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Vapor−Liquid Equilibria at 101.3 kPa for Binary Mixtures Containing 2-Methyl-1-propanol + 2-Methyl-1-butanol, 2-Methyl-1-propanol + 3-Methyl-1-butanol, and 2-Methyl-1-propanol + 1-Pentanol

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Densities, refractive indices, and speeds of sound at 298.15 K, and isobaric vapor−liquid equilibria data at 101.3 kPa were reported for the binary mixtures 2-methyl-1-propanol + 2-methyl-1-butanol, 2-methyl-1-propanol + 3-methyl-1-butanol, and 2-methyl-1-propanol + 1-pentanol. VLE experimental data were tested for thermodynamic consistency by means of the Dechema test and were demonstrated to be consistent. The activity coefficients were correlated with the Margules, van Laar, UNIQUAC, NRTL, and Wilson equations. The ASOG model also was used for prediction. Examining the results, three systems have near ideal behavior.

Introduction

This work is part of a research project whose objective is to measure thermodynamic properties and concentration in equilibrium for binary systems involved in wine distillation processes for further simulation.[1−11] In this process, multicomponent mixtures are seen. The main components are water and ethanol, and several minor compounds such as alcohols, aldehydes, and acetates are also present. These minor compounds are called congeners. For modeling and process simulation in which mixtures appear, binary data are needed. For this, it is very important to have available vapor−liquid equilibrium (VLE) data of mixtures formed by water + congeners, ethanol + congeners, and congeners + congeners. From the measurements parameters of some classic correlations such as Wilson, NRTL, and UNIQUAC could be calculated, and the results can be applied to study the distillation of wine.

Experimental Section

2-Methyl-1-propanol ($x > 99.5$) was supplied by Fluka and was used without further purification. 2-Methyl-1-butanol ($x > 99$) from Aldrich and 3-methyl-1-butanol ($x > 99$) and 1-pentanol ($x > 99$) from Fluka were purified by distillation in a laboratory column of 100 plates. The purity of the material was checked by gas−liquid chromatography (GLC, $x > 99.6$). All products were degassed using ultrasound and dried on molecular sieves (pore diameter $3 \cdot 10^{-10}$ m from Fluka) before use. Densities, refractive indices, and normal boiling points of the pure substances are given in Table 1 and compared with the literature values of Riddick et al.[12] The still used to measure the VLE data was a dynamic recirculating apparatus described by Resa et al.[10] The equilibrium temperature was measured with a digital platinum 100 resistance thermometer with an uncertainty of ± 0.01 K. For the pressure measurement, a digital manometer regulator (Divatronic DT1 model) manufactured by Leybold with an uncertainty of ± 0.1 kPa was used. Both vapor- and liquid-phase compositions for the two systems were determined by densimetry and refractometry. Densities were measured at 298.15 K by using an Anton Paar DMA 58 vibrating-tube densimeter with an uncertainty of ± 0.00001 $\mathrm{g \cdot cm^{-3}}$ that had been calibrated at atmospheric pressure with twice distilled water and dry air. The temperature of the densimeter was maintained at 298.15 K with a uncertainty of ± 0.01 K by means of a semiconductor Peltier element and measured by a calibrated platinum resistance thermometer. Refractive indices were measured with a Mettler RE50 refractometer with an uncertainty of ± 0.00001, and temperature was controlled like the densimeter with a temperature uncertainty of ± 0.01 K. Prior to measurements, density calibration and refractive index for these systems were obtained to calculate the compositions of the vapor and liquid phases. The binary mixtures were prepared by directly weighing the constituent components with an electronic balance (Salter model ER-182A) that has an uncertainty of ± 0.0001 g. Precautions were taken in order to minimize evaporation losses during storage and preparation of the solutions. The estimated uncertainty in the determination of both liquid- and vapor-phase mole fractions is ± 0.001. Table 2 shows the density and refractive index composition values.

Table 1. Physical Properties of Pure Compounds[a]

	$\rho/(\mathrm{kg \cdot m^{-3}})$		n_D		T_b/K	
	exptl	lit.[b]	exptl	lit.[b]	exptl	lit.[b]
2-methyl-1-propanol	797.84	797.8	1.39370	1.39389	381.00	381.036
2-methyl-1-butanol	814.87	815.0	1.40872	1.4086	401.93	401.9
3-methyl-1-butanol	806.78	807.1	1.40515	1.4052	404.27	403.7
1-pentanol	810.96	810.80	1.40770	1.4080	410.85	411.133

[a] Densities ρ and refractive indices n_D at 298.15 K and normal boiling points T_b. [b] Ref 12.

