J. Chem. Eng. Data **2009,** *54,* 945–949

Measurement and Correlation of Liquid−Liquid Equilibrium Data for Water + Acetic Acid + Methyl *tert*-Butyl Ether + NaCl

Hongxun Zhang* and Tiangui Wang

School of Chemistry and Chemical Engineering, Henan University of Technology, Zhengzhou, Henan 450001, China

Liquid−liquid equilibrium data of the quaternary system of water + acetic acid + methyl *tert*-butyl ether (MTBE) + NaCl at 298.15 K have been measured under atmospheric pressure. The liquid-phase splitting of the water + acetic acid + MTBE can be enhanced by the addition of salt NaCl. Selectivity of MTBE at different NaCl concentrations was calculated and compared. In addition, the experimental data were correlated with the NRTL and electrolyte−NRTL model. Also, parameters of these two models were regressed.

Introduction

The recovery of acetic acid from aqueous solutions is industrially and environmentally significant. There have been many various reports about the recovery of acetic acid, for example, distillation, extraction, neutralization, adsorption, membrane separation, and the integration of these methods.[1−5] The integration of extraction and azeotropic distillation is considered to be a comparatively effective method because of its lower energy cost. The key to its efficiency depends on extraction solvent selection. Kürüm et al.[6] evaluated 34 types of possible entrainers for acetic acid purification with extraction followed by azeotropic distillation in terms of selectivity, distribution coefficient, recoverability, density, chemical reactivity, viscosity, vapor pressure and freezing point, toxicity, cost, and so on, and concluded that ethyl acetate, diisopropyl ether, and methyl *tert*-butyl ether (MTBE) are the most promising entrainers. Among them, MTBE is the best because of its better selectivity, lesser density, lower vaporization enthalpy, and lower price. We measured the liquid−liquid equilibrium (LLE) data of the ternary system of water + acetic acid + MTBE experimentally and confirmed that MTBE is indeed an effective solvent for acetic acid extraction.[1] Because of this, salt-containing extraction has received a great deal of attention in recent years[7−9] and some practical purposes;[10] we investigated the effect of salt on the LLE of the system of water + acetic acid + MTBE in this work.

Table 1. Mass Fraction LLE Data for Water (1) + Acetic Acid (2) + MTBE (3) + NaCl (4) at Different Mass Fractions of NaCl (w_s) and T = 298.15 K

