

Liquid-liquid equilibrium for the quaternary system of *o*-xylene(1)+water(2)+butyric acid(3)+ethyl acetate(4) at 25 °C and atmospheric pressure

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Abstract—The liquid-liquid equilibrium for the quaternary system of *o*-xylene(1)+water(2)+butyric acid(3)+ethyl acetate(4) was measured at 25 °C and atmospheric pressure. Binodal curves, tie-lines, distribution, and selectivity for the quaternary system have been determined in order to investigate the effect of binary solvents, *o*-xylene and ethyl acetate, on extracting butyric acid from aqueous solution. In addition, these experimental tie line data were also compared with the values predicted by the UNIFAC model. It is found that UNIFAC group interaction parameters used for LLE could not provide a good prediction.

Key words: Liquid-Liquid Equilibrium, Quaternary System, Ternary System, UNIFAC, Butyric Acid

INTRODUCTION

The extraction of organic acids from aqueous media and other broth has been investigated in connection with the development of biotechnology in the last decade [1]. The efficient separation of organic acids from aqueous media and other broth is of economic importance in the chemical industry, for example, in the fermentation industry. The economics of the process, however, depends on the development of an effective recovery method for the organic acids from the broth [2]. Solvent extraction processes as a promising recovery technique have been proposed as an alternative to the conventional precipitation process [3], and many solvents have been tried to improve such recovery [4-8]. The extraction technique has the advantage of continuously removing the acid from the fermented broth and keeping the acids concentration in the broth to a low level. This is effective in suppressing product inhibition and increasing reactor efficiency [9].

Butyric acid is a special chemical with many applications in the chemical, food, and pharmaceutical industries. It is used in the form of pure acid to enhance butter-like notes in food flavors [10]. Esters of butyric acid are used as additives for increasing fruit fragrance and as aromatic compounds for production of perfumes [10-13].

The purpose of this work is to get LLE data of *o*-xylene(1)+water(2)+butyric acid(3)+ethyl acetate(4). The LLE data for the quaternary system were measured at 25 °C and atmospheric pressure. The effect of the mixed solvents to separate butyric acid from aqueous solution was investigated. The system studied was a type C quaternary system, and the ternary systems relevant to the quaternary mixtures at 25 °C have been reported for the type 1 systems of *o*-xylene(1)+water(2)+butyric acid(3) and water(2)+butyric acid(3)+ethyl acetate(4), and the type 2 system of *o*-xylene(1)+water(2)+ethyl acetate(4). The LLE data have also been predicted with the UNIFAC method [14-16], and the calculated data are compared with the experimental data at 25 °C.

EXPERIMENTAL SECTION

1. Materials

The *o*-xylene, ethyl acetate, and butyric acid used in this work were purchased from Aldrich and were of 99.5% purity, and ordinary bidistilled water was used. The purity of each component was checked by gas chromatography. The GC analysis did not show any appreciable peaks of impurities for all the components and determined >99.5 mass percent purities for *o*-xylene, ethyl acetate, and butyric acid. They were used without further purification.

2. Apparatus

The apparatus was consisted of microburet (0.1 mL minimum scale), sample bottle (about 200 mL), clear water jacket, and constant temperature water from water bath circulated as shown in Fig. 1.

3. Quaternary Equilibrium Data Determination

The preliminary experiment of buty acetate+water+acetic acid system was compared with reference [17] under the same condi-

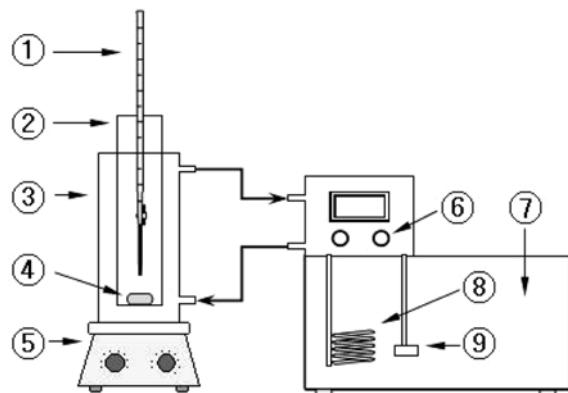


Fig. 1. Schematic diagram of experimental apparatus.

