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Analysis of {[N,N-dialkylamide or 1-methyl-2-pyrrolidone] + 1-chloroalkane or α,ω-dichloroalkane} mixtures in terms of group contribution

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ABSTRACT

The experimental vapour–liquid equilibria, VLE, excess molar Gibbs energies, G_m^E, and excess molar enthalpies, H_m^E, determined previously for the systems N,N-dimethylformamide, or N,N-dimethylacetamide, or 1-methyl-2-pyrrolidone + 1-chlorobutane, or +1-chlorohexane, or +1-chlorooctane, or +1,2-dichloroethane, or +1,4-dichlorobutane, or +1,6-dichlorohexane, are examined using the DISQUAC group contribution model. The dispersive and quasi-chemical interchange parameters for the contacts chloro-amide in the mixtures with N,N-dialkylamide or 1-methyl-2-pyrrolidone with chloro- or α,ω-dichloroalkane were calculated. The model reproduces quite well the experimental data.

1. Introduction

The low vapour pressure of the amides and of some chloroalkanes like 1-chlorooctane or 1,6-dichlorohexane makes it complicated to obtain experimental values for their thermodynamic properties. Hence the great interest in predictive methods. The DISQUAC group contribution method [1] pretends to improve the quality of the predictions by using structure-dependent interactions parameters. It has been applied successfully to many classes of substances including mixtures containing fluoro- [2], chloro- [3,4], bromo- [5,6], iodoalkanes [7,8], monohaloalkanes [9] and polyhaloalkanes [10–12]. In this work our previous experimental results [13–15] on vapour–liquid equilibria, VLE, excess molar Gibbs energies, G_m^E, and excess molar enthalpies, H_m^E, of the mixtures N,N-dimethylformamide, or N,N-dimethylacetamide, or 1-methyl-2-pyrrolidone + 1-chlorobutane, or +1-chlorohexane, or +1-chlorooctane, or +1,2-dichloroethane, or +1,4-dichlorobutane, or +1,6-dichlorohexane are interpreted in terms of the DISQUAC group contribution model.

2. Theory

N,N-dimethylformamide, or N,N-dimethylacetamide + 1-chloroalkane or α,ω-dichloroalkane systems are regarded as possessing three types of surfaces: (i) type *a* (CH_3, CH_2 or, in the case of N,N-dimethylformamide, H); (ii) type *n* (>N—C=O in N,N-dialkylamides); (iii) type *d* (chloro in 1-chloroalkanes and in α,ω-dichloroalkanes). The three types of surfaces *a*, *d*, *n*, generate three pairs of contacts: (*a*,*n*), (*a*,*d*) and (*n*,*d*).

1-Methyl-2-pyrrolidone + 1-chloroalkane or α,ω-dichloroalkane systems are regarded as possessing four types of surfaces: (i) type *a* (CH_3, CH_2); (ii) type *c* (CH_2 cyclic in 1-methyl-2-pyrrolidone); (iii) type *n* (>N—C=O in 1-methyl-2-pyrrolidone); (iv) type *d* (chloro in 1-chloroalkanes and in α,ω-dichloroalkanes). The four types of surfaces *a*, *c*, *d*, *n*, generate six pairs of contacts: (*a*,*c*), (*a*,*n*), (*a*,*d*), (*c*,*n*), (*c*,*d*) and (*n*,*d*).

The equations used to calculate G_m^E and H_m^E can be found in other publications [2–3]. The interactional terms for these thermodynamic properties are given by the DISQUAC model as the sum of a dispersive contribution, $G_{m,INT}^{E,dis}$ or $H_{m,INT}^{E,dis}$, and a quasi-chemical contribution, $G_{m,INT}^{E,quac}$ or $H_{m,INT}^{E,quac}$, thus:

$$G_m^E = G_{m,COMB}^E + G_{m,INT}^{E,dis} + G_{m,INT}^{E,quac} \tag{1}$$

$$H_m^E = H_{m,INT}^{E,dis} + H_{m,INT}^{E,quac} \tag{2}$$

where for a binary system, $G_{m,COMB}^E/RT = x_1 \ln(\phi_1/x_1) + x_2 \ln(\phi_2/x_2)$, that is the Flory-Huggins combinatorial term; $\phi_i = r_i x_i/(r_1 x_1 + r_2 x_2)$ is the volume fraction; x_i is the mole fraction and r_i is the total relative molecular volume of component *i* (*i* = 1, 2).

