Vapor–Liquid Equilibrium for the Systems 2-Methylpropane + Methanol, + 2-Propanol, + 2-Butanol, and + 2-Methyl-2-propanol at 364.5 K

Helena Laavi,* Petri Uusi-Kyyny, Juha-Pekka Pokki, Minna Pakkanen, and Ville Alopaeus

The Department of Biotechnology and Chemical Technology, Helsinki University of Technology, P.O. Box 6100, FIN-02015 TKK, Finland

Vapor–liquid equilibrium (VLE) data were measured using a static total pressure apparatus for binary pairs of 2-methylpropane (isobutane) + methanol, + 2-propanol, + 2-butanol, and + 2-methyl-2-propanol (TBA) at 364.51 K. The measured T, p, z data were fitted with Legendre polynomials and reduced by using the method of Barker to obtain the phase equilibrium data. In addition to Legendre polynomials, Wilson binary interaction parameters for the activity coefficient model were regressed. Azeotropic behavior was observed for the binary 2-methylpropane (1) + methanol ($x_1 = 0.871$, $p = 1822$ kPa).

Introduction

Knowledge of vapor–liquid equilibria (VLE) is a necessity for the process industry. VLE data are needed not only in developing new and improving old processes but also in process operation. Thermodynamic models relay on measured VLE data and are the basis of accurate modeling. Therefore, a solid database of accurate experimental VLE data is required. The parameters of the thermodynamic models used for process modeling are regressed from experimental data. An accurate thermodynamic model is fundamental in order to achieve a model of the process that is as close to reality as possible. Especially in the case of systems that may form azeotropic mixtures, it is crucial to predict VLE correctly. Azeotropic distillation processes serve as real examples because in these processes the accurate knowledge of VLE is exploited to achieve the successful separation of an azeotropic mixture. Due to the complexity of the process, it is clear how crucial the accuracy and availability of VLE data are in order to design, operate, and model such a process.

In this work, VLE data for the systems of isobutane + methanol, + 2-propanol, + 2-butanol, and + TBA at 364.5 K were measured by a static total pressure apparatus. The same type of apparatus was used in the previously published VLE data for isobutene + methanol, + 2-propanol, + 2-butanol, and + TBA at 313.15 K.[1] VLE data have been measured by Leu et al.[2] for the system isobutane + methanol in a variable-volume VLE cell in the temperature range (273 to 423) K and for the system isobutane + 2-propanol by Zabaloy et al.[3] in a static apparatus in the temperature range (320 to 390) K. Miyano et al. have measured Henry's constants and infinite dilution activity coefficients by a gas stripping method, for the system isobutane + methanol in the temperature range (255 to 320) K[4] and in the range (374 to 490) K.[5] For the systems isobutane + 2-propanol[6] + 2-butanol[7] and + TBA,[8] the reported temperature ranges were (250 to 330) K. Excess enthalpies have been measured for the isobutane + 2-propanol system at 298.15 K and at 325.15 K at the pressures of (5, 10, and 15) MPa[9] by using a high-temperature, high-pressure flow calorimeter. The azeotropic behavior of the isobutane + methanol binary system has been observed by Ouni et al.[1] and by Leu and Robinson.[2]

* Corresponding author. Tel.: +358-9-451 2644. Fax: +358-9-451 2694. E-mail address: helena.laavi@tkk.fi.

Table 1. Material Purities and Their Suppliers

component	supplier	purity
isobutane	Oy Aga Ab	99.95 %
methanol	Merck	99.8 %
2-propanol	Riedel-de Haën	99.8 %
2-butanol	Fluka	>99.8 %
TBA	Fluka	>99.7 %

Table 2. Measured Pure Component Vapor Pressures vs Literature Correlations

		p/kPa			
component	T/K	this work	[a]	[b]	[c]
isobutane	364.50	1685.6	1683.0	1685.1	1683.0
	364.51	1686.3	1683.3	1685.4	1683.3
	364.51	1686.4	1683.3	1685.4	1683.3
	364.52	1686.4	1683.6	1685.8	1683.7
methanol	364.51	267.5	266.9	267.4	266.8
2-propanol	364.51	144.2	142.2	142.7	142.2
2-butanol	364.51	74.1	73.7[d]	74.9	75.0
TBA	364.50	142.6	142.3[d]	143.0	139.1

[a] Ref 12. [b] Ref 13. [c] Ref 14. [d] Ref 15.

Table 3. Pure Component Physical Properties: Critical Temperature T_c, Critical Pressure p_c, and Acentric Factor ω[14]

component	T_c/K	p_c/MPa	ω
isobutane	408.14 ± 4.08	3.65 ± 0.04	0.1770
methanol	512.58 ± 5.13	8.10 ± 0.24	0.5656
2-propanol	508.31 ± 5.08	4.76 ± 0.14	0.6689
2-butanol	536.01 ± 5.36	4.19 ± 0.13	0.5711
TBA	506.20 ± 5.06	3.97 ± 0.12	0.6158

Experimental Section

Materials. Purities and suppliers of materials are presented in Table 1. The alcohols were dried over Merck 3A molecular sieves for at least 24 h prior to degassing. The degassing was performed by vacuum rectification[10] with modifications.[11] Isobutane was degassed by evacuation in a syringe pump. The vacuum line was opened 10 times in a 10 s period. The quality of the degassing was checked by comparing the measured vapor pressures to literature correlations as shown in Table 2.

10.1021/je800021r CCC: $40.75
Published on Web 03/15/2008

Table 4. VLE Data for Isobutane (1) and Methanol (2) at 364.5 K[a]

