

Vapor–liquid equilibria for the quaternary reactive system ethyl acetate + ethanol + water + acetic acid and some of the constituent binary systems at 101.3 kPa

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Abstract

In this work, vapour–liquid equilibria for the quaternary reactive system ethyl acetate + ethanol + water + acetic acid and constituent binary systems ethyl acetate + ethanol, ethyl acetate + acetic acid and water + acetic acid has been determined at 101.3 kPa. UNIQUAC equation was used to correlate the experimental phase equilibrium data of the quaternary system. Experimental binary data were correlated using Wilson, NRTL and UNIQUAC equations.

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1. Introduction

In the last years, reactive distillation has become an usual alternative to reaction separation processes [1–3], especially for equilibrium-limited reactions, like esterification reactions. The synthesis of ethyl acetate via reactive distillation is an interesting project because ethyl acetate is a widely used solvent in industrial processes. Even though there are some investigations regarding this process [4–7], there are not recently published vapour–liquid equilibrium data of the quaternary reactive system in the same conditions of our work. Kang et al. [8] present VLE data for this system adding 2 wt.% of para-toluenesulfonic acid to the liquid phase to obtain chemical reaction equilibrium.

In this work, experimental data of isobaric vapor–liquid equilibria for the reactive quaternary system ethyl acetate + ethanol + water + acetic acid and for some of the non-reactive constituent binary systems (ethyl acetate + acetic acid, water + acetic acid, ethyl acetate + ethanol) were carried out at 101.3 kPa. The binary system ethanol + water was not

experimentally determined in this paper, since it is a widely studied mixture [9–12], and the ethyl acetate + water system is immiscible in the most range of compositions. The experimental results of the binary systems were correlated using Wilson [13], NRTL [14] and UNIQUAC [15] equations, and the experimental data of the quaternary system were correlated by the UNIQUAC equation. To predict the VLE data of the binary systems studied, methods based on contribution groups ASOG [16] and UNIFAC [17] have been used.

2. Experimental

2.1. Chemicals

Ethyl acetate and ethanol were supplied by Merck and degassed ultrasonically, dried over molecular sieves type 0.4 nm and kept in an inert argon atmosphere. Acetic acid was supplied by Sigma, and water was bidistilled and deionized. The purities of these components were more than 99.8% for ethyl acetate and ethanol and more than 99% for acetic acid. Density, viscosity and boiling point of pure components were measured and compared with literature data (Table 1). Pure components were also analyzed by gas chromatography.

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Table 1

Comparison of density, ρ , and viscosity, η , at 298.15 and the boiling points of the pure components with literature data

Component	t_B (K)		ρ (g cm ⁻³)		$10^{-3}\eta$ (Pa s)	
	Experimental	Literature ^a	Experimental	Literature	Experimental	Literature
Acetic acid	390.95	391.035	1.0436	1.0436 ^b	1.115	1.115 ^b
Ethyl acetate	350.27	350.261	0.8944	0.8945 ^c 0.8944 ^d	0.426	0.424 ^d 0.429 ^c
Water	373.15	373.150	0.9971	0.9971 ^e	0.890	0.890 ^e
Ethanol	351.43	351.443	0.7854	0.7854 ^f	1.082	1.082 ^g

^a From [18].^b From [19].^c From [20].^d From [21].^e From [22].^f From [23].^g From [24].

2.2. Apparatus and procedure

A glass Fischer Labodest VLE apparatus model 602/D was used in the equilibrium determinations. The equilibrium vessel is a dynamic recirculating still, and it is equipped with a Cottrell circulation pump. The equilibrium temperature was measured with a digital thermometer Yokogawa model 7563, with an accuracy of ± 0.01 K. A digital pressure controller Ruska model 7218 with an accuracy of ± 0.001 kPa was used for the pressure measurement. For binary systems, when temperature remains constant for 30 min or longer, the condition of equilibrium is assumed, and then liquid and vapor samples are taken for analysis.

