

# Isobaric Vapor–Liquid Equilibria in the Systems Methyl 1,1-Dimethylethyl Ether + Hexane and + Heptane

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The vapor–liquid equilibrium at 94 kPa has been determined for the binary systems of methyl 1,1-dimethylethyl ether (MTBE) with hexane and with heptane. Both systems deviate slightly from ideal behavior, can be described as regular solutions, and do not present an azeotrope. The activity coefficients and boiling point of the solutions were correlated with its composition by the Redlich–Kister, Wohl, Wilson, UNIQUAC, NRTL, and Wisniak–Tamir equations.

Amendments of the U.S. Clean Air in 1990 have mandated that new gasoline formulations be sold in highly polluted areas of the country, with oxygenated gasolines being supplied particularly during the winter. MTBE is the primary oxygenated compound being used to reformulate gasolines to improve their octane rating and pollution-reducing capability. In addition, MTBE is finding acceptance as an effective replacement for methylene chloride, aromatics, and others, as well as a commercial outlet for 2-methyl-2-propanol. Unlike most ethers, MTBE minimizes handling and storage concerns associated with peroxide formation and exhibits excellent oxidative stability, resisting peroxide formation without added inhibitors. Phase equilibrium data of oxygenated mixtures are important for predicting the vapor phase composition that would be in equilibrium with hydrocarbon mixtures. The vapor–liquid equilibrium for the binary system MTBE (1) + hexane (2) has been measured by Plura et al. (1979) at 96 kPa and at 333.15 K, by Jin et al. (1985) at (323, 343, and 363) K, and by Lozano et al. (1995) at 313.15 K. According to Plura et al. the system can be described by the one-constant Redlich–Kister equation while according to Jin et al. by the Wilson equation. Vapor–liquid equilibrium data for the system MTBE (1) + heptane (3) and infinite dilution activity coefficients have been measured by Lee et al. (1994, 1995) at 318.15 and 338.15 K using head space gas chromatography. The activity coefficients at infinite dilution of the two binaries have been measured by Pividal et al. (1992) at (313.15 and 323.15) K. Tusel-Langer et al. (1991) have measured the heat of mixing in solutions of MTBE + heptane at 298.15 and 313.15 K and found the process to be endothermic. According to Tusel-Langer et al. a packing effect in the liquid state between chainlike molecules of heptane is responsible for the endothermic behavior, mixing with compact molecules like MTBE gradually destroys the packing effect. It is reasonable to suppose that solutions of MTBE + heptane will exhibit a similar behavior. The present work was undertaken to measure vapor–liquid equilibria (VLE) data for the title systems for which isobaric data are not available or are incomplete.

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**Table 1. Mole Percent GLC Purities (Mass %), Refractive Index  $n_D$  at the Na D line, and Normal Boiling Points  $T$  of Pure Components**

component (purity/mass %)	$n_D(298.15)$	$T/K$
methyl 1,1-dimethylethyl ether (99.93)	1.3661 <sup>a</sup>	328.29 <sup>a</sup>
	1.3663 <sup>b</sup>	327.85 <sup>b</sup>
hexane (99.73)	1.3730 <sup>a</sup>	341.84 <sup>a</sup>
	1.37226 <sup>b</sup>	341.89 <sup>b</sup>
heptane (99.57)	1.3851 <sup>a</sup>	371.54 <sup>a</sup>
	1.38511 <sup>b</sup>	371.57 <sup>b</sup>

<sup>a</sup> Measured. <sup>b</sup> TRC, 1974.

## Experimental Section

**Purity of Materials.** Methyl *tert*-butyl ether (99.93 mass %), hexane (99.73+ mass %), and heptane (99.57 mass %) were purchased from Aldrich. The reagents were used without further purification after gas chromatography failed to show any significant impurities. The properties and purity (as determined by GLC) of the pure components appear in Table 1.

