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Density, Refractive Index, and Speed of Sound at 298.15 K and Vapor−Liquid Equilibria at 101.3 kPa for Binary Mixtures of Ethyl Acetate + 1-Pentanol and Ethanol + 2-Methyl-1-propanol

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Densities, refractive indices, and speeds of sound at 298.15 K and isobaric vapor−liquid equilibria (VLE) data at 101.3 kPa were reported for the binary mixtures containing ethyl acetate + 1-pentanol and ethanol + 2-methyl-1-propanol. Excess molar volumes, refractive index deviations, and changes of speed of sound on mixing were calculated from the measurement results that were fitted with Redlich−Kister polynomials. VLE experimental data were tested for thermodynamic consistency by means of a modified Dechema test and were demonstrated to be consistent. The activity coefficients were correlated with the Margules, van Laar, UNIQUAC, nonrandom two-liquid, and Wilson equations. The analytical solution of groups model also was used for prediction.

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. 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. By 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, nonrandom two liquid (NRTL), and UNIQUAC would be calculated, and results can be applied to study the distillation of wine. We have measured the VLE at 101.3 kPa of ethyl acetate + 1-pentanol and ethanol + 2-methyl-1-propanol. There are no bibliographical references on ethyl acetate + 1-pentanol, and several references on ethanol + 2-methyl-1-propanol have been found, but these data are isothermal ones from Oracz[1] and Bhethanabotla and Campbell.[2]

Experimental Section

Materials. Ethyl acetate (99.9 mol %), 1-pentanol (99 mol %), and 2-methyl-1-propanol (99 mol %) were supplied by Fluka. Ethanol (99.5 mol %) was supplied by Panreac and was used without further purification. 1-Pentanol and 2-methyl-1-propanol were purified by distillation in a 100-plate laboratory column. The purity of the materials so obtained was checked by gas liquid chromatography and was found to be better than 99.7 mol %. All products were ultrasonically degassed and dried over molecular sieves (pore diameter of 3×10^{-10} m from Fluka) before use. Densities, refractive indices, speeds of sound, and normal boiling points of the pure substances are listed in Table 1 and compared with literature values from Riddick et al.[3]

Apparatus and Procedures. The still used to measure VLE data was a dynamic recirculating apparatus described by Resa et al.[4] The equilibrium temperature was measured with a digital platinum 100 resistance thermometer with an accuracy of ±0.1 K. For the pressure measurement, a digital manometer regulator (Divatronic DT1 model), manufactured by Leybold with an accuracy of ±0.1 kPa, was used. Both vapor- and liquid-phase compositions for the two systems were determined by densimetry. Densities were measured at 298.15 K by using an Anton Paar DMA 58 vibrating-tube densimeter with an accuracy of ±0.00001 $g\cdot cm^{-3}$ 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 precision 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 accuracy of ±0.00001, and temperature was controlled like the densimeter, with a temperature precision of ±0.01 K. Speeds of sound were measured with an Anton Paar DSA 48 sound analyzer with an accuracy of ±0.1 $m\cdot s^{-1}$, and temperature was controlled by a Peltier cooler to a precision of ±0.1 K. The binary mixtures were prepared by directly weighing the constituent components with an electronic balance (Salter model ER-182A) that has an accuracy 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 mol fractions is ±0.001.

Results and Discussion

Density, Refractive Index, and Speed of Sound. Table 2 lists the measured density ρ, refractive index n_D, and speed of sound u, data at 298.15 K with the corresponding excess molar volume V^E, refractive index deviation δn_D, and speed of sound deviation δu for the binary mixtures ethyl acetate + 1-pentanol and ethanol + 2-methyl-1-propanol.

