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DISQUAC predictions on thermodynamic properties of ternary and higher multicomponent mixtures.

I. Results for total pressure measurements at isothermal conditions of ternary systems

Juan Antonio González*, Javier Carmona, Isaías García De La Fuente, José Carlos Cobos

Dpto. Termodinamica y Fisica Aplicada, Facultad De Ciencias, Universidad De Valladolid, 47071 Valladolid, Spain

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Abstract

The ability of DISQUAC to represent vapor-liquid equilibria, VLE, particularly pressures for sets of (x_1, x_2) values at a given temperature, of ternary mixtures is investigated. Results are obtained using binary interaction parameters only, that is, ternary interactions are neglected.

At this end, a set of 39 ternary systems and of 98 related binaries is analyzed. The ternary mixtures treated are of widely different classes. They may be formed by only hydrocarbons; by one or two polar, but not self-associated, compounds and hydrocarbons; or by one or two alcohols and organic solvents, such as hydrocarbons, CCl_4 or polar components.

Deviations for ternary and binary systems are similar. The mean relative standard deviation in pressure is 0.020 for the former, and 0.017 for the latter. This means that good predictions on VLE of ternary mixtures can be obtained, taking into account binary interactions only. Moreover, DISQUAC results are practically independent of the system considered, and are valid over a wide range of temperature. Poorer predictions are obtained for those solutions with a binary showing a miscibility gap.

It is also noteworthy that, in most of the cases, the interaction parameters needed are already available in the literature. Only for a few binary systems considered, the VLE data are correlated to obtain interchange coefficients which are later used to predict VLE of the related ternary.

For the binary mixtures, DISQUAC predicts correctly: the coordinates of azeotropes and excess Gibbs energies, G^E.

A comparison between DISQUAC results and those obtained using different association models is also presented. DISQUAC and the UNIQUAC association theory yield similar predictions for ternary systems containing two alcohols and one *n*-alkane. © 1999 Elsevier Science B.V. All rights reserved.

Keywords: Binary interactions; Group contributions; Isothermal; Ternary systems; Vapor-liquid equilibria

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*Corresponding author. Fax: +34-83-423-136; e-mail: jagl@termo.uva.es

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1. Introduction

In the field of correlation and prediction of multicomponent data, many efforts have been developed on the basis of statistical thermodynamics methods, several theoretical solutions models and semiempirical equations. Nevertheless, each method seems to have its own area of application limited to particular types of systems. These difficulties continue to grow with increasing the number of components.

For instance, the semiempirical expressions more commonly used to predict excess enthalpies, H^E, of ternary systems should be regarded as different geometrical methods to establish the contribution of each one of the three binaries involved to the ternary H^E [1,2].

The more widely applied models to treat mixtures containing one self-associated component, such as alcohols, are the so-called continuous linear association models. As usually, the thermodynamic properties are obtained as the sum of different contributions. The chemical contribution is calculated assuming that the specific interactions lead to the formation of different associated species according to a well-defined set of chemical reactions, each characterized by an equilibrium constant. The problem is to elucidate what polymeric species are present in the system, because the treatment and results depend on this point. So different models have been proposed depending on the number of alcohols in the mixture [3–12]. On the other hand, the physical contribution can be represented by an equation of state (EOS), e.g. the Flory EOS [13] in the ERAS model [14], or by equations as Scatchard–Hildebrand [15], NRTL [16] or UNIQUAC [17]. This contribution plays an essential role while predicting liquid–liquid equilibria since the type of association normally observed is not sufficient by itself to produce phase separation [18]. It is also important to describe H^E of ternary systems involving one alkanol. Thus, the association model of Krestchmer and Wiebe [19], rederived by Renon and Prausnitz [20], which uses the Scatchard–Hildebrand equation to represent the physical interaction, and is generalized to take into account an unassociated active molecule as second compound [21–23] fails when representing H^E of the above ternary systems [3]. It has been shown that UNIQUAC is the most reliable equation to take into account the physical interactions in the framework of association models, leading to the so-called UNIQUAC association theory, widely applied with excellent results (see, e.g. [5,9,24,25]).

DISQUAC [26,27] is a purely physical model based on the rigid lattice theory developed by Guggenheim for liquid mixtures [28]. Here, the excess functions are also calculated as sum of two contributions: a dispersive term which represents the contribution from dispersive forces, and a quasichemical term which arises from the anisotropy of the field forces created by the solution molecules; in the case of G^E, the combinatorial entropy is represented by the Flory–Huggins equation [29]. Under the basic assumption of structure-dependent interaction parameters, we have shown that DISQUAC can be applied to treat any type of mixture. So systems which present strong positive (as methanol+n-alkanes) [30] or negative (1-alkanols+propanal) [31] deviations from the Raoult's law are well described by the model. Other complex mixtures as 1-alkanols+linear monocarboxylic acids can also be treated using DISQUAC [32]. Even, the model results close to the critical points are still valid [30,33].

On the other hand, we have also shown that DISQUAC is a reliable tool to predict VLE and H^E of ternary systems on the basis of binary parameters only, that is neglecting ternary interactions. So systems containing only hydrocarbons [34]; one or two 1-alkanols and hydrocarbons [35–37], or of the type 1-alkanol+linear ketone+hydrocarbon [38] are well represented by the model. The same occurs for VLE of the methanol+2-propanone+$CHCl_3$ mixture at different temperatures [31]. However, in this case H^E predictions are poorer, indicating that probably ternary interactions are needed for this system in order to improve results. This has been developed in terms of the UNIQUAC association model for the treatment of systems with two alkanoic alkanols and one hydrocarbon [8].

2. Ternary systems considered

The mixtures treated can be classified as follows:

– Group I: mixtures containing only hydrocarbons (or CCl_4).
– Group II: mixtures of one polar compound and two hydrocarbons (or CCl_4).

- Group III: mixtures of one alcohol and two hydrocarbons (or CCl_4).
- Group IV: mixtures containing one alcohol, one polar compound and one hydrocarbon (or CCl_4).
- Group V: mixtures of two alcohols and one hydrocarbon (or CCl_4).
- Group VI: mixtures of two alcohols and one polar compound.

Other two groups have also been studied by us previously:

- Group VII: mixtures containing one alcohol, one polar compound and one self-associated component (≠ alkanol; particularly $CHCl_3$) [31].
- Group VIII: mixtures containing two polar compounds and one self-associated component (≠ alkanol; particularly $CHCl_3$) [31].

