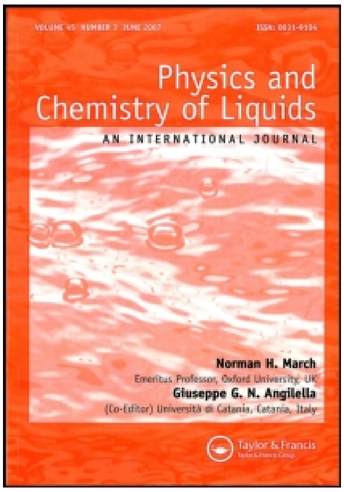

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Phase Equilibria in the Ternary System Methyl 1,1-Dimethylethyl Ether Oxolane Heptane

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PHASE EQUILIBRIA IN THE TERNARY SYSTEM METHYL 1,1-DIMETHYLETHYL ETHER + OXOLANE + HEPTANE

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Vapor-liquid equilibrium at 94 kPa has been determined for the ternary system methyl 1,1-dimethylethyl ether (MTBE) + oxolane + heptane. The system deviates positively from ideality and no azeotrope is present. The ternary activity coefficients and the boiling points of the system have been correlated with the composition using the Redlich-Kister, Wilson, NRTL, UNIQUAC, UNIFAC, and Wisniak-Tamir relations. Most of the models allow a good prediction of the activity coefficients of the ternary system from those of the pertinent binary systems.

Keywords: Vapor-liquid equilibrium; activity coefficients; ternary systems

1. INTRODUCTION

In 1992, the oxy-fuel program was introduced in the U.S. that required that gasoline had to contain 2.7% oxygen by mass in the winter months in areas that where in non-attainment on CO standards. These regulations have caused oxygenates like MTBE and ethanol to play a significant role as octane improvers. MTBE has been used as a gasoline blending agent since 1979. Now, it is the primary oxygenated compound being used to reformulate gasolines to improve their octane rating and pollution-reducing capability. Cyclic ethers are frequently

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used in the chemical industry as solvents and intermediates. Phase equilibrium data of oxygenated mixtures are important for predicting the vapor phase composition that would be in equilibrium with hydrocarbon mixtures. Two of the compounds tested here are oxygenates that may be considered as additives for gasoline. Vapor-liquid equilibrium data for the three binary systems methyl 1,1-dimethylethyl ether (1) + heptane (3), methyl 1,1-dimethylethyl ether (1) + oxolane (3), and oxolane + heptane (3) have already been reported at 94 kPa by Wisniak *et al.* [1, 2, 3], the three systems present slight to moderate positive deviations from ideality and do not have azeotropic points. The present work was undertaken to measure vapor-liquid equilibria (VLE) data for the ternary system for which no data are not available.

2. EXPERIMENTAL SECTION

Purity of Materials

Methyl 1,1-dimethylethyl ether (99.93 mass %), oxolane (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 I.

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 then

TABLE I Mole Percent GLC Purities (mass %), Refractive Index n_D at the Na D line, and Normal Boiling points T of Pure Componnents

Component (purity/mass %)	$n_D(298.15\ K)$	T/K
methyl 1,1-dimethylethyl ether (99.93)	1.3661[a] 1.3663[b]	328.29[a] 328.35[b]
oxolane (99.9)	1.4050[a] 1.4049[c]	339.15[a] 339.12[c]
heptane (99.57)	1.3851[a] 1.38513[d]	371.54[a] 371.553[e]

[a] Measured
[b] TRC Tables, a-6040[14]
[c] TRC Tables, a-6170[14]
[d] TRC Tables, f-1460[15]
[e] TRC Tables, k-1460[15]

returned again to the immersion heater. Temperature control is achieved by a 5 mm diameter Pt-100 temperature sensor, with an accuracy of 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 pressure transducer with an accuracy of 0.1 kPa. On the average the system reaches equilibrium conditions after 0.5 – 1 h of operation. Samples, taken by syringing 0.7 μL after the system has 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 cm in diameter, packed with SE-30. The column, injector, and detector temperatures were (343.15, 493.15, and 543.15) K. 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 fits had a correlation R^2 better than 0.99. Concentration measurements were accurate to better than 0.009 mole fraction.