The temperature dependence of the dispersive or quasi-chemical g_{st} parameters has been expressed by a three-constant

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Table 1
Relative volumes, r_i, relative total surfaces q_i, and molecular surface fractions α_{si} ($s = a$, aliphatic; $s = c$, cyclic aliphatic; $s = n$, amide and $s = d$, chloro) of studied compounds calculated from the group increments r_G and q_G of Bondi [16].

Compound	r_i	q_i	α_{ai}	α_{ci}	α_{ni}	α_{di}
$(CH_3)_2NCOH$	2.7342	2.3586	0.7324	0	0.2676	0
$(CH_3)_2NCOCH_3$	3.3318	2.8241	0.7766	0	0.2234	0
$CH_3(NCO(CH_2)_3)_r$	3.4942	2.7759	0.2634	0.5093	0.2273	0
$Cl{-}(CH_2)_3{-}CH_3$	3.2699	2.7483	0.7742	0	0	0.2258
$Cl{-}(CH_2)_5{-}CH_3$	4.4650	3.6793	0.8313	0	0	0.1687
$Cl{-}(CH_2)_7{-}CH_3$	5.6601	4.6104	0.8654	0	0	0.1346
$Cl{-}(CH_2)_2{-}Cl$	2.5526	2.1724	0.4286	0	0	0.5714
$Cl{-}(CH_2)_4{-}Cl$	3.7477	3.1035	0.6000	0	0	0.4000
$Cl{-}(CH_2)_6{-}Cl$	4.9428	4.0345	0.6923	0	0	0.3077

equation of the type:

$$\frac{g_{st}(T)}{RT} = C_{st.1} + C_{st.2}\left[\frac{T^0}{T} - 1\right] + C_{st.3}\left[\ln\frac{T^0}{T} - \frac{T^0}{T} + 1\right] \qquad (3)$$

where $T^0 = 298.15$ K is the scaling temperature. Accordingly,

$$\frac{h_{st}(T)}{RT} = C_{st.2}\frac{T^0}{T} + C_{st.3}\left[\frac{T^0}{T} - 1\right] \qquad (4)$$

$$C_{st.1} = \frac{g_{st}(T^0)}{RT^0}, \quad C_{st.2} = \frac{h_{st}(T^0)}{RT^0} \quad \text{and} \quad C_{st.3} = \frac{C_{p.st}(T)}{R}$$

are termed interchange energy coefficients. In our approach, we have considered zero as the third interchange coefficient.

The relative molecular volumes, r_i, the total relative surfaces, q_i, and the surface fractions, α_{si} (s,t = a, c, n, d) (i = 1, 2), of all molecular species have been calculated on the basis of the group volumes, V_G, and surfaces, A_G, recommended by Bondi [16], taking the volume and surface of methane as unity. Table 1 collects the geometrical parameters of all compounds used in this paper.

3. Estimation of the interaction parameters

In the application of DISQUAC to N,N-dimethylformamide or N,N-dimethylacetamide + monochloroalkane or α,ω-dichloroalkane mixtures, the interaction parameters used for the amide–alkane contacts were those obtained by González et al. [17] and Lobos et al. [18], and for the alkane–chlorine contacts were those obtained by Kehiaian and Marongiu [3]. Then, in this work the interaction parameters amide–chlorine have been calculated and the following assumptions have been made: (i) The terminal hydrogen atom of N,N-dimethylformamide have an aliphatic behaviour similar to that of CH_3 and CH_2; (ii) the interaction chlorine–amide in both amides, N,N-dimethylformamide and N,N-dimethylacetamide, can be described by the same set of parameters because their dipole moments and their environment are very similar; (iii) the dispersive coefficients are considered constant with the chain length, whereas quasi-chemical exchange energy coefficients are allowed to change. The values obtained for the latter coefficients are found to vary up to the 1,6-dichlorohexane and from then, onwards the value does not change and it is equal to the value for contacts with monochloroalkanes; this variation of the quasi-chemical coefficients suggest that the "proximity effect" diminishes noticeably or even disappears when the separation between the two chlorine atoms in the dihaloalkane molecule is equal or greater than six carbon atoms.