T/K	n_1/mol	n_2/mol	z_1	p_{exptl}/kPa	p_{Leg}/kPa	x_1	y_1	γ_1	γ_2
364.51	0.5962 ± 0.0063	0.0000 ± 0.0000	1.0000 ± 0.0000	1686.4	1686.4	1.0000 ± 0.0000	1.0000 ± 0.0000	1.00 ± 0.00	11.18 ± 0.24
364.52	0.5962 ± 0.0063	0.0062 ± 0.0006	0.9897 ± 0.0010	1721.4	1718.2	0.9902 ± 0.0010	0.9795 ± 0.0017	1.00 ± 0.00	10.41 ± 0.26
364.51	0.5962 ± 0.0063	0.0115 ± 0.0006	0.9811 ± 0.0012	1746.9	1741.0	0.9821 ± 0.0012	0.9648 ± 0.0016	1.00 ± 0.00	9.83 ± 0.23
364.51	0.5962 ± 0.0063	0.0187 ± 0.0007	0.9696 ± 0.0014	1773.8	1766.1	0.9709 ± 0.0014	0.9475 ± 0.0015	1.00 ± 0.00	9.10 ± 0.20
364.51	0.5962 ± 0.0063	0.0312 ± 0.0008	0.9503 ± 0.0017	1802.5	1795.4	0.9518 ± 0.0017	0.9240 ± 0.0015	1.01 ± 0.00	8.05 ± 0.15
364.50	0.5962 ± 0.0063	0.0663 ± 0.0012	0.8999 ± 0.0025	1822.4	1824.4	0.9007 ± 0.0026	0.8845 ± 0.0018	1.03 ± 0.00	6.00 ± 0.08
364.50	0.5962 ± 0.0063	0.1050 ± 0.0016	0.8503 ± 0.0032	1819.7	1824.7	0.8497 ± 0.0034	0.8637 ± 0.0021	1.07 ± 0.00	4.69 ± 0.05
364.51	0.5962 ± 0.0063	0.1493 ± 0.0020	0.7997 ± 0.0038	1811.7	1815.8	0.7976 ± 0.0040	0.8516 ± 0.0021	1.12 ± 0.00	3.78 ± 0.03
364.50	0.5962 ± 0.0063	0.1982 ± 0.0025	0.7505 ± 0.0043	1802.4	1804.9	0.7473 ± 0.0046	0.8446 ± 0.0020	1.18 ± 0.00	3.15 ± 0.03
364.51	0.5962 ± 0.0063	0.2553 ± 0.0031	0.7002 ± 0.0048	1792.9	1793.8	0.6962 ± 0.0050	0.8400 ± 0.0018	1.25 ± 0.01	2.69 ± 0.02
364.51	0.5962 ± 0.0063	0.3226 ± 0.0038	0.6489 ± 0.0051	1783.3	1783.3	0.6446 ± 0.0053	0.8369 ± 0.0015	1.34 ± 0.01	2.34 ± 0.02
364.51	0.5962 ± 0.0063	0.3989 ± 0.0045	0.5992 ± 0.0053	1773.8	1773.5	0.5950 ± 0.0055	0.8347 ± 0.0013	1.45 ± 0.01	2.07 ± 0.02
364.51	0.5962 ± 0.0063	0.4901 ± 0.0055	0.5488 ± 0.0054	1763.7	1763.5	0.5453 ± 0.0056	0.8329 ± 0.0011	1.57 ± 0.02	1.86 ± 0.01
364.51	0.5962 ± 0.0063	0.5978 ± 0.0066	0.4993 ± 0.0054	1752.5	1752.7	0.4969 ± 0.0056	0.8312 ± 0.0010	1.71 ± 0.02	1.69 ± 0.01
364.51	0.5951 ± 0.0063	0.6063 ± 0.0067	0.4953 ± 0.0054	1753.0	1752.4	0.4930 ± 0.0056	0.8311 ± 0.0010	1.73 ± 0.02	1.68 ± 0.01
364.51	0.4868 ± 0.0052	0.6063 ± 0.0067	0.4453 ± 0.0054	1737.1	1737.2	0.4378 ± 0.0056	0.8290 ± 0.0010	1.93 ± 0.03	1.52 ± 0.01
364.51	0.3963 ± 0.0043	0.6063 ± 0.0067	0.3953 ± 0.0052	1716.7	1716.6	0.3815 ± 0.0054	0.8265 ± 0.0011	2.19 ± 0.03	1.39 ± 0.01
364.51	0.3202 ± 0.0035	0.6063 ± 0.0067	0.3456 ± 0.0050	1686.6	1686.1	0.3249 ± 0.0051	0.8229 ± 0.0012	2.53 ± 0.04	1.29 ± 0.00
364.52	0.2548 ± 0.0028	0.6063 ± 0.0067	0.2959 ± 0.0046	1638.9	1638.1	0.2681 ± 0.0047	0.8176 ± 0.0014	2.99 ± 0.05	1.20 ± 0.00
364.52	0.1985 ± 0.0022	0.6063 ± 0.0067	0.2467 ± 0.0042	1563.0	1562.1	0.2128 ± 0.0041	0.8091 ± 0.0014	3.61 ± 0.06	1.13 ± 0.00
364.52	0.1502 ± 0.0017	0.6063 ± 0.0067	0.1986 ± 0.0036	1445.0	1444.8	0.1612 ± 0.0035	0.7951 ± 0.0013	4.44 ± 0.09	1.08 ± 0.00
364.51	0.1076 ± 0.0013	0.6063 ± 0.0067	0.1507 ± 0.0030	1269.1	1270.0	0.1139 ± 0.0028	0.7700 ± 0.0010	5.54 ± 0.14	1.04 ± 0.00
364.51	0.0701 ± 0.0009	0.6063 ± 0.0067	0.1036 ± 0.0023	1029.7	1031.0	0.0726 ± 0.0020	0.7223 ± 0.0008	6.94 ± 0.21	1.02 ± 0.00
364.52	0.0360 ± 0.0006	0.6063 ± 0.0067	0.0561 ± 0.0014	719.3	718.9	0.0363 ± 0.0012	0.6126 ± 0.0009	8.71 ± 0.32	1.00 ± 0.00
364.52	0.0228 ± 0.0004	0.6063 ± 0.0067	0.0363 ± 0.0011	569.7	569.4	0.0227 ± 0.0009	0.5174 ± 0.0012	9.56 ± 0.37	1.00 ± 0.00
364.51	0.0111 ± 0.0003	0.6063 ± 0.0067	0.0180 ± 0.0007	421.8	421.6	0.0110 ± 0.0006	0.3567 ± 0.0033	10.41 ± 0.42	1.00 ± 0.00
364.51	0.0000 ± 0.0000	0.6063 ± 0.0067	0.0000 ± 0.0000	267.5	267.5	0.0000 ± 0.0000	0.0000 ± 0.0000	11.30 ± 0.46	1.00 ± 0.00

[a] T is the experimental temperature; n_i is the amount of component in the cell; z_1 is the total mole fraction; p_{exptl} is the measured pressure and p_{Leg} is the pressure calculated from the Legendre polynomial fit; x_1 and y_1 are the liquid and vapor phase equilibrium mole fractions, respectively; γ_1 and γ_2 are the liquid activity coefficients.

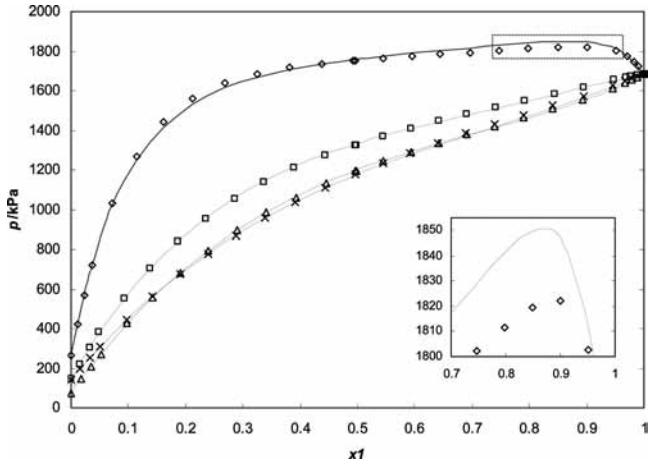

Figure 1. Pressure composition diagram of isobutane (1) ◇, + methanol; □, + 2-propanol; △, + 2-butanol; and ×, TBA. The resulting curves of Legendre polynomials are shown in lines, and the azeotropic point with methanol is shown enlarged.

