-a_{12}/T) \tag{9}$$

$$\tau_{21} = \exp(-\Delta u_{21}/RT) = \exp(-a_{21}/T) \tag{10}$$

The activity coefficients are given by the relation

$$RT\ln\gamma_i = \left[\frac{\partial(n_T g^E)}{\partial n_i}\right]_{P,T,n_j} \tag{11}$$

where n_i is the number of moles of component i, n_T is the total number of moles ($n_T = \sum_i n_i$), and $x_i = n_i/n_T$.

$$\begin{aligned}\ln\gamma_1 = {} & \ln\frac{\phi_1}{x_1} + 1 - \frac{\phi_1}{x_1} - \left(\frac{Z}{2}\right)q_1\left(\ln\frac{\phi_1}{\theta_1} + 1 - \frac{\phi_1}{\theta_1}\right) - q_1'\ln(\theta_1 + \theta_2\tau_{21}) \\ & + q_1\theta_2\left[\left(\frac{q_1'}{q_1}\right)\left(\frac{\tau_{12}}{\theta_1 + \theta_2\tau_{21}} - 1\right) - \left(\frac{q_2'}{q_2}\right)\left(\frac{\tau_{12}}{\theta_2 + \theta_1\tau_{12}} - 1\right)\right]\end{aligned} \tag{12}$$

$$\begin{aligned}\ln\gamma_2 = {} & \ln\frac{\phi_2}{x_2} + 1 - \frac{\phi_2}{x_2} - \left(\frac{Z}{2}\right)q_2\left(\ln\frac{\phi_2}{\theta_2} + 1 - \frac{\phi_2}{\theta_2}\right) - q_2'\ln(\theta_2 + \theta_1\tau_{12}) \\ & + q_2\theta_1\left[\left(\frac{q_2'}{q_2}\right)\left(\frac{\tau_{21}}{\theta_2 + \theta_1\tau_{12}} - 1\right) - \left(\frac{q_1'}{q_1}\right)\left(\frac{\tau_{21}}{\theta_1 + \theta_2\tau_{21}} - 1\right)\right]\end{aligned} \tag{13}$$

For a multicomponent mixture the extended UNIQUAC equation provides g^E and γ as follows.

$$\frac{g^E\text{ (combinatorial)}}{RT} = \sum_i x_i\ln\frac{\phi_i}{x_i} + \left(\frac{Z}{2}\right)\sum_i q_i x_i\ln\frac{\phi_i}{\theta_i} \tag{14}$$

$$\frac{g^E\text{ (residual)}}{RT} = -\sum_i q_i' x_i \ln\left(\sum_j \theta_j \tau_{ji}\right) \tag{15}$$

$$\ln \gamma_i = \ln\frac{\phi_i}{x_i} + 1 - \frac{\phi_i}{x_i} - \left(\frac{Z}{2}\right) q_i \left(\ln\frac{\phi_i}{\theta_i} + 1 - \frac{\phi_i}{\theta_i}\right)$$
$$-q_i' \ln\left(\sum_j \theta_j \tau_{ji}\right) + q_i \sum_j \left(\frac{q_j'}{q_j}\right) \theta_j - q_i \sum_j \frac{(q_j'/q_j)\theta_j \tau_{ij}}{\sum_k \theta_k \tau_{kj}} \tag{16}$$

where segment fraction, ϕ, and area fraction, θ, are given by

$$\phi_i = \frac{x_i r_i}{\sum_j x_j r_j} \tag{17}$$

$$\theta_i = \frac{x_i q_i}{\sum_j x_j q_j} \tag{18}$$

The vapor–liquid equilibrium relationship for any component i [5] is described by

$$\phi_i y_i P = \gamma_i x_i \phi_i^s P_i^s \exp\left[v_i^L (P - P_i^s)/RT\right] \tag{19}$$

where y is the vapor-phase mole fraction, P is the total pressure, P^s is the pure component vapor pressure, and v^L is the pure component molar liquid volume. The fugacity coefficient, ϕ, is calculated by

$$\ln \phi_i = \frac{P}{RT}\left(2\sum_j y_j B_{ij} - \sum_i \sum_j y_i y_j B_{ij}\right) \tag{20}$$

The pure-component and cross virial coefficients are calculated by the method of Hayden and O'Connell [6].

The equation of multicomponent liquid–liquid equilibria between two phases I and II, for each component i, is

$$(\gamma_i x_i)^{I} = (\gamma_i x_i)^{II} \tag{21}$$

RESULTS

The optimum parameters were found by satisfaction of a statistical criterion based on the maximum likelihood principle. Computer programs used in this work are similar to those described in the monograph of Prausnitz et al. [5]. r, q, and other parameters required to estimate the second virial coefficients are listed in the same monograph. The value of q' is independent of binary data after a single value is fixed by examining several binary systems. Table 1 lists a number of binary parameters. Figures 1–3