solvent-rich phase				water-rich phase			
w_1^{I}	w_2^{I}	w_3^{I}	w_4^{I}	w_1^{II}	w_2^{II}	w_3^{II}	w_4^{II}
$w_s = 0$							
0.0116	0.0000	0.9884		0.9548	0.0000	0.0452	
0.0259	0.0464	0.9277		0.9062	0.0476	0.0462	
0.0393	0.1122	0.8485		0.8384	0.1071	0.0545	
0.0721	0.1793	0.7486		0.7899	0.1525	0.0576	
0.1068	0.2554	0.6378		0.6950	0.2251	0.0799	
0.1620	0.2937	0.5443		0.6253	0.2686	0.1061	
0.2357	0.3300	0.4343		0.5302	0.3175	0.1523	
0.3065	0.3343	0.3592		0.4505	0.3248	0.2247	
0.3341	0.3372	0.3287		0.4247	0.3281	0.2472	
$w_s = 0.01$							
0.0188	0.0000	0.9800	0.0012	0.9740	0.0000	0.0207	0.0053
0.0414	0.1186	0.8383	0.0017	0.8579	0.0987	0.0373	0.0061
0.0743	0.2112	0.7109	0.0036	0.7729	0.1619	0.0545	0.0107
0.1004	0.2624	0.6325	0.0047	0.7250	0.1967	0.0662	0.0121
0.1305	0.3079	0.5558	0.0058	0.6757	0.2312	0.0800	0.0131
0.1933	0.3712	0.4274	0.0081	0.5732	0.2967	0.1160	0.0141
0.2153	0.3848	0.3906	0.0093	0.5289	0.3206	0.1358	0.0147
0.2730	0.4100	0.3120	0.0050	0.4423	0.3720	0.1792	0.0065
$w_s = 0.05$							
0.0152	0.0000	0.9775	0.0073	0.9509	0.0000	0.0171	0.0320
0.0411	0.1430	0.8041	0.0118	0.8439	0.0880	0.0338	0.0343
0.0778	0.2379	0.6665	0.0178	0.7448	0.1551	0.0561	0.0440
0.1097	0.3018	0.5687	0.0198	0.6880	0.1906	0.0739	0.0475
0.1541	0.3772	0.4434	0.0253	0.6013	0.2551	0.0896	0.0540
0.1842	0.4147	0.3732	0.0279	0.5316	0.3120	0.1081	0.0483
0.2035	0.4383	0.3290	0.0292	0.4801	0.3492	0.1220	0.0487
0.2477	0.4449	0.2755	0.0319	0.3857	0.3978	0.1641	0.0524
$w_s = 0.1$							
0.0031	0.0000	0.9909	0.0060	0.9613	0.0000	0.0104	0.0283
0.0398	0.1993	0.7467	0.0142	0.8761	0.0738	0.0151	0.0350
0.0647	0.3264	0.5778	0.0311	0.7700	0.1151	0.0251	0.0898
0.0823	0.3878	0.4914	0.0385	0.7005	0.1701	0.0316	0.0978
0.0933	0.4240	0.4412	0.0415	0.6569	0.2100	0.0357	0.0974
0.1052	0.4401	0.4046	0.0501	0.6125	0.2368	0.0423	0.1084
0.1378	0.4662	0.3265	0.0695	0.5194	0.2970	0.0583	0.1253
0.2254	0.4553	0.2092	0.1101	0.3917	0.3721	0.0940	0.1422

Experimental Section

Chemicals. MTBE with a purity of 99.9 % was purchased from Fisher. Acetic acid with a purity of 99.5 % and sodium chloride with analytical purity were purchased from Shanghai Chemical Reagent. Double-distilled water was prepared in our laboratory. The salt compounds were dried in an oven until the weight remained constant before being used. All liquid chemicals were checked with chromatographic analysis and used without further purification.

Apparatus and Procedure. All LLE data for the systems of interest were measured under atmospheric pressure. The experimental apparatus included a jacketed glass cell (internal volume about 50 cm^3), a thermostatically controlled bath, and a magnetic agitator. The circulating water from the thermostatically controlled bath was introduced to the jacket to keep the temperature of the liquid mixture essentially constant. The temperature fluctuation was controlled within ± 0.05 K. At the beginning of the experiments, water, acetic acid, MTBE, and NaCl were added to the cell by mass at known ratios. The weights of these reagents were determined by an electronic

---

* Corresponding author. E-mail: ztonghy@163.com. Tel: 86-371-67756725.

10.1021/je800724g CCC: $40.75

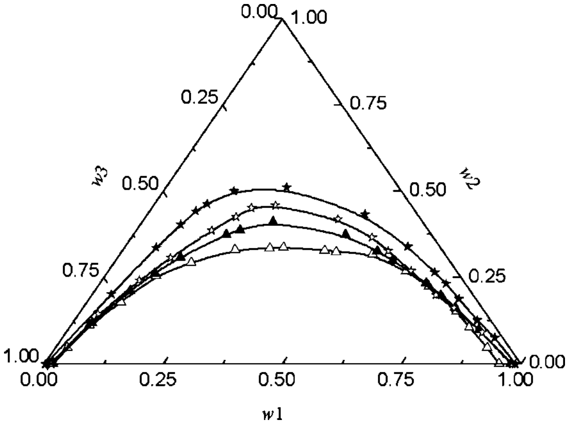

Figure 1. Comparison of the LLE phase diagram for water (1) + acetic acid (2) + MTBE (3) + NaCl (4) at different mass fractions of NaCl (w_s) and T = 298.15 K. Each coordinate variable is on a salt-free basis: △, w_s = 0 (salt free); ▲, w_s = 0.01; ☆, w_s = 0.05; ★, w_s = 0.1.

balance (BS110S, ± 0.1 mg).The heterogeneous mixtures were stirred for 2 h with a magnetic stirrer at 298.15 K and were allowed to settle for (3 to 6) h for complete phase separation. Samples were carefully taken from each phase and analyzed. The upper solvent-rich phase and the lower water-rich phase were sampled using different syringes.

