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Density, Refractive Index, Speed of Sound at 298.15 K, and Vapor−Liquid Equilibrium at 101.3 kPa for Binary Mixtures of Methanol + Ethyl Lactate and 1-Propanol + Ethyl Lactate

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Density, refractive index and speed of sound at 298.15 K, together with isobaric vapor−liquid equilibria data (VLE) at 101.3 kPa are reported for two binary mixtures, methanol + ethyl lactate and 1-propanol + ethyl lactate. Excess molar volumes on mixing were determined from the experimental data and consequently fitted with Redlich−Kister polynomials. VLE experimental data were tested by Dechema test, which proved their thermodynamic consistency. The activity coefficients were correlated with the Margules, van Laar, UNIQUAC, NRTL, and Wilson equations. The Analytical Solutions of Groups and Universal quasichemical Functional group Activity Coefficients models were also used for the prediction. Investigated binary systems did not show any azeotrope.

Introduction

This work is part of a research project (Resa et al.[1−9]) whose objective is to determine the thermodynamic properties for the binary systems involved in wine distillation processes. Experimental data are used for the simulation of these processes. In the wine distillation process, the presence of multicomponent mixtures based mainly on water and ethanol is typical. Remaining minor components, called congeners, are principally alcohols, aldehydes, and acetates. For modeling and process simulation in which these mixtures appear, binary data are needed. Thus, it is important to have available vapor−liquid equilibrium 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 can be calculated, and results can be applied to study the distillation of wine.

The main objective of this work was to determine density, refractive index, and the speed of sound at 298.15 K and to measure vapor−liquid equilibria data (VLE) at 101.3 kPa for the following systems: methanol + ethyl lactate and 1-propanol + ethyl lactate. No experimental data was found for these systems in the literature, although similar alcohol-lactate systems have been measured.[10−12] The aims of the introduced study were (a) the calculation of excess molar volumes on mixing and the correlation of the results, (b) testing the consistency of the equilibrium data, and (c) to fit the experimental data to classic models.

Experimental Section

Materials. Ethyl lactate (>99.0%), supplied by Fluka (Sigma-Aldrich Chemie GmbH), and methanol (> 99.9 %) and 1-propanol (99.5 %), supplied from Panreac Química S.A.B, were purified by distillation in a laboratory column of 100 plates. The purity of the materials was checked by gas−liquid chromatography that was higher than 99.6 %. All products were degassed before the experiment using a ultrasound technique and dried on molecular sieves with 3 Å pore diameter from Fluka manufacturer. Densities, refractive indexes, and normal boiling points of the pure substances are given in Table 1 and compared with the literature values.[4,13]

Apparatus and Procedure. The still used to measure VLE data was a dynamic recirculating apparatus designed by Resa et al.[4] The equilibrium temperature was measured with a Pt100 digital thermometer with a ± 0.01 K precision and a ± 0.05 K uncertainty. For the pressure measurement, a digital manometer regulator (Divatronic DT1 model), manufactured by Leybold with an estimated uncertainty of ± 0.03 kPa was used. Densities were measured at 298.15 K by using an Anton Paar DMA 58 vibrating tube densimeter with an uncertainty of ± 0.00005 $\text{g}\cdot\text{cm}^{-3}$, which had been calibrated at atmospheric pressure with twice distilled water and dry air. The temperature of the densimeter measured by the Pt resistive thermometer was maintained at a constant value of 298.15 K by a semiconductor Peltier element with a repeatability of ± 0.01 K. Refractive indices were measured with a Mettler RE50 refractometer with an uncertainty of ± 0.00005, and temperature was controlled with a ± 0.01 K precision. Speeds of sound were measured with an Anton Paar DSA 48 sound analyzer with an uncertainty of ± 0.1 $\text{m}\cdot\text{s}^{-1}$, and temperature was controlled by a Peltier cooler with an accuracy of ± 0.1 K. Prior to measurements, density-calibration curves were obtained to calculate the compositions of the vapor and liquid phases. The binary mixtures were prepared by directly weighing the constituent components using an electronic balance (Salter model ER-182A) having a given uncertainty of ± 0.0001 g. Precautions were taken in order to minimize evaporation losses during storage and preparation of the solutions. The estimated uncertainty of both liquid and vapor phase mole fractions in the vapor−liquid equilibrium experiments was found to be ± 0.001.