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Table 2. Densities ρ and Refractive Indices n_D for 2-Methyl-1-propanol (1) + 2-Methyl-1-butanol (2), 2-Methyl-1-propanol (1) + 3-Methyl-1-butanol (2), and 2-Methyl-1-propanol (1) + 1-Pentanol (2) at 298.15 K

2-methyl-1-propanol (1) + 2-methyl-1-butanol (2)			2-methyl-1-propanol (1) + 3-methyl-1-butanol (2)			2-methyl-1-propanol (1) + 1-pentanol (2)		
x_1	ρ/(g·cm^{-3})	n_D	x_1	ρ/(g·cm^{-3})	n_D	x_1	ρ/(g·cm^{-3})	n_D
0.051	0.81419	1.40802	0.051	0.80595	1.40469	0.050	0.81030	1.40710
0.101	0.81353	1.40742	0.099	0.80561	1.40418	0.100	0.80970	1.40650
0.151	0.81285	1.40674	0.150	0.80525	1.40371	0.150	0.80909	1.40588
0.200	0.81217	1.40606	0.199	0.80488	1.40318	0.200	0.80851	1.40524
0.250	0.81149	1.40538	0.254	0.80448	1.40260	0.249	0.80794	1.40460
0.300	0.81075	1.40466	0.300	0.80415	1.40212	0.299	0.80734	1.40394
0.350	0.81003	1.40397	0.350	0.80377	1.40161	0.350	0.80675	1.40326
0.401	0.80929	1.40328	0.400	0.80339	1.40109	0.400	0.80613	1.40259
0.450	0.80855	1.40252	0.445	0.80305	1.40060	0.450	0.80553	1.40193
0.500	0.80779	1.40178	0.500	0.80262	1.39997	0.499	0.80490	1.40124
0.549	0.80702	1.40107	0.549	0.80223	1.39940	0.549	0.80429	1.40055
0.599	0.80625	1.40029	0.600	0.80183	1.39879	0.600	0.80364	1.39983
0.650	0.80545	1.39946	0.650	0.80142	1.39821	0.650	0.80301	1.39910
0.701	0.80463	1.39866	0.700	0.80064	1.39764	0.699	0.80241	1.39834
0.749	0.80386	1.39790	0.751	0.80019	1.39701	0.750	0.80173	1.39758
0.800	0.80300	1.39706	0.799	0.79974	1.39636	0.800	0.80107	1.39683
0.850	0.80217	1.39620	0.849	0.79927	1.39567	0.850	0.80039	1.39604
0.899	0.80117	1.39533	0.899	0.79880	1.39500	0.900	0.79967	1.39524
0.950	0.80010	1.39450	0.950	0.79831	1.39435	0.950	0.79884	1.39447

Results and Discussion

The activity coefficients γ_i of the components were calculated from

$$\gamma_i = \frac{y_i \Phi_i P}{x_i P_i^0} \tag{1}$$

where x_i and y_i are the liquid and vapor mole fractions in equilibrium, Φ_i is a vapor-phase correction factor, P is the total pressure, and P_i^0 is the vapor pressure of pure component i. The constants were A_i, B_i, and C_i, and their values were obtained from Riddick et al.[12]

The vapor-phase correction factor is given by

$$\Phi_i = \frac{\phi_i}{\phi_i^{\text{sat}}} \exp\left[-\frac{V_i(P - P_i^0)}{RT}\right] \tag{2}$$

where ϕ_i is the fugacity coefficient of component i in the mixture, ϕ_i^{sat} is the fugacity coefficient at saturation, and V_i is the molar volume of component i in the liquid phase.

VLE data (T, x_1, y_1) for 2-methyl-1-propanol (1) + 2-methyl-1-butanol (2), 2-methyl-1-propanol (1) + 3-methyl-1-butanol (2). and 2-methyl-1-propanol + 1-pentanol binary systems at 101.3 kPa are presented in Table 3. The $T-x_1-y_1$ phase diagrams are shown in Figures 1, 2, and 3.

