Vapor−Liquid Equilibria for the Binary Systems of Methylcyclohexane with 1-Propanol, 2-Propanol, 1-Butanol, and 2-Butanol at 101.3 kPa

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Isobaric vapor−liquid equilibrium measurements at 101.3 kPa are reported for the binary systems methylcyclohexane + 1-propanol, + 2-propanol, + 1-butanol, and + 2-butanol. The systems show a strong positive deviation from ideality and present a minimum temperature azeotrope. The activity coefficients were found to be thermodynamically consistent by the methods of Van Ness−Byer−Gibbs and Wisniak. The date were correlated with five liquid-phase activity coefficient models (Margules, Van Laar, Wilson, NRTL, and UNIQUAC). According to the average deviations in temperature and in vapor composition the best model for the systems is NRTL.

Introduction

Distillation operation has been extensively used for the separation of liquid mixtures. Accurate and complete vapor−liquid equilibrium data are essential for the development and the rational design of distillation tower and other separation processes. Although these data can be estimated from available predictive VLE models, experimental data are required to update and improve the data bank used to fit the model parameters. Among these models, group-contribution methods such as UNIFAC (Fredenslund et al., 1977; Gmehling et al., 1982; Skjold-Jøergensen et al., 1979) have been shown to be useful. There is a lack of VLE measurements for mixtures that contain components with previously unstudied groups (Gmehling and Schiller, 1993).

Raju and Rao (1969) studied the systems 1-propanol + methylcyclohexane and methylcyclohexane + 1-butanol at 101.3 kPa, but the results were not thermodynamically consistent according to the point-to-point test. Seetharamaswamy et al. (1969) studied the system methylcyclohexane + 1-butanol at 101.3 kPa. Nagata (1965) measured VLE for the system 2-propanol + methylcyclohexane at a reduced pressure of 66.7 kPa, and the same system was studied by Nagata et al. (1973) at 323 and 333 K. Yamamoto et al. (1959) studied the system 2-butanol + methylcyclohexane at 101.3 kPa.

In this paper, we report VLE data at constant pressure (101.3 kPa), for four binary systems: methylcyclohexane with 1-propanol, 2-propanol, 1-butanol, and 2-butanol. The experimental data are shown to be thermodynamically consistent. Furthermore, we have determined the vapor pressures of pure methylcyclohexane.

Experimental Section

Chemicals. The reagents 1-propanol (99.5%), 2-propanol (99.5%), 1-butanol (99.8%), and methylcyclohexane (>99.0%) were obtained from Aldrich, and 2-butanol (>99.0%) was purchased from Fluka. The purity of the materials was checked by gas chromatography (GC), and this analysis showed that the impurities did not exceed 0.2 mass %. The liquids were used without further purification. The comparison of measured physical properties (densities, refractive indexes, and normal boiling point) with literature values is shown in Table 1.

Apparatus and Procedure. The still used to measure VLE was an all-glass dynamic recirculating one equipped with a Cottrell pump. The apparatus (Labodest model), manufactured by Fischer Labor und Verfahrenstechnik (Bonn, Germany), is capable of handling pressures from 0.25 up to 400 kPa and temperatures up to 523.15 K. This still ensures good mixing of the vapor and liquid phases and good separation of the phases once they reach equilibrium, and it prevents entrainment of liquid drops and partial condensation in the vapor phase. The equilibrium temperature was measured with a digital Fischer thermometer with an accuracy of ±0.1 K and the pressure with a digital manometer with an accuracy of ±0.01 kPa. The temperature probe was calibrated against the ice and steam points of distilled water. The vapor pressure of high-purity hexane (>99.9 mass %) was used for the manometer calibration.

In each experiment, the temperature was fixed and the heating and shaking system of the liquid mixture was turned on. The still was operated until equilibrium was reached. At this time, 0.2 cm^3 samples of liquid and condensed vapor were taken with special syringes without disruption of the operation of the still.