TABLE 1

Binary interaction parameters for extended UNIQUAC equation

System(1)–(2)	No. of points	Temp. (°C)	Energy parameters, K		Variance of fit	Ref.
			a_{12}	a_{21}		
Acetonitrile–benzene	11	45	−8.32	304.46	9.20	7
Benzene–*n*-heptane	15	45	69.51	61.71	1.90	7
Ethanol–*n*-octane	17	45	266.97	1228.39	4.27	8
	19	75	243.76	1129.06	3.51	8
Ethanol–acetonitrile	14	40	286.07	171.03	3.82	9
Ethanol–cyclohexane	7	35	202.70	1306.77	7.63	10
	7	50	197.24	1155.43	1.53	10
Tetrachloromethane–acetonitrile	13	45	641.58	78.08	4.22	11
Tetrachloromethane–cyclohexane	9	40	−66.31	91.69	0.08	12
	9	70	27.50	−2.87	0.09	12
Benzene–cyclohexane	7	39.99	−16.01	126.69	0.76	12
Acetone–acetonitrile	10	45	−61.63	120.93	0.67	13
Acetone–cyclohexane	12	25	123.68	435.40	8.78	14
	9	45	90.11	431.88	10.40	15
Ethanol–water	10	25	155.60	30.48	8.47	16
	13	40	78.55	153.28	12.68	17
	13	55	48.38	243.38	6.25	17
Acetonitrile–2-propanol	15	50	70.27	356.68	2.69	18
Methyl acetate–cyclohexane	8	35	182.19	250.13	13.72	19
	9	45	163.07	251.65	16.17	19
Ethanol–benzene	11	25	75.30	917.18	3.11	20
	12	45	82.40	768.33	1.52	21
Methanol–tetrachloromethane	9	35	122.16	1161.86	1.97	22
	6	55	138.36	1067.35	13.16	23
Methyl acetate–methanol	7	20	371.89	55.80	7.40	24
	7	30	356.37	27.63	14.30	24
Methanol–benzene	9	35	85.33	836.10	4.54	22
	9	55	80.04	825.10	5.45	22
Benzene–nitromethane	10	25	166.91	105.83	8.55	25
Benzene–furfural	7	25	358.80	−17.88	1.78	26
Benzene–2,2,4-trimethylpentane	7	35	2.77	133.53	0.37	27
	8	45	1.40	129.44	0.07	27
Cyclohexane–2,2,4–trimethylpentane	8	45	−20.96	30.70	1.79	28
Trichloromethane–benzene	19	50	−93.94	13.46	5.03	29
Methyl acetate–benzene	17	50	242.47	−164.78	5.00	29
Methyl acetate–trichloromethane	16	50	−185.65	12.47	6.65	29

TABLE 1 (continued)

System(1)–(2)	No. of points	Temp. (°C)	Energy parameters, K		Variance of fit	Ref.
			a_{12}	a_{21}		
Trichloromethane–methanol	25	50	933.25	−101.41	16.36	30
Acetone–trichloromethane	29	50	−178.99	0.73	11.69	30
Acetone–methanol	35	50	266.93	−26.70	12.39	30
Water–methanol	10	25	−67.54	85.58	2.15	16
Methanol–ethanol	11	25	−260.48	253.80	29.37	16
Ethanol–toluene	10	35	78.47	862.56	1.74	31
	10	55	84.08	774.14	1.74	31
2-Propanol–cyclohexane	9	50	137.84	916.74	6.63	32
n-Hexane–ethanol	16	40	1324.64	231.27	9.07	9
2-Propanol–benzene	12	45	122.05	532.56	1.63	33
1-Propanol–benzene	11	45	96.05	616.40	0.45	34
Benzene-1-butanol	9	45	543.70	100.09	0.68	34
Nitromethane–benzene	12	45	69.51	206.08	2.41	35
Nitromethane–tetrachloromethane	12	45	132.06	542.52	5.66	35
Ethanol-2-butanone	12	25	30.76	297.37	2.18	20
2-Butanone–benzene	10	25	−177.36	328.99	1.96	20
Tetrachloromethane–benzene	8	40	−4.54	12.68	0.08	36
n-Hexane–nitroethane	13	45	381.66	159.44	5.33	37
Acetonitrile–*n*-heptane	S[a]	45	391.74	959.88		7
Benzene–water	S	25	1447.40	906.28		38
Acetonitrile–cyclohexane	S	40	375.11	931.03		18
	S	45	373.36	909.36		39
	S	50	363.20	876.18		18
Methanol–cyclohexane	S	25	347.73	1349.8		18
Furfural–cyclohexane	S	25	361.42	752.69		40
Furfural–2,2,4-trimethylpentane	S	25	342.76	822.91		40
Nitromethane–cyclohexane	S	25	443.80	786.79		40

[a] S=Solubility data.

present examples of typical sets of binary data which I have correlated. The original UNIQUAC equation predicts phase separation for the ethanol-*n*-octane system [2]. The extended UNIQUAC equation correlates this system with good accuracy (Fig. 1) and works well for many types of mixtures including partially miscible ones. Table 2 represents the predicted results of vapor–liquid equilibria for six representative ternary systems. Calculated