**Apparatus and Procedure.** An all glass vapor–liquid equilibrium apparatus model 602, manufactured by Fischer Labor und Verfahrenstechnik (Germany), was used in the equilibrium determinations. In this circulation method apparatus, the solution is heated to its boiling point by a 250 W immersion heater (Cottrell pump). The vapor–liquid mixture flows through an extended contact line which guarantees an intense phase exchange and then enters a separation chamber whose construction prevents an entrainment of liquid particles into the vapor phase. The separated gas and liquid phases are condensed and returned to a mixing chamber, where they are stirred by a magnetic stirrer, and returned again to the immersion heater. Temperature control is achieved by a 5 mm diameter Pt-100 temperature sensor, with an accuracy of  $\pm 0.1$  K. The total pressure of the system is controlled by a vacuum pump capable of working under vacuum up to 0.25 kPa. The pressure is measured by a Vac Probs with an accuracy of  $\pm 0.1$  kPa. On average the system reaches equilibrium conditions after 1–2 h of operation. Samples, taken by syringing out 0.7  $\mu$ L after the system had achieved equilibrium, were analyzed by gas chromatography on a Gow-Mac series 550P apparatus provided with a thermal conductivity detector and a Spectra Physics Model SP 4290 electronic integrator. The column was 3 m long and 0.2

**Table 2. Experimental Vapor–Liquid Equilibrium Data for Methyl 1,1-Dimethylethyl Ether (1) + Hexane (2) at 94 kPa**

$T/K$	$x_1$	$y_1$	$\gamma_1$	$\gamma_2$	$-B_{11}/\text{cm}^3 \text{ mol}^{-1}$	$-B_{22}/\text{cm}^3 \text{ mol}^{-1}$	$-B_{12}/\text{cm}^3 \text{ mol}^{-1}$	$G^E/RT$
338.35	0.027	0.048	1.2031	1.0139	1097	1351	1200	0.018
337.75	0.051	0.088	1.1882	1.0145	1102	1357	1205	0.022
337.05	0.087	0.144	1.1633	1.0114	1108	1365	1212	0.024
335.95	0.142	0.220	1.1248	1.0150	1117	1377	1222	0.029
334.50	0.206	0.304	1.1186	1.0244	1130	1393	1236	0.042
333.65	0.256	0.367	1.1148	1.0216	1137	1403	1245	0.044
332.55	0.320	0.432	1.0854	1.0390	1147	1416	1256	0.052
330.85	0.422	0.536	1.0758	1.0551	1163	1436	1273	0.062
330.05	0.494	0.599	1.0528	1.0692	1170	1445	1281	0.059
329.75	0.520	0.622	1.0483	1.0731	1173	1449	1284	0.058
329.25	0.547	0.647	1.0529	1.0795	1177	1455	1290	0.063
328.25	0.651	0.733	1.0342	1.0956	1187	1467	1300	0.054
327.55	0.703	0.772	1.0313	1.1254	1194	1476	1308	0.057
327.20	0.770	0.822	1.0137	1.1480	1197	1480	1311	0.042
326.85	0.809	0.852	1.0113	1.1631	1200	1485	1315	0.038
326.35	0.859	0.891	1.0121	1.1802	1205	1491	1321	0.034
325.85	0.929	0.945	1.0086	1.2030	1210	1497	1326	0.021
325.65	0.951	0.962	1.0095	1.2126	1212	1500	1328	0.018
325.60	0.964	0.971	1.0068	1.2617	1212	1500	1329	0.015
325.59	1.000	1.000	1.0000		1213	1500		0.000
$\gamma_i^\infty$ <sup>a</sup>			1.21	1.23				

<sup>a</sup> Calculated according to the method suggested by Wisniak et al. (1996).

