Tie-line data for aqueous mixtures of butyric acid with isobutyl acetate at various temperatures

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Abstract–Experimental liquid-liquid equilibrium (LLE) data for the system of (water+butyric acid+*iso*-butyl acetate) were obtained at T=(298.2, 303.2, 308.2, and 313.2) K and atmospheric pressure. This ternary system exhibits type-1 behavior of LLE. The experimental tie-line data were correlated using the UNIQUAC and NRTL models. The reliability of the experimental tie-line data was determined by applying the Othmer-Tobias and Hand correlations. Distribution coefficients and separation factors were calculated over the immiscibility regions.

Key words: Butyric Acid, LLE Data, Ternary Mixture, NRTL Model, UNIQUAC Model

INTRODUCTION

Liquid phase equilibrium data for extraction of carboxylic acids from water are important in estimation of industrial solvent extraction units. The LLE data are always required for efficient separation operations, which can be obtained from direct measurements [1-8].

Butyric acid (BA) is one of the important carboxylic acids, which has many scientific and industrial applications. Therefore, the extraction of this acid from water is an important problem [9-11]. In recent years, some important data have been reported on the LLE measurements and the extraction of BA from aqueous solutions [12-18].

In this research, *iso*-butyl acetate was chosen as organic solvent for recovery of BA from water. This solvent has been used in the determination of LLE data for the aqueous mixture of phosphoric acid with organic solvent [19]. To investigate the change of equilibrium characteristics, four different temperatures were selected. At each temperature, T=(298.2, 303.2, 308.2 and 313.2) K, the phase compositions were measured.

Distribution coefficients and separation factors were calculated from the tie-line data to establish the possibility of the use of this solvent for the separation of BA from water. The experimental data were correlated using the UNIQUAC and NRTL models [20,21], and the values for the interaction parameters were obtained. In general, both of the correlation models are applicable for the LLE correlation of the investigated system.

EXPERIMENTAL SECTION

1. Materials

Butyric acid and *iso*-butyl acetate with stated mass fraction purity higher than 0.99, and 0.995, respectively, were obtained from Merck. The stated purity of the materials was checked based on their refractive indices and densities. Deionized and redistilled water was

Table 1. The refractive index (n) and density (ρ) of the pure components at T=298.2 K

Component	n		ρ/(kg·m^{-3})	
	Exp.	Lit.	Exp.	Lit.
Butyric acid	1.3976	1.3975 [13]	952.72	952.80 [13]
iso-Butyl acetate	1.3878	1.3876 [30]	866.29	866.12 [30]
Water	1.3324	1.3325 [31]	997.09	997.04 [31]

used throughout all the experiments. All materials were used as received without any further purification. The measured refractive index and density data of the chemicals used in this study along with the literature values are listed in Table 1.

2. Apparatus and Procedure

The solubility curves for the ternary mixtures were determined by the cloud point method in an equilibrium glass cell [22]. The binary mixtures of known compositions were prepared by weighting and introduced into the glass cell. The temperature of the cell was controlled by a water jacket and maintained with an accuracy of within ±0.1 K that was checked with a digital thermometer (Lutron TM-917). At each temperature, the third component was progressively added using a microburet. The end-point was determined by observing the transition from an appearance to a disappearance of the heterogeneous mixture. All the measurements were repeated at least three times. The average of these readings was taken for the component compositions and reported in Table 2.

To measure the LLE tie-line data for the investigated system, a 250 ml glass cell connected to a thermostat was used. The equilibrium data were determined by preparing the ternary mixtures of known compositions. The prepared mixtures were introduced into the extraction cell and were stirred for 4 h, and then left to settle for 6 h for phase separation. For this ternary system, this time is long enough to achieve equilibrium. When equilibrium was achieved, the system was separated into two liquid phases that became clear and transparent with a well-defined interface. Then, the organic liquid phase was almost completely collected with glass syringe into a

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Table 2. Experimental solubility data in mole fraction for (water+BA+*iso*-butyl acetate) at different temperatures[a]

