Isobaric Vapor−Liquid Equilibria for Binary and Ternary Mixtures of Ethanol, Methylcyclohexane, and *p*-Xylene

María del Carmen Sánchez-Russinyol,[†] Antonio Aucejo,[‡] and Sonia Loras*[,‡]

Departamento de Química e Ingeniería Química, Universidad de Matanzas, Autopista a Varadero Km. 3, Matanzas, Cuba, and Departamento de Ingeniería Química, Facultad de Química, Universitat de València, 46100 Burjassot, Valencia, Spain

Consistent vapor−liquid equilibria (VLE) were determined for the ternary system ethanol + methylcyclohexane + *p*-xylene and the three binary subsystems at 101.3 kPa at temperatures in the range from 345 to 408 K. The binary systems exhibit positive deviation from ideal behavior, and the system ethanol + methylcyclohexane presents a minimum-boiling-point azeotrope. The VLE data have been correlated by the Wilson, NRTL, and UNIQUAC equations. The ternary system does not present an azeotrope and is well predicted from binary interaction parameters.

Introduction

This work presents a continuation of our thermodynamic research on vapor−liquid equilibrium (VLE) data of mixtures formed by hydrocarbons and oxygenated additives (ethers and alkanols) to unleaded gasoline. Ethanol is of interest mostly because of the availability of its feedstock from renewable resources.

In this work, to improve our knowledge of the phase behavior of alkanes and aromatic hydrocarbons with ethanol mixtures, we measured isobaric vapor−liquid equilibrium (VLE) data at 101.3 kPa for the ternary system ethanol (1) + methylcyclohexane (2) + *p*-xylene (3) and the constituent binary systems ethanol (1) + methylcyclohexane (2), ethanol (1) + *p*-xylene (3), and methylcyclohexane (2) + *p*-xylene (3). The literature does not make reference to isobaric VLE data for the binary systems ethanol (1) + methylcyclohexane (2) and methylcyclohexane (2) + *p*-xylene (3); only isothermal data or azeotropic data have been reported.[1−6] For the other binary system, isobaric and isothermal VLE data have been reported in the literature.[7−12] References 7 and 12 report measurements at 101.3 kPa. For the ternary system, no VLE data have been previously published.

Experimental Section

Chemicals. Methylcyclohexane (99 mass %) and *p*-xylene (99+ mass %, HPLC grade) were supplied by Aldrich, and ethanol (99.5 vol %) was supplied by Prolabo. Ethanol was dried over 4-Å molecular sieves as soon as the bottles were opened. The reagents were used without further purification after chromatography failed to show any significant impurities. The densities of pure liquids were measured at 298.15 K using an Anton Paar DMA 55 densimeter. Temperature was controlled to ±0.01 K with a thermostated bath. The accuracies of density and refractive index measurements are $\pm 0.01\ \text{kg}\cdot\text{m}^{-3}$ and ±0.0002, respectively. The experimental values of these properties and the boiling points are given in Table 1, together with those given in the literature.

Apparatus and Procedure. An all-glass Fischer LABODEST vapor−liquid equilibrium apparatus model 602/D, manufactured by Fischer Labor und Verfahrenstechnik (Germany), was used in the equilibrium determinations. The equilibrium vessel was a dynamic recirculating still equipped with a Cottrell circulation pump. The still is capable of handling pressures from (0.25 to 400) kPa and temperature up to 523 K. The Cottrell pump ensures that both liquid and vapor phases are in intimate contact during boiling and also in contact with the temperature-sensing element. The equilibrium temperature was measured with a digital Hart Scientific thermometer model 1502A and a Pt 100 probe Hart Scientific model 5622 calibrated at the Spanish Instituto Nacional de Técnica Aeroespacial. The accuracy is estimated to be ±0.02 K. For the pressure measurement, a digital manometer with an accuracy of ±0.01 kPa was used. The temperature probe was calibrated against the ice and steam points of distilled water. The manometers were calibrated using the vapor pressure of ultrapure water.

In each VLE experiment, the pressure was fixed and held constant by using a vacuum pump, and the heating and shaking systems of the liquid mixture were turned on. The system was kept at the boiling point for at least 30 min to ensure that the steady state was reached. Then, samples of the liquid and condensate were taken for analysis. The sample extractions were carried out with special syringes that allowed the withdrawal of small-volume samples (0.1 μL). At least two analyses were made for each sample.

