VAPOR-LIQUID EQUILIBRIA OF METHYL OR ETHYL ACETATE WITH 1-CHLOROPENTANE OR 1-CHLOROHEXANE AT 101.32 kPa PRESSURE

JUAN ORTEGA* AND JUAN A. PEÑA

Laboratorio de Termodinámica y Fisicoquímica, Escuela Superior de Ingenieros Industriales, 35071-University of Las Palmas de G.C, Canary Islands, Spain

The *T-x-y* data for binary mixtures composed of methyl acetate + (1-chloropentane or 1-chlorohexane) and ethyl acetate + (1-chloropentane or 1-chlorohexane) have been measured ebulliometrically at 101.32 kPa using a dynamic method. In thermodynamic calculations using these data, the vapor phase was considered to be non-ideal, and the four systems studied exhibit positive deviations from Raoult's law. The activity coefficients and vapor phase compositions have been compared with those predicted by using UNIFAC and ASOG models.

Introduction

This paper reports experimental isobaric vapor-liquid equilibrium (VLE) data for four binary mixtures at 101.32 kPa. The four mixtures employed were: methyl acetate (MA) + 1-chloropentane (1Cl5), methyl acetate + 1-chlorohexane (1Cl6), ethyl acetate (EA) + 1-chloropentane, and ethyl acetate + 1-chlorohexane. The literature contains no values for these mixtures. The study of systems containing chloroalkanes is of great interest in the field of thermodynamics of mixtures, even though not many VLE data have been published for mixtures containing such components that would thereby make it possible to validate existing theoretical models. Moreover, there are very few reliable data on the physico-chemical properties of the above-mentioned halogenated hydrocarbons. A partial explanation for this situation is that the reactivity of such components increases as working temperature increases, making handling difficult. The literature consulted makes reference to similar systems, but we have only been able to find isothermal VLE values for ethyl acetate + 1-chlorobutane at various temperatures, reported by Khurma et al.[6]

We also investigated the usefulness of the ASOG and UNIFAC models in calculating equilibrium values for the mixtures considered.

1. Experimental Section

All the chemicals used in this study were purchased from Fluka. Table 1 lists the chemicals and their stated purities. However, before use they were degassed by ultrasound and stored in darkness on a molecular sieve (ref. 69828 from Fluka) for several days prior to use.

The most important physical properties of the ace-

tates and 1-chloroalkanes, NBP, ρ, and n_D, were determined experimentally, and the values so obtained did not differ significantly from the literature values (Table 1).

Isobaric vapor-liquid equilibria for binary systems containing these compounds were measured using a recirculating still made by us. The description and operation of this still were described previously by Ortega et al.[10] However, when working with 1-chloroalkanes, the system was modified slightly by hermetically sealing in glass the upper section of the still, which was subjected to high temperature. Temperature measurements were made by sensors enclosed in glass tubes welded to the outside of the still.

Pressure was regulated electronically, using a Fisher VKH100 controller, and the total pressure in the experimental equipment was measured using a MKS Instrument System with reading intervals of ±0.001 kPa. The accuracy of the reading was better than ±0.02 kPa. Temperature measurements were taken using a Comark-6800 digital thermometer with platinum probes and reading intervals of ±0.05 K. Calibration of this equipment recorded measurement errors of ±0.1 % in the readings.

Vapor and liquid samples were analyzed by densimetry, using an Anton Paar 60/602 vibrating-tube densimeter calibrated with water and n-nonane as described by Ortega et al.[9] employing standard curves for density-concentration obtained beforehand for binary mixtures (x_1 methyl acetate or x_1 ethyl acetate + x_2 1-chloropentane or x_2 1-chlorohexane) at (298.15 ±0.01) K. The polynomial equations used to estimate the vapor and liquid phase concentrations were:

$$\rho = 876.94 + 14.32\,x_1 + 26.75\,x_1^2 - 8.45\,x_1^3 + 17.47\,x_1^4 \quad (1)$$
$$(MA + 1Cl5)$$