3. The DISQUAC model

3.1. Assessment of geometrical parameters

The relative molecular volumes, r_i, the surfaces, q_i, and the surface fractions, α_{si}, of all the molecular species have been calculated on the basis of the group volumes and surfaces recommended by Bondi [39], taking the volume and the surface of methane arbitrarily as unity [29]. Values of r_i and q_i of the groups referred to in this work are given in the following references: CH_2 (aliphatic) [29]; C_6H_{12} (cyclohexane) [29]; c–CH_2 (cyclic) [40]; C_6H_6 (benzene) [29]; C_6H_5 (aromatic in toluene) [29]; CCl_4 (tetrachloromethane) [41]; c–O (oxygen in a cycle) [40]; CO (carbonyl in ketones) [42]; OH (hydroxyl in alkanols) [43].

3.2. Theory

The equations used to calculate thermodynamic properties in the framework of DISQUAC are given elsewhere [43]. The temperature dependence of the interaction parameters is expressed in terms of the dispersive (DIS) and quasichemical (QUAC) interchange coefficients $C^{\mathrm{DIS}}_{\mathrm{st},l}$ and $C^{\mathrm{QUAC}}_{\mathrm{st},l}$, where s, t are the different groups present in the mixture; and l is the order of the interchange coefficient: l=1 (Gibbs energy), l=2 (enthalpy), l=3 (heat capacity). For the QUAC part, the coordination number is z=4.

3.3. Values of the interchange coefficients

The interchange coefficients used in this work are listed in Table 1. As it can be seen, many of them are already available in the literature, and only a few have been estimated for this paper. They were fitted following the general procedure described elsewhere [37,43,44].

It should be emphasized that the DISQUAC interaction parameters are considered as structure-dependent, and so, in a given homologous series, each molecule of solute has its own set of interchange coefficients. Nevertheless, some general trends concerning their variation with the molecular structure have been pointed out. In mixtures formed by polar but not self-associated compounds, such variation is usually attributed to inductive effects on DIS coefficients, and steric effects on QUAC coefficients [45,46]. On the other hand, we have found in many series containing alcohols what seems to be a general rule: the QUAC coefficients for l=1 and l=3 are independent of the alcohol size. This is still valid for l=2, although some exceptions are found. So, in systems involving *sec*-alkanols one can distinguish two ensembles depending on the situation of the OH groups (e.g, 2-alkanols and 3-alkanols), each characterized by a different second QUAC interaction parameter [47].

4. Results

Table 2 lists, for the ternary mixtures investigated, the standard relative deviations in pressure, $\sigma_r(P)$, defined by:

$$\sigma_{\mathrm{r}}(P) = \left[1/N\sum(P_{\mathrm{exp}} - P_{\mathrm{calc}})^2/P^2_{\mathrm{exp}}\right]^{1/2} \quad (1)$$

for sets of (x_1, x_2) values at a given T. N is the number of experimental points for each system. In a similar way, and in order to check more carefully the model, values of $\sigma_r(P)$ are also given for the binaries involved in the ternary mixtures studied (Table 2). Note that we have considered, when possible, those binaries cited originally in papers regarding the ternary mixtures treated. For the binaries, it is also shown a comparison between experimental and calculated values of G^E at equimolar composition and temperature T (Table 2) and azeotrope coordinates (Table 3). Results are shown graphically, for selected mixtures, along Figs. 1–5.

It can be seen that DISQUAC provides a good description of the systems under study. The mean standard relative deviation in pressure, calculated as $\sum\sigma_r(P)$/(number of systems), is 0.020 for the ternaries, and 0.017 for the constituent binaries. For the latter, G^Es and azeotrope coordinates are well represented (Tables 2 and 3).

From the results obtained here, and from those obtained previously [31,36,38], summarized in Table 4, some interesting conclusions can be stated:

Table 1
Interaction parameters used in this work. The groups considered are: CH_2, aliphatic; C_6H_{12}, cyclohexane; c–CH_2, cyclic in tetrahydrofuran; C_6H_6, benzene; C_6H_5, aromatic in toluene; CCl_4, tetrachloromethane; CO, carbonyl in linear ketones; c–O, oxygen in a cyclic ether; 1-OH, hydroxyl in 1-alkanols; 2-OH, hydroxyl in 2-alkanols; t-OH, hydroxyl in 2-methyl-2-alkanols