Table 2

Vapor–liquid equilibrium data for ethyl acetate (1) + ethanol (2) system at 101.3 kPa

T (K)	x_1	y_1	γ_1	γ_2
350.77	0.0239	0.0602	2.471	0.988
350.08	0.0437	0.1024	2.349	0.989
349.52	0.0651	0.1367	2.143	0.994
348.76	0.1005	0.1936	2.014	0.994
347.53	0.1633	0.2778	1.851	1.004
347.16	0.1886	0.3066	1.790	1.008
346.74	0.2220	0.3389	1.704	1.019
346.07	0.2905	0.3899	1.531	1.059
345.70	0.3406	0.4296	1.457	1.081
345.61	0.3697	0.4407	1.381	1.113
345.51	0.3916	0.4586	1.361	1.120
345.41	0.4150	0.4729	1.329	1.139
345.35	0.4342	0.4844	1.304	1.154
345.27	0.4643	0.4981	1.257	1.191
345.22	0.4990	0.5178	1.218	1.225
345.19	0.5369	0.5410	1.184	1.263
345.21	0.5792	0.5657	1.147	1.314
345.29	0.6241	0.5930	1.113	1.374
345.43	0.6594	0.6165	1.090	1.421
345.61	0.7002	0.6438	1.066	1.488
345.89	0.7447	0.6762	1.043	1.571
346.33	0.7919	0.7164	1.025	1.658
346.99	0.8420	0.7699	1.014	1.726
347.91	0.8992	0.8357	1.001	1.863

The quaternary system studied is a reactive system, where the reactions of esterification of acetic acid and ethanol, and the corresponding hydrolysis take place. For the study of the quaternary VLE it is necessary to reach not only physical equilibrium, but chemical equilibrium too. For this reason, mixtures are kept in a bath during 1 or 2 days at 333.15 K to get close to the chemical equilibrium. Once in the Fischer Labodest, mixtures are kept for 4 or 5 h since the moment when temperature stills constant to ensure physical and chemical equilibrium.

Table 3

Vapor–liquid equilibrium data for ethyl acetate (1) + acetic acid (2) system at 101.3 kPa

T (K)	x_1	y_1	γ_1	γ_2
388.86	0.0238	0.0825	1.733	0.980
387.91	0.0366	0.1189	1.635	0.973
386.97	0.0476	0.1518	1.618	0.967
385.03	0.0776	0.2443	1.619	0.932
384.03	0.0907	0.2718	1.556	0.933
382.21	0.1170	0.3265	1.475	0.932
380.57	0.1410	0.3795	1.448	0.924
379.12	0.1657	0.4297	1.417	0.913
376.35	0.2203	0.5369	1.373	0.870
373.77	0.2661	0.6113	1.343	0.852
371.67	0.3085	0.6739	1.319	0.825
367.87	0.3924	0.7621	1.252	0.814
365.00	0.4617	0.8215	1.214	0.801
364.00	0.4924	0.8463	1.196	0.778
362.65	0.5332	0.8702	1.169	0.778
361.91	0.5559	0.8820	1.155	0.781
360.21	0.6068	0.9074	1.133	0.779
358.94	0.6568	0.9266	1.101	0.789
357.82	0.6986	0.9439	1.085	0.759
356.57	0.7426	0.9571	1.069	0.763
355.20	0.7974	0.9710	1.048	0.763
354.21	0.8415	0.9769	1.027	0.888
353.55	0.8644	0.9811	1.024	0.924
352.20	0.9197	0.9898	1.010	1.056
351.45	0.9482	0.9938	1.007	1.153
350.61	0.9809	0.9968	1.003	1.983

Table 4
Vapor–liquid equilibrium data for water (1)+acetic acid (2) system at 101.3 kPa

<i>T</i> (K)	<i>x</i> ₁	<i>y</i> ₁	<i>γ</i> ₁	<i>γ</i> ₂
386.54	0.0901	0.2253	2.274	0.938
384.47	0.1414	0.3177	2.087	0.925
382.96	0.1934	0.3954	1.920	0.915
381.86	0.2593	0.4654	1.680	0.923
380.39	0.3241	0.5311	1.554	0.944
379.83	0.3627	0.5622	1.470	0.965
378.65	0.4454	0.6371	1.362	0.989
378.35	0.4702	0.6593	1.336	0.993
377.84	0.5071	0.6844	1.291	1.026
377.47	0.5337	0.7047	1.267	1.043
377.18	0.5698	0.7271	1.222	1.080
376.70	0.6046	0.7536	1.200	1.105
376.28	0.6546	0.7961	1.171	1.106
375.98	0.6808	0.8200	1.163	1.092
375.68	0.7057	0.8394	1.153	1.094
375.45	0.7332	0.8566	1.133	1.119
375.00	0.7789	0.8819	1.103	1.196
374.62	0.8287	0.9064	1.067	1.341
374.45	0.8555	0.9183	1.048	1.464
374.11	0.8963	0.9419	1.029	1.629
373.99	0.9140	0.9515	1.020	1.736
373.84	0.9391	0.9651	1.008	1.942
373.66	0.9653	0.9809	0.999	2.120
373.31	0.9902	0.9951	0.999	2.279