Sample Analysis. First, the composition of salt in each phase was determined by gravimetric analysis. A measured amount of liquid sample was transferred to a weighed watch glass, which was then dried in a vacuum oven until the weight of the salt remained constant. A gas chromatograph (model GC-102D, Shanghai Precision Instrument, China) equipped with a thermal conductivity detector was used to analyze the composition of the salt-free samples. A stainless steel chromatographic column packed with Porapak Q ((2 m)·(3 mm)), which was clearly capable of separating the compounds of the salt-free samples, was used for chromatographic analysis. The temperatures of the oven, injection port, and detector were held at 385.15 K, 413.15 K, and 423.15 K, respectively. The flow rate of the carrier gas, hydrogen, was kept at 20 mL·min^{-1}. The external standard method was used to analyze the content of the three components.

Results and Discussion

Experimental Results. The LLE data of the quaternary system of water + acetic acid + MTBE + NaCl at mass fraction of NaCl of w_s = 0.01, w_s = 0.05, and w_s = 0.1 at 298.15 K have been measured. In a previous investigation,[1] we obtained the data of the ternary system of water + acetic acid + MTBE at 298.15 K. For the sake of comparison, LLE data of these two systems of interest are all listed in Table 1. The difference in behavior of the LLE data can be seen from the LLE phase diagram shown in Figure 1.

To estimate the capability of MTBE at different NaCl concentrations to separate acetic acid, the distribution ratios, D_1 for water and D_2 for acetic acid, and the selectivity, S_{MTBE}, for MTBE were calculated and are also listed in Table 2. D_i and S_{MTBE} were determined as follows

$$D_i = \frac{w_i^{\mathrm{I}}}{w_i^{\mathrm{II}}} \tag{1}$$

$$S_{\mathrm{MTBE}} = \frac{D_2}{D_1} \tag{2}$$

where w is the mass fraction; the superscript I represents the solvent-rich phase and II represents the water-rich phase; the subscript i is equal to 1 or 2 (i = 1 represents water and i = 2 acetic acid). The selections of MTBE at different NaCl concentrations are shown in Figure 2.

From Figure 1, it can be seen that the areas of two phases of each sample system become larger with the increase in concentration of NaCl. The addition of salt to the system is beneficial for separation and enlarges the operation range of extraction. Figure 2 clearly shows that the selectivity of MTBE also gradually increases with the increase in concentration of NaCl.

There are several varieties of equations that are useful for considering the reliability of the experimental data.[11−13] To extend its application to salt solution systems, Rajendran somewhat improved the Eisen−Joffe equation. The revised equation is[14,15]

$$\lg\frac{w_2^{\mathrm{I}}}{w_3^{\mathrm{I}}} = a + bw_s + (c + dw_s)\lg\frac{w_2^{\mathrm{II}}}{w_1^{\mathrm{II}}} \tag{3}$$

where w_2^{I} represents the mass fraction of solute (acetic acid) in the solvent-rich phase, w_3^{I} represents the mass fraction of solvent (MTBE) in the solvent-rich phase, w_1^{II} represents the mass fraction of water in the water-rich phase, w_2^{II} represents the mass fraction of solute (acetic acid) in the water-rich phase, and w_s represents the mass fraction of salt. The letters a, b, c, and d are parameters of the Eisen−Joffe equation.

Parameters of the Eisen−Joffe equation and results of correlation of the LLE data for water + acetic acid + MTBE + NaCl are listed in Table 3. All linear correlation coefficients (R) are greater than 0.9911, and standard deviations (SD) are less than 0.0427. These results suggest that it is reasonable to accept the LLE data of the considered systems as being reliable.