Table 1. Experimental physical characteristics of chemicals of and constants used in the treatment of VLE data and their comparison with data found in the literature

	purity %	NBP [K]	ρ [kg·m⁻³] 298.15 K	n_D 298.15 K	T_c [K]	p_c [MPa]	v_c [m³ kmol⁻¹]	$\mu/10^{-30}$ [cm]	ω	z_{RA}
MA	>99.95	330.05 330.0187 (15) 330.076 (1)	927.07 927.9 (15) 927.3 (21)	1.3590 1.3589 (15)	506.9 (16) 506.55 (15)	4.69 (16) 4.75 (15)	0.228 (16) 0.228 (15)	5.74 (15)	0.326 (14)	0.256 (13)
EA	>99.5	350.30 350.261 (15) 350.213 (1)	894.34 894.55 (15) 894.0 (21)	1.3701 1.3698 (15)	524.1 (16) 523.3 (15)	3.85 (16) 3.88 (15)	0.286 (16) 0.286 (15)	6.07 (15)	0.362 (14)	0.254 (13)
Cl5	>99	380.91 380.54 (15)	876.92 877.00 (15) 877.1 (21)	1.4099 1.4100 (15)	559.36[a]	3.35[a]	0.364[a]	6.47 (2)	0.316[b]	0.259[c]
Cl6	>99	408.05 408.07 (1)	873.33 873.9 (21)	1.4174	590.04[a]	2.98[a]	0.419[a]	6.54[a]	0.360[b]	0.256[c]

[a] values estimated using ref. (2)
[b] values estimated using ref. (15)
[c] values estimated using ref. (22)

Table 2. Experimental vapor pressures of pure compounds as a function of temperature

T [K]	p_i^o [kPa]	T [K]	p_i^o [kPa]
	Ethyl acetate		
334.1	59.3	352.0	109.4
336.8	65.3	352.5	111.3
338.6	69.7	353.1	113.5
340.2	73.7	354.0	116.9
342.1	78.6	354.5	118.8
343.5	82.5	355.3	121.9
345.1	87.1	356.3	125.9
345.8	89.3	356.9	128.3
347.6	94.7	357.7	131.6
348.6	98.1	358.5	134.9
349.5	100.8	359.1	137.4
349.9	102.3	-	-
	1-Chloropentane		
350.0	37.6	380.8	100.7
354.0	43.2	382.7	106.4
359.5	52.0	385.2	114.1
364.2	60.5	387.5	121.8
368.4	69.2	390.1	130.8
373.0	79.8	392.4	139.2
378.1	93.1	393.8	144.6
	1-Chlorohexane		
373.8	36.3	405.2	93.3
379.9	44.2	408.1	101.0
384.7	51.4	411.2	109.6
389.0	58.7	414.3	118.8
392.7	65.5	416.1	124.3
395.6	71.3	419.3	134.7
399.3	79.2	421.7	143.0
402.0	85.5	-	-

Table 3. Antoine constants obtained for pure compounds used in this work, standard deviations, $s(p_i^o)$, and from the literature

Compound	ref.	A	B	C	$s(p_i^o)$ [kPa]
Methyl acetate	(10)	6.4934	1329.46	33.52	0.04
	(15)	6.2441	1183.70	50.74	
Ethyl acetate	this work	7.1649	1856.18	10.21	0.06
	(15)	7.06309	1224.673	57.44	
1-Chloropentane	this work	6.0408	1294.54	60.15	0.07
	(15)	6.81151	1271.16	58.15	
1-Chlorohexane	this work	5.8292	1259.51	79.78	0.06
	(1)	7.05136	1461.72	57.58	

$$\rho = 873.29 + 12.69\, x_1 + 34.18\, x_1^2 - 25.70\, x_1^3 + 32.55\, x_1^4 \quad (2)$$