Vapour and liquid phases composition, except for the water+acetic acid system, were determined by density. Binary samples were prepared by mass using a Mettler AX-205 Delta Range balance with a precision of $\pm 10^{-5}$ g and analyzed with an Anton Paar DSA-5000 digital vibrating tube densimeter, with a precision of $\pm 10^{-5}$ g cm⁻³. The water+acetic acid mixtures were analyzed by gas chromatography because the density-composition curve for this binary system presents a very small slope zone. Vapor and liquid phases obtained in the experimental determination of quaternary VLE were also measured by gas chromatography with a Hewlett-Packard 6890GC instrument equipped with series-connected thermal conductivity and flame ionization detectors to detect water and the other components

Table 5
Consistency test for the binary systems

System	ΔP (mm Hg)	Δy_1
Ethyl acetate + ethanol	0.002	0.006
Ethyl acetate + acetic acid	0.003	0.009
Water + acetic acid	0.007	0.008

in the same run. The column was HP-FFAP polyethylene glycol TPA 30 m × 530 mm and helium was used as carrier gas. Injector, detectors and oven temperature were at 473.15, 503.15 and 423.15 K, respectively. To know the composition of these quaternary mixtures it is necessary to have quaternary patrons of known composition, so 249 quaternary mixtures were prepared by mass and measured by gas chromatography. So the esterification and hydrolysis reactions are really slow at room temperature, analysing these mixtures immediately after preparation there is no change in their composition.

3. Results and discussion

The activity coefficients, γ_i , for quaternary and binary systems, were calculated from the following equation:

$$\ln \gamma_i = \ln \frac{y_i P}{x_i P_i^0} + \frac{(B_{ii} - V_i^L)(P - P_i^0)}{RT} + \frac{P}{2RT} \sum_{j=1}^n \sum_{k=1}^n y_i y_k (2n_{ji} - n_{jk}) \quad (1)$$

$$\delta_{ji} = 2B_{ji} - B_{jj} - B_{ii} \quad (2)$$

where T is the absolute temperature, P is the total pressure, V_i^L is the molar liquid volume of component i , P_i^0 is the saturation pressure, and B_{ii} and B_{ij} are the virial coefficients estimated by Hayden and O'Connell [25] method. In systems containing acetic acid, the equilibrium constant for dimerization has been taken into account.

Table 6
Correlation parameters and standard deviations for the binary systems at 101.3 kPa

Model	Parameters (J mol ⁻¹)						ΔT (K)	Δy_1
Ethyl acetate + ethanol								
Wilson	$\Delta\lambda_{12}$	351.10	$\Delta\lambda_{21}$	2424.90	α	0.78	0.277	0.0068
NRTL	Δg_{12}	1239.80	Δg_{21}	1792.70			0.277	0.0070
UNIQUAC	Δu_{12}	1471.19	Δu_{21}	-185.81			0.276	0.0068
Ethyl acetate + acetic acid								
Wilson	$\Delta\lambda_{12}$	-590.92	$\Delta\lambda_{21}$	1400.96	α	0.31	0.699	0.0389
NRTL	Δg_{12}	1072.06	Δg_{21}	-322.69			0.385	0.0413
UNIQUAC	Δu_{12}	852.93	Δu_{21}	-370.66			0.311	0.0399
Water + acetic acid								
Wilson	$\Delta\lambda_{12}$	1062.63	$\Delta\lambda_{21}$	-278.59	α	0.30	0.436	0.0352
NRTL	Δg_{12}	692.78	Δg_{21}	60.09			0.431	0.0339
UNIQUAC	Δu_{12}	-479.96	Δu_{21}	969.61			0.337	0.0369