The ethyl lactate breaks down slowly at the experimental temperatures used in this work. When the analysis by GC was made, the decomposition of ethyl lactate was not detected in one run. So, a fresh sample was used for each run.

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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[b]	obs	lit[a]
methanol	786.8	786.37	1.32648	1.32652	1101	1102	337.80	337.69
1-propanol	800.3	799.60	1.3831	1.3837	1206	1206	370.4	370.93
ethyl lactate	1028.6	1027.2	1.41057	1.41050	1277	1277	427.6	427.70

[a] Riddick et al.[13] [b] Resa et al.[4]

Table 2. Densities, Refractive Indices, and Speed of Sounds for Methanol (1) + Ethyl Lactate (2) and Propanol (1) + Ethyl Lactate (2) at 298.15 K with Excess Molar Volume (V^E) Varying with Composition (x_i)

x_1	ρ/(g·cm^{-3})	V^E/(cm^3·mol^{-1})	n_D	u/m·s^{-1}
		methanol (1) + ethyl lactate (2)		
0.053	1.0245	−0.112	1.40940	1276
0.105	1.0199	−0.144	1.40821	1275
0.149	1.0165	−0.284	1.40729	1273
0.203	1.0116	−0.317	1.40539	1271
0.249	1.0067	−0.405	1.40364	1268
0.296	1.0017	−0.473	1.40214	1266
0.353	0.9946	−0.512	1.39965	1262
0.400	0.9889	−0.604	1.39771	1258
0.452	0.9807	−0.609	1.39589	1253
0.502	0.9726	−0.644	1.39367	1247
0.549	0.9635	−0.683	1.38953	1241
0.598	0.9528	−0.627	1.38628	1233
0.649	0.9407	−0.618	1.38192	1223
0.700	0.9266	−0.586	1.37770	1213
0.747	0.9126	−0.554	1.37193	1200
0.801	0.8925	−0.466	1.36488	1185
0.850	0.8737	−0.411	1.35905	1169
0.899	0.8496	−0.288	1.34922	1150
0.950	0.8198	−0.143	1.339	1126
		1-propanol (1) + ethyl lactate (2)		
0.058	1.0246	−0.023	1.40872	1272
0.100	1.0210	−0.029	1.40699	1266
0.150	1.0173	−0.038	1.40595	1261
0.200	1.0136	−0.061	1.40379	1255
0.250	1.0098	−0.071	1.40230	1250
0.300	1.0055	−0.068	1.39968	1244
0.350	1.0011	−0.069	1.39787	1238
0.400	0.9966	−0.069	1.39628	1232
0.450	0.9922	−0.080	1.39379	1226
0.500	0.9874	−0.082	1.39125	1220
0.550	0.9823	−0.074	1.38790	1213
0.600	0.9773	−0.081	1.38498	1207
0.650	0.9720	−0.087	1.38197	1200
0.700	0.9663	−0.078	1.37916	1193
0.750	0.9605	−0.072	1.37601	1186
0.800	0.9543	−0.060	1.37278	1180
0.850	0.9479	−0.040	1.36960	1172
0.900	0.9413	−0.035	1.36642	1164
0.950	0.9343	−0.018	1.36248	1157

Table 3. Adjustable Parameters (a_k) with the Standard Deviations (σ) for Excess Molar Volumes (V^E)

	methanol(1) + ethyl lactate (2) V^E/cm^3·mol^{-1}
a_0	−0.315
a_1	−0.064
a_2	0.041
a_3	0.109
a_4	−0.335
σ	0.007
	1-propanol (1) + ethyl lactate (2) V^E/cm^3·mol^{-1}
a_0	−0.324
a_1	−0.149
a_2	−0.038
a_3	0.439
a_4	−0.300
σ	0.013

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 sound deviation δu for the binary mixtures of methanol + ethyl lactate and 1-propanol + ethyl lactate.