T=298.2 K		T=303.2 K		T=308.2 K		T=313.2 K	
x_1 (water)	x_2 (BA)	x_1 (water)	x_2 (BA)	x_1 (water)	x_2 (BA)	x_1 (water)	x_2 (BA)
0.999	0.000	0.999	0.000	0.999	0.000	0.076	0.000
0.904	0.093	0.899	0.094	0.928	0.061	0.134	0.071
0.903	0.094	0.883	0.111	0.895	0.096	0.182	0.115
0.881	0.115	0.811	0.179	0.813	0.177	0.233	0.174
0.839	0.153	0.742	0.235	0.724	0.241	0.292	0.225
0.774	0.208	0.672	0.280	0.572	0.317	0.342	0.269
0.692	0.277	0.600	0.317	0.438	0.331	0.430	0.313
0.506	0.363	0.500	0.349	0.357	0.302	0.515	0.316
0.410	0.350	0.430	0.345	0.331	0.283	0.603	0.289
0.347	0.318	0.359	0.318	0.287	0.233	0.691	0.247
0.306	0.282	0.299	0.265	0.240	0.174	0.776	0.189
0.255	0.215	0.252	0.203	0.200	0.124	0.851	0.127
0.181	0.108	0.187	0.117	0.137	0.047	0.912	0.076
0.100	0.000	0.100	0.000	0.100	0.000	0.995	0.005
0.999	0.000	0.999	0.000	0.999	0.000	0.076	0.000

[a]Standard uncertainties u are u(T)=0.1 K, and u(x)=0.001

container and weighed. The remaining aqueous phase was also collected and weighed. After separation, samples of both phases were transparent and were carefully analyzed to determine their compositions.

3. Analysis

The aqueous and organic phases were weighed and analyzed to find the composition of the components. For the complete determination of the LLE data, the following method was used. The concentrations of the acid (BA) in both phases were determined by potentiometric NaOH titration in the presence of phenolphthalein as an indicator. The water content of the organic phase was measured by the Karl-Fisher method [23] using Metrohm-870 KF Titrino plus Karl-Fisher titrator. The solvent content in the aqueous phase was determined using refractive index measurements [24]. The refractive indices of both phases at equilibrium were measured and used for building standard curves.

Therefore, knowing the two known variables in the organic phase (i.e., mass of water and mass of the acid in the organic phase) one can evaluate the mole fraction of the third component (*iso*-butyl acetate) using the following equation:

$$\sum_i x_i = 1 \tag{1}$$

Furthermore, concentration analyses of BA and *iso*-butyl acetate were also performed for the aqueous phase using acid-base titration and refractive index measurements, respectively. Therefore, knowing mass of the solvent and mass of the acid, one can calculate the mole fraction of water in the aqueous phase using Eq. (1).

The calculated composition of tie-line samples was checked for data consistency using mass balance equations and the experimental information on the binodal curves. In other words, knowing the mass of the three components in the initial mixtures, the mass of separated phases, and the three known variables, one can estimate the other unknown variables. It should be noted that the composition of the feeds was on the straight lines of the organic and the aqueous phases. Thus, using mass balance equations, the water content in the aqueous phase and the amount of the solvent in both phases were determined.

All weighing was done with an analytical balance accurate to within $\pm 1 \times 10^{-4}$ g. The uncertainty in the mole fractions was estimated as ±0.001.

RESULTS AND DISCUSSION

1. Experimental LLE Data

The experimental LLE data for the (water+BA+*iso*-butyl acetate) ternary system were measured at T=(298.2, 303.2, 308.2 and 313.2) K. The experimental values for the ternary system at each temperature are listed in Table 3. The LLE diagrams at various temperatures for the investigated system are plotted in Figs. 1-4. Because (BA+water) and (BA+*iso*-butyl acetate) are two liquid pairs that are completely miscible and the only liquid pair (water+*iso*-butyl acetate) is partially miscible, this ternary system behaves as type-1 LLE.

To indicate the ability of the solvent in the recovery of the acid, separation factors (S) were calculated from the experimental data. The separation factor is defined as,

