Measurement and Correlation of Isobaric Vapor-Liquid Equilibria for Binary and Ternary Systems Containing Methyl Tertiary Butyl Ether (MTBE), Methanol and Alkanes

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Abstract−Isobaric vapor-liquid equilibria (VLE) of four binary systems--methyl tertiary butyl ether (MTBE)+methanol, MTBE+heptane, MTBE+octane and MTBE+i-octane--were measured at atmospheric pressure by using Othmer-type circulation method. The VLE of a ternary system, MTBE+methanol+heptane, were also measured at atmospheric pressure. These VLE data were predicted by ASOG and correlated by Wilson equation, and the prediction and correlation performances were discussed.

Key words: Vapor-Liquid Equilibria, Measurement, Correlation, MTBE, Alkane

INTRODUCTION

Ethers and alcohols used as gasoline additives have been remarked as octane number enhancing and environmental protection agents. The vapor-liquid equilibria (VLE) are required to assess the phase behavior of those mixtures in producing and blending operations. Methyl tertiary butyl ether (MTBE) seems to be a promising ether.

In this study, isobaric VLE data of four binary systems (MTBE+methanol, MTBE+heptane, MTBE+octane and MTBE+i-octane), and of a ternary system (MTBE+methanol+heptane) were measured at atmospheric pressure by using an Othmer-type circulation still. Although a few isobaric VLE data of binary systems containing MTBE have been reported to date in the literature for MTBE+methanol [Aim and Ciprin, 1980; Arce et al., 1996; Toghiani et al., 1996; Komatsu et al., 1997; Loras et al., 1999], for MTBE+heptane [Wisniak et al., 1997a] and for MTBE+octane [Wisniak et al., 1997b; Hiaki et al., 1999], there is no isobaric VLE data for MTBE+i-octane and the present ternary system. The VLE data obtained in this work were predicted by ASOG [Tochigi et al., 1990] and correlated by Wilson equation [Wilson et al., 1964]. Their prediction and correlation performances were discussed.

EXPERIMENTAL

1. Materials

MTBE supplied by Tokyo Kasei Kogyo and methanol, heptane, octane and i-octane supplied by Wako Pure Chemical Industries were special grade reagents. They were used without further purification because no impurities were detected by the gas chromatographic analysis. The purities are estimated to be more than 99.8%, 99%, 99%, 99% and 98% for methanol, MTBE, heptane, i-octane and octane, respectively.

2. Apparatus and Procedure

An all-glass Othmer-type apparatus modified by the author [Watanabe, 1985] was used to measure the VLE relation at atmospheric pressure. The equilibium still is about 160 mL in volume and is equipped with four baffle plates and a propeller agitator to sufficiently mix the liquid mixture. After the liquid mixture was evaporated, liquid drops of the condensed vapor were adjusted to be a proper rate for one hour. Liquid and vapor phase samples were analyzed with a gas chromatograph (Yanagimoto G2800-F) equipped with a flame ionization detector (FID) and an integrator. To determine their compositions, an internal standard method was adopted for calibration of the FID response.

The temperatures were measured with copper- or iron-constantan thermocouples. The uncertainties of the present experiment are believed to be ±0.1 K in temperature measurements. From the reproducibility, the uncertainties of mole fractions reported are estimated to be within ±0.001.

RESULTS AND DISCUSSION

1. Fundamental Equation

VLE relation at low pressure can be calculated based on the following equation when no significant interaction such as association occurs in vapor phase.

$$\pi y_i = \gamma_i x_i p_i^0 \tag{1}$$

Table 1. Antoine constants[a]

Component	Boiling point [K]	Constants			Lit.
		A	B	C	
MTBE	328.3	6.038757	1149.261	−43.150	b
Methanol	337.7	7.025886	1474.078	−44.020	b
Heptane	371.6	6.020230	1263.909	−56.718	c
Octane	398.9	6.043940	1351.938	−64.030	d
i-Octane	372.4	5.936790	1257.840	−53.415	e

[a]$\log(P/kPa) = A - B/[(T/K) + C]$. [b]Arce et al. [1996]. [c]Wisniak et al. [1997a]. [d]Hiaki et al. [1999]. [e]Loras et al. [2000].

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where π is the total pressure, γ_i is the liquid phase activity coefficient, p_i^0 is the vapor pressure of pure component, x_i and y_i are liquid and vapor phase mole fractions, respectively. In this case π= 101.3 kPa and p_i^0 can be calculated by using Antoine equation of which constants are presented in Table 1.

