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Isobaric Vapor−Liquid Equilibria for the Binary System 3-Methylpentane + 2-Methyl-2-propanol and for the Ternary System Methyl 1,1-Dimethylethyl Ether + 3-Methylpentane + 2-Methyl-2-propanol at 101.3 kPa

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Isobaric vapor−liquid equilibria data were obtained for the 3-methylpentane + 2-methyl-2-propanol binary system and methyl 1,1-dimethylethyl ether + 3-methylpentane + 2-methyl-2-propanol ternary system at 101.3 kPa in a temperature range from 329 to 356 K. The data were found to be thermodynamically consistent according to the Van Ness−Byer−Gibbs method for the binary system and according to the McDermott−Ellis method for the ternary one. The binary system shows a minimum boiling azeotrope that boils at 333.4 K and contains 82.6 mol % of 3-methylpentane. The binary interaction parameters obtained from this work and literature data are used to predict the vapor−liquid equilibrium for the ternary system using the UNIQUAC, NRTL, and Wilson models. The ternary system is well predicted from binary data.

Introduction

Environmental legislation around the world has forced the reformulation of gasolines to phase out the lead additives and to reduce the reactive evaporative and exhaust emissions. Oxygenated compounds, ethers and alkanols, have been shown to be substances with excellent antiknock and pollution-reducing capabilities. The commonly used oxygenated additive is methyl 1,1-dimethylethyl ether (MTBE). 2-Methyl-2-propanol (TBA) is added in the oxygenated gasolines with ethanol to avoid the formation of two liquid phases in the presence of small quantities of water. Phase equilibrium data of oxygenated and hydrocarbon mixtures are important to predict the vapor-phase composition that would be in equilibrium with gasoline mixtures, for which iso-paraffins may constitute up to 30% weight. The systems reported here constitute a particular case of such mixtures.

In this work, isobaric vapor−liquid data at 101.3 kPa for the binary system 3-methylpentane (1) + TBA (2) and the ternary system MTBE (1) + 3-methylpentane (2) + TBA (3) have been obtained, tested for thermodynamic consistency, and VLE modeled. For these systems no VLE data have been previously reported.

Experimental Section

Chemicals. High-purity methyl 1,1-dimethylethyl ether (MTBE), 3-methylpentane, and 2-methyl-2-propanol (TBA) (Aldrich Chemical Co.) were used without further purification. The purities of all chemicals, checked by gas chromatography (GC), were as follows: MTBE, 99.8 mass %; 3-methylpentane, 100.0 mass % TBA, 99.9 mass %. The experimental densities of the pure liquids at 293.15 K were measured in a digital precision densimeter, Anton Paar DMA55. The refractive indexes at 293.15 K were measured using an Abbe refractometer (ATAGO 3T). Temperature was controlled to ±0.01 K with a thermostated bath. The accuracies in density and refractive index measurements were ±0.01 kg·m^{-3} and ±0.0002, respectively. The boiling points were determined using the apparatus described below. The experimental values of these properties are listed in Table 1 together with the literature values.

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

	d(298.15K)/kg m^{-3}		n_D(298.15K)		T_b(101.3 kPa)/K	
component	exptl	lit.	exptl	lit.	exptl	lit.
MTBE	735.20	735.28[a]	1.3664	1.3663[a]	328.2	328.11[b]
3-methyl-pentane	659.51	660.04[c]	1.3738	1.3739[a]	336.3	336.43[d]
TBA	775.40[e]	775.43[e,f]	1.3851	1.3859[a]	355.6	355.52[g]

[a] Daubert and Danner (1995). [b] Martínez-Ageitos (1996). [c] Awwad and Pethrick (1983). [d] Willingham et al. (1945). [e] Measured at 303.15 K. [f] Hales et al. (1983). [g] Ambrose and Sprake (1970).

Table 2. Antoine Coefficients, Eq 1

compound	A_i	B_i	C_i
MTBE[a]	14.6212	3022.19	−26.03
3-methylpentane[b]	14.3708	2999.76	−28.69
TBA[c]	14.8533	2649.89	−96.69

[a] Aucejo et al. (1998). [b] Loras et al. (1999a). [c] Aucejo et al. (1999).