$$S = \frac{\text{distribution coefficient of the aicd } (D_2)}{\text{distribution coefficient of water } (D_1)} \tag{2a}$$
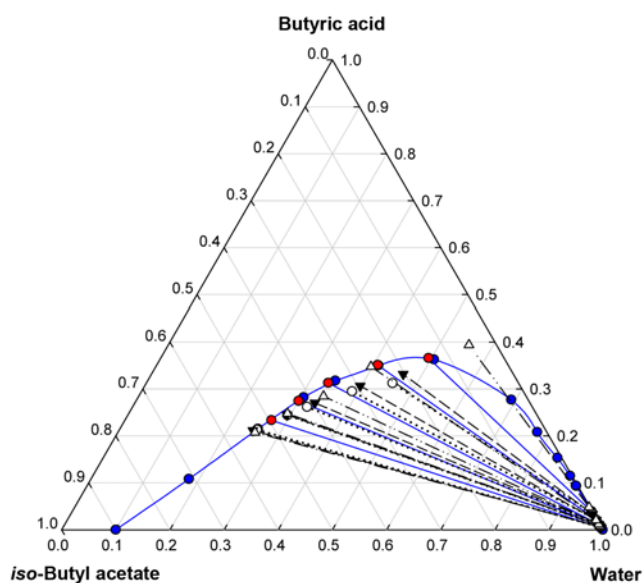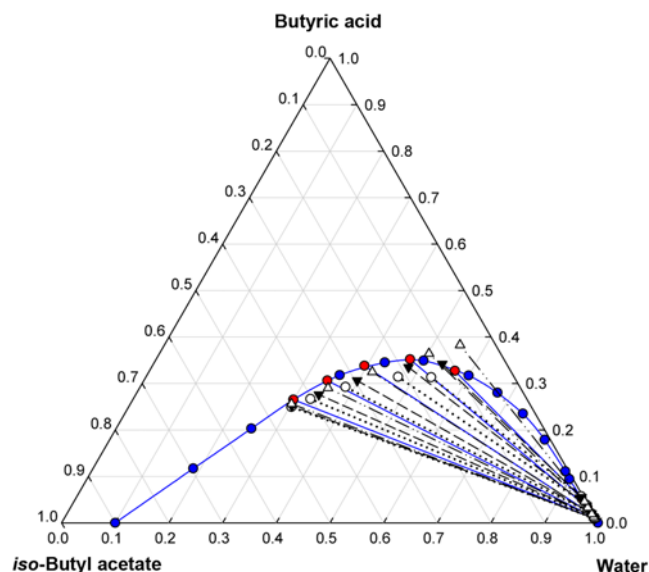
$$S = \frac{x_{23}/x_{21}}{x_{13}/x_{11}} \tag{2b}$$

x_{13} and x_{23} are the mole fractions of water and BA in the organic-rich phase, respectively. x_{11} and x_{21} are the mole fractions of water and BA in the aqueous phase, respectively.

The distribution coefficients and separation factors for the ternary system are also given in Table 3. The variation of experimental separation factor as a function of the mole fraction of the solute in aqueous phase for the ternary system is shown in Fig. 5. As can be seen, the temperature effect on the separation factor is notice-

Table 3. Experimental tie-line values in mole fraction for (water+BA+*iso*-butyl acetate) along with separation factor (S) and distribution coefficient of the acid (D_2) and water (D_1) at different temperatures[a]

Aqueous phase mole fraction			Organic phase mole fraction			D_2	D_1	S
x_{11} (water)	x_{21} (BA)	x_{31} (i-BA)	x_{13} (water)	x_{23} (BA)	x_{33} (i-BA)			
			T=298.2 K					
0.992	0.005	0.003	0.271	0.233	0.496	5×10^1	0.273	2×10^2
0.990	0.008	0.002	0.300	0.274	0.426	3×10^1	0.303	1×10^2
0.986	0.011	0.003	0.336	0.313	0.351	28	0.341	84
0.977	0.021	0.002	0.408	0.351	0.241	17	0.418	40
0.959	0.039	0.002	0.495	0.366	0.139	9.4	0.516	18
			T=303.2 K					
0.989	0.009	0.002	0.300	0.265	0.435	3×10^1	0.303	1×10^2
0.985	0.012	0.003	0.342	0.306	0.352	26	0.347	73
0.978	0.019	0.003	0.395	0.338	0.267	18	0.404	44
0.963	0.033	0.004	0.473	0.352	0.175	11	0.491	22
0.943	0.053	0.004	0.569	0.327	0.104	6.2	0.603	10
			T=308.2 K					
0.991	0.007	0.002	0.247	0.175	0.578	3×10^1	0.249	1×10^2
0.988	0.010	0.002	0.288	0.236	0.476	24	0.291	81
0.985	0.012	0.003	0.335	0.289	0.376	24	0.340	71
0.982	0.014	0.004	0.384	0.318	0.298	23	0.391	58
0.979	0.017	0.004	0.447	0.335	0.218	20	0.457	43
			T=313.2 K					
0.989	0.010	0.001	0.291	0.220	0.489	22	0.294	75
0.985	0.013	0.002	0.340	0.270	0.390	21	0.345	60
0.981	0.017	0.002	0.383	0.294	0.323	17	0.390	44
0.977	0.021	0.002	0.422	0.309	0.269	15	0.432	34
0.972	0.025	0.003	0.455	0.317	0.228	13	0.468	27

