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Calorimetric and phase equilibrium data for linear carbonates + hydrocarbons or $+\mathrm{CCl_4}$ mixtures. Comparison with DISQUAC predictions [1]

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Abstract

The DISQUAC interaction parameters for the linear organic carbonate–alkane, carbonate–cyclohexane, carbonate–benzene or –toluene, and carbonate–$\mathrm{CCl_4}$ contacts are revised on the basis of new experimental data on vapor–liquid equilibria for dimethyl or diethyl carbonate + n-alkane mixtures.

The new parameters differ slightly from the previous ones. The main conclusions remain valid. The quasichemical interchange coefficients for carbonate–alkane or –cyclohexane contacts, and the purely dispersive interchange coefficients for carbonate–benzene or –toluene and carbonate–$\mathrm{CCl_4}$ contacts, show a relatively weak steric effect. The model provides a fairly consistent description of low pressure fluid phase equilibria (vapor–liquid, liquid–liquid and solid–liquid) and related excess functions (Gibbs energy and enthalpy) using the same set of parameters.

INTRODUCTION

We are interested in the characterization of the interactions between organic carbonates and organic solvents. Therefore, we have reported experimental data on (i) vapor–liquid equilibria (VLE) for systems involving di-n-alkyl carbonate + cyclohexane, +benzene or +tetrachloromethane [2,3], (ii) liquid–liquid equilibria (LLE) for dimethyl carbonate + n-alkane mixtures [4,5], and (iii) molar excess enthalpies H^E for systems containing di-n-alkyl carbonate + n-alkanes, +cyclohexane, +methylcyclohexane, +benzene, +toluene or +tetrachloromethane [6–8].

In a theoretical treatment using the DISQUAC model [9], we have characterized the interactions for linear carbonate–alkane, carbonate–

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[1] Thermodynamics of binary mixtures containing organic carbonates. Part 9 (Part 8 is ref. 1).

cyclohexane, carbonate–benzene or –toluene, and carbonate–CCl_4 [10, 11]. In the absence of VLE data for organic carbonate + *n*-alkane mixtures, the interchange coefficients for the carbonate–alkane interactions were estimated on the basis of VLE data of mixtures containing cyclohexane, to reproduce as well as possible the available data on LLE and H^E.

We have recently reported data on molar excess volumes V^E for systems of dimethyl carbonate + hydrocarbons or $+CCl_4$ [1], VLE and molar excess Gibbs energies G^E for carbonate + *n*-alkane mixtures [12–14], and G^E for systems of carbonates + c-C_6H_{12}, $+C_6H_6$, or $+CCl_4$ [12, 15]. Using our previously published DISQUAC interaction parameters, the G^E of diethyl carbonate + *n*-alkane systems are fairly well represented by the model. However, the predicted G^E values for dimethyl carbonate + *n*-alkane mixtures are somewhat smaller than the experimental ones [12]. Therefore, the purpose of this paper is to provide revised interchange coefficients for the linear organic carbonate–alkane interactions which reproduce better the whole set of experimental data. We also study the influence of this change on the coefficients of the other carbonate–solvent interactions.

NEW EXPERIMENTAL G^E DATA CONSIDERED

Table 1 lists the Redlich–Kister coefficients A_i given by

$$G^E/RT = x_1(1 - x_1) \sum_{i=0} A_i(2x_1 - 1)^i \tag{1}$$

and the standard deviations for the vapor phase mole fraction of carbonate y_1 given by

$$\sigma(y_1) = \left[\sum (y_1^{calc} - y_1^{exp})^2/N\right]^{1/2} \tag{2}$$

obtained in the fitting of all the recently investigated mixtures involving

TABLE 1

Coefficients A_i and standard deviations in the vapor mole fraction $\sigma(y_1)$ for least squares representation of G^E/RT by eqn. (1) for diethyl carbonate(1) + solvent(2) mixtures at $T = 298.65$ K

Solvent	A_0	A_1	$\sigma(y_1)$	Ref.
CH_3–$(CH_2)_4$–CH_3	1.170	−0.112	0.0030	14
CH_3–$(CH_2)_6$–CH_3	1.3223	−0.0901	0.0098	14
CH_3–$(CH_2)_{10}$–CH_3	1.558	0.133	0.0005	14
c-C_6H_{12}	1.3387		0.0026	15
C_6H_6	−0.1590	0.099	0.0041	15
CCl_4	0.08446	−0.1059	0.0014	15

diethyl carbonate. N is the number of experimental data and x_1 is the liquid mole fraction of carbonate.

