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Vapor–Liquid Equilibria for the Ternary System Ethanol + Water + 1-Ethyl-3-methylimidazolium Ethylsulfate and the Corresponding Binary Systems Containing the Ionic Liquid at 101.3 kPa

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In this work, experimental data of isobaric vapor–liquid equilibria (VLE) for the ternary system ethanol + water + 1-ethyl-3-methylimidazolium ethylsulfate (EMISE) and for the corresponding binary systems containing the ionic liquid (ethanol + EMISE, water + EMISE) were carried out at 101.3 kPa. VLE data of binary systems were correlated using the e-NRTL and NRTL equations. With the obtained parameters, we can predict the behavior of the ternary system. In addition, the ternary system was correlated using the NRTL equation. A study of the evolution of the ethanol + water azeotrope composition with different amounts of ionic liquid was made.

Introduction

Ionic liquids (ILs) are substances formed by ions, and they show a negligible vapor pressure at normal temperature and pressure conditions. This and other properties, such as their thermal stability and their capability as solvents, make the IL a new alternative for processes.

The prediction of the most suitable IL for each separation process is limited since there is not enough information about the influence of the structure of the IL on its physical and solvent properties. Besides, experimental phase equilibrium data are required for developing thermodynamic (G^E) models and for understanding their thermodynamic behavior. Vapor–liquid equilibria (VLE) data permit checking the potential of G^E models which are used for the description of the nonideal behavior of systems containing IL.

There have been some studies involving IL, where models such as the Margules equation, the Wilson equation, the NRTL equation, and the UNIQUAC equation have been used.[1,2] In these works, it is stated that the NRTL equation gives the best empirical description of activity coefficients. Usually the model used to correlate the experimental data of systems containing ILs is the NRTL model.[3] Although this model is theoretically developed for systems involving nonelectrolytes, it usually leads to good correlations for systems containing IL.[4–8] Nevertheless, there is an extension of the NRTL model that considers the fact that ILs are organic salts that present ionic character, the e-NRTL model.[9–11] This model has also been used in the literature,[12,13] although it is less common. In any case, perhaps for the lack of models specifically created for systems with ILs and for its relative simplicity, the most used model is the NRTL model.

In this work, experimental VLE for the binary systems ethanol (1) + EMISE (2) and water (1) + EMISE (2) and for the ternary system ethanol (1) + water (2) + EMISE (3) were carried out at 101.3 kPa. In addition, a comparison of the correlation models commonly used for the treatment of these experimental data, the NRTL and the e-NRTL models, was made.

Table 1. Comparison of Measured Pure Component Properties Data with Literature Values at T = 298.15 K

	ρ/g·cm^{-3}		$10^3\eta$/Pa·s	
component	exptl	lit.	exptl	lit.
ethanol	0.78546	0.78546[a]	1.082	1.082[a]
water	0.99705	0.99705[b]	0.890	0.890[b]
EMISE	1.23763	1.23915[c] 1.2296[d]	97.58	100.77[c]

[a] From ref 16. [b] From ref 17. [c] From ref 18. [d] From ref 19.

Literature experimental data have been found for systems containing EMISE,[4,14] but only the work of Arce et al. has comparable conditions.

Experimental

Chemicals. Ethanol was purchased from Merck. It was degassed ultrasonically and dried over molecular sieves type 0.4 nm. Its purity was more than 99.8 %. Water was bidistilled and deionized. The IL used in this work was synthesized in our laboratory using the method described in a previous paper.[15] To ensure its purity, NMR measurements were made. The water concentration ($<3 \cdot 10^{-4}$ mass fraction) was checked by Karl Fischer titration. The IL was kept in bottles with an inert gas. To reduce the water content to negligible, a vacuum ($2 \cdot 10^{-1}$ Pa) and moderate temperature (343.15 K) were applied to the IL for several days, always immediately prior to its use.

The density and viscosity of ethanol, water, and EMISE were measured and compared with the literature (Table 1). These two components were also analyzed by gas chromatography to check its purity. The presented results show that there is a good agreement between experimental and literature data.