[a]Standard uncertainties u are u(T)=0.1 K, and u(x)=0.001

Fig. 1. Ternary phase diagram for LLE of [water (1)+BA (2)+*iso*-butyl acetate (3)] at T=298.2 K; (●) Experimental binodal curve and (●) tie-lines, (○) UNIQUAC calculated points, (▼) NRTL calculated points (α=0.3), (△) NRTL calculated points (regressed α).
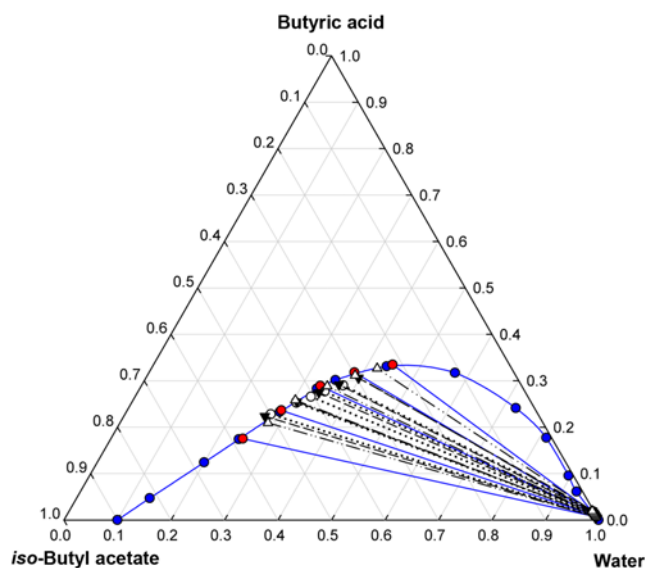

Fig. 2. Ternary phase diagram for LLE of [water (1)+BA (2)+*iso*-butyl acetate (3)] at T=303.2 K; (●) Experimental binodal curve and (●) tie-lines, (○) UNIQUAC calculated points, (▼) NRTL calculated points (α=0.3), (△) NRTL calculated points (regressed α).

Fig. 3. Ternary phase diagram for LLE of [water (1)+BA (2)+iso-butyl acetate (3)] at T=308.2 K; (●) Experimental binodal curve and (●) tie-lines, (○) UNIQUAC calculated points, (▼) NRTL calculated points (α=0.3), (△) NRTL calculated points (regressed α).
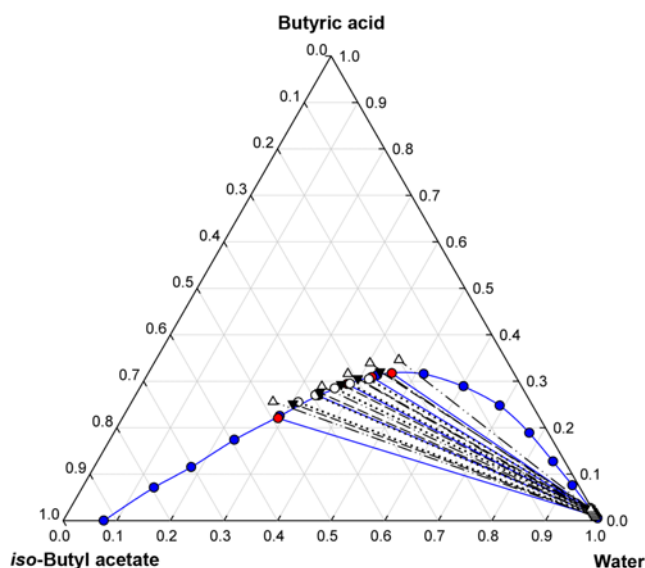

Fig. 4. Ternary phase diagram for LLE of [water (1)+BA (2)+iso-butyl acetate (3)] at T=313.2 K; (●) Experimental binodal curve and (●) tie-lines, (○) UNIQUAC calculated points, (▼) NRTL calculated points (α=0.3), (△) NRTL calculated points (regressed α).

able. The average relative uncertainties for water distribution coefficient, acid distribution coefficient and separation factor are 0.003, 0.08 and 0.08, respectively.

