Thermochimica Acta, 117 (1987) 271–281
Elsevier Science Publishers B.V., Amsterdam – Printed in The Netherlands

THE SIMULTANEOUS CORRELATION OF EXCESS GIBBS ENERGIES OF BINARY SYSTEMS AT DIFFERENT TEMPERATURES BY MEANS OF THE UNIQUAC EQUATION

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(Received 19 January 1987)

ABSTRACT

The temperature dependence of the UNIQUAC interaction parameters A_{12} and A_{21} is introduced in the form of the equation $A_{ij} = a_{ij} + b_{ij}/T$. Values of the temperature-independent parameters a_{ij} and b_{ij} are obtained for several binary systems. This procedure enables us to use a unique set of four parameters (a_{12}, b_{12}, a_{21} and b_{21}) to accurately represent the excess Gibbs energy (G^E) in a certain temperature range provided that values for G^E derived from vapour–liquid equilibrium data show a regular variation with temperature. The correlation equation for A_{12} and A_{21} proposed by Luecke is also confirmed for the systems considered in this study.

INTRODUCTION

In recent years, much effort has been devoted to the development of equations for the excess Gibbs energy (G^E) of liquid solutions. The equations based on the concept of local composition have been of great practical value and among them one of the most effective seems to be the UNIversal QUAsiChemical (UNIQUAC) equation introduced by Abrams and Prausnitz [1]. Many sets of experimental data for binary systems have been analysed using this equation which requires two binary interaction parameters, A_{12} and A_{21}, and pure-component structural parameters. Data for higher order systems can be also analysed using this equation and two interaction parameters for each binary combination. Values for the parameters A_{12} and A_{21} must be found from binary experimental data: vapour–liquid equilibrium (VLE) data at constant temperature, total pressure data at constant temperature, VLE data at constant pressure, etc. Different sets of values for the pair (A_{12}, A_{21}) can be found to represent a set of data for a particular system within a certain confidence level [2]. It has been also observed that while both parameters may vary over a wide range, their variations are related so that the set of (A_{12}, A_{21}) values lies within an area of elliptical contours for the chosen confidence level. On the other

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hand, the areas corresponding to pairs of parameters obtained from VLE data taken at different temperatures very often do not overlap [3]. This indicates a significant dependence of the (A_{12}, A_{21}) values on temperature.

The strong correlation of the interaction parameters has been confirmed by Luecke [4] who studied the (A_{12}, A_{21}) values published in the Dechema Data Series [5] for the systems for which the largest numbers of sets of experimental data are available. For instance the parameters of the ethanol(1) + water(2) system (for which 43 sets of data taken at temperatures ranging from 278 to 418 K are available) are shown to be correlated by the equation

$$(A_{12}+c)(A_{21}+c)=2\times 10^{6} \qquad (1)$$

where $c=1290$ when A_{12} and A_{21} are given in cal mol^{-1}.

Hyperbolas with varying values of c were obtained by Luecke for another eight binary systems. No significant correlation between temperature and the location of the pair (A_{12}, A_{21}) on the hyperbola was found. Therefore, it is necessary to assume relations equating A_{12} and A_{21} to a certain function of temperature if the temperature dependence of these parameters is to be included in the UNIQUAC equation. Values for the new parameters appearing in the equations relating A_{12} and A_{21} with temperature could be obtained using several sets of VLE data taken at different temperatures. Consequently, a unique set of parameters would represent the behaviour of the system in a wide range of temperatures.