Table 2 presents the dispersive and the quasi-chemical interchange energy coefficients for the three types of contacts occurring in the mixtures of N,N-dimethylformamide or N,N-dimethylacetamide + monochloroalkane or +α,ω-dichloroalkane. The variation of the quasi-chemical parameters with the separation of the chlorine atoms in the α,ω-dichloroalkane can be seen in Fig. 1a.

In the application of DISQUAC to the mixtures of 1-methyl-2-pyrrolidone + monochloroalkane or α,ω-dichloroalkane interaction parameters used for the cyclic amide–alkane and cyclic amide–cycloalkane contacts were those obtained by González et al. [17], and for the alkane–chlorine, alkane–cycloalkane and chlorine–cycloalkane contacts those obtained by Kehiaian and Marongiu [3]. To calculate the interaction parameters

Table 2
Interchange energy coefficients $C^{dis}_{st./}$ and $C^{quac}_{st./}$ for contacts (s,t) (a, aliphatic; n, amide and d, chloro) in mixtures with N,N-dimethylformamide and N,N-dimethylacetamide (/ = 1, Gibbs energy; / = 2, enthalpy); n is the number of atoms of carbon in the α,ω-dichloroalkane.

Contact	n	$C^{dis}_{st.1}$	$C^{dis}_{st.2}$	$C^{quac}_{st.1}$	$C^{quac}_{st.2}$
(a,n)[a]	2	4.8/4.75[d]	12.000	8.000	6.000
	4	4.8/4.75[d]	12.000	8.000	6.000
	≥6	4.8/4.75[d]	12.000	8.000	6.000
	1-Chloroalkanes	4.8/4.75[d] 4.65[**]/4.4[d,**]	12.000	8.000	6.000
(a,d)[b]	2	0.093	0.180	1.670	3.200
	4	0.093	0.180	2.280[*]	3.680
	≥6	0.093	0.180	2.340	3.750
	1-Chloroalkanes	0.093	0.180	2.340	3.750
(n,d)[c]	2	1.500	0.500	−0.100	0.200
	4	1.500	0.500	0.600	1.000
	≥6	1.500	0.500	0.800[*]	1.200
	1-Chloroalkanes	1.500	0.500	0.800	1.200

[*] Estimated values.

[**] 1-Chlorooctane.

[a] Ref. [17] N,N-dimethylformamide.

[b] Ref. [3].

[c] This work.

[d] Ref. [18] N,N-dimethylacetamide.

Table 3

Interchange energy coefficients $C^{dis}_{st.l}$ and $C^{quac}_{st.l}$ for contacts (*s*,*t*) (*a*, aliphatic; *c*, cyclo aliphatic; *n*, amide and *s*,*t* = *d*, chloro) in mixtures with 1-methyl-2-pyrrolidone (*l* = 1, Gibbs energy; *l* = 2, enthalpy); *n* is the number of atoms of carbon in the α,ω-dichloroalkane.

Contact	*n*	$C^{dis}_{st.1}$	$C^{dis}_{st.2}$	$C^{quac}_{st.1}$	$C^{quac}_{st.2}$
(*a*,*c*)[a]	2	0.030	0.117	0.000	0.000
	4	0.030	0.117	0.000	0.000
	≥6	0.030	0.120	0.000	0.000
	1-Chloroalkanes	0.030/0.032**	0.117/0.129**	0.000	0.000
(*a*,*c*)[b]	2	5,250	12.000	8.000	6.000
	4	5.250	12.000	8.000	6.000
	≥6	5.250	12.000	8.000	6.000
	1-Chloroalkanes	5.250/5.000**	12.000	8.000	6.000
(*a*,*d*)[a]	2	0.093	0.180	1.670	3.200
	4	0.093	0.180	2.280*	3.680
	≥6	0.093	0.180	2.340	3.750
	1-Chloroalkanes	0.093	0.180	2.340	3.750
(*c*,*n*)[b]	2	5.000	12.000	8.000	6.000
	4	5.000	12.000	8.000	6.000
	≥6	5.000	12.000	8.000	6.000
	1-Chloroalkanes	5.000	12.000	8.000	6.000
(*c*,*d*)[a]	2	0.049	0.105	1.670	3.200
	4	0.049	0.105	2.280*	3.680
	≥6	0.049	0.105	2.340	3.750
	1-Chloroalkanes	0.049	0.105	2.340	3.750
(*n*,*d*)[c]	2	1.500	0.500	−0.600	−1.000
	4	1.500	0.500	0.300*	−0.300
	≥6	1.500	0.500	0.800*	−0.050
	1-Chloroalkanes	1.500	0.500	0.800	−0.050

* Estimated values.