System [a]	Interaction	$C^{DIS}_{st,1}$	$C^{DIS}_{st,2}$	$C^{DIS}_{st,3}$	$C^{QUAC}_{st,1}$	$C^{QUAC}_{st,2}$	$C^{QUAC}_{st,3}$	Ref.
C_6H_{12}+n-C_6	C_6H_{12}/CH_2	0.058	0.120	−0.214	0.00	0.00	0.00	[44]
C_6H_{12}+n-C_7	C_6H_{12}/CH_2	0.034	0.123	−0.250	0.00	0.00	0.00	[44]
C_5H_{10}+n-C_n	c–CH_2/CH_2 [b]	0.000	0.000	0.000	0.00	0.00	0.00	[40]
C_6H_6+n-C_6	C_6H_6/CH_2	0.267	0.565	−0.593	0.00	0.00	0.00	[51]
C_6H_6+n-C_7	C_6H_6/CH_2	0.254	0.559	−0.600	0.00	0.00	0.00	[51]
C_7H_8+n-C_7	C_6H_5/CH_2	0.332	0.570	−0.421	0.00	0.00	0.00	[51]
C_6H_{12}+C_6H_6	C_6H_{12}/C_6H_6	0.2445	0.5619	0.00	0.00	0.00	0.00	[29]
C_6H_{12}+C_7H_8	C_6H_{12}/C_6H_5 [c,d]	0.33	0.65	0.00	0.00	0.00	0.00	—
CCl_4+n-C_5	CCl_4/CH_2	0.103	0.177	−0.220	0.00	0.00	0.00	[51]
C_6H_6+CCl_4	C_6H_6/CCl_4	0.0613	0.0831	0.00	0.00	0.00	0.00	[29]
1CO1+n-C_n	CO/CH_2	2.90	5.05	0.00	6.08	8.25	0.00	[45]
1CO2+n-C_n	CO/CH_2	3.25	6.16	0.00	5.35	6.35	0.00	[45]
1CO1+C_6H_{12}	CO/C_6H_{12}	3.09	5.24	0.00	6.08	8.25	0.00	[45]
1CO2+C_6H_{12}	CO/C_6H_{12}	3.46	6.39	0.00	5.35	6.35	0.00	[45]
1CO2+C_6H_6	CO/C_6H_6	5.50	5.50	0.00	0.00	0.00	0.00	[38]
THF+n-C_n	c–O/CH_2	15.50	24.90	0.00	6.40	9.24	0.00	[40]
MeOH+n-C_n	1-OH/CH_2	1.35	1.60	−9.10	12.20	8.10	71.10	[30]
EtOH+n-C_n	1-OH/CH_2	1.84	0.81	−9.10	12.20	12.20	71.10	[43]
1-PrOH+n-C_n	1-OH/CH_2	2.55	0.18	−15.50	12.20	15.20	71.10	[43]
1-BuOH+n-C_n	1-OH/CH_2	3.00	0.40	−15.50	12.20	15.20	71.10	[43]
i-BuOH+n-C_n	1-OH/CH_2 [c]	3.00	0.40	−15.50	12.20	15.20	71.10	—
MeOH+C_6H_{12}	1-OH/C_6H_{12}	1.67	2.10	−9.10	12.20	8.10	71.10	[52]
EtOH+C_6H_{12}	1-OH/C_6H_{12}	2.20	0.92	−9.10	12.20	12.20	71.10	[52]
1-PrOH+C_6H_{12}	1-OH/C_6H_{12}	2.80	0.18	−14.00	12.20	15.20	71.10	[52]
MeOH+C_6H_6	1-OH/C_6H_6	1.15	−1.15	−9.10	10.30	16.70	21.20	[53]
EtOH+C_6H_6	1-OH/C_6H_6	1.84	−2.60	−9.10	8.90	16.70	21.20	[53]
1-PrOH+C_6H_6	1-OH/C_6H_6	2.45	−2.60	−14.00	8.90	16.70	21.20	[53]
EtOH+C_7H_8	1-OH/C_6H_5	1.90	−2.25	−9.10	8.90	16.70	21.20	[53]
EtOH+CCl_4	1-OH/CCl_4	1.30	−1.45	−9.10	12.20	20.50	62.50	[54]
MeOH+1CO1	1-OH/CO	0.20	−2.80	0.00	1.65	5.00	0.00	[55]
2-PrOH+n-C_n	2-OH/CH_2	3.00	2.50	−15.50	10.00	9.75	68.00	[47]
2-BuOH+n-C_n	2-OH/CH_2	4.15	5.70	−15.50	10.00	9.75	68.00	[47]
2-PrOH+C_6H_{12}	2-OH/C_6H_{12}	3.10	2.60	−12.00	10.00	9.75	68.00	[47]
2-PrOH+C_6H_6	2-OH/C_6H_6	2.90	1.30	−10.00	8.50	12.75	32.50	[56]
2-PrOH+C_7H_8	2-OH/C_6H_5	2.80	1.85	−10.00	8.50	12.75	32.50	[56]
2-PrOH+1CO1	2-OH/CO	3.15	9.70	0.00	1.25	3.00	0.00	[56]
2-PrOH+1CO2	2-OH/CO	3.15	11.20	0.00	1.25	3.00	0.00	[56]
t-PeOH+n-C_n	t-OH/CH_2	0.85	1.20	−21.50	10.00	13.00	80.00	[57]
t-PeOH+C_6H_{12}	t-OH/C_6H_{12}	1.10	0.90	−21.50	10.00	13.00	80.00	[57]
t-PeOH+C_6H_6	t-OH/C_6H_6 [c,e]	1.15	1.30	0.00	8.50	12.00	0.00	—
1-PrOH+THF	1-OH/c-O [c,f]	1.90	34.90	0.00	0.75	2.30	0.00	—
MeOH+EtOH	1-OH/1-OH	0.00	0.00	−5.00	0.00	0.00	0.00	[37]

Table 1 (*Continued*)

System [a]	Interaction	$C_{st,1}^{DIS}$	$C_{st,2}^{DIS}$	$C_{st,3}^{DIS}$	$C_{st,1}^{QUAC}$	$C_{st,2}^{QUAC}$	$C_{st,3}^{QUAC}$	Ref.
MeOH+*i*-BuOH	1-OH/1-OH	1.25	0.70	−5.00	0.00	0.00	0.00	—
EtOH+1-PrOH	1-OH/1-OH	0.25	0.00	−5.00	0.00	0.00	0.00	[37]
MeOH+2-PrOH	1-OH/2-OH [c,g]	0.60	−0.45	−5.00	0.00	0.00	0.00	—
MeOH+2-BuOH	1-OH/2-OH [c,h]	−0.10	0.50	−5.00	0.00	0.00	0.00	—
EtOH+2-PrOH	1-OH/2-OH [c,i]	−0.30	−0.10	−5.00	0.00	0.00	0.00	—
2-PrOH+2-BuOH	2-OH/2-OH [c]	0.00 [j]	0.00 [j]	−5.00 [j]	0.00	0.00	0.00	—

[a] The abbreviations used are: 1CO1, 2-propanone; 1CO2, 2-butanone; THF, tetrahydrofuran; MeOH, methanol; EtOH, ethanol; 1-PrOH, 1-propanol; 1-BuOH, 1-butanol; 2-PrOH, 2-propanol; 2-BuOH, 2-butanol; *t*-PeOH, *tert*-pentanol; *i*-BuOH, isobutanol.

[b] Interaction in THF+n-C_n.

[c] Interaction characterized in this work.

[d] Data used in the fitting: [58] (VLE); [59] (H^E).

[e] Data used in the fitting: [60] (VLE); [61] (H^E).

[f] Data used in the fitting: [62] (VLE and H^E).

[g] Data used in the fitting: [63] (VLE); [64] (H^E).

[h] Data used in the fitting: [65] (VLE); [66] (H^E).

[i] Data used in the fitting: [50] (VLE); [67] (H^E).

[j] Estimated values.

Table 2
Relative standard deviations in pressure, $\sigma_r(P)$, defined by Eq. (1) for the ternary and binary systems considered in this work. For the binary mixtures, a comparison between experimental (exp.) and calculated (calc.) G^E, at equimolar composition and temperature T(K), is also listed. N is the number of data points for each system (for abbreviations, see Table 1)