2. VLE Data and Consistency Tests

VLE data of four binary systems at atmospheric pressure are presented in Tables 2-5. The activity coefficients were evaluated by Eq. (1) using the present x-y data and they are given in Tables 2-5. The experimental data were examined by thermodynamic consistency tests: the area test [Herington, 1951] and the point test [Van Ness et al., 1973]. The results are summarized in Table 6. As shown in Table 6, the consistency tests are not satisfied for MTBE+heptane and MTBE+octane. This may be due to the fact that these binary mixtures are almost ideal and therefore activity coefficients are nearly unity [Miyamoto et al., 2001]. The present VLE data of MTBE+methanol and MTBE+octane are compared with the literature data. As shown in Figs. 1 and 2, they are in good agreement though the results of Komatsu et al. [1997] for MTBE+methanol show deviation.

VLE data at atmospheric pressure of a ternary system, MTBE+methanol+heptane, are shown in Table 7. The data were obtained in the homogeneous liquid region. The activity coefficients evalu-

Table 2. Experimental VLE data for MTBE(1)+methanol(2) at atmospheric pressure

T/K	x_1	y_1	γ_1	γ_2
336.7	0.0091	0.055	4.617	0.992
335.6	0.024	0.090	2.998	1.013
334.6	0.037	0.131	2.910	1.021
333.9	0.042	0.150	3.012	1.031
334.1	0.059	0.191	2.688	0.992
332.1	0.085	0.246	2.545	1.031
331.6	0.120	0.311	2.315	1.001
330.2	0.132	0.351	2.501	1.011
329.6	0.174	0.383	2.102	1.037
328.5	0.190	0.422	2.201	1.036
328.5	0.218	0.458	2.082	1.006
327.7	0.250	0.473	1.925	1.055
327.3	0.274	0.471	1.769	1.113
327.5	0.288	0.500	1.776	1.064
326.1	0.313	0.497	1.697	1.178
325.7	0.357	0.546	1.658	1.154
325.8	0.394	0.556	1.524	1.194
325.7	0.438	0.575	1.423	1.237
325.2	0.484	0.592	1.349	1.320
324.7	0.523	0.613	1.313	1.387
324.9	0.575	0.643	1.245	1.421
324.6	0.664	0.672	1.137	1.676
324.2	0.697	0.693	1.133	1.768
324.7	0.725	0.708	1.094	1.814
325.1	0.826	0.760	1.018	2.312
325.2	0.847	0.787	1.024	2.327
325.3	0.884	0.798	0.991	2.900
325.4	0.933	0.891	1.045	2.702

Table 3. Experimental VLE data for MTBE(1)+heptane(2) at atmospheric pressure

T/K	x_1	y_1	γ_1	γ_2
366.6	0.046	0.195	1.405	0.979
365.8	0.056	0.217	1.315	0.986
364.4	0.069	0.233	1.190	1.022
360.0	0.135	0.381	1.110	1.019
354.5	0.216	0.515	1.088	1.051
353.1	0.250	0.554	1.049	1.059
351.0	0.277	0.588	1.064	1.090
349.9	0.299	0.615	1.065	1.089
345.0	0.384	0.704	1.090	1.130
342.0	0.489	0.778	1.036	1.132
341.2	0.502	0.786	1.042	1.156
340.6	0.514	0.795	1.049	1.158
337.4	0.572	0.824	1.076	1.269
336.8	0.588	0.839	1.085	1.233
336.2	0.615	0.846	1.066	1.291
334.4	0.664	0.865	1.067	1.386
335.2	0.674	0.875	1.037	1.286
334.2	0.723	0.899	1.025	1.269
332.5	0.746	0.914	1.066	1.259
332.1	0.816	0.940	1.015	1.225
330.4	0.881	0.967	1.021	1.126
329.7	0.924	0.981	1.009	1.052
330.0	0.934	0.980	0.988	1.267