** 1-Chlorooctane.

[a] Ref. [3].

[b] Ref. [17].

[c] This work.

amide–chlorine the following assumptions have been made: (i) the dispersive exchange energy coefficients for the cyclic amide are the same than for the linear amides; (ii) the "proximity effect" present in α,ω-dichloroalkanes is taken into account letting the quasi-chemical parameters to change while the dispersive parameters used are the same in the mixtures with monochloroalkanes as in the mixtures with α,ω-dichloroalkanes. Similarly to the two linear amides instances, the values obtained for the quasi-chemical coefficients seem to show that the proximity effect between the two chlorine atoms disappears, or diminishes perceptibly, when their separation in the α,ω-dichloroalkane molecule is equal or greater than six carbon atoms. The above mentioned variation can be seen in Fig. 1b. Table 3 presents the quasi-chemical and dispersive exchange energy coefficients for the six types of contacts existing in the mixtures 1-methyl-2-pyrrolidone + monochloroalkane or +α,ω-dichloroalkane.

4. Comparison with experiment and discussion

Table 4 and Figs. 2–8 show a comparison of DISQUAC with experiment for the excess molar enthalpies, isothermal vapour–liquid equilibria and excess Gibbs energies of N,N-dimethylformamide, or N,N-dimethylacetamide, or 1-methyl-2-pyrrolidone + 1-chloroalkane or α,ω-dichloroalkane. Standard deviation for DISQUAC predictions of H^E_m, $\sigma(H^E_m)$, and of vapour pressure, $\sigma(P)$, calculated as:

$$\sigma(H^E_m) = \left\{\sum \frac{(H^E_{\mathrm{DQ}} - H^E_{\mathrm{calc}})^2}{N-m}\right\}^{1/2} \quad \text{and}$$

$$\sigma(P) = \left\{\sum \frac{(P_{\mathrm{DQ}} - P_{\mathrm{exp}})^2}{N}\right\}^{1/2} \tag{5}$$

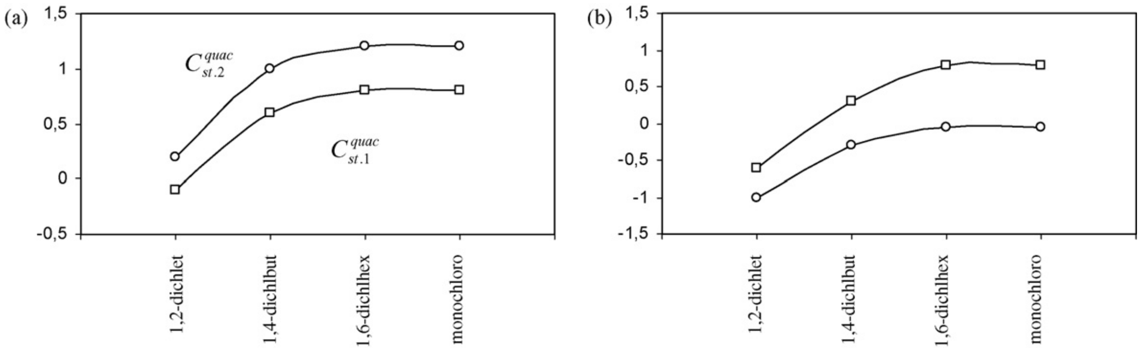

Fig. 1. Quasi-chemical interchange energy coefficients parameters $C^{quac}_{st.l}$ for contacts (*s*,*t*) (a, aliphatic; r, cyclo aliphatic; n, amide and h, chloro) in mixtures with N,N-dimethylformamide, N,N-dimethylacetamide (a) or 1-methyl-2-pyrrolidone (b) (*l* = 1, Gibbs energy; *l* = 2, enthalpy) plotted against *u*, the number of C atoms in the α,ω-dichloroalkane.