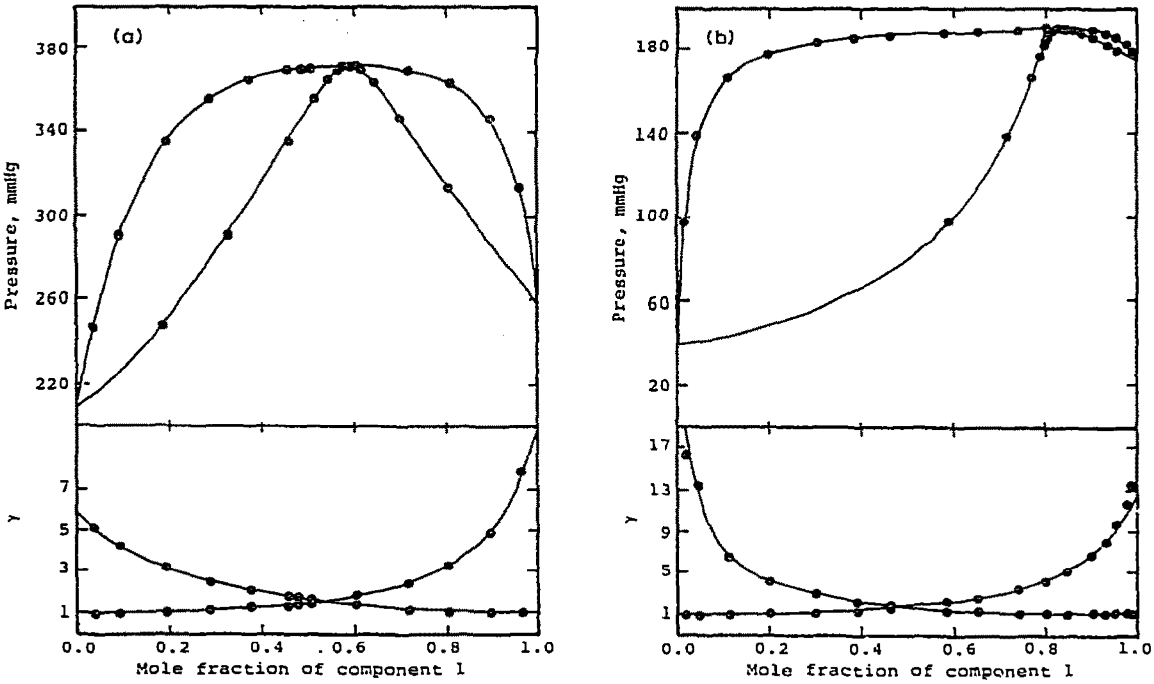

Fig. 1. Representation of vapor–liquid equilibria for (a) the tetrachloromethane(1)–acetonitrile(2) and (b) the ethanol(1)–*n*-octane(2) systems at 45°C. ———, Calculated; ●, experimental. Data of Brown and Smith [11] for tetrachloromethane–acetonitrile; data of Boublikova and Lu [8] for ethanol–*n*-octane.

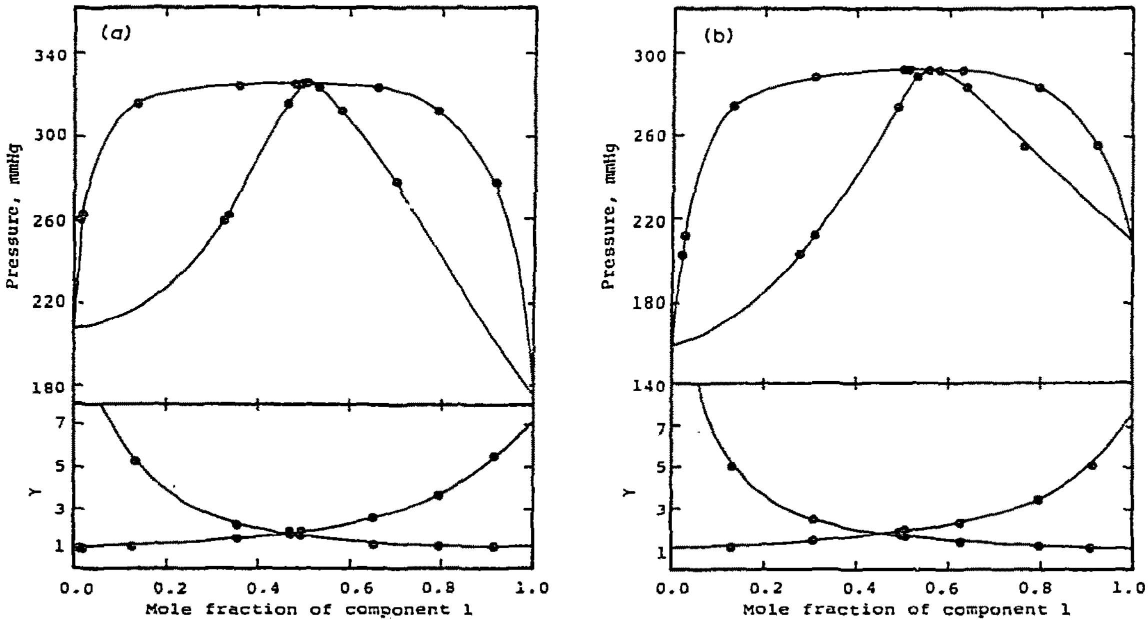

Fig. 2. Representation of vapor–liquid equilibria for (a) the methanol(1)–tetrachloromethane(2) and (b) the methanol(1)–benzene(2) systems at 35°C. ———, Calculated; ●, experimental. Data of Scatchard et al. [22].

