SIMULTANEOUS CORRELATION
OF THE CYCLOHEXANE–n-ALCOHOL SYSTEMS*

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The characterization and comparison of phase behaviour of binary systems of primary alcohols with cyclohexane was carried out by simultaneous correlating the equilibrium and the enthalpy data using the Wilson, Renon and the formerly derived multiconstant equation.

An effort has been exerted for a long time towards a more generalized description of thermodynamic behaviour of solutions the result of which would be the possibility of wider extrapolation and more accurate estimation of phase equilibria and consequently also the substantial limitation of hitherto inevitable extensive experimental measurements. The shortcoming of most those works is the circumstance that they used as a rule a casual choice of data with regard both to the character of systems and to the way and accuracy of measurements. It was so above all because, in spite of a relatively large amount of measured phase equilibria of binary systems, the area covered by the experimental data has been still sparse to enable such detailed description.

We have undertaken a systematic study of phase equilibria in homologous series, at first in binary systems of normal primary alcohols with cyclohexane. A detailed description of experimental works and correlations of single systems is presented in foregoing papers[1-3]. Considering that the experimental data for some systems have been already known from the literature, a relatively detailed system resulted from their additional measurements, which is suitable for investigating simultaneous ways of correlation of the equilibrium and the enthalpy data. This idea is not a new one and has been worked out by several authors, but either with a very narrow choice of systems, contingently with only one[4,5], or by using expressions with a large number of constants[6-8], or by separate determining constants from the equilibrium data and the enthalpy data[4,9] and the like[10-12].

In this work an attempt is made to treat the equilibrium and enthalpy data by means of three relations, *i.e.* by a multiconstant nonlinear expansion, the Wilson and the Renon equations.

THEORETICAL

In one of the foregoing papers[2], an equation was described and checked which stems out of the Redlich–Kister expansion but includes also the dependence of molar excess free enthalpy on temperature. For a dimensionless quantity g^E/RT it is possible to write it in the form

$$g^E/RT = x_1 x_2 \exp\left[(\alpha + \beta T)(x_1 - x_2)\right] \cdot$$
$$\cdot \left[A_0 + BT + A_1(x_1 - x_2) + A_2(x_1 - x_2)^2\right]. \tag{1}$$

By using

$$h^E/RT = -T\left[\frac{\partial(g^E/RT)}{\partial T}\right]_{x,\,P} \tag{2}$$

the relation is obtained for the excess enthalpy

$$h^E/RT = -Tx_1 x_2 \exp\left[(\alpha + \beta T)(x_1 - x_2)\right] \cdot$$
$$\cdot \left\{B + \beta(x_1 - x_2)\left[A_0 + BT + A_1(x_1 - x_2) + A_2(x_1 - x_2)^2\right]\right\}. \tag{3}$$

This equation provided good interpolated and extrapolated results[13,14] keeping up a good agreement in other dependences related to the excess quantities ($\log \gamma_1$, $\log \gamma_2$, $\log \gamma_1/\gamma_2$, g^M, and $\partial g^M/\partial x$). A certain disadvantage of this relation is still a large number of constants (six, for simpler systems when it is possible to put $A_2 = 0$, five), which brings about a danger of occurrence of false inflex points apart from the fact that these constants are without any physical interpretation.

Another way of the simultaneous correlation is allowed by the relation which proposed Wilson[15] on the basis of the assumption of local composition by using the Flory[16] and Huggins[17] equations. As modified by Orye and Prausnitz[18], the molar excess free enthalpy of a binary solution is described by the equation

$$g^E/RT = -x_1 \ln\left(x_1 + x_2 H_1^w\right) - x_2 \ln\left(x_2 + x_1 H_2^w\right). \tag{4}$$

where

$$H_1^w = \left(V_2^L/V_1^L\right)\exp\left(-h_1^w/RT\right) \quad \text{and} \quad H_2^w = \left(V_1^L/V_2^L\right)\exp\left(-h_2^w/RT\right),$$

for

$$h_1^w = \lambda_{12} - \lambda_{11} \quad \text{and} \quad h_2^w = \lambda_{21} - \lambda_{11}.$$

If the assumption is accepted that $\lambda_{12} = \lambda_{21}$ then it is possible to calculate only the constants h_1^w and h_2^w, which was used in this work. For single isothermal or isobaric data with a small difference in temperatures it would be sufficient to calculate even the constants H_1^w and H_2^w only.