Table 4
Molar excess Gibbs energies, G_m^E and molar excess enthalpies, H_m^E of amide + chloroalkane mixtures at various temperatures and equimolar composition ($x_1 = 0.5$). Comparison of direct experimental results (Exp.) [13–15] with values calculated (Calc.) using the coefficients from Tables 2 and 3. Standard deviations of molar excess enthalpies, H_m^E, and pressure, P, of amide + chloroalkane mixtures at various temperatures.

System	T (K)	H_m^E (J mol^{-1})		$\sigma(H_m^E)$ (J mol^{-1})	G_m^E (J mol^{-1})		$\sigma(P)$ (kPa)
		Exp.	Calc.		Exp.	Calc.	
$(CH_3)_2NCOH$ (1) + $CH_3–(CH_2)_3–Cl$ (2)	298.15	572	534	38			
	308.15	579	531	46			
	313.15				603	525	0.59
	323.15				607	525	0.72
+ $CH_3–(CH_2)_5–Cl$ (2)	298.15	841	887	47			
	308.15	864	889	29			
	313.15				680	755	0.09
	323.15				679	751	0.14
	333.15				678	746	0.20
+ $CH_3–(CH_2)_7–Cl$ (2)	298.15	1081	1179	103			
	308.15	1127	1186	70			
+ $Cl–(CH_2)_2–Cl$ (2)	298.15	−664	−793	106			
	308.15	−662	−797	111			
	313.15				−368	−481	0.36
	323.15				−359	−471	0.30
+ $Cl–(CH_2)_4–Cl$ (2)	298.15	−56	−97	35			
	308.15	−56	−104	42			
+ $Cl–(CH_2)_6–Cl$ (2)	298.15	340	303	34			
	308.15	346	297	47			
$(CH_3)_2NCOCH_3$ (1) + $CH_3–(CH_2)_3–Cl$ (2)	298.15	363	314	51			
	308.15	374	310	63			
	313.15				353	352	0.29
	323.15				354	353	0.40
+ $CH_3–(CH_2)_5–Cl$ (2)	298.15	606	598	38			
	308.15	638	598	54			
+ $CH_3–(CH_2)_7–Cl$ (2)	298.15	823	844	42			
	308.15	870	848	49			
+ $Cl–(CH_2)_2–Cl$ (2)	298.15	−887	−742	133			
	308.15	−916	−744	154			
	313.15				−555	−524	0.14
	323.15				−544	−517	0.13
+ $Cl–(CH_2)_4–Cl$ (2)	298.15	−314	−191	96			
	308.15	−323	−199	120			
+ $Cl–(CH_2)_6–Cl$ (2)	298.15	90	115	37			
	308.15	95	107	28			
$CH_3NCO(CH_2)_3$ (1) + $CH_3–(CH_2)_3–Cl$ (2)	298.15	150	131	40			
	308.15	189	134				
	313.15				390	434	0.38
	323.15				398	443	0.24
+ $CH_3–(CH_2)_5–Cl$ (2)	298.15	414	453	48			
	308.15	468	461	52			
	313.15				809	692	0.09
	323.15				816	700	0.15
	333.15				824	706	0.28
+ $CH_3–(CH_2)_7–Cl$ (2)	298.15	634	735	98			
	308.15	696	747	43			
+ $Cl–(CH_2)_2–Cl$ (2)	298.15	−1265	−1294	51			
	308.15	−1276	−1285	26			
	313.15				−768	−736	0.19
	323.15				−750	−718	0.42
+ $Cl–(CH_2)_4–Cl$ (2)	298.15	−678	−640	32			
	308.15	−651	−638	15			
+ $Cl–(CH_2)_6–Cl$ (2)	298.15	−199	−213	20			
	308.15	−160	−211	51			

where H_{calc}^E are the values obtained with Redlich–Kister equation [13–15], N are the total number of experimental measurements and m are the number of coefficients of fitted equations, are also included. However, the values for $\sigma(P)$ are not very significant and must be taken carefully given that in various systems, the experimental measurements have been carried out along a reduced interval of molar fractions, because of experimental difficulties due to the low vapour pressures of the pure components.