For the investigated system, the experimental results confirm that the organic solvent has high separation factor, indicating the ability of the solvent to extract the acid from water. However, iso-butyl acetate has a larger separation factor at 298.2 K (separation factors varying between 75 and 200), meaning that it is a more appropriate solvent for the extraction of BA from water at T=298.2 K.
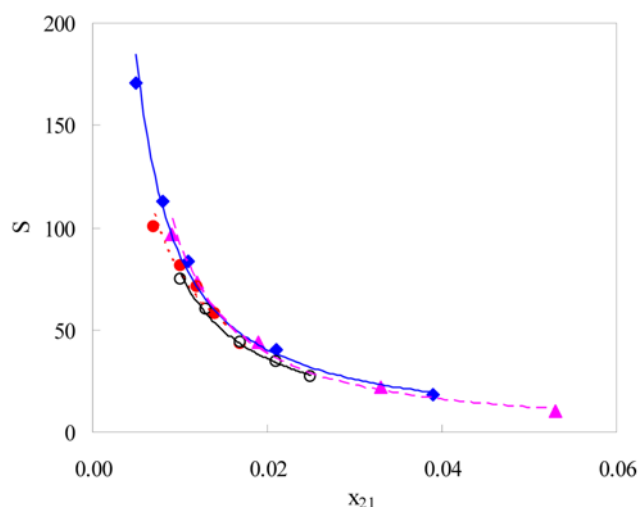
Fig. 5. Plot of the separation factor (S) of BA as a function of mole fraction of the acid in the aqueous phase; (◆) 298.2 K, (▲) 303.2 K, (●) 308.2 K and (○) 313.2 K.

Table 4. The UNIQUAC structural parameters (r and q) for pure components

Components	r	q
Water	0.92	1.40
Butyric acid	3.55	3.15
iso-Butyl acetate	4.83	4.19

2. Correlation Models

The experimental LLE data were correlated using the universal quasi-chemical (UNIQUAC) method of Abrams and Prausnitz [20] and the nonrandom two-liquid (NRTL) model of Renon and Prausnitz [21]. The UNIQUAC structural parameters r and q used for this ternary system (Table 4) have been computed from the number of molecular groups and the individual values of the van der Waals volume and area of the molecule [25,26].

In this work, the value of the non-randomness α was fixed at 0.3. Moreover, in the NRTL equation α was regressed (correlated α) and used for the investigated system. The optimum values for the regressed α (i.e. α_{12}, α_{13}, and α_{23}) were 0.46, 0.40, and 1.00, respectively.

The correlated results together with the experimental values for the ternary system of (water+BA+iso-butyl acetate) were plotted and are shown in Figs. 1-4. The experimental LLE results were also used to determine the optimum UNIQUAC and NRTL global binary interaction parameters between each pair of components (see Table 5). The detailed description of the meaning of parameters and equations is widely defined in the current literature [27]. The calculated data for the ternary system at each temperature are listed in Table 6.

The quality of the correlation is measured by the root-mean square deviation (rmsd). The rmsd value was calculated from the difference between the experimental and calculated mole fractions according to the following equation:

$$\% \text{ rmsd} = \sqrt{\frac{\sum_{k=1}^{n}\sum_{j=1}^{2}\sum_{i=1}^{3}(x_i - \hat{x}_{ijk})^2}{6n}} \times 100 \tag{3}$$

Table 5. The rmsd values for correlated results from the UNIQUAC and NRTL model and the corresponding binary interaction parameters (a_{ij} and a_{ji}) for the ternary system

Model	α_{ij}	i-j	a_{ij}	a_{ji}	rmsd %
	-	1-2	0.0865	−0.4367	
UNIQUAC	-	1-3	−0.0804	−3.4806	1.58
	-	2-3	1.5196	−50.0000	
	0.3	1-2	2.7847	−0.1306	
NRTL	0.3	1-3	4.9007	2.5703	1.33
	0.3	2-3	−2.6482	5.0079	
	0.45	1-2	2.8219	1.1673	
NRTL (Regressed α)	0.40	1-3	5.6081	1.8278	1.27
	1.00	2-3	−0.6736	5.7291	