**Table 3. Experimental Vapor–Liquid Equilibrium Data for Methyl 1,1-Dimethylethyl Ether (1) + Heptane (3) at 94 kPa**

$T/K$	$x_1$	$y_1$	$\gamma_1$	$\gamma_3$	$-B_{11}/\text{cm}^3 \text{ mol}^{-1}$	$-B_{33}/\text{cm}^3 \text{ mol}^{-1}$	$-B_{13}/\text{cm}^3 \text{ mol}^{-1}$	$G^E/RT$
366.45	0.021	0.078	1.2041	1.0145	896	1599	1169	0.018
365.35	0.038	0.134	1.1886	1.0008	903	1612	1178	0.007
364.25	0.048	0.167	1.1878	1.0051	910	1626	1188	0.013
363.65	0.060	0.202	1.1697	0.9924	913	1634	1193	0.002
360.55	0.100	0.314	1.1719	0.9779	933	1674	1220	-0.004
359.95	0.105	0.323	1.1652	0.9882	937	1682	1226	0.005
356.25	0.157	0.429	1.1351	0.9917	962	1733	1260	0.013
350.55	0.239	0.551	1.1102	1.0357	1002	1816	1316	0.052
349.25	0.272	0.600	1.0996	1.0064	1011	1836	1329	0.030
348.55	0.292	0.618	1.0750	1.0114	1016	1846	1337	0.029
346.25	0.337	0.677	1.0861	0.9859	1034	1883	1361	0.018
342.85	0.415	0.743	1.0637	0.9979	1060	1938	1398	0.024
340.75	0.465	0.782	1.0605	0.9955	1077	1974	1422	0.025
340.65	0.470	0.786	1.0576	0.9900	1078	1976	1423	0.021
336.55	0.597	0.853	1.0181	1.0345	1112	2049	1472	0.024
334.85	0.638	0.876	1.0292	1.0332	1127	2080	1492	0.030
333.75	0.674	0.892	1.0254	1.0404	1136	2101	1506	0.030
332.15	0.730	0.915	1.0196	1.0490	1151	2132	1527	0.027
331.15	0.771	0.929	1.0113	1.0649	1160	2151	1540	0.023
330.45	0.803	0.941	1.0046	1.0636	1166	2165	1549	0.016
329.15	0.853	0.957	1.0016	1.0913	1178	2191	1566	0.014
328.15	0.886	0.967	1.0058	1.1152	1188	2212	1580	0.018
327.85	0.899	0.971	1.0046	1.1258	1191	2218	1584	0.016
326.85	0.944	0.984	1.0009	1.1644	1200	2239	1597	0.009
326.45	0.958	0.988	1.0030	1.1826	1204	2248	1603	0.010
325.59	1.000	1.000	1.0000		1213	2266	1615	0.000
$\gamma_i^\infty$ <sup>a</sup>			1.11	1.17				

<sup>a</sup> Calculated according to the method suggested by Wisniak et al. (1996).

cm in diameter, packed with SE-30. Column, injector, and detector temperatures were (333.15, 453.15, and 543.15) K for the system MTBE + hexane and (353.15, 493.15, and 543.15) K for the system MTBE + heptane. Very good separation was achieved under these conditions, and calibration analyses were carried out to convert the peak ratio to the mass composition of the sample. The pertinent polynomial fit had a correlation coefficient  $R^2$  better than 0.99. Concentration measurements were accurate to better than  $\pm 0.009$  mole fraction.

## Results

The temperature  $T$  and liquid-phase  $x_i$  and vapor-phase  $y_i$  mole fraction at  $P = 94$  kPa are reported in Tables 2 and 3 and Figures 1 and 2, together with the activity coefficients  $\gamma_i$  that were calculated from the following

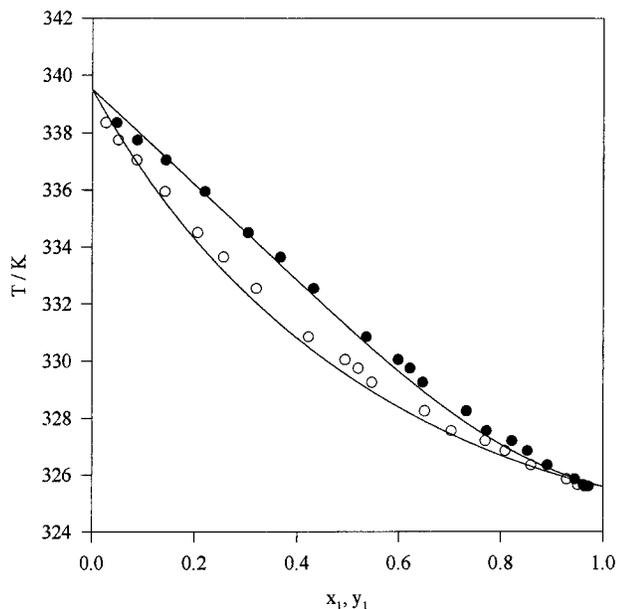
equation (Van Ness and Abbott, 1982):