All the samples were analyzed using a Hewlett-Packard 5890 S-II gas chromatograph (GC), with calibration with gravimetrically prepared standard solutions. A flame ionization detector was used with a 60-m, 0.2-mm-i.d. fused silica capillary column, SUPERCOWAX 10. The gas carrier was helium flowing at 1 cm^3/min, and the column, injector, and detector temperatures were 393, 493, and 523 K, respectively. The GC response peaks were integrated by using a Hewlett-Packard 3396 integrator. The accuracy of the measured mole fraction was ±0.001. At least two analyses were made for each liquid and each vapor sample.

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Table 1. Physical Properties of Chemicals: Densities *d* and Refractive Indexes *n* at 298.15 K and Normal Boiling Points T_b of Pure Compounds

	$d/(\mathrm{kg \cdot m^{-3}})$		n		T_b/K	
compound	exptl	lit.	exptl	lit.	exptl	lit.
1-butanol	805.64	805.60[a]	1.3972	1.3972[a]	390.5	390.55[a]
2-butanol	802.62	780.93[a]	1.3950	1.3950[a]	372.1	372.35[a]
1-propanol	799.51	799.50[a]	1.3830	1.3831[a]	369.9	370.01[a]
2-propanol	780.95	780.93[a]	1.3748	1.3750[a]	355.1	355.39[a]
methylcyclohexane	764.83	764.59[a]	1.4194	1.4206[b]	373.6	374.08[a]

[a] *TRC Thermodynamic Tables Hydrocarbons*, 1996. [b] *Data Compilation Tables of Properties of Pure Compounds* (Daubert and Danner, 1989).

Table 2. Vapor−Liquid Equilibrium Data, Liquid Phase Mole Fraction x_1, Vapor Phase Mole Fraction y_1, Temperature *T*, and Activity Coefficients γ_i for the 1-Propanol (1) + Methylcyclohexane (2) System at 101.3 kPa

x_1	y_1	T/K	γ_1	γ_2
0.000	0.000	373.6		
0.018	0.125	370.3	7.020	0.975
0.033	0.191	367.8	6.376	0.983
0.051	0.275	364.6	6.686	0.984
0.066	0.319	363.1	6.334	0.982
0.134	0.394	359.9	4.352	1.037
0.175	0.407	359.4	3.516	1.080
0.240	0.429	359.0	2.741	1.144
0.296	0.438	358.9	2.277	1.220
0.362	0.445	358.8	1.899	1.333
0.428	0.457	358.8	1.649	1.455
0.487	0.471	358.8	1.491	1.583
0.501	0.473	358.8	1.456	1.621
0.573	0.484	358.8	1.302	1.856
0.636	0.493	358.9	1.191	2.131
0.751	0.546	359.2	1.102	2.769
0.791	0.575	359.7	1.079	3.048
0.833	0.609	361.0	1.031	3.377
0.859	0.638	361.8	1.014	3.621
0.926	0.732	363.8	0.997	4.840
0.971	0.873	366.7	1.013	5.430
0.988	0.937	368.3	1.007	6.192
1.000	1.000	369.9		

Table 3. Vapor−Liquid Equilibrium Data, Liquid Phase Mole Fraction x_1, Vapor Phase Mole Fraction y_1, Temperature *T*, and Activity Coefficients γ_i for the 2-Propanol (1) + Methylcyclohexane (2) System at 101.3 kPa

x_1	y_1	T/K	γ_1	γ_2
0.000	0.000	373.6		
0.011	0.136	369.2	7.505	0.987
0.020	0.219	366.2	7.369	0.980
0.032	0.297	363.3	6.918	0.972
0.057	0.397	359.0	6.071	0.973
0.102	0.477	355.6	4.632	0.983
0.155	0.522	353.6	3.602	1.017
0.213	0.547	352.4	2.878	1.075
0.273	0.561	351.6	2.377	1.156
0.330	0.574	351.1	2.052	1.237
0.393	0.586	350.6	1.794	1.349
0.464	0.601	350.3	1.577	1.487
0.532	0.613	350.2	1.408	1.658
0.595	0.629	350.2	1.291	1.838
0.653	0.648	350.2	1.211	2.037
0.707	0.667	350.2	1.151	2.284
0.754	0.687	350.3	1.107	2.551
0.798	0.712	350.5	1.075	2.843
0.838	0.741	350.7	1.056	3.173
0.870	0.769	351.1	1.038	3.487
0.905	0.810	351.7	1.026	3.859
0.934	0.850	352.4	1.014	4.299
0.960	0.897	353.3	1.004	4.749
0.975	0.931	353.9	1.002	5.006
0.987	0.961	354.4	1.002	5.367
0.993	0.979	354.7	1.003	5.323
1.000	1.000	355.1		

