JOURNAL OF
CHEMICAL &
ENGINEERING
DATA

ARTICLE

pubs.acs.org/jced

Isobaric Vapor Liquid Equilibrium of 3-Methyl-1-butanol + Ethyl Lactate and 1-Pentanol + Ethyl Lactate at (13.0 and 101.3) kPa

Emilio A. Cepeda*,† and Cipriano Urbano‡

†Departamento de Ingeniería Química and ‡Departamento de Ingeniería Química y Medio Ambiente, Universidad del País Vasco, Apartado 450, 01006, Vitoria, Spain

ABSTRACT: Isobaric vapor−liquid equilibrium (VLE) data were determined for the binary systems 3-methylbutan-1-ol (3-methyl-1-butanol) + ethyl (2*S*)-2-hydroxypropanoate (ethyl lactate) and pentan-1-ol (1-pentanol) + ethyl lactate at two pressures (13.0 and 101.3 kPa). The data were obtained using a vapor recirculating-type equilibrium device. Calculations of the nonideality of the vapor phase were made with the second virial coefficients evaluated from the Hayden−O'Connell method. VLE experimental data were tested by Herrington and a modified DECHEMA test, which proved their thermodynamic consistency. The activity coefficients and boiling points of the solutions were well correlated with the mole fraction using Wilson, NRTL, and UNIQUAC equations. The mixtures do not present azeotropes.

INTRODUCTION

This work is part of a research project[1−11] whose objective is to determine the thermodynamic properties for the binary systems involved in the wine distillation processes. The main components of the mixtures present in wine distillation were water and ethanol. There are also other minor compounds such as alcohols, aldehydes, acetates, and esters which are known as congeners. These secondary products comprise the characteristic aroma of each wine and contribute to their taste.[12] For modeling and process simulation in which mixtures appear, binary data are needed. In this work, the binary mixtures of congeners, 3-methyl-1-butanol, and 1-pentanol with ethyl lactate were considered. There was found no experimental data for these systems in the literature, although similar alcohol−lactate[13−15] systems have been measured. The alcohols with five carbons, 3-methyl-1-butanol, and 1-pentanol are well-known[16,17] components of the wine. 3-Methyl-1-butanol on its own or dissolved in water has an unpleasant odor but, in wine, it favorably contributes to its aroma.[12] 1-Pentanol is one of the most important legal congeners in common alcoholic distillation. These alcohols are the main components of fusel oil obtained from the distillation of wine. Ethyl lactate is the main compound produced during the aging process of grape wine by acidic fermentation of malic and lactic acid. The odor of ethyl lactate when dilute is mild, buttery, and creamy, with hints of fruit and coconut. Moreover, the ethyl lactate is a solvent used to replace organic solvents because it is biodegradable and the vapor−liquid equilibrium (VLE) data is essential for the design of the separation processes.

The aim of this work was the study of the vapor−liquid equilibrium of the systems 3-methyl-1-butanol + ethyl lactate and 1-pentanol + ethyl lactate. The ethyl lactate breaks down at high temperatures, so the VLE was measured at two pressures (13 and 101.3 kPa). This objective included the testing of the consistency of the equilibrium data and to fit the experimental data to classic models.

EXPERIMENTAL SECTION

Materials. Ethyl lactate (ethyl (2*S*)-2-hydroxypropanoate, CAS 687-47-8, $x > 99.0$), 3-methyl-1-butanol (3-methylbutan-1-ol, CAS 123-51-3; AR grade, ≥ 0.99 mass fraction) and 1-pentanol (pentan-1-ol, CAS 71-41-0; AR grade ≥ 0.99 mass fraction) were supplied by Fluka (Sigma-Aldrich Chemie GmbH). The alcohols were treated with potassium bicarbonate as an agent of neutralization and drying and next were vacuum distilled in a 1 m height and 30 mm diameter adiabatic distillation column (packed with 2 mm × 2 mm stainless Dixon rings), working at a 1:100 reflux ratio. All products were degassed using ultrasound and dried on molecular sieves (pore diameter 3 Å from Fluka) before use. The purity of the materials was checked by gas chromatography and was found to be better than 0.998 mass fraction. The density, refractive indices and normal boiling points for the pure compounds are listed in the Table 1 along with their literature values. The estimated uncertainties in the measurements were ± 0.0005 in mole fraction, ± 0.0002 in refractive index, ± 0.05 K in temperature, and ± 14 kPa in pressure.