- | | |
|-------------------------|--------------------------|
| ① Micro buret | ⑥ Temperature controller |
| ② Sample bottle | ⑦ Water bath |
| ③ Water jacket | ⑧ Heater |
| ④ Magnetic stirring bar | ⑨ Circulation pump |
| ⑤ Magnetic stirrer | |

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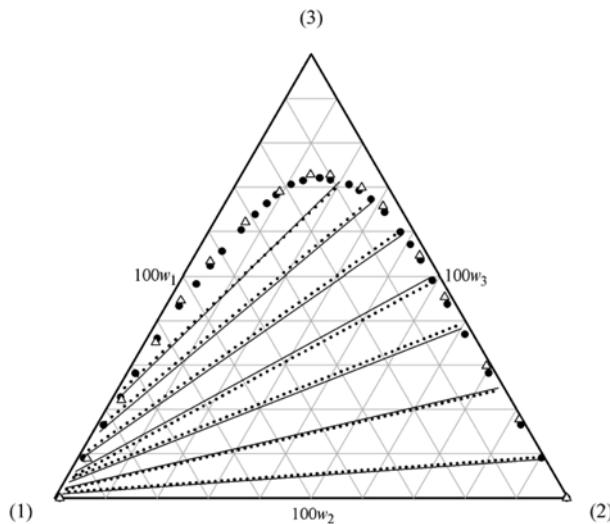


Fig. 2. Liquid-liquid equilibrium data for butyl acetate(1)+water(2)+acetic acid(3) system at 25 °C compared to the reference [17]; ●, Binodal curve Ref. Data, △, Binodal curve Exp. Data, —, Tie-line Ref. Data, ···, Tie-line Exp. Data.

tions in order to ascertain the reliability of the apparatus. The binodal curve and tie-line of preliminary experiments agreed with those of the reference and are shown in Fig. 2.

According to Prausnitz's classification [18], the quaternary system in this study was formed type C, which contains two ternary LLE mixtures of type 1 having a plait point, and one ternary LLE mixture of type 2 having two pairs of partially miscible compounds in Treybal's classification [19]. The study of the quaternary solubility surface was carried out for a system made up of water, butyric acid, and mixtures of *o*-xylene and ethyl acetate in the ratios of 100/0, 75/25, 50/50, 25/75, and 0/100 (pseudoternary systems) in mass percent with the following analytical method. Fig. 3 shows the method schematically, where R is the ratio of the solvents.

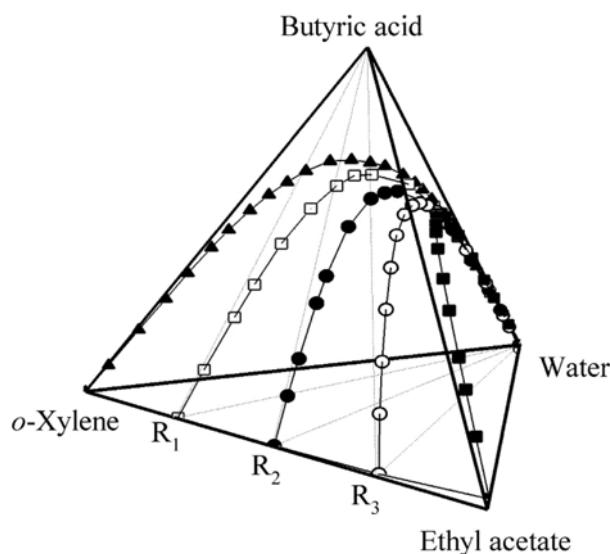


Fig. 3. Phase equilibrium of *o*-xylene(1)+water(2)+butyric acid(3)+ethyl acetate(4). R₁, R₂, and R₃ denote quaternary sectional planes.