Results and Discussion

Vapor−liquid equilibrium data (T, x_1, and y_1) along with calculated activity coefficients at 101.3 kPa are presented in Tables 2−5 and Figures 1 and 2. The systems show a minimum boiling azeotrope. The composition and boiling temperature of the azeotropes are summarized in Table 6 along with a comparation with literature values. Azeotropic compositions have been obtained, in all cases, by determining x_1 values that make zero the best polynomial fit of the fuction $(x_1 - y_1) = f(x_1)$. Azeotropic temperatures have been obtained from the best polynomial fit for $T = f(x_1)$, using the x_1 values previously determined.

The activity coefficients γ_i were calculated, taking into account the nonideality of the vapor phase, from the following equations:

$$\gamma_i = \frac{y_i P}{x_i P^\circ_i} \exp\left[\frac{(B_{ii} - V^\circ_i)(P - P^\circ_i) + (1 - y_i)^2 P \delta_{ij}}{RT}\right] \tag{1}$$

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

where x_i and y_i are the liquid and vapor phase compositions, P is the total pressure, P°_i are the vapor pressures of the pure compounds calculated by the Antoine equation, B_{ii} are the second virial coefficients of the pure gases, B_{ij} are the cross second virial coefficients, and V°_i are the molar volumes of the saturated liquids. The correction for the nonideality of the vapor, represented by the exponential term in eq 1, contributed less than 1.5% to the activity coefficients; in general, its influence was important only at very dilute concentrations.

Antoine constants for methylcyclohexane were obtained from our own vapor pressure measurements over the temperature range 335.55−377.95 K using the same still as for the binary systems; the results are shown in Table 7. The constants were calculated by a nonlinear optimization method to minimize the mean relative deviation, MRD-(P) = 0.16%. The Antoine constants for 1-propanol and 2-propanol were obtained from Aucejo et al. (1995) and for 1-butanol and 2-butanol were taken from Dejoz et al. (1995). The molar virial coefficients B_{ii} and B_{ij} were estimated by means of the Pitzer and Curl equations (1957) with the correction proposed by Tsonopoulos (1974). Critical properties of both components and the molar volumes of the saturated liquids were taken from DIPPR (Daubert and Danner, 1989).

The thermodynamic consistency of the experimental results was checked using the point-to-point test of Van Ness−Byer−Gibbs (1973), described by Fredenslund et al. (1977), and the $L-W$ method of Wisniak (1993). For the Van Ness method, a five-parameter Legendre polynomial was used for the excess Gibbs free energy. According to this test, the experimental data are consistent if the average deviation in y (Δy) is smaller than 0.01.

The $L-W$ method of Wisniak requires the evaluation of the integrals L and W as described in the paper by Wisniak

Table 4. Vapor–Liquid Equilibrium Data, Liquid Phase Mole Fraction x_1, Vapor Phase Mole Fraction y_1, Temperature T, and Activity Coefficients γ_i for the 1-Butanol (1) + Methylcyclohexane (2) System at 101.3 kPa