$$\ln \gamma_i = \ln \frac{y_i P}{x_i P_i^0} + \frac{(B_{ii} - V_i^L)(P - P_i^0)}{RT} + y_j^2 \frac{\delta_{ij} P}{RT} \quad (1)$$

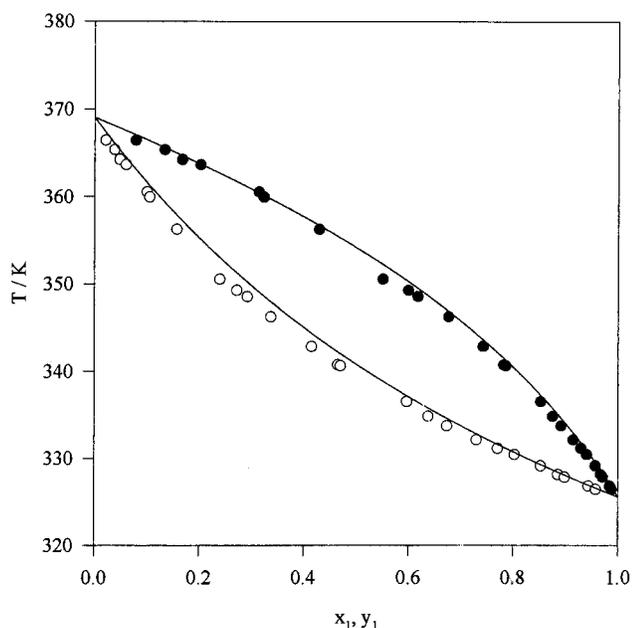
where  $T$  and  $P$  are the boiling point and the total pressure,  $V_i^L$  is the molar liquid volume of component  $i$ ,  $B_{ii}$  and  $B_{jj}$  are the second virial coefficients of the pure gases,  $P_i^0$  is the vapor pressure,  $B_{ij}$  is the cross second virial coefficient, and

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

The standard state for calculation of activity coefficients is the pure component at the pressure and temperature of the solution. Equation 1 is valid at low and moderate



**Figure 1.** Boiling temperature diagram for the system methyl 1,1-dimethylethyl ether (1) + hexane (2) at 94 kPa: bubble-point temperature curve (○); dew-point temperature curve (●); UNIFAC prediction (—).



**Figure 2.** Boiling temperature diagram for the system methyl 1,1-dimethylethyl ether (1) + heptane (3) at 94 kPa: bubble-point temperature curve (○); dew-point temperature curve (●); UNIFAC prediction (—).

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 liquid volumes of the pure components are incompressible over the pressure range under consideration. The pure component vapor pressures  $P_i^0$  were calculated according to the Antoine equation:

$$\log(P_i^0/\text{kPa}) = A_i - \frac{B_i}{(TK) - C_i} \quad (3)$$

where the Antoine constants  $A_i$ ,  $B_i$ , and  $C_i$  are reported in Table 4. The molar virial coefficients  $B_{ii}$  and  $B_{ij}$  were estimated by the method of Hayden and O'Connell (1975)