Liquid−Liquid Equilibrium Calculations. The relationship of LLE can be represented by an activity coefficient model. In this model, the basic relationships for every component i in two coexistent liquid phases of a system at equilibrium are

$$x_i^{\mathrm{I}} r_i^{\mathrm{I}} = x_i^{\mathrm{II}} r_i^{\mathrm{II}} \tag{4}$$

$$\sum x_i^{\mathrm{I}} = 1 \tag{5}$$

Table 2. Distribution Coefficients (D_i) of Water (1) and Acetic Acid (2) and Separation Factors of MTBE (S_{MTBE}) at Different Mass Fractions of NaCl (w_s) and T = 298.15 K

D_1	D_2	S_{MTBE}	D_1	D_2	S_{MTBE}	D_1	D_2	S_{MTBE}	D_1	D_2	S_{MTBE}
	w_s = 0			w_s = 0.01			w_s = 0.05			w_s = 0.1	
0.0286	0.9748	34.1064	0.0483	1.2016	24.9003	0.0487	1.6250	33.3659	0.0454	2.7005	59.4459
0.0469	1.0476	22.3492	0.0961	1.3045	13.5701	0.1045	1.5338	14.6839	0.0840	2.8358	33.7490
0.0913	1.1757	12.8809	0.1385	1.3340	9.6330	0.1594	1.5834	9.9307	0.1175	2.2798	19.4049
0.1537	1.1346	7.3834	0.1931	1.3317	6.8955	0.2563	1.4786	5.7697	0.1420	2.0190	14.2156
0.2591	1.0934	4.2206	0.3372	1.2511	3.7099	0.3465	1.3292	3.8360	0.1718	1.8585	10.8208
0.4445	1.0394	2.3380	0.4071	1.2002	2.9485	0.4239	1.2552	2.9612	0.2653	1.5697	5.9166
0.6804	1.0292	1.5128	0.6172	1.1022	1.7856	0.6422	1.1184	1.7415	0.5754	1.2236	2.1264
0.7867	1.0277	1.3064									

$$\sum x_i^{\mathrm{II}} = 1 \qquad (6)$$

where x_i^{I} and x_i^{II} are the mole fractions of component i in phases I and II, and r_i^{I} and r_i^{II} are the activity coefficients of component i in phases I and II. The key to solving the set of equations is to calculate the activity coefficients. The activity coefficients of the quaternary system above were calculated with the electrolyte−NRTL model by Aspen Plus, and the activity coefficients of the ternary system were calculated with the NRTL model.

The electrolyte−NRTL model was originally proposed by Chen et al. for aqueous electrolyte systems.[16] It was later extended to mixed solvent electrolyte systems.[17] When binary and pair parameters are employed, the model can represent aqueous electrolyte systems as well as mixed-solvent electrolyte systems over the entire range of electrolyte concentrations. The model reduces to the well-known NRTL model when electrolyte concentrations become zero.[18] Chen proposed an excess Gibbs energy expression that contains two contributions: one contribution for the long-range ion−ion interactions that exist beyond the immediate neighborhood of a central ionic species, and the other related to the local interactions that exist at the immediate neighborhood of any central species. The unsymmetrical Pitzer−Debye−Hückel model and the Born equation are used to represent the contribution of the long-range ion−ion interactions, and the NRTL theory is used to represent the local interactions. This NRTL expression for the local interactions, the Pitzer−Debye−Hückel expression, and the Born equation are added to give eq 7 for the excess Gibbs energy

$$\frac{G_{\mathrm{m}}^{*\mathrm{E}}}{RT} = \frac{G_{\mathrm{m}}^{*\mathrm{E,PDH}}}{RT} + \frac{G_{\mathrm{m}}^{*\mathrm{E,Born}}}{RT} + \frac{G_{\mathrm{m}}^{*\mathrm{E,lc}}}{RT} \qquad (7)$$

This leads to

$$\ln \gamma_i^{*} = \ln \gamma_i^{*\mathrm{PDH}} + \ln \gamma_i^{*\mathrm{Born}} + \ln \gamma_i^{*\mathrm{lc}} \qquad (8)$$

where $\gamma^{*\mathrm{PDH}}$ is the long-range interaction contribution term (the Pitzer−Debye−Hückel equation), $\gamma^{*\mathrm{Born}}$ is the Born term that transfers the reference state of ions from the infinitely dilute state in a mixed solvent to an aqueous infinitely dilute state, and $\gamma^{*\mathrm{lc}}$ is the short-range interaction contribution term (based on the local composition concept of the original NRTL model).

