Table IV (Continued)

E. For Hydroxides

M	deviation (calcd − obsd), kcal/mol from $\Delta H^\circ_{(CO_3)}$, $\Delta H^\circ_{(O)}$ (eq $1_{m,h}$, $1_{b,h}$)	from $\Delta H^\circ_{(SO_4)}$, $\Delta H^\circ_{(O)}$ (eq $2_{m,h}$, $2_{b,h}$)	from $\Delta H^\circ_{(O)}$, $\Delta H^\circ_{(NO_3)}$ (eq $4_{m,h}$, $4_{b,h}$)	M	deviation (calcd − obsd), kcal/mol from $\Delta H^\circ_{(CO_3)}$, $\Delta H^\circ_{(O)}$ (eq $1_{m,h}$, $1_{b,h}$)	from $\Delta H^\circ_{(SO_4)}$, $\Delta H^\circ_{(O)}$ (eq $2_{m,h}$, $2_{b,h}$)	from $\Delta H^\circ_{(O)}$, $\Delta H^\circ_{(NO_3)}$ (eq $4_{m,h}$, $4_{b,h}$)
Li	−1.3	0.3	1.3	Ba	1.2	1.3	1.3
Na	−0.9	0.7	0.8	Ra			
K	−0.4	1.1	1.1	Pb			
Rb	0.2	1.0	1.0	Cd	−2.4	−1.6	−2.1
Cs	−0.6	0	−0.8	Mn	−2.2	−0.1	0.6
Tl	−2.7	−3.7	−4.2	Fe	−1.2	−1.5	
Ag				Co	−0.7	0.1	−1.1
Be	3.3	2.1		Ni		−1.4	−3.4
Mg	2.6	2.9	3.8	Cu		0.6	−0.6
Ca	1.0	1.1	1.7	Zn	−1.6	−0.6	0.5
Sr	−1.2	−0.4	−0.8				

Table V. Estimated $\Delta H_f{}^\circ{}_{298}$ Values (kcal/mol)

compd	ref compds and (eq used)	$\Delta H_f{}^\circ{}_{298}$, kcal/mol estimated value	av value
AgOH	$\Delta H^\circ_{(CO_3)}$ and $\Delta H^\circ_{(O)}$ ($1_{m,h}$)	−38.6	−38.8
	$\Delta H^\circ_{(SO_4)}$ and $\Delta H^\circ_{(O)}$ ($2_{m,h}$)	−39.3	
	$\Delta H^\circ_{(O)}$ and $\Delta H^\circ_{(NO_3)}$ ($4_{m,h}$)	−38.6	
$Be(NO_3)_2$	$\Delta H^\circ_{(SO_4)}$ and $\Delta H^\circ_{(O)}$ ($2_{b,n}$)	−172.0	−172.0
	$\Delta H^\circ_{(SO_4)}$ and $\Delta H^\circ_{(OH)}$ ($4_{b,n}$)	−171.9	
	$\Delta H^\circ_{(SO_4)}$ and $\Delta H^\circ_{(CO_3)}$ ($6_{b,n}$)	−172.0	
$RaCO_3$	$\Delta H^\circ_{(SO_4)}$ and $\Delta H^\circ_{(O)}$ ($2_{b,c}$)	−289.2	−288.9
	$\Delta H^\circ_{(NO_3)}$ and $\Delta H^\circ_{(O)}$ ($4_{b,c}$)	−288.6	
$Ra(OH)_2$	$\Delta H^\circ_{(SO_4)}$ and $\Delta H^\circ_{(O)}$ ($2_{b,h}$)	−219.6	−219.6
	$\Delta H^\circ_{(O)}$ and $\Delta H^\circ_{(NO_3)}$ ($4_{b,h}$)	−219.5	
$Fe(NO_3)_2$	$\Delta H^\circ_{(SO_4)}$ and $\Delta H^\circ_{(O)}$ ($2_{b,n}$)	−106.2	−106.2
	$\Delta H^\circ_{(SO_4)}$ and $\Delta H^\circ_{(OH)}$ ($4_{b,n}$)	−106.3	
	$\Delta H^\circ_{(SO_4)}$ and $\Delta H^\circ_{(CO_3)}$ ($6_{b,n}$)	−106.1	
$NiCO_3$	$\Delta H^\circ_{(SO_4)}$ and $\Delta H^\circ_{(O)}$ ($2_{b,c}$)	−166.1	−167.8
	$\Delta H^\circ_{(OH)}$ and $\Delta H^\circ_{(SO_4)}$ ($3_{b,c}$)	−165.5	
	$\Delta H^\circ_{(NO_3)}$ and $\Delta H^\circ_{(O)}$ ($4_{b,c}$)	−170.6	
	$\Delta H^\circ_{(OH)}$ and $\Delta H^\circ_{(NO_3)}$ ($5_{b,c}$)	−169.1	
$CuCO_3$	$\Delta H^\circ_{(SO_4)}$ and $\Delta H^\circ_{(O)}$ ($2_{b,c}$)	−143.1	−144.4
	$\Delta H^\circ_{(OH)}$ and $\Delta H^\circ_{(SO_4)}$ ($3_{b,c}$)	−143.3	
	$\Delta H^\circ_{(NO_3)}$ and $\Delta H^\circ_{(O)}$ ($4_{b,c}$)	−145.6	
	$\Delta H^\circ_{(OH)}$ and $\Delta H^\circ_{(NO_3)}$ ($5_{b,c}$)	−145.5	
$Pb(OH)_2$	$\Delta H^\circ_{(CO_3)}$ and $\Delta H^\circ_{(O)}$ ($1_{b,h}$)	−136.4	−136.0
	$\Delta H^\circ_{(SO_4)}$ and $\Delta H^\circ_{(O)}$ ($2_{b,h}$)	−135.6	
	$\Delta H^\circ_{(O)}$ and $\Delta H^\circ_{(NO_3)}$ ($4_{b,h}$)	−136.1	