$$(MA + 1Cl6)$$

$$\rho = 876.90 + 1.50\, x_1 + 11.20\, x_1^2 + 4.73\, x_1^3 \quad (3)$$

$$(EA + 1Cl5)$$

$$\rho = 873.36 + 4.47\, x_1 + 4.23\, x_1^2 + 12.22\, x_1^3 \quad (4)$$

$$(EA + 1Cl6)$$

which yielded correlation coefficients very close to unity. In addition, the uniform distribution of v^E values

for each of the mixtures was also verified, these results being compared with those determined by us and published previously[8, 11)], with average errors for all mixtures of less than 1 %. The concentration values for the liquid phase obtained using Eqs. (1)-(4) were accurate to ±0.0025; however, the uncertainty for those of the vapor phase was slightly higher, including the high volatility of the methyl acetate in the least favorable cases.

2. Results and Discussion

2.1 Vapor pressures

The correlations employed for the vapor pressures exert a considerable influence on the thermodynamic treatment of VLE data. The Antoine equation is the best known, and the literature contains values for the components used in this experiment, though values for the 1-chloroalkanes are not abundant. Therefore, experimental (T, p_i^o) values were determined over a temperature range similar to that for the working conditions employed in this study using the same equilibrium still. **Table 2** sets out the vapor pressure measurement values for ethyl acetate, 1-chloropentane, and 1-chlorohexane. The vapor

Table 4. Experimental Vapor-Liquid Equilibrium Data at (101.32 ±0.02) kPa for the mixtures
$x_1 H_3CCOOC_uH_{2u+1}$ ($u = 1, 2$) + x_2 1 – ClC_vH_{2v+1}
($v = 5, 6$)