The mutual solubility (binodal curve) in *o*-xylene(1)+water(2)+butyric acid(3)+ethyl acetate(4) was determined at 25 °C and atmospheric pressure by the cloud-point method as described by Othmer et al. [20]. For the determination of the mutual solubility data, an accurately known mass of a homogeneous mixture of two compounds was placed in a thermostated glass-stoppered bottle, one of the components was titrated into the bottle from a microburet with an uncertainty of ±0.1 mL until the solution became turbid, and the amount titrated was recorded. The bottle was kept in a constant-temperature bath. The temperature of the bath was maintained at (25±0.1) °C. The end point was confirmed by observing the transition from a homogenous to a heterogeneous mixture. The procedure was repeated to obtain the phase-boundary curve.

For the tie line measurement, an equilibrium cell was immersed in a thermostat controlled at the desired temperature (±0.1 °C). The pure components were added, and the mixture was stirred for at least 1 hr with a magnetic stirrer. The two-phase mixture was allowed to settle for at least 24 hr. Samples were taken by a syringe from the upper and lower mixtures.

The mixtures of aqueous and organic phase were analyzed on an HP 5890 series II gas chromatograph, equipped with a thermal conductivity detector (TCD). A 6 ft×1/8 in column packed with Porapack Q was used. The injector and detector temperatures were maintained at 200 °C and 230 °C, respectively. The column temperature was programmed for an initial temperature of 160 °C and a final temperature of 200 °C. The heating rate was 4 °C/min, and the flow rate of the helium carrier gas was 30 mL/min. The uncertainty of the mass fraction measurements for the overall composition determination was ±0.002.

The reliability of measured tie-line compositions was ascertained by making Othmer-Tobias and Hand plots for the quaternary system [21,22].

The plots are presented in Figs. 4 and 5. The linear correlation coefficients for the system in ratios of 75/25, 50/50, and 25/75 in

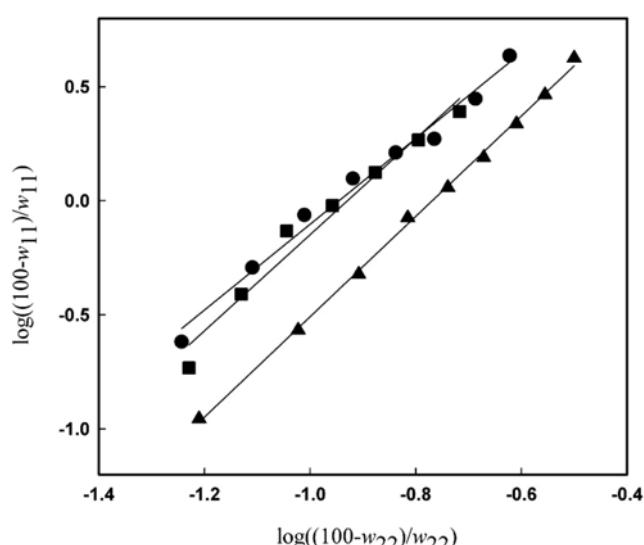


Fig. 4. Othmer-Tobias plots of *o*-xylene(1)+water(2)+butyric acid(3)+ethyl acetate(4) at 25 °C: ▲, *o*-xylene/ethyl acetate (75/25); ●, *o*-xylene/ethyl acetate (50/50); ■, *o*-xylene/ethyl acetate (25/75).