Table 4. Experimental VLE data for MTBE(1)+octane(2) at atmospheric pressure

T/K	x_1	y_1	γ_1	γ_2
395.0	0.010	0.058	1.031	1.060
396.0	0.011	0.060	0.906	1.030
395.0	0.017	0.097	0.955	1.025
393.4	0.025	0.140	0.985	1.029
388.7	0.056	0.296	1.042	0.997
387.7	0.061	0.305	1.012	1.019
386.1	0.062	0.339	1.147	1.017
385.2	0.077	0.327	0.907	1.082
380.8	0.092	0.440	1.119	1.049
379.4	0.106	0.468	1.076	1.055
377.8	0.114	0.501	1.111	1.052
379.0	0.115	0.463	0.983	1.092
378.8	0.118	0.487	1.016	1.053
377.2	0.118	0.503	1.092	1.071
373.4	0.154	0.590	1.075	1.041
365.9	0.204	0.681	1.127	1.105
366.4	0.204	0.682	1.113	1.086
363.4	0.235	0.721	1.106	1.096
358.8	0.272	0.753	1.123	1.202
357.1	0.299	0.792	1.125	1.116
352.7	0.369	0.840	1.091	1.121
348.7	0.415	0.859	1.108	1.238
343.2	0.505	0.902	1.120	1.263

Table 4. Continued

T/K	x_1	y_1	γ_1	γ_2
340.5	0.587	0.926	1.073	1.270
338.2	0.665	0.949	1.041	1.195
334.9	0.736	0.962	1.054	1.292
333.5	0.797	0.972	1.029	1.295
331.6	0.863	0.983	1.020	1.296
329.7	0.943	0.993	1.002	1.336

Table 5. Experimental VLE data for MTBE(1)+i-octane(2) at atmospheric pressure

T/K	x_1	y_1	γ_1	γ_2
366.6	0.047	0.167	1.169	1.034
361.3	0.114	0.326	1.086	1.054
358.0	0.172	0.454	1.097	1.011
353.6	0.208	0.540	1.213	1.023
354.2	0.226	0.538	1.091	1.033
351.0	0.283	0.632	1.123	0.981
348.9	0.332	0.663	1.065	1.033
347.4	0.359	0.689	1.068	1.046
346.5	0.360	0.688	1.090	1.084
344.8	0.406	0.737	1.086	1.044
345.1	0.408	0.739	1.075	1.028
342.3	0.477	0.778	1.052	1.085
342.2	0.496	0.794	1.036	1.048
341.0	0.499	0.792	1.064	1.112
340.2	0.514	0.807	1.077	1.096
339.8	0.550	0.825	1.042	1.088
339.8	0.568	0.838	1.025	1.050
337.9	0.618	0.862	1.026	1.081
336.8	0.643	0.871	1.030	1.123
334.0	0.753	0.917	1.009	1.164
331.8	0.834	0.950	1.014	1.116
329.6	0.925	0.978	1.009	1.176

Table 6. Thermodynamic consistency tests

System	Area test		Point test	
	D-J	Results	Δy	Results
MTBE+methanol	1.6	+	0.008	+
MTBE+heptane	31.4	−	0.021	−
MTBE+octane	70.1	−	0.021	−
MTBE+i-octane	28.3	−	0.007	+

Criterion of consistency (character: +) $D-J<10$ $\Delta y<0.01$

where $D=100\left|\int_0^1 \log(\gamma_1/\gamma_2)dx_1\right| / \int_0^1 \left|\log(\gamma_1/\gamma_2)\right| dx_1$,

$J=150(T_{max}-T_{min})/T_{min}$, $\Delta y=\frac{1}{N}\sum_k |y_{cal}-y_{exp}|_k$,

N=number of data points.

ated by Eq. (1) are also given in Table 7. The present ternary system does not form a ternary azeotrope, though the minimum boiling point azeotrope is found in MTBE+methanol as shown in Fig. 1.