**Table 4. Antoine Coefficients, Eq 3**

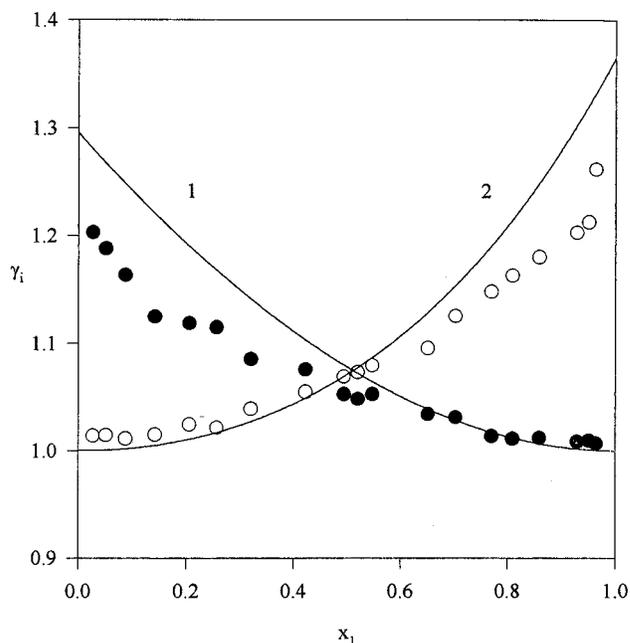
compound	$A_i$	$B_i$	$C_i$
methyl 1,1-dimethylethyl ether <sup>a</sup>	5.860 78	1032.988	59.876
hexane <sup>b</sup>	6.000 91	1171.170	48.740
heptane <sup>b</sup>	6.021 67	1264.900	56.610

<sup>a</sup> Reich (1996). <sup>b</sup> TRC (1974).

**Table 5. Activity Coefficients at Infinite Dilution for Methyl 1,1-Dimethylethyl Ether (1) + Hexane (2) and + Heptane (3)**

system	$\gamma_i^\infty$	$\gamma_j^\infty$	$T/K$
MTBE (1) + hexane (2) [ $i = 1, j = 2$ ]	1.26 <sup>a</sup>	1.16 <sup>a</sup>	303.15
	1.20 <sup>a</sup>	1.15 <sup>a</sup>	323.15
		1.23 <sup>b</sup>	326.04
	1.21 <sup>b</sup>		339.51
		1.36 <sup>c</sup>	325.59
MTBE (1) + heptane (3) [ $i = 1, j = 3$ ]	1.29 <sup>c</sup>		339.51
	1.31 <sup>a</sup>	1.01 <sup>a</sup>	313.15
	1.172 <sup>d</sup>	1.165 <sup>d</sup>	318.15
	1.22 <sup>a</sup>	0.94 <sup>a</sup>	323.15
		1.17 <sup>b</sup>	325.59
	1.11 <sup>b</sup>		369.09
	1.41 <sup>c</sup>	326.04	
	1.22 <sup>c</sup>		369.09

<sup>a</sup> Pividal et al. (1992). <sup>b</sup> Extrapolated from present VLE data according to the method of Wisniak et al. (1996). <sup>c</sup> Predicted by modified UNIFAC (Larsen, 1987). <sup>d</sup> Lee et al. (1994).



**Figure 3.** Activity coefficients for the system methyl 1,1-dimethylethyl ether (1) + hexane (2) at 94 kPa:  $\gamma_1^{\text{exptl}}$  (○);  $\gamma_2^{\text{exptl}}$  (●); UNIFAC prediction (—).

by assuming the association parameter  $\eta$  to be zero. Critical properties of MTBE were taken from Ambrose et al. (1974). The last two terms in eq 1, particularly the second one that expresses the correction due to the nonideal behavior of the vapor phase, contributed less than 2% to the activity coefficients in the binary of MTBE with hexane and less than 7% in the binary with heptane; in general, their influence was important only at very dilute concentrations. The calculated activity coefficients reported in Tables 2 and 3 are estimated accurate to within  $\pm 3\%$ . The results reported in these tables indicate that both systems exhibit small positive deviations from ideal behavior and that no azeotrope is present. Tables 2 and 3 contain also the activity coefficients at infinite dilution calculated by the method suggested by Wisniak et al. (1996). These