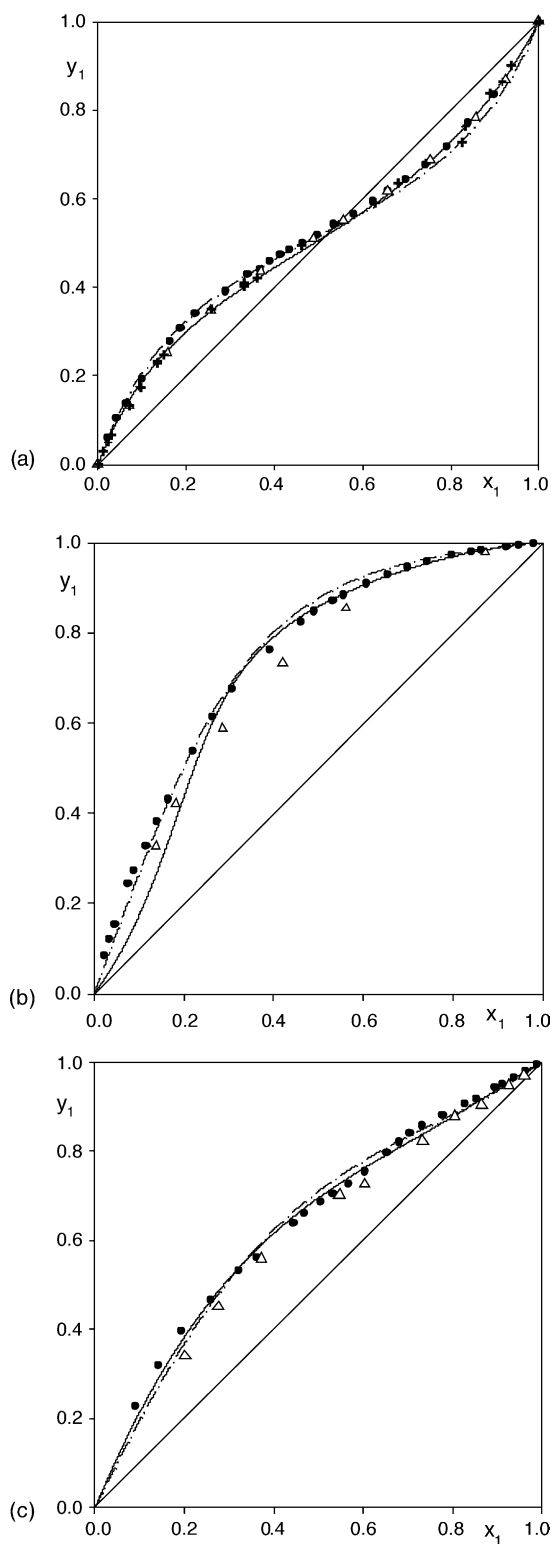


Fig. 1. Binary xy diagram of binary systems at 101.3 kPa; (a) ethyl acetate + ethanol, (●) this work, (Δ) from [28], (+) from [29]; (b) ethyl acetate + acetic acid, (●) this work, (Δ) from [30]; (c) water + acetic acid, (●) this work, (Δ) from [31]. (—) ASOG and (— · —) UNIFAC models.