Apparatus and Procedures. The equilibrium vessel used in this work is an all-glass, dynamic recirculating still described by Walas (1985), equipped with a Cottrell pump. The still (Labodest model), manufactured by Fischer Labor und Verfahrenstechnik (Bonn, Germany), is capable of handling pressures from 0.25 up to 400 kPa and temperatures up to 523.15 K. In the boiler, vapor is generated by external heating. The Cottrell pump ensures intimate contact between the liquid and vapor phases and also with the temperature-sensing element. The equilibrium tem-

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Table 3. Experimental Vapor−Liquid Equilibrium Data for 3-Methylpentane (1) + TBA (2) at 101.3 kPa

T/K	x_1	y_1	γ_1	γ_2	$-B_{11}$/cm^3·mol^{-1}	$-B_{22}$/cm^3·mol^{-1}	$-B_{12}$/cm^3·mol^{-1}
355.6	0.000	0.000					
352.5	0.028	0.142	3.261	0.996	1129	1184	852
351.0	0.043	0.203	3.132	0.998	1141	1200	861
347.5	0.086	0.344	2.914	0.990	1171	1239	882
344.2	0.140	0.458	2.622	0.997	1201	1277	903
342.5	0.179	0.513	2.404	1.010	1216	1298	914
340.5	0.240	0.570	2.107	1.053	1235	1323	927
339.6	0.270	0.601	2.026	1.058	1244	1334	933
338.0	0.335	0.650	1.851	1.095	1260	1355	944
337.0	0.383	0.673	1.728	1.151	1270	1368	951
336.1	0.440	0.699	1.604	1.217	1279	1380	958
335.2	0.501	0.729	1.508	1.284	1288	1393	964
334.4	0.584	0.753	1.370	1.454	1296	1404	970
334.0	0.640	0.770	1.292	1.599	1300	1409	973
333.8	0.690	0.786	1.231	1.743	1302	1412	974
333.6	0.751	0.804	1.164	2.010	1304	1415	976
333.3	0.805	0.821	1.117	2.371	1307	1419	977
333.4	0.857	0.840	1.071	2.892	1306	1418	977
333.6	0.907	0.865	1.036	3.725	1304	1415	976
334.2	0.954	0.902	1.009	5.308	1298	1407	971
335.2	0.982	0.951	1.002	6.553	1288	1393	964
336.3	1.000	1.000					

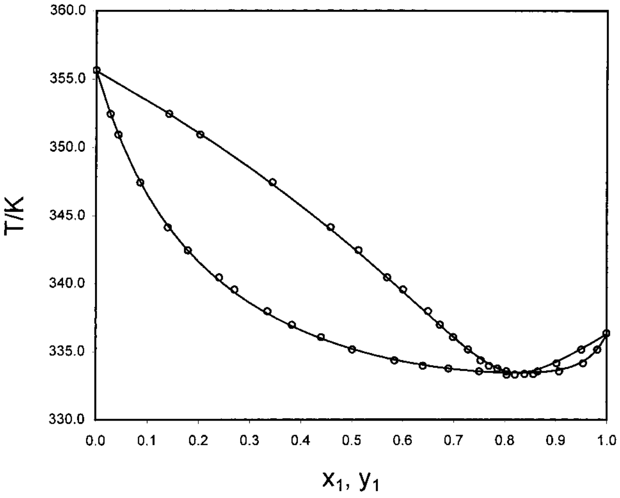

Figure 1. Temperature versus composition (x_1, y_1) for the system 3-methylpentane (1) + TBA (2) at 101.3 kPa: (○) experimental; (—) NRTL model.

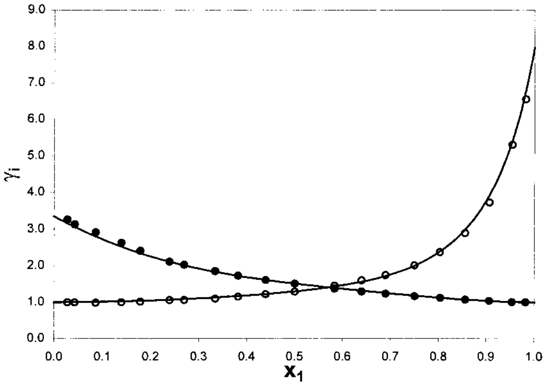

Figure 2. Activity coefficient plot for the system 3-methylpentane (1) + TBA (2) at 101.3 kPa: γ_1 (●); γ_2 (○); smoothed with the Legendre polynomial used in the consistency test (—).