INTERACTION PARAMETERS IN THE UNIQUAC EQUATION

The UNIQUAC equation for the excess Gibbs energy may be written as the sum of a combinatorial and a residual contributions

$$G^{\mathrm{E}}=G_{\mathrm{c}}^{\mathrm{E}}+G_{\mathrm{r}}^{\mathrm{E}} \qquad (2)$$

The combinatorial term depends on the mole fractions, x, the area fractions, θ, and the segment fractions, Φ, and is given by

$$G_{\mathrm{c}}^{\mathrm{E}}/RT=x_1\ \ln(\Phi_1/x_1)+x_2\ \ln(\Phi_2/x_2)+(Z/2)$$
$$\times(q_1x_1\ \ln(\theta_1/\Phi_1)+q_2x_2\ \ln(\theta_2/\Phi_2)) \qquad (3)$$

where Z is the coordination number which is set equal to 10. Φ and θ can be calculated using the parameters r and q, respectively, and are given by

$$\Phi_1=x_1r_1/(x_1r_1+x_2r_2) \qquad \Phi_2=x_2r_2/(x_1r_1+x_2r_2) \qquad (4)$$

$$\theta_1=x_1q_1/(x_1q_1+x_2q_2) \qquad \theta_2=x_2q_2/(x_1q_1+x_2q_2) \qquad (5)$$

r and q are pure component molecular structure constants depending on molecular size and external surface areas.

The residual term of G^E is given by

$$G_r^E/RT = -q_1'x_1 \ln(\theta_1' + \theta_2'\Gamma_{21}) - (q_2'x_2 \ln(\theta_2' + \theta_1'\Gamma_{12})) \tag{6}$$

The parameter q' was later introduced to be used in mixtures containing water or alcohols for which the surface of interaction q' is smaller than the geometrical external surface q. Values for the structural parameters have been tabulated by Prausnitz et al. [3]. Γ_{12} and Γ_{21} are given by

$$\Gamma_{12} = \exp(-(u_{12} - u_{22})/RT) = \exp(-A_{12}/RT) \tag{7}$$

$$\Gamma_{21} = \exp(-(u_{21} - u_{11})/RT) = \exp(-A_{21}/RT) \tag{8}$$

where u_{12} and u_{21} are the characteristic energies of each binary combination, also referred to as A_{12} and A_{21}, the two binary interaction parameters. In order to include in the UNIQUAC equation the temperature dependence of A_{12} and A_{21}, we have assumed the following relations

$$A_{12} = a_{12} + b_{12}/T \tag{9}$$

$$A_{21} = a_{21} + b_{21}/T \tag{10}$$

These relations are the same assumed by Prausnitz et al. [3] in their study of the isothermal VLE data of the ethanol (1) + cyclohexane (2) system. In the next section, isothermal VLE data of several binary systems will be analyzed using the UNIQUAC model and eqns. (9) and (10). The use of a unique set of parameters (a_{12}, b_{12}, a_{21}, b_{21}) which can be used to simultaneously represent several isothermal sets of data of a binary system will be discussed.

RESULTS AND DISCUSSION

Table 1 lists the components of the binary systems, the conditions of the VLE data, the values of the parameters A_{12} and A_{21}, the standard deviation in pressure, σ_p, and the source of data. When the data are isothermal, both the temperature and the range of total pressures are indicated. When the data are isobaric, the pressure and the temperature interval are stated. Values of A_{12} and A_{21} were calculated using a non-linear regression method based on the maximum likelihood principle [30]. Values for the structural parameters, critical constants, association parameters, constants for fugacity equation, etc., required for this calculation were taken from Prausnitz et al. [3]. When vapour compositions, y, are reported together with liquid compositions, x, vapour pressure, P, and temperatures, T, the regression is performed on the (P, T, x, y) data set. When vapour compositions are not reported, the (P, T, x) data set is considered.

Figure 1 shows a plot of values of A_{12} and A_{21} taken from Table 1. The correlation hyperbola obtained by Luecke [4] for the ethanol (1) + water (2) system is also shown for comparison. Points are scattered because of the different nature of systems studied. Figures 2 and 3 show the typical results obtained when values of A_{12} and A_{21} calculated in this study and those reported by Gmehling and Onken [5] for the same sets of VLE data of methanol (1) + ethanol (2) and cyclohexane (1) + 2,2,4-trimethylpentane (2)

TABLE 1

Binary systems studied, conditions and source of VLE data, values of the UNIQUAC parameters A_{12} and A_{21}, and standard deviation in pressure, σ_p