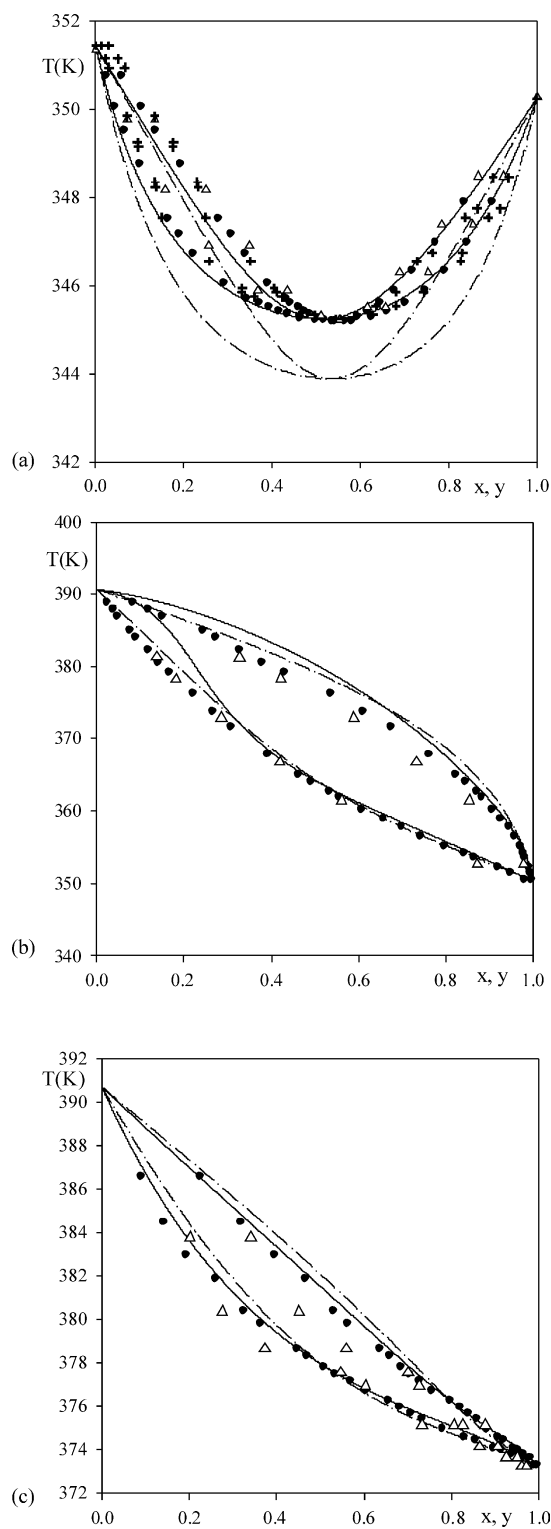


Fig. 2. Boiling temperature diagram of the binary systems at 101.3 kPa; (a) ethyl acetate + ethanol, (●) this work, (Δ) from [28], (+) from [29]; (b) ethyl acetate + acetic acid, (●) this work, (Δ) from [30]; (c) water + acetic acid, (●) this work, (Δ) from [31]. (—) ASOG and (— · —) UNIFAC models.

3.1. Non-reactive binary systems

Experimental isobaric VLE data of the binary systems ethyl acetate + ethanol, ethyl acetate + acetic acid, water + acetic acid are reported in Tables 2–4.

The experimental VLE data were found to be thermodynamically consistent by the point-to-point test of Van Ness et al. [26], modified by Fredenslund et al. [27], and results are shown in Table 5.

Experimental data were correlated using Wilson, NRTL and UNIQUAC equations, by minimizing the objective function:

$$\text{O.F.} = \sum_{j=1}^{np} \sum_{i=1}^{nc} \left[\frac{\gamma_{ij}^{\text{exp}} - \gamma_{ij}^{\text{calc}}}{\gamma_{ij}^{\text{exp}}} \right]^2 \quad (3)$$

Correlation parameters and the standard deviations are summarized in Table 6.

To predict the VLE of the binary systems, ASOG and UNIFAC methods have been used. Fig. 1 shows the xy diagrams and Fig. 2 shows the boiling temperature diagrams of the experimental, predicted and literature data [28–31] for the binary systems determined. Fig. 3 shows experimental and predicted activity coefficients. For the systems ethyl acetate + ethanol and water + acetic acid, ASOG method gives better results than UNIFAC. Table 7 presents the standard deviation of the boiling points and the vapor-phase mole fractions for the binary systems.

3.2. Quaternary system

Table 8 summarizes the experimental VLE isobaric data for the system ethyl acetate + ethanol + water + acetic acid.

To correlate the experimental phase equilibrium data of the quaternary system, the UNIQUAC equation was used. In this case, the objective function to minimize is defined by:

$$\text{O.F.} = \sum_{i=1}^{np} (\gamma_{ij}^{\text{exp}} - \gamma_{ij}^{\text{calc}}) \quad (4)$$

The correlation parameters and the standard deviations are shown in Table 9. UNIQUAC parameters calculated in

Table 7

Standard deviations of boiling points, σT , and vapor-phase mole fraction, σy_1 , resulting from the prediction of VLE using ASOG and UNIFAC methods

Method	σy_1	σT (K)
Ethyl acetate + ethanol		
ASOG	0.01263	0.1668
UNIFAC	0.01063	1.1775
Ethyl acetate + acetic acid		
ASOG	0.05463	2.6978
UNIFAC	0.01910	1.0592
Water + acetic acid		
ASOG	0.01462	0.3074
UNIFAC	0.02286	0.6201