3. Correlation and Prediction

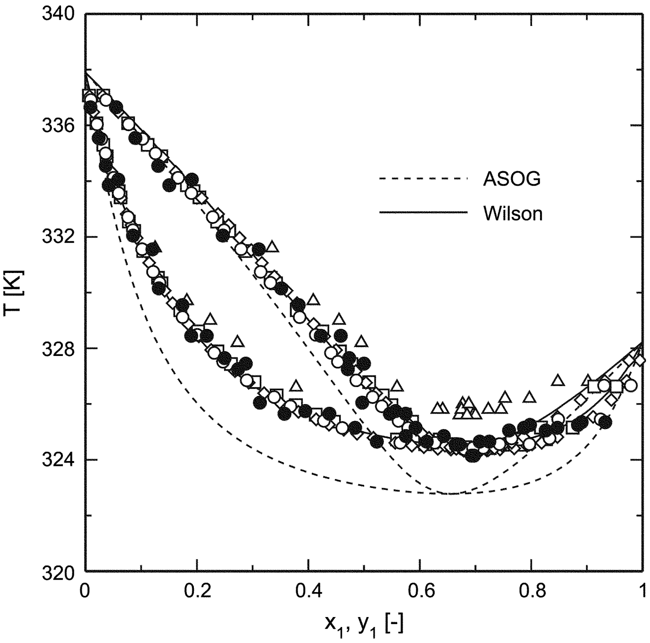

Fig. 1. VLE relation for MTBE(1)+methanol(2) at atmospheric pressure: (●) this work; (○) Aim and Ciprin; (◇) Arce et al., (△) Komatsu et al., (□) Toghiani et al.

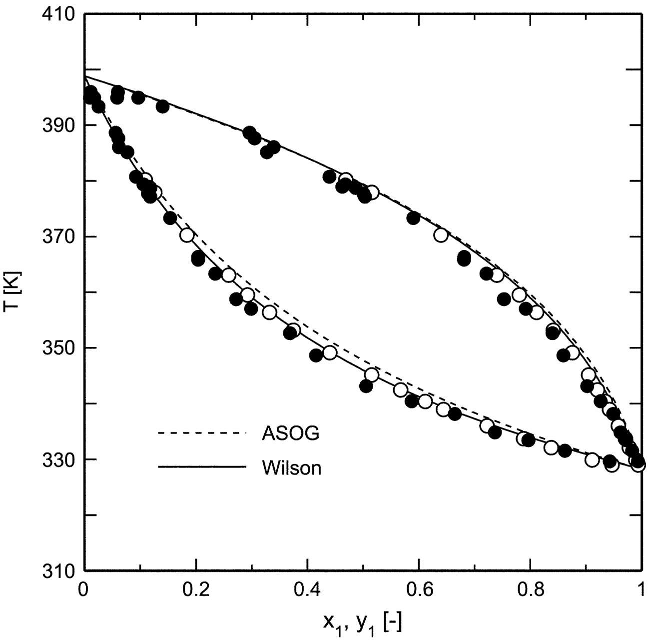

Fig. 2. VLE relation for MTBE(1)+octane(2) at atmospheric pressure: (●) this work; (○) Hiaki et al.

VLE relation x_i~y_i can be obtained by using Eq. (1) when the liquid phase activity coefficients are given by any model. First, we attempted to predict the activity coefficients by ASOG because it is widely used, as well as UNIFAC, in the prediction of phase equilibria for many mixtures. The ASOG parameters required were cited from the literature [Tochigi et al., 1990]. For the binary systems the prediction performances are listed in Table 8 and the comparisons between experimental and predicted results are illustrated in Figs. 1

Table 7. Experimental VLE data for MTBE(1)+methanol(2)+heptane(3) at atmospheric pressure