3. RESULTS

The temperature T and liquid-phase x_i, and vapor-phase y_i mole fraction measurements at $P = 94$ kPa are reported in Table II, together

TABLE II Experimental Vapor-Liquid Equilibria Data for Methyl 1,1-Dimethylethyl Ether (1) + Oxolane (2) + Heptane (3) at 94 kPa

					Activity coefficients			*Virial coefficients* $(cm^3\ mol^{-1})$					
T/k	X_1	X_2	Y_1	Y_2	γ_1	γ_2	γ_3	$-B_{12}$	$-B_{13}$	$-B_{23}$	$-B_{11}$	$-B_{22}$	$-B_{33}$
327.75	0.863	0.062	0.929	0.049	1.0052	1.0807	1.1132	1126	1052	2143	1089	1557	1529
328.05	0.753	0.204	0.817	0.169	1.0035	1.1107	1.2127	1124	1049	2137	1087	1553	1525
328.05	0.860	0.052	0.93	0.043	0.9989	1.0985	1.2222	1124	1049	2137	1087	1553	1525
328.15	0.747	0.219	0.809	0.181	0.9983	1.1009	1.1339	1123	1048	2135	1086	1552	1524
328.15	0.788	0.156	0.852	0.132	0.9954	1.1270	1.1523	1123	1048	2135	1086	1552	1524
328.45	0.812	0.100	0.881	0.090	0.9914	1.1867	1.2314	1120	1045	2130	1083	1548	1520
329.15	0.808	0.067	0.902	0.060	0.9972	1.1491	1.1289	1114	1039	2117	1077	1539	1511
329.35	0.748	0.137	0.849	0.116	1.0064	1.0841	1.1321	1112	1038	2113	1076	1537	1509
329.85	0.798	0.037	0.917	0.032	1.0042	1.1042	1.1046	1108	1034	2104	1071	1531	1502
330.05	0.694	0.182	0.794	0.167	0.9932	1.1483	1.1329	1106	1032	2101	1070	1528	1500
330.65	0.659	0.189	0.775	0.179	1.0018	1.1579	1.0691	1101	1027	2090	1065	1521	1492
330.75	0.614	0.255	0.725	0.232	1.0028	1.1089	1.1581	1100	1026	2088	1064	1520	1491
331.05	0.583	0.318	0.672	0.294	0.9702	1.1191	1.1898	1098	1024	2083	1061	1516	1487
331.15	0.544	0.351	0.645	0.318	0.9952	1.0922	1.2139	1097	1023	2081	1060	1515	1486
332.15	0.475	0.403	0.584	0.373	1.0019	1.0801	1.1496	1089	1015	2063	1052	1503	1474
333.05	0.393	0.479	0.493	0.456	0.9943	1.0792	1.2623	1081	1007	2048	1045	1492	1463
333.15	0.302	0.654	0.391	0.591	1.0221	1.0203	1.319	1080	1006	2046	1044	1491	1462
333.15	0.408	0.454	0.513	0.435	0.9925	1.0815	1.1976	1080	1006	2046	1044	1491	1462
333.45	0.410	0.433	0.517	0.421	0.9871	1.0868	1.2414	1078	1004	2041	1041	1487	1458
333.95	0.423	0.371	0.555	0.365	1.0122	1.0840	1.1962	1074	1000	2032	1037	1481	1452
334.15	0.342	0.500	0.451	0.488	1.0100	1.0671	1.1860	1072	998	2029	1036	1479	1450
334.45	0.456	0.290	0.603	0.301	1.0032	1.1234	1.1545	1070	996	2024	1033	1475	1446
334.55	0.358	0.450	0.466	0.456	0.9866	1.0915	1.2371	1069	995	2022	1033	1474	1445
334.75	0.194	0.753	0.270	0.705	1.0471	1.0045	1.4029	1067	994	2019	1031	1472	1443
334.85	0.300	0.531	0.406	0.524	1.0146	1.0552	1.2421	1066	993	2017	1030	1471	1442
334.85	0.350	0.450	0.459	0.461	0.9845	1.0967	1.1916	1066	993	2017	1030	1471	1442
335.15	0.361	0.411	0.485	0.428	0.9983	1.1025	1.1315	1064	991	2012	1028	1467	1438
335.45	0.310	0.472	0.424	0.491	1.0070	1.0916	1.1476	1061	988	2007	1025	1464	1435