Analysis. The concentrations of the liquid and condensed phases were determined using a CE Instruments GC 8000 Top gas chromatograph (GC), after calibration with gravimetrically prepared standard solutions. A flame ionization detector was used together with a 30-m, 0.454-mm-i.d. capillary column (DB-MTBE, J & W Scientific). The GC response peaks were treated with Chrom-Card for Windows, version 1.21. Column, injector, and detector temperatures were (393, 473, and 498) K, respectively. Very good peak separation was achieved under these conditions, and calibration analyses were carried out to convert the peak area ratio to the mass composition of the sample. The standard deviation in the mole fraction was usually less than 0.001.

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* Corresponding author. E-mail: sonia.loras@uv.es. Fax: +34 963 544898.

[†] Universidad de Matanza.

[‡] Universitat de València.

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Table 1. Density *d*, Refractive Index n_D, and Normal Boiling Point T_b, of the Chemicals

	d(298.15 K)/kg·m^{-3}		n_D(298.15 K)		T_b(101.3 kPa)/K	
component	exptl	lit	exptl	lit	exptl	lit
ethanol	785.04	785.07[a]	1.3592	1.3663[a]	351.41	351.48[b]
methylcyclohexane	764.87	764.59[c]	1.4204	1.42060[c]	373.96	374.10[b]
p-xylene	856.55	856.70[c]	1.4930	1.49325[c]	411.23	411.50[b]

[a] Reference 24. [b] Reference 14. [c] Reference 25.

Table 2. Experimental Vapor−Liquid Equilibrium Data for Ethanol (1) + Methylcyclohexane (2) at 101.3 kPa

T/K	x_1	y_1	γ_1	γ_2
368.84	0.008	0.161	12.130	1.000
360.02	0.024	0.342	10.650	1.002
354.69	0.040	0.464	9.413	1.007
349.67	0.086	0.554	6.824	1.028
347.79	0.143	0.583	4.876	1.074
346.93	0.197	0.594	3.743	1.133
346.64	0.221	0.604	3.378	1.164
346.03	0.307	0.612	2.486	1.299
345.82	0.358	0.615	2.153	1.395
345.68	0.399	0.622	1.948	1.483
345.59	0.440	0.628	1.784	1.580
345.40	0.523	0.632	1.537	1.814
345.36	0.549	0.636	1.476	1.901
345.32	0.607	0.643	1.359	2.128
345.30	0.660	0.650	1.272	2.387
345.35	0.718	0.664	1.192	2.757
345.46	0.754	0.673	1.149	3.051
345.66	0.797	0.690	1.105	3.495
346.12	0.849	0.722	1.061	4.227
346.92	0.899	0.765	1.029	5.239
348.35	0.948	0.839	1.008	6.705
349.75	0.976	0.909	1.002	7.864

Table 3. Experimental Vapor−Liquid Equilibrium Data for Ethanol (1) + *p*-Xylene (3) at 101.3 kPa

T/K	x_1	y_1	γ_1	γ_3
404.93	0.008	0.189	4.291	0.972
391.42	0.031	0.479	4.067	0.937
372.38	0.082	0.734	4.253	0.922
366.03	0.123	0.795	3.808	0.926
361.22	0.196	0.835	2.973	0.965
359.31	0.255	0.847	2.483	1.036
358.12	0.313	0.857	2.137	1.097
357.30	0.349	0.863	1.989	1.144
356.39	0.397	0.875	1.833	1.167
355.72	0.464	0.881	1.619	1.282
355.34	0.488	0.883	1.565	1.338
354.74	0.553	0.887	1.419	1.514
354.28	0.600	0.893	1.339	1.631
353.89	0.650	0.898	1.261	1.805
353.45	0.706	0.902	1.186	2.099
353.09	0.750	0.906	1.137	2.402
352.70	0.802	0.915	1.089	2.785
352.32	0.850	0.927	1.057	3.208
351.99	0.896	0.940	1.029	3.856
351.63	0.949	0.962	1.008	5.061
351.48	0.979	0.982	1.003	5.869

Table 4. Experimental Vapor−Liquid Equilibrium Data for Methylcyclohexane (2) + *p*-Xylene (3) at 101.3 kPa