Apparatus and Procedure. The still used to measure VLE data was our own design of a dynamic recirculating apparatus.[6] In each

Table 1. Physical Properties of the Pure Compounds[a]

	ρ/kg m^{-3}		n_D		T_b/°C	
compound	expt.	lit.	expt.	lit.	expt.	lit.
3-methyl-1-butanol	807.6	807.1[24]	1.4051	1.4052[24]	131.4	131.9[25] 130.5[24] 131.2[26] 132.0[27]
1-pentanol	811.2	810.8[24]	1.4077	1.4080[24]	137.8	137.98[24] 137.8[26] 137.6[28]
ethyl lactate	1028.6	1027.2[24]	1.41057	1.41050[24]	154.5	154.55[24]

[a] Densities, ρ (285.15 K), refractive indices, n_D (298.15 K), and boiling points, T_b (101.3 kPa).

Received: January 28, 2011
Accepted: March 19, 2011
Published: March 30, 2011

Table 2. Vapor Pressure Equation Used ($\ln P/\text{kPa} = A + B/T + C \ln T + DT^E$) and Parameters[26]

compound	A	B	C	D	E
3-methyl-1-butanol	102.842	−10394	−12.083	6.2×10^{-18}	6
1-pentanol	162.052	−12659	−21.366	1.2×10^{-5}	2
ethyl lactate	71.8662	−6715.3	−9.5666	0.01499	1

Table 3. Vapor−Liquid Equilibrium Data of the 3-Methyl−1-Butanol + Ethyl Lactate System at 13.0 and 101.3 kPa

13.0 kPa					101.3 kPa				
x_1	y_1	T/K	γ_1	γ_2	x_1	y_1	T/K	γ_1	γ_2
0.000	0.000	366.55			0.000	0.000	427.65		
0.036	0.077	365.45	1.252	1.004	0.024	0.057	426.25	1.204	1.050
0.063	0.129	364.75	1.235	1.001	0.066	0.147	424.25	1.189	1.055
0.093	0.181	363.85	1.221	1.008	0.112	0.233	422.25	1.178	1.061
0.124	0.234	363.05	1.221	1.008	0.128	0.261	421.65	1.178	1.060
0.186	0.321	361.85	1.180	1.008	0.149	0.295	420.85	1.168	1.062
0.271	0.426	360.15	1.163	1.019	0.197	0.365	419.15	1.147	1.070
0.329	0.485	359.25	1.138	1.030	0.238	0.421	417.85	1.139	1.071
0.425	0.576	357.85	1.116	1.048	0.251	0.434	417.35	1.130	1.082
0.470	0.612	357.45	1.093	1.058	0.284	0.476	416.55	1.119	1.076
0.542	0.667	356.85	1.064	1.076	0.314	0.509	415.75	1.110	1.079
0.568	0.688	356.45	1.066	1.089	0.328	0.526	415.25	1.114	1.082
0.658	0.754	355.65	1.048	1.120	0.363	0.559	414.35	1.101	1.092
0.759	0.821	354.95	1.023	1.193	0.407	0.602	413.25	1.094	1.097
0.806	0.854	354.65	1.017	1.224	0.458	0.646	412.35	1.071	1.101
0.875	0.904	354.35	1.006	1.264	0.517	0.691	411.05	1.057	1.126
0.984	0.987	353.85	1.000	1.398	0.604	0.756	409.35	1.044	1.148
1.0000	1.0000	353.75			0.649	0.786	408.55	1.037	1.165
					0.707	0.822	407.75	1.021	1.193
					0.785	0.872	406.55	1.013	1.221
					0.862	0.918	405.45	1.007	1.267
					0.933	0.961	404.55	1.003	1.284
					0.966	0.980	404.05	1.004	1.349
					0.997	0.998	403.85	0.998	1.372
					1.0000	1.0000	403.75		

Table 4. Vapor−Liquid Equilibrium Data of the 1-Pentanol + Ethyl Lactate System at 13.0 and 101.3 kPa