Recently Tardy et al. (*6*–*9*) and Sverjensky (*4*, *5*) have also shown the existence of empirical relations among Gibbs free energies and enthalpies of formation of solid compounds such as silicates, carbonates, and sulfates, etc. However, they related $\Delta H_f{}^\circ$ (salts) to $\Delta H_f{}^\circ$ for the corresponding aqueous cations. Moreover, the relationships given are based on limited groups of compounds and the compounds are classified according to structural types. With the relationships illustrated here, we are able to correlate any three types of compounds only on the basis of valence states. We have also found that the relationship described above are not confined to the oxygen-containing compounds. The halides also show similar relations, and these will be described later.

Literature Cited

(1) Hisham, M. W. M.; Benson, S. W. *J. Phys. Chem.* **1985**, *89*, 1905.
(2) Hisham, M. W. M.; Benson, S. W. *J. Phys. Chem.* **1985**, *89*, 3417.
(3) Hisham, M. W. M.; Benson, S. W. *J. Phys. Chem.* **1986**, *90*, 885.
(4) Sverjensky, D. A. *Geochim. Cosmochim. Acta* **1984**, *48*, 1127.
(5) Sverjensky, D. A. *Geochim. Cosmochim. Acta* **1985**, *49*, 853.
(6) Tardy, Y.; Garrels, R. M. *Geochim. Cosmochim. Acta* **1976**, *41*, 1051.
(7) Tardy, Y.; Garrels, R. M. *Geochim. Cosmochim. Acta* **1977**, *41*, 87.
(8) Tardy, Y.; Viellard, P. *Contrib. Mineral Petrol.* **1977**, *63*, 75.
(9) Tardy, Y.; Gartner, L. *Contrib. Mineral Petrol.* **1977**, *63*, 89.
(10) Wagman, D. D.; Evans, W. H.; Parker, V. B.; Schumm, R. H.; Halow, I.; Bailey, S. M.; Churney, K. L.; Nuttall, R. L. *J. Phys. Chem. Ref. Data*, **1982**, Suppl. 11.

Received for review June 17, 1986. Accepted November 3, 1986. We are grateful to the National Science Foundation (Grant CHE-78-26623) and to the Army Research Office (Grant DAAG-29-85-K-0019) for their support.