The excess molar volumes of binary mixtures were calculated from density measurements by applying

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Table 1. Physical Properties of Pure Compounds: Densities ρ, Refractive Indices n_D, Speeds of Sound u, at 298.15 K, and Normal Boiling Points T_b

	ρ/(kg·m^{-3})		n_D		u/(m·s^{-1})		T_b/K	
	obs	lit[a]	obs	lit[a]	obs	lit[a]	obs	lit[a]
ethyl acetate	894.24	894.55	1.36980	1.36978	1137.66	not available	350.25	350.26
1-pentanol	810.96	810.80	1.40770	1.40800	1273.31	not available	410.59	411.13
ethanol	785.36	784.93	1.35916	1.35941	1143.11	not available	351.50	351.44
2-methyl-1-propanol	797.84	797.80	1.39370	1.39389	1185.63	not available	381.00	381.04

[a] Riddick et al.[4]

Table 2. Densities, Refractive Indices, and Speeds of Sound for Ethyl Acetate (1) + 1-Pentanol (2) and Ethanol (1) + 2-Methyl-1-propanol (2) at 298.15 K with Excess Molar Volume V^E, Refractive Index Deviation δn_D, and Speeds of Sound Deviation δu

x_1	ρ/(kg·m^{-3})	V^E/(cm^3·mol^{-1})	n_D	δn_D	u/(m·s^{-1})	δu/(m·s^{-1})
		Ethyl Acetate (1) + 1-Pentanol (2)				
0.050	814.16	0.075	1.40579	−0.00003	1263.5	−3.1
0.101	817.63	0.138	1.40382	−0.00005	1253.8	−5.8
0.150	821.07	0.180	1.40193	−0.00008	1245.0	−7.9
0.200	824.60	0.226	1.40012	−0.00010	1236.5	−9.7
0.251	828.30	0.262	1.39803	−0.00013	1227.5	−11.8
0.300	831.90	0.297	1.39629	−0.00014	1219.9	−12.7
0.350	835.73	0.320	1.39423	−0.00015	1211.9	−13.9
0.400	839.69	0.328	1.39250	−0.00014	1204.5	−14.5
0.450	843.69	0.335	1.39052	−0.00014	1198.2	−14.1
0.500	847.81	0.335	1.38863	−0.00014	1190.5	−15.0
0.549	851.89	0.333	1.38676	−0.00013	1184.1	−14.8
0.599	856.06	0.334	1.38493	−0.00014	1178.8	−13.3
0.650	860.53	0.315	1.38303	−0.00012	1172.1	−13.0
0.700	865.01	0.291	1.38107	−0.00012	1166.8	−11.7
0.749	869.40	0.278	1.37919	−0.00011	1161.7	−10.0
0.800	874.19	0.232	1.37733	−0.00009	1157.1	−7.7
0.850	879.00	0.187	1.37548	−0.00008	1152.2	−5.8
0.898	883.69	0.141	1.37360	−0.00005	1148.0	−3.6
0.947	888.68	0.082	1.37188	−0.00002	1144.1	−0.7
		Ethyl Acetate (1) + 2-Methyl-1-propanol (2)				
0.052	797.95	−0.061	1.39253	0.00062	1186.09	2.7
0.104	797.48	−0.054	1.39136	0.00123	1184.42	3.2
0.155	796.99	−0.047	1.39012	0.00176	1182.91	3.9
0.206	796.46	−0.038	1.38890	0.00231	1181.47	4.6
0.258	795.94	−0.032	1.38753	0.00270	1179.29	4.6
0.308	795.35	−0.021	1.38611	0.00303	1177.47	5.0
0.359	794.79	−0.016	1.38470	0.00337	1175.62	5.3
0.410	794.16	−0.006	1.38330	0.00371	1174.15	5.9
0.460	793.58	−0.004	1.38169	0.00383	1171.26	5.2
0.510	793.00	−0.004	1.38030	0.00416	1169.01	5.1
0.560	791.89	0.043	1.37842	0.00400	1165.22	3.4
0.609	791.22	0.045	1.37674	0.00403	1162.97	3.2
0.659	790.55	0.044	1.37505	0.00404	1160.41	2.8
0.708	789.90	0.038	1.37301	0.00370	1158.25	2.7
0.757	789.15	0.038	1.37111	0.00349	1155.32	1.9
0.806	788.40	0.034	1.36884	0.00291	1152.79	1.4
0.855	787.68	0.025	1.36675	0.00250	1150.10	0.8
0.904	786.87	0.020	1.36431	0.00173	1147.39	0.2
0.952	786.08	0.009	1.36189	0.00097	1144.67	−0.5