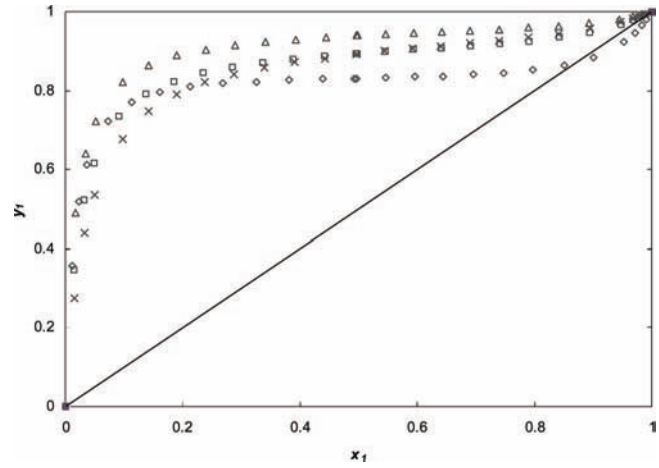

Figure 2. Composition diagram of isobutane (1) ◇, + methanol; □, + 2-propanol; △, + 2-butanol; and ×, + TBA. The method of Barker data reduction is used to obtain equilibrium data from total pressure data.

Apparatus. The measurements were performed by using a static total pressure apparatus. The apparatus consisted of an equilibrium cell, which was equipped with a magnetic stirrer and submersed into a water bath. The volume of the cell was 112.68 cm^3. Further details of the apparatus are explained in Uusi-Kyyny et al.[16] with the modifications by Hynynen et al.[17]

Procedure. The details of the experimental procedure are explained in Ouni et al.,[1] and thus only a short overview is presented here. For each data point, the temperature (T), the total pressure (p), and the volumes of injected components were recorded. From this data, the injected moles of component i (n_i) could be calculated. The measurement of each binary pair was divided into two parts. First, isobutane was introduced into the cell, and alcohol was then gradually added until a mole fraction of about 0.5 was reached. Then, the cell was emptied and cleaned. In the second part, the alcohol was introduced into the cell, and isobutane was gradually added until the same target mole fraction of 0.5 was achieved. The coinciding of the measured vapor pressures at equimolar composition when approaching it from both ends of the composition scale was considered as an indication of good data quality.

Theoretical Calculations

Data Reduction. The measured p, T, n_1, and n_2 data were reduced into phase equilibrium data through the method of Barker[18] by using the VLEFIT software.[19] By equating fugacities of vapor and liquid phases, eq 1 can be derived.

$$\frac{y_i}{x_i} = \frac{\gamma_i \phi_i^S p_i^S}{\phi_i p} \exp \int_{p_i^S}^{p} \frac{V_i^L}{RT} dp \qquad (1)$$

In the data reduction, in order to calculate the liquid (x_i) and vapor (y_i) compositions, the Legendre polynomials[20] were

Table 5. VLE Data for Isobutane (1) and 2-Propanol (2) at 364.5 K[a]

T/K	n_1/mol	n_2/mol	z_1	p_{exptl}/kPa	p_{Leg}/kPa	x_1	y_1	γ_1	γ_2
364.51	0.4849 ± 0.0052	0.0000 ± 0.0000	1.0000 ± 0.0000	1686.3	1686.3	1.0000 ± 0.0000	1.0000 ± 0.0000	1.00 ± 0.00	6.56 ± 0.18
364.51	0.4849 ± 0.0052	0.0053 ± 0.0003	0.9893 ± 0.0007	1680.7	1681.9	0.9891 ± 0.0008	0.9909 ± 0.0008	1.00 ± 0.00	6.12 ± 0.12
364.51	0.4849 ± 0.0052	0.0109 ± 0.0004	0.9780 ± 0.0010	1674.4	1676.2	0.9776 ± 0.0010	0.9825 ± 0.0010	1.00 ± 0.00	5.70 ± 0.09
364.50	0.4849 ± 0.0052	0.0151 ± 0.0004	0.9698 ± 0.0011	1669.2	1671.3	0.9691 ± 0.0012	0.9770 ± 0.0011	1.00 ± 0.00	5.42 ± 0.07
364.50	0.4849 ± 0.0052	0.0262 ± 0.0005	0.9488 ± 0.0015	1654.8	1657.5	0.9474 ± 0.0016	0.9652 ± 0.0013	1.01 ± 0.00	4.80 ± 0.04
364.51	0.4849 ± 0.0052	0.0552 ± 0.0008	0.8977 ± 0.0023	1619.5	1620.6	0.8942 ± 0.0026	0.9454 ± 0.0014	1.03 ± 0.00	3.71 ± 0.01
364.51	0.4849 ± 0.0052	0.0863 ± 0.0011	0.8490 ± 0.0030	1585.4	1585.0	0.8435 ± 0.0033	0.9335 ± 0.0014	1.06 ± 0.00	3.02 ± 0.02
364.52	0.4849 ± 0.0052	0.1223 ± 0.0015	0.7986 ± 0.0037	1551.3	1549.9	0.7918 ± 0.0040	0.9249 ± 0.0014	1.10 ± 0.00	2.54 ± 0.01
364.51	0.4849 ± 0.0052	0.1621 ± 0.0019	0.7494 ± 0.0042	1517.2	1516.1	0.7419 ± 0.0045	0.9184 ± 0.0015	1.15 ± 0.01	2.20 ± 0.01
364.51	0.4849 ± 0.0052	0.2088 ± 0.0024	0.6990 ± 0.0046	1482.4	1481.6	0.6915 ± 0.0050	0.9128 ± 0.0016	1.21 ± 0.01	1.95 ± 0.01
364.51	0.4849 ± 0.0052	0.2622 ± 0.0029	0.6490 ± 0.0050	1446.6	1446.4	0.6421 ± 0.0052	0.9078 ± 0.0016	1.28 ± 0.01	1.76 ± 0.00
364.51	0.4849 ± 0.0052	0.3234 ± 0.0035	0.5999 ± 0.0052	1409.7	1410.0	0.5940 ± 0.0054	0.9030 ± 0.0016	1.35 ± 0.01	1.61 ± 0.00
364.52	0.4849 ± 0.0052	0.3972 ± 0.0043	0.5498 ± 0.0053	1369.4	1370.1	0.5454 ± 0.0055	0.8981 ± 0.0015	1.43 ± 0.01	1.49 ± 0.00
364.51	0.4849 ± 0.0052	0.4853 ± 0.0052	0.4998 ± 0.0053	1325.0	1326.6	0.4975 ± 0.0054	0.8930 ± 0.0015	1.52 ± 0.02	1.39 ± 0.00
364.51	0.4836 ± 0.0052	0.4877 ± 0.0052	0.4979 ± 0.0053	1326.2	1325.6	0.4955 ± 0.0054	0.8928 ± 0.0015	1.53 ± 0.02	1.38 ± 0.00
364.51	0.3971 ± 0.0043	0.4877 ± 0.0052	0.4488 ± 0.0053	1272.5	1272.0	0.4429 ± 0.0054	0.8865 ± 0.0014	1.64 ± 0.02	1.30 ± 0.00
364.51	0.3244 ± 0.0035	0.4877 ± 0.0052	0.3995 ± 0.0052	1210.0	1209.7	0.3898 ± 0.0053	0.8792 ± 0.0014	1.78 ± 0.02	1.22 ± 0.00
364.52	0.2634 ± 0.0029	0.4877 ± 0.0052	0.3507 ± 0.0050	1138.2	1137.9	0.3375 ± 0.0050	0.8704 ± 0.0014	1.95 ± 0.03	1.16 ± 0.00
364.51	0.2115 ± 0.0024	0.4877 ± 0.0052	0.3025 ± 0.0046	1054.7	1054.5	0.2862 ± 0.0046	0.8592 ± 0.0015	2.13 ± 0.03	1.12 ± 0.00
364.51	0.1654 ± 0.0019	0.4877 ± 0.0052	0.2533 ± 0.0042	954.2	954.4	0.2348 ± 0.0042	0.8438 ± 0.0016	2.36 ± 0.04	1.08 ± 0.00
364.50	0.1260 ± 0.0015	0.4877 ± 0.0052	0.2053 ± 0.0037	840.2	840.2	0.1860 ± 0.0036	0.8225 ± 0.0017	2.61 ± 0.05	1.05 ± 0.00
364.51	0.0906 ± 0.0011	0.4877 ± 0.0052	0.1566 ± 0.0031	705.9	706.0	0.1382 ± 0.0029	0.7893 ± 0.0018	2.90 ± 0.06	1.03 ± 0.00
364.50	0.0591 ± 0.0008	0.4877 ± 0.0052	0.1082 ± 0.0024	553.3	553.3	0.0927 ± 0.0022	0.7327 ± 0.0016	3.24 ± 0.08	1.01 ± 0.00
364.51	0.0308 ± 0.0005	0.4877 ± 0.0052	0.0595 ± 0.0015	380.4	380.6	0.0494 ± 0.0014	0.6149 ± 0.0014	3.62 ± 0.11	1.00 ± 0.00
364.51	0.0200 ± 0.0004	0.4877 ± 0.0052	0.0394 ± 0.0012	304.2	303.8	0.0323 ± 0.0011	0.5199 ± 0.0015	3.80 ± 0.13	1.00 ± 0.00
364.52	0.0094 ± 0.0003	0.4877 ± 0.0052	0.0188 ± 0.0008	221.8	222.0	0.0152 ± 0.0007	0.3467 ± 0.0033	3.98 ± 0.16	1.00 ± 0.00
364.51	0.0000 ± 0.0000	0.4877 ± 0.0052	0.0000 ± 0.0000	144.2	144.2	0.0000 ± 0.0000	0.0000 ± 0.0000	4.16 ± 0.18	1.00 ± 0.00