Apparatus and Procedure. A glass Fischer Labodest apparatus model 602/D was used for the VLE determinations. The equilibrium vessel is a dynamic recirculating still, and it is equipped with a Cottrell pump. A thermometer Yokogawa model 7563, with an uncertainty of ± 0.01 K, was used to measure the equilibrium temperature. For the pressure measurement, a digital pressure controller Ruska model 7218 with an uncertainty of 0.001 kPa was used.

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Table 2. Vapor–Liquid Equilibrium Data for the Ethanol (1) + EMISE (2) System at 101.3 kPa

T/K	x_1	γ_1
351.44	1.000	1.000
351.99	0.980	0.999
352.49	0.960	1.000
353.15	0.936	0.999
353.77	0.912	1.001
354.67	0.889	0.992
355.65	0.863	0.984
356.89	0.832	0.973
358.92	0.789	0.950
361.25	0.742	0.926
363.32	0.708	0.899
366.92	0.649	0.861
369.60	0.618	0.823
373.05	0.572	0.789
386.38	0.443	0.656
402.88	0.338	0.521

The initial sample for the binary VLE was the pure solvent (ethanol or water), then IL was added for each experimental point. For the ternary VLE, the initial sample was a binary mixture. When equilibrium was reached, vapor and liquid samples were obtained, and different amounts of solvents or IL were then added, in order to cover the whole composition range. In the range of low compositions of solvents, the temperature becomes unstable, and the IL could decompose. So, the experimental VLE were carried out as long as the equilibrium temperature was stable. For the binary system ethanol (1) + EMISE (2), x_2 reaches a composition of 0.66 approximately, corresponding to a mass fraction $w_2 \approx 0.91$ at a temperature of 402.88 K, and for the binary system water (1) + EMISE (2), x_2 reaches 0.42 approximately, corresponding to $w_2 \approx 0.905$ at a temperature of 410.58 K. In the ternary system ethanol (1) + water (2) + EMISE (3), the mass fraction of IL reached, w_3, was approximately 0.73 at a temperature of 375.60 K.

The liquid phase composition of the binary systems was determined by density measurements. The vapor phase composition was also measured to ensure that only ethanol or water was evaporated. To determine the liquid phase composition of the ternary system, it is necessary to know at least two physical properties of the ternary mixture. In this case, the density and refractive index physical properties were determined. The vapor phase, formed by a binary mixture ethanol + water, was measured by density. The mole fraction uncertainty of the technique for the determination of liquid and vapor composition is ± 0.001. Physical properties of these binary and ternary systems were determined in previous works.[15,20]

The densities of pure liquids and mixtures were measured with an Anton Paar DSA-5000 densimeter, and the repeatability and the uncertainty in experimental measurements have been found to be lower than ($\pm\ 2 \cdot 10^{-6}$ and $\pm\ 2.6 \cdot 10^{-5}$) $\text{g} \cdot \text{cm}^{-3}$ with a precision of $\pm\ 2 \cdot 10^{-6}\ \text{g} \cdot \text{cm}^{-3}$. To measure refractive indices, an automatic refractometer (Abbemat-HP Dr. Kernchen) with a resolution of $\pm\ 10^{-6}$ and an uncertainty in the experimental measurements of $\pm\ 4 \cdot 10^{-5}$ was used.

Results and Discussion

VLE for the binary systems ethanol (1) + EMISE (2) and water (1) + EMISE (2) as well as the VLE for the ternary system ethanol (1) + water (2) + EMISE (3) have been determined at 101.3 kPa, and the results are summarized in Tables 2 to 4. In Figure 1, a comparison between the experimental and literature data[4] of the VLE of the binary system ethanol (1) + EMISE (2) is shown. In this figure, the mass fraction is plotted because of the high difference between the molecular weight of the components ($M_{\text{EtOH}} = 46.069\ \text{g} \cdot \text{mol}^{-1}$, $M_{\text{EMISE}} = 236.29\ \text{g} \cdot \text{mol}^{-1}$). No literature isobaric VLE data have been found for the other studied systems.