the equation

$$V^E = x_1M_1(1/\rho - 1/\rho_1) + x_2M_2(1/\rho - 1/\rho_2) \qquad (1)$$

where ρ is the density of the mixture, ρ_1 and ρ_2 are the densities of the pure substances, M_1 and M_2 are the molar masses, and x_1 and x_2 are the mol fractions. The uncertainty in the calculation of V^E from density measurements was estimated to be ±0.001 cm^3·mol^{-1}. Figure 1 illustrates the excess molar volumes of the two binary systems at 298.15 K.

The changes of refractive index δn_D at 298.15 K from the linear additive value of the mol fraction is obtained by

$$\delta n_D = n_D - (x_1n_{D1} + x_2n_{D2}) \qquad (2)$$

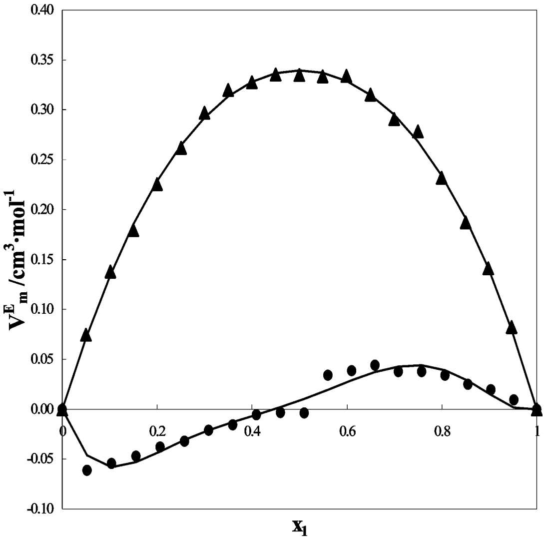

Figure 1. Excess molar volumes of mixtures of ▲, ethyl acetate (1) + 1-pentanol (2); ●, ethanol(1) + 2-methyl-1-propanol (2). Solid lines are Redlich−Kister fit curves at 298.15 K.

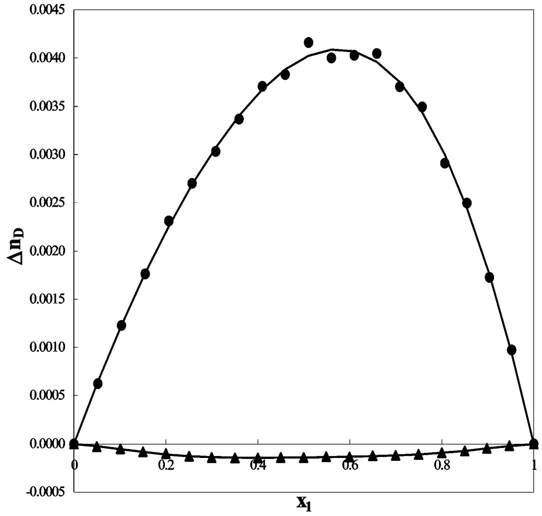

Figure 2. Change of refractive indices on mixing ▲, ethyl acetate (1) + 1-pentanol (2); ●, ethanol (1) + 2-methyl-1-propanol (2). Solid lines are Redlich−Kister fit curves at 298.15 K.

where n_D is the refractive index of the mixture and n_{D1} and n_{D2} are the refractive indices of the pure compounds. The plot of δn_D vs the mol fraction x_1 of the most volatile compound of each binary system is given in Figure 2.