[a] T is the experimental temperature; n_i is the amount of component in the cell; z_1 is the total mole fraction; p_{exptl} is the measured pressure and p_{Leg} is the pressure calculated from the Legendre polynomial fit; x_1 and y_1 are the liquid and vapor phase equilibrium mole fractions, respectively; γ_1 and γ_2 are the liquid activity coefficients.

Table 6. VLE Data for Isobutane (1) and 2-Butanol (2) at 364.5 K[a]

T/K	n_1/mol	n_2/mol	z_1	p_{exptl}/kPa	p_{Leg}/kPa	x_1	y_1	γ_1	γ_2
364.52	0.4508 ± 0.0048	0.0000 ± 0.0000	1.0000 ± 0.0000	1686.3	1686.3	1.0000 ± 0.0000	1.0000 ± 0.0000	1.00 ± 0.00	5.86 ± 0.24
364.51	0.4508 ± 0.0048	0.0047 ± 0.0003	0.9896 ± 0.0007	1669.7	1670.6	0.9890 ± 0.0007	0.9952 ± 0.0005	1.00 ± 0.00	5.46 ± 0.17
364.51	0.4508 ± 0.0048	0.0095 ± 0.0003	0.9794 ± 0.0009	1653.8	1655.5	0.9782 ± 0.0010	0.9910 ± 0.0007	1.00 ± 0.00	5.11 ± 0.13
364.52	0.4508 ± 0.0048	0.0142 ± 0.0004	0.9695 ± 0.0011	1639.0	1641.2	0.9676 ± 0.0012	0.9875 ± 0.0008	1.00 ± 0.00	4.80 ± 0.10
364.52	0.4508 ± 0.0048	0.0247 ± 0.0005	0.9480 ± 0.0015	1609.8	1612.2	0.9447 ± 0.0016	0.9810 ± 0.0009	1.01 ± 0.00	4.23 ± 0.06
364.51	0.4508 ± 0.0048	0.0507 ± 0.0007	0.8989 ± 0.0023	1554.2	1555.1	0.8929 ± 0.0026	0.9710 ± 0.0010	1.03 ± 0.00	3.31 ± 0.01
364.51	0.4508 ± 0.0048	0.0800 ± 0.0010	0.8493 ± 0.0030	1507.2	1506.5	0.8414 ± 0.0034	0.9644 ± 0.0010	1.06 ± 0.00	2.70 ± 0.01
364.51	0.4508 ± 0.0048	0.1133 ± 0.0014	0.7991 ± 0.0036	1463.5	1462.4	0.7900 ± 0.0040	0.9597 ± 0.0010	1.10 ± 0.01	2.29 ± 0.01
364.52	0.4508 ± 0.0048	0.1507 ± 0.0017	0.7495 ± 0.0042	1421.9	1421.0	0.7399 ± 0.0046	0.9559 ± 0.0011	1.15 ± 0.01	2.00 ± 0.00
364.51	0.4508 ± 0.0048	0.1937 ± 0.0022	0.6995 ± 0.0046	1379.9	1379.6	0.6903 ± 0.0049	0.9525 ± 0.0011	1.20 ± 0.01	1.79 ± 0.00
364.51	0.4508 ± 0.0048	0.2432 ± 0.0027	0.6496 ± 0.0049	1336.9	1337.2	0.6414 ± 0.0052	0.9493 ± 0.0012	1.26 ± 0.01	1.62 ± 0.00
364.51	0.4508 ± 0.0048	0.3008 ± 0.0033	0.5998 ± 0.0052	1291.9	1292.7	0.5931 ± 0.0054	0.9461 ± 0.0012	1.33 ± 0.01	1.50 ± 0.00
364.52	0.4508 ± 0.0048	0.3689 ± 0.0040	0.5500 ± 0.0053	1244.3	1245.2	0.5453 ± 0.0054	0.9428 ± 0.0012	1.40 ± 0.01	1.40 ± 0.00
364.51	0.4508 ± 0.0048	0.4500 ± 0.0048	0.5005 ± 0.0053	1192.8	1194.1	0.4981 ± 0.0054	0.9393 ± 0.0012	1.48 ± 0.02	1.32 ± 0.00
364.51	0.4511 ± 0.0048	0.4527 ± 0.0048	0.4991 ± 0.0053	1193.9	1193.3	0.4968 ± 0.0054	0.9392 ± 0.0012	1.48 ± 0.02	1.31 ± 0.00
364.51	0.3709 ± 0.0040	0.4527 ± 0.0048	0.4503 ± 0.0053	1132.0	1131.2	0.4447 ± 0.0054	0.9348 ± 0.0012	1.58 ± 0.02	1.24 ± 0.00
364.51	0.3034 ± 0.0033	0.4527 ± 0.0048	0.4013 ± 0.0052	1061.7	1061.1	0.3924 ± 0.0052	0.9295 ± 0.0012	1.69 ± 0.02	1.18 ± 0.00
364.52	0.2466 ± 0.0027	0.4527 ± 0.0048	0.3527 ± 0.0050	983.4	983.0	0.3407 ± 0.0050	0.9232 ± 0.0012	1.82 ± 0.03	1.13 ± 0.00
364.52	0.1973 ± 0.0022	0.4527 ± 0.0048	0.3036 ± 0.0046	893.8	893.9	0.2891 ± 0.0046	0.9150 ± 0.0013	1.97 ± 0.03	1.09 ± 0.00
364.51	0.1549 ± 0.0018	0.4527 ± 0.0048	0.2549 ± 0.0042	794.0	794.3	0.2388 ± 0.0042	0.9040 ± 0.0014	2.13 ± 0.04	1.06 ± 0.00
364.51	0.1178 ± 0.0014	0.4527 ± 0.0048	0.2065 ± 0.0037	682.3	683.0	0.1899 ± 0.0036	0.8883 ± 0.0016	2.31 ± 0.04	1.04 ± 0.00
364.51	0.0847 ± 0.0011	0.4527 ± 0.0048	0.1576 ± 0.0031	558.0	558.2	0.1420 ± 0.0029	0.8636 ± 0.0018	2.51 ± 0.05	1.02 ± 0.00
364.51	0.0563 ± 0.0008	0.4527 ± 0.0048	0.1106 ± 0.0024	426.4	426.1	0.0975 ± 0.0023	0.8221 ± 0.0019	2.72 ± 0.06	1.01 ± 0.00
364.51	0.0290 ± 0.0005	0.4527 ± 0.0048	0.0602 ± 0.0016	272.4	272.2	0.0517 ± 0.0015	0.7238 ± 0.0021	2.97 ± 0.09	1.00 ± 0.00
364.52	0.0190 ± 0.0004	0.4527 ± 0.0048	0.0403 ± 0.0012	208.5	208.6	0.0343 ± 0.0011	0.6409 ± 0.0023	3.07 ± 0.11	1.00 ± 0.00
364.53	0.0100 ± 0.0003	0.4527 ± 0.0048	0.0215 ± 0.0009	146.5	146.7	0.0181 ± 0.0008	0.4914 ± 0.0041	3.17 ± 0.12	1.00 ± 0.00
364.51	0.0000 ± 0.0000	0.4527 ± 0.0048	0.0000 ± 0.0000	74.1	74.1	0.0000 ± 0.0000	0.0000 ± 0.0000	3.28 ± 0.15	1.00 ± 0.00