Table 3. Vapor–Liquid Equilibrium Data for the Water (1) + EMISE (2) System at 101.3 kPa

T/K	x_1	γ_1
373.15	1.000	1.000
373.54	0.995	0.991
373.83	0.987	0.989
374.16	0.980	0.984
374.50	0.973	0.980
374.86	0.967	0.973
375.34	0.958	0.966
376.02	0.945	0.956
376.85	0.932	0.942
378.02	0.915	0.921
379.36	0.893	0.901
381.00	0.875	0.869
382.72	0.855	0.839
385.42	0.820	0.800
389.07	0.787	0.740
392.90	0.747	0.689
397.52	0.705	0.631
400.29	0.677	0.604
402.33	0.658	0.584
403.64	0.648	0.571
405.32	0.634	0.555
410.58	0.580	0.520

Table 4. Vapor–Liquid Equilibrium Data for the Ternary System Ethanol (1) + Water (2) + EMISE (3) at 101.3 kPa

T/K	x_1	x_2	y_1	γ_1	γ_2
357.34	0.346	0.545	0.690	2.300	3.329
358.58	0.232	0.690	0.604	3.267	2.503
359.34	0.179	0.755	0.581	4.129	2.218
360.40	0.137	0.810	0.544	5.167	1.985
361.50	0.104	0.851	0.507	6.544	1.812
362.69	0.082	0.880	0.464	7.901	1.674
359.01	0.152	0.811	0.534	4.905	2.092
357.20	0.210	0.756	0.574	3.801	2.412
356.23	0.262	0.706	0.610	3.166	2.685
355.20	0.347	0.623	0.629	2.489	3.167
354.48	0.430	0.542	0.645	2.065	3.748
353.93	0.538	0.435	0.668	1.687	4.776
355.12	0.347	0.635	0.623	2.500	3.118
356.54	0.229	0.755	0.586	3.581	2.477
357.40	0.175	0.813	0.561	4.527	2.226
358.40	0.137	0.852	0.550	5.588	2.040
362.41	0.062	0.922	0.444	10.639	1.615
364.45	0.062	0.906	0.390	9.818	1.521
360.81	0.102	0.866	0.498	6.820	1.828
363.79	0.089	0.847	0.465	7.057	1.667
366.52	0.074	0.840	0.420	7.692	1.518
370.40	0.053	0.836	0.351	9.416	1.320
370.46	0.119	0.754	0.379	4.130	1.462
375.60	0.060	0.755	0.370	6.917	1.214
354.18	0.520	0.445	0.672	1.729	4.621
356.00	0.408	0.513	0.689	2.051	3.727
357.32	0.374	0.520	0.695	2.129	3.487
359.24	0.347	0.521	0.696	2.136	3.231
361.72	0.301	0.530	0.693	2.245	2.884
365.11	0.268	0.518	0.689	2.226	2.593
353.65	0.638	0.336	0.673	1.438	6.254
353.79	0.606	0.355	0.694	1.506	5.893
354.60	0.518	0.420	0.707	1.707	4.813
355.81	0.456	0.453	0.717	1.851	4.257
357.32	0.415	0.464	0.643	1.917	3.912
356.35	0.495	0.394	0.743	1.670	4.788
355.71	0.546	0.347	0.761	1.551	5.572
357.33	0.510	0.352	0.762	1.562	5.158
359.48	0.446	0.375	0.777	1.645	4.447
364.48	0.387	0.366	0.787	1.575	3.760
360.05	0.531	0.264	0.835	1.352	6.165