The coefficients were determined by regression through minimization of the sum of the deviations in the vapor phase mole fraction y_i, all the points weighted equally [14–15]. Vapor phase imperfection was accounted for in terms of the second virial coefficients estimated by the method of Hayden and O'Connell [16]. All the direct experimental isothermal x–y data have been published elsewhere [3, 14, 15].

For mixtures involving dimethyl carbonate, G^E data can be found in refs. 12, 13 and 15.

THEORY

The molecules under study, i.e. linear organic carbonates, alkanes, benzene, toluene and tetrachloromethane, are regarded as possessing three types of surface:

(1) type a, CH_3 and CH_2 groups in *n*-alkanes, carbonates, methylcyclohexane or toluene;

(2) type s, s = b, C_6H_6 (benzene) or C_6H_5 (phenyl group in toluene); s = c (*c*-CH_2 group in cyclohexane); or s = t, CCl_4 (tetrachloromethane);

(3) type d, O–CO–O group in carbonates.

The equations used to calculate G^E and H^E are the same as in other cases [17]. All the geometrical parameters of the compounds referred to in this

TABLE 2

Volumes r_i, total surfaces q_i, and molecular surface fractions α_{ai} and $\alpha_{fi} = 1 - \alpha_{ai}$ (f = s, d) for the compounds referred to in this work

Compound	r_i	q_i	α_{ai}	α_{fi}	Ref.
CH_3–O–CO–O–CH_3	2.6893	2.4276	0.6023	0.3977 [a]	10
CH_3–CH_2–O–CO–O–CH_2–CH_3	3.8844	3.3586	0.7125	0.2875 [a]	10
CH_3–$(CH_2)_4$–CH_3	3.9871	3.3241	1.0000	0	18
CH_3–$(CH_2)_5$–CH_3	4.5847	3.7897	1.0000	0	18
CH_3–$(CH_2)_6$–CH_3	5.1822	4.2552	1.0000	0	18
CH_3–$(CH_2)_8$–CH_3	6.3773	5.1862	1.0000	0	18
CH_3–$(CH_2)_{10}$–CH_3	7.5724	6.1172	1.0000	0	18
CH_3–$(CH_2)_{12}$–CH_3	8.7675	7.0483	1.0000	0	18
CH_3–$(CH_2)_{14}$–CH_3	9.9626	7.9793	1.0000	0	18
c-C_6H_{12}	3.5187	2.5966	0	1.0000 [b]	18, 19
c-C_6H_{11}–CH_3	4.1157	3.0587	1.0000	0	10
C_6H_6	2.8248	2.0724	0	1.0000 [c]	18
C_6H_5–CH_3	3.4760	2.5690	0.2846	0.7154 [c]	18
CCl_4	3.0543	2.5104	0	1.0000 [d]	17

[a] f = d. [b] f = c. [c] f = b. [d] f = t.

work are listed in Table 2. The parameters for the CO_3 group were calculated using the values reported by Bondi [20] for –O–, –CO– and –O–CO–, and are $r_{CO_3} = 1.09229$ and $q_{CO_3} = 0.96552$. Methylcyclohexane was treated as an *n*-alkane owing to the small G^E and H^E values of methylcyclohexane + *n*-alkanes.

The temperature dependence of the interaction parameters is expressed in terms of the dispersive (DIS) or quasichemical (QUAC) interchange coefficients $C^{DIS}_{ed,l}$ and $C^{QUAC}_{ed,l}$, where e = a or s; and $l = 1$ (Gibbs energy), or $l = 2$ (enthalpy). Heat capacity coefficients ($l = 3$) have not been considered.

ESTIMATION OF THE INTERACTION PARAMETERS

The three types of surface generate three pairs of contacts: (a, s), (a, d) and (s, d), where s = b, c or t. The non-polar alkane–benzene or alkane–phenyl (a, b), alkane–cyclohexane (a, c), and alkane–CCl_4 (a, t) interactions are represented by the dispersive parameters [17, 18, 21] listed in Table 3.

Carbonates + n-*alkanes*

These systems are characterized by a single type of contact (a, d). With the G^E and H^E data available, the fitting of the parameters was straightforward. We noted that:

(1) the quasichemical coefficients $C^{QUAC}_{ad,l}$ decrease with increasing chain length of the alkyl groups;

(2) the dispersive coefficients $C^{DIS}_{ad,l}$ remain constant (Table 4).

Compared to the previous values [10], $C^{QUAC}_{ad,1}$ of dimethyl carbonate is 3.15 (instead of 2.90) and that of methyl ethyl carbonate is 3.06 (instead of 2.85). All the other parameters change by less than 5%.