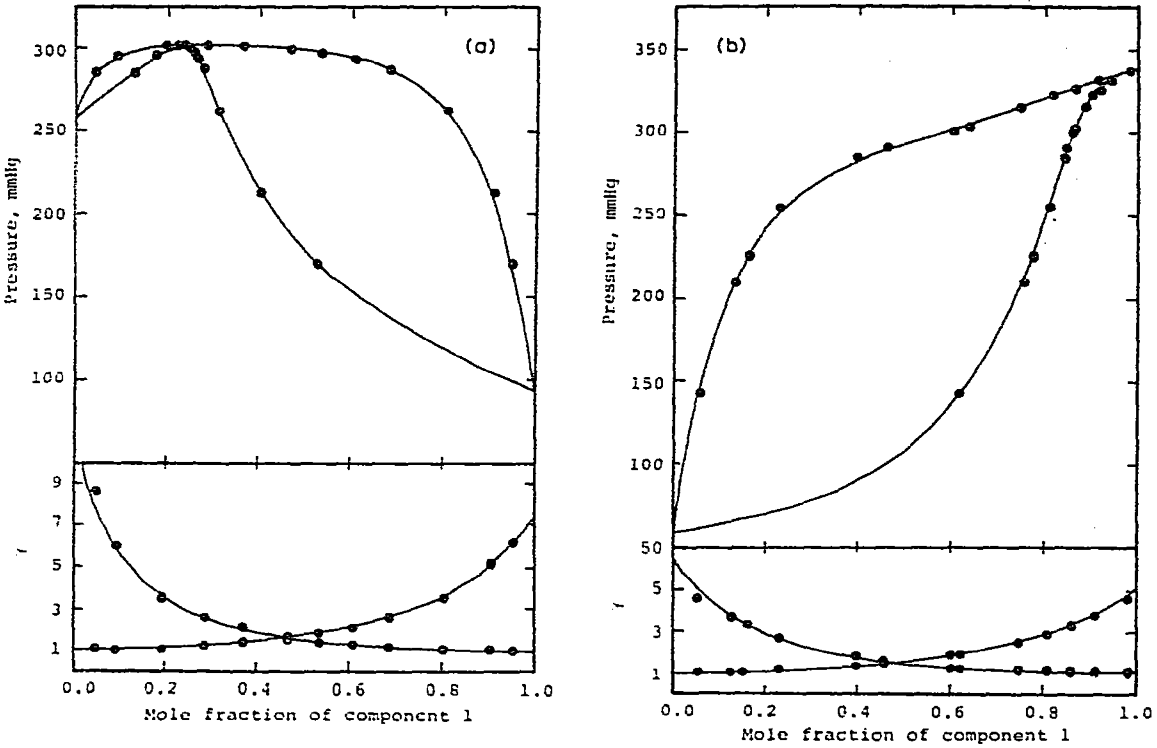

Fig. 3. Representation of vapor–liquid equilibria for (a) the nitromethane(1)–tetrachloromethane(2) and (b) the *n*-hexane(1)–nitroethane(2) systems at 45°C. ———, Calculated; ●, experimental. Data of Brown and Smith [35] for nitromethane–tetrachloromethane; data of Edwards [37] for *n*-hexane–nitroethane.

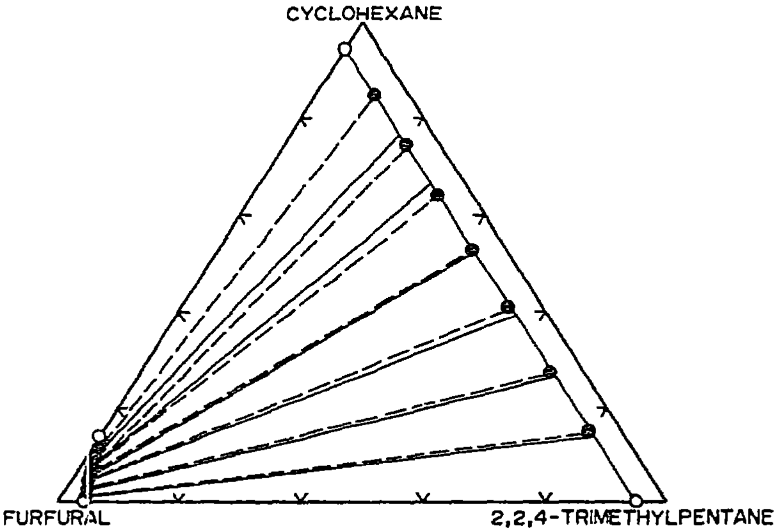

Fig. 4. Predicted ternary liquid–liquid equilibria for the furfural–cyclohexane-2,2,4-trimethylpentane system at 25°C. ●— — —, Tie-line data of Henty et al. [41]; ———, calculated. Concentrations are in mole fractions.

TABLE 2

Prediction of ternary vapor–liquid equilibria

System	Temp. (°C)	No. of points	Absolute arith. mean dev.			Ref.
			Vapor compn (mole %)	Press. (mm Hg)	Press. (%)	
Acetonitrile– benzene– *n*-heptane	45	51	0.48 0.40 0.43	3.6	1.24	7
Water– methanol– ethanol	25	37	0.42 0.95 0.94	1.3	1.88	16
Ethanol– 2-butanone– benzene	25	33	0.32 0.24 0.36	1.0	0.90	20
Methanol– tetrachloromethane– benzene	55	8	0.25 0.15 0.13	1.4	0.20	23
Methyl acetate– trichloromethane– benzene	50	66	0.32 0.29 0.32	3.9	0.92	29
Acetone– trichloromethane– methanol	50	30	0.95 1.11 1.05	8.5	1.57	30

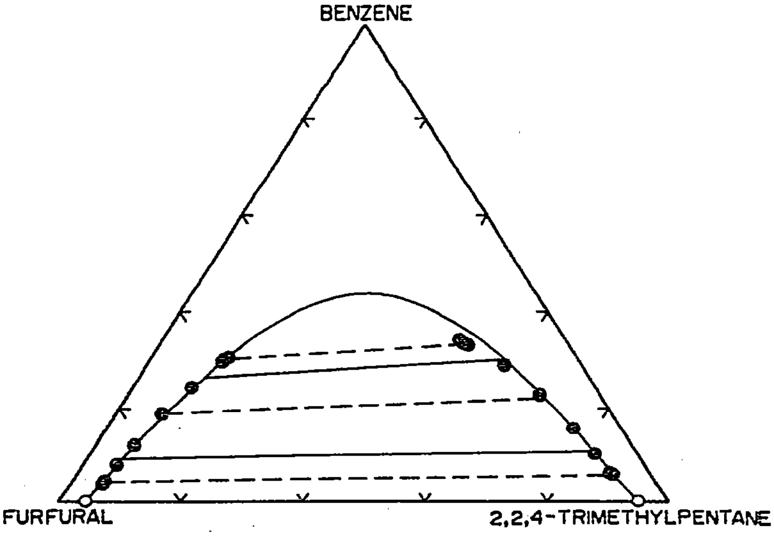

Fig. 5. Predicted ternary liquid–liquid equilibria for the furfural–benzene-2,2,4-trimethylpentane system at 25°C ●— — —, Tie-line data of Henty et al. [41]; ————, calculated. Concentrations are in mole fractions.