335.55	0.265	0.564	0.360	0.569	0.9975	1.0538	1.2178	1061	987	2005	1025	1463	1434
335.55	0.358	0.363	0.501	0.394	1.0266	1.1350	1.1002	1061	987	2005	1025	1463	1434
335.75	0.324	0.421	0.452	0.450	1.0185	1.1079	1.1172	1059	986	2002	1023	1460	1431
336.05	0.569	0.026	0.823	0.031	1.0477	1.2353	1.0336	1057	984	1997	1021	1457	1428
336.65	0.533	0.049	0.780	0.062	1.0404	1.2897	1.0603	1052	979	1987	1016	1450	1421
336.75	0.099	0.781	0.147	0.794	1.0589	1.0223	1.3681	1051	978	1986	1015	1449	1420
336.95	0.041	0.895	0.063	0.900	1.0813	1.0046	1.6003	1050	977	1982	1014	1447	1417
337.05	0.060	0.845	0.092	0.857	1.0676	1.0102	1.5080	1049	976	1981	1013	1445	1416
337.15	0.250	0.461	0.372	0.508	1.0453	1.0948	1.1384	1048	975	1979	1012	1444	1415
337.25	0.089	0.771	0.132	0.796	1.0418	1.0218	1.4121	1047	974	1978	1011	1443	1414
337.35	0.098	0.732	0.148	0.767	1.0468	1.0327	1.3840	1046	974	1976	1011	1442	1413
337.65	0.287	0.378	0.424	0.441	1.0216	1.1402	1.0919	1044	971	1971	1008	1439	1410
337.75	0.117	0.677	0.176	0.726	1.0414	1.0447	1.2756	1043	971	1969	1008	1438	1408
338.15	0.098	0.671	0.153	0.742	1.0676	1.0629	1.2133	1040	968	1963	1005	1433	1404
338.25	0.146	0.587	0.224	0.657	1.0386	1.0729	1.1911	1039	967	1961	1004	1432	1403
338.65	0.391	0.166	0.602	0.219	1.0344	1.2496	1.0562	1036	964	1955	1001	1428	1398
339.15	0.189	0.451	0.295	0.549	1.0337	1.1362	1.1111	1032	960	1947	997	1422	1393
339.25	0.096	0.605	0.158	0.703	1.0855	1.0784	1.1950	1032	959	1946	996	1421	1392
339.55	0.201	0.405	0.333	0.490	1.0829	1.1134	1.1427	1029	957	1941	994	1418	1388
339.65	0.447	0.059	0.707	0.081	1.0311	1.2734	1.0834	1029	956	1939	993	1417	1387
339.75	0.098	0.580	0.160	0.693	1.0594	1.0934	1.1548	1028	956	1938	992	1415	1386
340.45	0.118	0.507	0.194	0.635	1.0490	1.1220	1.1189	1022	951	1927	987	1408	1379
340.45	0.283	0.240	0.466	0.322	1.0477	1.2026	1.0928	1022	951	1927	987	1408	1379
340.55	0.173	0.407	0.294	0.516	1.0794	1.1322	1.1078	1022	950	1925	987	1407	1378
341.15	0.171	0.373	0.295	0.498	1.0754	1.1692	1.0892	1017	945	1916	982	1400	1371
341.45	0.235	0.280	0.397	0.390	1.0492	1.2101	1.0380	1015	943	1911	980	1397	1368
341.85	0.208	0.294	0.359	0.413	1.0569	1.2074	1.0689	1012	940	1905	977	1393	1364
342.65	0.147	0.339	0.275	0.476	1.1244	1.1756	1.1022	1006	935	1893	971	1384	1355

TABLE II (Continued)