Each type of electrolyte−NRTL parameter consists of both the nonrandomness factor, α, and energy parameters, τ. The detailed temperature dependency relations of the electrolyte−NRTL parameters for each term have been given elsewhere.[16−18]

The significant derived parameters obtained in this work were regressed and listed in Tables 4 and 5. The LLE diagrams under atmospheric pressure for water + acetic acid + MTBE + NaCl at different NaCl concentrations are shown in Figures 3, 4, 5, and 6. These LLE diagrams compare the experimental results with the calculated binodal locus and tie lines from the NRTL and electrolyte−NRTL models.

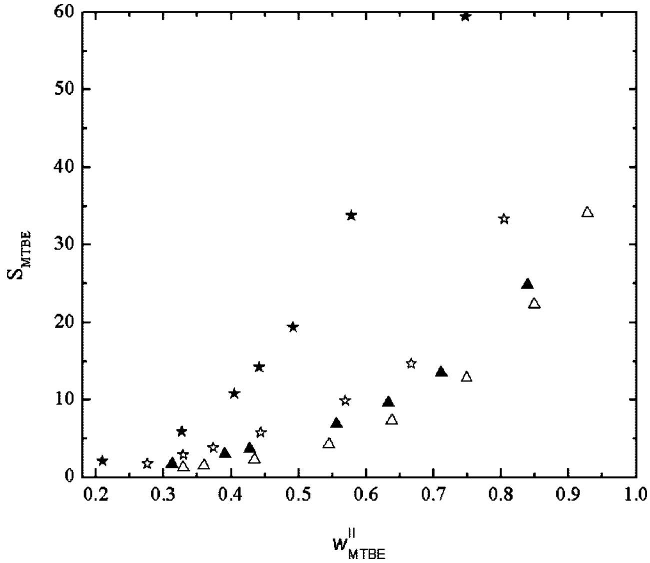

Figure 2. Selectivity of MTBE in two liquid phases at different mass fractions of NaCl (w_s) and T = 298.15 K: △, w_s = 0 (salt free); ▲, w_s = 0.01; ☆, w_s = 0.05; ★, w_s = 0.1.

Table 3. Results of Correlation of the LLE Data Correlation for Water (1) + Acetic acid (2) + MTBE (3) + NaCl (4) at Different Mass Fractions of NaCl (w_s) and T = 298.15 K

w_s	a	b	c	d	R^a	SD^a
0	0.1389		1.1173		0.9992	0.0196
0.01		0.1026		0.0146	0.9972	0.0276
0.05		0.0229		−0.0242	0.9943	0.0403
0.1		0.0237		−0.0293	0.9911	0.0427

[a] Where R is the linear correlation coefficient and SD is the standard deviation.

Table 4. Calculated Results from the NRTL Model for Water (1) + Acetic Acid (2) + MTBE (3) at T = 298.15 K

i-j	α_{ij}^a	τ_{ij}^a	τ_{ji}^a
1-2	0.47	0.3540	− 1.2151
1-3	0.20	3.9737	1.2998
2-3	0.37	− 0.2774	− 2.8068

[a] α_{ij} is the nonrandomness factor of the NRTL model, and τ_{ij} and τ_{ji} are interaction energy parameters of the NRTL model.