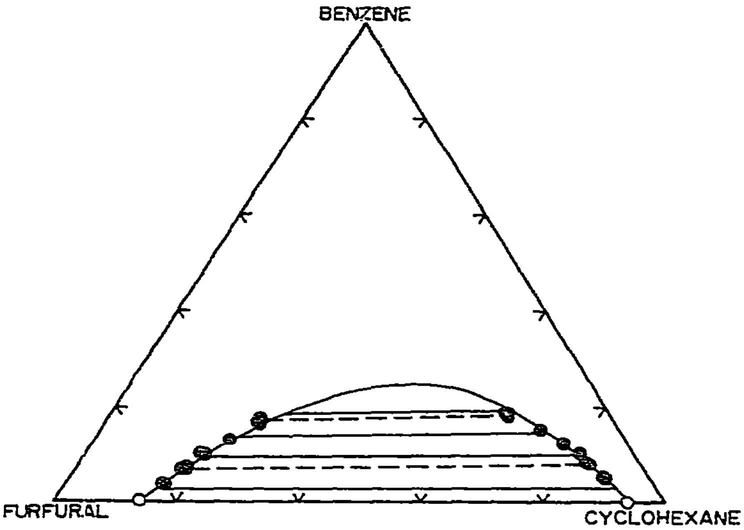

Fig. 6. Predicted ternary liquid–liquid equilibria for the furfural–benzene–cyclohexane system at 25°C. ●— — —, Tie-line data of Henty et al. [41]; ————, calculated. Concentrations are in mole fractions.

vapor-phase compositions and pressures agree well with experimental values. Especially excellent agreement is obtained for the methanol–tetrachloromethane–benzene system, probably because the binary data of superior quality are adequately correlated with the extended UNIQUAC equation.

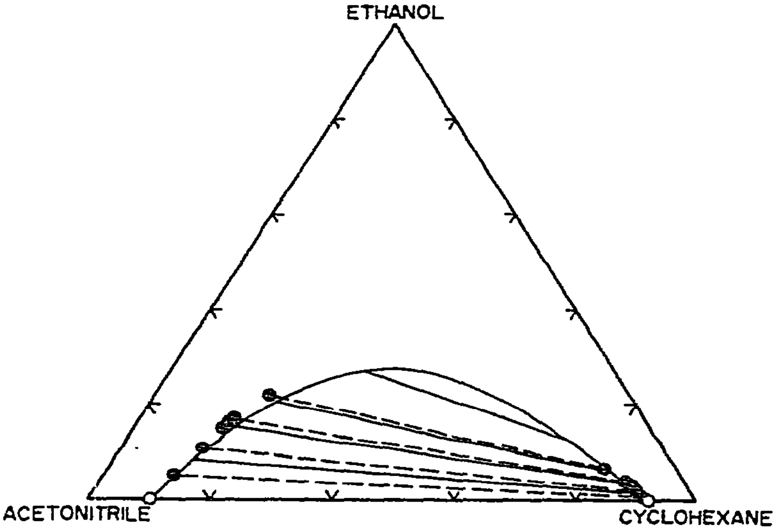

Fig. 7. Predicted ternary liquid–liquid equilibria for the acetonitrile–ethanol–cyclohexane system at 40°C. ●— — —, Tie-line data of Nagata and Katoh [18]; ————, calculated. Concentrations are in mole fractions.

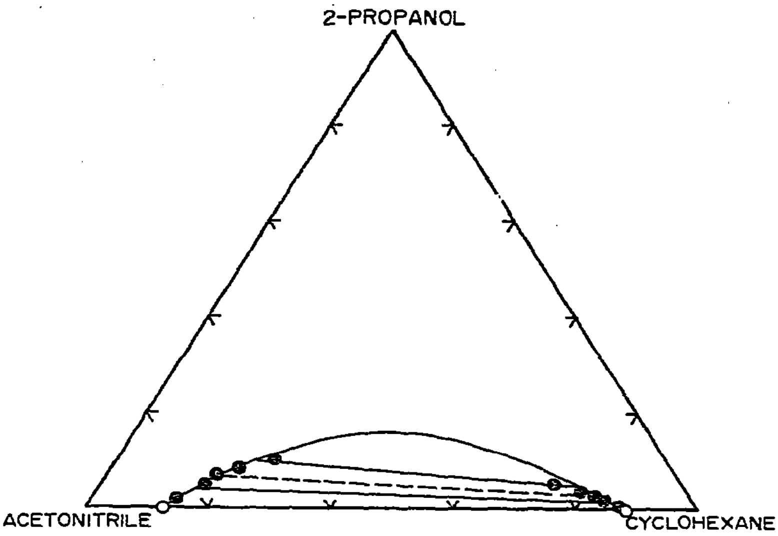

Fig. 8. Predicted ternary liquid–liquid equilibria for the acetonitrile–2-propanol–cyclohexane system at 50°C. ●— — —, Tie-line data of Nagata and Katoh [18]; ————, calculated. Concentrations are in mole fractions.