The local composition was also used by Renon[19,20] in his considerations about the interactions of components in solution. By comparing with the Scott theory[21], he obtained the dependence well-known in the literature as the NRTL equation

$$g^E/RT = \frac{x_1 x_2}{RT}\left[\frac{h_1^R H_1^R}{x_1 + x_2 H_1^R} + \frac{h_2^R H_2^R}{x_2 + x_1 H_2^R}\right], \qquad (5)$$

where

$$H_1^R = \exp\left(-\alpha_{12} h_1^R/RT\right) \quad \text{and} \quad H_2^R = \exp\left(-\alpha_{12} h_2^R/RT\right),$$

for

$$h_1^R = g_{21} - g_{11} \quad \text{and} \quad h_2^R = g_{12} - g_{22}.$$

For calculating the constants h_1^R and h_2^R, or H_2^R and H_1^R holds the same as for the case of the previous equation. By differentiating with respect to temperature, the corresponding relations for h^E/RT are obtained from Eqs (4) and (5).

Both relations contain an implicit dependence of excess functions on temperature and for that reason the constants h_i^w and h_i^R should be the real constants characteristic for a given system. When checking practically this assumption, the best results were not gained. Therefore, for a more accurate description of reality, it was recommended to introduce the temperature dependences of the constants h_i^w and $h_i^{R\,4,19,22}$ (in case of the NRTL equation, α_{12} as well[5]). This dependence is usually chosen empirically, *i.e.* either a linear one or in a form of expansions which, however, anyway must approach to the linear course. But the number of constants required is necessarily increased (with the Wilson relation to four and with NRTL to six) in this way, which results in a loss in physical interpretation. However, it is not impossible that these recommendations are rather premature and follow above all from the fact that both equations were not used to correlate both kinds of data (the equilibrium and the enthalpy ones) simultaneously in a more extensive set of data. Besides, it seems to be interesting to try to compare those two equations with the above-mentioned multiconstant equation suitability of which was proved formerly.

RESULTS

Survey of single sets of values which were used for simultaneous correlating every binary system is presented in Table I.

The correlation consisted in finding the minimum of a function Φ given by

$$\Phi = \left(\sum_{J=1}^{M}\sum_{I=1}^{N_J} W_I^J |E_I^J - F_I^J|\right)\Big/\sum_{J=1}^{M}\sum_{I=1}^{N_J} W_I^J. \qquad (6)$$

TABLE I

Survey of Data Sets

Component (2)	Symbol of system	Type of data	t, °C or P (Torr)	Symbol of set	Number of points	Ref.
Methanol	C 1	VLE[a]	55	g 55	13	1
		VLE	50	g 50	11	1,24
		HM[b]	50·5	h 50	12	25
Ethanol	C 2	VLE	0	g 0	12	26
		VLE	5	g 5	7	27
		VLE	10	g 10	19	26
		VLE	20	g 20A	19	26
		VLE	20	g 20B	7	27
		VLE	25	g 25	9	28
		VLE	30	g 30	19	26
		VLE	35	g 35	7	27
		VLE	50	g 50	7	27
		VLE	65	g 65	7	27
		VLE	(760)	g 760	20	29
		HM	20	h 20	5	31
		HM	25	h 25	16	30
1-Propanol	C 3	VLE	49·86	g 50	15	32
		VLE	55	g 55	17	1
		VLE	59·81	g 60	14	32
		VLE	65	g 65	14	1
		VLE	70	g 70	10	32
		VLE	(760)	g 760	18	33
		HM	25	h 25	20	30
1-Butanol	C 4	VLE	25	g 25	14	34
		VLE	35	g 35	13	34
		VLE	45	g 45	12	34
		VLE	50	g 50	15	2
		VLE	70	g 70	16	2
		VLE	80	h 80	9	35
		VLE	90	g 90	9	35
		VLE	100	g 100	9	35
		VLE	110	g 110	9	35
		VLE	(760)	g 760	20	36
		HM	25	h 25	20	30

TABLE 1
(continued)

Component (2)	Symbol of system	Type of data	t, °C or P (Torr)	Symbol of set		Number of points	Ref.
1-Hexanol	C 6	VLE	50	g	50	13	3
		VLE	60	g	60	14	3
		VLE	70	g	70	15	3
		VLE	81·2	g	80	18	3
		VLE	(70)	gP	70	17	3
		VLE	(100)	gP	100	17	3
		HM	25	h	25	18	30

[a] VLE Vapour–liquid equilibrium; [b] HM heats of mixing.