No.	System	T/K	N	$\sigma_r(P)$	G^E/J mol		Ref.
					exp.	calc.	
Group I							
1	C_6H_{12}+C_6H_6+n-C_6	283.15	10	0.037			[68]
1	C_6H_{12}+C_6H_6	283.15	6	0.008	355	358	[69]
2	C_6H_{12}+n-C_6	283.15	6	0.010	118	106	[69]
3	C_6H_6+n-C_6	283.15	7	0.006	411	413	[69]
2	C_6H_{12}+C_6H_6+n-C_6	288.15	10	0.033			[68]
4	C_6H_{12}+C_6H_6	288.15	6	0.007	343	350	[69]
5	C_6H_{12}+n-C_6	288.15	6	0.007	108	104	[69]
6	C_6H_6+n-C_6	288.15	7	0.006	402	403	[69]
3	C_6H_{12}+C_6H_6+n-C_6	298.15	10	0.030			[68]
7	C_6H_{12}+C_6H_6	298.15	6	0.006	329	334	[69]
8	C_6H_{12}+n-C_6	298.15	6	0.007	91	100	[69]
9	C_6H_6+n-C_6	298.15	7	0.005	384	386	[69]
4	C_6H_{12}+C_6H_6+n-C_6	343.15	6	0.014			[68]
10	C_6H_{12}+C_6H_6	333.15	36	0.004	270	279	[70]
5	C_6H_{12}+C_7H_8+n-C_7	298.15	16	0.029			[71]
11	C_6H_{12}+C_7H_8	298.15	11	0.042	304	296	[71]
12			10	0.002	293 [a]		[58]
13	C_6H_{12}+n-C_7	298.15	11	0.010	74	43	[71]
14			30	0.002	49		[72]
					43		[73]
15	C_7H_8+n-C_7	298.15	11	0.026	282	299	[71]
Group II							
6	1CO1+C_6H_{12}+n-C_6	313.15	53	0.013			[74]
16	1CO1+C_6H_{12}	313.15	30	0.005	1091	1094	[74]
17	1CO1+n-C_6	313.15	30	0.007	1070	1063	[75]
18	C_6H_{12}+n-C_6	313.15	9	0.010	64	95	[74]

Table 2 (*Continued*)

No.	System	T/K	N	$\sigma_r(P)$	G^E/J mol		Ref.
					exp.	calc.	
Group III							
7	$MeOH+C_6H_{12}+n\text{-}C_6$	313.15	51	0.064			[76]
19	$MeOH+C_6H_{12}$	313.15	34	0.059	885 [b]	845	[77]
20	$MeOH+n\text{-}C_6$	313.15	53	0.038	1640	1579	[78]
18	$C_6H_{12}+n\text{-}C_6$	313.15	9	0.010	64	95	[74]
8	$EtOH+C_6H_6+n\text{-}C_6$	308.15	27	0.018			[79]
21	$EtOH+C_6H_6$	313.15	15	0.006	1128	1130	[80]
22	$EtOH+n\text{-}C_6$	308.15	9	0.012	1420	1406	[81]
23	$C_6H_6+n\text{-}C_6$	313.15	15	0.003	362	361	[82]
9	$EtOH+C_6H_6+n\text{-}C_6$	318.15	27	0.017			[79]
24	$EtOH+C_6H_6$	318.15	12	0.009	1120	1132	[83]
25	$EtOH+n\text{-}C_6$	318.15	9	0.013	1450	1428	[81]
10	$EtOH+C_6H_6+n\text{-}C_6$	328.15	43	0.037			[84]
11	$EtOH+C_6H_6+n\text{-}C_6$	328.15	27	0.016			[79]
26	$EtOH+C_6H_6$	328.15	9	0.017	1150	1130	[84]
27	$EtOH+n\text{-}C_6$	328.15	17	0.031	1500	1444	[84]
28	$C_6H_6+n\text{-}C_6$	328.15	14	0.010	358	339	[84]
12	$1\text{-}PrOH+C_6H_6+n\text{-}C_7$	348.15	77	0.015			[85]
29	$1\text{-}PrOH+C_6H_6$	348.15	13	0.007	976	969	[86]
30	$1\text{-}PrOH+n\text{-}C_7$	348.15	14	0.029	1360	1369	[86]
31	$C_6H_6+n\text{-}C_7$	348.15	11	0.006	249	266	[86]
13	$MeOH+C_6H_6+C_6H_{12}$	311.65	10	0.043			[87,111]
32	$MeOH+C_6H_6$	311.65	5	0.004	1259	1293	[87,111]
33	$MeOH+C_6H_6$	313.15	73	0.038	1282	1295	[88]
19	$MeOH+C_6H_{12}$	313.15	34	0.059	885	845	[77]
34	$C_6H_6+C_6H_{12}$	313.15	7	0.006	295	311	[89]
14	$MeOH+C_6H_6+C_6H_{12}$	328.15	10	0.035			[87,111]
35	$MeOH+C_6H_6$	328.15	5	0.014	1288	1313	[87,111]
36	$MeOH+C_6H_{12}$	328.15	6	0.012	1678	1631	[87,111]
37	$MeOH+C_6H_{12}$	328.15	13	0.054	1670		[90]
10	$C_6H_{12}+C_6H_6$	333.15	36	0.004	270	279	[70]
15	$EtOH+C_6H_6+C_6H_{12}$	323.15	19	0.018			[91,111]
26	$EtOH+C_6H_6$	328.15	9	0.017	1150	1130	[84]
38	$EtOH+C_6H_{12}$	323.15	5	0.019	1457	1486	[91,111]
39	$EtOH+C_6H_{12}$	323.15	7	0.028	1433		[92]
40	$EtOH+C_6H_{12}$	323.15	5	0.027	1433		[93]
41	$C_6H_{12}+C_6H_6$	323.15	35	0.004	284	295	[70]
16	$EtOH+C_7H_8+C_6H_{12}$	323.15	19	0.026			[93]
42	$EtOH+C_7H_8$	323.15	5	0.004	1196	1211	[93]
43	$EtOH+C_7H_8$	313.15	40	0.003	1197 [a]	1207	[88]
44	$EtOH+C_7H_8$	333.15	13	0.008	1203	1208	[94]
38	$EtOH+C_6H_{12}$	323.15	5	0.019	1457	1486	[91,111]
39	$EtOH+C_6H_{12}$	323.15	7	0.028	1433		[92]
40	$EtOH+C_6H_{12}$	323.15	5	0.027	1433		[93]
17	$EtOH+C_6H_6+CCl_4$	307.95	36	0.014			[95]
21	$EtOH+C_6H_6$	313.15	15	0.006	1128	1130	[80]
45	$EtOH+CCl_4$	307.95	12	0.020	1140	1166	[95]
46	$C_6H_6+CCl_4$	313.15	14	0.001	82	83	[96]
18	$EtOH+C_6H_6+CCl_4$	323.15	35	0.018			[95]
19			36	0.020			[97,111]
47	$EtOH+C_6H_6$	323.15	10	0.010	1120	1132	[95]
48	$EtOH+C_6H_6$	323.15	9	0.038	1210		[97,111]

Table 2 (*Continued*)