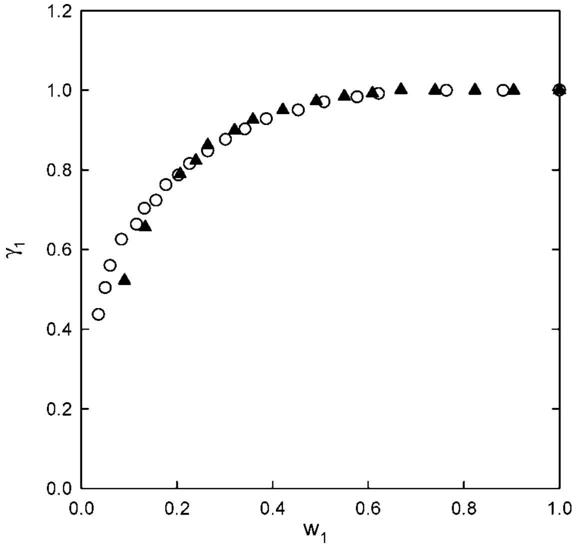

Figure 1. Comparison between ▲, experimental and ○, literature data (ref 4) as a function of mass fraction of the VLE of the binary system ethanol (1) + EMISE (2).

For the correlation of experimental data of VLE, two models are usually used: the NRTL model[3] has proved to be a good model for the correlation of VLE of systems containing IL; the e-NRTL model is an extension of the NRTL model which Chen et al.[9] derived as a model for single-solvent + electrolyte systems. Mock et al.[10,11] extended it to mixed-solvent + electrolyte systems. The three fundamental assumptions for the mixed-solvent electrolyte NRTL are: (1) the local composition of cations (anions) around a central cation (anion) is zero; (2) the distribution of cations and anions around a central solvent molecule is such that the net local ionic charge is zero; and (3) the long-range interaction contribution can be neglected.

Binary Systems. With comparison purposes, the binary VLE have been correlated using the NRTL and the e-NRTL models. In these correlations, the nonrandomness parameters α are also adjusted, obtaining better results than fixing them as constants. When these models are used for systems constituted by a solvent + salt, the equilibrium condition has to do only with the solvent, which is the component that is present in both phases. These correlations were made by minimizing the following objective function

$$\mathrm{OF} = \sum_j h|y_1 - y_{1,\mathrm{calcd}}| + \sum_j k|T - T_{\mathrm{calcd}}| \qquad (1)$$

where y_1 is the solvent mole fraction in the vapor phase (y_1 = 1 for binary systems); T is the equilibrium temperature; and h and k are the weighting factors. The calculated parameters and root-mean-square deviations for both correlation models are summarized in Table 5. The vapor pressures of the components were calculated using the Antoine equation.

Figure 2a shows the boiling temperature diagram of the experimental and calculated data, and Figure 2b shows the experimental and calculated activity coefficients for both binary systems and both correlation models. As can be observed in Table 5 and Figure 2, both models correlate satisfactorily the experimental data, giving very similar deviations.

Ternary Systems. From the binary adjustable parameters obtained in the correlations of the constituent binary systems, we can predict the VLE of the ethanol (1) + water (2) + EMISE (3) ternary system. In our system, nine binary adjustable parameters must be determined for each model, from which six are energy parameters and three are nonrandomness factors.

Table 5. Binary Correlation Parameters, Δ*g* and α, and Root-Mean-Square Deviations for the Binary Systems Ethanol (1) + EMISE (2) and Water (1) + EMISE (2) at 101.3 kPa

	Ethanol + EMISE			
NRTL	Δg_{12}/J·mol^{-1}	−5469.0	Δ*T*/K	0.17
	Δg_{21}/J·mol^{-1}	1074.8	Δ*y*	0.006
	α	0.21	Δγ	0.005
e-NRTL	Δg_{12}/J·mol^{-1}	−2016.9	Δ*T*/K	0.16
	Δg_{21}/J·mol^{-1}	−310.6	Δ*y*	0.006
	α	1.06	Δγ	0.005
	Water + EMISE			
NRTL	Δg_{12}/J·mol^{-1}	−139.9	Δ*T*/K	0.22
	Δg_{21}/J·mol^{-1}	−5161.0	Δ*y*	0.008
	α	0.65	Δγ	0.007
e-NRTL	Δg_{12}/J·mol^{-1}	998.6	Δ*T*/K	0.26
	Δg_{21}/J·mol^{-1}	−6053.6	Δ*y*	0.009
	α	0.16	Δγ	0.008