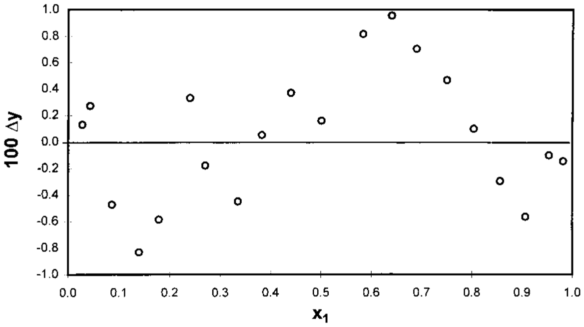

Figure 3. Deviation between calculated and measured vapor-phase mole fraction y_1 versus liquid-phase mole fraction x_1 for the system 3-methylpentane (1) + TBA (2) at 101.3 kPa.

Table 4. Correlation Parameters for Activity Coefficients, Activity Coefficients at Infinite Dilution γ_i^∞, and Mean Absolute Deviations MAD(y) and MAD(T) for the Binary System 3-Methylpentane (1) + TBA (2) at 101.3 kPa

model	A_{ij}/J·mol^{-1}	A_{ji}/J·mol^{-1}	α_{ij}	γ_1^∞	γ_2^∞	MAD(y)	MAD(T)/K
Wilson	146.56	5634.38		3.21	6.66	0.0053	0.23
NRTL	4530.95	1968.56	0.674	3.35	7.73	0.0027	0.08
UNIQUAC	2933.79	−1015.96		3.23	7.32	0.0102	0.35

perature is measured with a digital Fischer thermometer with an accuracy of ±0.1 K. For the pressure measurement, a digital manometer with an accuracy of ±0.01 kPa is used. The temperature probe was calibrated against the ice and steam points of distilled water. High-purity (>99.9 mass %) hexane vapor pressures were used for the manometer calibration.

In each VLE experiment, the pressure was fixed and held constant 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 at least for 30 min to ensure that a steady-state equilibrium was reached. Then, samples of liquid and condensed vapor were taken for analysis. The sample extractions were carried out with special syringes that allowed one to withdraw small-volume samples (<1.0 μL).

Analysis. All the samples were analyzed by using a Hewlett-Packard 5890 S-II gas chromatograph with a flame ionization detector. The chromatographic column was a 60 m, 0.2 mm i.d., fused silica capillary column, SUPELCOWAX 10. The chromatographic conditions were the following: oven temperature, 343 K; gas carrier, helium (50 cm^3·min^{-1}). The GC response peaks were integrated with a Hewlett-Packard 3396 integrator. The gas chromatograph was calibrated with gravimetrically prepared standard solutions. Twenty binary and sixty ternary samples covering the entire composition range were prepared to calibrate the gas chromatograph. The uncertainty of the composition measurements was estimated to be ±0.001 mole fraction. At least two analyses were made for each liquid and each condensed vapor sample.

Results and Discussion

Vapor pressures $P_i°$ were calculated with the Antoine equation

$$\ln(P_i°/\text{kPa}) = A - \frac{B}{(C + T/\text{K})} \qquad (1)$$

The Antoine constants for MTBE, 3-methylpentane, and TBA were taken from Aucejo et al. (1998), Loras et al. (1999a), and Aucejo et al. (1999), respectively, and are summarized in Table 2.