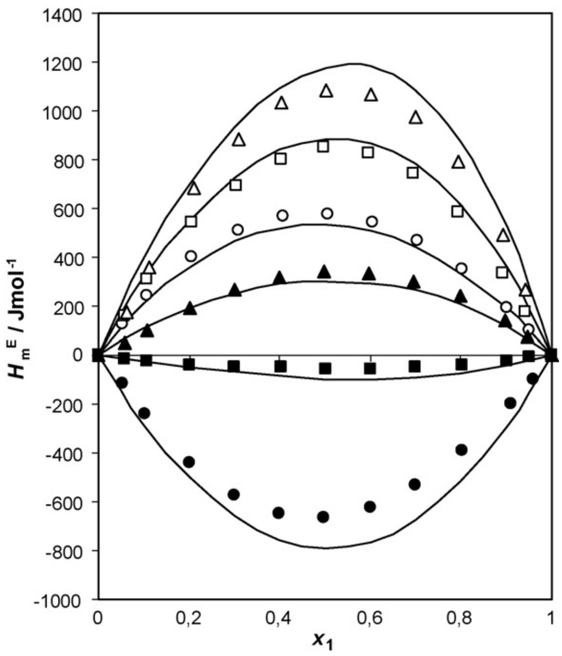

Fig. 2. Comparison of theory with experiment for the molar excess enthalpies, H_m^E for the mixtures N,N-dimethylformamide + 1-chlorobutane (○); +1-chlorohexane (□); +1-chlorooctane (△); +1,2-dichloroethane (●); +1,4-dichlorobutane (■); +1,6-dichlorohexane (▲) at 298.15 K. DISQUAC (—).

DISQUAC reproduces satisfactorily the vapour–liquid equilibria and both the symmetry and magnitude of the H_m^E-composition curves, particularly taking into account the number of fitted parameters, and the occurrence of intramolecular effects (especially the "proximity effect") and other effects that have not been taken into account such as the "orientational effect" [19]. DISQUAC reproduces less adequately the magnitude of G_m^E-composition curves.

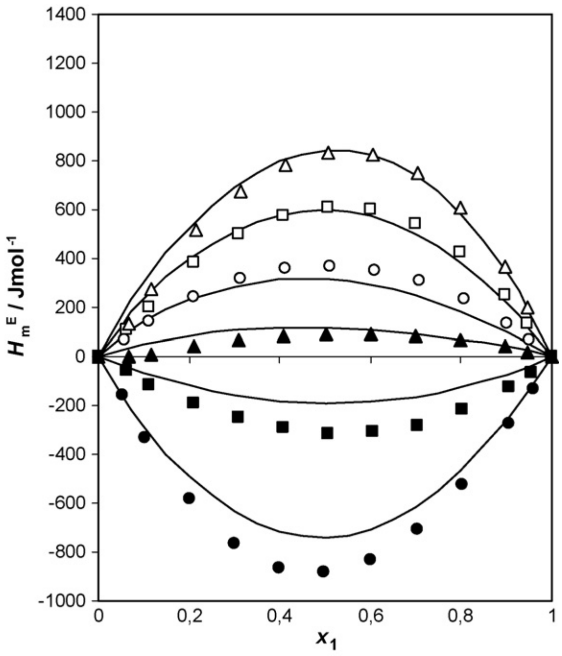

Fig. 3. Comparison of theory with experiment for the molar excess enthalpies, H_m^E for the mixtures N,N-dimethylacetamide + 1-chlorobutane (○); +1-chlorohexane (□); +1-chlorooctane (△); +1,2-dichloroethane (●); +1,4-dichlorobutane (■); +1,6-dichlorohexane (▲) at 298.15 K. DISQUAC (—).

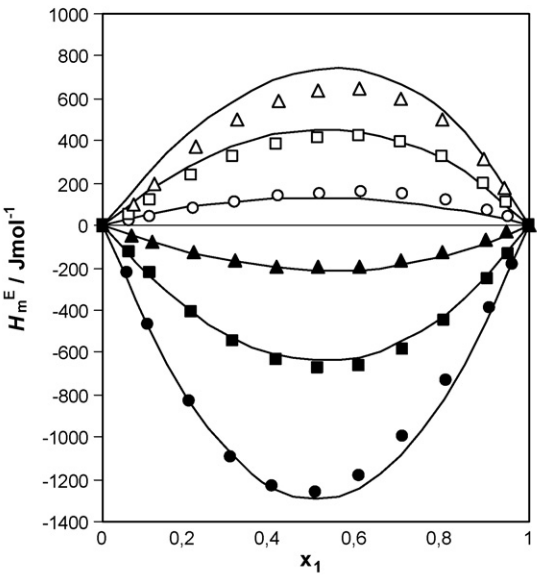

Fig. 4. Comparison of theory with experiment for the molar excess enthalpies, H_m^E for the mixtures 1-methyl-2-pyrrolidone + 1-chlorobutane (○); +1-chlorohexane (□); +1-chlorooctane (△); +1,2-dichloroethane (●); +1,4-dichlorobutane (■); +1,6-dichlorohexane (▲) at 298.15 K. DISQUAC (—).