T [K]	x_1	y_1	γ_1	γ_2
		Methyl acetate + 1-Chloropentane		
376.65	0.0207	0.1131	1.449	1.019
373.10	0.0447	0.2195	1.418	1.020
370.35	0.0649	0.2891	1.377	1.029
366.70	0.1011	0.3802	1.274	1.042
364.70	0.1208	0.4291	1.267	1.044
361.75	0.1579	0.5009	1.223	1.045
358.65	0.2026	0.5697	1.178	1.051
356.75	0.2285	0.6034	1.165	1.066
354.00	0.2673	0.6532	1.164	1.076
353.00	0.2805	0.6682	1.167	1.084
351.05	0.3156	0.7036	1.154	1.089
346.95	0.3891	0.7701	1.153	1.093
345.65	0.4183	0.7899	1.144	1.099
342.65	0.4917	0.8319	1.122	1.122
340.95	0.5424	0.8538	1.100	1.155
339.05	0.6032	0.8784	1.079	1.190
336.35	0.7001	0.9102	1.049	1.289
335.05	0.7550	0.9274	1.033	1.343
334.15	0.7854	0.9388	1.035	1.383
332.15	0.8835	0.9659	1.010	1.488
331.15	0.9446	0.9805	0.991	1.863
		Methyl acetate + 1-Chlorohexane		
395.65	0.0407	0.3067	1.306	1.008
390.65	0.0609	0.7085	1.292	1.012
387.40	0.0756	0.4677	1.279	1.018
382.30	0.1015	0.5547	1.267	1.022
377.60	0.1272	0.6244	1.270	1.028
373.50	0.1504	0.6807	1.283	1.027
369.70	0.1794	0.7245	1.267	1.041
366.70	0.2022	0.7579	1.266	1.043
364.50	0.2245	0.7802	1.245	1.052
360.60	0.2613	0.8185	1.244	1.049
355.85	0.3224	0.8590	1.204	1.059
353.05	0.3551	0.8789	1.210	1.064
350.20	0.3971	0.8978	1.199	1.074
347.85	0.4427	0.9128	1.171	1.089
345.80	0.4925	0.9244	1.132	1.127
342.55	0.5795	0.9421	1.081	1.193
340.00	0.6390	0.9545	1.075	1.217
339.70	0.6597	0.9561	1.052	1.262
337.85	0.7088	0.9650	1.048	1.275
336.90	0.7487	0.9699	1.027	1.324
335.70	0.7873	0.9749	1.020	1.376
335.15	0.8099	0.9771	1.011	1.440
333.75	0.8631	0.9832	0.999	1.563
333.00	0.8887	0.9863	0.997	1.624
332.60	0.9147	0.9876	0.983	1.951
331.95	0.9254	0.9905	0.991	1.975
		Ethyl acetate + 1-Chloropentane		
376.85	0.0289	0.1249	2.023	1.010
375.95	0.0505	0.1714	1.626	1.003
375.55	0.0576	0.1891	1.589	1.000
374.85	0.0701	0.2173	1.530	0.998
374.35	0.0797	0.2357	1.477	0.999
373.95	0.0867	0.2505	1.458	0.999
373.65	0.0963	0.2651	1.401	0.999
372.55	0.1084	0.2944	1.423	1.004
371.65	0.1218	0.3168	1.396	1.013
370.25	0.1546	0.3792	1.367	0.997
369.45	0.1792	0.4158	1.321	0.990
367.85	0.2175	0.4683	1.281	0.991
366.35	0.2497	0.5087	1.264	1.001
364.95	0.2897	0.5526	1.230	1.005
362.85	0.3634	0.6191	1.166	1.019
361.15	0.4388	0.6697	1.097	1.058
360.05	0.4728	0.6949	1.091	1.078
359.05	0.5143	0.7253	1.078	1.088
358.25	0.5534	0.7507	1.062	1.102
357.25	0.5958	0.7800	1.056	1.110
356.65	0.6263	0.7963	1.044	1.134
355.75	0.6674	0.8235	1.041	1.138
355.15	0.7054	0.8451	1.029	1.150
354.35	0.7454	0.8670	1.023	1.174
353.75	0.7843	0.8878	1.014	1.193
352.95	0.8292	0.9118	1.010	1.217
352.25	0.8726	0.9329	1.003	1.271
351.45	0.9167	0.9544	1.002	1.358
350.85	0.9538	0.9737	1.001	1.442
350.55	0.9787	0.9868	0.998	1.586
		Ethyl acetate + 1-Chlorohexane		
403.05	0.0221	0.1409	1.626	1.003
399.35	0.0418	0.2298	1.516	1.013
396.15	0.0579	0.2955	1.510	1.030
393.65	0.0739	0.3493	1.478	1.038
392.75	0.0803	0.3657	1.454	1.046
391.05	0.0910	0.4026	1.468	1.047
390.45	0.1026	0.4147	1.360	1.057
388.85	0.1180	0.4494	1.330	1.061
386.75	0.1358	0.4927	1.331	1.062
384.65	0.1554	0.5356	1.330	1.060
384.25	0.1707	0.5437	1.241	1.074
382.35	0.1939	0.5815	1.224	1.075
379.85	0.2277	0.6259	1.194	1.086
377.55	0.2683	0.6675	1.145	1.097
374.55	0.3149	0.7167	1.132	1.101
372.35	0.3658	0.7525	1.084	1.119
369.35	0.4282	0.7959	1.062	1.134
366.35	0.4853	0.8330	1.065	1.145
364.05	0.5398	0.8603	1.055	1.164
361.35	0.6061	0.8886	1.048	1.195
359.25	0.6647	0.9121	1.043	1.199
357.35	0.7336	0.9315	1.021	1.264
355.35	0.8054	0.9515	1.008	1.323
353.85	0.8601	0.9646	1.002	1.425
352.65	0.9043	0.9742	0.999	1.591
351.95	0.9369	0.9824	0.993	1.693
351.45	0.9666	0.9874	0.983	2.336

pressure values for methyl acetate were measured by the authors earlier and were reported in a recent paper[13]. The data in Table 2 were regressed using the Antoine equation with three constants:

$$\log p_i^o = A - B / (T - C) \tag{5}$$

The values for the three constants A, B, and C so obtained appear in **Table 3** together with the standard deviations, $s(p_i^o)$, for the experimental values and other literature values.