TABLE 3

Dispersive interchange coefficients $C^{DIS}_{as,l}$ for contacts (a, s), s = b, c, or t: type a, CH_3 or CH_2 in *n*-alkane, carbonate, methylcyclohexane or toluene; type b, C_6H_6 (benzene) or C_6H_5 (phenyl group in toluene); type c, *c*-CH_2 in cyclohexane; type t, CCl_4 (tetrachloromethane) (the quasichemical coefficients are equal to zero)

$C^{DIS}_{ab,1}$	$C^{DIS}_{ab,2}$	$C^{DIS}_{ac,1}$	$C^{DIS}_{ac,2}$	$C^{DIS}_{at,1}$	$C^{DIS}_{at,2}$
0.260 [a]	0.562 [a]	0.030 [b]	0.117 [b]	0.093 [b]	0.180 [b]
0.332 [c]	0.570 [c]				

[a] Values for benzene group [18]. [b] Ref. 17. [c] Values for phenyl group [21].

TABLE 4

Interchange coefficients, dispersive $C^{DIS}_{ed,l}$ and quasichemical $C^{QUAC}_{ed,l}$ for contacts (e, d), e = a or c: type a, CH_3 or CH_2 in *n*-alkane, carbonate, methylcyclohexane or toluene; type c, *c*-CH_2 in cyclohexane; type d, CO_3 in CH_3–$(CH_2)_{u-1}$–CO_3–$(CH_2)_{v-1}$–CH_3

u	v	$C^{DIS}_{ad,1}$	$C^{DIS}_{ad,2}$	$C^{DIS}_{cd,1}$	$C^{DIS}_{cd,2}$	$C^{QUAC}_{cd,1}$	$C^{QUAC}_{cd,2}$
1	1	1.20	3.00	1.50	3.25	3.15	4.10
1	2	1.20	3.00	1.50	3.25	3.06 [a]	3.98 [a]
2	2	1.20	3.00	1.50	3.25	2.80	3.70
2	>2	1.20	3.00	1.50	3.25	2.75 [a]	3.60 [a]
>2	>2	1.20	3.00	1.50	3.25	2.70 [a]	3.50 [a]

[a] Estimated value.

Carbonates + cyclohexane

These mixtures are characterized by three types of contact: (a, c), (a, d) and (c, d). The (a, c) and (a, d) parameters are well-known, so it is easy to obtain the (c, d) parameters. The important point is that the rules found for carbonates are similar to the rules for other polar compounds:

(1) the quasichemical coefficients $C^{QUAC}_{cd,l} = C^{QUAC}_{ad,l}$, i.e. they are independent of the class of alkane;

(2) the dispersive coefficients $C^{DIS}_{cd,l}$ remain constant, but are larger than $C^{DIS}_{ad,l}$ (Table 4).

Carbonates + benzene, +toluene, or +tetrachloromethane

These mixtures are characterized by three types of contact: (a, s), (a, d) and (s, d), with s = b or t. The adjustment of the (s, d) parameters was developed as before, assuming that these contacts are now purely dispersive

TABLE 5

Dispersive interchange coefficients $C^{DIS}_{sd,l}$ for contacts (s, d): s = b, C_6H_6 or C_6H_5; s = t, CCl_4: d, CO_3 in CH_3 –$(CH_2)_{u-1}$–CO_3–$(CH_2)_{v-1}$–CH_3 (the quasichemical coefficients are equal to zero)

u	v	$C^{DIS}_{bd,1}$	$C^{DIS}_{bd,2}$	$C^{DIS}_{td,1}$	$C^{DIS}_{td,2}$
1	1	2.44 2.37 [a,b]	3.37 3.30 [a]	3.03	4.10
2	2	1.86 1.72 [a,b]	2.80 2.61 [a]	2.55	3.61

[a] Values for phenyl group. [b] Estimated value.

[11], because only in this case is there a regular change of the interchange coefficients from one system to another (Table 5). Again, all the parameters change by less than 5% of the previous ones.

RESULTS AND DISCUSSION

Tables 6 and 7 list for comparison the calculated and experimental values for G^E and H^E (at equimolar composition and temperature T) of systems containing a linear carbonate and an organic solvent. The standard deviations $\sigma(Q)$ ($Q = y_1$ or H^E) defined similarly to eqn. (2), are also listed. For a better comparison, we have also included the $\sigma(Q)$ values obtained from the direct Redlich–Kister fit of the same experimental data.