13.0 kPa					101.3 kPa				
x_1	y_1	T/K	γ_1	γ_2	x_1	y_1	T/K	γ_1	γ_2
0.000	0.000	366.65			0.000	0.000	427.62		
0.013	0.028	366.25	1.492	1.000	0.032	0.066	426.35	1.278	1.000
0.041	0.084	365.45	1.442	1.001	0.038	0.077	426.15	1.273	1.001
0.055	0.107	365.25	1.419	1.002	0.043	0.087	426.05	1.268	1.001
0.096	0.170	364.35	1.357	1.006	0.054	0.106	425.55	1.260	1.001
0.111	0.190	364.05	1.335	1.008	0.067	0.130	425.15	1.249	1.002
0.128	0.215	363.65	1.313	1.010	0.085	0.159	424.45	1.235	1.003
0.139	0.223	363.35	1.300	1.012	0.099	0.181	424.15	1.225	1.003
0.162	0.253	363.15	1.273	1.015	0.105	0.191	423.95	1.221	1.004
0.214	0.312	362.45	1.221	1.025	0.118	0.211	423.75	1.212	1.005
0.282	0.382	361.75	1.165	1.041	0.138	0.241	422.95	1.198	1.007
0.371	0.464	360.85	1.111	1.065	0.151	0.258	422.65	1.190	1.008
0.444	0.526	360.45	1.077	1.088	0.168	0.281	422.15	1.179	1.009
0.552	0.614	359.85	1.043	1.124	0.197	0.320	421.65	1.162	1.013
0.653	0.696	359.35	1.023	1.158	0.210	0.334	421.25	1.155	1.014
0.789	0.813	358.95	1.007	1.204	0.240	0.369	420.55	1.139	1.018
0.912	0.919	358.65	1.001	1.244	0.283	0.416	419.85	1.119	1.025
0.957	0.960	358.45	1.000	1.258	0.322	0.457	419.15	1.102	1.032
1.000	1.000	358.35			0.375	0.508	418.05	1.083	1.042
					0.412	0.544	417.45	1.071	1.049
					0.473	0.596	416.75	1.054	1.063
					0.593	0.694	415.05	1.029	1.092
					0.663	0.751	414.25	1.019	1.111
					0.740	0.810	413.35	1.010	1.133
					0.816	0.865	412.55	1.005	1.155
					0.893	0.922	412.05	1.002	1.178
					0.895	0.924	411.95	1.002	1.179
					0.934	0.952	411.55	1.001	1.190
					0.972	0.979	411.35	1.000	1.202
					0.993	0.995	411.15	1.000	1.208
					1.000	1.000	411.05		

VLE experiment, the pressure was fixed and held constant by using a vacuum pump and the heating and stirring systems of the liquid mixture were turned on. Equilibrium conditions were assumed when constant temperature and pressure were obtained for 60 min or longer. Then, samples of liquid and condensate were taken. At least two analyses were made for each sample. The still was operated under a nitrogen atmosphere. The total pressure of the system was monitored with a digital manometer and controlled to the desired value (0.09 kPa) by means of a pressure controller (Normastat 75). Temperature was measured with a digital thermometer (Ertco-Hart, model 850) with a precision of ± 0.01 K and an uncertainty of ± 0.05 K. Both vapor-phase and liquid-phase compositions for the two systems were determined by densimetry and refractometry. Densities were measured at 298.15 K using an Anton Paar DMA 58 vibrating-tube densimeter with an uncertainty of ± 0.00005 g 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 an uncertainty of 0.01 K by means 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 ± 0.00005, with a temperature precision of ± 0.01 K. Prior to measurements, the 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 ± 0.00005 g. The estimated uncertainty in the determination of both liquid- and vapor-phase mole fractions is ± 0.001. This estimate is based on the agreement between multiple samples taken from the same phase at the same condition and on results obtained by analyzing standards of known composition using the same analytical techniques. The ethyl lactate breaks down at high temperatures into ethanol and lactic acid, so the still was filled in each experiment.

RESULTS AND DISCUSSION

Isobaric vapor−liquid equilibrium data for the two systems were obtained at (13.0 and 101.3) kPa and are shown in Tables 3 and 4. The liquid-phase activity coefficients of the components in the mixtures were calculated from[18]

$$\phi_i y_i P = \gamma_i x_i \phi_i^s P_i^s \exp[v_i^L (P - P_i^s)/RT] \quad (1)$$

where ϕ_i is the fugacity coefficient of component i in the vapor phase, y_i and x_i are the molar fractions in the vapor and liquid phases, respectively, γ_i is the activity coefficient of i with respect to the reference fugacity, ϕ_i^S is the fugacity coefficient of the pure saturated vapor of component i, P_i^S is the vapor pressure of component i at temperature T, and v_i^L is the molar volume of pure liquid. In the expression, it is assumed that the molar volume of component i is equal to the partial molar volume of component i at these conditions. The vapor pressures were calculated from the equation given in Table 2.