When considering the excess molar enthalpies, H_m^E, the concordance with the experimental values is good for those systems containing the cyclic amide; for the mixtures with N,N-dimethylformamide and N,N-dimethylacetamide it can be considered quite acceptable, particularly bearing in mind that in this model the excess functions are the sum of two contributions, dispersive and quasi-chemical, and that in some cases the magnitude of them is large, with opposite sign and similar in absolute value. In addition, the model predicts correctly the symmetry of the H_m^E-composition curves and the sign of their coefficient of temperature.

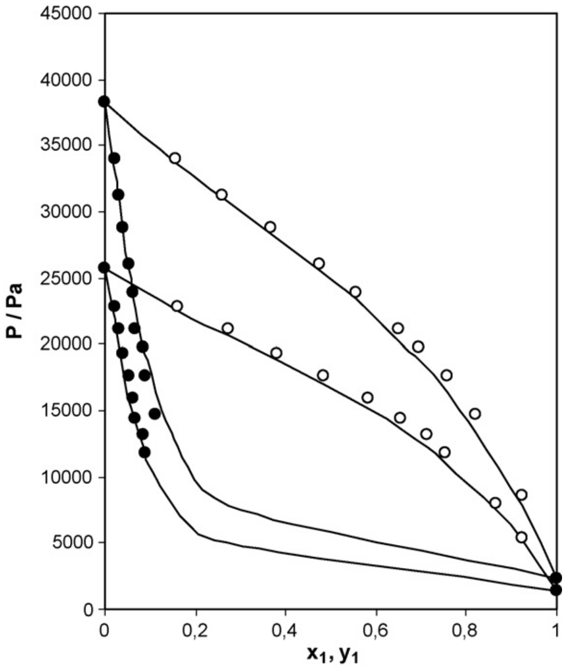

Fig. 5. Isothermal vapour–liquid equilibria for the mixture N,N-dimethylformamide + 1-chlorobutane. Liquid molar fraction (○); vapour molar fraction (●) at 313.15 and 323.15 K. DISQUAC (—).

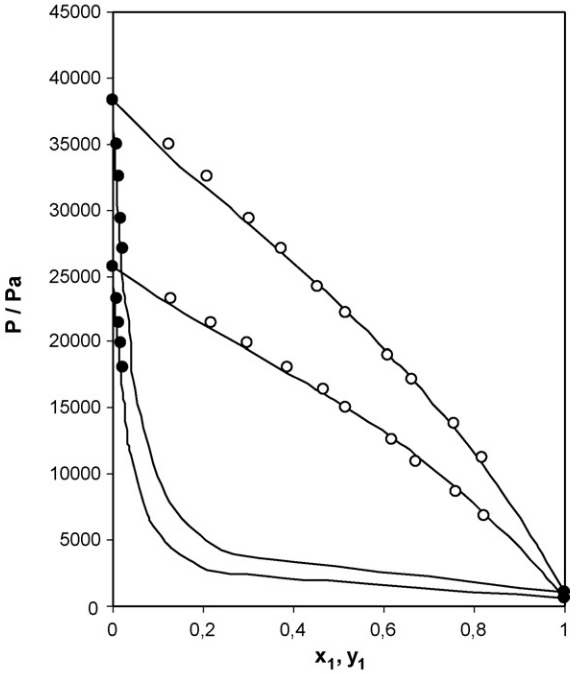

Fig. 6. Isothermal vapour–liquid equilibria for the mixture N,N-dimethylacetamide + 1-chlorobutane. Liquid molar fraction (○); vapour molar fraction (●) at 313.15 and 323.15 K. DISQUAC (—).