					Activity coefficients			*Virial coefficients* $(cm^3\ mol^{-1})$					
T/k	X_1	X_2	Y_1	Y_2	γ_1	γ_2	γ_3	$-B_{12}$	$-B_{13}$	$-B_{23}$	$-B_{11}$	$-B_{22}$	$-B_{33}$
343.65	0.188	0.257	0.341	0.392	1.0552	1.2389	1.0580	999	928	1878	964	1374	1345
344.55	0.295	0.058	0.584	0.104	1.1258	1.4267	1.0276	992	921	1865	958	1365	1335
345.65	0.034	0.401	0.069	0.634	1.1294	1.2076	1.0831	984	914	1849	950	1353	1324
346.25	0.153	0.218	0.309	0.369	1.0978	1.2692	1.0325	980	910	1840	946	1347	1318
346.85	0.140	0.191	0.281	0.343	1.0711	1.3271	1.1107	976	906	1832	942	1341	1312
349.05	0.195	0.080	0.423	0.162	1.0893	1.4024	1.0514	960	891	1801	927	1320	1290
351.75	0.095	0.134	0.239	0.294	1.1791	1.4001	1.0199	942	874	1764	909	1294	1265
356.25	0.064	0.090	0.176	0.223	1.1551	1.3976	1.0366	913	846	1705	881	1252	1223
359.55	0.045	0.065	0.131	0.169	1.1092	1.3532	1.0371	892	827	1664	861	1223	1195

with the activity coefficients γ_i which were calculated from the following equation [4]:

$$\ln \gamma i = \ln\left(\frac{Py_i}{P_i^0 X_i}\right) + \frac{(B_{ii} - V_i^L)(P - P_i^0)}{RT} + \frac{P}{2RT}\sum\sum y_j y_k (2\delta_{ji} - \delta_{jk}) \tag{1}$$

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

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

The standard state for calculation of activity coefficients is the pure component at the pressure and temperature of the solution. The pure component vapor pressures P_i^0 were calculated according to the Antoine equation:

$$\log(P_i^0/kPa) = A_i - \frac{B_i}{(T/K) - C_i} \tag{3}$$

where the Antoine constants A_i, B_i and C_i are reported in Table III. The molar virial coefficients B_{ii} and B_{ij} were estimated by the method of Hayden and O'Connell [5] by assuming the association and solvation parameters to be negligible. The last two terms in Eqn. (1) contributed less than 3 % to the activity coefficients, and their influence was important only at very dilute concentrations. The calculated activity coefficients reported in Table II are estimated accurate to within 3 % and were found to be thermodynamically consistent as tested by the L-W method of Wisniak [6] and the McDermot-Ellis method [7] modified by Wisniak and Tamir [8]. According to these references two experimental points a and b are considered thermodynamically consistent if the following condition is fulfilled:

$$D < D_{\max} \tag{4}$$

TABLE III Antoine Coefficients, Eq. 3

Compound	A_i	B_i	C_i
methyl 1,1-dimethylethyl ether[a]	5.860 78	1032.988	59.876
oxolane[b]	6.121 42	1203.11	46.795
heptane[c]	6.020 23	1263.909	56.718

[a] Reich [16]
[b] TRC Tables, k-1440 [14]
[c] TRC Tables, k-1460 [15]

The local deviation D is given by

$$D = \sum_{i=1}^{N} (x_{ia} + x_{ib})(\ln\gamma_{ia} - \ln\gamma_{ib}) \tag{5}$$

where N is the number of components and the maxium deviation $D_{\max}$ is:

$$\begin{aligned} D_{\max} = {} & \sum_{i=1}^{N} (x_{ia} + x_{ib})\left(\frac{1}{x_{ia}} + \frac{1}{y_{ia}} + \frac{1}{x_{ib}} + \frac{1}{y_{ib}}\right)\Delta x \\ & + \sum_{i=1}^{N} (x_{ia} + x_{ib})\frac{\Delta P}{P} + 2\sum_{i=1}^{N} |\ln\gamma_{ib} - \ln\gamma_{ia}|\Delta x \\ & + \sum_{i=1}^{N} (x_{ia} + x_{ib})B_j\{(T_a + C_j)^{-2} + (T_b + C_j)^{-2}\}\Delta T \end{aligned} \tag{6}$$

The errors in the measurements x, P and T were as previously indicated. The first term in Eqn. (6) was the dominant one. For the experimental points reported here D never exceeded 0.041 while the smallest value of $D_{\max}$ was 0.209.