2.2 Vapor-liquid equilibrium data

The isobaric vapor-liquid equilibrium data p, T, x_1 and y_1 for the binary mixtures $\{x_1 CH_3COOC_uH_{2u+1}$ ($u = 1, 2$) + 1-Cl C_vH_{2v+1} ($v = 5, 6$)$\}$ obtained by direct experimentation at a pressure of (101.32 ±0.02) kPa, (760 ±0.15) Torr, are given in **Table 4**. To shed light on

the behavior of the above mixtures, the activity coefficient, γ_i, for both mixture components were calculated by applying the equation:

$$\phi_i \, p y_i = \gamma_i \, p_i^o \, x_i \, \phi_i^o \, \exp[v_i^L (p - p_i^o) / RT] \tag{6}$$

where the fugacity coefficients, ϕ_i and ϕ_i^o, were calculated using the second virial coefficients computed from Tsonopoulos empirical correlations[23]. The molar volumes of the liquid pure components, v_i^L, and changes in these values with temperature were calculated using a modified version of Rackett's equation [see Spencer and Danner[18]]. Rackett's parameters, z_{RA}, for the 1-chloroalkanes were estimated from the (ρ, T) values published[22]. As set out in Table 1, the critical properties and other properties for the 1-chloroalkanes were estimated

Table 5. Coefficients obtained for Eqs. (7) and (8) and standard deviations s (Q) for the different correlations

correlation	k	A_0	A_1	A_2	A_3	s
x_1 Methyl acetate + x_2 1-Chloropentane						
$(y_1 - x_1)$ vs. x_1	0.236	5.458	−9.875	9.495	−4.421	0.002
T vs. x_1	0.228	−189.0	402.3	−477.5	232.5	0.1
T vs. y_1	4.788	15.1	71.7	−89.8	−2.0	0.1
x_1 Methyl acetate + x_2 1-Chlorohexane						
$(y_1 - x_1)$ vs. x_1	0.170	8.721	−10.293	2.444	-	0.002
T vs. x_1	0.142	−279.7	214.7	−131.3	145.1	0.2
T vs. y_1	0.836	4.3	270.7	−502.9	392.9	0.2
x_1 Ethyl acetate + x_2 1-Chloropentane						
$(y_1 - x_1)$ vs. x_1	0.048	11.367	−35.644	48.046	−23.285	0.003
T vs. x_1	0.030	−833.7	2807.5	−3471.3	1481.0	0.1
T vs. y_1	0.672	−4.9	36.1	−34.2	-	0.1
x_1 Ethyl acetate + x_2 1-Chlorohexane						
$(y_1 - x_1)$ vs. x_1	0.292	6.243	−12.920	12.448	−5.042	0.004
T vs. x_1	0.058	−92.0	−459.3	787.4	−272.9	0.3
T vs. y_1	0.178	−139.1	811.7	−1302.9	683.1	0.1

by a variety of methods, always trying to ensure that the empiricalexpressionsincludedatleastonecharacteristic parameter value for the components determined by the authorsthemselves,e.g.,molarrefractionortheParachor index, as in the case of the expressions recommended by Meissner [see Bretsznajder[2]].