System No.	Components	T (K)	p (kPa)	A_{12} (J mol^{-1})	A_{21} (J mol^{-1})	σ_p (kPa)	Ref.
I	n-Hexane (1) + n-hexadecane (2)	293.15	0.4– 13	−447.2	621.4	0.01	6
		303.15	0.7– 21	−346.8	499.8	0.01	6
		313.15	1– 31	−383.6	543.8	0.02	6
		323.15	1– 44	−422.3	590.3	0.03	6
		333.15	2– 62	−441.4	614.7	0.04	6
II	Cyclohexane (1) + 2,2,4-trimethylpentane (2)	308.15	13– 19	−1001.9	1268.7	0.03	7
		318.15	17– 28	−1011.1	1257.8	0.05	7
		328.15	26– 41	−936.4	1136.8	0.02	7
		338.15	37– 59	−852.1	1003.0	0.03	7
		348.15	57– 81	−896.7	1053.8	0.03	7
III	Benzene (1) + 2,2,4-trimethylpentane (2)	298.15	7– 12	−569.6	1329.3	0.40	8
		298.15	8– 13	−1030.4	2299.7	0.21	9
		308.15	12– 20	−270.7	841.1	0.04	10
		318.15	18– 30	127.1	357.6	0.14	10
		328.15	27– 44	−143.1	648.4	0.05	10
		338.15	45– 62	−124.1	604.6	0.08	10
		348.15	54– 86	−266.0	747.9	0.04	10
		82–97	101.35	−373.4	686.7	0.49	8
		80–96	101.35	−371.1	849.8	0.10	11
IV	Hexafluorobenzene (1) + cyclohexane (2)	303.15	17– 21	−27.97	17.54	0.16	12
		313.15	27– 32	32.81	−96.77	0.20	12
		323.15	39– 47	446.3	−519.4	0.21	12
		333.15	57– 67	−113.4	−50.94	0.25	12
		343.15	80– 93	290.7	−481.6	0.18	12
V	Hexafluorobenzene (1) + toluene (2)	303.15	6– 13	−1705.1	1143.7	0.03	12
		313.15	9– 21	−1708.1	1135.9	0.02	12
		323.15	14– 31	−1695.3	1097.2	0.03	12
		333.15	21– 46	−1674.6	1046.5	0.04	12
		343.15	31– 66	−1646.0	986.1	0.05	12
VI	Hexafluorobenzene (1) + *p*-xylene (2)	303.15	14– 16	−1503.6	571.9	0.06	12
		313.15	22– 24	−1505.4	577.6	0.07	12
		323.15	34– 36	−1650.9	787.7	0.30	12
		333.15	50– 53	−1544.4	639.8	0.11	12
		343.15	71– 74	−1530.8	604.8	0.37	12

TABLE 1 (continued)