[a] T is the experimental temperature; n_i is the amount of component in the cell; z_1 is the total mole fraction; p_{exptl} is the measured pressure and p_{Leg} is the pressure calculated from the Legendre polynomial fit; x_1 and y_1 are the liquid and vapor phase equilibrium mole fractions, respectively; γ_1 and γ_2 are the liquid activity coefficients.

used as the liquid activity coefficient (γ_i) model, and the fugacities (ϕ) were calculated by the Soave−Redlich−Kwong (SRK) equation of state[21] with quadratic mixing rules. The binary interaction parameters in the quadratic mixing rules were set to zero. The molar volumes (V_i^L) in the Poynting correction were calculated by using the Rackett equation.[22] The symbols R and T denote the gas constant and the system temperature. The superscript S in the component fugacity and pressure (p_i) refers to the saturated state of vapor pressure. The number of parameters in Legendre polynomials for each system was chosen as suggested by Pokki[23] so that the most accurate results were achieved but overfitting was avoided. The details of the data reduction are presented in Uusi-Kyyny et al.[16] The component critical properties and acentric factors[14] used in the data reduction are presented in Table 3.

Table 7. VLE Data for Isobutane (1) and 2-Methyl-2-propanol (2) at 364.5 K[a]

T/K	n_1/mol	n_2/mol	z_1	p_{exptl}/kPa	p_{Leg}/kPa	x_1	y_1	γ_1	γ_2
364.50	0.4303 ± 0.0046	0.0000 ± 0.0000	1.0000 ± 0.0000	1685.6	1685.6	1.0000 ± 0.0000	1.0000 ± 0.0000	1.00 ± 0.00	4.34 ± 0.15
364.50	0.4303 ± 0.0046	0.0048 ± 0.0003	0.9891 ± 0.0007	1673.1	1673.7	0.9885 ± 0.0007	0.9932 ± 0.0006	1.00 ± 0.00	4.09 ± 0.11
364.51	0.4303 ± 0.0046	0.0090 ± 0.0003	0.9795 ± 0.0009	1661.8	1663.1	0.9785 ± 0.0009	0.9879 ± 0.0008	1.00 ± 0.00	3.89 ± 0.09
364.51	0.4303 ± 0.0046	0.0136 ± 0.0004	0.9693 ± 0.0011	1649.9	1651.6	0.9677 ± 0.0012	0.9827 ± 0.0010	1.00 ± 0.00	3.70 ± 0.07
364.52	0.4303 ± 0.0046	0.0231 ± 0.0004	0.9491 ± 0.0014	1627.2	1629.2	0.9464 ± 0.0016	0.9737 ± 0.0012	1.01 ± 0.00	3.37 ± 0.05
364.51	0.4303 ± 0.0046	0.0480 ± 0.0007	0.8996 ± 0.0023	1574.6	1575.9	0.8942 ± 0.0025	0.9570 ± 0.0014	1.02 ± 0.00	2.75 ± 0.02
364.51	0.4303 ± 0.0046	0.0771 ± 0.0010	0.8481 ± 0.0030	1524.1	1524.1	0.8405 ± 0.0034	0.9446 ± 0.0015	1.05 ± 0.00	2.32 ± 0.00
364.51	0.4303 ± 0.0046	0.1086 ± 0.0013	0.7985 ± 0.0037	1477.4	1476.6	0.7896 ± 0.0040	0.9353 ± 0.0015	1.08 ± 0.00	2.02 ± 0.00
364.50	0.4303 ± 0.0046	0.1449 ± 0.0017	0.7481 ± 0.0042	1430.5	1429.4	0.7387 ± 0.0046	0.9273 ± 0.0016	1.12 ± 0.01	1.81 ± 0.00
364.51	0.4303 ± 0.0046	0.1857 ± 0.0021	0.6985 ± 0.0046	1383.5	1382.8	0.6893 ± 0.0050	0.9202 ± 0.0017	1.17 ± 0.01	1.64 ± 0.00
364.51	0.4303 ± 0.0046	0.2329 ± 0.0026	0.6489 ± 0.0050	1335.7	1335.2	0.6405 ± 0.0052	0.9133 ± 0.0018	1.21 ± 0.01	1.51 ± 0.00
364.51	0.4303 ± 0.0046	0.2885 ± 0.0031	0.5986 ± 0.0052	1284.6	1285.0	0.5917 ± 0.0054	0.9065 ± 0.0018	1.27 ± 0.01	1.41 ± 0.00
364.50	0.4303 ± 0.0046	0.3535 ± 0.0038	0.5490 ± 0.0053	1231.4	1232.8	0.5440 ± 0.0055	0.8995 ± 0.0018	1.33 ± 0.01	1.33 ± 0.00
364.50	0.4303 ± 0.0046	0.4317 ± 0.0046	0.4992 ± 0.0054	1174.8	1177.1	0.4964 ± 0.0054	0.8919 ± 0.0017	1.39 ± 0.01	1.26 ± 0.00
364.51	0.4299 ± 0.0046	0.4332 ± 0.0046	0.4981 ± 0.0054	1178.8	1177.2	0.4953 ± 0.0054	0.8917 ± 0.0017	1.39 ± 0.01	1.26 ± 0.00
364.51	0.3532 ± 0.0038	0.4332 ± 0.0046	0.4491 ± 0.0053	1112.3	1111.2	0.4433 ± 0.0054	0.8824 ± 0.0017	1.47 ± 0.02	1.20 ± 0.00
364.51	0.2881 ± 0.0032	0.4332 ± 0.0046	0.3994 ± 0.0052	1038.0	1037.6	0.3907 ± 0.0052	0.8713 ± 0.0017	1.57 ± 0.02	1.15 ± 0.00