No.	System	T/K	N	$\sigma_r(P)$	G^E/J mol		Ref.
					exp.	calc.	
49	$EtOH+CCl_4$	323.15	12	0.011	1190	1194	[95]
50	$EtOH+CCl_4$	323.15	9	0.020	1198		[97,111]
20	$EtOH+C_6H_6+CCl_4$	333.15	36	0.014			[95]
51	$EtOH+C_6H_6$	333.15	10	0.047	1150	1129	[95]
26	$EtOH+C_6H_6$	328.15	9	0.017	1150	1130	[84]
52	$EtOH+CCl_4$	333.15	12	0.010	1220	1200	[95]
21	$EtOH+C_6H_6+CCl_4$	339.15	36	0.018			[95]
53	$EtOH+CCl_4$	339.15	12	0.011	1220	1198	[95]
54	$C_6H_6+CCl_4$	343.15	7	0.004	86	79	[98]
22	$1\text{-}PrOH+C_6H_6+C_6H_{12}$	323.15	63	0.011			[70]
55	$1\text{-}PrOH+C_6H_6$	323.15	26	0.006	1010	1000	[70]
56	$1\text{-}PrOH+C_6H_{12}$	323.15	26	0.026	1260	1318	[70]
41	$C_6H_{12}+C_6H_6$	323.15	35	0.004	284	295	[70]
23	$1\text{-}PrOH+C_6H_6+C_6H_{12}$	333.15	50	0.013			[70]
57	$1\text{-}PrOH+C_6H_6$	333.15	34	0.005	980	990	[70]
58	$1\text{-}PrOH+C_6H_{12}$	333.15	30	0.029	1260	1328	[70]
10	$C_6H_{12}+C_6H_6$	333.15	36	0.004	270	279	[70]
24	$2\text{-}PrOH+C_6H_6+C_6H_{12}$	313.15	6	0.012			[99]
59	$2\text{-}PrOH+C_6H_6$	313.15	4	0.029	1012	1041	[99]
60	$2\text{-}PrOH+C_6H_6$	318.15	12	0.013	1030 [a]	1033	[100]
61	$2\text{-}PrOH+C_6H_{12}$	313.15	4	0.014	1187	1190	[99]
34	$C_6H_6+C_6H_{12}$	313.15	7	0.006	295	311	[89]
25	$2\text{-}PrOH+C_6H_6+C_6H_{12}$	328.15	6	0.005			[99]
62	$2\text{-}PrOH+C_6H_6$	328.15	4	0.018	978	1015	[99]
63	$2\text{-}PrOH+C_6H_6$	333.15	12	0.013	995	1004	[101,111]
64	$2\text{-}PrOH+C_6H_{12}$	328.15	4	0.008	1194	1192	[99]
65	$2\text{-}PrOH+C_6H_{12}$	323.15	9	0.025	1230 [a]	1192	[102]
41	$C_6H_{12}+C_6H_6$	323.15	35	0.004	284	295	[70]
26	$2\text{-}PrOH+C_6H_6+C_6H_{12}$	342.15	6	0.001			[99]
66	$2\text{-}PrOH+C_6H_{12}$	333.15	10	0.021	1198	1189	[102]
67		342.15	4	0.006	1204	1180	[99]
68	$2\text{-}PrOH+C_6H_6$	342.15	4	0.014	952	981	[99]
69	$2\text{-}PrOH+C_6H_6$	343.15	10	0.016	962	979	[102]
10	$C_6H_{12}+C_6H_6$	333.15	36	0.004	270	279	[70]
27	$2\text{-}PrOH+C_7H_8+C_6H_{12}$	318.15	23	0.021			[103]
70	$2\text{-}PrOH+C_7H_8$	313.15	10	0.015	1060	1069	[104]
65	$2\text{-}PrOH+C_6H_{12}$	323.15	9	0.025	1230 [a]	1192	[102]
28	$t\text{-}PeOH+C_6H_6+C_6H_{12}$	343.15	22	0.024			[60]
71	$t\text{-}PeOH+C_6H_6$	343.15	8	0.028	613 [a]	591	[60]
72	$t\text{-}PeOH+C_6H_{12}$	343.15	8	0.014	794	816	[60]
10	$C_6H_{12}+C_6H_6$	333.15	36	0.004	270	279	[70]
Group IV							
29	$1\text{-}PrOH+THF+n\text{-}C_7$	298.15	60	0.026			[62]
73	$1\text{-}PrOH+THF$	298.15	15	0.010	295 [a]	310	[62]
74	$1\text{-}PrOH+n\text{-}C_7$	298.15	16	0.015	1290	1320	[62]
75	$THF+n\text{-}C_7$	298.15	9	0.013	358	403	[62]
30	$2\text{-}PrOH+1CO2+C_6H_6$	323.15	19	0.013			[105,111]
76	$2\text{-}PrOH+1CO2$	323.15	5	0.010	395	441	[105,111]
77	$2\text{-}PrOH+1CO2$	323.15	11	0.004	446		[106]
78	$2\text{-}PrOH+C_6H_6$	323.15	15	0.019	1040	1024	[102]
79	$1CO2+C_6H_6$	313.15	22	0.012	116	150	[107]

Table 2 (*Continued*)

No.	System	T/K	N	$\sigma_r(P)$	G^E/J mol		Ref.
					exp.	calc.	
Group V							
31	MeOH+EtOH+n-C_5	303.15	66	0.032			[108]
80	MeOH+EtOH	303.15	22	0.005	−7	4	[108]
81	MeOH+n-C_5	303.15	22	0.046	1570	1515	[65]
82	EtOH+n-C_5	303.15	24	0.027	1370	1319	[108]
32	MeOH+2-BuOH+n-C_5	303.15	67	0.022			[65]
83	MeOH+2-BuOH	303.15	22	0.020	−73 [a]	−36	[65]
78	MeOH+n-C_5	303.15	22	0.046	1570	1515	[65]
84	2-BuOH+n-C_5	303.15	26	0.016	1050 [a]	1045	[65]
33	EtOH+1-PrOH+n-C_7	303.15	66	0.022			[50]
85	EtOH+1-PrOH	303.15	22	0.003	23	16	[50]
86	EtOH+n-C_7	303.15	23	0.014	1450	1435	[50]
87	1-PrOH+n-C_7	303.15	22	0.012	1300	1331	[50]
34	EtOH+2-PrOH+n-C_7	303.15	66	0.019			[50]
88	EtOH+2-PrOH	303.15	22	0.002	−31 [a]	−26	[50]
86	EtOH+n-C_7	303.15	23	0.014	1450	1435	[50]
89	2-PrOH+n-C_7	303.15	22	0.018	1270	1262	[50]
35	EtOH+i-BuOH+n-C_7	303.15	78	0.016			[109]
90	EtOH+i-BuOH	303.15	21	0.004	45	44	[109]
91	EtOH+n-C_7	303.15	22	0.008	1430	1435	[109]
92	i-BuOH+n-C_7	303.15	22	0.018	1180	1211	[109]
36	2-PrOH+2-BuOH+n-C_3	328.15	3	0.008			[110]
93	2-BuOH+n-C_3	328.15	11	0.059		778	[110]
37	2-PrOH+2-BuOH+n-C_3	348.15	3	0.012			[110]
94	2-BuOH+n-C_3	348.15	11	0.062		762	[110]
38	2-PrOH+2-BuOH+n-C_3	368.15	3	0.021			[110]
95	2-BuOH+n-C_3	368.15	11	0.078		727	[110]
Group VI							
39	MeOH+2-PrOH+1CO1	328.15	67	0.011			[63]
96	MeOH+2-PrOH	328.15	20	0.015 [a]	71	68	[63]
97	MeOH+1CO1	328.15	28	0.008	434	411	[63]
98	2-PrOH+1CO1	328.15	14	0.032	496	435	[63]