System No.	Components	T (K)	p (kPa)	A_{12} (J mol^{-1})	A_{21} (J mol^{-1})	σ_p (kPa)	Ref.
VII	Methanol (1) + ethanol (2)	273.15	2 - 4	−788.1	−2069.1	0.04	13
		293.15	6 - 12	−2163.2	4112.2	0.15	13
		298.15	9 - 16	683.4	−532.5	0.04	14
		303.15	12 - 21	1151.9	−684.5	0.10	13
		313.15	20 - 33	−1847.7	3288.1	0.17	13
		323.15	32 - 53	439.9	−68.68	0.05	13
		333.15	51 - 79	−1778.3	3323.4	0.12	13
		353.15	114 -162	−2433.8	4423.2	0.26	13
		373.15	236 -318	−2158.0	3758.4	0.08	13
		373.15	233 -344	7455.5	−2709.7	0.21	15
		393.15	427 -621	8354.2	−2957.1	0.17	15
		338–351	101.35	5903.0	−2510.4	0.14	16
		339–350	101.35	5297.7	−2387.1	0.15	17
		339–350	101.35	3293.0	−1668.3	0.16	18
		339–351	101.35	4472.1	−2006.0	0.16	19
VIII	Methanol (1) + 1-propanol (2)	273.15	0.7- 4	4081.9	−1650.6	0.06	13
		283.15	2 - 7	28.56	490.8	0.05	13
		293.15	3 - 12	−766.7	1799.9	0.07	13
		303.15	6 - 20	3029.8	−1066.2	0.02	13
		313.15	10 - 33	2140.8	−639.0	0.08	13
		323.15	17 - 52	3514.1	−1187.2	0.15	13
		333.15	27 - 79	4668.2	−1787.2	0.26	13
		333.17	23 - 83	3434.0	−1428.6	0.20	20
		340–362	101.35	−1366.1	4901.1	1.20	21
		339–367	101.35	4665.3	−1771.4	0.22	22
		340–361	101.35	5075.4	−1573.9	0.24	23
IX	Methanol (1) + 2-propanol (2)	328.15	33 - 66	2942.1	−1327.8	0.21	24
		338–352	101.35	2328.8	−458.0	0.39	25
		339–354	101.35	5320.7	−2417.3	0.13	26
		340–353	101.35	4502.6	−2190.9	0.15	23
		341–353	101.35	2205.7	−1456.2	0.32	27
		339–355	101.35	5006.3	−2276.9	0.07	28
X	Ethanol (1) + 1-propanol (2)	323.15	14 - 28	−389.5	92.83	0.10	29
		333.15	24 - 45	−1053.5	891.6	0.07	29
		343.15	9 - 17	4131.4	−2511.6	0.02	29
		353.15	14 - 25	3777.9	−2556.8	0.04	29
		353–367	101.35	−1252.7	293.0	0.16	23

systems are plotted. The correlation hyperbolas obtained by fitting these values of A_{12} and A_{21} to eqn. (1) are also shown in Figs. 2 and 3. The same correlation curve is adequate for the (A_{12}, A_{21}) pairs of the remaining systems. Table 2 lists the total number of pairs (A_{12}, A_{21}) (those from Table 1 and those reported by Gmehling and Onken [5]) used to establish the correlation hyperbola of each system and the values of parameter c. The

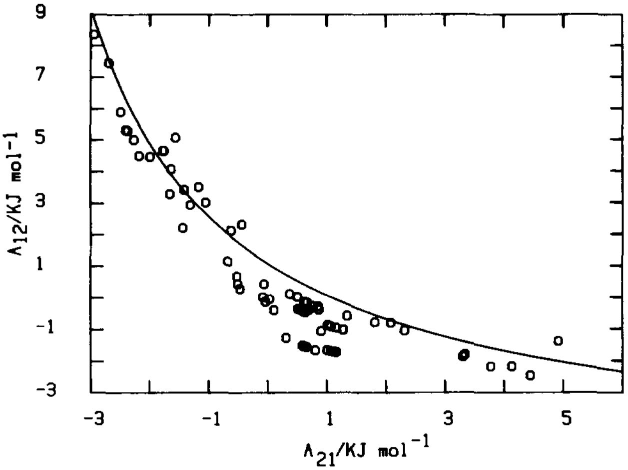

Fig. 1. Correlation of UNIQUAC interaction parameters for the binary systems listed in Table 1. The correlation curve obtained by Luecke [4] for the ethanol (1) + water (2) system is also shown for comparison purposes.

methanol (1) + ethanol (2) system was also studied by Luecke who reported a value of *c* ($c = 1411.1$ cal mol^{-1}) similar to that calculated in this study. The other eight systems studied by Luecke were formed by an alcohol as a first component and water, ethanol, chloroform, cyclohexane, benzene, 2-butanone and cyclohexanone as a second component. Therefore, it is confirmed that this correlation is also valid for (A_{12}, A_{21}) pairs of systems formed by two alcohols, two hydrocarbons or a hydrocarbon + hexafluorobenzene.