Table 5. Properties of the Pure Compounds Used in Calculating the Second Virial Coefficients and UNIQUAC Parameters[a]

	3-methyl-1-butanol	1-pentanol	ethyl lactate
T_c/K	579.4	597.1	588.00
P/kPa	3880	2760	3936.1
V_c/m^3/kmol	0.327	0.46	0.354
$R_D \times 10^{-10}$/m	3.684	3.679	3.622
DM $\times 10^{-30}$/C m	6.0042	6.0042	8.0055
MV/m^3/kmol	0.1092	0.1085	0.1150
R	4.1279	4.1286	3.2815
Q	3.588	3.592	2.7360
q'	1.15	1.15	2.7360
$\eta_{\text{association}}$	1.9	2.2	1.5
$\eta_{\text{solvation}}$	(1)-(2) 1.45	(2)-(3) 1.45	
ω	0.5558	0.5938	0.7926

[a] Critical temperature T_c, critical pressure P_c, critical volume V_c, mean gyration radius RD, dipole moment DM, molar volume MV (298 K), structure volume parameter for the UNIQUAC equation R, structure area parameter for the UNIQUAC equation Q, structure volume parameter for the modified UNIQUAC equation q', association or solvation parameter η, acentric factor w.

The fugacity coefficients were estimated by using the virial equation of state truncated after the second term. The second virial coefficients were obtained by using the method of Hayden and O'Connell[19] (HOC). The physical properties of the pure components required in this calculation are presented in Table 5. The association parameter for 3-methyl-1-butanol, 1-pentanol, and ethyl lactate were estimated by analogy from the components given in Fredenslund and others.[20] The η solvation parameter value for ethyl lactate-alcohol was assumed to be 1.45. This value was based on the assumption that solvation of ethyl lactate would be intermediate to that of alcohol—alcohol (1.55) and alcohol—ester (1.3). The association parameters were assumed to be 2.20 for 1-pentanol (similar to butan-1-ol) and 1.9 for 3-methyl-1-butanol (similar to 2-methyl-1-propanol) and 1.5 for ethyl lactate assuming that the self-associations of pure product are intermediate between pure alcohol and pure ester. Because of low pressures, the uncertainty in the estimation of the fugacity coefficients is negligible and the values of ϕ_i ($\phi_i y_i P = \gamma_i x_i P_i^\circ$) were next to 1 with ± 0.17% of deviation.

The 3-methyl-1-butanol—ethyl lactate system did not present an azeotrope both at 13 kPa and 101.3 kPa and exhibited positive deviation from ideality (Table 3). The infinite dilution activity coefficients (calculated from NRTL) were (1.26 and 1.41) at 13 kPa

Table 6. Mathematical Forms of the Activity Coefficient Equations

Wilson	$\ln \gamma_k = -\ln[\sum_{j=1}^{c} x_j \Lambda_{kj}] + 1 - \sum_{i=1}^{c} \frac{x_i \Lambda_{ik}}{\sum_{j=1}^{c} x_j \Lambda_{ij}}$	$\Lambda_{ji} = \frac{v_i}{v_j} \exp\left(-\frac{\lambda_{ij} - \lambda_{jj}}{RT}\right)$	$A = \lambda_{12} - \lambda_{11}$ $B = \lambda_{21} - \lambda_{22}$
NRTL	$\ln \gamma_i = \frac{\sum_{j=1}^{c} \tau_{ji} G_{ji} x_j}{\sum_{l=1}^{c} G_{li} x_l} + \sum_{j=1}^{c} \frac{x_j G_{ij}}{\sum_{l=1}^{c} G_{lj} x_l} \left(\tau_{ij} - \frac{\sum_{r=1}^{c} x_r \tau_{rj} G_{rj}}{\sum_{l=1}^{c} G_{lj} x_l}\right)$ $G_{ji} = \exp(-\alpha_{ji} \tau_{ji}),\ (\alpha_{ji} = \alpha_{ij})$	$\tau_{ji} = \frac{g_{ji} - g_{ii}}{RT}$	$A = g_{ji} - g_{ii}$ $B = g_{ji} - g_{ii}$
UNIQUAC	$\ln \gamma_i = \ln \frac{\Phi_i^*}{x_i} + \frac{z}{2} q_i \ln \frac{\theta_i}{\Phi_i^*} + l_i - \frac{\Phi_i^*}{x_i} \sum_{j=1}^{c} x_j l_j - q_i \ln\left(\sum_{j=1}^{c} \theta_j \tau_{ji}\right) + q_i l_i - q_i \sum_{j=1}^{c} \frac{\theta_j \tau_{ij}}{\sum_{k=1}^{c} \theta_k \tau_{kj}}$	$\tau_{ij} = \exp\left(-\frac{\Delta u_{ij}}{RT}\right)$	$A = \Delta u_{ij}$ $B = \Delta u_{ij}$