The activity coefficients for the ternary system were correlated from the following Redlich-Kister expansion [9]:

$$\begin{aligned} \frac{G^E}{RT} = {} & x_1x_2[b_{12} + c_{12}(x_1 - x_2) + d_{12}(x_1 - x_2)^2] \\ & + x_1x_3[b_{13} + c_{13}(x_1 - x_3) + d_{13}(x_1 - x_3)^2] \\ & + x_2x_3[b_{23} + c_{23}(x_2 - x_3) + d_{23}(x_2 - x_3)^2] \\ & + x_1x_2x_3(C + D_1x_1D_2x_2) \end{aligned} \tag{7}$$

TABLE VI Coefficients in Correlation of Boiling Points, eq. 10 and 11, Root Mean Square Deviations in Temperature, rmsd (T/K), and Percent Deviation

A.-Equation 10

System	C_0	C_1	C_2	C_3	*rmsd(T/K)*	*%dev*[a]	*max % dev*[b]
MTBE (1) + oxolane (2)[d]	−5.32981	2.96623	−0.45661	–	0.02	0.1	0.25
MTBE (1) + heptane (3)[e]	46.4787	8.41682	−66.3331	89.8022	0.20	1.3	3.1
oxolane (2) + heptane (3)[f]	−29.0692	16.0822	−10.2034	–	0.03	0.15	0.46
		A	*B*	*C*	*rmsd(T/K)*[a]	*% dev*[b]	*max %dev*[c]
MTBE (1) + oxolane (2) + heptane (3)	−181.9704	−32.6047	–	–	0.4	3.7	9.7

B.-Equation 11

$A_{12} = -1.798$	$B_{12} = -1.351$	$C_{12} = -2.827$	rmsd(T/K) = 0.03
$A_{13} = -30.484$	$B_{13} = 18.213$	$C_{13} = -11.579$	%dev[b] = 0.3
$A_{23} = -29.069$	$B_{23} = 16.507$	$C_{23} = -12.541$	max % dev[c] = 0.8

[a] rmsd (T/K) : Root mean square deviation, $\{\Sigma(T_{\text{expt}} - T_{\text{calc}})^2\}^{0.5}/N$

[b] Average % deviations

[c] Maximum % deviations

[d] Wisniak [1]

[e] Wisniak *et al.* [3]

[f] Wisniak *et al.* [2]

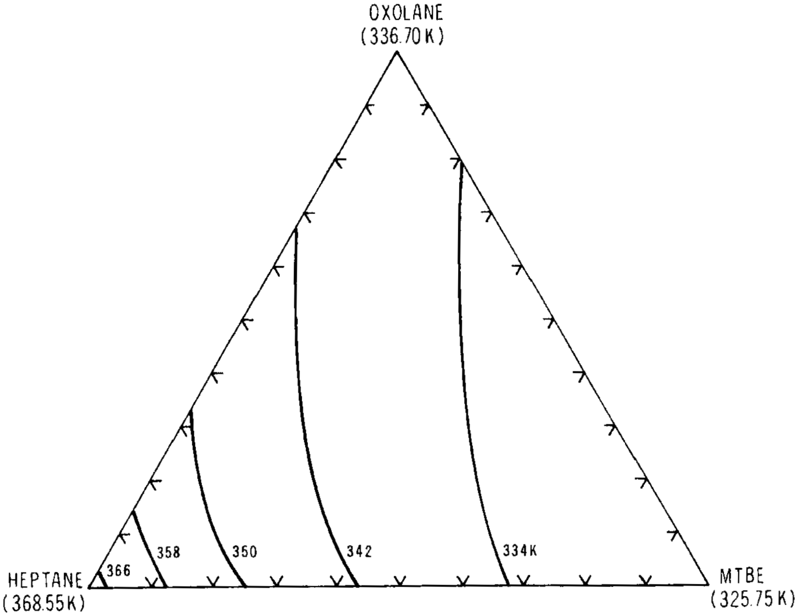

FIGURE 1 Isothermals for the ternary system MTBE (1) + oxolane (2) + heptane (3) at 94 kPa from 344 K to 366 K, every 8 K. Coefficients from eq. 11.