[a] System used in the estimation of interchange coefficients.

[b] Non-miscible system, value at x_1=0.125.

1. Deviations for the investigated ternary and binary mixtures are similar. In consequence, it is possible to neglect ternary interactions and provide good predictions of isothermal VLE.
2. In general, results are independent of the type of system considered. Solutions with a high number of contacts (up to 6) are usually well represented. Of course, larger deviations are obtained when the ternary involves a binary with a miscibility gap, or when the system temperature is close to the critical, but even then predictions are still valid.
3. The model yields good results over a rather wide range of temperature.

The results are more valuable because only a few binary systems of those tested here were correlated to obtain interaction parameters which were later used to predict VLE of the related ternary.

Finally, it should be mentioned that for solutions containing 2-methyl-1-propanol, the interaction parameters of 1-butanol were used. In spite of this, the good results obtained are noteworthy. Previous calculations suggest that the

Table 3
Comparison of experimental (exp.) coordinates of azeotropes: temperature (T^{az}/K); mole fraction (x_1^{az}) and pressure (P^{az}/kPa) with DISQUAC results (calc.) for some binary mixtures considered in this work

System [a]	T^{az}/K	x_1^{az}		P^{az}/kPa		Ref.
		exp.	calc.	exp.	calc.	
C_6H_6+n-C_6	283.15	0.117	0.082	10.18	10.12	[69]
	298.15	0.099	0.067	20.28	20.22	[69]
C_6H_{12}+C_6H_6	283.15	0.522	0.508	7.261	7.231	[69]
	298.15	0.525	0.496	14.77	14.74	[69]
	323.15	0.487	0.474	40.36	40.54	[70]
	333.15	0.478	0.463	57.62	57.79	[70]
1CO1+n-C_6	313.15	0.636	0.640	71.73	71.68	[75]
1CO1+C_6H_{12}	313.15	0.748	0.746	63.08	63.03	[74]
MeOH+n-C_5	303.15	0.193	0.189	99.99	97.91	[65]
MeOH+n-C_6	313.15	0.479	0.487	69.03	66.82	[78]
EtOH+n-C_5	303.15	0.092	0.088	86.78	86.16	[108]
EtOH+n-C_6	308.15	0.275	0.279	39.95	39.46	[81]
	328.15	0.333	0.332	90.35	88.05	[84]
EtOH+n-C_7	303.15	0.556	0.558	16.26	16.17	[50]
1-PrOH+n-C_7	298.15	0.256	0.261	7.819	7.846	[62]
	303.15	0.278	0.286	10.22	10.29	[50]
	348.15	0.440	0.420	70.80	71.03	[86]
2-PrOH+n-C_7	303.15	0.476	0.483	13.04	12.98	[50]
i-BuOH+n-C_7	303.15	0.149	0.156	8.706	8.847	[109]
EtOH+C_6H_{12}	323.15	0.410	0.415	56.66	57.98	[92]
1-PrOH+C_6H_{12}	323.15	0.178	0.197	42.06	42.66	[70]
	333.15	0.206	0.219	61.18	62.44	[70]
2-PrOH+C_6H_{12}	323.15	0.326	0.316	48.57	47.83	[102]
	333.15	0.366	0.349	72.11	70.42	[102]
t-PeOH+C_6H_{12}	343.15	0.107	0.143	73.20	73.18	[60]
MeOH+C_6H_6	313.15	0.558	0.548	48.82	48.85	[88]
EtOH+C_6H_6	328.15	0.398	0.413	62.63	61.93	[84]
1-PrOH+C_6H_6	323.15	0.145	0.150	39.27	39.03	[70]
	333.15	0.168	0.167	56.70	56.90	[70]
	348.15	0.215	0.194	95.10	94.71	[86]
2-PrOH+C_6H_6	318.15	0.291	0.283	36.49	36.84	[100]
	333.15	0.352	0.323	66.90	66.43	[101,111]
	343.15	0.386	0.368	99.55	95.55	[102]
t-PeOH+C_6H_6	343.15	0.066	0.091	71.02	71.82	[60]
EtOH+C_7H_8	313.15	0.719	0.718	20.37	20.36	[88]
	333.15	0.762	0.787	51.04	51.45	[94]
2-PrOH+C_7H_8	313.15	0.666	0.653	16.40	16.27	[104]
EtOH+CCl_4	323.15	0.340	0.344	57.42	57.50	[95]
	333.15	0.371	0.379	85.07	84.02	[95]
MeOH+1CO1	328.15	0.202	0.202	100.8	100.5	[63]
2-PrOH+1CO2	298.15	0.070	0.083	12.12	12.14	106]
	323.15	0.191	0.191	36.23	36.46	[106]

[a] For abbreviations, see Table 1.

properties of this type of isomeric alcohols can be represented modifying only the second DIS interaction parameter of the corresponding 1-alkanol.