For di-n-alkyl carbonate + n-alkanes or +cyclohexane systems, the DISQUAC model describes fairly well the G^E and H^E values (Tables 6, 7; Figs. 1, 2). Large discrepancies are encountered for the G^E of diethyl carbonate(1) + n-dodecane(2) mixture, probably due to experimental inaccuracies related to the extremely flat VLE curve of y_1 vs. x_1 [14]. However, in general, the model yields a rather good representation of the experimental VLE data for these systems. In particular, DISQUAC gives a good prediction for the coordinates of the azeotropes (Table 8 and Fig. 3).

TABLE 6

Molar excess Gibbs energies G^E at equimolar composition and at temperature T, and standard deviation $\sigma(y_1)$ of di-n-alkyl carbonate(1) + solvent(2): comparison of direct experimental results (exp) with values calculated (calc) using the coefficients $C_{jk,l}^{DIS}$ and $C_{jk,l}^{QUAC}$ from Tables 3–5

Mixture	G^E (J mol^{-1})		$\sigma(y_1)$		Ref.
	Calc	Exp	Calc	Exp	
Dimethyl carbonate [a] +					
CH_3–$(CH_2)_4$–CH_3	1161	1175	0.0075	0.0053	12, 13
CH_3–$(CH_2)_6$–CH_3	1223	1189	0.0063	0.0015	12, 13
CH_3–$(CH_2)_8$–CH_3	1244	1250	0.0025	0.0008	12, 13
c-C_6H_{12}	1256	1231	0.0163	0.0061	12, 15
C_6H_6	275	274	0.0044	0.0022	12, 15
CCl_4	505	506	0.0053	0.0036	12, 15
Diethyl carbonate [b] +					
CH_3–$(CH_2)_4$–CH_3	741	727	0.0048	0.0030	14
CH_3–$(CH_2)_6$–CH_3	819	821	0.0081	0.0098	14
CH_3–$(CH_2)_{10}$–CH_3	860	967	0.0023	0.0005	14
c-C_6H_{12}	834	831	0.0142	0.0026	15
C_6H_6	−97	−99	0.0057	0.0041	15
CCl_4	51	52	0.0054	0.0014	15

[a] $T = 298.15$ K. [b] $T = 298.65$ K.

TABLE 7

Molar excess enthalpies H^E at equimolar composition and at 298.15 K, and standard deviation $\sigma(H^E)$[a] of di-n-alkyl carbonate(1) + solvent(2): comparison of direct experimental results (exp) with values calculated (calc) using the coefficients $C_{jk,l}^{DIS}$ and $C_{jk,l}^{QUAC}$ from Tables 3–5

Mixture	H^E (J mol^{-1})		$\sigma(H^E)$ (J mol^{-1})		Ref.
	Calc	Exp	Calc	Exp	
Dimethyl carbonate+					
CH_3–$(CH_2)_4$–CH_3	1901	1902	57	6.7	7, 8
CH_3–$(CH_2)_5$–CH_3	2031	1988	39	8.4	7, 8
CH_3–$(CH_2)_6$–CH_3	2147	2053	58	8.5	7, 8
CH_3–$(CH_2)_8$–CH_3	2346	2205	100	8.6	7, 8
c-C_6H_{12}	1926	1947	63	7.6	7, 8
c-C_6H_{11}–CH_3	1819	1862	79	5.8	7, 8
C_6H_6	394	401	22	2.1	7, 8
C_6H_5–CH_3	511	512	4	2.2	7, 8
CCl_4	528	527	32	2.6	7, 8
Diethyl carbonate+					
CH_3–$(CH_2)_4$–CH_3	1248	1264	16	5.8	6
CH_3–$(CH_2)_5$–CH_3	1344	1328	15	6.3	6
CH_3–$(CH_2)_6$–CH_3	1431	1399	21	4.3	6
CH_3–$(CH_2)_8$–CH_3	1582	1536	29	4.4	6
CH_3–$(CH_2)_{12}$–CH_3	1816	1798	39	7.7	6
c-C_6H_{12}	1355	1320	26	6.6	6
c-C_6H_{11}–CH_3	1188	1184	25	2.6	6
C_6H_6	7	7	21	0.4	6
C_6H_5–CH_3	−14	−10	4	0.5	6
CCl_4	−61	−66	21	1.0	6

[a] $\sigma(H^E) = [\sum (H^E_{calc} - H^E_{exp})^2/N]^{1/2}$; N = number of experimental points.

The usual behavior of H^E for systems close to the critical solution point [23–25] is found in the case of dimethyl carbonate(1) + n-decane(2) mixture, with experimental values smaller than the calculated ones (Table 7 and Fig. 2).