Some consistency tests were applied to reduce the equilibrium data. So, for each set of isobaric data the ln (γ_1/γ_2) values were plotted against x_1 and the values of the areas above and below the x_1-axis were calculated. In accordance with the method put forward by Herington[5], the value of D was compared with the empirical quantity, J. The value of $D-J$ was not less than 10 % for any of the four systems considered herein, although $D<J$ was verified in all cases, on account of the large differences in the boiling points of the pure components, which ranged between 30.6 K for the mixtures consisting of (ethyl acetate + 1-chloropentane) and 78 K for the mixture consisting of (methyl acetate + 1-chlorohexane). A modified version of the test of Van Ness *et al.*[24] was also applied, using a subroutine based on Tsonopoulos'[23] empirical expressions instead of the procedure of Hayden and O'Connell [see Fredenslund *et al.*[3]] that had yielded good results in earlier studies[12, 13]. Using this version of the point-to-point test, the four systems considered herein yield positive consistency values overall, that is, average deviations values for the vapor phase, y_1, of less than 0.01.
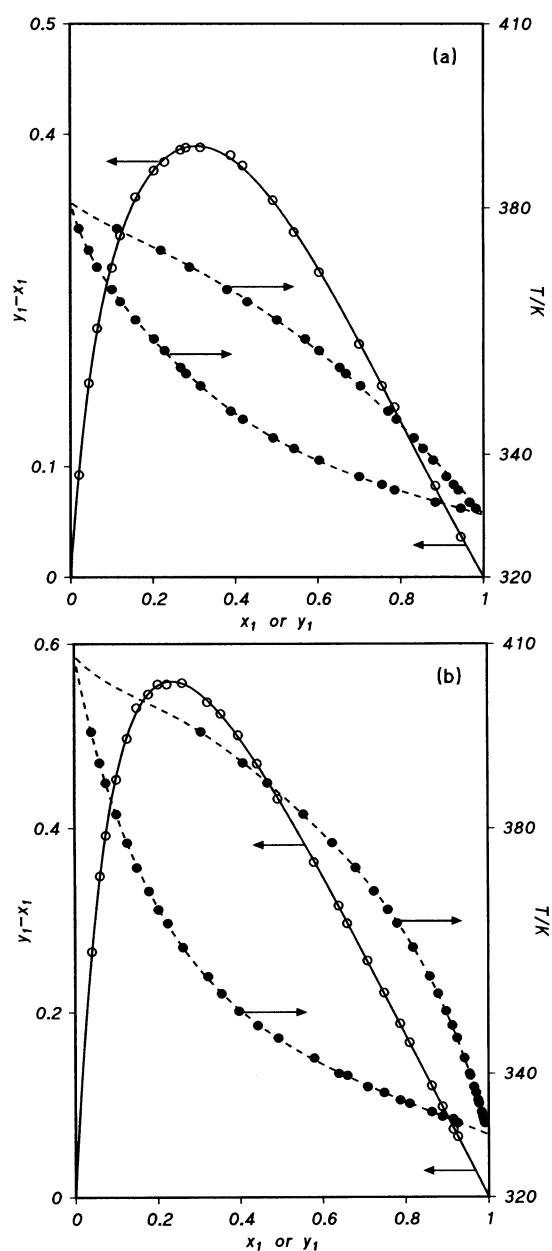
The data were correlated using the equation:

$$Q = x_1 x_2 \Sigma A_i [x_1 / (x_1 + kx_2)]^i \tag{7}$$

Thus, composition values were fitted by setting Q equal to y_1-x_1, whereas the temperature values were fitted using an equation similar to that for binary mixtures proposed by Tamir[19], which for the liquid phase took the form:

$$T = \Sigma x_i T_i^o + Q \tag{8}$$

where Q is identical to Q in Eq. (7), and x_i and T_i^o are,

respectively, the mole fraction and the boiling point for the pure component i at the working pressure. Application of Eq. (7) to the data was carried out using a method of least squares for a given value of k, and the degree of the polynomial was optimized using a statistical criterion (F-test). The minimum standard deviation, s (Q), for the data was calculated iteratively by applying this procedure for various values of k.

Table 5 shows the values for the coefficients of Eq. (7) and for s (Q) obtained by correlating the function (y_1-x_1) and T on the composition values. **Figure 1** depicts thecorrespondingplotsforthefourmixturesconsidered.