x_1	y_1	T/K	γ_1	γ_2
0.000	0.000	373.6		
0.016	0.049	372.4	6.113	0.999
0.028	0.090	371.4	6.659	0.995
0.045	0.121	370.7	5.717	0.997
0.070	0.158	369.6	5.005	1.011
0.108	0.190	369.0	3.989	1.032
0.156	0.212	368.7	3.116	1.070
0.203	0.226	368.5	2.571	1.120
0.259	0.240	368.5	2.139	1.183
0.320	0.252	368.6	1.809	1.265
0.382	0.263	368.6	1.581	1.372
0.445	0.274	368.8	1.402	1.497
0.508	0.289	369.1	1.279	1.640
0.575	0.309	369.5	1.188	1.826
0.625	0.324	370.1	1.118	1.991
0.683	0.351	370.8	1.077	2.219
0.720	0.375	371.9	1.044	2.349
0.773	0.407	373.0	1.010	2.669
0.838	0.477	375.7	0.982	3.072
0.881	0.574	379.6	0.968	3.080
0.910	0.645	381.5	0.981	3.237
0.946	0.750	384.7	0.976	3.513
0.970	0.861	387.2	0.999	3.316
0.984	0.917	388.6	0.999	3.593
0.988	0.940	389.1	1.002	3.424
0.994	0.966	389.7	1.002	3.828
1.000	1.000	390.5		

Table 5. Vapor–Liquid Equilibrium Data, Liquid Phase Mole Fraction x_1, Vapor Phase Mole Fraction y_1, Temperature T, and Activity Coefficients γ_i for the 2-Butanol (1) + Methylcyclohexane (2) System at 101.3 kPa

x_1	y_1	T/K	γ_1	γ_2
0.000	0.000	373.6		
0.013	0.079	371.5	6.367	0.988
0.030	0.130	370.0	4.732	0.991
0.038	0.167	369.0	4.971	0.984
0.065	0.225	367.1	4.245	0.993
0.096	0.264	366.0	3.506	1.007
0.117	0.295	365.1	3.303	1.015
0.152	0.330	364.2	2.935	1.032
0.201	0.359	363.3	2.502	1.076
0.232	0.374	363.0	2.287	1.102
0.272	0.382	362.8	2.002	1.156
0.324	0.398	362.5	1.775	1.222
0.364	0.413	362.4	1.648	1.270
0.415	0.431	362.3	1.510	1.345
0.468	0.448	362.3	1.389	1.437
0.529	0.468	362.3	1.285	1.563
0.597	0.497	362.3	1.209	1.728
0.659	0.527	362.6	1.147	1.905
0.714	0.563	363.3	1.100	2.057
0.766	0.598	363.9	1.063	2.277
0.810	0.641	364.8	1.040	2.447
0.867	0.707	366.3	1.011	2.729
0.907	0.768	367.5	1.002	3.013
0.939	0.825	368.5	1.002	3.344
0.956	0.871	369.7	0.993	3.328
0.976	0.925	370.7	0.996	3.489
0.992	0.974	371.6	0.998	3.603
1.000	1.000	372.1		

(1993), and values of the deviation D defined as

$$D = 100\frac{|L - W|}{L + W} \qquad (3)$$

less than 5 indicate thermodynamic consistency. All the experimental data are consistent according to both methods, as shown in Table 8.

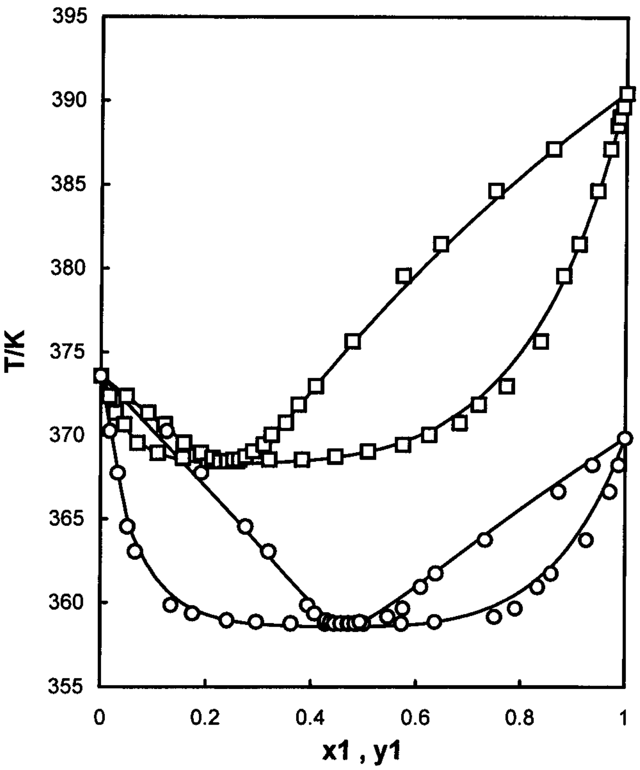

Figure 1. T–x_1–y_1 diagram at constant pressure, $P = 101.3$ kPa: (□) experimental data of 1-butanol (1) + methylcyclohexane (2); (○) experimental data of 1-propanol (1) + methylcyclohexane (2); (—) Van Laar model.