364.50	0.2336 ± 0.0026	0.4332 ± 0.0046	0.3503 ± 0.0050	957.6	957.7	0.3389 ± 0.0050	0.8581 ± 0.0017	1.67 ± 0.02	1.11 ± 0.00
364.50	0.1864 ± 0.0021	0.4332 ± 0.0046	0.3009 ± 0.0046	869.2	869.3	0.2873 ± 0.0046	0.8413 ± 0.0018	1.78 ± 0.03	1.08 ± 0.00
364.50	0.1469 ± 0.0017	0.4332 ± 0.0046	0.2532 ± 0.0042	776.0	776.1	0.2384 ± 0.0042	0.8201 ± 0.0019	1.90 ± 0.03	1.05 ± 0.00
364.50	0.1119 ± 0.0013	0.4332 ± 0.0046	0.2052 ± 0.0037	673.9	673.9	0.1902 ± 0.0036	0.7909 ± 0.0021	2.03 ± 0.04	1.03 ± 0.00
364.51	0.0804 ± 0.0010	0.4332 ± 0.0046	0.1565 ± 0.0031	562.0	561.8	0.1424 ± 0.0030	0.7474 ± 0.0021	2.18 ± 0.04	1.02 ± 0.00
364.51	0.0528 ± 0.0007	0.4332 ± 0.0046	0.1087 ± 0.0024	443.0	443.2	0.0970 ± 0.0023	0.6784 ± 0.0019	2.35 ± 0.05	1.01 ± 0.00
364.51	0.0265 ± 0.0005	0.4332 ± 0.0046	0.0577 ± 0.0015	307.7	307.6	0.0503 ± 0.0014	0.5356 ± 0.0015	2.54 ± 0.08	1.00 ± 0.00
364.51	0.0174 ± 0.0004	0.4332 ± 0.0046	0.0387 ± 0.0012	254.5	254.6	0.0334 ± 0.0011	0.4389 ± 0.0017	2.62 ± 0.09	1.00 ± 0.00
364.51	0.0081 ± 0.0003	0.4332 ± 0.0046	0.0184 ± 0.0008	196.8	196.7	0.0158 ± 0.0008	0.2742 ± 0.0033	2.71 ± 0.10	1.00 ± 0.00
364.50	0.0000 ± 0.0000	0.4332 ± 0.0046	0.0000 ± 0.0000	142.6	142.6	0.0000 ± 0.0000	0.0000 ± 0.0000	2.79 ± 0.12	1.00 ± 0.00

[a] T is the experimental temperature; n_i is the amount of component in the cell; z_1 is the total mole fraction; p_{exptl} is the measured pressure and p_{Leg} is the pressure calculated from the Legendre polynomial fit; x_1 and y_1 are the liquid and vapor phase equilibrium mole fractions, respectively; γ_1 and γ_2 are the liquid activity coefficients.

Error Analysis. The overall uncertainty in the temperature measurements was ± 0.02 K for the water bath and ± 0.1 K for the syringe pumps. The uncertainty in the pressure measurements was ± 0.7 kPa for the cell and ± 20 kPa for the syringe pumps. The uncertainty of the injected volumes was 0.02 cm^3, and it was obtained from calibration with distilled water. The densities of the components were calculated from the Hankinson−Brobst−Thompson (HBT) density correlation.[24,15] The reported uncertainties for the density correlations of pure components were 1 % for all the components.

The error analysis procedure is presented in Laakkonen et al.[25] with the modifications by Hynynen et al.[17] The maximum errors of n_i, T, and p_{exptl} were calculated based on the known uncertainties of the equipment and the correlations used. Then, the maximum errors for the variables x_i, y_i, p_{Leg}, and γ_i throughout the range of experiments were calculated by the VLEFIT program. In the program, the values of these variables were separately set to their upper and lower values as the theoretical maximum errors are not necessarily obtained when all the variables are simultaneously set either to their upper or lower value. Thus, 16 combinations of upper and lower values were made to reveal the theoretical maximum error. In the tables, the reported uncertainty is the average absolute error calculated from the combinations.

Results and Discussion

The measured VLE data together with the calculated equilibrium data and activity coefficients are presented in Tables 4 to 7. The measured total pressure as a function of the calculated liquid mole fraction is shown in Figure 1. The composition diagram is shown in Figure 2. An azeotropic behavior was observed for the system of isobutane + methanol at $x_{isobutane}$ = 0.871, p_{exptl} = 1822 kPa, and T = 364.5 K. The Legendre parameters obtained through data reduction and the Wilson model parameters[26] are presented in Table 8. The activity coefficients obtained by Legendre polynomials are shown in Figure 3.