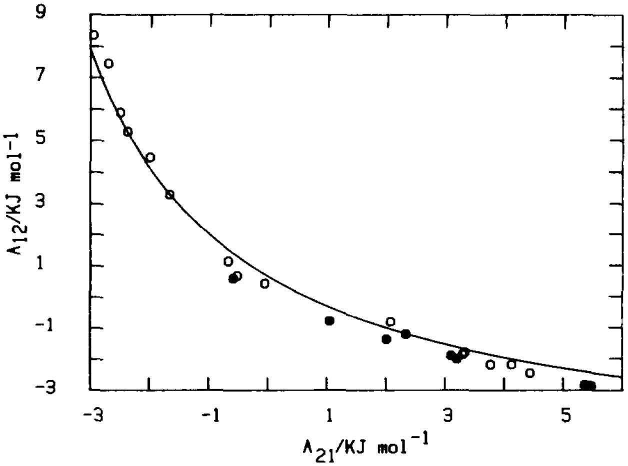

Fig. 2. Correlation of UNIQUAC interaction parameters A_{12} and A_{21} for the cyclohexane (1) + 2,2,4-trimethylpentane (2) system: (○) (A_{12}, A_{21}) pairs from Table 1; (●) (A_{12}, A_{21}) pairs calculated by Gmehling and Onken [5]; (———) correlation curve.

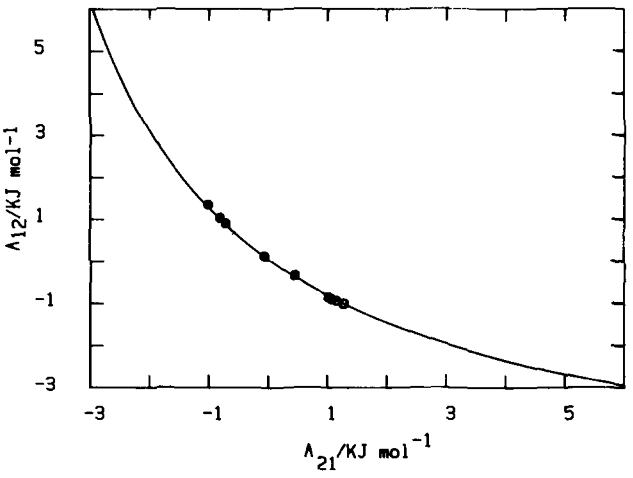

Fig. 3. Correlation of UNIQUAC interaction parameters A_{12} and A_{21} for the methanol (1) + ethanol (2) system: (○) (A_{12}, A_{21}) pairs from Table 1; (●) (A_{12}, A_{21}) pairs calculated by Gmehling and Onken [5]; (———) correlation curve.

Isothermal VLE data for the methanol (1) + 2-propanol (2) system were taken only at one temperature. The excess Gibbs energy values which can be obtained at several temperatures and mole fractions from the isothermal VLE data of the other nine systems studied were used to check the performance of the UNIQUAC equation with four adjustable parameters. Values of a_{12}, b_{12}, a_{21} and b_{21} can be calculated by minimizing the sum of squares of the differences between these values of G^E and those obtained from the UNIQUAC equation. Except for data taken at 298.15 K for the benzene (1) + 2,2,4-trimethylpentane (2) system, the values of G^E were reported together with the (P, T, x, y) measurements for systems I–VI. For a particular system, values of G^E always show a regular variation with

TABLE 2

Correlation of UNIQUAC interaction parameters according to eqn. (1)

System No.	No. of (A_{12}, A_{21}) pairs	c (J mol^{-1})
I	10	5856.8
II	10	5892.0
III	18	5677.3
IV	10	5755.1
V	11	6073.5
VI	10	6245.9
VII	23	5591.0
VIII	18	5378.8
IX	12	5647.1
X	10	5994.0

temperature. For some systems, G^E values are small, thus indicating a nearly ideal behaviour. For other systems, these G^E values are considerably endothermic or exothermic. This variety of behaviour makes the group of systems studied a convenient sample to test a modification in an excess Gibbs energy equation.