Binary System. The experimental VLE data for the binary system are given in Table 3 and Figure 1. The system presents a minimum boiling azeotrope. Figure 2

Table 5. Experimental Vapor−Liquid Equilibrium Data for MTBE (1) + 3-Methylpentane (2) + TBA (3) at 101.3 kPa

T/K	x_1	x_2	y_1	y_2	γ_1	γ_2	γ_3	$-B_{11}$[a]	$-B_{22}$[a]	$-B_{33}$[a]	$-B_{12}$[a]	$-B_{13}$[a]	$-B_{23}$[a]
335.1	0.044	0.463	0.059	0.676	1.088	1.519	1.273	1083	1289	1394	1159	1233	965
347.7	0.051	0.053	0.135	0.224	1.472	3.094	0.978	989	1169	1236	1056	1128	881
333.8	0.052	0.905	0.067	0.851	1.090	1.015	4.814	1094	1302	1412	1171	1245	974
333.4	0.060	0.638	0.072	0.727	1.029	1.246	1.709	1097	1306	1418	1175	1249	977
339.8	0.062	0.208	0.109	0.493	1.234	2.147	1.043	1047	1242	1332	1119	1192	932
337.4	0.091	0.286	0.138	0.540	1.141	1.834	1.101	1065	1266	1363	1139	1213	949
342.9	0.102	0.105	0.211	0.323	1.331	2.562	0.981	1024	1213	1293	1093	1166	911
334.3	0.106	0.470	0.135	0.641	1.057	1.452	1.299	1090	1297	1405	1166	1240	971
332.8	0.111	0.790	0.134	0.746	1.038	1.053	3.219	1102	1313	1426	1180	1254	981
332.8	0.111	0.692	0.130	0.710	1.013	1.143	2.150	1102	1313	1426	1180	1254	981
338.0	0.189	0.185	0.297	0.378	1.158	1.953	1.077	1060	1260	1355	1134	1208	944
334.2	0.194	0.384	0.247	0.535	1.055	1.492	1.272	1090	1298	1407	1167	1241	971
335.8	0.204	0.257	0.286	0.440	1.109	1.747	1.161	1078	1282	1384	1153	1227	960
332.9	0.212	0.476	0.252	0.566	1.026	1.324	1.526	1101	1312	1425	1179	1253	981
332.0	0.213	0.686	0.247	0.647	1.029	1.077	2.901	1109	1321	1438	1187	1262	987
332.3	0.214	0.591	0.245	0.610	1.005	1.169	2.012	1106	1318	1434	1185	1259	985
340.1	0.222	0.090	0.402	0.230	1.263	2.301	1.006	1044	1239	1328	1116	1190	930
332.0	0.247	0.707	0.295	0.644	1.058	1.040	3.684	1109	1321	1438	1187	1262	987
334.0	0.294	0.291	0.375	0.418	1.063	1.548	1.241	1092	1300	1409	1169	1243	973
331.9	0.303	0.495	0.342	0.523	1.004	1.213	1.835	1109	1322	1439	1188	1263	988
335.5	0.307	0.189	0.429	0.323	1.116	1.761	1.136	1080	1285	1389	1156	1230	962
331.5	0.312	0.586	0.351	0.555	1.015	1.099	2.586	1113	1327	1445	1192	1266	991
332.5	0.321	0.383	0.375	0.460	1.021	1.351	1.490	1104	1316	1431	1183	1257	984
338.5	0.345	0.055	0.567	0.122	1.199	2.093	1.044	1057	1255	1348	1130	1203	941
334.0	0.392	0.200	0.504	0.298	1.072	1.607	1.203	1092	1300	1409	1169	1243	973
331.6	0.398	0.399	0.445	0.428	1.004	1.243	1.739	1112	1325	1444	1191	1265	990
331.0	0.411	0.488	0.453	0.464	1.010	1.120	2.352	1117	1332	1452	1197	1271	995
335.7	0.412	0.102	0.587	0.182	1.132	1.831	1.087	1078	1283	1386	1154	1228	960
332.3	0.421	0.286	0.494	0.352	1.030	1.398	1.415	1106	1318	1434	1185	1259	985
330.7	0.445	0.506	0.494	0.459	1.026	1.080	2.800	1119	1335	1457	1199	1274	997
331.4	0.494	0.299	0.549	0.331	1.005	1.289	1.627	1114	1328	1447	1193	1267	992
330.5	0.507	0.392	0.549	0.377	1.007	1.152	2.151	1121	1337	1460	1201	1276	999
332.0	0.530	0.194	0.617	0.243	1.034	1.432	1.378	1109	1321	1438	1187	1262	987
334.3	0.541	0.056	0.720	0.091	1.102	1.738	1.140	1090	1297	1405	1166	1240	971
330.2	0.602	0.299	0.640	0.292	0.999	1.182	2.011	1124	1341	1464	1204	1279	1001
332.0	0.629	0.100	0.739	0.130	1.044	1.493	1.304	1109	1321	1438	1187	1262	987
331.1	0.643	0.151	0.716	0.175	1.016	1.370	1.495	1116	1331	1451	1196	1270	994
329.7	0.644	0.307	0.682	0.283	1.010	1.133	2.178	1128	1346	1472	1209	1283	1005
329.9	0.697	0.203	0.737	0.203	1.001	1.222	1.814	1126	1344	1469	1207	1281	1003
330.5	0.786	0.054	0.859	0.062	1.017	1.379	1.428	1121	1337	1460	1201	1276	999
329.8	0.791	0.110	0.831	0.114	0.999	1.265	1.674	1127	1345	1470	1208	1282	1004
329.0	0.888	0.063	0.910	0.062	1.000	1.245	1.722	1134	1354	1482	1216	1290	1010