Table 9 shows a comparison between calculated and predicted coordinates of critical solution points for dimethyl carbonate(1) + n-alkane(2) mixtures. In comparison with the results obtained from our previous interaction parameters, the present critical temperatures are less accurate by about 10 K. The critical compositions are predicted to better than 0.007 in mole fraction (previous result 0.012); but (as usual) the calculated LLE curves are not as flat as the experimental ones [9, 25] (Fig. 4).

A comparison between calculated and experimental values for the solid–liquid equilibrium (SLE) of the dimethyl carbonate(1) + n-hexadecane(2) mixture is also shown in Fig. 4. The theoretical liquidus

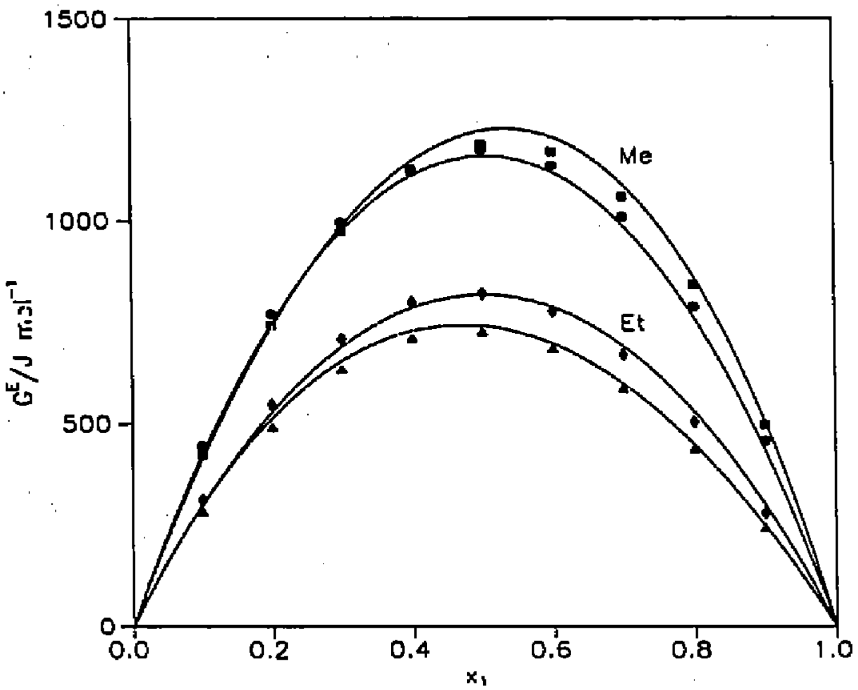

Fig. 1. Comparison of theory with experiment for the molar excess Gibbs energy G^E at 298.15 K or 298.65 K of dimethyl (Me) or diethyl (Et) carbonate + n-alkane. Lines show predicted values. Experimental results: dimethyl carbonate + n-hexane (●), +n-octane (■) [12, 13]; diethyl carbonate + n-hexane (▲), +n-octane (◆) [14].