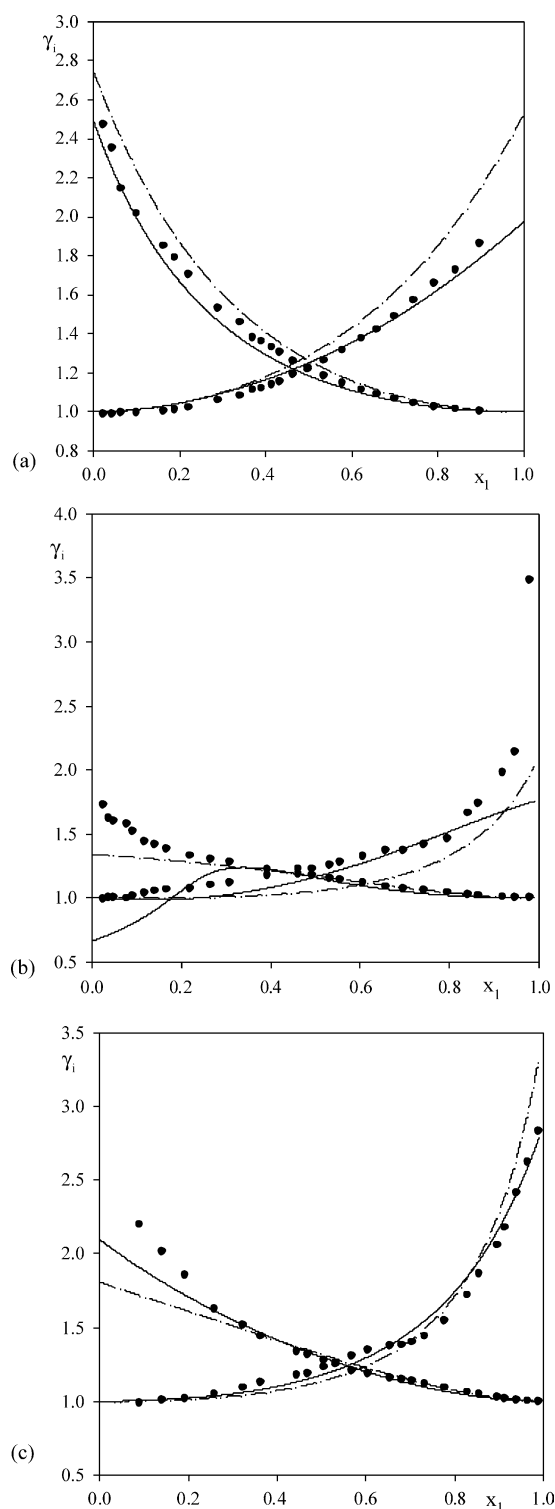


Fig. 3. Activity coefficients plots of the binary systems at 101.3 kPa; (●) this work, (—) ASOG and (---) UNIFAC models: (a) ethyl acetate + ethanol; (b) ethyl acetate + acetic acid; (c) water + acetic acid.

the quaternary system correlation, have been used to predict the VLE of the binary mixtures ethyl acetate + ethanol, ethanol + water, ethyl acetate + acetic acid, water + acetic acid and ethyl acetate + water, with the aim of checking the