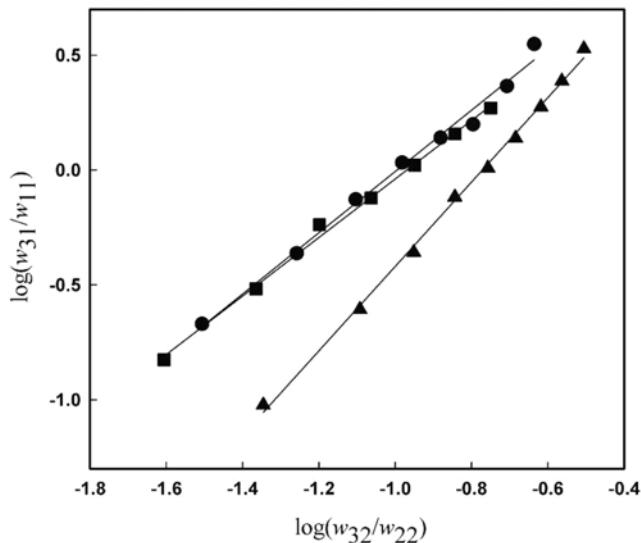


Fig. 5. Hand plots of *o*-xylene(1)+water(2)+butyric acid(3)+ethyl acetate(4) at 25 °C: ▲, *o*-xylene/ethyl acetate (75/25); ●, *o*-xylene/ethyl acetate (50/50); ■, *o*-xylene/ethyl acetate (25/75).

mass percent as mixed solvents are 0.9992, 0.9927, and 0.9849 in Othmer-Tobias and 0.9988, 0.9922, and 0.9981 in Hand, respectively. The linearity of the plots (a correlation factor close to 1) indicates the degree of consistency of the related data.

RESULTS AND DISCUSSION

The liquid-liquid equilibrium for the quaternary system of *o*-xylene(1)+water(2)+butyric acid(3)+ethyl acetate(4) was measured at 25 °C and atmospheric pressure. The quaternary experimental data exhibited type C quaternary LLE behavior as shown in Fig. 3 and were composed of two ternary LLE mixtures of type 1 having a

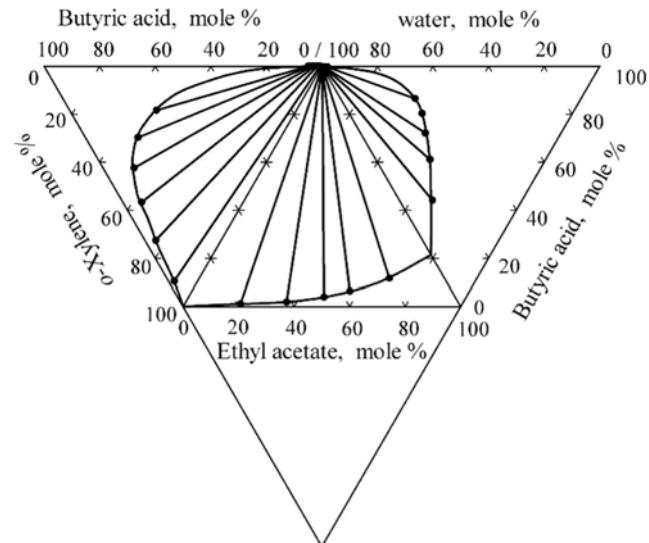


Fig. 6. Binodal curves and tie lines of three ternary mixtures making up *o*-xylene(1)+water(2)+butyric acid(3), water(2)+butyric acid(3)+ethyl acetate(4), and *o*-xylene(1)+water(2)+ethyl acetate(4) at 25 °C, respectively.

plait point (*o*-xylene(1)+water(2)+butyric acid(3) and water(2)+butyric acid(3)+ethyl acetate(4)) and one ternary LLE mixture of type 2 having two immiscible binary pairs (*o*-xylene(1)+water(2)+ethyl acetate(4)). Fig. 6 shows the experimental results for the ternary mixtures constituting the quaternary mixtures for *o*-xylene(1)+water(2)+butyric acid(3)+ethyl acetate(4).