In the same way, the changes of speed of sound on mixing were calculated by the equation

$$\delta u = u - (x_1u_1 + x_2u_2) \qquad (3)$$

where u is the speed of sound of the mixture and u_1 and u_2 are the speeds of sound of the pure compounds. The plot

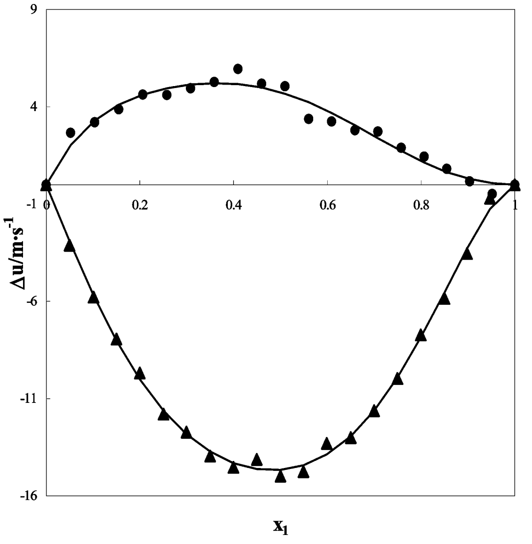

Figure 3. Change of speed sounds on mixing ▲, ethyl acetate (1) + 1-pentanol (2); ●, ethanol (1) + 2-methyl-1-propanol (2). Solid lines are Redlich−Kister fit curves at 298.15 K.

Table 3. Adjustable Parameters a_k with the Standard Deviations σ for Excess Molar Volumes V^E, Refractive Index Deviations δn_D, and Speeds of Sound Deviations δu

	V^E (cm^3·mol^{-1})	δn_D	δu (m·s^{-1})
	Ethyl Acetate (1) + 1-Pentanol (2)		
a_0	1.358	−0.0006	−58.7
a_1	0.007	0.0001	4.0
a_2	0.250	−0.0004	−0.9
a_3	0.055	−0.0001	20.3
a_4	−0.020	0.0006	25.0
σ	0.004 (cm^3·mol^{-1})	0.0000	0.3 (m·s^{-1})
	Ethanol (1) + 2-Methyl-1-propanol (2)		
a_0	0.034	0.0160	19.0
a_1	0.344	0.0048	−14.0
a_2	0.248	0.0013	−7.3
a_3	0.255	−0.0008	−9.7
a_4	−1.045	−0.0004	13.0
σ	0.056 (cm^3·mol^{-1})	0.0006	1.0 (m·s^{-1})

of δu vs the mol fraction x_1 of the more volatile compound of each binary system is given in Figure 3.

Excess molar volumes, changes of refractive indices, and speeds of sound on mixing of the binary systems were fitted to Redlich−Kister polynomials of the form

$$(V^E \text{ or } \delta n_D \text{ or } \delta u) = x_1 x_2 \sum_{k\geq 0} a_k (x_1 - x_2)^k \qquad (4)$$

where a_k is the adjustable parameter obtained by a least-squares fit method and k is the degree of the polynomial expansion. Table 3 lists the parameters with their standard deviations σ. The coefficients a_k were used to calculate the solid curves; see Figures 1−3. The standard deviations σ are defined as follows

$$\sigma = \left(\frac{\sum (F_{\text{cal}} - F_{\text{exp}})_i^2}{N - m}\right)^{1/2} \qquad (5)$$

where N is the number of experimental data, m is the number of equation parameters, and F is the considered property (V^E or δn_D or δu).

VLE Data. Vapor−liquid equilibrium data (T,x_1,y_1) for the ethyl acetate + 1-pentanol and ethanol+ 2-methyl-1-propanol binary systems at 101.3 kPa are presented in Table 4. The T−x_1−y_1 phase diagrams are shown in Figures 4 and 5.