The binary parameters of the binary system ethanol (1) + water (2) have been taken from the literature.[21] The other necessary parameters have been determined in this work. Using these binary parameters previously determined, we can predict the behavior of the ternary system using the NRTL and e-NRTL models. Similar calculation of the behavior of ternary systems

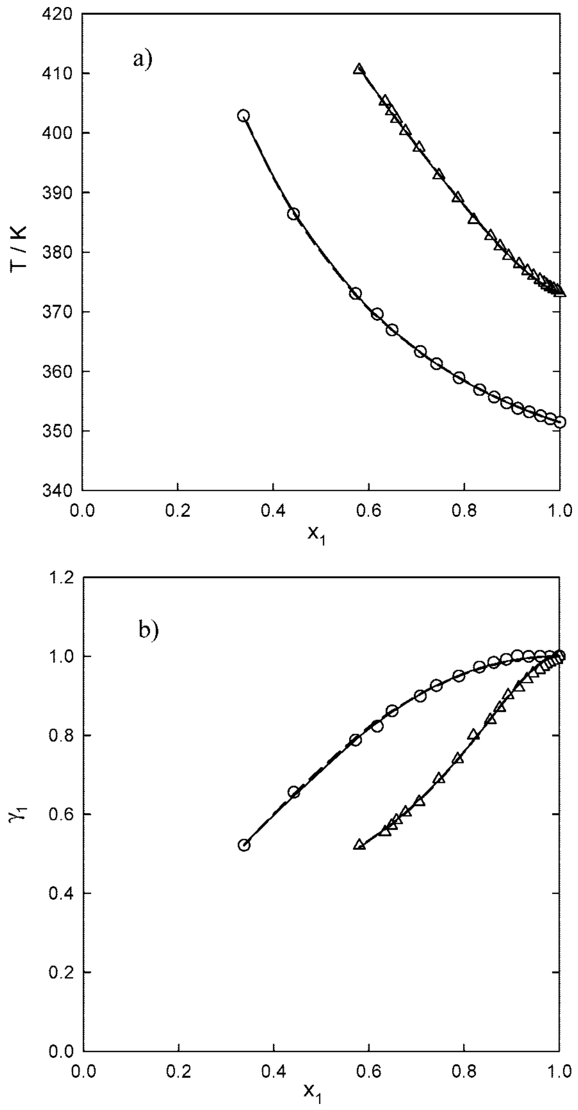

Figure 2. (a) Boiling temperature diagram and (b) activity coefficients diagram of experimental VLE data (○, EtOH + EMISE; Δ, H_2O + EMISE) and calculated correlation curves: —, NRTL; and — —, e-NRTL.