Table 6. Calculated UNIQUAC and NRTL tie-line values in mole fraction for (water+BA+*iso*-butyl acetate) at different temperatures

Aqueous phase mole fraction						Organic phase mole fraction					
x_{11} (Water)			x_{21} (BA)			x_{13} (Water)			x_{23} (BA)		
UNIQ.	NRTL α=0.3	NRTL reg. α	UNIQ.	NRTL α=0.3	NRTL reg. α	UNIQ.	NRTL α=0.3	NRTL reg. α	UNIQ.	NRTL α=0.3	NRTL reg. α
						T=298.2 K					
0.992	0.992	0.992	0.005	0.005	0.006	0.254	0.247	0.253	0.213	0.212	0.208
0.989	0.989	0.990	0.008	0.008	0.008	0.295	0.293	0.293	0.244	0.245	0.248
0.986	0.986	0.986	0.011	0.012	0.012	0.323	0.332	0.341	0.262	0.271	0.285
0.976	0.977	0.976	0.021	0.020	0.021	0.389	0.398	0.397	0.295	0.307	0.348
0.962	0.964	0.949	0.035	0.033	0.048	0.455	0.465	0.555	0.312	0.332	0.394
						T=303.2 K					
0.988	0.989	0.989	0.009	0.009	0.009	0.303	0.303	0.300	0.249	0.252	0.256
0.985	0.985	0.985	0.012	0.013	0.013	0.330	0.341	0.351	0.266	0.276	0.290
0.977	0.977	0.978	0.020	0.020	0.019	0.383	0.397	0.416	0.292	0.306	0.325
0.958	0.960	0.964	0.039	0.037	0.033	0.470	0.479	0.502	0.314	0.335	0.364
0.939	0.941	0.951	0.057	0.055	0.046	0.532	0.538	0.551	0.313	0.342	0.383
						T=308.2 K					
0.991	0.992	0.992	0.006	0.006	0.006	0.273	0.263	0.277	0.228	0.224	0.209
0.988	0.988	0.989	0.010	0.009	0.009	0.308	0.308	0.303	0.253	0.255	0.258
0.986	0.985	0.985	0.012	0.013	0.012	0.329	0.339	0.348	0.266	0.275	0.288
0.983	0.982	0.981	0.015	0.016	0.016	0.349	0.368	0.388	0.277	0.291	0.312
0.978	0.977	0.978	0.019	0.020	0.020	0.377	0.397	0.422	0.290	0.306	0.327
						T=313.2 K					
0.988	0.989	0.990	0.010	0.009	0.008	0.311	0.303	0.263	0.255	0.252	0.256
0.985	0.985	0.985	0.013	0.013	0.012	0.336	0.341	0.338	0.270	0.276	0.289
0.980	0.981	0.982	0.017	0.017	0.016	0.365	0.372	0.373	0.284	0.294	0.316
0.977	0.977	0.977	0.021	0.020	0.020	0.388	0.397	0.403	0.294	0.306	0.339
0.971	0.971	0.972	0.027	0.026	0.024	0.418	0.431	0.453	0.305	0.321	0.346

where n is the number of tie-lines, x indicates the experimental mole fraction, $\hat{x}$ is the calculated mole fraction, and the subscript i indexes components, j indexes phases and k=1, 2, ..., n (tie-lines). The rmsd values for the data obtained through UNIQUAC and NRTL correlations are also listed in Table 5.