Vapor–Liquid Equilibrium of the System Ethanol + Benzene + Cyclohexane at 760 mmHg

Alberto Arce,* Antonio Blanco, and Jose Tojo

Chemical Engineering Department, University of Santiago de Compostela, Santiago de Compostela, Spain

Vapor–liquid equilibrium data for the ternary mixture ethanol + benzene + cyclohexane at a constant pressure of 760 mmHg have been determined experimentally and predicted by using the group contribution methods UNIFAC and ASOG-KT and the NRTL, UNIQUAC, and Wilson equations with parameters estimated from data for the corresponding binary mixtures. The predictions compare satisfactorily with the experimental results.

Introduction

Apart from their intrinsic value for the design of distilling plants, experimental VLE data provide an important means of testing the validity of thermodynamic models of liquid mixtures and the associated methods of data processing and prediction. For binary mixtures experimental VLE data are generally available and reliable, but the same is not true for most multicomponent systems. This article reports experimental VLE

0021-9568/87/1732-0247$01.50/0

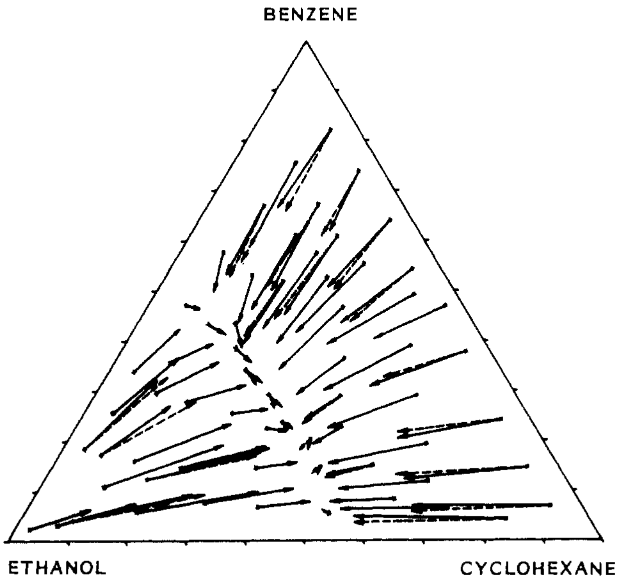

Figure 1. VLE data for ethanol + benzene + cyclohexane at 760 mmHg: — experimental; --- predicted by ASOG-KT.

data for the ternary mixture ethanol + benzene + cyclohexane at a constant pressure of 760 mmHg, and compares the results with those predicted by the group contribution methods ASOG-KT (*1*) and UNIFAC (*2*) and by the NRTL (*3*), Wilson (*4*), and UNIQUAC (*5*, *6*) equations (the latter as modified for alcohols), the NRTL, Wilson, and UNIQUAC parameters being estimated from data for the binary mixtures. In all cases the deviations of both phases from ideal behavior were taken into account by employing the equilibrium criterion

$$y_i \phi_i P = x_i \gamma_i f_i^0 \tag{1}$$

where the coefficients of fugacity ϕ_i were obtained by using second virial coefficients B_{ij} calculated by the Hayden-O'Connell method (*7*).

Experimental Methods and Results

Experimental vapor-liquid equilibrium data for the mixture ethanol + benzene + cyclohexane at 760 mmHg were obtained by using an Othmer ebulliometer as modified by Ocón and Espantoso (*8*, *9*). All chemical products used were Merck chromatographic grade and were used as supplied without further purification. The densities at 25 °C, refractive indices at 25 °C, and boiling points at 760 mmHg of the pure components of the mixtures were measured and agreed with published values. All experiments were carried out under an atmosphere of argon so as to prevent ethanol taking up water from the environment. Samples of both the liquid and vapor phases were analyzed by measuring their refractive indices and densities at 25 °C and interpolating in previously determined diagrams of refractive index and density as functions of composition (*10*).

The experimental VLE data for the ternary mixture are shown in Table I and are shown as unbroken arrows in Figure 1, where the flight end of each arrow marks the composition of the liquid phase and the arrowhead that of the vapor phase in equilibrium. Figure 2, in which equilibrium isotherms are plotted on the liquid-phase composition diagram, shows a ternary minimum azeotrope of boiling point 64.8 °C at a mole fraction composition of ethanol:benzene:cyclohexane = 0.42:0.07:0.51. The points marked A, B, and C in Figure 2 show the azeotropes of the three binary mixtures.