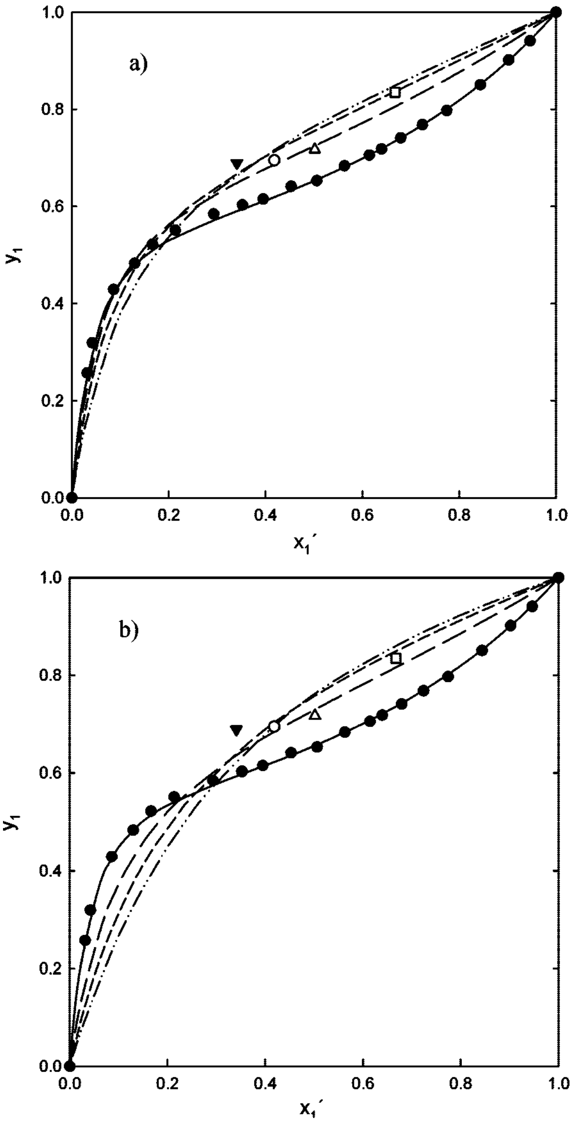

Figure 3. $x'y$ diagrams of the ternary system ethanol (1) + water (2) + EMISE (3). Prediction from binary parameters calculated with the (a) NRTL model and (b) e-NRTL model, at IL mole fractions of —, 0 %; — —, 10 %; -, 20 %; and — •• — ••, 30 %. •, experimental VLE data from ref 21; ○, experimental ternary point x_1 = 0.374, x_3 = 0.106; Δ, experimental ternary point x_1 = 0.456, x_3 = 0.091; ▼, experimental ternary point x_1 = 0.268, x_3 = 0.214; □, experimental ternary point x_1 = 0.531, x_3 = 0.205.

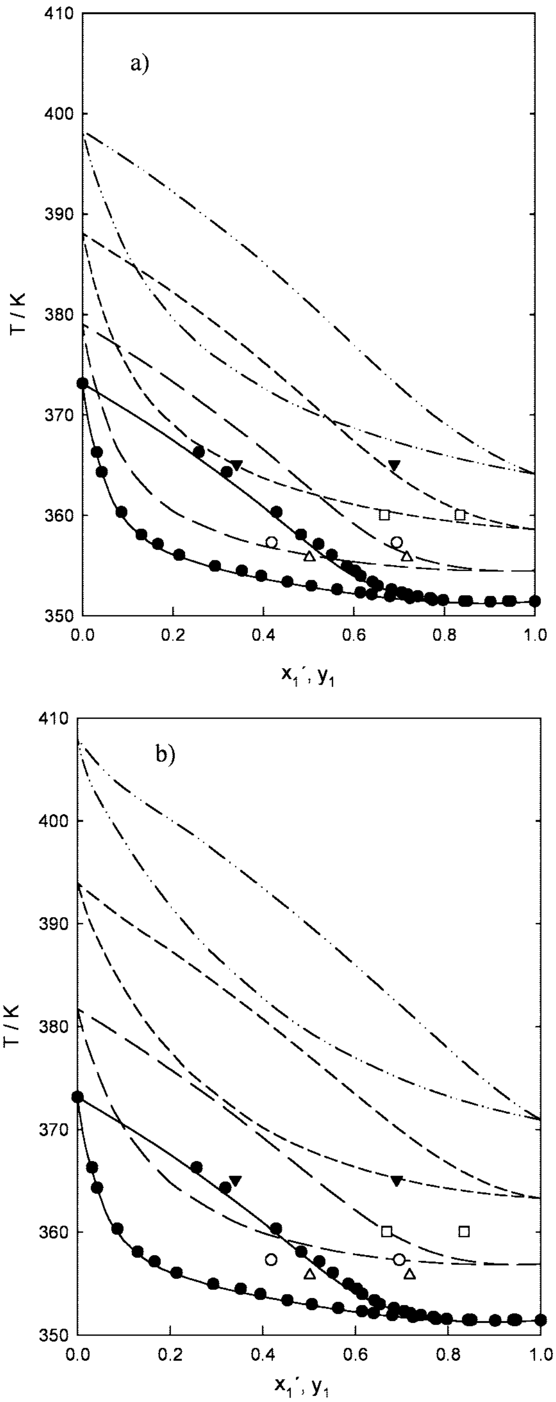

Figure 4. $Tx'y$ diagrams of the ternary system ethanol (1) + water (2) + EMISE (3). Prediction from binary parameters calculated with the (a) NRTL model and (b) e-NRTL model, at IL mole fractions of —, 0 %; — —, 10 %; -, 20 %; and — •• — ••, 30 %. •, experimental VLE data from ref 21; ○, experimental ternary point x_1 = 0.374, x_3 = 0.106; Δ, experimental ternary point x_1 = 0.456, x_3 = 0.091; ▼, experimental ternary point x_1 = 0.268, x_3 = 0.214; □, experimental ternary point x_1 = 0.531, x_3 = 0.205.