The activity coefficients, γ_i and the composition values for the vapor phase, y_i, at 101.32 kPa were predicted on the basis of the T vs. x_1 data for the (alkyl acetate+1-chloroalkane) systems using the UNIFAC and ASOG theoretical models. As applied, the UNIFAC model assumed that ester/chlorine interaction took the
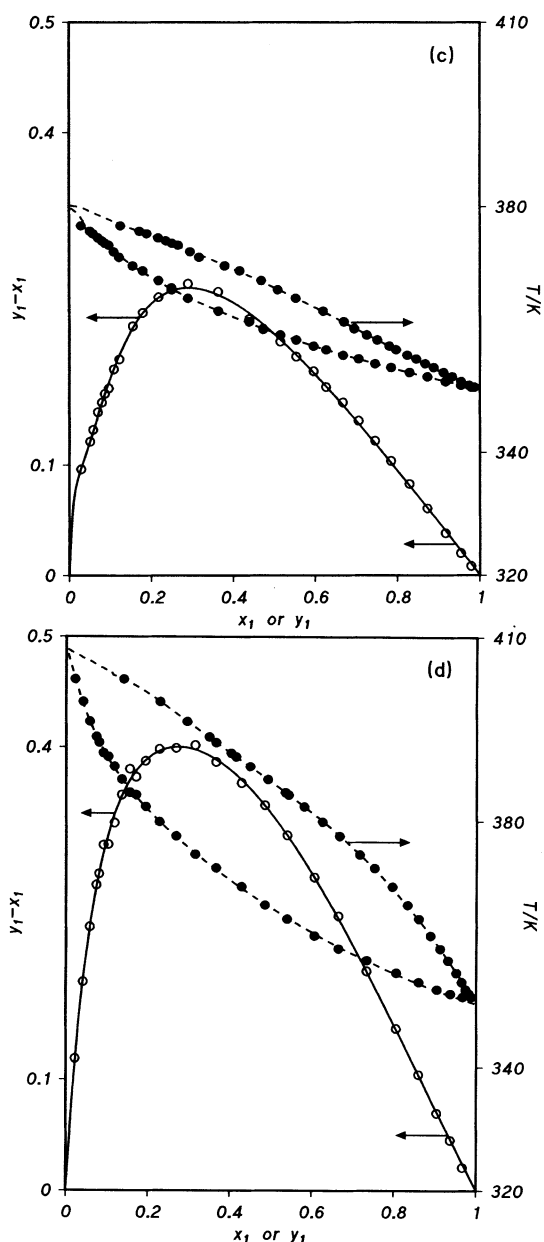
Fig. 1 Experimental vapor-liquid equilibria for the binary systems (x_1 alkyl acetate + x_2 1-chloroalkanes) at 101.32 kPa: (O), (y_1-x_1) vs x_1; (O), T vs x_1, y_1; (—) calculated by Eq. (7); (- - -) calculated by Eq. (8), (a), MA + 1Cl5; (b), MA + 1Cl6; (c), EA + 1Cl5; (d) EA + 1Cl6
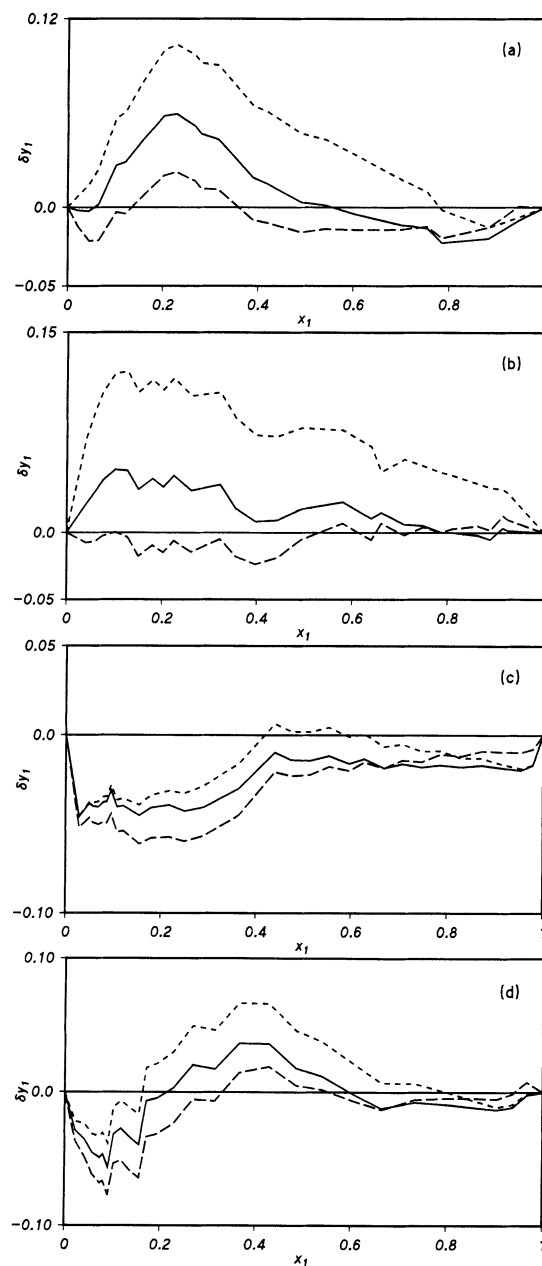
Fig. 2 Differences between the experimental values and those estimated by different theoretical models for the binaries (x_1 alkyl ethanoate + x_2 1-chloroalkanes). (—), UNIFAC[20]; (- - -), Modified UNIFAC[4]; (- - -), ASOG[21]. (a), (MA + 1Cl5); (b), (MA + 1Cl6); (c), (EA + 1Cl5) (d), (EA + 1Cl6).