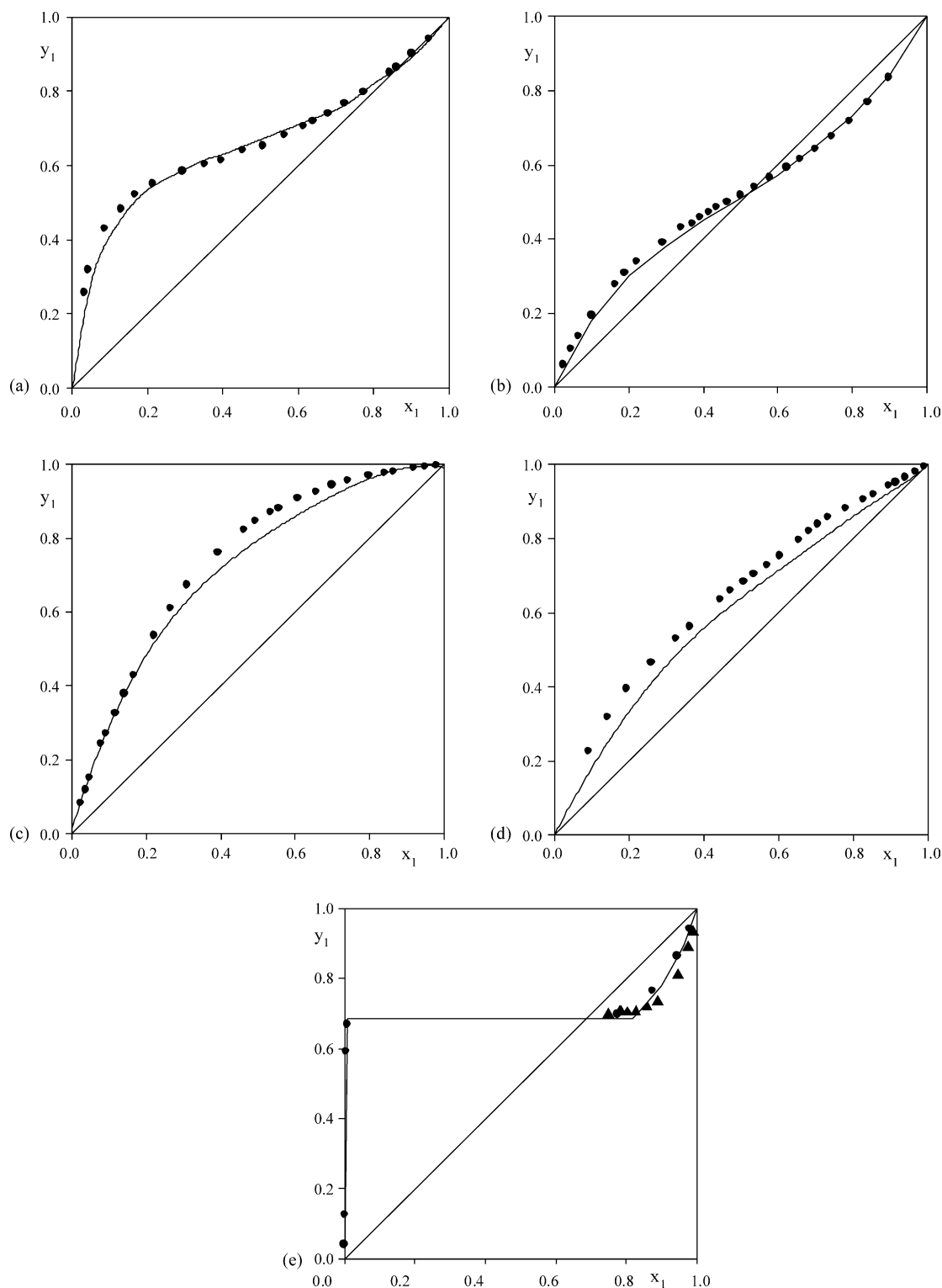


Fig. 4. Comparison between experimental and calculated data (—) using UNIQAC parameters from quaternary system correlation: (a) ethanol + water system, (●) from [9]; (b) ethyl acetate + ethanol system, (●) this work; (c) ethyl acetate + acetic acid, (●) this work; (d) water + acetic acid, (●) this work; (e) ethyl acetate + water, (▲) from [32], (●) from [33].

Table 8

Vapor–liquid equilibrium data for the quaternary system ethyl acetate (1) + ethanol (2) + water (3) + acetic acid (4) at 101.3 kPa

<i>T</i> (K)	<i>x</i> ₁	<i>x</i> ₂	<i>x</i> ₃	<i>y</i> ₁	<i>y</i> ₂	<i>y</i> ₃	<i>γ</i> ₁	<i>γ</i> ₂	<i>γ</i> ₃	<i>γ</i> ₄
352.11	0.2632	0.2036	0.3004	0.5274	0.2471	0.2179	1.8702	1.1343	2.2697	0.9888
352.38	0.3474	0.1837	0.2887	0.5590	0.2052	0.2278	1.6111	1.1582	2.5734	1.0104
355.17	0.2698	0.1354	0.2797	0.5908	0.1684	0.2249	1.8253	1.153	2.2899	0.9932
347.71	0.3588	0.1527	0.3698	0.6015	0.1701	0.2284	1.6938	1.1865	2.3955	0.9966
353.14	0.2649	0.3484	0.1579	0.4625	0.3763	0.1521	1.6211	1.0954	2.6916	1.0240
353.88	0.2228	0.1481	0.3362	0.5550	0.1953	0.2432	2.1306	1.1585	2.0439	0.9823
354.93	0.1554	0.1629	0.4387	0.4548	0.2200	0.3069	2.9457	1.2147	1.7087	0.9865
351.56	0.2831	0.2448	0.3139	0.4945	0.2545	0.2472	1.8238	1.1277	2.3321	0.9867
357.8	0.1190	0.1568	0.4611	0.4074	0.2555	0.3239	3.4342	1.2581	1.5929	1.0006
347.82	0.5143	0.1683	0.2327	0.6118	0.1708	0.2157	1.2685	1.2574	3.4035	1.0920
351.1	0.0858	0.4163	0.4142	0.5357	0.2964	0.1679	3.4175	1.1306	1.7148	0.9896
351.28	0.0912	0.4616	0.3533	0.4642	0.3575	0.1782	3.0098	1.0902	1.8426	0.9859
349.79	0.1818	0.3347	0.3929	0.3971	0.3516	0.2513	2.4823	1.1153	1.9350	0.9657
347.82	0.2639	0.1052	0.5088	0.4650	0.1310	0.4039	2.4398	1.2471	1.8333	0.9893
347.83	0.2804	0.2224	0.3860	0.5382	0.2173	0.2445	1.9736	1.1488	2.1557	0.9741
347.91	0.2797	0.3295	0.2888	0.4835	0.3086	0.2079	1.7796	1.1042	2.4190	0.9822
347.44	0.3605	0.1152	0.4004	0.6526	0.1210	0.2263	1.7453	1.2076	2.3128	0.9974
347.17	0.3665	0.1986	0.3292	0.5551	0.2058	0.2392	1.6076	1.1697	2.5399	1.0001
347.8	0.4083	0.4235	0.0898	0.4977	0.4026	0.0997	1.3103	1.1636	3.5627	1.0929
349.22	0.1985	0.5425	0.1771	0.3118	0.5255	0.1527	1.8663	1.0456	2.5376	1.0003
355.7	0.2026	0.2758	0.2049	0.4261	0.3283	0.2168	1.8769	1.0995	2.3100	0.9956
358.89	0.1668	0.1110	0.3749	0.4804	0.1792	0.3172	2.5743	1.2004	1.8052	0.9808
348.46	0.1787	0.5608	0.0923	0.4066	0.4965	0.0969	1.7895	1.0423	2.6719	1.0187
354.77	0.1446	0.4385	0.1638	0.3165	0.5033	0.1731	2.0037	1.0573	2.3104	1.0053
353.57	0.1244	0.4066	0.2902	0.3346	0.4456	0.2178	2.5743	1.2004	1.8052	0.9808
354.96	0.1514	0.2343	0.4681	0.4346	0.3018	0.2636	3.1898	1.2763	1.5954	1.0092
347.31	0.4510	0.1027	0.3317	0.5635	0.1137	0.2228	1.4456	1.2383	2.7883	1.0375
347.56	0.5047	0.2086	0.1968	0.5938	0.2115	0.1947	1.2549	1.2463	3.5062	1.0983
355.54	0.1366	0.1488	0.4646	0.4870	0.2257	0.2872	3.2833	1.2505	1.6226	0.9978
351.23	0.1430	0.4549	0.2554	0.3255	0.4730	0.2015	2.2661	1.0593	2.1434	0.9856
351.17	0.1449	0.3434	0.3655	0.3712	0.3818	0.2470	2.6315	1.1114	1.8909	0.9753
351.89	0.1206	0.2290	0.4590	0.4339	0.3068	0.2592	3.3674	1.2105	1.6337	0.9975
353.84	0.1192	0.1413	0.5528	0.4680	0.2393	0.2928	4.2287	1.3481	1.4715	1.0302
356.05	0.0683	0.0623	0.6553	0.4551	0.1503	0.3946	7.3726	1.6857	1.2471	1.1714