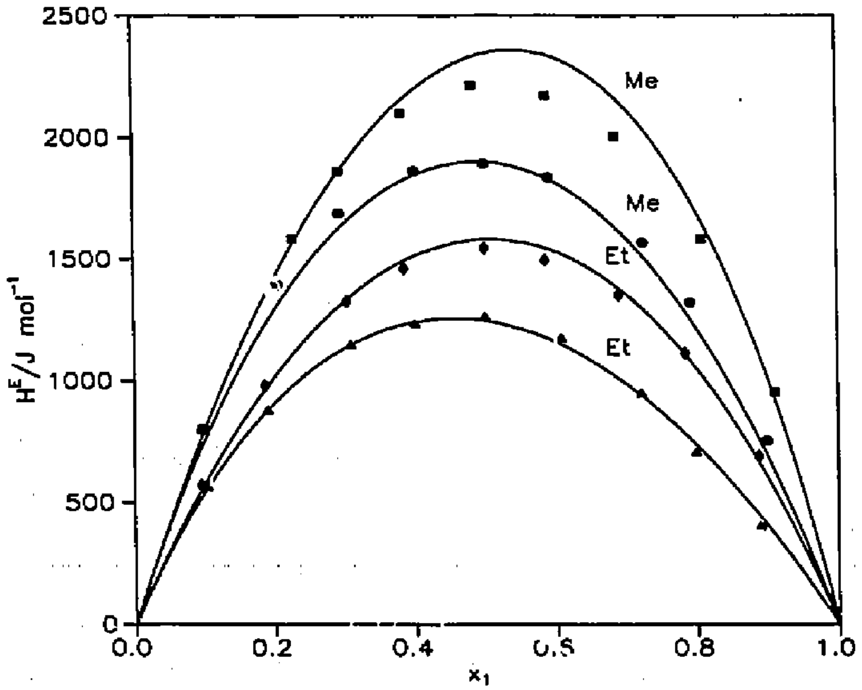

Fig. 2. Comparison of theory with experiment for the molar excess enthalpy H^E at 298.15 K of dimethyl (Me) or diethyl (Et) carbonate + n-alkane. Lines show predicted values. Experimental results: dimethyl carbonate + n-hexane (●), +n-decane (■) [7, 8]; diethyl carbonate + n-hexane (▲), +n-decane (◆) [6].

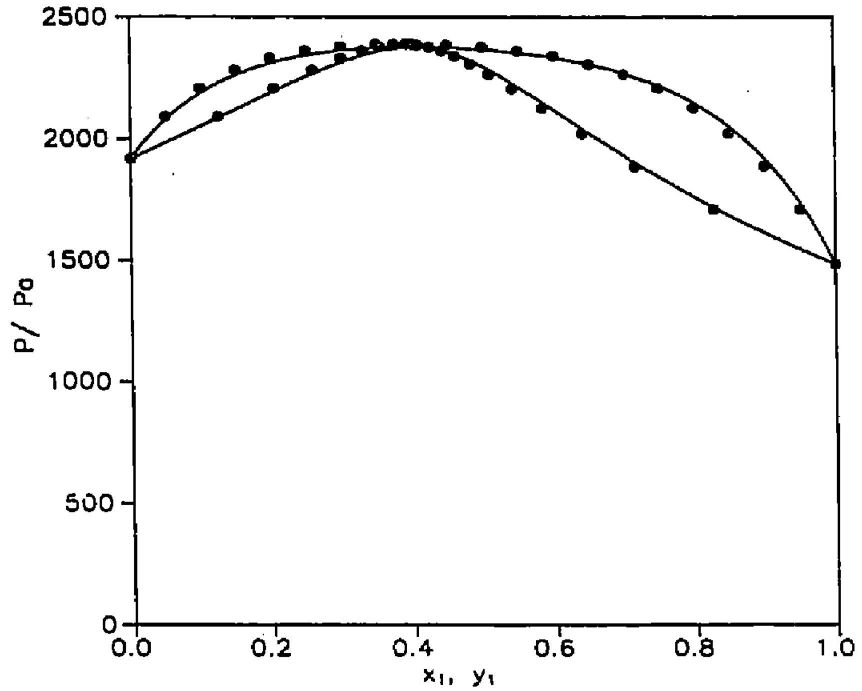

Fig. 3. Comparison of theory with experiment for the isothermal vapor–liquid phase equilibrium diagram of diethyl carbonate(1) + *n*-octane(2) at 298.65 K: total pressure, *P* vs. x_1 or y_1 (the mole fractions of diethyl carbonate in the liquid and vapor phases, respectively). Full lines show predicted values. Experimental results [14]: (●).