The mutual solubility data and the equilibrium tie line compositions measured at 25 °C for the ternary mixtures of *o*-xylene(1)+water(2)+butyric acid(3), water(2)+butyric acid(3)+ethyl acetate(4), and *o*-xylene(1)+water(2)+ethyl acetate(4) system are shown in Tables 1 and 2. Quaternary solubility data concerning the planes R₁(75/25), R₂(50/50), and R₃(25/75) are given in Table 3, and the

Table 1. Binodal curve data for the ternary systems; *o*-xylene(1)+water(2)+butyric acid(3), water(2)+butyric acid(3)+ethyl acetate(4), and *o*-xylene(1)+water(2)+ethyl acetate(4)

T/K	<i>o</i> -xylene(1)+water(2)+butyric acid(3)			water(2)+butyric acid(3)+ethyl acetate(4)			<i>o</i> -xylene(1)+water(2)+ethyl acetate(4)		
	100x ₁	100x ₂	100x ₃	100x ₁	100x ₂	100x ₃	100x ₁	100x ₂	100x ₃
298.15	99.88	0.12	0.00	5.28	0.00	94.72	99.83	0.00	0.17
	88.21	2.16	9.62	10.60	21.23	68.17	93.37	0.38	6.25
	74.75	3.52	21.73	12.24	27.57	60.19	87.98	0.67	11.35
	63.26	5.62	31.12	15.43	36.83	47.74	80.68	0.97	18.35
	54.91	6.49	38.61	17.97	43.11	38.92	74.84	1.24	23.92
	45.89	9.42	44.69	24.08	51.75	24.17	65.79	1.67	32.54
	39.71	11.92	48.37	29.73	53.63	16.64	57.74	2.31	39.95
	26.44	21.47	52.08	36.88	52.41	10.71	47.86	3.48	48.66
	16.35	33.67	49.98	41.02	50.46	8.52	39.63	5.51	54.86
	11.25	42.28	46.47	52.06	42.73	5.21	31.82	7.97	60.21
	7.68	51.26	41.06	62.81	33.95	3.24	25.07	9.79	65.17
	3.74	63.86	32.40	71.88	25.71	2.41	17.58	12.55	69.87
	1.23	76.49	22.28	79.18	18.90	1.92	12.76	15.13	72.11
	0.35	86.29	13.36	90.62	8.45	0.93	7.22	17.99	74.79
	0.01	99.99	0.00	97.65	0.00	2.35	0.00	21.42	78.58

tie line results of the quaternary mixtures for *o*-xylene(1)+water(2)+butyric acid(3)+ethyl acetate(4) system at 25 °C are given in Table

Table 2. Liquid-liquid equilibrium compositions for the ternary systems; *o*-xylene(1)+water(2)+butyric acid(3), water(2)+butyric acid(3)+ethyl acetate(4), and *o*-xylene(1)+water(2)+ethyl acetate(4)

		<i>o</i> -xylene(1)+water(2)+butyric acid(3)					
T/K		Organic phase			Aqueous phase		
		100x ₁	100x ₂	100x ₃	100x ₁	100x ₂	100x ₃
298.15	89.48	2.11	8.41	0.01	99.34	0.65	
	72.62	3.88	23.50	0.01	98.27	1.72	
	56.64	6.80	36.56	0.01	97.35	2.64	
	42.38	11.22	46.40	0.01	96.50	3.49	
	29.70	18.96	51.34	0.01	95.73	4.26	
	18.33	31.31	50.36	0.01	95.11	4.88	
water(2)+butyric acid(3)+ethyl acetate(4)							
T/K		Organic phase			Aqueous phase		
		100x ₁	100x ₂	100x ₃	100x ₁	100x ₂	100x ₃
298.15	33.22	11.86	55.92	99.24	0.27	0.49	
	41.64	19.48	38.88	99.03	0.56	0.41	
	48.82	23.33	27.85	98.76	0.90	0.34	
	54.10	26.28	19.62	98.42	1.31	0.27	
	59.76	26.82	13.42	97.97	1.82	0.21	
<i>o</i> -xylene(1)+water(2)+ethyl acetate(4)							
T/K		Organic phase			Aqueous phase		
		100x ₁	100x ₂	100x ₃	100x ₁	100x ₂	100x ₃
298.15	78.77	1.10	20.13	0.01	99.86	0.13	
	61.75	1.91	36.34	0.00	99.79	0.21	
	47.28	3.96	48.76	0.00	99.74	0.26	
	36.76	6.35	56.89	0.00	99.69	0.31	
	0.00	99.56	0.44	19.87	11.65	68.48	