5. Comparison with other models

Equations such as NRTL [16], UNIQUAC [17] or Wilson [48] have been applied to predict VLE of

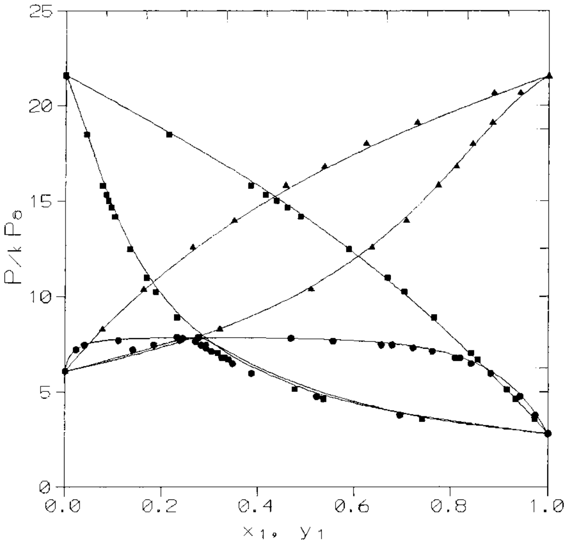

Fig. 1. Comparison of theory with experiment for the VLE diagram at 298.15 K for the binaries involved in the ternary system 1-propanol+THF+n-C_7. Total pressure, P, vs. x_1 and y_1, the mole fractions of the first compound in the liquid and vapor phases, respectively. Lines, predicted values, points, experimental results [62]: ●, 1-propanol(1)+n-C_7 (2); ■, 1-propanol(1)+THF(2); ▲, THF(1)+n-C_7 (2).

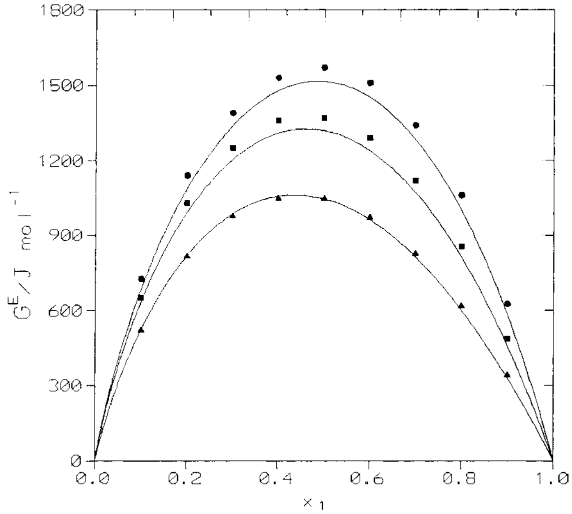

Fig. 3. Comparison of theory with experiment for G^E at 303.15 K for alkanols(1)+n-C_5 (2) mixtures. Lines, predicted values; points, experimental results: ●, methanol [65]; ■, ethanol [108]; ▲, 2-butanol [65].

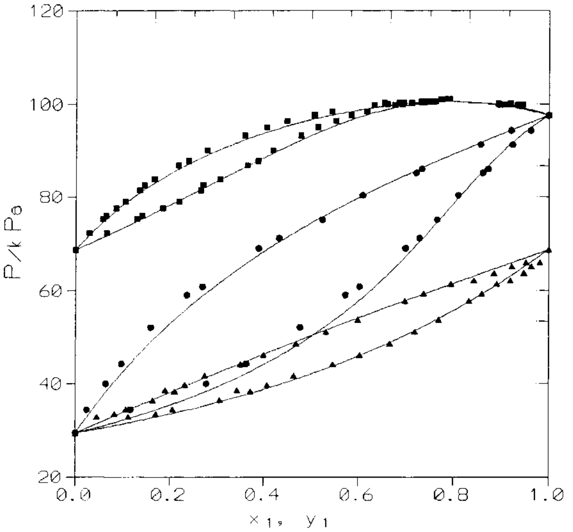

Fig. 2. Comparison of theory with experiment for the VLE diagram at 328.15 K for the binaries involved in the ternary system methanol+2-propanol+2-propanone. Total pressure, P, vs. x_1 and y_1, the mole fractions of the first compound in the liquid and vapor phases, respectively. Lines, predicted values, points, experimental results [63]: ●, methanol(1)+2-propanol(2); ■, 2-propanone(1)+methanol(2); ▲, 2-propanone(1)+2-propanol(2).

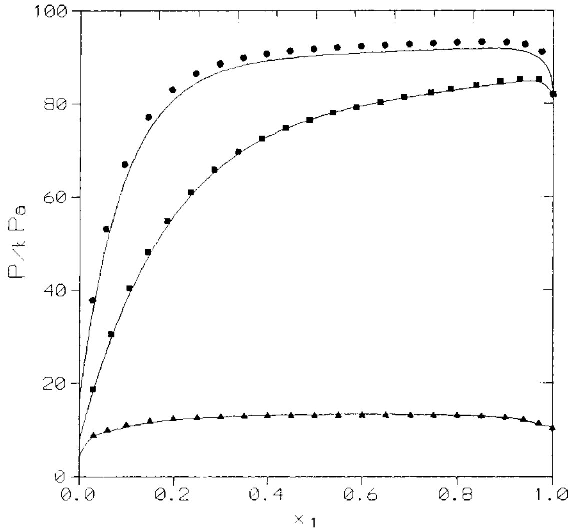

Fig. 4. Comparison of theory with experiment for the VLE, at 303.15 K and different C values (C=zz_2/(zz_2+zz_3), where zz is the overall mole fraction in the equilibrium cell), for ternary systems containing two alcohols and one n-alkane. Solid lines, DISQUAC predictions; points experimental results: ●, n-C_5 (1)+methanol(2)+ethanol(3) (C=0.2459) [108]; ■, n-C_5 (1)+methanol(2)+2-butanol(3) (C=0.5061) [65]; ▲, ethanol(1)+n-C_7 (2)+1-propanol(3) (C=0.4943) [50].

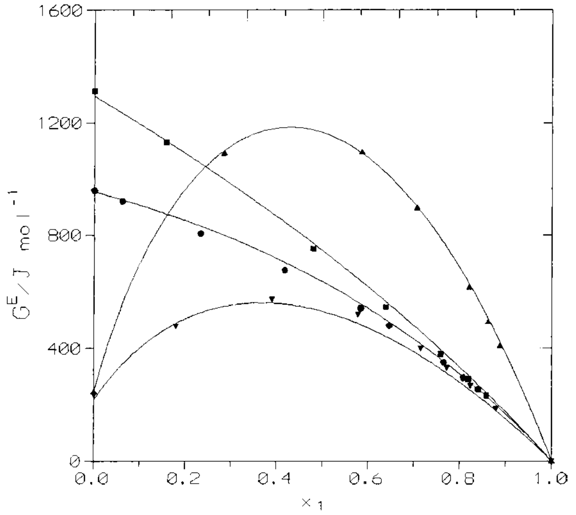

Fig. 5. Comparison of DISQUAC with UNIQUAC results for G^E, at 298.15 K and different x_i/x_j values, for the 1-propanol(1)+THF(2)+n-C_7 (3) system. Lines, DISQUAC calculations; points, UNIQUAC results [62]: ●, x_1/x_3=1/4.3; ■, x_1/x_3=1/1.68; ▲, x_1/x_3=1/0.27; ▼, x_2/x_3=1/0.19.

ternary data using only the parameters obtained for the constituent binaries. A careful study for some selected systems is presented in Ref. [49].