T/K	x_1	x_2	y_1	y_2	γ_1	γ_2	γ_3
340.1	0.077	0.071	0.105	0.612	0.930	7.883	0.929
330.7	0.087	0.478	0.149	0.667	1.574	1.851	1.681
330.7	0.090	0.841	0.190	0.674	1.942	1.062	7.886
329.7	0.091	0.777	0.159	0.660	1.667	1.174	5.657
333.6	0.096	0.095	0.111	0.654	0.976	8.114	1.032
330.6	0.098	0.587	0.126	0.683	1.186	1.550	2.421
329.9	0.098	0.832	0.207	0.655	1.997	1.078	8.113
330.1	0.100	0.748	0.191	0.644	1.789	1.170	4.441
330.0	0.101	0.816	0.193	0.662	1.797	1.108	7.148
330.7	0.102	0.575	0.113	0.675	1.019	1.557	2.609
330.3	0.105	0.654	0.147	0.662	1.307	1.363	3.209
331.8	0.119	0.312	0.135	0.657	1.011	2.671	1.395
336.0	0.119	0.090	0.146	0.586	0.961	6.960	1.098
330.0	0.120	0.777	0.213	0.644	1.676	1.132	5.642
332.0	0.130	0.194	0.138	0.638	0.938	4.132	1.257
331.6	0.139	0.220	0.141	0.645	0.908	3.749	1.285
329.4	0.143	0.737	0.223	0.640	1.498	1.216	4.764
333.3	0.149	0.144	0.168	0.601	0.952	4.976	1.180
329.7	0.161	0.718	0.249	0.617	1.472	1.188	4.570
329.5	0.166	0.670	0.296	0.570	1.704	1.186	3.413
334.7	0.168	0.063	0.204	0.539	0.985	9.607	1.141
329.5	0.176	0.672	0.258	0.604	1.402	1.252	3.798
329.9	0.189	0.456	0.220	0.604	1.098	1.816	2.034
329.2	0.198	0.592	0.239	0.632	1.169	1.506	2.580
330.4	0.221	0.276	0.236	0.582	0.993	2.834	1.459
329.7	0.231	0.358	0.253	0.574	1.040	2.217	1.745
331.5	0.237	0.140	0.225	0.581	0.853	5.336	1.200
335.1	0.253	0.067	0.316	0.452	1.001	7.485	1.146
328.7	0.270	0.582	0.296	0.623	1.079	1.544	2.335
331.4	0.280	0.158	0.293	0.543	0.940	4.424	1.136
329.5	0.289	0.274	0.294	0.545	0.971	2.778	1.536
329.2	0.303	0.434	0.313	0.564	0.999	1.833	1.972
326.6	0.304	0.642	0.449	0.503	1.551	1.234	4.187
329.2	0.308	0.445	0.342	0.530	1.074	1.682	2.185
328.7	0.313	0.455	0.357	0.520	1.120	1.647	2.279
329.0	0.314	0.346	0.335	0.528	1.038	2.169	1.719
329.2	0.314	0.342	0.332	0.527	1.022	2.178	1.721
330.4	0.332	0.175	0.359	0.477	1.005	3.663	1.337
329.7	0.351	0.368	0.369	0.532	0.998	1.999	1.460
329.0	0.368	0.386	0.391	0.496	1.032	1.832	1.949
332.4	0.374	0.077	0.431	0.393	1.007	6.316	1.192
329.4	0.398	0.219	0.398	0.474	0.960	3.024	1.403
326.5	0.435	0.512	0.514	0.457	1.248	1.412	2.557
327.8	0.448	0.277	0.459	0.438	1.036	2.371	1.662
329.7	0.479	0.166	0.495	0.387	0.983	3.218	1.380
328.5	0.490	0.226	0.490	0.410	0.988	2.639	1.536
325.1	0.503	0.467	0.560	0.408	1.231	1.468	5.206
325.3	0.529	0.407	0.572	0.395	1.188	1.618	2.469
327.7	0.532	0.264	0.529	0.396	1.008	2.255	1.660
330.2	0.535	0.086	0.582	0.292	1.018	4.584	1.353
328.5	0.568	0.166	0.589	0.329	1.025	2.886	1.338
327.3	0.614	0.187	0.605	0.328	1.014	2.684	1.516
328.8	0.661	0.089	0.696	0.222	1.031	3.571	1.416
325.8	0.727	0.231	0.705	0.280	1.047	1.969	1.841
326.7	0.769	0.088	0.753	0.201	1.028	3.581	1.491

Table 8. Prediction and correlation performances for binary systems

System	N	ASOG		Wilson	
		ΔT [K]	Δy_1 [-]	ΔT [K]	Δy_1 [-]
MTBE+methanol	28	1.8	0.034	0.3	0.009
MTBE+heptane	23	1.9	0.015	0.9	0.012
MTBE+octane	29	2.1	0.010	1.0	0.013
MTBE+i-octane	22	1.9	0.011	0.4	0.006

$\Delta T=\frac{1}{N}\sum_k |T_{cal}-T_{exp}|_k,\ \Delta y_1=\frac{1}{N}\sum_k |y_{1,cal}-y_{1,exp}|_k,$

N=number of data points.

and 2 for example. As shown in Table 8 and Figs. 1 and 2, the agreement seems to be insufficient. The ASOG parameters should be re-evaluated in the future work.