**Table 6. Parameters and Deviations between Experimental and Calculated Values for Different  $G^E$  Models**

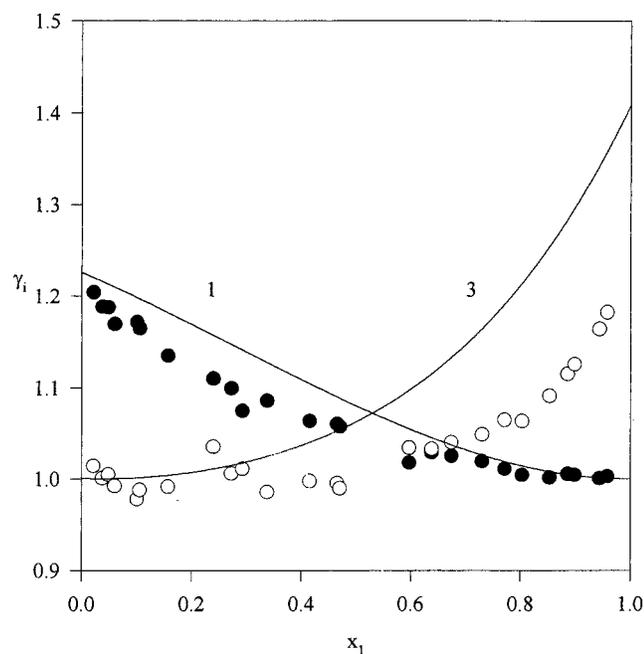
A. Redlich–Kister, Eq 4						
system	$B$	$C$	max dev % <sup>a</sup>	av dev % <sup>b</sup>	rmsd <sup>c</sup>	
methyl 1,1-dimethylethyl ether (1) + hexane (2)	0.0847	4.49	1.52	0.004		
	0.0847	0.0054	3.47	1.40	0.004	
methyl 1,1-dimethylethyl ether (1) + heptane (3)	0.0740		6.05	3.21	0.008	
	0.0742	0.0014	5.89	3.23	0.008	
B. Other Models <sup>d</sup>						
model	system	$A_{12}$	$A_{21}$	$q_1/q_2$	$\alpha$	$\delta(y)^e$
Wohl	1 + 2	0.2123	0.2674	0.7197		0.0044
	1 + 3	0.1707	0.1546	1.1220		0.0040
Wilson	1 + 2	901.75 <sup>f</sup>	-244.71 <sup>f</sup>			0.0029
	1 + 3	788.27 <sup>f</sup>	-291.12 <sup>f</sup>			0.0042
NRTL	1 + 2	259.32 <sup>f</sup>	356.36 <sup>f</sup>		0.358	0.0028
	1 + 3	-746.85 <sup>f</sup>	932.06 <sup>f</sup>		0.352	0.0040
UNIQUAC	1 + 2	68.537 <sup>f</sup>	-16.509 <sup>f</sup>			0.0038
	1 + 3	-62.123 <sup>f</sup>	-444.90 <sup>f</sup>			0.0040

<sup>a</sup> Maximum deviation, %. <sup>b</sup> Average deviation, %. <sup>c</sup> Root mean square deviation. <sup>d</sup> All equations in  $\ln \gamma_i$  form. <sup>e</sup>  $\delta(y) = \sum |y_{\text{exptl}} - y_{\text{calcd}}|/N$  ( $N$ : number of data points). <sup>f</sup> Parameters in J/mol.

**Table 7. Coefficients in Correlation of Boiling Points, Eq 6, Average Deviation, and Root Mean Square Deviations in Temperature, rmsd**

Redlich–Kister, Eq 4						
system	$C_0$	$C_1$	$C_2$	max dev <sup>a</sup> /K	av dev <sup>b</sup> /K	rmsd <sup>c</sup> /K
methyl 1,1-dimethylethyl ether (1) + hexane (2)	-9.1536	4.5569	-9.8725	0.6	0.23	0.06
methyl 1,1-dimethylethyl ether (1) + heptane (3)	-29.6561	18.4664	-14.1848	0.67	0.27	0.07

<sup>a</sup> Maximum deviation. <sup>b</sup> Average deviation. <sup>c</sup> Root mean square deviation.