The activity coefficients were correlated with the Margules,[13] van Laar,[14] Wilson,[15] NRTL,[16] and UNIQUAC[17] equations. To determine the constants of each model, we have used the method "VLE calc" suggested by Gess et al.[18] Estimation of the parameters for the equation was based on the iterative solution, using the maximum likelihood regression of the objective function Q_i,[19] with the activity coefficients obtained from the consistency test as experimental values:

$$Q_i = \sum\left(\frac{\gamma_{\text{exptl}} - \gamma_{\text{calcd}}}{\gamma_{\text{exptl}}}\right)^2 \tag{3}$$

where γ_{exptl} are the activity coefficients calculated from experimental data and γ_{calcd} are the coefficients calculated with the correlations. The parameters, the average deviation in T (ΔT), and the average deviation in y (Δy) are listed in Table 4. Also, the ASOG[20] method was used to obtain predictions in Figures 1 to 3.

The thermodynamic consistency of the experimental data was checked by means od the Dechema test,[21] where the fugacity coefficients are calculated by the method of Hayden and O'Connel[22] and activity coefficients are calculated by using the four-suffix Margules equation:

$$\bar{g}^E/RT = x_1 x_2 [A x_2 + B x_1 - D x_1 x_2] \tag{4}$$

with the corresponding activity coefficients:

$$\ln \gamma_1 = x_2^2[A + 2(B - A - D)x_1 + 3Dx_1^2] \tag{5}$$

$$\ln \gamma_2 = x_1^2[B + 2(A - B - D)x_2 + 3Dx_2^2] \tag{6}$$

Parameters A, B, and D were estimated using the error-in-variables regression maximum likelihood technique. The constraint equation for the regression was

$$F = P - \left(\frac{x_1 \gamma_1{}^* f_1^0}{\phi_1} + \frac{x_2 \gamma_2{}^* f_2^0}{\phi_2}\right) \tag{7}$$

Here the asterisk (*) denotes a calculated or predicted value. An experimental value has no asterisk; f_1^0 and f_2^0 are the standard state fugacities. The errors in the prediction of y_1 were calculated. Predicted y_1* values were obtained using the equation

$$y_1{}^* = \frac{x_1 \gamma_1{}^* f_1^0}{\phi_1 P^*} \tag{8}$$

An average deviation was calculated from

$$\text{average deviation} = \frac{\sum_{i=1}^{n} |\Delta y|}{n} \tag{9}$$

Here $\Delta y = y_1 - y_1{}^*$ and n = number of experimental data points. To pass the consistency test, a system must have an average deviation less than 0.01. The three systems included in this work have passed this consistency test. In Table 5, we show these results and the values A, B and D of eqs 5 and 6.

Table 3. Vapor−Liquid Equilibrium Data for 2-Methyl-1-propanol (1) + 2-Methyl-1-butanol (2), 2-Methyl-1-propanol (1) + 3-Methyl-1-butanol (2), and 2-Methyl-1-propanol (1) + 1-Pentanol (2) Systems[a]