T/K	x_2	y_2	γ_2	γ_3
374.38	0.980	0.993	1.005	1.061
375.07	0.946	0.981	1.009	1.043
376.11	0.901	0.965	1.012	1.012
377.28	0.850	0.945	1.017	1.010
378.36	0.810	0.926	1.015	1.036
379.88	0.745	0.898	1.027	1.013
381.22	0.695	0.870	1.029	1.034
382.27	0.648	0.847	1.044	1.019
383.80	0.598	0.819	1.050	1.006
385.62	0.544	0.791	1.063	0.967
387.07	0.502	0.759	1.064	0.976
388.79	0.454	0.725	1.075	0.964
392.34	0.353	0.628	1.094	0.988
394.46	0.300	0.569	1.106	0.994
396.93	0.244	0.506	1.138	0.981
399.38	0.198	0.432	1.127	0.991
401.80	0.153	0.371	1.182	0.969
407.55	0.056	0.164	1.247	0.985

Table 5. Antoine Coefficients[a] of Equation 2

compound	A_i	B_i	C_i	T range/K
ethanol	18.9119	3803.98	−41.68	369−270
methylcyclohexane	15.7105	2926.04	−51.75	400−270
p-xylene	16.0963	3346.65	−57.84	440−300

[a] Reference 14.

Results and Discussion

Binary Systems. The temperature, T, and the liquid-phase and vapor-phase mole fractions, x_i and y_i, at 101.3 kPa are reported in Tables 2 to 4, together with the activity coefficients γ_i that were calculated from the equation[13]

$$\gamma_i = \frac{\gamma_i \mathrm{P}}{x_i P_i^{\mathrm{o}}} \tag{1}$$

for the system methylcyclohexane (2) + *p*-xylene (3). In eq 1, P is the total pressure, and P_i^{o} is the pure-component vapor pressure that was calculated with the Antoine equation

$$\ln(P_{\mathrm{i}}^{\mathrm{o}}/\mathrm{mmHg}) = A_i - \frac{B_i}{(T/\mathrm{K}) - C_i} \tag{2}$$

where the Antoine constants A_i, B_i, and C_i were taken from Reid et al.[14] and are reported in Table 5. When activity coefficients are calculated according to eq 1, the vapor phase is assumed to be an ideal gas, and the pressure dependence of the liquid-phase fugacity is neglected. Equation 1 was selected to calculate activity coefficients for this system because the low pressures observed in the present VLE data make these simplifications reasonable. In addition, in such almost ideal mixtures the activity coefficients become very sensitive to the vapor-phase corrections, and the estimation methods of vapor-phase corrections can introduce uncertainties into the calculated activity coefficients.[15] Activity coefficients for the systems ethanol (1) + methylcyclohexane (2) and ethanol (1) + *p*-xylene (3) were calculated according to[13]

$$\ln \gamma_i = \ln \frac{\gamma_i P}{x_i P_i^{0}} + \frac{(B_{ii} - V_i^{L})(P - P_i^{0})}{RT} + \frac{P}{2RT} \sum\sum y_i y_k (2\delta_{ji} - \delta_{jk}) \tag{3}$$

where V_i^L is the molar liquid volume of component i, B_{ii} and B_{jj} are the second virial coefficients of the pure gases,

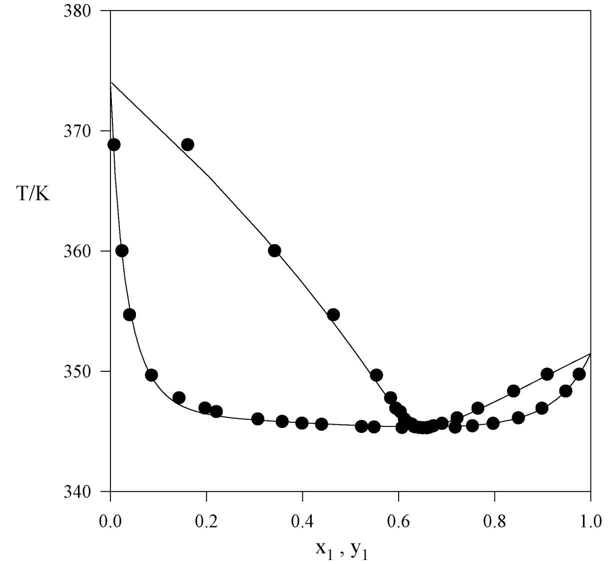

Figure 1. Experimental data for the system ethanol (1) + methylcyclohexane (2) at 101.3 kPa: ●, experimental data reported in this work; −, data smoothed with the NRTL model.