Table 4. Vapor−Liquid Equilibrium Data for the Ethyl Acetate + 1-Pentanol and Ethanol + 2-Methyl-1-propanol Systems: Liquid-Phase Mol Fraction x_1, Vapor-Phase Mol Fraction y_1, Boiling Temperature T, Activity Coefficients γ_1 and γ_2, Fugacity Coefficients ϕ_1 and ϕ_2, and Fugacity Coefficients at Saturation ϕ_1^s and ϕ_2^s at 101.3 kPa

x_1	y_1	T/K	γ_1	γ_2	ϕ_1	ϕ_2	ϕ_1^s	ϕ_2^s
		Ethyl Acetate (1) + 1-Pentanol (2)						
0.000	0.000	410.59						
0.022	0.145	406.75	1.526	1.009	0.978	0.961	0.890	0.966
0.065	0.327	400.66	1.328	1.019	0.976	0.959	0.899	0.971
0.119	0.495	393.93	1.276	1.028	0.974	0.956	0.909	0.976
0.153	0.564	390.53	1.226	1.047	0.973	0.955	0.913	0.978
0.199	0.652	385.89	1.218	1.053	0.971	0.953	0.919	0.980
0.250	0.724	381.19	1.210	1.072	0.970	0.951	0.925	0.983
0.292	0.777	377.32	1.227	1.073	0.969	0.950	0.930	0.985
0.341	0.810	374.23	1.188	1.116	0.968	0.948	0.934	0.986
0.391	0.843	371.67	1.155	1.113	0.967	0.947	0.937	0.987
0.442	0.869	368.96	1.134	1.140	0.966	0.946	0.940	0.988
0.489	0.888	366.77	1.113	1.173	0.965	0.944	0.942	0.989
0.528	0.905	364.74	1.112	1.180	0.964	0.943	0.944	0.990
0.573	0.918	363.12	1.088	1.212	0.964	0.942	0.946	0.991
0.608	0.927	361.85	1.074	1.247	0.963	0.942	0.947	0.991
0.637	0.934	360.87	1.063	1.274	0.963	0.941	0.948	0.991
0.683	0.944	359.42	1.045	1.325	0.962	0.940	0.950	0.992
0.698	0.946	359.02	1.036	1.367	0.962	0.940	0.950	0.992
0.745	0.959	357.33	1.035	1.333	0.962	0.939	0.952	0.992
0.781	0.964	356.36	1.020	1.429	0.961	0.939	0.953	0.993
0.882	0.982	353.45	1.007	1.529	0.960	0.937	0.956	0.993
0.936	0.991	351.96	0.995	1.520	0.959	0.936	0.957	0.994
1.000	1.000	350.25						
		Ethanol (1) + 2-Methyl-1-propanol (2)						
0.000	0.000	381.00						
0.012	0.035	380.36	1.058	0.996	0.982	0.965	0.946	0.970
0.085	0.205	377.01	0.980	0.999	0.981	0.964	0.950	0.973
0.121	0.290	375.21	1.034	0.991	0.981	0.963	0.952	0.974
0.163	0.365	373.55	1.021	0.993	0.980	0.965	0.953	0.976
0.199	0.427	372.07	1.026	0.988	0.979	0.962	0.955	0.977
0.238	0.480	370.69	1.011	0.993	0.979	0.961	0.956	0.978
0.270	0.527	369.51	1.016	0.987	0.979	0.961	0.958	0.978
0.299	0.567	368.50	1.022	0.979	0.978	0.961	0.959	0.979
0.328	0.597	367.65	1.008	0.984	0.978	0.960	0.960	0.979
0.362	0.632	366.54	1.006	0.988	0.978	0.960	0.961	0.980
0.419	0.686	364.78	1.004	0.991	0.977	0.959	0.962	0.981
0.455	0.717	363.66	1.004	0.998	0.977	0.959	0.963	0.982
0.497	0.753	362.59	1.003	0.985	0.977	0.959	0.964	0.982
0.535	0.778	361.63	0.995	0.998	0.976	0.958	0.965	0.983
0.600	0.821	359.96	0.995	1.003	0.976	0.958	0.967	0.984
0.632	0.842	359.09	1.000	0.997	0.976	0.957	0.967	0.984
0.667	0.861	358.27	0.999	1.004	0.975	0.957	0.968	0.985
0.802	0.924	355.37	0.993	1.040	0.975	0.956	0.970	0.986
0.856	0.948	354.17	0.998	1.029	0.974	0.955	0.971	0.987
0.912	0.969	353.20	0.993	1.067	0.974	0.955	0.972	0.987
0.947	0.981	352.50	0.994	1.082	0.974	0.954	0.973	0.988
1.000	1.000	351.50						