The excess molar volumes of binary mixtures were calculated from density measurements by applying the following 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 the densities of the pure substances, M_1 and M_2 are the molar masses, and x_1, x_2 are the mole fractions. The uncertainty in the calculation of V^E from density measurements was estimated to be ± 0.0005 cm^3·mol^{-1}.

Excess molar volumes were fitted to Redlich−Kister polynomials of the following form,

$$V^E = x_1x_2\sum_{k\geq 0} a_k(x_1 - x_2)^k \qquad (2)$$

where a_k are the adjustable parameters obtained by a least-squares correlation method, and k is the polynomial degree. Table 3 lists the parameters for the Redlich−Kister eq 4 with their standard deviations σ. The standard deviations σ are defined as follows

$$\sigma = \sqrt{\frac{\sum (V^E_{\text{cal}} - V^E_{\text{exp}})_i^2}{N - m}} \qquad (3)$$

where N is the number of experimental data, and m is the number of equation parameters.

As seen from Table 3, the experimentally derived excess molar volumes were correlated satisfactorily by the Redlich−Kister equation. Table 2. shows that the excess molar volumes V^E are asymmetric and negative for the systems studied over the entire composition range and that a minimum in V^E is reached with mole fraction of alcohol $x_1 = 0.6$. The negative excess molar volumes indicated that a more efficient packing and/or attractive interaction occurred when the alcohol and lactate were mixed, due to intermolecular associations via H-bonding, resulting into complex formation producing displacement of electron and nuclei.

VLE Data. Vapor−liquid equilibrium data (T, x_1, y_1) for methanol (1) + methyl lactate (2) and 1-propanol (1) + ethyl lactate (2) binary systems at 101.3 kPa are presented in Table 4 and Table 5. The T, x_1, y_1 phase diagrams are shown in Figure 1 and 2. No azeotropes were found in any of these systems

The activity coefficients γ_i of the components were calculated from

$$\gamma_i = \frac{y_i \Phi_i P}{x_i P_i^0} \tag{4}$$

where x_i and y_i are the liquid and vapor mole fractions in equilibrium, respectively, Φ_i is a vapor phase correction factor, P is the total pressure, and P_i^o is the vapor pressure of pure component i. These vapor pressures were calculated from the equation

$$\log(P_i^o/\mathrm{kPa}) = A_i - \frac{B_i}{T/\mathrm{K} + C_\mathrm{i}} + C \ln T/\mathrm{K} + D(T/\mathrm{K})^\mathrm{E} \tag{5}$$

The constants are reported in Table 6.

The vapor phase correction factor is given by the following equation,

$$\Phi_i = \frac{\varphi_i}{\varphi_i^{\mathrm{sat}}} \exp\left[-\frac{V_i(P - P_i^o)}{RT}\right] \tag{6}$$

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 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 Hayden and O'Connell[14] method. The physical properties of the pure components required in this calculation are presented in Table 7. The value of the solvation parameter for the binary mixtures studied was regarded as 1.3, typical in the alcohol-ester mixtures.