x_1	y_1	T/K	γ_1	γ_2	ϕ_1	ϕ_2	ϕ_1^s	ϕ_2^s
2-Methyl-1-propanol (1) + 2-Methyl-1-butanol (2)								
0.060	0.119	400.18	1.22	0.99	0.9715	0.9620	0.9526	0.9640
0.111	0.204	398.73	1.19	0.99	0.9710	0.9614	0.9541	0.9652
0.155	0.284	397.23	1.24	0.99	0.9705	0.9609	0.9556	0.9664
0.201	0.350	395.63	1.24	1.00	0.9700	0.9602	0.9572	0.9677
0.251	0.416	394.26	1.23	1.00	0.9695	0.9597	0.9585	0.9688
0.276	0.451	393.43	1.25	1.00	0.9692	0.9594	0.9593	0.9694
0.297	0.461	393.11	1.20	1.03	0.9691	0.9593	0.9596	0.9697
0.322	0.503	392.66	1.23	1.00	0.9690	0.9591	0.9601	0.9700
0.352	0.525	392.18	1.19	1.02	0.9688	0.9589	0.9605	0.9704
0.394	0.579	391.35	1.21	0.99	0.9685	0.9586	0.9613	0.9710
0.511	0.686	389.42	1.17	0.98	0.9678	0.9579	0.9630	0.9724
0.543	0.708	388.65	1.17	1.00	0.9676	0.9576	0.9637	0.9729
0.606	0.756	387.44	1.17	1.01	0.9673	0.9573	0.9644	0.9735
0.653	0.793	386.61	1.17	1.01	0.9672	0.9571	0.9648	0.9738
0.697	0.823	385.75	1.17	1.02	0.9669	0.9568	0.9655	0.9744
0.735	0.852	385.07	1.18	1.00	0.9666	0.9564	0.9662	0.9749
0.793	0.891	384.07	1.18	0.98	0.9663	0.9561	0.9668	0.9754
0.842	0.925	383.30	1.19	0.91	0.9660	0.9557	0.9676	0.9761
0.897	0.951	382.55	1.18	0.95	0.9657	0.9554	0.9683	0.9766
0.962	0.983	381.52	1.18	0.94	0.9654	0.9551	0.9689	0.9770
0.976	0.989	381.23	1.18	0.94	0.9651	0.9547	0.9697	0.9777
2-Methyl-1-propanol (1) + 3-Methyl-1-butanol (2)								
0.056	0.120	402.33	1.17	0.98	0.9722	0.9617	0.9503	0.9634
0.103	0.209	400.73	1.17	0.97	0.9716	0.9611	0.9520	0.9648
0.305	0.490	395.19	1.10	0.98	0.9697	0.9591	0.9576	0.9693
0.328	0.515	394.73	1.09	0.98	0.9696	0.9589	0.9581	0.9697
0.357	0.561	393.77	1.13	0.96	0.9693	0.9586	0.9590	0.9704
0.385	0.598	393.05	1.14	0.94	0.9690	0.9583	0.9597	0.9710
0.408	0.622	392.41	1.14	0.94	0.9688	0.9581	0.9603	0.9714
0.447	0.657	391.51	1.14	0.94	0.9685	0.9577	0.9611	0.9721
0.482	0.682	390.74	1.12	0.96	0.9683	0.9574	0.9618	0.9727
0.516	0.700	390.22	1.09	0.99	0.9681	0.9572	0.9623	0.9731
0.540	0.727	389.51	1.11	0.97	0.9678	0.9569	0.9630	0.9736
0.574	0.753	388.88	1.11	0.97	0.9676	0.9567	0.9635	0.9740
0.594	0.765	388.43	1.10	0.98	0.9675	0.9565	0.9639	0.9743
0.633	0.789	387.87	1.09	0.99	0.9673	0.9563	0.9644	0.9747
0.647	0.808	387.39	1.11	0.96	0.9671	0.9561	0.9648	0.9751
0.690	0.837	386.27	1.12	0.97	0.9667	0.9556	0.9658	0.9758
0.746	0.873	385.18	1.12	0.96	0.9664	0.9552	0.9667	0.9765
0.787	0.900	384.51	1.12	0.92	0.9661	0.9549	0.9673	0.9770
0.839	0.925	383.60	1.12	0.95	0.9658	0.9546	0.9680	0.9776
0.885	0.952	382.80	1.12	0.88	0.9655	0.9542	0.9687	0.9781
0.914	0.963	382.33	1.12	0.93	0.9654	0.9540	0.9691	0.9784
2-Methyl-1-propanol (1) + 1-Pentanol (2)								
0.033	0.091	409.10	1.13	1.00	0.9744	0.9616	0.9427	0.9641
0.077	0.189	407.16	1.06	1.00	0.9737	0.9609	0.9449	0.9657
0.118	0.270	405.35	1.04	1.00	0.9731	0.9602	0.9470	0.9671
0.165	0.344	403.60	1.00	1.01	0.9725	0.9596	0.9489	0.9685
0.218	0.431	401.45	1.01	1.00	0.9718	0.9588	0.9512	0.9701
0.270	0.496	399.87	0.99	1.00	0.9713	0.9583	0.9529	0.9713
0.293	0.528	398.85	1.00	1.01	0.9709	0.9579	0.9540	0.9720
0.327	0.581	397.60	1.02	0.98	0.9705	0.9575	0.9552	0.9729
0.356	0.605	396.64	1.01	1.00	0.9702	0.9571	0.9562	0.9736
0.383	0.636	395.89	1.01	0.99	0.9700	0.9568	0.9569	0.9741
0.405	0.668	394.94	1.03	0.96	0.9696	0.9565	0.9579	0.9747
0.446	0.696	393.87	1.01	0.98	0.9663	0.9530	0.9589	0.9754
0.471	0.718	393.07	1.01	0.99	0.9690	0.9557	0.9597	0.9760
0.503	0.739	392.28	1.00	1.00	0.9688	0.9554	0.9604	0.9765
0.517	0.750	391.92	1.00	1.00	0.9686	0.9553	0.9607	0.9767
0.546	0.768	391.13	0.99	1.01	0.9684	0.9550	0.9615	0.9772
0.658	0.847	388.36	1.00	0.99	0.9674	0.9539	0.9640	0.9789
0.759	0.907	385.66	1.01	0.94	0.9665	0.9527	0.9663	0.9804
0.869	0.956	383.48	1.01	0.89	0.9658	0.9518	0.9681	0.9816
0.917	0.975	382.53	1.00	0.82	0.9655	0.9515	0.9687	0.9820

[a] x_1, liquid-phase mole fraction; y_1, vapor-phase mole fraction; T, boiling temperature; γ_1 and γ_2, activity coefficients; ϕ_1 and ϕ_2, fugacity coefficients; and ϕ_1^s and ϕ_2^s, fugacity coefficients at saturation at 101.3 kPa.