Table 9

UNIQUAC parameters (J mol^{-1}) and standard deviations resulting from the correlation of VLE data of quaternary system ethyl acetate (1) + ethanol (2) + water (3) + acetic acid (4)

	EtAc	EtOH	H ₂ O	HAc
EtAc	0	−18.05	388.05	148.28
EtOH	263.90	0	172.09	469.57
H ₂ O	668.50	111.40	0	460.57
HAc	63.07	−327.48	−290.26	0
ΔT (K)	Δy_1	Δy_2	Δy_3	Δy_4
3.63	0.051	0.030	0.041	0.019

quality of these parameters. Fig. 4 shows a comparison between experimental and calculated data using the UNIQUAC parameters. As it can be seen in this figure, there is a good agreement between predicted and experimental data.

4. Conclusions

In this work, experimental isobaric vapor–liquid equilibria data for the reactive quaternary system ethyl

acetate + ethanol + water + acetic acid have been determined, insuring we reach not only physical equilibrium, but chemical equilibrium too.

Non-reactive binary systems ethyl acetate + ethanol, ethyl acetate + acetic acid and water + acetic acid have been also determined at 101.3 kPa.

Wilson, NRTL and UNIQUAC equations have been used to correlate experimental binary data, and VLE of quaternary system has been correlated using UNIQUAC equation.

To predict the behaviour of the binary systems, group contribution methods ASOG and UNIFAC have been used.

List of symbols

B_{ii}	second virial coefficients for component <i>i</i>
B_{ij}	second virial coefficients for interactions between molecules <i>i</i> – <i>j</i>
P	total pressure
P_i^0	saturation pressure
T	temperature
V_i^L	molar liquid volume for the component <i>i</i>
x_i	liquid-phase mole fraction for component <i>i</i>
y_i	vapour-phase mole fraction for component <i>i</i>

Greek symbols

- δ_{ji}, δ_{jk} functions for virial coefficients
 γ_i liquid-phase activity coefficient for component i

Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at [doi:10.1016/j.fluid.2005.07.010](https://doi.org/10.1016/j.fluid.2005.07.010)

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