Table 8. Liquid Activity Coefficient Model Parameters for Legendre and Wilson Models[a]

isobutane (1)	+ methanol	+ 2-propanol	+ 2-butanol	+ TBA
Legendre, $a_{1,0}$	2.21802	1.54496	1.37761	1.16606
Legendre, $a_{2,0}$	−0.000803	0.205511	0.262086	0.205023
Legendre, $a_{3,0}$	0.191102	0.103683	0.0971993	0.0759488
Legendre, $a_{4,0}$	−0.004607	0.0224098	0.0272624	0.015444
Legendre, $a_{5,0}$	−	0.0043871	0.0035757	0.0050932
γ_1^∞	11.30	4.16	3.28	2.79
γ_2^∞	11.18	6.56	5.86	4.34
$\|\Delta p\|$/kPa	1.69	0.68	0.68	0.67
Wilson $\lambda_{1,2}$/K	254.61	129.913	111.566	84.9909
Wilson $\lambda_{2,1}$/K	1013.32	626.539	567.351	454.455
Wilson volume ratio at 298.1 K	2.561	1.356	1.129	1.099
γ_1^∞	11.95	4.13	3.29	2.75
γ_2^∞	14.08	6.67	5.95	4.18
$\|\Delta p\|$/kPa	13.89	3.92	3.98	5.46

[a] γ_1^∞, γ_2^∞, infinite dilution activity coefficients; $|\Delta p|$, Absolute Average Pressure Residuals. Legendre model parameters obtained from data reduction of measured values, and Wilson parameters fit from regressed values.

In all the experiments, the pressures coincided when approaching equimolarity from both ends of the binary composition scale. Therefore, the measured data were considered to be of good quality. The regressed parameters of the Legendre polynomials and the calculated infinite dilution activity coefficients by the Legendre polynomials and the Wilson model are presented in Table 8.

The regressed parameters depict the binaries properly as the average absolute pressure residuals are small. In addition, the average absolute pressure deviations of Legendre polynomial fits are close to the uncertainty of the measured pressure. However, in the system isobutane + methanol, which forms an azeotrope, the pressure deviation is higher. In this specific binary mixture, the quality of the Legendre polynomial fit did not improve even with a larger number of Legendre parameters. In

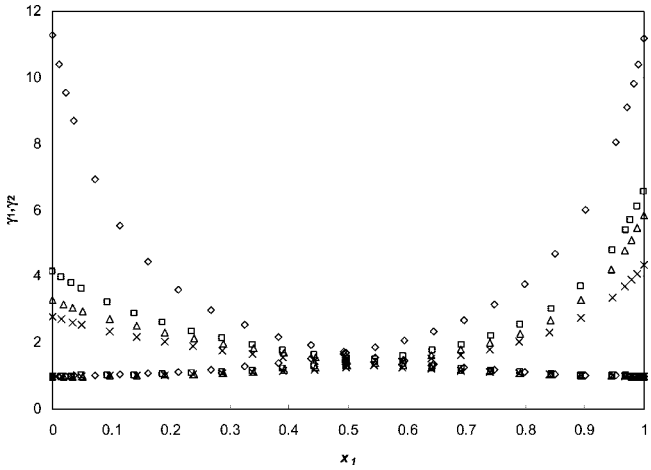

Figure 3. Activity coefficient composition diagram of isobutane (1) ◇, + methanol; □, + 2-propanol; △, + 2-butanol; and ×, + TBA at 364.5 K.

Table 9. Comparison of Measured Azeotropic Composition of Isobutane + Methanol to Literature

	x_1	T/K	p_{measd}/kPa
this work	0.871	364.51	1821.8
literature[a]	0.9906	273.15	159.1
	0.947[b]	313.06[b]	548.25[b]
	0.9562	323.15	733.2
	0.9007[c]	364.51[c]	
	0.8883	373.15	2190
	0.832	398.15	3438
	0.8126	404.15	3808
	0.8048	406.15	3959

[a] Ref 2. [b] Ref 1. [c] Correlation from ref 2.

Table 10. Comparison of Infinite Dilution Activity Coefficients from Different Sources

T/K	+ methanol	+ 2-propanol	+ 2-butanol	+ TBA
		γ_1^∞		
250		5.4[a]	4.4[b]	
255	17.5[c]			
260	17.1[c]			
270	16.5[c]	5.3[a]	4.3[b]	
280	15.8[c]			
290	15.1[c]	5.2[a]	4.2[b]	
300	14.6[c]			3.6[d]
310	14.1[c]	5.0[a]	4.1[b]	3.44[d]
313	13.86[e]	5.04[e]	3.73[e]	3.38[e]
320	13.6[c]			3.35[d]
330		4.7[a]	3.8[b]	3.24[d]
365	11.30	4.16	3.28	2.79
374	9.6[f]			
395	8.3[f]			
		γ_2^∞		
313	44.39[e]	22.14[e]	17.30[e]	12.83[e]
365	11.18	6.56	5.86	4.34

[a] Ref 6. [b] Ref 7. [c] Ref 4. [d] Ref 8. [e] Ref 1. [f] Ref 5.

the table, the minimum number of parameters above which the fit did not improve is reported. The mathematical difficulties of finding a better fit for this binary can be due to the azeotropic behavior.

In the case of the binary isobutane + methanol, a crucial indicator of the quality of the model can be regarded as its capability to predict the location of the azeotropic point. As seen in the enlarged figure of the azeotropic region in Figure 1, the vapor pressure curve is rather flat, which complicates the definition of the exact location of the azeotropic point. Nonetheless, the Legendre model successfully predicts the location of the azeotropic point in relation to our measured data. Even though the Legendre model precisely predicts the location, it overestimates the vapor pressure of the azeotropic point. Nevertheless, the correct location of the azeotropic point is more critical than the exact vapor pressure.

A comparison of measured azeotropic compositions is shown in Table 9. Azeotropic data were found only for the system isobutane + methanol. No experimental data at 364.5 K were found, but the experimental compositions at other temperatures are listed in together with the azeotropic point measured in this work. Our results agree with other literature sources. In addition, Leu and Robinson[2] presented a correlation for calculating the azeotropic composition for the system of isobutane + methanol. The correlation was based on their own experimental data. By using their correlation, the location of the azeotropic point at 364.5 K was calculated and added to Table 9.

Our results complement other literature sources even though there seems to be slight discrepancies in the evolution of the azeotropic point with temperature. As shown in Table 9, the azeotropic data measured by Leu and Robinson[2] seems to overestimate the mole fraction of isobutane in comparison to the measurements performed in our laboratory. Nevertheless, a comparison of the results of this work and the previous work in our laboratory[1] with the data by Leu and Robinson shows that in both cases the data by Leu and Robinson overestimate the azeotropic composition of isobutane. However, due to the flatness of the curve shown in Figure 1, even small errors in the measurements can cause larger errors in the composition and thus lead to discrepancies in the evolution of the azeotropic point with temperature.

When considering the infinite dilution activity coefficients, the differences between the predictions by the Legendre model and the Wilson model are in average minor as seen in Table 8. However, in the case of isobutane + methanol, where an azeotrope is formed, the difference is higher. A comparison of reported infinite dilution activity coefficients at different temperatures is shown in Table 10. At 364.5 K, where the measurements of this work were performed, there were no reported values for infinite dilution activity coefficients for these systems. Nevertheless, literature values of this property are reported at other temperatures. As seen in Table 10, our results properly complement the literature data.