The activity coefficients γ_i of the components were calculated from

$$\gamma_i = \frac{y_i \Phi_i P}{x_i P_i^0} \qquad (6)$$

where x_i and y_i are the liquid and vapor mol 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. These vapor pressures were calculated from the Antoine equation

$$\log(P_i^0/\text{kPa}) = A_i - \frac{B_i}{(T/\text{K}) + C_i} \qquad (7)$$

The constants A_i, B_i, and C_i are reported in Table 5, and their values were obtained from Riddick et al.[3]

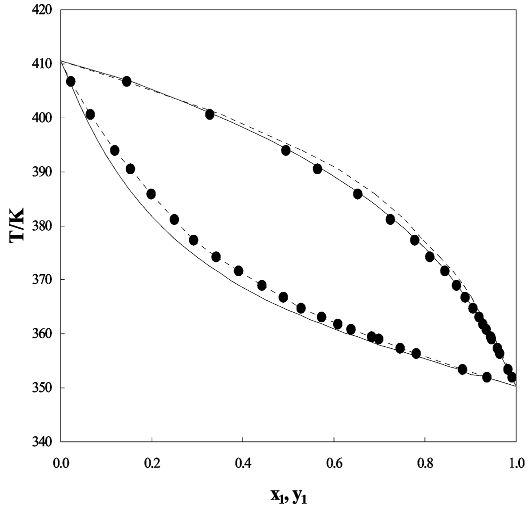

Figure 4. $T-x_1-y_1$ diagram for ethyl acetate (1) + 1-pentanol (2) at 101.3 kPa: ●, experimental data; dashed line, Wilson correlation; solid line, ASOG prediction.

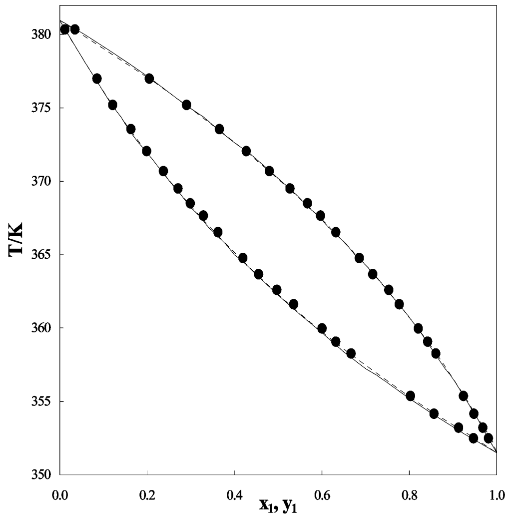

Figure 5. $T-x_1-y_1$ diagram for ethanol (1) + 2-methyl-1-propanol (2) at 101.3 kPa: ●, experimental data; dashed line, Wilson correlation; solid line, ASOG prediction.

Table 5. Antoine Coefficients from Equation 7

compound	A_i	B_i	C_i
ethyl acetate	6.18799	1224.673	−57.430
1-pentanol	6.30306	1286.333	−111.843
ethanol	7.16879	1552.601	−50.731
2-methyl-1-propanol	6.35648	1107.060	−101.048

Table 6. Correlation Parameters for Activity Coefficients and Average Deviation for the Studied Systems

equation	A_{12}	A_{21}	ΔT/K	Δy_1
Ethyl Acetate (1) + 1-Pentanol (2)				
Margules[a]	0.3053	0.4869	0.20	0.0041
van Laar[a]	0.3285	0.5035	0.19	0.0039
Wilson[b] 2	−11.72	1537.13	0.19	0.0036
NRTL[c] ($\alpha_{12} = 0.08$)	2476.23	−896.31	0.19	0.0037
UNIQUAC[d]	4268.99	−2081.38	0.19	0.0047
Ethanol (1) + 2-Methyl-1-propanol (2)				
Margules[a]	0.0089	0.0351	0.06	0.0049
van Laar[a]	0.0132	0.0534	0.06	0.0050
Wilson[b] 2	368.75	−64.15	0.05	0.0051
NRTL[c] ($\alpha_{12} = 0.30$)	2097.86	−1648.64	0.06	0.0048
UNIQUAC[d]	−1241.40	1863.36	0.06	0.0048