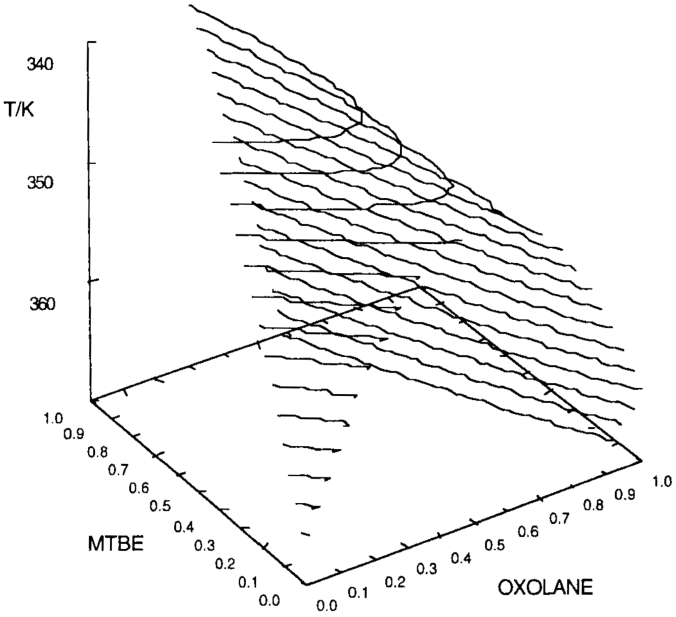

FIGURE 2 Three-dimensional graph T-x_1-x_2.

TABLE V Parameters and prediction statistics for different G^E models

Model	*ij*	A_{ij} / $J\ mol^{-1}$	A_{ji} / $J\ mol^{-1}$	α_{ij}	*Bubble-point pressures*			*Dew-point pressures*		
					$\Delta P(\%)$	y_1	Δy_2	$\Delta P(\%)$	Δx_1	Δx_2
	1–2	56.44	266.13	0.300						
NRTL	1–3	−375.85	932.01	0.352	1.04	0.0046	0.0045	0.90	0.0047	0.0058
	2–3	1165.60	228.94	0.300						
	1–2	−398.92	833.18							
[a]Wilson	1–3	788.24	−291.11	–	0.92	0.0042	0.0036	0.91	0.0043	0.0047
	2–3	1936.96	−362.65							
	1–2	130.81	129.62							
[b]UNIQUAC	1–3	−145.97	319.33	–	1.05	0.0148	0.0230	2.01	0.0109	0.0288
	2–3	117.59	280.03							
[c]UNIFAC	–	–	–	–	1.70	0.0083	0.0059	2.51	0.0090	0.0076

[a] liquid volumes have been estimated from the Rackett equation

[b] molecular parameters are those calculated from UNIFAC

[c] calculations based on modified UNIFAC (Larsen, 1987)

In this equation n is the number of components ($n=2$ or 3), T_i^0 is the boiling point of the pure component i and m is the number of terms in the series expansion of (x_i-x_j). C_k are the binary constants where A, B, C, and D are ternary constants. The following equation, of the same structure, has been suggested by Tamir [13] for the direct correlation of ternary data, without use of binary data:

$$\begin{aligned} T/K = \sum_{i=1}^{3} x_i T_i^0 &+ x_1x_2[A_{12} + B_{12}(x_1 - x_2) + C_{12}(x_1 - x_2)^2 + \cdots] \\ &+ x_1x_3[A_{13} + B_{13}(x_1 - x_3) + C_{13}(x_1 - x_3)^2 + \cdots] \\ &+ x_2x_3[A_{23} + B_{23}(x_2 - x_3) = C_{23}(x_2 - x_3)^2 + \cdots] \end{aligned} \quad (11)$$

In Eqn. (11) coefficients A_{ij}, B_{ij}, and C_{ij} are not binary constants, they are multicomponent parameters determined directly from the data. Direct correlation of $T(x)$ for ternary mixtures can be very efficient as reflected by a lower % average deviation and root mean square deviation (rmsd) and a smaller number of parameters than those for Eqn. (10). Both equations may require similar number of constants for similar accuracy, but the direct correlation allows an easier calculation of boiling isotherms (Figs. 1 and 2). The various constants of Eqns. (10) and (11) are reported in Table VI, which also contains information indicating the degree of goodness of the corrleation. It is clear that for the ternary system in question a direct fit of the data gives a much better fit.

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