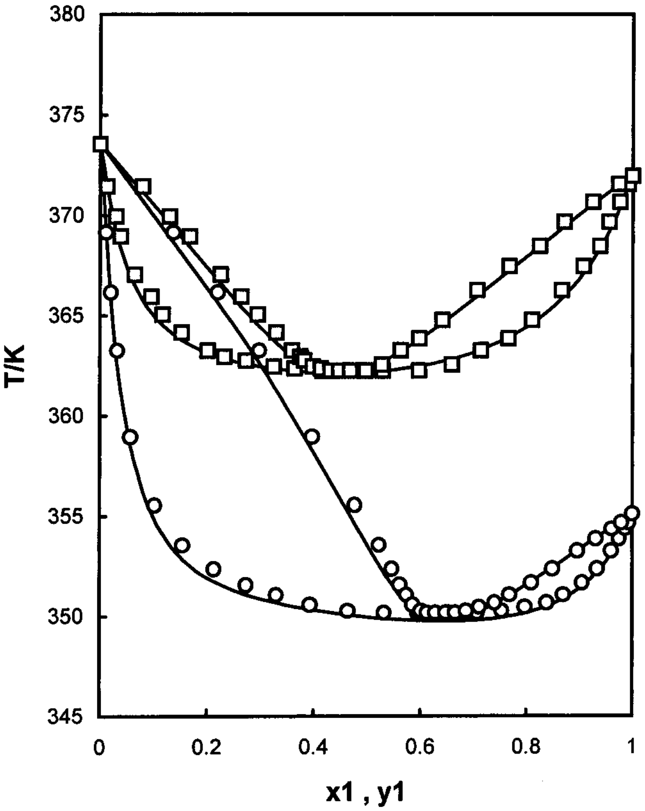

Figure 2. T–x_1–y_1 diagram at constant pressure, $P = 101.3$ kPa: (□) experimental data of 2-butanol (1) + methylcyclohexane (2); (○) experimental data of 2-propanol (1) + methylcyclohexane (2); (—) NRTL (α_{12} adjustable) model.

The activity coefficients were correlated with the Margules, Van Laar, Wilson, NRTL, and UNIQUAC equations (Gmehling et al, 1977). To fit the binary parameters, a

Table 6. Composition ($y_{1(\mathrm{az})}$) and Boiling Temperature ($T_{(\mathrm{az})}$) of the Azeotropic Mixtures

	exptl data		lit. data		
system	$y_{1(\mathrm{az})}$	$T_{(\mathrm{az})}$/K	$y_{1(\mathrm{az})}$	$T_{(\mathrm{az})}$/K	ref
1-propanol (1) + methylclohexane (2)	0.465	358.75	0.465	360.15	Raju and Rao, 1969
2-propanol (1) + methylclohexane (2)	0.645	350.15			
1-butanol (1) + methylclohexane (2)	0.230	368.45	0.248	369.35	Seetharamaswamy et al., 1969
			0.255	369.65	Raju and Rao, 1969
2-butanol (1) + methylclohexane (2)	0.439	362.25	0.460	362.80	Yamamoto et al., 1959[a]

[a] Data from the DECHEMA *Chemistry Data Series* (Gmehling and Onken, 1977).