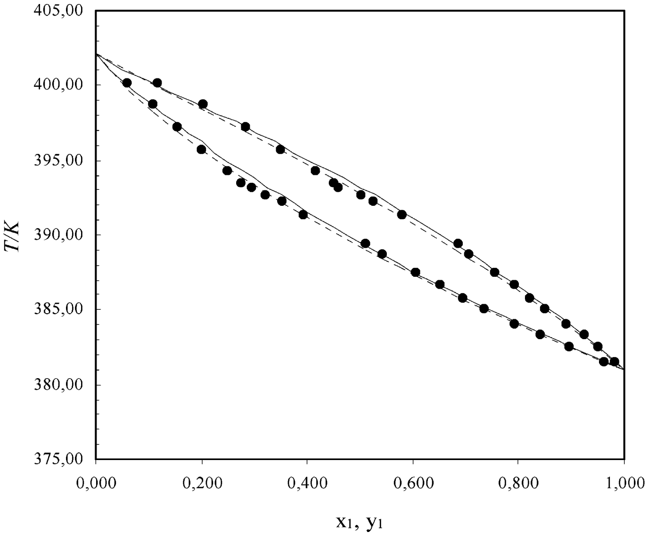

Figure 1. $T-x_1-y_1$ diagram for 2-methyl-1-propanol (1) + 2-methyl-1-butanol (2) at 101.3 kPa: ●, experimental data; - - -, Wilson correlation; —, ASOG prediction.

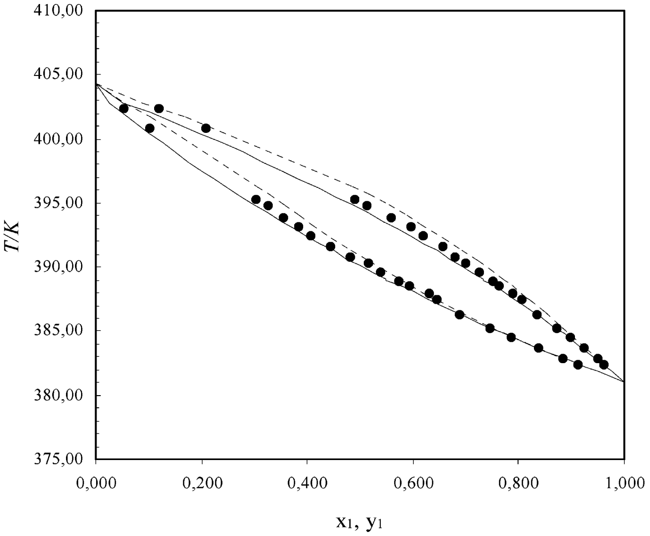

Figure 2. $T-x_1-y_1$ diagram for 2-methyl-1-propanol (1) + 3-methyl-1-butanol (2) at 101.3 kPa: ●, experimental data; - - -, Wilson correlation; —, ASOG prediction.

We also carried out the Margules constant test using the program of Gess et al.[17] The Margules constant can be used to indicate the ideality of a system. Systems that yield a Margules constant whose absolute value is less than 0.60 can be considered ideal, while those that yield an absolute value greater than 0.60 can be considered nonideal. This criterion for classification, however, is not rigorous. Table 6 shows the values of this constant.

Conclusions

New VLE data not previously reported in the literature have been measured for the systems 2-methyl-1-propanol with (2-methyl-1-butanol, 3-methyl-1-butanol, and 1-pentanol) as well as binary parameters, values of different correlations, and necessary physical properties for modeling and simulation of wine distillation.