For systems where two binaries are partially miscible and the third binary is entirely miscible, it is rather easy to predict the ternary equilibria as pointed out by several authors. For systems where only one binary is partially miscible, liquid–liquid equilibrium calculation is more sensitive than vapor–liquid equilibrium calculation to small errors in the predicted activity coefficient values. Figures 4–14 present several representative exam-

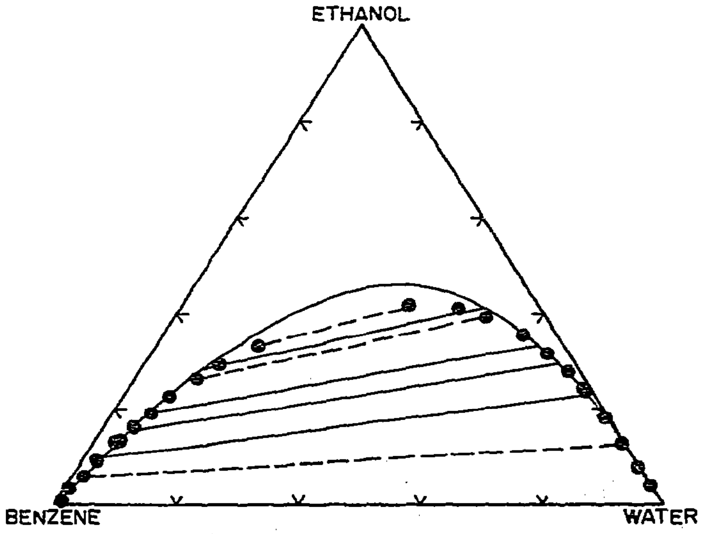

Fig. 9. Predicted ternary liquid–liquid equilibria for the benzene—ethanol–water system at 25°C. ●— — —, Tie-line data of Bancroft and Hubard [42]; ————, calculated. Concentrations are in mole fractions.

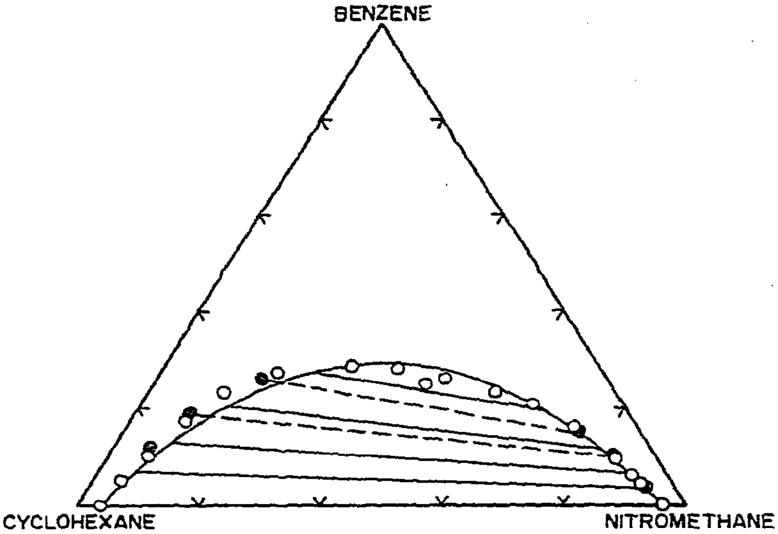

Fig. 10. Predicted ternary liquid–liquid equilibria for the cyclohexane–benzene–nitromethane systems at 25°C. Experimental data of Weck and Hunt [43], ○ (solubility), ●— — — (tie-line); ————, calculated. Concentrations are in mole fractions.

ples to indicate successful predictions of ternary liquid–liquid equilibria by using only binary parameters. Anderson and Prausnitz [2] have much improved ternary prediction of liquid–liquid equilibria for mixtures containing methanol from binary data with their modified UNIQUAC equation, and have used a calculation method which utilizes some ternary data in the

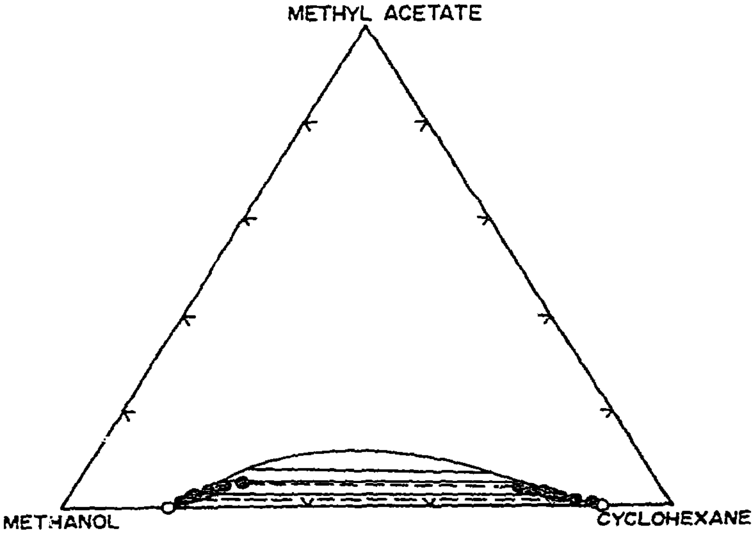

Fig. 11. Predicted ternary liquid–liquid equilibria for the methanol–methyl acetate–cyclohexane system at 25°C. ●— — —, Tie-line data of Sugi et al. [44]; —, calculated. Concentrations are in mole fractions.