Table I. Experimental VLE Temperatures and Compositions for the Mixture Ethanol + Benzene + Cyclohexane at 760 mmHg

		liq phase mole fracn		vap phase mole fracn	
run	t, °C	ethanol	benzene	ethanol	benzene
1	73.81	0.953	0.023	0.837	0.064
2	70.63	0.901	0.035	0.749	0.072
3	68.79	0.858	0.047	0.638	0.094
4	66.31	0.749	0.061	0.525	0.100
5	65.24	0.628	0.081	0.465	0.104
6	64.92	0.544	0.070	0.448	0.081
7	64.81	0.429	0.057	0.438	0.061
8	64.87	0.306	0.085	0.416	0.082
9	65.04	0.228	0.070	0.412	0.065
10	65.80	0.141	0.045	0.393	0.051
11	68.98	0.055	0.069	0.290	0.069
12	69.28	0.053	0.148	0.272	0.127
13	70.26	0.043	0.245	0.242	0.207
14	70.41	0.040	0.379	0.241	0.312
15	71.88	0.033	0.470	0.188	0.401
16	70.54	0.045	0.540	0.229	0.434
17	71.02	0.045	0.635	0.215	0.504
18	73.37	0.036	0.742	0.153	0.616
19	72.45	0.049	0.818	0.219	0.663
20	69.41	0.137	0.758	0.302	0.588
21	67.85	0.234	0.671	0.371	0.532
22	67.16	0.338	0.581	0.409	0.498
23	67.06	0.465	0.475	0.445	0.467
24	67.41	0.614	0.345	0.495	0.427
25	67.71	0.693	0.259	0.527	0.370
26	68.74	0.776	0.190	0.586	0.325
27	67.52	0.748	0.181	0.589	0.273
28	66.19	0.701	0.166	0.507	0.235
29	65.35	0.627	0.150	0.467	0.191
30	64.95	0.507	0.149	0.432	0.157
31	64.88	0.407	0.144	0.413	0.136
32	64.89	0.309	0.153	0.400	0.134
33	65.09	0.230	0.119	0.396	0.104
34	65.36	0.200	0.195	0.389	0.154
35	65.81	0.166	0.291	0.355	0.225
36	66.95	0.129	0.389	0.324	0.301
37	68.87	0.070	0.491	0.273	0.389
38	67.88	0.123	0.555	0.310	0.415
39	67.61	0.142	0.611	0.314	0.452
40	68.33	0.145	0.670	0.311	0.501
41	67.10	0.216	0.609	0.363	0.463
42	66.44	0.328	0.527	0.395	0.438
43	66.35	0.445	0.437	0.423	0.417
44	66.44	0.546	0.361	0.457	0.395
45	66.37	0.595	0.302	0.468	0.359
46	65.78	0.556	0.282	0.445	0.312
47	65.31	0.504	0.257	0.428	0.264
48	65.03	0.450	0.229	0.418	0.222
49	64.91	0.390	0.204	0.407	0.187
50	65.02	0.324	0.228	0.393	0.196
51	65.14	0.299	0.288	0.387	0.242
52	65.39	0.252	0.364	0.377	0.289
53	65.93	0.207	0.465	0.372	0.346
54	66.30	0.201	0.526	0.354	0.394
55	66.83	0.195	0.577	0.360	0.426
56	66.39	0.283	0.515	0.412	0.393
57	66.16	0.397	0.438	0.413	0.392
58	65.80	0.420	0.385	0.414	0.353
59	65.58	0.427	0.345	0.408	0.321
60	65.27	0.416	0.293	0.403	0.272
61	65.05	0.414	0.240	0.411	0.225
62	65.82	0.700	0.127	0.482	0.181
63	67.45	0.778	0.112	0.539	0.192

Analysis of the Data for the Binary Mixtures

The VLE data for the binary mixtures benzene + cyclohexane (*11*), ethanol + cyclohexane (*12*), and ethanol + benzene (*13*) were subjected to the thermodynamical consistency test proposed by Fredenslund et al. (*14*) and the data passing the test were used to estimate the parameters of the UNIQUAC, NRTL, and Wilson equations by the method of

Table II. Optimized Parameters of the UNIQUAC, NRTL, and Wilson Equations, with the Values of the Objective Function *S*