Table 5. Calculated Results from the Electrolyte−NRTL Model for Water (1) + Acetic acid (2) + MTBE (3) + NaCl (4) at T = 298.15 K

i-j	α_{ij}^a	τ_{ij}^a	τ_{ji}^a
1-4	0.20	8.9012	− 4.5721
2-4	0.10	15.0335	− 8.0067
3-4	0.10	10.0335	− 2.0067

[a] α_{ij} is the nonrandomness factor of the electrolyte−NRTL model, and τ_{ij} and τ_{ji} are the interaction energy parameters of the electrolyte−NRTL model. The values of the nonrandomness parameters (α and τ) for (1-2), (1-3), and (2-3) pairs are the same as those in Table 4.

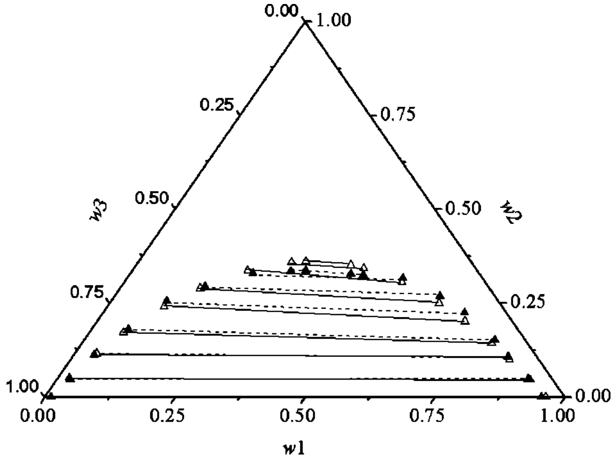

Figure 3. LLE phase diagram for water (1) + acetic acid (2) + MTBE (3) at T = 298.15 K: △, exptl data; ▲, NRTL correlation; —, exptl tie line; ---, NRTL tie line.

The root-mean-square deviations (RMSD) of the correlations with different models were calculated according to the following

Table 6. Root-Mean-Square Deviations Obtained from NRTL and Electrolyte−NRTL Models at Different Mass Fractions of NaCl (w_s) and T = 298.15 K

w_s	NRTL	electrolyte−NRTL
0	0.0108	
0.01		0.0033
0.05		0.0129
0.1		0.0091
av		0.0084

equation. Equation 9 is applicable to the ternary system of water + acetic acid + MTBE, and eq 10 is applicable to the quaternary system of water + acetic acid + MTBE + NaCl

$$\mathrm{RMSD}=\left(\sum_{k=1}^{n}\sum_{j=1}^{2}\sum_{i=1}^{3}\left(\frac{(w_{ijk}^{\mathrm{calcd}}-w_{ijk}^{\mathrm{obsd}})^2}{6n}\right)\right)^{1/2} \qquad (9)$$

$$\mathrm{RMSD}=\left(\sum_{k=1}^{n}\sum_{j=1}^{2}\sum_{i=1}^{3}\left(\frac{(w_{ijk}^{\mathrm{calcd}}-w_{ijk}^{\mathrm{obsd}})^2}{8n}\right)\right)^{1/2} \qquad (10)$$

where w_{ijk}^{calcd} and w_{ijk}^{obsd} are the calculated and the observed mass fractions of component i in phase j on tie line k, and n is the number of tie lines.

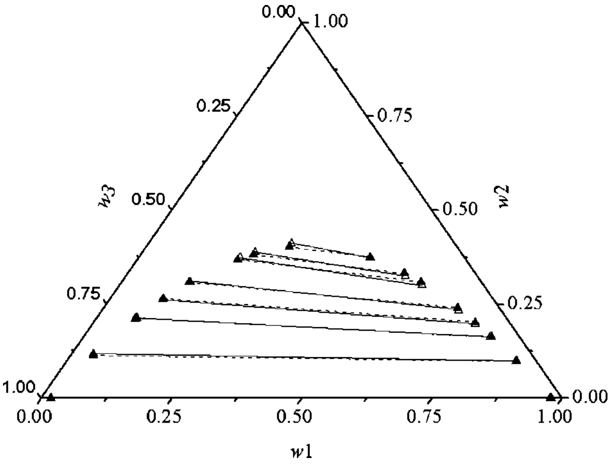

Figure 4. LLE phase diagram for water (1) + acetic acid (2) + MTBE (3) + NaCl (4) at mass fraction of NaCl w_s = 0.01and T = 298.15 K. Each coordinate variable is on a salt-free basis: △, exptl data; ▲, electrolyte−NRTL correlation; —, exptl tie line; ---, electrolyte−NRTL tie line.