G^E values at 298.15 K for the benzene (1) + 2,2,4-trimethylpentane (2) system were obtained from VLE data taken by Chu et al. [8] and Kenny [9] using Barker's method. Details of the application of the method are given elsewhere [30]. These values for the excess Gibbs energy are in disagreement with those calculated by Weissman and Wood [10] from VLE data taken at 308.15, 318.15, 328.15, 338.15 and 348.15 K. Consequently, a_{12}, b_{12}, a_{21} and b_{21} were determined from data taken in the interval 308.15–348.15 K. For the remaining five systems (I, II, IV–VI), parameters a_{12}, b_{12}, a_{21}, and b_{21} were obtained considering simultaneously all isothermal data available.

Values of G^E were not reported for most of the systems formed by two alcohols. As a part of this study, G^E values were evaluated from the (P, T, x, y) or (P, T, x) sets of data using a regression method discussed elsewhere [30]. These resulting sets of G^E values are moderately endothermic and do not show a regular variation with temperature. For a given alcohol (1) + alcohol (2) system, G^E curves corresponding to different temperatures cross each other sometimes and G^E values either increase or decrease with temperature. As can be seen in Table 1, the isothermal VLE data for these systems cover considerable temperature intervals (120 K for the methanol + ethanol (2) system, 60 K for the methanol + 1-propanol (2) system and 30 K for the ethanol + 1-propanol (2) system). However, due to the observed discrepancies, it is impossible to simultaneously consider these

TABLE 3

Values of the UNIQUAC parameters a_{12}, b_{12}, a_{21}, and b_{21} and comparison between G^E values obtained from VLE data and G^E values calculated from the UNIQUAC equation

System No.	T (K)	a_{12} (J mol^{-1})	b_{12} (J K mol^{-1})	a_{21} (J mol^{-1})	b_{21} (J K mol^{-1})	σ (J mol^{-1})	σ/G^E_{max} (%)
I	293–333	400.77	−22313.2	−1116.4	180096.5	0.4	1
II	308–348	−484.41	−41638.2	261.48	144812.4	0.3	1
III	308–348	4370.4	−477633.0	−4286.1	1223725	2.4	1
IV	303–343	−1699.4	1059841	−121.17	931806.1	2.7	1
V	303–343	611.54	−1006979	−3996.4	2855273	8.5	4
VI	303–343	6818.4	−1848355	−1798.3	−66003.3	12	3
VII	323–333	−9867.8	2848589	15907.9	−4715776	2.8	6
VIII	283–293	−25521.8	7392916	33402.4	−9457733	4.1	6
	303–323	−2780.5	1403918	1075.9	−705211.4	2.7	2
	313–323	2947.7	−447119.9	−1851.7	240794.1	4.9	4
X	323–333	14746.1	−4130114	−12846.4	3903680	6.3	4
	343–353	10103.8	−3205118	−13567.8	4787403	6.3	3

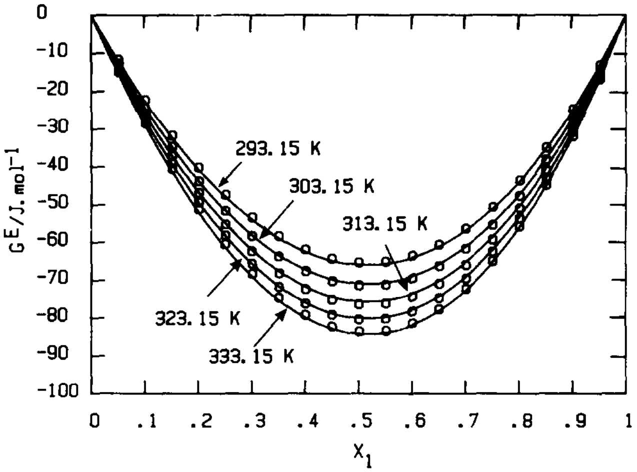

Fig. 4. Excess Gibbs energies for the n-hexane (1) + n-hexadecane (2) system: (○) experimental; (———) calculated from the UNIQUAC equation with the introduction of the temperature-independent parameters given in Table 3.

sets of data. Only those sets which are consistent with each other can be taken into account in the regression. New isothermal measurements would be necessary in order to obtain coherent sets of G^E values for these systems.