	A_{12}	A_{21}	α	S
	Benzene + Cyclohexane			
UNIQUAC	27.71	74.12		13.49
NRTL	353.02	-87.61	0.3	13.43
Wilson	130.05	132.93		13.71
	Ethanol + Cyclohexane			
UNIQUAC	-222.98	2582.19		96.69
NRTL	888.26	1431.97	0.47	34.74
Wilson	2062.21	378.63		70.64
	Ethanol + Benzene			
UNIQUAC	-256.50	1716.77		150.00
NRTL	529.69	971.14	0.47	116.41
Wilson	1325.45	218.98		158.83

Prausnitz et al. (*15*), a maximum likelihood nonlinear regression procedure in which the optimal parameter values are taken to be those that minimize the objective function

$$S = \sum^{m} \left\{ \frac{(P_i^{c} - P_i^{e})^2}{\sigma_{P_i}^{2}} + \frac{(T_i^{c} - T_i^{e})^2}{\sigma_{T_i}^{2}} + \frac{(x_{1i}^{c} - x_{1i}^{e})^2}{\sigma_{x_{1i}}^{2}} + \frac{(y_{1i}^{c} - y_{1i}^{e})^2}{\sigma_{y_{1i}}^{2}} \right\} \qquad (2)$$

where the superscripts c and e indicate calculated and experimental values, respectively, σ^2 is the estimated variance of each of the variables measured, and the sum is taken over the m experimental determinations. The σ values assumed in the present work were as follows: $\sigma_P = 0.5$ mmHg; $\sigma_T = 0.1$ °C; $\sigma_x = 0.001$ mole fraction; $\sigma_y = 0.005$ mole fraction. The values of the parameters r, q, and q' required by the UNIQUAC equation were taken from the literature (*15*). The liquid molar volumes needed for the correlation with the Wilson equation were calculated by using the modified Rackett equation (*16*). The results of these optimizations are listed in Table II, where the A_{ij} for the three theoretical methods are defined as follows:

UNIQUAC $\quad A_{ij} = u_{ij} - u_{jj}$ (cal/(g mol))

NRTL $\quad A_{ij} = g_{ij} - g_{jj}$ (cal/(g mol))

Wilson $\quad A_{ij} = \lambda_{ij} - \lambda_{ii}$ (cal/(g mol))

Prediction of the VLE of the Mixture Ethanol + Benzene + Cyclohexane

The activity coefficients of the components of the ternary mixture was calculated by using the group contribution methods ASOG-KT (*1*) and UNIFAC (*2*, *17*) and the UNIQUAC (*5*, *6*), NRTL (*3*), and Wilson (*4*) equations for three-component mixtures (for the values of whose parameters those determined for the two-component mixtures were employed). Once the activity coefficients have been obtained, the equilibrium temperatures and compositions of the ternary mixture at 760 mmHg were calculated taking into account the nonideal nature of both phases. Table III lists the root mean square deviations of the predictions from the experimental values determined by ourselves and from those published by Morachevskii and Zharov (*18*) and Deshpande and Lu (*19*). In all cases the ASOG-KT method proved to yield the best predictions of equilibrium compositions (shown in Figure 1 by broken arrows) and the worst predictions of temperatures, though none of the theoretical methods can be considered to have produced more than small deviations. Table IV lists the root mean square deviations between the experimental data for the three binary mixtures and the predictions of the ASOG-KT and UNIFAC methods for these systems.

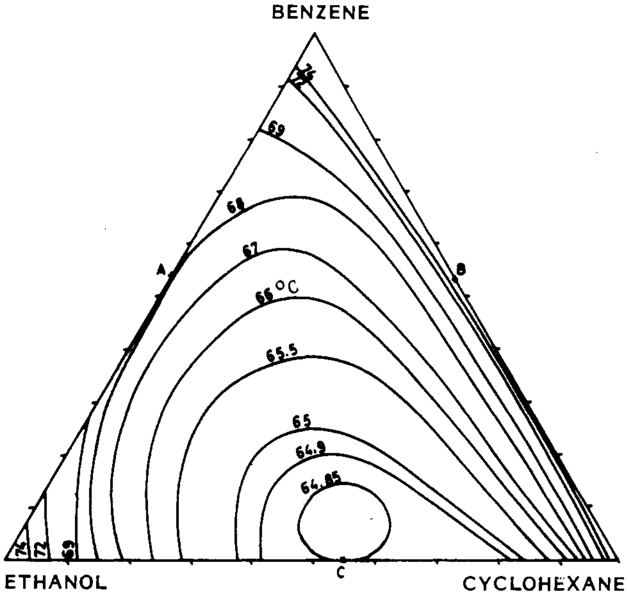

Figure 2. Isotherms for ethanol + benzene + cyclohexane at 760 mmHg.