Conclusions

Isothermal VLE data of four binary systems consisting of isobutane and alcohols were measured at 364.5 K by using a static total pressure apparatus. The obtained p, T, z data were converted into the P, T, x, y form through the method of Barker. Azeotropic behavior was observed for the system isobutane + methanol at $x_{isobutane} = 0.871$, $p_{exptl} = 1822$ kPa, and $T = 364.5$ K. Our results entirely fit in the list of the other reported azeotropic compositions of the isobutane + methanol system and confirm the azeotropic behavior in this temperature range. The parameters of the Legendre polynomials and of the Wilson model were regressed, and an error analysis was performed. The infinite dilution activity coefficients were compared to literature data and our measured data proved to complement other literature sources.

Literature Cited

(1) Ouni, T.; Zaytseva, A.; Uusi-Kyyny, P.; Pokki, J.-P.; Aittamaa, J. Vapour-liquid equilibrium for the 2-methylpropane + methanol, +ethanol, + 2-propanol, + 2-butanol and + 2-methyl-2-propanol systems at 313.15 K. *Fluid Phase Equilib.* **2005**, *232*, 90–99.

(2) Leu, A.-D.; Robinson, D. B. Equilibrium phase properties of the methanol-isobutane binary system. *J. Chem. Eng. Data* **1992**, *37*, 10–13.

(3) Zabaloy, M. S.; Mabe, G. D. B.; Bottini, S. B.; Brignole, E. A. Vapor liquid equilibria in ternary mixtures of water-alcohol-non polar gases. *Fluid Phase Equilib.* **1993**, *83*, 159–166.

(4) Miyano, Y.; Nakanishi, K.; Fukuchi, K. Henry's constants of butane, isobutane, 1-butene and isobutene in methanol at 255–320 K. *Fluid Phase Equilib.* **2003**, *208*, 223–238.

(5) Miyano, Y.; Fujihara, I.; Sato, K. Henry's law constants of propane, propene, butane, and 2-methylpropane in methanol at 374–490 K. *Fluid Phase Equilib.* **2006**, *240*, 56–62.

(6) Miyano, Y. Henry's constants and infinite dilution activity coefficients of propane, propene, butane, isobutane, 1-butene, isobutene, *trans*-2-butene, and 1,3-butadiene in 2-propanol at 250–330 K. *J. Chem. Eng. Data* **2004**, *49*, 368–371.

(7) Miyano, Y. Henry's constants and infinite dilution activity coefficients of propane, propene, butane, isobutane, 1-butene, isobutene, trans-2-butene, and 1,3-butadiene in 1-butanol and 2-butanol from (250–330) K. *J. Chem. Eng. Data* **2004**, *49*, 1285–1289.

(8) Miyano, Y. Henry's constants and infinite dilution activity coefficients of propane, propene, butane, isobutane, 1-butene, isobutene, trans-2-butene, and 1,3-butadiene in isobutanol and tert-butanol. *J. Chem. Thermodyn.* **2004**, *36*, 865–869.

(9) Moore, J. D.; Brown, P R.; Ott, J. B. Excess molar enthalpies and excess molar volumes for the binary mixtures of 2-methylpropane, 2-methylpropene, and propan-2-ol at the temperatures (298.15 and 323.15) K and the pressures (5, 10, and 15) MPa. *J. Chem. Thermodyn.* **1997**, *29*, 179–195.

(10) Van Ness, H. C.; Abbott, M. M. A procedure for rapid degassing of liquids. *Ind. Eng. Chem. Fundam.* **1978**, *17:1*, 66–67.

(11) Fischer, K.; Gmehling, J. P-x and γ^{∞} data for the different binary butanol-water systems at 50°C. *J. Chem. Eng. Data* **1994**, *39*, 309–315.

(12) Yaws, C. L. *Chemical properties handbook: physical, thermodynamic, environmental, transport, safety, and health related properties for organic and inorganic chemicals*; McGraw-Hill: New York, 1999.

(13) Perry, R. H.; Green, D. W. *Perry's chemical engineers' handbook*, 7th ed.; McGraw-Hill: New York, 1997.

(14) Daubert, T. E.; Danner, R. P. *Physical and thermodynamic properties of pure chemicals: Data compilation*; Hemisphere: New York, 1989.

(15) Reid, R. C.; Prausnitz, J. M.; Poling, B. E. *The properties of gases & liquids*, 4th ed.; McGraw-Hill: New York, 1987.

(16) Uusi-Kyyny, P.; Pokki, J.-P.; Laakkonen, M.; Aittamaa, J.; Liukkonen, S. Vapor liquid equilibrium for the binary systems 2-methylpentane + 2-butanol at 329.2 K and n-hexane + 2-butanol at 329.2 and 363.2 K with a static apparatus. *Fluid Phase Equilib.* **2002**, *201*, 343–358.

(17) Hynynen, K.; Uusi-Kyyny, P.; Pokki, J.-P.; Pakkanen, M.; Aittamaa, J. Isothermal vapor liquid equilibrium for 2-methylpropene + methanol, + 1-propanol, + 2-propanol, + 2-butanol, and + 2-methyl-2-propanol binary systems at 364.5 K. *J. Chem. Eng. Data* **2006**, *51*, 562–568.

(18) Barker, J. A. Determination of activity coefficients from total pressure measurements. *Aust. J. Chem.* **1953**, *6*, 207–210.

(19) Aittamaa, J.; Pokki, J.-P. *VLEFIT A Program package to obtain parameters in models of activity coefficient from VLE data (Manual)*; Helsinki University of Technology, Laboratory of chemical engineering: Espoo, 2004.

(20) Gmehling, J.; Onken, U. *Vapor-liquid equilibrium data collection. Aqueous-organic systems*, DECHEMA Chemistry Data Series; DECHEMA: Frankfurt/Main, 1977; Vol. 1, Part 1.

(21) Soave, G. Equilibrium constants from a modified Redlich-Kwong equation of state. *Chem. Eng. Sci.* **1972**, *27*, 1192–1203.

(22) Rackett, H. G. Equation of state for saturated liquids. *J. Chem. Eng. Data* **1970**, *15* (4), 514–517.

(23) Pokki, J.-P. *Development of Vapour Liquid Equilibrium Calculation Methods for Chemical Engineering Design*, Dissertation, Chemical Engineering Report Series No. 46; Espoo, 2004.

(24) Poling, B. E.; Prausnitz, J. M.; O'Connell, J. P. *The properties of gases & liquids*, 5th ed.; McGraw-Hill: NY, 2000.

(25) Laakkonen, M.; Pokki, J.-P.; Uusi-Kyyny, P.; Aittamaa, J. Vapour-liquid equilibrium for the 1-butene + methanol, + ethanol, + 2-propanol, + 2-butanol and + 2-methyl-2-propanol systems at 326 K. *Fluid Phase Equilib.* **2003**, *206*, 237–252.

(26) Wilson, G. M. Vapor-liquid equilibrium. XI. A new expression for the excess free energy of mixing. *J. Am. Chem. Soc.* **1964**, *86*, 127–130.

Received for review January 8, 2008. Accepted January 28, 2008. The Graduate School of Chemical Engineering (GSCE) is gratefully acknowledged for financial support.

JE800021R