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

Table 7. Results of the Thermodynamic Consistency Test

system	average deviation Δy_1	A	B	D
ethyl acetate (1) + 1-pentanol (2)	0.0037	0.3154	0.4948	0.0389
ethanol (1) + 2-methyl-1-propanol (2)	0.0053	0.0335	0.0417	0.0527

The vapor-phase correction factor is given by

$$\Phi_i = \frac{\phi_i}{\phi_i^{\text{sat}}} \exp\left[-\frac{V_i(P - P_i^0)}{RT}\right] \qquad (8)$$

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.

The activity coefficients were correlated with the Margules,[5] van Laar,[6] Wilson,[7] NRTL,[8] and UNIQUAC[9] equations. To determine the constants of each model, we have used the method "VLE calc" suggested by Gess et al.[10] Estimation of the parameters for the equation was based on the iterative solution, using the maximum likelihood regression of the objective function Q_i[11] with the activity coefficients obtained from the consistency test as experimental values

$$Q_i = \sum\left(\frac{\gamma_{\text{exp}} - \gamma_{\text{calc}}}{\gamma_{\text{exp}}}\right)^2 \qquad (9)$$

where γ_{exp} are the activity coefficients calculated from experimental data and γ_{calc} 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 6. Also, the analytical solution of groups (ASOG)[12] method was used to obtain predictions in Figures 4 and 5.

The thermodynamic consistency of the experimental data was checked by means of a modified Dechema test[13] in which the fugacity coefficients are calculated by the method of Hayden and O'Connel[14] and activity coefficients are calculated by use of the four-suffix Margules equation

$$\bar{g}^{\text{E}}/RT = x_1x_2[Ax_2 + Bx_1 - Dx_1x_2] \qquad (10)$$

with the corresponding activity coefficients

$$\ln \gamma_1 = x_2^{\,2}[A + 2(BAD)x_1 + 3Dx_1^{\,2}] \qquad (11)$$

$$\ln \gamma_2 = x_1^{\,2}[B + 2(ABD)x_2 + 3Dx_2^{\,2}] \qquad (12)$$

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

$$F = P - \left(\frac{x_1\gamma_1^*f_1^{\,0}}{\phi_1} + \frac{x_2\gamma_2^*f_2^{\,0}}{\phi_2}\right) \qquad (13)$$

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

$$y_1^* = \frac{x_1 \gamma_1^* f_1^{\,0}}{\phi_1 P^*} \tag{14}$$

An average deviation was calculated from

$$\text{average deviation} = \frac{\sum_{i=1}^{n} |\Delta y|}{n} \tag{15}$$

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 two systems included in this work have passed this consistency test.

Conclusions

Ethanol + 2-methyl-1-propanol form an ideal solution according to the values of activity coefficients, which are close to unity at all composition studied; the ASOG method prediction is in good agreement with experimental data. As the behavior of the binary system ethyl acetate + 1-pentanol is less ideal, the ASOG method shows fair agreement with the experimental values. The five correlation methods tested gave very similar average deviations for both temperatures as vapor-phase mol fractions. The values of the excess molar volume of ethanol + 2-methyl-1-propanol are close to zero, and ethyl acetate + 1-pentanol has a positive value due to an expansive trend. In case of changes of refractive indices, we can point out opposite behavior and values that are very close to zero for the ethanol−1-propanol system. The variation of the speed of sound shows very big values, negative for ethanol + 1-propanol and positive for the ethyl acetate + 1-pentanol system. New data of VLEs are presented for ethanol + 2-methyl-1-propanol and ethyl acetate + 1-pentanol systems not found in the literature, and values of correlation constants necessary for the modeling and simulation in wine distillation has been calculated.

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