form *COOC/CCl* and used the parameter values published by Tiegs *et al.*[20] The estimation of activity coefficients gave an average error smaller than 5 %. The other version of UNIFAC (Modified UNIFAC), by Gmehling *et al.*[4], was also applied to our mixtures and the estimations for γ_i values were very similar in all cases. The ASOG model considered that the *ester/chloride* interaction took the form *COO/Cl* and two sets of values for that interaction were used: those from original version of Kojima and Tochigi[6], and others published more recently[21]. Using these last values, predictions of γ_i were good with an average error smaller than 6 % for the four mixtures. However, using the parameters of Kojima and

Tochigi[7] the errors obtained were higher. In **Fig. 2** we have represented the differences obtained for the values of y_i, between the experimentals from Table 4 and those theoreticals calculated with the models above mentioned. In these cases, major discrepancies appear in the mixtures, (a) (MA + 1Cl5) 10 %, (b) (MA + 1Cl6) 11 % using the ASOG model.

Acknowledgments

Nomenclature

A, B, C	=	Antoine equation constants	
A_i	=	Coefficients in Eq. (7)	
k	=	Parameter in Eq. (7)	
NBP	=	Normal boiling point	[K]
n_D	=	Refractive index	
p	=	Total pressure	[kPa]
p_i^o	=	Vapor pressure of pure component i	[kPa]
s	=	Standard deviation	
T	=	Temperature	[K]
T_i^o	=	Normal boiling temperature of pure component i, NBP	[K]
R	=	Universal gas constant	$[\text{JK}^{-1}\,\text{kmol}^{-1}]$
v_i^L	=	Molar volume of pure component i	$[\text{m}^3\,\text{kmol}^{-1}]$
x	=	Liquid-phase molar fraction	[-]
y	=	Vapor-phase molar fraction	[-]
ρ	=	Density	$[\text{kgm}^{-3}]$
γ_i	=	Activity coefficient of component i	
ϕ_i	=	Fugacity coefficient of component i	
μ	=	Dipolar moment	[Cm]
ω	=	Acentric factor	

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