Table V lists the ternary azeotrope boiling points and compositions determined experimentally by ourselves and by others and calculated by theoretical methods. The empirical values differ appreciably, which is readily explained by the experimental difficulties, but the theoretical predictions agree quite well with each other, only the UNIFAC results being slightly discrepant from the rest.

Conclusions

The VLE characteristics predicted by the UNIQUAC, NRTL, and Wilson models and by the UNIFAC and ASOG-KT methods for the mixture ethanol + benzene + cyclohexane all compare satisfactorily with the experimental data obtained by ourselves and reported in this article, particularly good agreement being achieved by the ASOG-KT method (Table III). The deviations of the predictions from the experimental values reported by Morachevskii and Zharov (*18*) and Deshpande and Lu (*19*) are

Table III. Root Mean Square Differences between Experimental VLE Temperatures and Compositions of the Mixture Ethanol + Benzene + Cyclohexane and Those Predicted by Various Methods

	this work[a]				Morachevskii and Zharov[b] (*18*)				Deshpande and Lu[c] (*19*)			
		RMS y, mole fracn				RMS y, mole fracn				RMS y, mole fracn		
method	RMS t, °C	ethanol	benzene	cyclohexane	RMS t, °C	ethanol	benzene	cyclohexane	RMS t, °C	ethanol	benzene	cyclohexane
UNIFAC	0.37	0.0220	0.0122	0.0145	0.43	0.0192	0.0092	0.0179	0.79	0.0274	0.0206	0.0174
ASOG	0.62	0.0153	0.0064	0.0124	0.57	0.0163	0.0040	0.0169	1.23	0.0240	0.0193	0.0162
UNIQUAC	0.37	0.0200	0.0094	0.0146	0.33	0.0174	0.0071	0.0179	0.90	0.0253	0.0196	0.0163
NRTL (α = 0.47)	0.30	0.0204	0.0095	0.0145	0.33	0.0189	0.0081	0.0179	0.96	0.0265	0.0204	0.0169
Wilson	0.31	0.0205	0.0100	0.0145	0.32	0.0179	0.0081	0.0175	0.88	0.0258	0.0200	0.0165

[a] 63 experimental data. [b] 19 experimental data. [c] 57 experimental data.

Table IV. Root Mean Square Differences between Experimental VLE Temperatures and Compositions for the Binary Mixtures and Those Predicted by the ASOG-KT and UNIFAC Methods

	ASOG method		UNIFAC method	
	RMS t, °C	RMS y, mole fracn	RMS t, °C	RMS y, mole fracn
benzene + cyclohexane	0.16	0.002	0.08	0.003
ethanol + cyclohexane	0.73	0.009	0.10	0.013
ethanol + benzene	0.60	0.017	0.18	0.006

Table V. Boiling Point and Composition of the Ternary Azeotrope of the Mixture Ethanol + Benzene + Cyclohexane at 760 mmHg

		mole fraction		
method	t, °C	ethanol	benzene	cyclohexane
exptl (this work)	64.8	0.42	0.07	0.51
exptl (Zieborak et al. (*20*))	65.05	0.441	0.092	0.467
exptl (Morachevskii et al. (*18*))	64.7	0.43	0.11	0.46
exptl (Deshpande and Lu (*19*))	65.1	0.424	0.158	0.46
UNIFAC	64.81	0.433	0.033	0.534
ASOG-KT	65.48	0.442	0.074	0.483
UNIQUAC	65.15	0.442	0.050	0.508
NRTL	65.15	0.438	0.073	0.489
Wilson	65.10	0.438	0.076	0.486

generally somewhat greater (Table III). It may be pointed out that in our own experimental work 63 determinations spanning the whole space of ternary compositions were carried out (Figure 1), as compared with the 19 experimental points of Morachevskii and Zharov and the 57 of Deshpande and Lu.