Table 7. Correlation Parameters for Activity Coefficients and Average Mean Deviation of Vapor Molar Fraction (Δy)

	Wilson			NRTL[a]			UNIQUAC		
P/kPa	A/J·mol^{-1}	B/J·mol^{-1}	(Δy)	A/J·mol^{-1}	B/J·mol^{-1}	(Δy)	A/J·mol^{-1}	B/J·mol^{-1}	(Δy)
3-Methyl-1-butanol + Ethyl Lactate									
13	3232.14	−2238.03	0.0010	1821.73	−771.55	0.0010	−1432.58	2466.73	0.0011
101.3	4842.99	−4064.19	0.0046	−1398.55	2585.84	0.0028	2330.33	−1357.20	0.0117
1-Pentanol + Ethyl Lactate									
13	5523.58	−4134.00	0.002	−1270.273	2681.39	0.002	−1983.95	3800.50	0.0033
101.3	5817.22	−4765.76	0.0025	−1398.55	2585.84	0.0028	2330.33	−1357.20	0.0117

[a] α = 0.3.

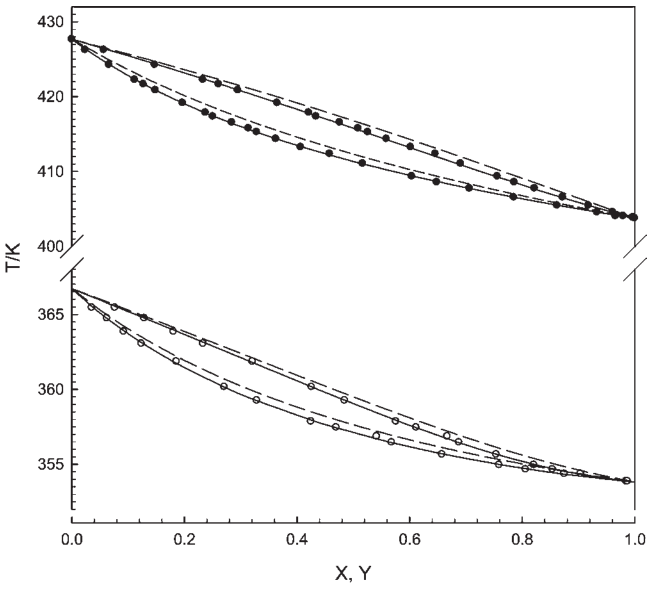

Figure 1. System 3-methyl-1-butanol + ethyl lactate. Symbols refer to experimental data at (○) 13.0 kPa; (●) 101.3 kPa. Lines: (—) smoothed using NRTL; (- - -) UNIFAC Dormund prediction.[29]

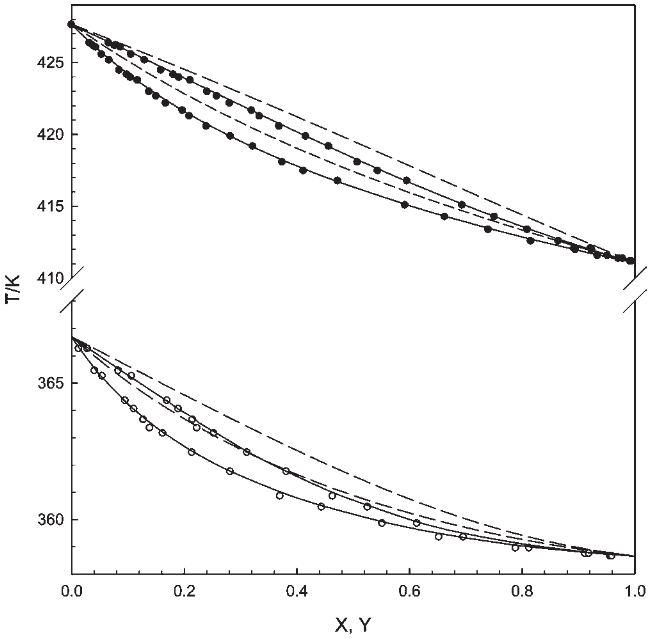

Figure 2. System 1-pentanol + ethyl lactate at 101.3 kPa. Symbols refer to experimental data at (○) 13.0 kPa; (●) 101.3 kPa. Lines: (—) smoothed using NRTL; (- - -) UNIFAC Dormund prediction.[29]