Table 4. Experimental VLE Data for the Binary Methanol (1) + Ethyl Lactate (2) System at 101.33 kPa

x_1	y_1	T/K	γ_1	γ_2
0.000	0.000	427.60		
0.015	0.194	420.65	1.19	1.00
0.045	0.448	408.95	1.19	1.01
0.057	0.516	405.06	1.18	1.02
0.086	0.639	397.35	1.17	1.01
0.103	0.682	393.60	1.15	1.02
0.112	0.711	391.42	1.17	1.01
0.122	0.723	389.35	1.15	1.05
0.143	0.765	385.72	1.14	1.04
0.181	0.809	380.23	1.11	1.08
0.203	0.843	377.15	1.13	1.02
0.231	0.859	374.07	1.11	1.06
0.248	0.874	372.51	1.10	1.03
0.360	0.926	362.65	1.08	1.04
0.419	0.941	359.24	1.06	1.05
0.478	0.951	355.81	1.05	1.12
0.513	0.959	354.25	1.04	1.07
0.599	0.969	350.65	1.01	1.14
0.822	0.990	342.55	1.01	1.19
0.921	0.996	339.75	1.00	1.15
0.960	0.998	338.70	1.00	1.13
1.000	1.000	337.80		

Table 5. Experimental VLE Data for the Binary 1-Propanol (1) + Ethyl Lactate (2) System at 101.33 kPa

x_1	y_1	T/K	γ_1	γ_2
0.000	0.000	427.60		
0.008	0.070	422.25	1.53	1.00
0.022	0.162	419.85	1.50	1.00
0.033	0.224	417.45	1.48	1.00
0.045	0.284	412.05	1.47	1.00
0.079	0.419	401.95	1.41	1.00
0.169	0.622	399.35	1.30	1.02
0.206	0.665	397.15	1.26	1.02
0.239	0.700	394.75	1.23	1.03
0.275	0.732	392.65	1.20	1.04
0.310	0.759	390.25	1.17	1.05
0.340	0.789	387.45	1.15	1.06
0.408	0.820	385.85	1.11	1.08
0.438	0.843	382.85	1.10	1.09
0.522	0.878	379.65	1.06	1.12
0.625	0.908	377.65	1.04	1.16
0.701	0.932	376.35	1.02	1.20
0.750	0.942	374.15	1.01	1.22
0.836	0.970	371.75	1.01	1.26
0.935	0.988	370.65	1.00	1.30
1.000	1.000	370.4		

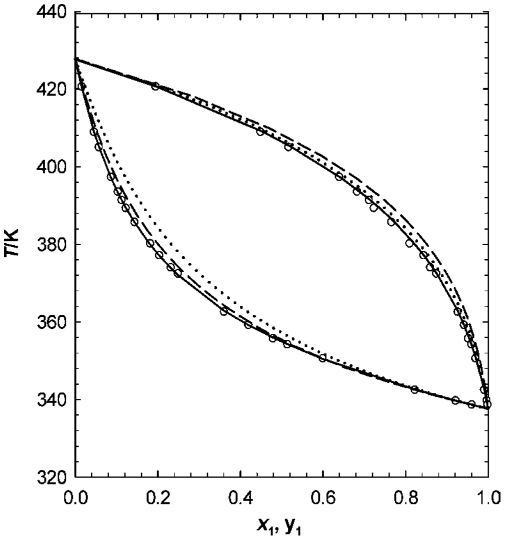

Figure 1. T-x_1-y_1 diagram for methanol (1) + ethyl lactate (2) at 101.3 kPa. ○, Experimental data; —, Wilson correlation; - -, ASOG prediction; ••••, UNIFAC Dortmund prediction.

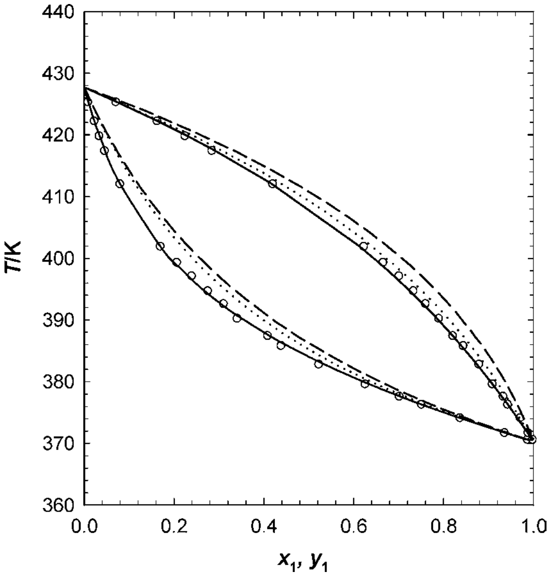

Figure 2. T-x_1-y_1 diagram for 1-propanol (1) + ethyl lactate (2) at 101.3 kPa. ○, Experimental data; —, Wilson correlation; - -, ASOG prediction; ••••, UNIFAC Dortmund prediction.