TABLE II

Values of Physico-Chemical Quantities of Pure Substances

Substance	Antoine equation			Equation for dependence of molar volume on temperature	
	A	B	C	OA $\left(\dfrac{ml}{mol}\right)$	OB $\left(\dfrac{ml}{mol\ grad}\right)$
Cyclohexane	6·85875	1 212·014	223·956[37]	64·54	0·1483[38]
Methanol	7·87863	1 473·11	230[39]	24·16	0·05734[38]
Ethanol	8·195854	1 641·263	230·479[40]	39·46	0·0646[38]
1-Propanol	7·8323	1 485·84	203[41]	50·06	0·084[38]
1-Butanol	7·56483	1 416·99	184·99[41]	66·66	0·085[38]
1-Hexanol	8·16353	1 903·29	205·046[42]	94	0·1049[38]

Substance	T_k, K	P_k, atm	$v_k \left(\dfrac{ml}{mol}\right)$	ω	μ, D	η
Cyclohexane	553·2	40	308[43]	0·186[43]	0	0
Methanol	513·2	78·5	118[43]	0·105	1·66	1·21[23]
Ethanol	516·3	63	167[43]	0·152	1·69	1[23]
1-Propanol	536·7	51	218·2[43]	0·201	1·68	0·57[23]
1-Butanol	563	43·6	274·6[43]	0·252	1·65	0·45[23]
1-Hexanol	610·2	34·1	388[44]	0·634[43]	·0	0

The values of excess functions were calculated in a form of dimensionless quantities g^E/RT or h^E/RT from experimental data. Molar excess free enthalpy was calculated with respect to the non-ideality of the vapour phase according to the relation

$$g^E/RT = x_1 \left[\ln \frac{y_1 P}{x_1 P_1^0} + \frac{(B_{11} - V_1^L)(P - P_1^0) + y_2^2 \delta_{12} P}{RT} \right] +$$
$$+ x_2 \left[\ln \frac{y_2 P}{x_2 P_2^0} + \frac{(B_{22} - V_2^L)(P - P_2^0) + y_1^2 \delta_{12} P}{RT} \right], \qquad (7)$$

where $\delta_{12} = 2B_{12} - B_{11} - B_{22}$. The virial coefficients were estimated by the O'Connell and Prausnitz method[23]. The temperature dependence of molar volumes of liquids was assumed linear, the constants were found from literary data. The dependence of saturated vapour pressures on temperature was calculated by means of the Antoine equation. All data of physico-chemical properties of pure substances used for calculating g^E/RT are listed in Table II.

For characterization of each point, a statistical weight was used which can be expressed in the form

$$W_I^J = \frac{MAX^{(g)} + MAX^{(h)}}{MAX^{(F)}} \cdot \frac{Q^{(F)}}{1 + (F_I^J/x_{11})^2}. \qquad (8)$$

This weight allows to minimize the distance perpendicular to the tangent of the dependence of function in a given point on composition of the liquid phase. The first

TABLE III
Values of Calculated Constants

System		C 1	C 2	C 3	C 4	C 6
Wilson equation	h_1^W	1 048·9	628·4	306·1	292·2	412·6
	h_2^W	1 252·7	1 541·2	1 358·7	1 194·9	1 185·4
NRTL equation	h_1^R	957·1	867·2	501·2	395·4	413·1
$\alpha_{12} = 0.47$	h_2^R	932·4	1 130·6	1 194·0	1 102·0	1 084·1
Multiconstant	α	0·0950	0·0285	0·5250	0·5956	0·5500
equation	β	0	0	0	0·0001	0
	A_0	4·3479	3·2205	2·7499	2·5948	2·6831
	B	−0·0061	−0·0035	−0·0031	−0·0031	−0·0032
	A_1	−0·1779	−0·2709	−0·4636	−0·4840	0·0074
	A_2	0·7694	0·4127	0·2814	0·2022	−0·7029

TABLE IV

Separate and Simultaneous Correlations in the Cyclohexane–n-Alcohol Systems (average weighted per cent deviations)