Table 3. Binodal curve data for the *o*-xylene(1)+water(2)+butyric acid(3)+ethyl acetate(4) quaternary system

T/K	w(1)/w(4)=75/25				w(1)/w(4)=50/50				w(1)/w(4)=25/75			
	100w ₁	100w ₂	100w ₃	100w ₄	100w ₁	100w ₂	100w ₃	100w ₄	100w ₁	100w ₂	100w ₃	100w ₄
298.15	74.57	0.58	0.00	24.86	49.70	0.60	0.00	49.70	24.57	1.74	0.00	73.70
	64.11	0.98	13.54	21.37	43.08	1.21	12.63	43.08	20.86	2.94	13.61	62.59
	52.53	1.95	28.02	17.51	37.96	2.23	21.85	37.96	17.48	4.97	25.11	52.44
	45.22	2.84	36.87	15.07	30.26	3.95	35.54	30.26	13.92	7.86	36.48	41.75
	35.82	4.48	47.76	11.94	26.32	5.26	42.11	26.32	10.84	11.27	45.39	32.51
	26.62	7.73	56.78	8.87	18.46	9.35	53.73	18.46	8.17	15.21	52.12	24.50
	19.44	11.87	62.21	6.48	12.40	15.71	59.50	12.40	5.98	20.31	55.79	17.93
	15.08	15.48	64.41	5.03	9.64	20.73	59.99	9.64	3.40	30.29	56.11	10.20
	11.71	20.51	63.88	3.90	7.43	25.80	59.35	7.43	1.89	41.11	51.33	5.67
	5.63	33.23	59.26	1.88	3.13	40.77	52.98	3.13	1.28	48.65	46.24	3.83
	2.97	44.70	51.34	0.99	1.48	53.84	43.21	1.48	0.70	60.95	36.27	2.09
	1.40	57.84	40.29	0.47	0.95	65.92	32.19	0.95	0.31	74.90	23.86	0.93
	1.14	65.90	32.58	0.38	0.55	77.57	21.33	0.55	0.26	87.27	11.71	0.77
	0.60	79.02	20.18	0.20	0.31	85.74	13.65	0.31	0.21	91.91	7.25	0.63
	0.22	99.71	0.00	0.07	0.28	99.44	0.00	0.28	0.28	98.90	0.00	0.83

4. The effect of the mixing ratio on the binodal curves for the quaternary system at 25 °C is shown in Fig. 7. As shown in Fig. 7, the heterogeneous zone of ethyl acetate was increased with the addition of *o*-xylene, but mixed solvents, *o*-xylene and ethyl acetate in the ratios of 75/25, 50/50, and 25/75 were not better than the pure solvent, *o*-xylene, as an extraction solvent.

Because the most important factor in liquid-liquid extraction is the selectivity of the solvent, the solvents having a higher distribution and selectivity for the solute should be selected after considering the recovery, chemical stability, boiling and freezing points, corrosion, density, and so forth. For those properties, the most important factors are distribution and selectivity. They are shown in the following equations.

$$D = \frac{\text{butyric acid mol\% in solvent layer}}{\text{butyric acid mol\% in water layer}} = \frac{x_{31}}{x_{32}} \quad (1)$$

$$S = \frac{\text{butyric acid mol\% in solvent-free solvent layer}}{\text{butyric acid mol\% in solvent-free water layer}} = \frac{x_{31}/(x_{31}+x_{21})}{x_{32}/(x_{32}+x_{22})} \quad (2)$$

Compositions are expressed in mole fraction x_{CL} , where the first index refers to the component (2=water, 3=butyric acid) and the second index refers to the phase (1=upper layer, 2=lower layer).

Figs. 8 and 9 illustrate the effect of the distribution and selectivity for the mixtures in the ratios 75/25, 50/50, and 25/75. As shown in the result of Fig. 7, they were also not better than the pure solvent, *o*-xylene, as an extraction solvent. Perhaps this is because of the higher mutual solubility of water and ethyl acetate when compared with that for water and *o*-xylene. The results indicate that the solubility of *o*-xylene is high enough in butyric acid.