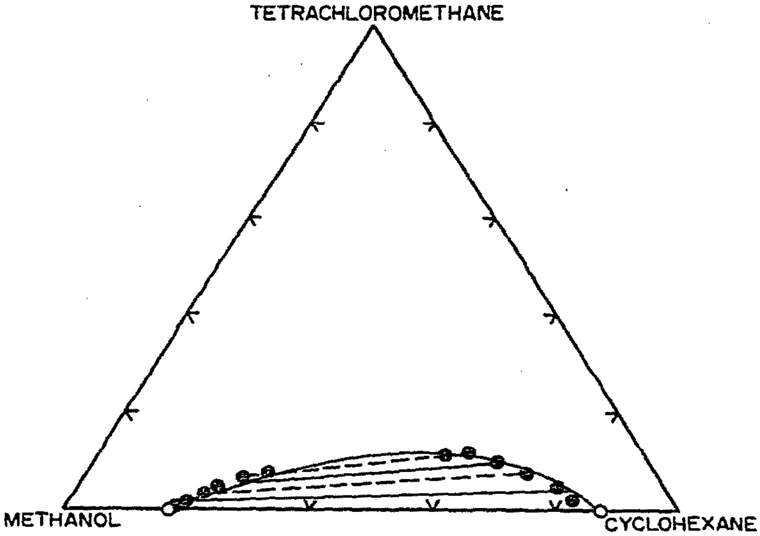

Fig. 12. Predicted ternary liquid–liquid equilibria for the methanol–tetrachloromethane–cyclohexane system at 25°C. ● — — —, Tie-line data of Yasuda et al. [45]; ————, calculated. Concentrations are in mole fractions.

parameter estimation to represent well the ternary systems. In most cases only a single tie-line is required. Inclusion of the single ternary tie-line into the method of data reduction gives satisfactory ternary representation but does so at a high price of some loss of accuracy in the representation of vapor–liquid equilibria for component binaries as shown by results for the acetonitrile–benzene–*n*-heptane system. On the contrary, the present method

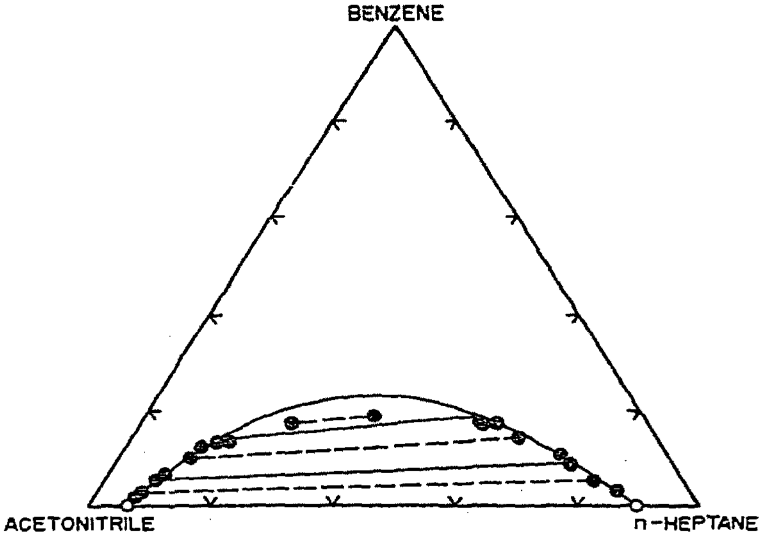

Fig. 13. Predicted ternary liquid–liquid equilibria for the acetonitrile–benzene–*n*-heptane system at 45°C. ● — — —, Tie-line data of Palmer and Smith [7]; ————, calculated. Concentrations are in mole fractions.

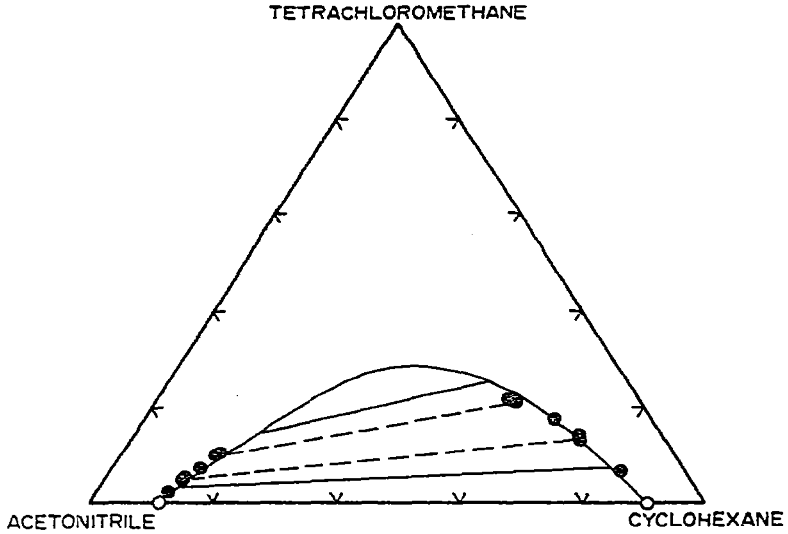

Fig. 14. Predicted ternary liquid–liquid equilibria for the acetonitrile–tetrachloromethane–cyclohexane system at 45°C. ●— — —, Tie-line data of Nagata et al. [46]: ————, calculated. Concentrations are in mole fractions.

can predict considerably well ternary liquid–liquid equilibria without loss of accuracy in the representation of the binary vapor–liquid equilibria. The extended UNIQUAC equation as well as other common models for the excess Gibbs energy cannot properly describe the flat region near the plait point.