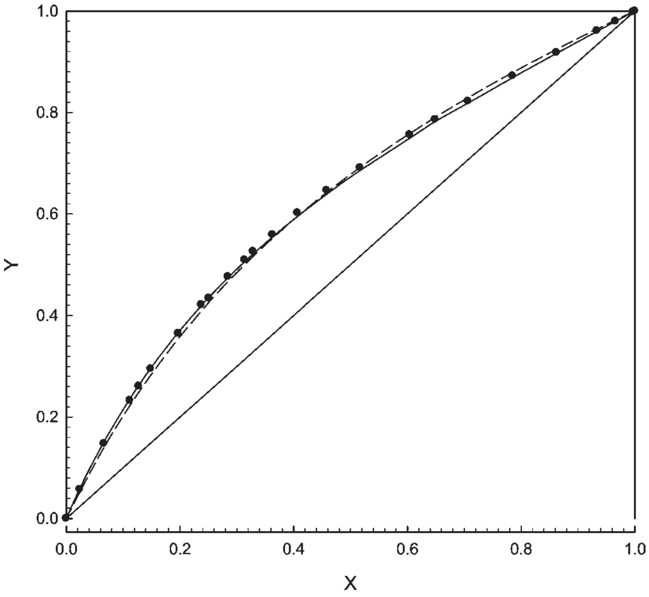

Figure 3. System 3-methyl-1-butanol + ethyl lactate at 101.30 kPa. (●) experimental data. Lines: (—) smoothed using NRTL model; (— — —) predicted by UNIFAC Dormund method.

Table 8. Results of the Thermodynamic of Modified DECHEMA Consistency Test

		Margules constants		
	average deviation δ	A	B	C
	3-Methyl-1-butanol + Ethyl Lactate			
13 kPa	0.0012	0.2644	0.3491	0.0877
101.3 kPa	0.0056	0.2679	0.2397	0.1697
	1-Pentanol + Ethyl Lactate			
13 kPa	0.0020	0.4660	0.3447	0.2438
101.3 kPa	0.0027	0.2949	0.2030	0.0645

and (1.26 and 1.35) at 101.3 kPa for $x_1 = 0$ and $x_1 = 1$, respectively. The 1-pentanol−ethyl lactate system did not present an azeotrope either and presented positive deviations of ideal behavior with the following values of the infinite dilution activity coefficients (calculated from NRTL) (1.57 and 1.28) at 13 kPa and (1.34 and 1.20) at 101.3 kPa for $x_1 = 0$ and $x_1 = 1$, respectively. So, both mixtures can be considered almost ideal because the values of the activity coefficients are close to unity. That indicated that the alcohol−lactate interactions were similar to those of the pure components, probably due to the OH group that can form hydrogen bonds with a like or an unlike molecule. Similar results were found by other authors with alcohol−lactate systems.[14,15,21]

The Wilson, NTRL, and UNIQUAC models were used for the correlation of the activity coefficients. The exact mathematical forms of the equations are shown in Table 6. The values of r_k and q_k were taken from the Distil[22] program in the UNIQUAC model.

The estimation of the parameters was done with the maximum likelihood technique, where the pressure, temperature, and liquid and vapor concentrations are considered simultaneously. The computer subroutines given by Gess et al.[23] were used. The fitted parameters along with the average mean deviations in the vapor-phase mole fraction, (Δy) are listed in Table 7. All the activity coefficient models listed in Table 6 provide a similar correlation of experimental data. Figures 1−3 shows the $T-x-y$ representations of experimental data and the results with the NRTL correlation. The best description of the equilibrium of the system 3-methyl-1-butanol + ethyl lactate was given by the NRTL equation, with an average mean deviation of vapor molar fraction (Δy) of 0.001 at 13 kPa and 0.003 at 101.3 kPa. The results of correlations for the system 1-pentanol + ethyl lactate by use of the Wilson and NTRL models present better correlations (Δy = 0.004) than the UNIQUAC model. Considering the large differences in boiling points of the binary systems, the deviations are within an acceptable range.

The two systems are consistent according to the criteria Herrington because of $|D - J| \leq 10\%$. The values of D and J for 3-methyl-1-butanol + ethyl lactate were found to be 7.64% and 5.43% at 13 kPa and 13.05 and 8.88 at 101.3 kPa, respectively, and for the 1-pentanol + ethyl lactate the values are 0.47% and 3.35% at 13 kPa and 0.54% and 5.54% at 101.3 kPa, respectively. Also the consistency was checked by means of the modification of the DECHEMA test proposed by Gess et al.[23] for the prediction of the mole fraction in the vapor phase. In the method, the activity coefficients were calculated with the four-suffix Margules equation

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

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

The pressure was calculated with the following equation,

$$P* = \frac{x_1\gamma_1{}^* f_1^0}{\phi_1} + \frac{x_2\gamma_2{}^* f_2^0}{\phi_2} \tag{4}$$

Here the asterisk (∗) denoted a calculated or predicted value. The parameters of the Margules equation were calculated by minimization of $(P - P^*)$. The values of y_1 were estimated by

$$y_1 = \frac{x_1\gamma_1 f_{1_1}^0}{P*} \tag{5}$$

To pass the consistency test, a system must have an average deviation $\delta = \Sigma|y - y^*|/n$ lower than 0.01. The two systems have passed the test. The values of the constants A, B, D, and δ are shown in Table 8.