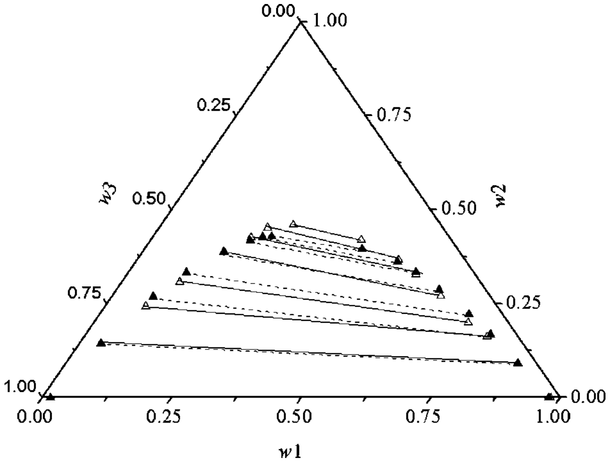

Figure 5. LLE phase diagram for water (1) + acetic acid (2) + MTBE (3) + NaCl (4) at mass fraction of NaCl w_s = 0.05 and T = 298.15 K. Each coordinate variable is on a salt-free basis: △, exptl data; ▲, electrolyte−NRTL correlation; —, exptl tie line; ---, electrolyte−NRTL tie line.

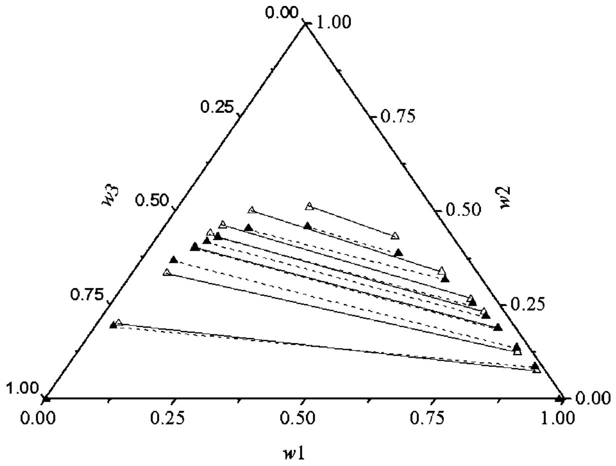

Figure 6. LLE phase diagram for water (1) + acetic acid (2) + MTBE (3) + NaCl (4) at mass fraction of NaCl w_s = 0.1 and T = 298.15 K. Each coordinate variable is on a salt-free basis: △, exptl data; ▲, electrolyte−NRTL correlation; —, exptl tie line; ---, electrolyte−NRTL tie line.

Table 6 gives the RMSD values for the two models at different salt concentrations. As seen from Table 6 and Figures 3, 4, 5, and 6, good agreements between the calculated values and experimental results have been obtained for the two systems.

Conclusions

LLE data for the quaternary system of water + acetic acid + MTBE + NaCl at 298.15 K under atmospheric pressure were determined at different sodium chloride concentrations. The two-phase region increased with increasing concentrations of sodium chloride. In addition, parameters of NRTL and electrolyte−NRTL models for the investigated systems were regressed. A comparison between experimental data and calculated values was drawn. The results show that the LLE of the investigated systems could be well correlated with electrolyte−NRTL.

Literature Cited

(1) Miao, X.; Zhang, H.; Wang, T.; He, M. Liquid−Liquid Equilibria of the Ternary System Water + Acetic Acid + Methyl *tert*-Butyl Ether. *J. Chem. Eng. Data* **2007**, *52*, 789–793.

(2) Xiao, X. J.; Wang, L. J.; Ding, G. H.; Li, X. Liquid−Liquid Equilibria for the Ternary System Water + Acetic Acid + Propyl Acetate. *J. Chem. Eng. Data* **2006**, *51*, 582–583.