Three systems present near to ideal behavior according to Margules test and activity coefficients. The ASOG method prediction has a perfect agreement with experimental data in the case of the 2-methyl-1-propanol + 1-pentanol system. For the 2-methyl-1-propanol + 2-methyl-1-butanol system, the ASOG prediction presents a slight deviation; for the 2-methyl-

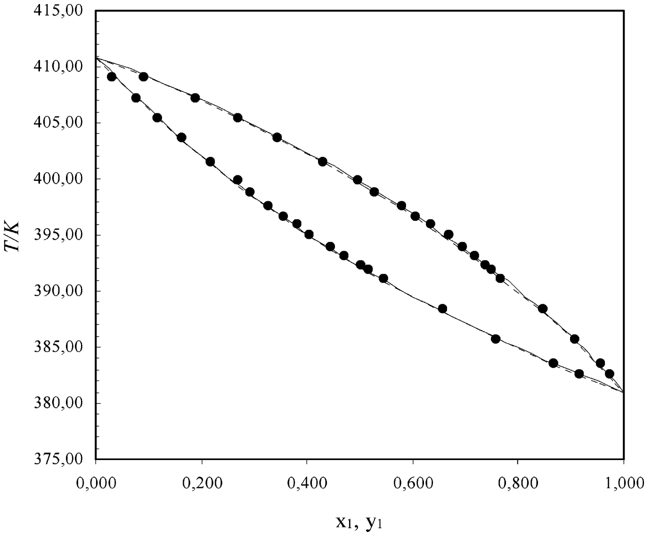

Figure 3. $T-x_1-y_1$ diagram for 2-methyl-1-propanol (1) + 1-pentanol (2) at 101.3 kPa: ●, experimental data; - - -, Wilson correlation; —, ASOG prediction.

Table 4. Correlation Parameters for Activity Coefficients and Average Deviation for the Studied Systems

equation	A_{12}	A_{21}	ΔT/K	Δy_1
2-Methyl-1-propanol (1) + 2-Methyl-1-butanol (2)				
Margules[a]	0.1043	0.0157	0.19	0.008
van Laar[a]	0.0882	0.0433	0.21	0.007
Wilson[b]	2630.7	−1937.4	0.20	0.007
NRTL[c] ($\alpha_{12} = 0.28$)	−2303.1	3148.1	0.19	0.007
UNIQUAC[d]	2062.3	−1483.5	0.22	0.006
2-Methyl-1-propanol (1) + 3-Methyl-1-butanol (2)				
Margules[a]	−0.3176	0.0004	0.24	0.011
van Laar[a]	−6.6165	−0.0734	0.10	0.006
Wilson[b]	1991.4	−2059.7	0.27	0.016
NRTL[c] ($\alpha_{12} = 2.23$)	−705.36	572.07	0.24	0.015
UNIQUAC[d]	3408.4	−2273.3	0.24	0.012
2-Methyl-1-propanol (1) + 1-Pentanol (2)				
Margules[a]	0.0136	0.0042	0.11	0.008
van Laar[a]	0.0013	−17.327	0.12	0.008
Wilson[b]	1716.8	−1505.4	0.11	0.008
NRTL[c] ($\alpha_{12} = 0.30$)	11.106	16.4048	0.11	0.008
UNIQUAC[d]	2315.7	−1697.6	0.14	0.007

[a] Margules and van Laar constants (dimensionless). [b] Wilson's interaction parameters ($J\cdot mol^{-1}$). [c] NRTL's interaction parameters ($J\cdot mol^{-1}$). [d] UNIQUAC's interaction parameters ($J\cdot mol^{-1}$).

Table 5. Results of the Thermodynamic Consistency Test

system	average deviation Δy_1	*A*	*B*	*D*
2-methyl-1-propanol (1) + 2-methyl-1-butanol (2)	0.007	0.0141	−0.0523	−0.3533
2-methyl-1-propanol (1) + 3-methyl-1-butanol (2)	0.008	−0.5736	−0.1534	−0.8434
2-methyl-1-propanol (1) + 1-pentanol (2)	0.009	0.0484	0.118	0.0842

Table 6. Results of the Margules Constant Test

system	Margules constant
2-methyl-1-propanol (1) + 2-methyl-1-butanol (2)	0.0328
2-methyl-1-propanol (1) + 3-methyl-1-butanol (2)	−0.1836
2-methyl-1-propanol (1) + 1-pentanol (2)	−0.0242

1-propanol + 3-methyl-1-butanol system, this deviation is bigger.

Correlations for the 2-methyl-1-propanol + 2-methyl-1-butanol system are very similar. Better correlation for the 2-methyl-1-propanol + 3-methyl-1-butanol system is van Laar's equation. Those of 2-methyl-1-propanol + 1-pentanol are very similar for all correlation deviations (shown in Table 4).

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