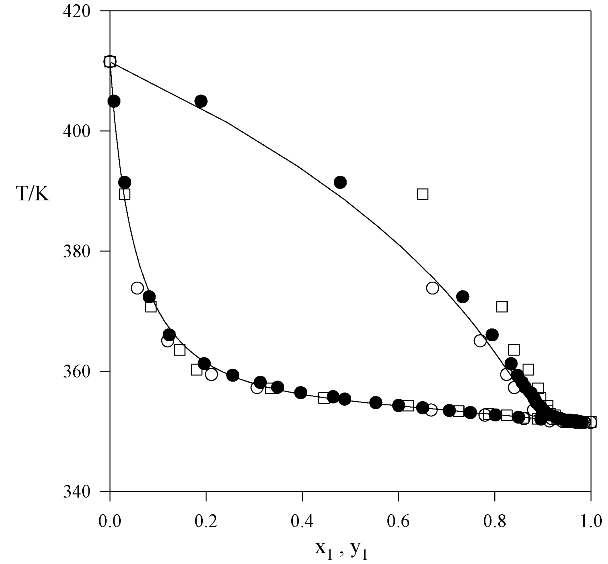

Figure 2. Experimental data for the system ethanol (1) + *p*-xylene (3) at 101.3 kPa: ●, experimental data reported in this work; ○, data of Galska-Krajewska;[7] □, data of Yakushina and Koshelkov;[12] (−), data smoothed with the NRTL model.

B_{ij} is the cross second virial coefficient, and

$$\delta_{ij} = 2B_{ij} - B_{jj} - B_{ii} \tag{4}$$

The standard state for the calculation of activity coefficients is the pure component at the pressure and temperature of the solution. Equation 3 is valid at low and moderate pressures when the virial equation of state truncated after the second coefficient is adequate to describe the vapor phase of the pure components and their mixtures and when liquid volumes of the pure components are incompressible over the pressure range under consideration. The molar virial coefficients B_{ii} and B_{jj} were estimated by the method of Hayden and O'Connell[16] using the molecular parameters suggested by Prausnitz et al.[17] Critical properties of all components were taken from DIPPR.[18]

Figures 1 to 3 show the $T-x-y$ diagrams at 101.3 kPa for each binary system. Figure 2 presents a graphical comparison between boiling-point temperatures measured for the system ethanol (1) + *p*-xylene (3) in this work and those found in the literature.[7,12] From this Figure, good agreement among the data of this work and those determined by Galska-Krajewska[7] is deduced. Nevertheless, the data determined by Yakushina and Koshelkov[12] do not agree very well with the other sets of data. These literature data are thermodynamically inconsistent by the point-to-point method,[19,20] as pointed out in DECHEMA.[21]

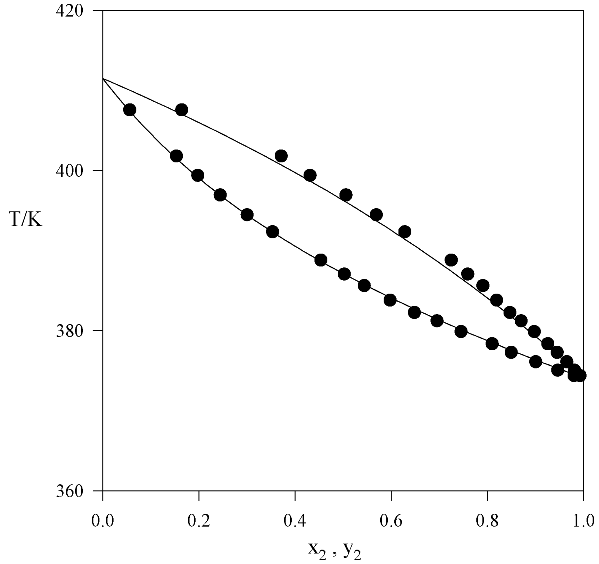

Figure 3. Experimental data for the system methylcyclohexane (2) + *p*-xylene (3) at 101.3 kPa: ●, experimental data reported in this work; −, data smoothed with the NRTL model.

Table 6. Consistency Test Statistics for the Binary Systems Determined in This Work

system	N_p[a]	100MAD(y)[b]	MAD(P)[c]/kPa
(1) + (2)	3	0.70	0.91
(1) + (3)	2	0.91	2.21
(2) + (3)	2	0.96	0.31

[a] Number of parameters for the Legendre polynomial used for consistency. [b] Mean absolute deviation in vapor-phase composition. [c] Mean absolute deviation in pressure.

According to the obtained results at 101.3 kPa, the system ethanol (1) + methylcyclohexane (2) deviates remarkably from ideal behavior, presenting a minimum-boiling azeotrope at $x_1 \cong 0.65$ and $T = 345.30$ K. The system ethanol (1) + *p*-xylene (3) shows important positive deviations from ideality but does not present an azeotrope, and the binary system methylcyclohexane (2) + *p*-xylene (3) is almost ideal, with very small positive deviations.