REFERENCES

1 D.S. Abrams and J.M. Prausnitz, AIChE J., 21 (1975) 116.
2 T.F. Anderson and J.M. Prausnitz, Ind. Eng. Chem., Process Des. Dev., 17 (1978) 552, 561.
3 I. Nagata and K. Katoh, Fluid Phase Equilibria, 5 (1981) 225.
4 G. Maurer and J.M. Prausnitz, Fluid Phase Equilibria, 2 (1978) 91.
5 J.M. Prausnitz, T.F. Anderson, E.A. Grens, C.A. Eckert, R. Hsieh and J.P. O'Connell, Computer Calculations for Multicomponent Vapor–Liquid and Liquid–Liquid Equilibria, Prentice-Hall, Englewood Cliffs, 1980.
6 J.G. Hayden and J.P. O'Connell, Ind. Eng. Chem., Process Des. Dev., 14 (1975) 209.
7 D.A. Palmer and B.D. Smith, J. Chem. Eng. Data, 17 (1972) 71.
8 L. Boublikova and B.C.-Y. Lu, J. Appl. Chem. (London), 19 (1969) 89.
9 H. Sugi and T. Katayama, J. Chem. Eng. Jpn., 11 (1978) 167.
10 G. Scatchard and F.G. Satkiewicz, J. Am. Chem. Soc., 86 (1964) 130.
11 I. Brown and F. Smith, Aust. J. Chem., 7 (1954) 269.
12 G. Scatchard, S.E. Wood and J.M. Mochel, J. Am. Chem. Soc., 61 (1939) 3206.
13 I. Brown and F. Smith, Aust. J. Chem., 13 (1960) 30.
14 P.S. Puri, J. Polak and J.A. Ruether, J. Chem. Eng. Data, 19 (1974) 87.
15 A.N. Marinichev and M.P. Susarev, Zh. Prikl. Khim., 38 (1965) 378.

16 D.J. Hall, C.J. Mash and R.C. Pemberton, Natl. Phys. Lab. Rep. Chem., 95 (1979).
17 I. Mertl, Collect. Czech. Chem. Commun., 37 (1972) 366.
18 I. Nagata and K. Katoh, Thermochim. Acta, 39 (1980) 45.
19 I. Nagata, T. Ohta, T. Takahashi and K. Gotoh, J. Chem. Eng. Jpn., 6 (1973) 129.
20 T. Ohta, J. Koyabu and I. Nagata, Fluid Phase Equilibria, 7 (1981) 65.
21 I. Brown and F. Smith, Aust. J. Chem., 7 (1954) 264.
22 G. Scatchard, S.E. Wood and J.M. Mochel, J. Am. Chem. Soc., 68 (1946) 1960.
23 G. Scatchard and L.B. Ticknor, J. Am. Chem. Soc., 74 (1952) 3724.
24 V. Bekárek, Collect. Czech. Chem. Commun., 33 (1968) 2608.
25 D.F. Saunders and A.J.B. Spaull, Z. Phys. Chem. N.F., 28 (1961) 332.
26 J. Kenny, Chem. Eng. Sci., 6 (1957) 116.
27 S. Weissman and S.E. Wood, J. Chem. Phys., 32 (1960) 1153.
28 R. Battino, J. Phys. Chem., 70 (1966) 3408.
29 I. Nagata and H. Hayashida, J. Chem. Eng. Jpn., 3 (1970) 161.
30 W.H. Severns, Jr., A. Sesonske, R.H. Perry and R.L. Pigford, AIChE J., 1 (1955) 401.
31 C.B. Kretschmer and R. Wiebe, J. Am. Chem. Soc., 71 (1949) 1793.
32 I. Nagata, T. Ohta and Y. Uchiyama, J. Chem. Eng. Data, 18 (1973) 54.
33 I. Brown, W. Fock and F. Smith, Aust. J. Chem., 9 (1956) 364.
34 I. Brown and F. Smith, Aust. J. Chem., 12 (1959) 407.
35 I. Brown and F. Smith, Aust. J. Chem., 8 (1955) 501.
36 G. Scatchard, S.E. Wood and J.M. Mochel, J. Am. Chem. Soc., 62 (1940) 712.
37 J.B. Edwards, Ph.D. Dissertation, Georgia Institute of Technology, 1962.
38 J. Polak and B.C.-Y. Lu, Can. J. Chem., 51 (1973) 4018.
39 I. Nagata, Y. Nakamiya, K. Katoh and J. Koyabu, Thermochim. Acta, 45 (1981) 153.
40 J.M. Sørensen and W. Arlt, Liquid–Liquid Equilibrium Data Collection, Binary Systems, DECHEMA Chem. Data Ser., Vol. V, Part 1, Frankfurt/M, 1979.
41 C.J. Henty, W.J. McManamey and R.G.H. Prince, J. Appl. Chem. (London), 14 (1964) 148.
42 W.B. Bancroft and S.S. Hubard, J. Am. Chem. Soc., 64 (1942) 347.
43 H.I. Weck and H. Hunt, Ind. Eng. Chem., 46 (1954) 2521.
44 H. Sugi, T. Nitta and T. Katayama, J. Chem. Eng. Jpn., 9 (1976) 12.
45 M. Yasuda, H. Kawade and T. Katayama, Kagaku Kogaku Ronbunshu, 1 (1975) 172.
46 I. Nagata, K. Katoh and J. Koyabu, Thermochim. Acta, 47 (1981) 225.