Our data are coincident with the predicted by the UNIFAC Dortmund method in the $x-y$ diagram (Figure 3) and are slightly different in the $T-x-y$ diagram for both systems (Figures 1 and 2). The same behavior was found in the systems methanol−ethyl lactate and propan-1-ol−ethyl lactate.[13]

■ CONCLUSIONS

Sets of experimental VLE data were obtained for the binary 3-methyl-1-butanol + ethyl lactate and 1-pentanol + ethyl lactate systems at (13.0 and 101.3) kPa. No azeotropes were observed. The experimental data were correlated satisfactorily with the Wilson, NRTL, and UNIQUAC models. The consistency of experimental data was proved on the basis of the Herrington as well as the point to point direct test of thermodynamic consistency. According to the tests, the quality of measured data was classified from good to excellent.

■ AUTHOR INFORMATION

Corresponding Author

*Fax: 34 945 013014. E-mail: emilio.cepeda@ehu.es.

■ REFERENCES

(1) Goenaga, J. M.; Gonzalez-Olmos, R.; Iglesias, M.; Resa, J. M. Measurement and modelling of phase equilibria for ethanol+water+1-pentanol at isobaric condition. *Korean J. Chem. Eng.* **2006**, *23*, 631–637.

(2) Goenaga, J. M.; Gayol, A.; Concha, R. G.; Iglesias, M.; Resa, J. M. Effect of temperature on thermophysical properties of ethanol plus aliphatic alcohols (C-4-C-5) mixtures. *Monatsh. Chem.* **2007**, *138*, 402–435.

(3) Resa, J. M.; Gonzalez, C.; Goenaga, J. M.; Iglesias, M. Influence of temperature on ultrasonic velocity measurements of ethanol plus water plus ethyl acetate mixtures. *Phys. Chem. Liq.* **2005**, *43*, 65–89.

(4) Resa, J. M.; Goenaga, J. M.; Gonzalez-Olmos, R.; Iglesias, M. Measurement and modeling of phase equilibria for ethanol plus water plus methanol at isobaric condition. *J. Chem. Eng. Data* **2006**, *51*, 2114–2120.

(5) Resa, J. M.; Goenaga, J. M.; Iglesias, M. 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. *J. Chem. Eng. Data* **2006**, *51*, 1892–1895.

(6) Resa, J. M.; Goenaga, J. M.; Sanchez-Ruiz, A. I.; Iglesias, M. Density, refractive index, speed of sound at 298.15 K, and vapor-liquid equilibria at 101.3 kPa for binary mixtures of ethyl acetate plus ethyl lactate and methyl acetate plus ethyl lactate. *J. Chem. Eng. Data* **2006**, *51*, 1294–1299.

(7) Resa, J. M.; Goenaga, J. M.; Iglesias, M.; Gonzalez-Olmos, R.; Pozuelo, D. Liquid-liquid equilibrium diagrams of ethanol plus water plus (ethyl acetate or 1-pentanol) at several temperatures. *J. Chem. Eng. Data* **2006**, *51*, 1300–1305.

(8) Resa, J. M.; Goenaga, J. M.; Lanz, J.; Iglesias, M. Vapor-liquid equilibrium of binary mixtures containing ethyl acetate plus 2-methyl-1-propanol and ethyl acetate plus 2-methyl-1-butanol at 101.3 kPa. *J. Chem. Eng. Data* **2006**, *51*, 595–598.

(9) Resa, J. M.; Gonzalez, C.; Goenaga, J. M. Density, refractive index, speed of sound at 298.15 K, and vapor-liquid equilibria at 101.3 kPa for binary mixtures of propanol+2-methyl-1-butanol and propanol+3-methyl-1-butanol. *J. Chem. Eng. Data* **2006**, *51*, 73–78.

(10) Resa, J. M.; Goenaga, J. M.; Iglesias, M. Temperature dependence of excess molar volumes of ethanol+water+1-propanol. *J. Therm. Anal. Calorim.* **2007**, *88*, 549–555.