At 343.15 K, the standard deviations $\sigma(P)$ in pressure is defined by:

$$\sigma(P)/kPa = \left[1/N\sum(P_{\text{exp}} - P_{\text{calc}})^2\right]^{1/2} \quad (3)$$

given by the NRTL, UNIQUAC or Wilson equations for the cyclohexane+benzene+n-C_6 are 2.6, 2.8 and 2.8 kPa, respectively [49]. The DISQUAC result is 1.2 kPa. At 298.15 K, the three equations give $\sigma(P)$=0.215 kPa [49], while the DISQUAC value gives 0.230 kPa.

The main problem when applying such equations is related to the parameter temperature-dependence [49]. As already mentioned, DISQUAC predictions are valid over a rather wide range of temperature.

It is also interesting to compare DISQUAC results with those given by different association models (Table 5). Some of the main features of the models considered here are:

Table 4
Summary of the results obtained in this work and in Refs. [31,36,38]

Type of system	Number of ternary/binary systems	Range of temperature (K)	$\bar{\sigma}_r(P)$
Group I	5	283.15–343.15	0.029
	15		0.010
Group II	1	298.15	0.013
	3		0.007
Group III	24 [c]	307.95–348.15	0.020
	61 [d]		0.016
Group IV	14 [e]	293.15–372.70	0.025
	40 [f]		0.017
Group V	9	303.15–368.15	0.020
	19		0.023
Group VI	1	328.15	0.011
	3		0.018
Group VII [a]	3	298.15–323.15	0.034
	9		0.013
Group VIII [b]	1	303.15	0.013
	3		0.009

[a] Systems of the type: 1-alkanol+polar compound+self-associated compound (≠ alcohol; particularly $CHCl_3$); see Ref. [31].
[b] Systems of the type: polar compound+polar compound+self-associated compound (≠ alcohol; particularly $CHCl_3$); see Ref. [31].
[c] Included two ternary systems treated in Ref. [36].
[d] Included five binary systems treated in Ref. [36].
[e] Included 12 ternary systems treated in Ref. [38].
[f] Included 33 binary systems treated in Ref. [38].

Table 5
Comparison of deviations given by different models for some mixtures considered in this work: Model I [50]; Model II [12]; Model III [8]; Model IV [9]; Model V [10] (see text); DQ, DISQUAC (this work). Decimal numbers are standard relative deviations (Eq. (1)); integer numbers are standard deviations (Eq. (3)) given in Pa. Also are indicated the temperature, T (in K), and the number of data points, N, of each system (see Table 1 for abbreviations.)

	N	T/K	Deviations of models					
			I	II	III	IV	V	DQ
MeOH+EtOH+n-C_5 [a]	66	303.15	0.003	0.003		0.060		0.032
MeOH+EtOH [a]	22	303.15	0.001	0.001		10		0.005/68
MeOH+n-C_5 [b]	22	303.15	0.005	0.005	760	50		0.046/3600
EtOH+n-C_5 [a]	24	303.15	0.003	0.003		80		0.027/1400
MeOH+2-BuOH+n-C_5 [b]	67	303.15	0.006	0.009	0.021	0.009		0.022
MeOH+2-BuOH [b]	22	303.15	0.011	0.018	170	10		0.024/170
2-BuOH+n-C_5 [b]	26	303.15	0.006	0.006	640	30		0.016/700
EtOH+1-PrOH+n-C_7 [c]	66	303.15	0.003	0.003				0.022
EtOH+1-PrOH [c]	22	303.15	0.001	0.001				0.003
EtOH+n-C_7 [c]	23	303.15	0.003	0.003	40	20		0.014/200
1-PrOH+n-C_7 [c]	22	303.15	0.001	0.001				0.012
EtOH+2-PrOH+n-C_7 [c]	66	303.15	0.002	0.005				0.019
EtOH+2-PrOH [c]	22	303.15	0.001	0.001				0.002
2-PrOH+n-C_7 [c]	22	303.15	0.003	0.003				0.018
EtOH+i-BuOH+n-C_7 [d]	78	303.15	0.007	0.003	0.019	0.039		0.016
EtOH+i-BuOH [d]	21	303.15	0.005	0.003	10	20		0.004/27
i-BuOH+n-C_7 [d]	22	303.15	0.002	0.002	80	40		0.018/150
2-PrOH+C_6H_6+C_6H_{12} [e]	6	313.15					0.006	0.012
	6	328.15					0.010	0.005

[a] [108].
[b] [65].
[c] [50].
[d] [109].
[e] [99].

MODEL I [50]: valid for mixtures of two alcohols with one hydrocarbon. It needs an equilibrium constant for each alcohol, a single parameter for each alcohol–hydrocarbon pair and a cross-association constant for each alcohol–alcohol pair. The latter magnitude can be used as an adjustable parameter, or calculated as the geometrical mean of the self-association constants of the pure alcohols. Results shown in Table 5 were obtained using the cross-association constant as an adjustable parameter.

MODEL II [12]: improves model I; can be applied to mixtures of any number of alcohols and n-alkanes. It takes into account physical interactions between alcohol molecules, which are characterized by a parameter to be fitted. In exchange, the cross-association constant were obtained using the geometric mean rule.

MODEL III [8]: UNIQUAC association model.

MODEL IV [9]: similar to model III, but using four adjustable parameters in the residual term of the original UNIQUAC association model, characterized by only two adjustable parameters.

MODEL V [10]: It is similar to model III, but with a different residual term, due to the segment fractions of components are redefined.

Models I and II yield much better results than the remainder solutions theories considered here. They predict very accurately the isothermal VLE of ternary systems on the basis of the parameters obtained correlating the data of the constituent binaries. However, their field of application is very limited. Models III, IV, V and DISQUAC provide similar predictions on VLE for the ternary systems listed in Table 5. Results for the binaries involved are better using association theories, probably because they also correlate the binary data. In exchange, the DISQUAC interaction parameters were obtained, in the most of the cases, using different data to those treated in the present article.

6. Conclusions

DISQUAC is a reliable tool to predict isothermal VLE of any type of ternary system on a wide range of temperature. Properties of the constituent binaries, such as G^E or coordinates of azeotropes are also well represented.

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