(3) Ince, E. Liquid−Liquid Equilibria of the Ternary System Water + Acetic Acid + Dimethyl Adipate. *Fluid Phase Equilib.* **2005**, *230*, 58–63.

(4) Colombo, A.; Battilana, P.; Ragaini, V.; Bianchi, C. L. Liquid−Liquid Equilibria of the Ternary Systems Water + Acetic Acid + Ethyl Acetate and Water + Acetic Acid + Isophorone (3,5,5-Trimethyl-2-cyclohexen-1-one). *J. Chem. Eng. Data* **1999**, *44*, 35–39.

(5) Fahim, M. A.; Al-Muhtaseb, S. A.; Al-Nashef, I. M. Liquid−Liquid Equilibria of the Ternary System Water + Acetic Acid + 1-Hexanol. *J. Chem. Eng. Data* **1997**, *42*, 183–186.

(6) Kürüm, S.; Fonyo, Z.; Kut, Ö. M. Design Strategy for Acetic Acid Recovery. *Chem. Eng. Commun.* **1995**, *136*, 161–176.

(7) Chen, J. T.; Chen, M. C. Liquid−Liquid Equilibria for the Quaternary Systems of Water + *N*-Methyl-2-pyrrolidone + 1-Hexanol + NaCl, + KCl, or + KAc. *J. Chem. Eng. Data* **2008**, *53*, 217–222.

(8) Aznar, M.; Araujo, R. N.; Romanato, J. F.; Santos, G. R.; d'Avila, S. G. Salt Effects on Liquid−Liquid Equilibrium in Water + Ethanol + Alcohol + Salt Systems. *J. Chem. Eng. Data* **2000**, *45*, 1055–1059.

(9) Wang, J. G.; Zhang, Y. M. Liquid−Liquid Equilibrium for 1-Propanol (or 2-Propanol) + Water Systems Containing Potassium Fluoride. *J. Chem. Eng. Data* **2002**, *47*, 110–112.

(10) Zhang, H.; Miao, X.; Jiang, Sh.; Xu, M.; Wang, T. Recovery of Acidic Acid from Process of Waste Solution. *Chem. Eng.* **2006**, 57–59.

(11) Othmer, D. F.; Tobias, P. E. Tie-Line Correlation. *Ind. Eng. Chem.* **1942**, *34*, 693–700.

(12) Hand, D. B. The Distribution of Consolute Liquid between Two Immiscible Liquids. *J. Phys. Chem.* **1930**, *34*, 1961–2000.

(13) Eisen, E. O.; Joffe, J. Salt Effects in Liquid−Liquid Equilibria. *J. Chem. Eng. Data* **1966**, *11*, 480–484.

(14) Rajendran, M.; Srinivasan, D. Measurement and Prediction of Liquid−Liquid Equilibria of Ethyl Acetate + Methanol + Water in the Presence of Dissolved Inorganic Salts. *Fluid Phase Equilib.* **1988**, *44*, 53–75.

(15) Rajendran, M.; Renganarayanan, S.; Srinivasan, D. Salt Effect in Phase Equilibria: Effect of Dissolved Inorganic Salts on the Liquid−Liquid Equilibria of Benzene-2−Propanol−Water System and the Vapor−Liquid Equilibria of its Constituent Binaries. *Fluid Phase Equilib.* **1989**, *50*, 133–164.

(16) Chen, C. C.; Evans, L. B. A Local Composition Model for the Excess Gibbs Energy of Aqueous Electrolyte Systems. *AIChE J.* **1986**, *32*, 444–454.

(17) Mock, B.; Evans, L. B.; Chen, C. C. Thermodynamic Representation of Phase Equilibria of Mixed-Solvent Electrolyte Systems. *AIChE J.* **1986**, *32*, 1655–1664.

(18) Renon, H.; Prausnitz, J. M. Local Compositions in Thermodynamic Excess Functions for Liquid Mixtures. *AIChE J.* **1968**, *14*, 135–144.

Received for review September 29, 2008. Accepted December 11, 2008.

JE800724G