**Figure 4.** Activity coefficients for the system methyl 1,1-dimethylethyl ether (1) + heptane (3) at 94 kPa:  $\gamma_{1,\text{exptl}}$  (○);  $\gamma_{2,\text{exptl}}$  (●); UNIFAC prediction (—).

values are compared in Table 5 with those reported in the literature; a fair agreement is observed for the activity coefficients at infinite dilution of MTBE solutions in either hydrocarbon. Heptane diluted in MTBE shows the expected positive deviation from ideal behavior, in contrast to the negative deviation reported by Pividal et al. (1992).

The vapor–liquid equilibria data reported in Tables 2 and 3 were found to be thermodynamically consistent by the  $L$ – $W$  method of Wisniak (1993) and the point-to-point method of Van Ness et al. (1973) as modified by Fredenslund et al. (1977). For both binaries, the residuals of the Fredenslund test were randomly distributed, as measured

by the Durbin–Watson statistic. The variation of  $G^E/RT$  with composition appears in Tables 2 and 3, the value of  $G^E$  ( $x = 0.5$ ) for the system MTBE + hexane is almost double that of the system MTBE + heptane, illustrating the closer packing of the hexane molecules as compared to heptane molecules.

The activity coefficients were correlated with the Redlich–Kister, Wohl, Wilson, NRTL, and UNIQUAC equations (Walas, 1985) and compared with those of the modified UNIFAC group contribution method (Larsen et al., 1987). The following expression was used for the Redlich–Kister (1948) expansion

$$\log(\gamma_1/\gamma_2) = B(x_2 - x_1) + C(6x_1x_2 - 1) \quad (4)$$

The values of the constants  $B$  and  $C$  were determined by multilinear regression and appear in Table 6 together with the pertinent statistics. It is seen that the Redlich–Kister model gives a good representation of the data for the system MTBE + hexane and a reasonable fit of the second binary system, with the largest deviations occurring at the dilute end of the components. In addition, it is seen that both binary systems behave like a regular solution, this result is in excellent accordance with that of Plura et al. (1979) who found that the system MTBE + hexane behaves regularly with  $B = 0.038$  at 333.15 K and  $B = 0.0834$  at 96 kPa. The parameters of the Wohl, Wilson, NRTL, and UNIQUAC equations were obtained by minimizing the following objective function (OF)

$$\text{OF} = \sum_{i=1}^N \left( \frac{\gamma_{1,i}^{\text{exptl}} - \gamma_{1,i}^{\text{calc}}}{\gamma_{1,i}^{\text{exptl}}} \right)^2 + \left( \frac{\gamma_{2,i}^{\text{exptl}} - \gamma_{2,i}^{\text{calc}}}{\gamma_{2,i}^{\text{exptl}}} \right)^2 \quad (5)$$

and are reported in Table 6, together with the relative deviation of the vapor composition. Inspection of the results given in Table 7 shows that all four models fitted well both systems, the best fit corresponding to the NRTL model for the MTBE + hexane system and MTBE +

heptane system. The capability of predicting the vapor-phase composition has been used as the ranking factor. The boiling point diagram and the activity coefficients predicted by the modified UNIFAC group contribution method (Larsen et al., 1987) are shown in Figures 1–4. According to these figures it can be concluded that the modified UNIFAC method overpredicts the activity coefficients. In addition, an apparent considerable experimental noise is observed for the MTBE + heptane system (Figure 4), which can be explained in terms of the almost ideal behavior of the system, as well as the narrow scale of the figure.

The boiling point of the solution was correlated with its composition by the equation proposed by Wisniak and Tamir (1976):

$$T/K = x_1 T_1^0 + x_2 T_2^0 + x_1 x_2 \sum_{k=1}^m C_k (x_1 - x_2)^k \quad (6)$$

In this equation  $T_i^0/K$  is the boiling point of the pure component  $i$  and  $m$  are the number of terms in the series expansion of  $(x_1 - x_2)$ . The various constants of eq 6 are reported in Table 6, which also contains information indicating the degree of goodness of the correlation.

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