Table 6. Vapor Pressure Parameters Equation 5[a]

compound	A_i	B_i	C_i	D	E
methanol	59.8373	−6282.89	−6.37870	4.61746×10^{6}	2.00000
1-propanol	71.8662	−6715.29	−9.56659	1.49929×10^{-02}	1.00000
ethyl lactate	79.4625	−8294.91	−8.90961	1.81967×10^{-06}	2.00000

[a] Distil Library.[25]

The activity coefficients were correlated with the Margules,[15] van Laar,[16] Wilson,[17] NRTL,[18] and UNIQUAC[19] equations. The

Table 7. Properties of the Pure Compounds Used in Calculating the Second Virial Coefficients and UNIQUAC Parameters: Critical Presure P_c, Mean Gyration Radius RD, Dipole Moment μ, Association Parameter ETA, Critical Temperature T_c, Critical Compressibility Factor Z_c, Acentric Factor w, UNIQUAC Parameters r, q

	P_c	RD × 10^{-10}	$\mu \times 10^{-30}$		V	T_c			UNIQUAC	
compound	kPa	m	C·m	ETA	m^3/kmol	K	Zc	w	r	q
methanol	8256.6	1.552	5.6706	1.63	0.0407	512.64	0.224	0.5640	1.4311	1.4320
1-propanol	5170.0	2.825	5.6039	1.32	0.0751	536.75	0.254	0.6279	2.7799	2.5120
ethyl lactate	3936.1	3.622	8.0055	0.80	0.0853	588.00	0.280	0.7926	4.4555	3.9280

Table 8. Correlation Parameters for the Activity Coefficients A_{12} and A_{21} and Average Deviations in Temperature (ΔT) and Composition (Δy_1)

equation	A_{12}	A_{21}	ΔT/K	Δy_1
methanol (1) + ethyl lactate (2)				
Margules[a]	0.2041	0.16322	0.11	0.004
van Laar[a]	0.2065	0.16389	0.11	0.004
Wilson[b]	406.02	−323.93	0.11	0.003
NRTL[c] ($\alpha_{12} = 0.304$)	−445.23	812.11	0.11	0.003
UNIQUAC[d]	−141.93	268.67	0.12	0.004
1-propanol (1) + ethyl lactate (2)				
Margules[a]	0.3060	0.4647	1.19	0.016
van Laar[a]	0.3316	0.3972	0.4	0.008
Wilson[b]	655.09	−501.99	0.45	0.010
NRTL[c] ($\alpha_{12} = 0.302$)	142.83	7.48	0.42	0.009
UNIQUAC[d]	−197.51	416.72	0.58	0.011

[a] Margules and van Laar constants (dimensionless). [b] Wilson's interaction parameters (K). [c] NRTL's interaction parameters (K). [d] UNIQUAC's interaction parameters (K).