[a] Units: $\mathrm{cm^3 \cdot mol^{-1}}$.

shows the experimental liquid-phase activity coefficients γ_i that were calculated from

$$\ln \gamma_i = \ln\left(\frac{Py_i}{x_i P_i^{\circ}}\right) + (B_{ii} - v_i^{\mathrm{L}})(P - P_i^{\circ})/RT + (P/2RT)\sum_1^n \sum_1^n y_j y_k (2\delta_{ji} - \delta_{jk}) \quad (2)$$

$$\delta_{ji} = 2B_{ji} - B_{jj} - B_{ii} \quad (3)$$

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, and B_{ij} is the cross second virial coefficient. The molar virial coefficients B_{ii} and B_{ij} were estimated by the method of Hayden and O'Connell (1975) using the molecular parameters suggested by Prausnitz et al. (1980). Critical properties of all components were taken from DIPPR (1995). The last two terms in eq 2 contributed <1.5% to the activity coefficient. As can be observed from the activity coefficients, the system shows positive deviations from ideality.

The thermodynamic consistency of the VLE experimental data was checked by the point-to-point test of Van Ness−Byer−Gibbs (1973) using a four-parameter Legendre polynomial for the excess Gibbs free energy. The objective function selected to minimize was the sum of the squared relative deviations in the total pressure. The consistency criteria in this test are that the mean absolute deviation between calculated and measured mole fractions of component 1 in the vapor phase, MAD(y), is <0.01 and that deviations scatter randomly about zero. To check the absence of bias in the data, a graphical inspection of the error in y_1 is performed; y_1 residuals are plotted versus x_1 in Figure 3. Experimental VLE data were found to be thermodynamically consistent according to this test, with a value of MAD(y) of 0.0040 and a sum of y_1 residuals (BIAS) of 0.0074.

The activity coefficients were correlated with the Wilson, NRTL, and UNIQUAC equations (Gmehling and Onken, 1977). To fit the binary parameters, a nonlinear optimization method was used to minimize the function

$$F = \sum_n \sum_i \left(\frac{\gamma_{\mathrm{exptl}} - \gamma_{\mathrm{calcd}}}{\gamma_{\mathrm{exptl}}}\right)_{i,n}^2 \quad (4)$$

where n is the number of data points. The parameters A_{12}, A_{21}, and α_{12} for the correlation equations, mean absolute deviations, and activity coefficients at infinite dilution γ_i^{∞} are given in Table 4. The best fit was obtained with the NRTL equation. An azeotropic composition and temperature were calculated with the NRTL equation at the conditions $x_1 = 0.826$ and $T = 333.4$ K.

Table 6. Correlation of Experimental Ternary Data with Wilson, NRTL, and UNIQUAC Equations Using Binary Interaction Parameters for MTBE (1) + 3-Methylpentane (2) + TBA (3) at 101.3 kPa

model	*ij*	A_{ij}/J·mol^{-1}	A_{ji}/J·mol^{-1}	α_{ij}	system	MAD(y_1)	MAD(y_2)	MAD(y_3)	max. dev in *y*	MAD(*T*)/K
Wilson	1−2[a]	309.33	225.19							
	1−3[b]	−687.80	2263.47		1 + 2 + 3	0.0024	0.0022	0.0037	0.018	0.16
	2−3	146.56	5634.38							
NRTL	1−2[a]	675.98	−139.21	0.30						
	1−3[b]	3080.07	−1211.97	0.30	1 + 2 + 3	0.0023	0.0032	0.0026	0.017	0.12
	2−3	4530.95	1968.56	0.674						
UNIQUAC	1−2[a]	141.87	−11.48							
	1−3[b]	2267.26	−1295.30		1 + 2 + 3	0.0021	0.0052	0.0052	0.026	0.21
	2−3	2933.79	−1015.96							

[a] Loras et al. (1999a). [b] Loras et al. (1999b).