System	Data		Separate correlation by Wilson Eq.	Simultaneous correlation		
				Wilson Eq.	NRTL Eq. $\alpha_{12} = 0.47$	multiconstant Eq.
C 1	g	55	9·8	22·5	27·1	12·0
	g	50	15·2	27·9	36·3	28·2
	h	50	32·6	34·6	44·1	12·1
C 2	g	0	1·1	2·4	7·1	5·1
	g	5	1·0	3·9	6·6	1·8
	g	10	0·7	4·3	5·6	1·1
	g	20A	0·6	4·2	5·5	0·9
	g	20B	0·9	4·1	6·3	1·5
	g	25	5·0	9·3	9·4	6·5
	g	30	0·9	6·9	6·2	2·7
	g	35	1·9	4·2	6·2	1·6
	g	50	0·3	4·1	4·2	1·3
	g	65	0·5	4·8	2·7	3·6
	g	760	2·0	7·4	5·8	3·3
	h	25	3·6	3·6	7·9	10·2
	h	20	2·7	3·9	2·7	6·2
C 3	g	50	4·3	10·6	8·3	6·0
	g	55	6·8	8·8	8·1	8·3
	g	60	3·8	10·5	7·3	4·8
	g	65	4·1	9·4	6·2	5·1
	g	70	6·9	8·5	7·0	6·9
	g	760	2·7	4·7	3·1	3·7
	h	25	5·1	5·5	5·3	6·9
C 4	g	25	2·1	20·4	20·1	20·3
	g	35	7·3	18·4	17·2	14·8
	g	45	25·1	37·9	35·6	31·2
	g	50	8·6	16·3	16·2	17·0
	g	70	5·4	9·9	9·9	10·5
	g	80	16·3	19·5	18·2	18·1
	g	90	4·1	14·3	13·9	13·1
	g	100	3·1	16·2	15·7	14·4
	g	110	4·5	25·3	24·6	22·9
	g	760	7·4	18·9	16·9	13·8
	h	25	4·1	7·9	4·5	4·8

TABLE IV
(Continued)

| System | Data | | Separate correlation by Wilson Eq. | Simultaneous correlation | | |
				Wilson Eq.	NRTL Eq. $\alpha_{12} = 0.47$	multiconstant Eq.
C 6	g	50	19·3	31·1	32·2	20·1
	g	60	25·1	47·7	48·6	22·8
	g	70	58·3	99·5	101·3	57·5
	g	80	10·8	24·0	25·6	15·2
	gP	70	62·6	67·1	67·2	48·0
	gP	100	56·2	57·4	57·6	37·8
	h	25	3·5	10·7	6·2	4·5

term of the product in Eq. (8) has the aim to bring the data to the same level as for the maximum absolute values and the value $Q^{(F)}$ as for the mutual number of data for g^E/RT and h^E/RT.

The results of calculations are presented in Tables III–V. In Table III the constants of single equations are summarized which were calculated by simultaneous correlation of all sets of each system. In Table IV the accuracy of calculation in single sets is expressed. To judge the suitability of single equation relations in detail the values of minimized functions were calculated with all sets for each single point together with the values of the derived dependences ($\log \gamma_i$, g^E, g^M, $\partial g^M/\partial x_1$) and compared with the values obtained from the experimental data. This manner is unsuitable for presentation owing to its extent. Therefore the average weighted percentual deviation of minimized function was used for comparison which is sufficiently sensitive to indicate the mutual differences of used equations in single sets. To judge the application of single equations to practical purposes, above all for the construction of phase diagrams at chosen conditions requiring the interpolation of data, the results of back calculation of composition of the gas phase are summarized in Table V, again as the average deviations of the measured and in this way calculated data.

DISCUSSION

The results indicate that all used equations describe with a fairly good agreement the phase behaviour of the systems studied. The lowest deviations were reached with the system C 2; with increasing number of carbons of the polar component the deviations gradually increase as far as the system C 6, which is already according to its character

TABLE V

Back Calculations of Composition of the Vapour Phase (average deviations)

System	Data		Separate correlation by Wilson Eq.	Simultaneous correlation		
				Wilson Eq.	NRTL Eq.	multiconstant Eq.
C 1	g	55	0·032	0·099	0·099	0·031
	g	50	0·017	0·065	0·058	0·027
C 2	g	0	0·003	0·021	0·013	0·007
	g	5	0·001	0·015	0·014	0·010
	g	10	0·003	0·016	0·014	0·008
	g	20A	0·004	0·020	0·017	0·007
	g	20B	0·001	0·019	0·016	0·009
	g	25	0·012	0·024	0·024	0·015
	g	30	0·012	0·030	0·026	0·015
	g	35	0·010	0·022	0·017	0·008
	g	50	0·001	0·023	0·017	0·007
	g	65	0·001	0·024	0·016	0·007
	g	760	0·012	0·033	0·023	0·014
C 3	g	50	0·009	0·016	0·011	0·008
	g	55	0·010	0·019	0·014	0·011
	g	60	0·011	0·018	0·011	0·009
	g	65	0·004	0·012	0·011	0·012
	g	70	0·016	0·020	0·016	0·015
	g	760	0·007	0·009	0·008	0·017
C 4	g	25	0·013	0·010	0·010	0·011
	g	35	0·011	0·011	0·011	0·010
	g	45	0·004	0·011	0·010	0·011
	g	50	0·006	0·012	0·010	0·006
	g	70	0·005	0·011	0·008	0·006
	g	80	0·023	0·020	0·020	0·021
	g	90	0·019	0·016	0·015	0·013
	g	100	0·021	0·15	0·014	0·013
	g	110	0·024	0·018	0·018	0·022
	g	760	0·005	0·014	0·010	0·011
C 6	g	50	0·002	0·002	0·002	0·003
	g	60	0·005	0·003	0·003	0·006
	g	70	0·003	0·010	0·010	0·012
	g	80	0·005	0·004	0·004	0·010
	gP	70	0·007	0·008	0·009	0·027
	gP	100	0·007	0·011	0·012	0·024