3. Consistency of Tie-line Data

The reliability of experimental tie-line data was determined by the Othmer-Tobias [28] and the Hand [29] correlation equations, for the investigated ternary system:
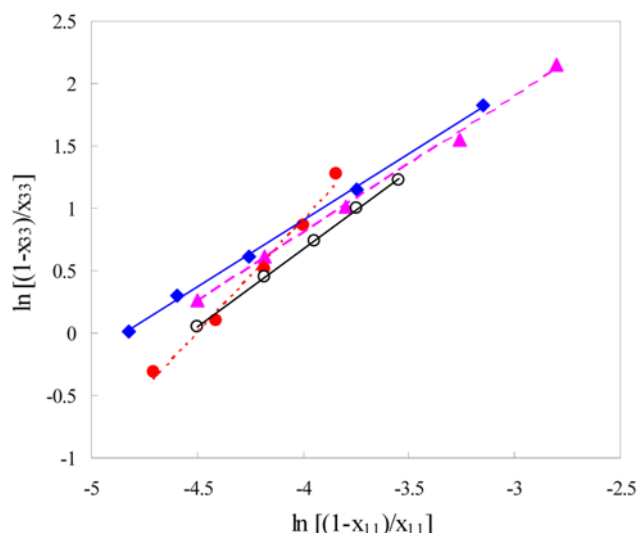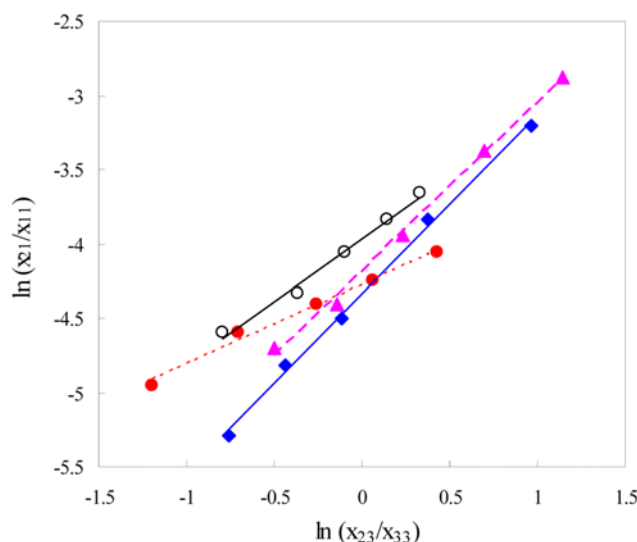
$$\ln\left(\frac{1-x_{33}}{x_{33}}\right) = A + B\ln\left(\frac{1-x_{11}}{x_{11}}\right)$$

$$\ln\left(\frac{x_{21}}{x_{11}}\right) = A' + B'\ln\left(\frac{x_{23}}{x_{33}}\right) \tag{5}$$

where the components definition are same as those previously defined. A, B, A′, and B′ are the parameters of the Othmer-Tobias correlation and the Hand correlation, respectively. The parameters of the Othmer-Tobias and the Hand correlations are listed in Table 7.

Table 7. Othmer-Tobias and Hand equations constants for the ternary system of (water+BA+*iso*-butyl acetate)

T/K	Othmer-Tobias correlation			Hand correlation		
	A	B	R²	A′	B′	R²
298.2	5.1768	1.0687	0.9990	−4.3372	1.2040	0.9971
303.2	5.1855	1.0970	0.9972	−4.1886	1.1371	0.9967
308.2	8.2048	1.8262	0.9869	−4.2729	0.5365	0.9894
313.2	5.6440	1.2432	0.9993	−3.9624	0.8398	0.9900

Fig. 6. Othmer-Tobias plot of the (water+BA+*iso*-butyl acetate) ternary system at different temperatures; (◆) 298.2 K, (▲) 303.2 K, (●) 308.2 K and (○) 313.2 K.

Fig. 7. Hand plot of the (water+BA+*iso*-butyl acetate) ternary system at different temperatures; (◆) 298.2 K, (▲) 303.2 K, (●) 308.2 K and (○) 313.2 K.

For this system, the Othmer-Tobias and the Hand plots are also shown in Figs. 6 and 7, respectively. The correlation factor (R^2) being approximately unity and the linearity of the plots indicate the degree of regularity of the measured tie-line data.

CONCLUSIONS

The LLE data for the ternary system of (water+butyric acid+*iso*-butyl acetate) were experimentally obtained at T=(298.2, 303.2, 308.2 and 313.2) K. The ternary system exhibits type-1 behavior of the LLE. The UNIQUAC and NRTL (α=0.3 and regressed α) models were satisfactorily used to correlate the experimental data and to calculate the phase compositions of the mixture studied. The corresponding optimized binary interaction parameters were also calculated. Both the models give good results for the investigated system.

The separation factors and distribution coefficients for the organic solvent used in this work were calculated. The experimental data indicated that *iso*-butyl acetate has relatively high separation factors and distribution coefficients, which shows the ability of the solvent to extract the acid from water.

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