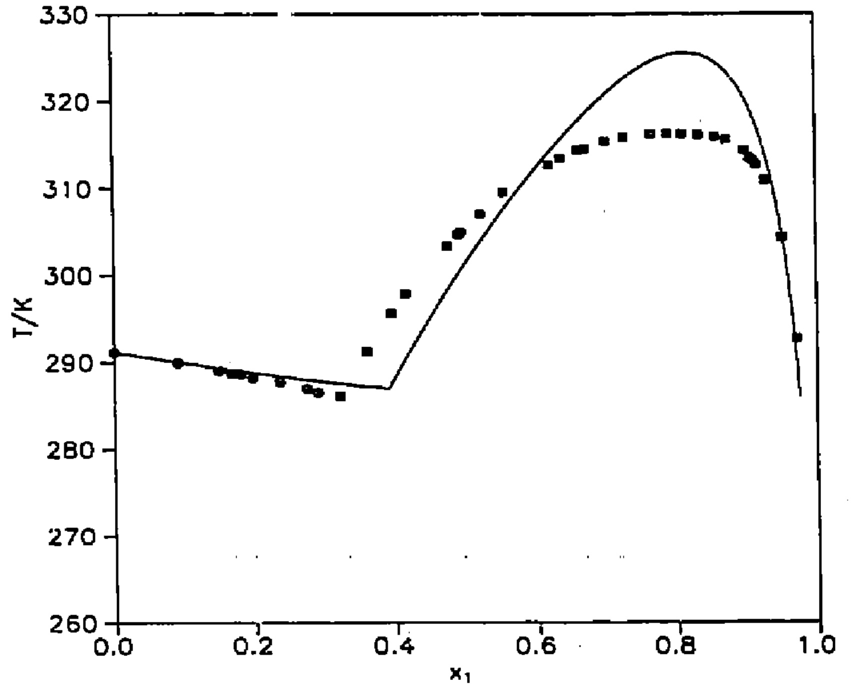

Fig. 4. Comparison of theory with experiment for the liquid–liquid and solid–liquid equilibrium temperatures *T* vs. the mole fraction x_1 of dimethyl carbonate in dimethyl carbonate(1) + *n*-hexadecane(2) mixture. The full line shows predicted values. Experimental results [4, 5]: (●), SLE; (■), LLE.

TABLE 8

Comparison of experimental (exp) coordinates of azeotropes: pressure P^{az}, temperature T^{az} and mole fraction x_1^{az} for di-*n*-alkyl carbonate(1) + solvent(2) mixtures with values calculated (calc) using the coefficients $C_{jk,l}^{DIS}$ and $C_{jk,l}^{QUAC}$ from Tables 3–5

Mixture	T^{az} (K)	x_1^{az}		P^{az} (10^3 Pa)		Ref.
		Calc	Exp	Calc	Exp	
Dimethyl carbonate+						
CH_3–$(CH_2)_4$–CH_3	298.15	0.2207	0.2188	22.249	22.473	12, 13
CH_3–$(CH_2)_6$–CH_3	298.15	0.8334	0.8421	7.807	7.759	12, 13
c-C_6H_{12}	298.15	0.3353	0.328	17.016	16.83	12, 15
CCl_4	298.15	0.0580	0.0937	15.281	15.446	12, 15
	348.9	0.1430	0.189	106.59	101.3	22
Diethyl carbonate+						
CH_3–$(CH_2)_6$–CH_3	298.65	0.4008	0.388	2.375	2.388	14

T–x_2 curve was calculated according to the equation

$$-\ln x_2 = [1/T - 1/T^*_{m2}]\,\Delta H^*_{m2}/R + \ln \gamma_2 \qquad (3)$$

where $T^*_{m2} = 291.15$ K and $\Delta H^*_{m2} = 53.36$ kJ mol^{-1} are the experimental temperature [4, 5] and the molar enthalpy of fusion [26] of pure *n*-hexadecane, respectively. T is the mixture temperature and γ_2 is the activity coefficient. Solid–solid transitions and contributions from terms related to differences in heat capacities between liquid and solid phases were neglected. It is noteworthy that a similar region of demixing is obtained using LLE or SLE conditions.

The low pressure VLE and related excess functions of solutions of di-*n*-alkyl carbonate as solute in a non-polar but 'active' solvent (benzene, toluene, or CCl_4) are also fairly well represented by DISQUAC (Tables 6–8;

TABLE 9

Upper critical solution temperatures T^c and compositions x_1^c of dimethyl carbonate(1) + *n*-alkane(2) mixtures: comparison of experimental results (exp) [4, 5] with values calculated (calc) using the coefficients $C_{nd,l}^{DIS}$ and $C_{nd,l}^{QUAC}$ from Table 4

m [a]	T^c (K)		x_1^c	
	Calc	Exp	Calc	Exp
10	304.0	286.6	0.692	0.686
12	312.6	297.8	0.745	0.738
14	319.7	307.6	0.785	0.783
16	325.6	316.2	0.816	0.814

[a] The number of carbon atoms in the *n*-alkane.