In the application of DISQUAC to liquid-vapour equilibria the agreement between the *P*–*x* values obtained with the model and the experimental can be considered satisfactory. Unlike the *P*–*x* values, the molar excess Gibbs energies, G_m^E, present larger deviations, therefore, the predictions from the model are less satisfactory. As for the molar excess enthalpies, DISQUAC predictions are better in the mixtures of 1-methyl-2-pyrrolidone + 1-chloroalkane or +α,ω-dichloroalkane than in those mixtures containing the N,N-alkylamides. On the other hand, the azeotrope that N,N-dimethylformamide + 1-chlorohexane system presents at 313.15, 323.15, and 333.15 K is reproduced by the model adequately. However, the predicted variation of the properties with temperature is not always in accord with the experimental data.

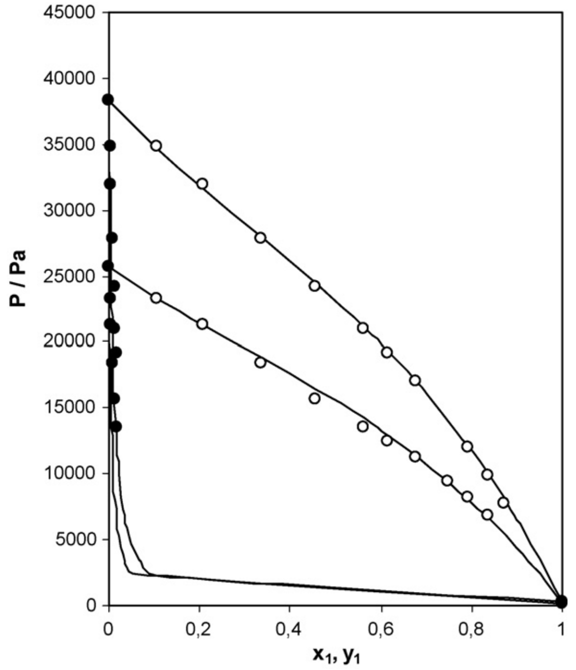

Fig. 7. Isothermal vapour–liquid equilibria for the mixture 1-methyl-2-pyrrolidone + 1-chlorobutane. Liquid molar fraction (○); vapour molar fraction (●) at 313.15 and 323.15 K. DISQUAC (—).

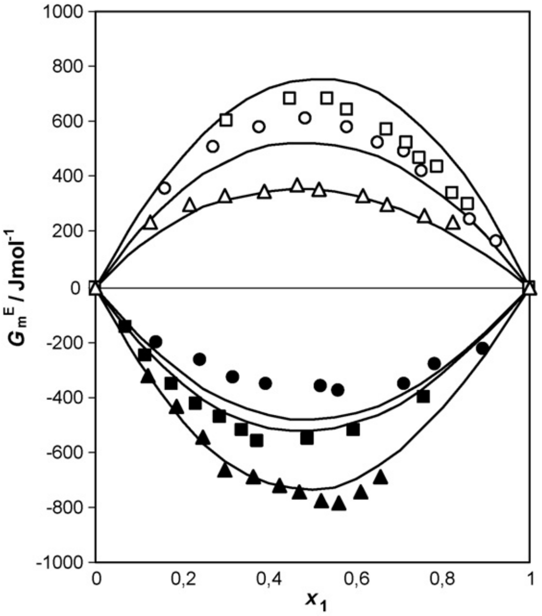

Fig. 8. Comparison of theory with experiment for the molar excess Gibbs energies, G_m^E for the mixtures N,N-dimethylformamide + 1-chlorobutane (○); +1-chlorohexane (□); N,N-dimethylacetamide + 1-chlorobutane (△); N,N-dimethylformamide (●), or N,N-dimethylacetamide (■), or 1-methyl-2-pyrrolidone (▲) +1,2-dichloroethane at 313.15 K. DISQUAC (—).

When comparing DISQUAC with other group contribution models it should be remarked that in our application, the model has reproduced the experimental curves of the two N,N-alkylamides using the same dispersive parameters for both the mixtures with 1-chloroalkanes and the mixtures with α,ω-dichloroalkanes, and varying only the quasi-chemical exchange energy coefficients with the length of the aliphatic chain of the α,ω-dichloroalkane until reaching the value of the monochloroalkane, and thus confirming that the “proximity effect” between the two chlorine atoms in the molecule tends to disappear when the separation between them is equal to or greater than 6 methylene groups. This had already been seen in previous works on mixtures of α,ω-dihaloalkanes with various solvents [20].

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