using the binary NRTL correlation parameters has been already reported by Döker and Gmehling.[22] The deviations obtained using the binary parameters for the prediction of the ternary system are ΔT = 1.14 and Δy = 0.04 for the NRTL parameters and ΔT = 3.35 and Δy = 0.05 for the e-NRTL parameters. According to these deviations, NRTL parameters give better results.

Pseudobinary figures have been plotted to make a comparison between models. In Figure 3a and b the predicted $x'y$ diagrams and in Figure 4a and b the predicted $Tx'y$ diagrams calculated with the NRTL and e-NRTL models, respectively, are presented. To verify that the predictions considering the mole fraction of IL = 0 (binary system) are satisfactory, experimental data of the binary system ethanol + water from the literature[21] are also plotted. In these figures, the liquid phase composition of the low-boiling component is the amount of this substance in the volatile part of the liquid phase. The IL concentration is stated for each curve separately for x_3 = (0, 10, 20, and 30) %. Different experimental points are also included to test the goodness of the predictions. These experimental points represent $x_3 \approx 0.1$ (x_1 = 0.456, x_3 = 0.091, x_1 = 0.374, x_3 = 0.106) and $x_3 \approx 0.2$ (x_1 = 0.268, x_3 = 0.214, x_1 = 0.531, x_3 = 0.205). As can be observed in these figures, both models have a similar prediction in the xy diagram, but the NRTL model predicts much better the equilibrium temperature, as could be concluded from the deviations.

Table 6. Ternary NRTL Correlation Parameters, Δg and α, Obtained from the Correlation of VLE Experimental Data of the Ternary System Ethanol (1) + Water (2) + EMISE (3) and Root-Mean-Square Deviations at 101.3 kPa

parameters	Δg_{12}/J·mol^{-1}	4459.63	Δg_{23}/J·mol^{-1}	−1749.17	α_{12}	−0.404
	Δg_{13}/J·mol^{-1}	−4647.33	Δg_{31}/J·mol^{-1}	731067	α_{13}	0.3018
	Δg_{21}/J·mol^{-1}	−3099.34	Δg_{32}/J·mol^{-1}	−4045.64	α_{23}	0.996
rmsd	ΔT	0.83	Δy	0.046		

To find out if it is worth correlating the ternary system separately or if it is enough with the prediction from binary parameters, we correlated the experimental data of the VLE of the ternary system ethanol (1) + water (2) + EMISE (3) using the NRTL model. The calculated parameters and deviations of this correlation are summarized in Table 6, and the pseudobinary representation of the *xy* and *Txy* diagrams is plotted in Figure 5.