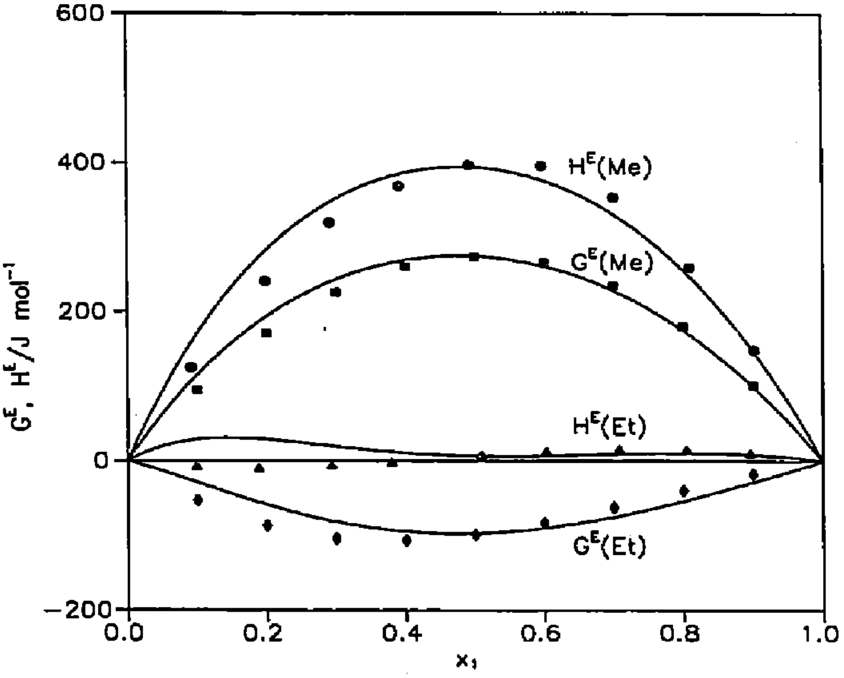

Fig. 5. Comparison of theory with experiment for the molar excess Gibbs energy G^E and enthalpy H^E at 298.15 K or 298.65 K of dimethyl (Me) or diethyl (Et) carbonate + benzene. Full lines show predicted values. Points show experimental results [6–8, 12, 15].

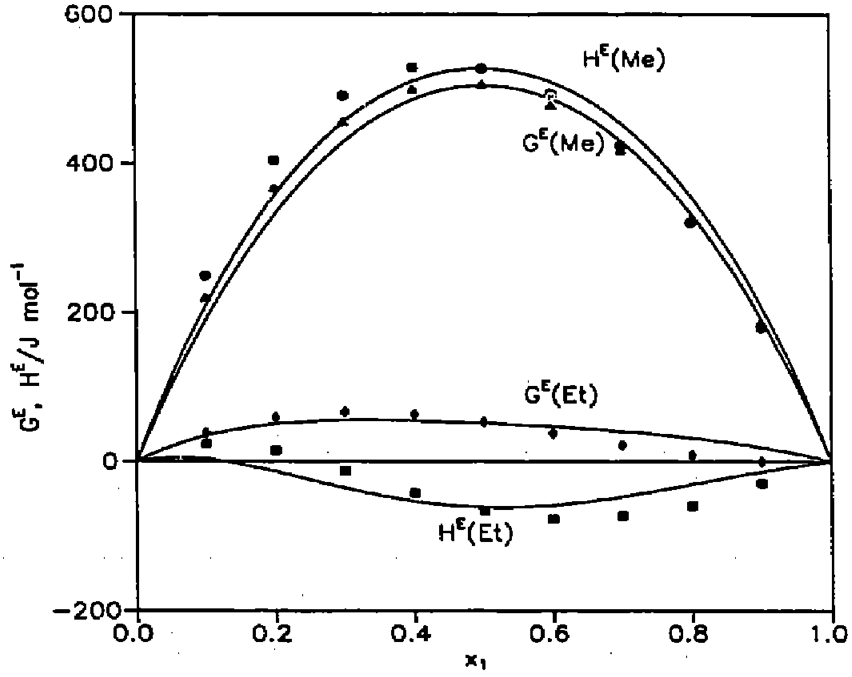

Fig. 6. Comparison of theory with experiment for the molar excess Gibbs energy G^E and enthalpy H^E at 298.15 K or 298.65 K of dimethyl (Me) or diethyl (Et) carbonate + tetrachloromethane. Full lines show predicted values. Points show experimental results [6–8, 12, 15].

Figs. 5 and 6). The model yields a reasonably good description of the rather complex shape of the G^{E} and H^{E} curves, which are less positive than in *n*-alkanes, and may even be S-shaped or negative for systems containing diethyl carbonate. This may be due to exothermic contributions related to charge transfer or dipole/induced-dipole interactions between the polar group of the solute and the polarizable solvent molecules [11, 27].

The parameters listed in Tables 4 and 5 show that the *n*-alkyl groups attached to –O–CO–O– exert a relatively weak steric effect on the $C_{\mathrm{ed},l}^{\mathrm{QUAC}}$ (e = a, c) and $C_{\mathrm{sd},l}^{\mathrm{DIS}}$ (s = b, t) coefficients [10, 11]. This behavior is also encountered for ether + *n*-alkane, or +cyclohexane mixtures [23] and in systems containing C_6H_6 or CCl_4 and iodoalkanes [28], bromoalkanes [29], or chloroalkanes [30, 31].

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