(11) Resa, P.; Elvira, L.; de Espinosa, F. M.; Gomez-Ullate, Y. Ultrasonic velocity in water-ethanol-sucrose mixtures during alcoholic fernientation. *Ultrasonics* **2005**, *43*, 247–252.

(12) Peynaud, E.; Blouin, J. *Enología Práctica. Conocimiento Y Elaboración Del Vino*; Mundi-Prensa: Madrid, Spain, 2003.

(13) Resa, J. M.; Cepeda, E. A.; Goenaga, J. M.; Ramos, A.; Aguirre, S.; Urbano, C. Density, Refractive Index, Speed of Sound at 298.15 K, and Vapor-Liquid Equilibrium at 101.3 kPa for Binary Mixtures of Methanol plus Ethyl Lactate and 1-Propanol + Ethyl Lactate. *J. Chem. Eng. Data* **2010**, *55*, 1017–1021.

(14) Sanz, M. T.; Calvo, B.; Beltran, S.; Cabezas, J. L. Vapor-liquid equilibria at (33.33, 66.66, and 101.33) kPa and densities at 298.15 K for the system methanol plus methyl lactate. *J. Chem. Eng. Data* **2002**, *47*, 1003–1006.

(15) Vu, D. T.; Lira, C. T.; Asthana, N. S.; Kolah, A. K.; Miller, D. J. Vapor-liquid equilibria in the systems ethyl lactate plus ethanol and ethyl lactate plus water. *J. Chem. Eng. Data* **2006**, *51*, 1220–1225.

(16) Verginer, M.; Leitner, E.; Berg, G. Production of Volatile Metabolites by Grape-Associated Microorganisms. *J. Agric. Food Chem.* **2010**, *58*, 8344–8350.

(17) Boulton, R. B.; Singleton, V. L.; Bisson, L. F.; Kunkee, R. E. *Principle and Practices of Winemaking*; Chapman & Hall: New York, 1996.

(18) Walas, S. M. *Phase Equilibria in Chemical Engineering*; Butterworth: Boston, MA, 1985.

(19) Hayden, J. G.; Oconnell, J. P. Generalized method for predicting second virial-coefficients. *Ind. Eng. Chem. Proc. Des. Dev.* **1975**, *14*, 209–216.

(20) Fredenslund, A.; Gmehling, J.; Rasmussen, P. *Vapor-Liquid Equilibria Using UNIFAC: A Group-Contribution Method*; Elsevier: Amsterdam, The Netherlands, 1977.

(21) Pena-Tejedor, S.; Murga, R.; Sanz, M. T.; Beltran, S. Vapor-liquid equilibria and excess volumes of the binary systems ethanol plus ethyl lactate, isopropanol plus isopropyl lactate and n-butanol plus n-butyl lactate at 101.325 kPa. *Fluid Phase Equilib.* **2005**, *230*, 197–203.

(22) *Distil 5.0*; Hyprotech Ltd.: Calgary Alberta, 2001.

(23) Gess, M. A.; Danner, R. P.; Nagvekar, M. *Thermodynamic Analysis of Vapor-Liquid Equilibria: Recommended Models and a Standard Data Base*. AIChE: New York, 1991.

(24) Riddick, J. A.; Bunger, W. B.; Sakano, T. K. *Organic Solvents: Physical Properties and Methods of Purification*, 4th ed.; Wiley: New York, 1986.

(25) Krokhin, N. G. Equilibrium liquid-vapor ratios in the system acetic acid-isoamyl alcohol-isoamyl acetate. II. Binary systems acetic acid-isoamyl acetate and isoamyl alcohol-isoamyl acetate. *Zh. Fiz. Khim.* **1967**, *41*, 1509–1511.

(26) Daubert, T. E.; Danner, R. P. *Physical and Thermodynamic Properties of Pure Chemicals Data Compilation*; Hemisphere Pub. Corp: New York, 1984.

(27) Reddy, M. S.; Rao, C. V. Isothermal and Isobaric Vapor-Liquid Equilibrium Data System - Toulene-Isoamyl Alcohol. *J. Chem. Eng. Data* **1965**, *10*, 309.

(28) Diaz, M. A. E.; Guetachew, T.; Landy, P.; Jose, J.; Voilley, A. Experimental and estimated saturated vapour pressures of aroma compounds. *Fluid Phase Equilib.* **1999**, *157*, 257–270.

(29) *ChemCAD 5.2.0 Professional*; Chemstations Inc.: Houston, TX, 2002.