Table 7. Experimental Vapor Pressures and Constants of the Antoine Equation, ln(P/kPa) = $A - B/(C + T/\mathrm{K})$, of Methylcyclohexane

$P°_i$/kPa	T/K	$P°_i$/kPa	T/K	$P°_i$/kPa	T/K	$P°_i$/kPa	T/K
29.97	335.6	55.00	353.4	79.96	365.6	101.33	373.6
34.96	339.9	59.82	356.0	84.83	367.6	105.00	374.9
40.00	343.8	65.01	358.6	90.09	369.6	109.61	376.4
44.97	347.3	69.92	361.0	94.84	371.4	114.43	378.0
49.95	350.4	74.94	363.5	100.00	373.2		
	A			B		C	
	14.5844			3490.08		−23.413	

Table 8. Thermodynamic Consistency Tests

system:	Van Ness−Byer−Gibbs		Wisniak		
methylcyclohexane +	MAD(y)	MAD(P)/kPa	L	W	D
1-propanol	0.0092	0.73	10.86	10.39	2.21
2-propanol	0.0061	0.45	11.18	11.42	1.05
1-butanol	0.0088	0.65	10.79	11.34	2.47
2-butanol	0.0067	0.46	8.21	8.66	2.67

Table 9. Correlation Parameters for Activity Coefficients, Activity Coefficients at Infinite Dilution γ_i^∞, and Mean Absolute Deviations MAD(y) and MAD(T)

model	A_{12}	A_{21}	α_{12}	γ_1^∞	γ_2^∞	MAD(y)	MAD(T)
		1-Propanol (1) + Methylcyclohexane (2) System					
Margules	2.0392[a]	1.64578[a]		7.68	5.19	0.0096	0.53
Van Laar	2.0667[a]	1.5681[a]		7.90	4.80	0.0099	0.46
Wilson	6361.68[b]	1493.52[b]		8.57	6.09	0.0096	0.37
NRTL	3517.18[b]	4711.16[b]	0.516[a]	8.56	6.32	0.0094	0.37
UNIQUAC	−1151.71[b]	3278.11[b]		8.10	5.11	0.0089	0.55
		2-Propanol (1) + Methylcyclohexane (2) System					
Margules	2.0332[a]	1.5624[a]		7.64	4.78	0.0112	0.51
Van Laar	2.0549[a]	1.5683[a]		7.81	4.80	0.0097	0.46
Wilson	6037.89[b]	1484.77[b]		7.83	5.81	0.0082	0.41
NRTL	3448.65[b]	4680.39[b]	0.537[a]	8.31	6.04	0.0077	0.49
UNIQUAC	−1100.28[b]	3210.43[b]		7.31	4.68	0.0107	0.49
		1-Butanol (1) + Methylcyclohexane (2) System					
Margules	1.9601[a]	1.1684[a]		7.10	3.22	0.0124	0.32
Van Laar	2.0091[a]	1.2683[a]		7.46	3.55	0.0118	0.25
Wilson	6199.45[b]	675.80[b]		8.04	3.92	0.0141	0.33
NRTL	2348.55[b]	4875.32[b]	0.584[a]	7.80	4.04	0.0157	0.41
UNIQUAC	−1392.51[b]	3290.47[b]		8.28	3.49	0.0111	0.41
		2-Butanol (1) + Methylcyclohexane (2) System					
Margules	1.7486[a]	1.1275[a]		5.75	3.09	0.0085	0.41
Van Laar	1.7867[a]	1.1662[a]		5.97	3.21	0.0066	0.37
Wilson	5689.87[b]	383.33[b]		6.45	3.44	0.0051	0.33
NRTL	2669.99[b]	4307.20[b]	0.662[a]	6.50	4.16	0.0054	0.38
UNIQUAC	−1148.97[b]	2698.72[b]		5.62	3.27	0.0055	0.33

[a] Dimensionless. [b] $J \times \mathrm{mol}^{-1}$.

nonlinear optimization method was used to minimize the following objective function

$$F = \sum_{i=1}^{i=N} \left[\left(\frac{y_{\mathrm{exp}} - y_{\mathrm{cal}}}{y_{\mathrm{exp}}} \right)^2 + \left(\frac{T_{\mathrm{exp}} - T_{\mathrm{cal}}}{T_{\mathrm{exp}}} \right)^2 \right]_i \tag{4}$$

where N is the number of experimental data. The adjustable parameters A_{12}, A_{21}, and α_{12} for the correlation equations, mean absolute deviation, and activity coefficients at infinite dilution γ_i^∞ are given in Table 9.

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