Then VLE relations for the binary systems were correlated by using the Wilson equation:

$$\ln\gamma_1=-\ln(x_1+\Lambda_{12}x_2)+x_2\left(\frac{\Lambda_{12}}{x_1+\Lambda_{12}x_2}-\frac{\Lambda_{21}}{\Lambda_{21}x_1+x_2}\right) \tag{2}$$

$$\ln\gamma_2=-\ln(\Lambda_{21}x_1+x_2)-x_1\left(\frac{\Lambda_{12}}{x_1+\Lambda_{12}x_2}-\frac{\Lambda_{21}}{\Lambda_{21}x_1+x_2}\right) \tag{3}$$

where Λ_{12} and Λ_{21} are the parameters adjusted by using experimental VLE data. The binary parameters determined by using present VLE data are in Table 9. The correlation performances of Wilson equation presented in Table 8 and Figs. 1 and 2 for binary systems. A good agreement between experiment and correlation is obtained. An advantage of Wilson equation is that the VLE relation for a multicomponent system can be calculated with the binary parameters of the constituent binary systems. VLE of the present ternary system, MTBE+methanol+heptane, were calculated by using the binary parameters given in Table 9. The correlation performance is presented in Table 10 in which the prediction performance of ASOG is also given for comparison. Comparisons of the experimental data with the correlated values for typical tie-lines are shown in Fig. 3. As shown in Table 10 and Fig. 3, a fairly good agreement is ob-

Table 9. Wilson parameters

System	Λ_{12} [-]	Λ_{21} [-]
MTBE(1)+methanol(2)	0.50044	0.48033
MTBE(1)+heptane(2)	0.42008	1.49185
MTBE(1)+octane(2)	2.08019	0.21056
MTBE(1)+i-octane(2)	0.88652	0.92845

Table 10. Prediction and correlation performances for MTBE(1)+methanol(2)+heptane(3)

Model	ΔT [K]	Δy_1 [-]	Δy_2 [-]	Δy_3 [-]
ASOG	1.1	0.020	0.026	0.013
Wilson	1.0	0.009	0.017	0.014

$\Delta T=\frac{1}{N}\sum_k |T_{cal}-T_{exp}|_k,\ \Delta y_i=\frac{1}{N}\sum_k |y_{i,cal}-y_{i,exp}|_k,$ (i=1, 2, 3),

N=number of data points.

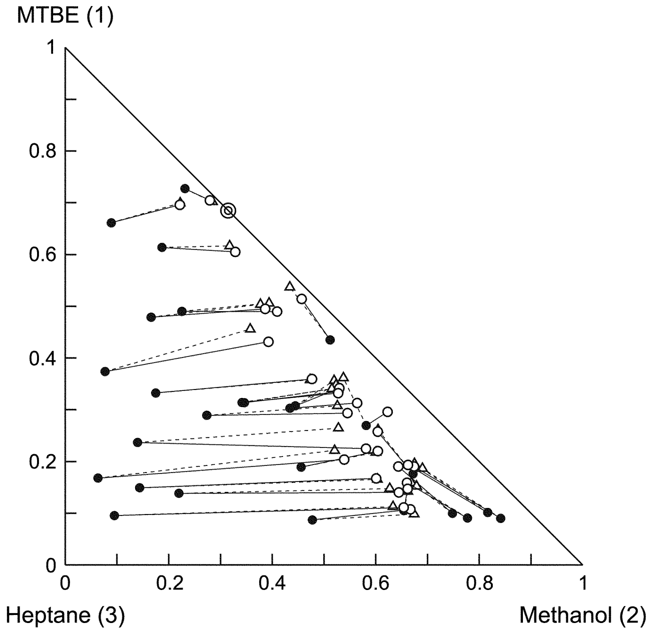

Fig. 3. Tie-lines for MTBE(1)+methanol(2)+heptane(3) at atmospheric pressure: (●) experimental data of liquid phase; (○) experimental data of vapor phase; (△) predicted values of vapor phase by Wilson equation; (◎) azeotrope.

tained.

CONCLUSION

Isobaric VLE relations of binary and ternary systems containing MTBE were measured at atmospheric pressure by using a modified Othmer-type circulation apparatus. The data are reported for four binary systems--MTBE+methanol, MTBE+heptane, MTBE+octane and MTBE+i-octane, and also for a ternary system--MTBE+methanol+heptane. For the present data, the prediction performance of ASOG and the correlation performance of Wilson equation were discussed. It is noted that ASOG parameters should be re-evaluated in the future work to give better prediction results. The Wilson equation can be adopted to correlate VLE of the present binary and ternary systems.

NOMENCLATURE

p_i^0 : vapor pressure of pure component i [Pa]
x_i : liquid phase mole fraction of component i [-]
y_i : vapor phase mole fraction of component i [-]

Greek Letters

γ_i : liquid phase activity coefficient of component i [-]
Λ_{12}, Λ_{21} : Wilson interaction parameters between components 1 and 2 for binary system [-]
π : total pressure [Pa]

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