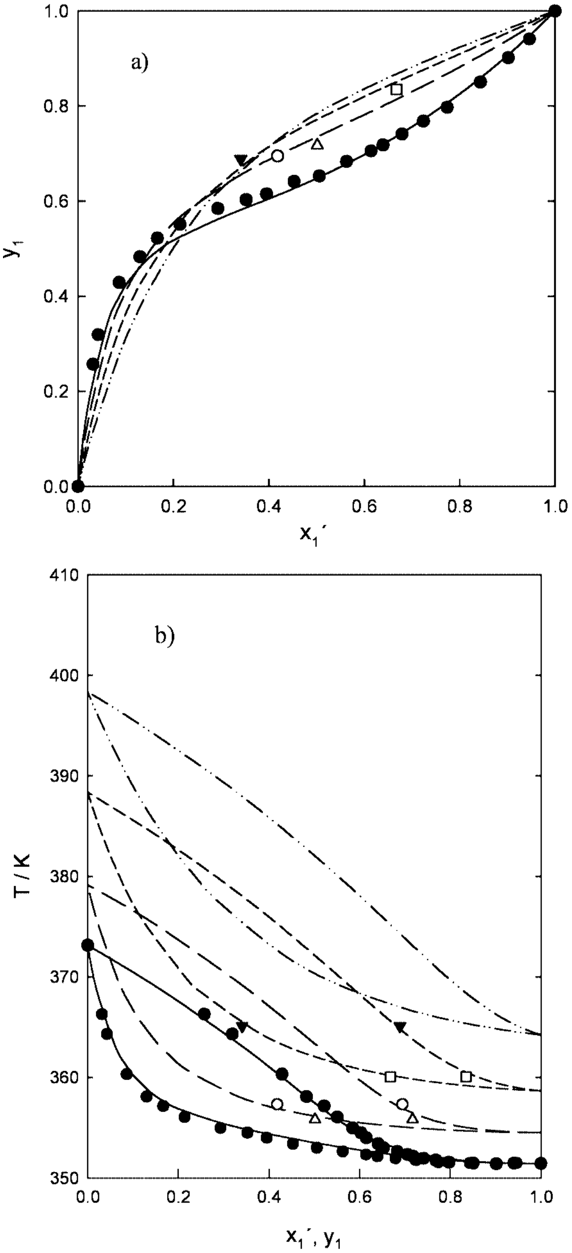

Figure 5. (a) *x′y* and (b) *Tx′y* diagrams of the ternary system ethanol (1) + water (2) + EMISE (3) from the NRTL ternary correlation, at IL mole fractions of —, 0 %; — —, 10 %; -, 20 %; and — •• — ••, 30 %. •, experimental VLE data from ref 21; ○, experimental ternary point x_1 = 0.374, x_3 = 0.106; Δ, experimental ternary point x_1 = 0.456, x_3 = 0.091; ▼, experimental ternary point x_1 = 0.268, x_3 = 0.214; □, experimental ternary point x_1 = 0.531, x_3 = 0.205.

In Figure 5a, we can observe that the addition of EMISE to this binary azeotropic mixture leads to the break of the binary azeotrope ethanol + water, producing a crossover effect, a salting-out effect near the azeotropic point, and a salting-in effect at small concentrations of ethanol.

Conclusions

In this work, experimental VLE of the ternary system ethanol (1) + water (2) + EMISE (3) at 101.3 kPa have been carried out, as well as the constituent binary systems containing the IL, ethanol (1) + EMISE (2) and water (1) + EMISE (2).

For the data treatment, two correlation models have been tested, the NRTL and e-NRTL models, usually used in the correlation of these kinds of systems. In the correlation of the binary systems, both models give similar results, making the correlations very satisfactory. Nevertheless, in the prediction of the behavior of the VLE of the ternary system from the binary correlation parameters, the NRTL model shows a better prediction, especially in the prediction of the equilibrium temperature.

The experimental data of the VLE of the ternary system have been correlated using the NRTL model, showing a good agreement with the experimental data. The deviations of this correlation are similar to the ones obtained in the prediction from the correlation binary parameters using the NRTL model. Comparing the Figures 3 to 5, we can observe that when the NRTL model is used the prediction of the behavior of the VLE of the ternary system is similar using the parameters obtained from the correlation of the binary systems or the ones obtained from the correlation of the ternary system. Slight differences are observed in the *Txy* diagram since the deviation of the equilibrium temperature obtained using the parameters of the correlation of the ternary system is lower. It can be concluded that the prediction of the behavior of the VLE of the ternary system can be calculated from the parameters obtained from the correlation of the binary systems using the NRTL model, obtaining very good results, but correlating the ternary system with the NRTL model, the agreement with experimental data is slightly higher, as expected.

From these correlations, it can be observed that with the addition of EMISE to the mixture ethanol + water the existing azeotrope can be broken, opening a new possibility as an entrainer for this IL.

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