Results of the regressions are shown in Table 3. Figures 4–6 also show some typical examples of the results obtained. Values of a_{12}, b_{12}, a_{21} and b_{21}, are given in Table 3. The temperature interval, the standard deviation, σ, between G^E values obtained from VLE data and G^E values calculated

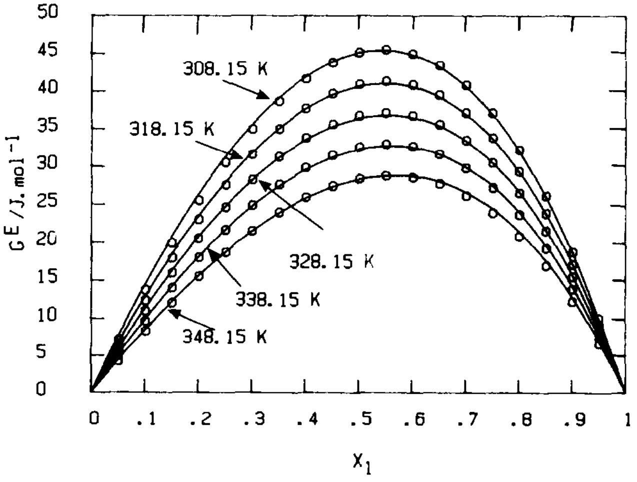

Fig. 5. Excess Gibbs energies for the cyclohexane (1) + 2,2,4-trimethylpentane (2) system: (○) experimental; (———) calculated from the UNIQUAC equation with the introduction of the temperature-independent parameters given in Table 3.

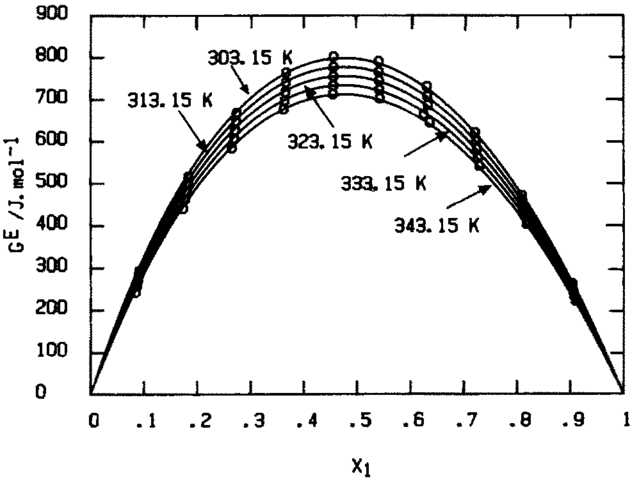

Fig. 6. Excess Gibbs energies for the hexafluorobenzene (1) + cyclohexane (2) system: (O) experimental; (———) calculated from the UNIQUAC equation with the introduction of the temperature-independent parameters given in Table 3.

from the UNIQUAC equation, and the percentage of this standard deviation with respect to the highest absolute value of G^E, are also indicated. Values of σ/G^E_{max} are as high as 6% for the systems formed by two alcohols and considerably lower (1–4%) for the other systems. This could be related to the observed discrepancies in VLE data for these systems. Values of A_{12} and A_{21} calculated from eqns. (9) and (10) by substituting the values of a_{12}, b_{12}, a_{21}, and b_{21} reported in Table 3 and the temperatures at which isothermal data were taken are also correlated by eqn. (1) after substitution of the value for the parameter c given in Table 2 for each system.