The VLE data reported in Tables 2 to 4 were found to be thermodynamically consistent by the point-to-point method of van Ness et al.[19] as modified by Fredenslund et al.[20] Pertinent consistency details and statistics are presented in Table 6.

The VLE data were also correlated with Wilson, NRTL, and UNIQUAC equations. The parameters of these models were obtained by minimizing the following objective function (OF)

$$\mathrm{OF} = \sum_{i=1}^{N} 100 \times \left(\left| \frac{P_i^{\mathrm{exptl}} - P_i^{\mathrm{calcd}}}{P_i^{\mathrm{exptl}}} \right| + \left| y_i^{\mathrm{exptl}} - y_i^{\mathrm{calcd}} \right| \right) \tag{5}$$

and are reported in Table 7, together with the pertinent statistics of VLE interpolation. An inspection of the results

Table 7. Parameters and Correlation Statistics for Different G^E Models

model	ij	A_{ij}/J·mol^{-1}	A_{ji}/J·mol^{-1}	α_{ij}	100MAD(y_i)	ΔP^a/%
Wilson[b]	(1) + (2)	8274.27	1516.58		0.61	0.61
	(1) + (3)	5517.71	1222.69		1.06	1.33
	(2) + (3)	−1047.94	2000.11		1.51	0.31
NRTL	(1) + (2)	3992.78	5674.68	0.47	0.52	0.65
	(1) + (3)	3437.80	3174.69	0.47	1.07	1.41
	(2) + (3)	−1059.35	1822.17	0.20	1.68	0.49
UNIQUAC[c]	(1) + (2)	−797.20	4898.65		1.52	2.06
	(1) + (3)	−528.09	3235.29		1.04	1.69
	(2) + (3)	−1008.92	1385.09		1.73	0.78

[a] Average percent deviation in pressure. [b] Liquid volumes have been estimated from the Rackett equation.[26] [c] Volume and surface parameters from ref 21.

Table 8. Experimental Vapor−Liquid Equilibrium Data for Ethanol (1) + Methylcyclohexane (2) + *p*-Xylene (3) at 101.3 kPa

T/K	x_1	x_2	y_1	y_2	γ_1	γ_2	γ_3
388.10	0.030	0.045	0.524	0.087	5.0978	1.3588	0.8076
375.71	0.048	0.210	0.533	0.248	4.7644	1.1396	0.8324
365.56	0.046	0.509	0.502	0.403	6.5924	1.0099	0.8437
355.85	0.043	0.899	0.470	0.519	9.3768	0.9797	1.0579
351.60	0.082	0.818	0.564	0.422	6.9027	1.0025	0.9198
353.00	0.090	0.694	0.585	0.384	6.1801	1.0300	0.8954
354.54	0.094	0.594	0.598	0.355	5.7051	1.0613	0.8878
356.00	0.099	0.504	0.613	0.325	5.2560	1.0956	0.8726
357.85	0.100	0.411	0.635	0.288	5.0315	1.1266	0.8232
360.29	0.091	0.308	0.664	0.244	5.2868	1.1856	0.7340
358.15	0.194	0.100	0.802	0.090	3.2279	1.4466	0.7995
355.64	0.195	0.197	0.758	0.156	3.3319	1.3707	0.8083
353.95	0.206	0.297	0.722	0.210	3.2026	1.2869	0.8308
352.45	0.198	0.405	0.687	0.260	3.3583	1.2222	0.8557
350.99	0.205	0.496	0.659	0.299	3.2925	1.1999	0.9500
348.60	0.215	0.638	0.632	0.348	3.3053	1.1702	1.0072
347.87	0.179	0.765	0.610	0.382	3.9458	1.0956	1.0866
347.43	0.299	0.601	0.639	0.346	2.5153	1.2833	1.1629
348.72	0.294	0.505	0.668	0.306	2.5401	1.2976	0.9555
353.55	0.286	0.201	0.768	0.164	2.4885	1.5079	0.8198
353.87	0.396	0.097	0.823	0.098	1.9011	1.8549	0.9560
351.83	0.403	0.198	0.768	0.171	1.8851	1.6847	1.0091
349.75	0.394	0.292	0.724	0.232	1.9708	1.6515	0.9986
348.53	0.405	0.399	0.685	0.286	1.9040	1.5460	1.1024
346.29	0.441	0.510	0.645	0.346	1.8004	1.5702	1.4899
346.97	0.500	0.402	0.665	0.318	1.5927	1.7927	1.3719
348.85	0.498	0.302	0.710	0.246	1.5840	1.7413	1.6216
350.86	0.499	0.183	0.761	0.181	1.5927	1.7927	1.3719
349.00	0.609	0.201	0.736	0.222	1.3341	2.3535	1.6232
346.25	0.611	0.339	0.665	0.322	1.3410	2.2039	2.1154
347.53	0.702	0.201	0.714	0.260	1.1898	2.8870	2.0812
351.45	0.707	0.057	0.835	0.098	1.1839	3.4096	1.9113
348.83	0.797	0.1	0.780	0.185	1.0864	3.9758	2.5208
349.81	0.898	0.053	0.831	0.143	0.9880	5.6391	3.8060