closer to the liquid–gas equilibrium, where the differences between the measured and calculated values are higher. A worse agreement was also observed with the sets of the system C1, which is caused, in all probability, by a close neighbourhood of the heterogeneous region. This circumstance enhances considerably the requirements on analytic expression of dependences of excess quantities which show extreme courses.

The mutual comparison of results attained by correlating by means of single equations is rather made difficult because of different number of constants. As it was possible to expect the equation with six constants provided the best results. The disadvantage of the considerable number of constants with sets studied in this work is compensated to a certain extent by finding that the constant β can be put equal zero and the constant B generalized by only one value for all systems. This circumstance giving evidence for a similar course of the temperature dependence of g^E/RT and h^E/RT for all systems, led, above all in the relation for expressing
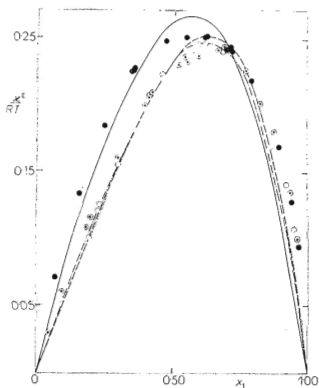

FIG. 1

Comparison of the Experimental Dependence of Excess Enthalpy of Systems C2, C3, and C4 at 25°C with the Data Calculated from Eq. (9)

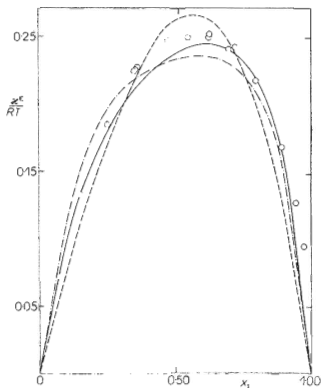Measured points C2 ○, C3 ●, C4 ⊙; calculated dependences C2 solid line, C3 dot-and-dash line, C4 dash line.

FIG. 2

Comparison of the Experimentally Found and Calculated Dependence of Excess Enthalpy of the Cyclohexane–Ethanol System at 25°C on Composition

Measured points ○; calculated dependences by Wilson equation solid line, NRTL equation dot-and-dash line, and Eq. (9) dash line.

excess enthalpy, to substantial simplification. Eq. (3) changes by this rearranging to a form with two constants

$$h^E/RT = -TB \exp \left[\alpha(x_1 - x_2) \right] x_1 x_2 \qquad (9)$$

and is consequently well comparable, as for the number of constants and the accuracy reached, with both remaining relations. The situation is apparent well from the following figures. In Fig. 1 the experimental data of the mixing enthalpy for systems C2, C3, and C4 at 25°C are plotted together with the data calculated from Eq. (9). In Fig. 2 the experimental data of h^E/RT for the system C2 at 25°C are compared with the data calculated from the equations considered here. The results indicate that the agreement is approximately the same, which warrants to conclude that all three equations are mutually well comparable and provide a sufficiently accurate characterization of vapour–liquid equilibrium in the region bounded by experimental values.

LIST OF SYMBOLS

$A_0, A_1, A_2, B, \alpha, \beta$ constants of Eq. (1)
B_{ii} virial coefficient of pure component i
g molar free enthalpy
g_{ij} molar free enthalpy of interactions $i - j$
h molar enthalpy
h_1, h_2, H_1, H_2 constants
I, J summation indices
P pressure
R gas constant
T temperature (K)
V molar volume
x, y mole fractions

α_{12} constant of NRTL equation
λ_{ij} interaction energy
M number of sets of given system
N number of points in single sets of data
F calculated value of function g^E/RT or h^E/RT
E experimental value corresponding to F
w statistical weight corresponding to F
Q factor of weight w
MAX maximum value of thermodynamic quantity of given system

Superscripts

E denotes excess quantity
L liquid phase
R refers to NRTL equation
w refers to Wilson's equation

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