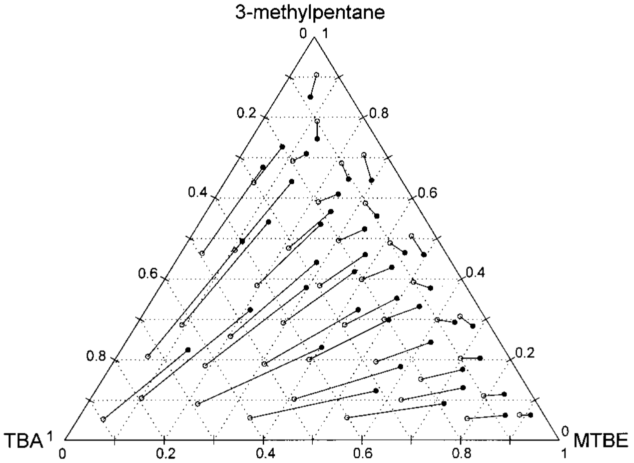

Figure 4. Vapor−liquid equilibrium tie lines for the MTBE (1) + 3-methylpentane (2) + TBA (3) system at 101.3 kPa: (○) liquid-phase mole fractions; (●) vapor-phase mole fractions.

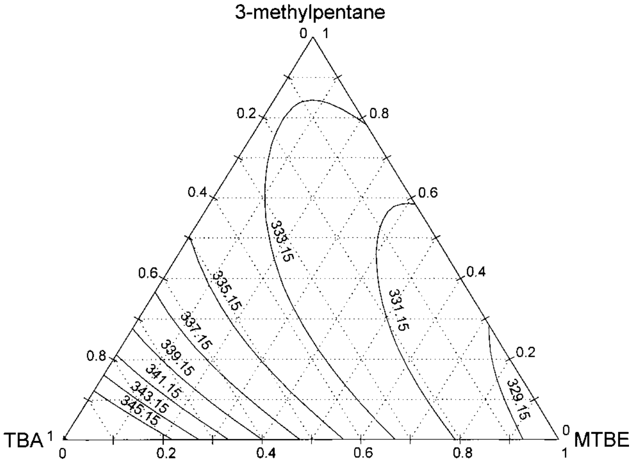

Figure 5. Vapor−liquid isotherms for the MTBE (1) + 3-methylpentane (2) + TBA (3) system at 101.3 kPa calculated with the NRTL model as a function of the liquid mole fraction.

Ternary System. The VLE data for the ternary system are shown in Table 5 and Figure 4. The activity coefficients γ_i were calculated from eq 2. The last two terms in eq 2 contributed <2.7% to the activity coefficient. As can be observed, the system shows positive deviations from ideality. The ternary data were found to be thermodynamically consistent, as tested by the McDermott−Ellis method (1965) modified by Wisniak and Tamir (1977). The test requires that $D_i < D_{\max}$ for every experimental point. Values calculated of $D_{\max} - D_i$ were >0 for every experimental point with a mean value of 0.048.

Vapor−liquid equilibrium for the ternary system has been predicted by using the Wilson, NRTL, and UNIQUAC models for the activity coefficients of the components with the binary interaction parameters obtained from the regression of binary data. The values of the binary interaction parameters have been taken from this study and from the literature (Loras et al., 1999a,b) and are summarized in Table 6 for the readers' convenience. Table 6 also lists the mean absolute deviations between experimental and calculated temperatures and vapor-phase mole fractions of the components, and the maximum deviations in vapor-phase mole fractions. The three models yield similar deviations, with each representing the data successfully. Thus, the models can be used to predict the vapor-phase compositions and 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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