At 760 mmHg, the ternary mixture ethanol + benzene + cyclohexane was found experimentally to have a minimum azeotrope of boiling point 64.8 °C and mole fraction composition ethanol:benzene:cyclohexane = 0.42:0.07:0.51. These proportions are very close to those predicted by the UNIFAC method.

Glossary

A_{ij}	optimized UNIQUAC, NRTL, and Wilson parameters
B	second virial coefficient
f^0	reference fugacity
g_{ij}, g_{jj}	NRTL interaction parameters, cal/(g mol)
m	number of data points
P	total pressure of the system
q	UNIQUAC areal structure parameter
q'	UNIQUAC areal structure parameter for water and alcohols
RMS	root mean square difference
r	UNIQUAC volume structure parameter
S	objective function
t	temperature, °C
u_{ij}, u_{jj}	UNIQUAC interaction parameters, cal/(g mol)
x	mole fraction in the liquid phase
y	mole fraction in the vapor phase

Greek Letters

α_{ij}	third NRTL parameter
γ	activity coefficient
λ_{ij}, λ_{ii}	Wilson interaction parameters, cal/(g mol)
$\sum$	summation
σ^2	estimated variance of each of the variables measured
ϕ	coefficient of fugacity

Subscripts

i	component i (eq 1)
i	i-th experimental point (eq 2)
ij	mixture of components i and j

Superscripts

c	calculated
e	experimental

Registry No. Ethanol, 64-17-5; benzene, 71-43-2; cyclohexane, 110-82-7.

Literature Cited

(1) Kojima, K.; Tochigi, K. *Prediction of Vapor-Liquid Equilibria by the ASOG Method*; Elsevier: Tokyo, 1979.
(2) Fredenslund, Aa.; Jones, R. L.; Prausnitz, J. M. *AIChE J.* **1975**, *21*, 1086.
(3) Renon, H.; Prausnitz, J. M. *AIChE J.* **1968**, *14*, 135.
(4) Wilson, G. M. *J. Am. Chem. Soc.* **1964**, *86*, 127.
(5) Abrams, D. S.; Prausnitz, J. M. *AIChE J.* **1975**, *21*, 116.
(6) Anderson, T. F.; Prausnitz, J. M. *Ind. Eng. Chem. Process Des. Dev.* **1978**, *17*, 552.
(7) Hayden, J. G.; O'Connell, J. P. *Ind. Eng. Chem. Process Des. Dev.* **1975**, *14*, 209.
(8) Ocôn, J.; Espantoso, J. *Quim. Ind.* **1956**, *3*, 109.
(9) Ocôn, J.; Espantoso, J. *An. Quim.* **1958**, *54B*, 413.
(10) Arce, A.; Blanco, A.; Antorrena, G.; Quintela, M. D. *An. Quim.* **1980**, *76*, 405.
(11) Nagata, I. *J. Chem. Eng. Data* **1962**, *7*, 461.
(12) Arce, A.; Blanco, A.; Mendez, M. R.; Tojo, J. *An. Quim.*, in press.
(13) Niini, A. *Ann. Acad. Sci. Fennicae* **1940**, *A-55*, No. 8.
(14) Fredenslund, Aa.; Gmehling, J.; Rasmussen, P. *Vapor-liquid Equilibria using UNIFAC*; Elsevier: Amsterdam, 1977.
(15) Prausnitz, J. M.; Anderson, T. F.; Grens, E.; Eckert, C.; Hsieh, R.; O'Connell, J. *Computer Calculations for Multicomponent Vapor-liquid and Liquid-liquid Equilibria*; Prentice-Hall: New Jersey, 1980.
(16) Spencer, C. F.; Danner, R. P. *J. Chem. Eng. Data* **1972**, *17*, 236.
(17) Gmehling, J.; Rasmussen, P.; Fredenslund, Aa. *Ind. Eng. Chem. Process Des. Dev.* **1982**, *21*, 118.
(18) Morachevskii, A. G.; Zharov, V. T. *Zh. Prikl. Khim.* **1963**, *36*, 2771.
(19) Deshpande, A. K.; Lu, B. C.-Y. *J. Appl. Chem.* **1965**, *15*, 136.
(20) Zieborak, K.; Golska-Krajewska, A. *Bull. Acad. Polon. Sci. Chim. Geôl. Géogr.* **1985**, *6*, 763.

Received for review May 5, 1986. Accepted December 5, 1986.