We may conclude that the success of the simultaneous correlation of excess Gibbs energies of binary mixtures evaluated at different temperatures depends on the accuracy of isothermal VLE data. When data are accurate enough so as to provide consistent sets of G^E values, a unique set of four parameters can be obtained. These parameters represent the behaviour of the system in a wide temperature interval. The sign and magnitude of the excess Gibbs energies of a system do not seem to influence the result. The modification introduced in the UNIQUAC equation seems to be valid for systems exhibiting either endothermic or exothermic values of G^E and either small, moderate or large absolute values of G^E.

REFERENCES

1 D.S. Abrams and J.M. Prausnitz, A.I.Ch.E. J., 21 (1975) 116.
2 R.C. Reid, J.M. Prausnitz and T.K. Sherwood, The Properties of Gases and Liquids, 3rd edn. McGraw-Hill, New York, 1977.

3 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, NJ, 1980.
4 R.H. Luecke, Fluid Phase Equilibria, 14 (1983) 373.
5 J. Gmehling and U. Onken, Vapor–Liquid Equilibrium Data Collection, Chemistry Data Series, Dechema, Frankfurt, 1977.
6 M.L. McGlashan and A.G. Williamson, Trans. Faraday Soc., 57 (1961) 588.
7 R. Battino, J. Phys. Chem., 70 (1966) 3408.
8 J. Gmehling and U. Onken, Vapor–Liquid Equilibrium Data Collection, Chemistry Data Series, Dechema, Frankfurt, 1977, Vol. I, Part 6B, p. 303.
9 J.W. Kenny, Chem. Eng. Sci., 6 (1957) 116.
10 S. Weissman and S.E. Wood, J. Chem. Phys., 32(4) (1960) 1153.
11 L. Sieg, Chem. Ing. Tech., 22 (1950) 322.
12 W.J. Gaw and F.L. Swinton, Trans. Faraday Soc., 12 (1968) 637, 2023.
13 G.C. Schmidt, Z. Phys. Chem., 121 (1926) 221.
14 D.J. Hall, C.J. Mash and R.C. Pemberton, NPL Rep. Chem. 95, Jan. 1979.
15 K.L. Butcher and W.I. Robinson, J. Appl. Chem., 16 (1966) 289.
16 H.H. Amer, R.R. Paxton and M. Van Winkle, Anal. Chem., 25 (1953) 1204.
17 H.H. Amer, R.R. Paxton and M. Van Winkle, Ind. Eng. Chem., 48 (1956) 142.
18 A.O. Delzenne, Ind. Eng. Chem., Chem. Eng. Data Ser., 3 (1958) 224.
19 I.P. Slovodyanik and E.M. Babuskhina, Zh. Prikl. Khim., 39 (1966) 1555.
20 C. Berro, R. Deyrieux and A. Peneloux, J. Chim. Phys., Phys.-Chim. Biol., 72 (1975) 1118.
21 D.W. Hill and M. Van Winkle, Ind. Eng. Chem., 44 (1952) 205.
22 K. Kojima, K. Tochigi, H. Seki and K. Watase, Kagaku Kogaku, 32 (1968) 149.
23 K. Ochi and K. Kojima, Kagaku Kogaku, 33 (1969) 352.
24 D.C. Freshwater and K.A. Pike, J. Chem. Eng. Data, 12 (1967) 179.
25 J.G. Dunlop, Thesis, Brooklyn University, 1948.
26 L.H. Ballard and M. Van Winkle, Ind. Eng. Chem., 44 (1952) 2450.
27 J. Kohoutova, J. Suska, J.P. Novak and J. Pick, Collect. Czech. Chem. Commun., 35 (1970) 3210.
28 L. Verhoeye and H. de Schepper, J. Appl. Chem. Biotechnol., 23 (1973) 607.
29 V.V. Udovenko and T.B. Frid, Zh. Fiz. Khim., 22 (1948) 1135.
30 R.G. Rubio, J.A.R. Renuncio and M. Díaz Peña, Fluid Phase Equilibria, 12 (1983) 217.