constants of each model were determined in sense of "VLE calc" method suggested by Gess et al.[20] The parameters estimation of the equations was solved iteratively using the maximum likelihood regression of the objective function Q_i,[21] with the activity coefficients obtained from the consistency test as experimental values

$$Q_i = \sum \left(\frac{\gamma_{\text{exp}} - \gamma_{\text{cal}}}{\gamma_{\text{exp}}} \right)^2 \qquad (7)$$

where γ_{exp} are the activity coefficients calculated from experimental data and γ_{cal} are the coefficients calculated with the correlations. The parameters of the activity coefficient equation, the average deviation in temperature (ΔT), and the average deviation in composition (Δy) are listed in Table 8. Comparison between experimental data and Wilson prediction is shown in Figures 1 and 2. The ASOG[22] and UNIFAC Dormund method[23] were also used to predict the VLE of both systems. Comparison of the results of both predictions with the experimental data is shown in Figures 1 to 2. The predictions of both methods are good for the methanol-ethyl lactate system in the x-y diagram but the temperatures predicted with those methods are different to the experimental ones in the T-x-y diagram (Figure 1). In the system propanol-ethyl lactate, the deviations are higher in the ASOG method.

The thermodynamic consistency of the experimental data was checked by means of a modified Dechema test[24] where the fugacity coefficients were calculated by the method of Hayden and O'Connell,[14] and activity coefficients were calculated by using the four-suffix Margules equation,

$$\frac{G^{\text{E}}}{RT} = x_1 x_2 (A x_2 + B x_1 - D x_1 x_2) \qquad (8)$$

with the corresponding activity coefficients

$$\begin{aligned} \ln \gamma_1 &= x_2^2 (A + 2(B - A - D)x_1 + 3Dx_1^2) \\ \ln \gamma_2 &= x_1^2 (A + 2(B - A - D)x_2 + 3Dx_2^2) \end{aligned} \qquad (9)$$

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

$$F = P - \left(\frac{x_1 \gamma_1^* f_1^{\text{o}}}{\varphi_1} + \frac{x_2 \gamma_2^* f_2^{\text{o}}}{\varphi_2} \right) \qquad (10)$$

where P is the total pressure. Here the asterisk (*) denotes a calculated or predicted value. Experimental values are represented with no asterisk; f_1^{o} and f_2^{o} are the standard state fugacities. The errors in the prediction of y_1 were calculated. Predicted y_1^* values were obtained using the equation

$$y_1^* = \frac{x_1 \gamma_1^* f_1^{\text{o}}}{\varphi_1 P^*} \qquad (11)$$

The average deviation (δy) was calculated from,

$$\delta y = \frac{\sum_{i=1}^{n} |\Delta y|}{N} \qquad (12)$$

Here $\Delta y = y_1 - y_1^*$ and N = number of experimental data points. To pass the consistency test, a system must generally achieve an average deviation below 0.01. Both of the mixtures investigated in this work have passed this test as can be seen from Table 9.

We also carried out a Margules constant test using the Gess et al.[20] program. The Margules constant can be used to indicate the ideality of a system. The systems with an absolute value of the Margules constant less than 0.60 should be considered ideal. However this classification criterion is not rigorous. Table 10 shows the values of the Margules constant evaluated for the investigated binary mixtures. Both mixtures can be considered ideal under this criterion. That indicates that the alcohol-lactate interactions are similar to those of the pure components, probably due to 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.[10−12]

Table 9. Results of the Thermodynamic Consistency Test

		Margules constants		
system	average deviation Δy_1	A	B	D
methanol (1) + ethyl lactate (2)	0.0025	0.1811	0.1429	−0.0712
1-propanol (1) + ethyl lactate (2)	0.0065	0.4359	0.4453	0.2792

Table 10. Results of the Margules Constant Test

system	Margules constant
methanol(1)+ ethyl lactate (2)	0.2939
1-propanol (1) + ethyl lactate (2)	0.3624

Conclusions

New vapor−liquid equilibria data not previously reported in literature have been measured for the systems methanol + ethyl lactate and 1-propanol + ethyl lactate. Both systems present ideal behavior. All considered correlations provided comparable results for the methanol + ethyl lactate mixture. The VLE data for 1-propanol + ethyl lactate was fitted less accurately. The best prediction was obtained from the NRTL correlation.

The trend of the excess molar volumes is quite similar for both systems, since their behavior shows slightly contractive (negative) character.

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