Table 9. Estimation of Experimental Ternary Data with Wilson, NRTL, and UNIQUAC Equations Using Binary Interaction Parameters from Table 7

model	100MAD(y_1)	100MAD(y_2)	ΔP/%
Wilson	2.01	1.04	1.94
NRTL	1.87	0.81	1.84
UNIQUAC	2.24	1.17	2.89

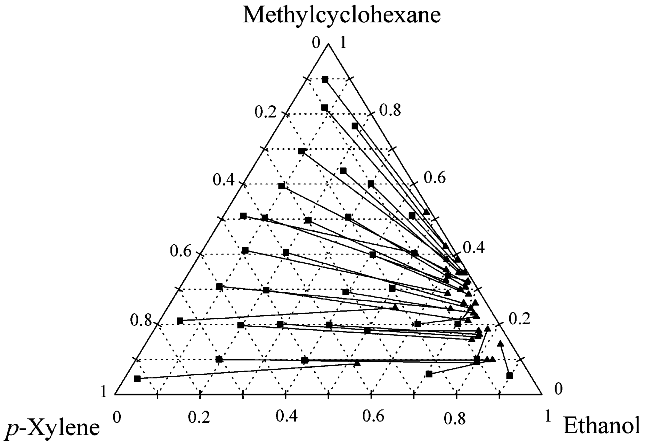

Figure 4. Diagram of VLE for the ternary system ethanol (1) + methylcyclohexane (2) + *p*-xylene (3) at 101.3 kPa: ■, liquid-phase mole fractions; ▲, vapor-phase mole fractions.

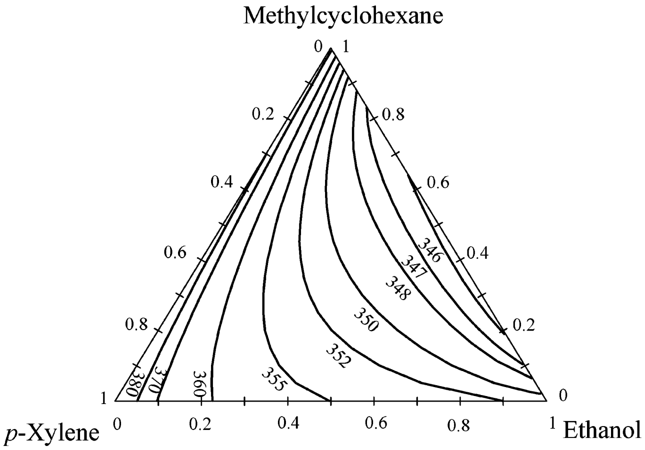

Figure 5. Boiling isotherms (K) for the ternary system ethanol (1) + methylcyclohexane (2) + *p*-xylene (3) at 101.3 kPa calculated with the NRTL model.

given in Table 7 shows that these models are adequate for the prediction of binary data.

Ternary System. The VLE data for the ternary system are shown in Table 8 and Figure 4. The activity coefficients γ_i were calculated from eq 3, and the molar virial coefficients were estimated for the binary system. The ternary data were found to be thermodynamically consistent by the McDermott−Ellis method[22] as modified by Wisniak and Tamir.[23]

The VLE data for the ternary systems have been estimated by using the Wilson, NRTL, and UNIQUAC models with the binary interaction parameters obtained from the regression of binary data. Table 9 lists the mean absolute deviations between experimental and calculated pressures and vapor-phase mole fractions of the components. The three models represent the data successfully. Thus, the models can be used to calculate boiling points from liquid-phase compositions at the system pressure. As an example, boiling isotherms calculated with the NRTL model are presented in Figure 5.

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