1. UNIFAC Predicted LLE Data

Because experimental data are often missing or of poor quality, group contribution methods have become increasingly valuable. The great advantage of the group contribution makes it possible to predict the compositions of a large number of systems using only a

Table 4. Calculated and experimental liquid-liquid equilibrium compositions for the *o*-xylene(1)+water(2)+butyric acid(3)+ethyl acetate(4) quaternary system at 25 °C

	Organic phase				Aqueous phase			
	100x ₁	100x ₂	100x ₃	100x ₄	100x ₁	100x ₂	100x ₃	100x ₄
x(1)/x(4)=75/25								
Exp.	60.23	7.44	9.08	23.25	0.02	98.75	0.91	0.31
Calc.	61.69	2.08	12.30	23.93	0.01	99.29	0.53	0.18
Exp.	42.17	12.68	29.24	15.91	0.02	97.54	2.23	0.21
Calc.	43.40	7.16	32.42	17.02	0.01	98.57	1.30	0.12
Exp.	24.85	24.36	40.77	10.02	0.02	96.41	3.45	0.12
Calc.	28.31	15.70	44.79	11.20	0.01	97.90	2.02	0.07
Exp.	14.95	34.18	44.98	5.89	0.02	95.23	4.69	0.07
Calc.	17.22	25.77	50.13	6.88	0.01	97.19	2.76	0.04
Exp.	7.39	49.52	40.11	2.98	0.02	93.93	6.00	0.05
Calc.	9.50	36.06	50.64	3.80	0.01	96.41	3.55	0.03
x(1)/x(4)=50/50								
Exp.	17.45	10.27	16.43	55.85	0.02	98.86	0.63	0.50
Calc.	37.99	5.36	12.10	44.54	0.01	99.35	0.36	0.28
Exp.	9.16	25.91	32.43	32.50	0.02	98.05	1.58	0.35
Calc.	25.72	13.51	30.41	30.36	0.01	98.88	0.91	0.20
Exp.	8.06	33.82	39.36	18.76	0.02	97.12	2.61	0.25
Calc.	16.85	22.13	40.94	20.07	0.01	98.35	1.51	0.14
Exp.	6.55	42.08	41.41	9.96	0.02	95.97	3.85	0.16
Calc.	10.46	30.71	46.3	12.52	0.01	97.66	2.24	0.09
Exp.	4.50	53.11	36.97	5.42	0.02	94.54	5.34	0.10
Calc.	5.93	39.18	47.76	7.13	0.01	96.81	3.12	0.06
x(1)/x(4)=25/75								
Exp.	15.63	13.72	11.62	59.03	0.01	98.81	0.50	0.68
Calc.	16.91	11.38	11.90	59.82	0.00	99.36	0.27	0.37
Exp.	10.22	33.91	24.95	30.92	0.01	98.19	1.27	0.53
Calc.	11.31	21.23	27.38	40.08	0.00	99.02	0.69	0.29
Exp.	6.47	40.77	31.01	21.75	0.02	97.36	2.24	0.38
Calc.	7.44	29.14	36.81	26.62	0.01	98.56	1.22	0.21
Exp.	3.66	51.04	32.32	12.98	0.02	96.22	3.5	0.25
Calc.	4.72	35.99	42.33	16.96	0.01	97.93	1.92	0.14
Exp.	2.35	55.26	33.9	8.49	3.00	40.55	45.21	11.23
Calc.	2.52	54.07	34.64	8.76	2.26	55.86	33.53	8.35

relatively small number of group interaction parameters. The most common group contribution method for the prediction of phase equilibrium is UNIFAC.

This model requires two group interaction parameters (a_{mn} and a_{nm}) per pair of groups and the group volume and surface areas of individual groups for the estimation of the activity coefficient γ . The UNIFAC group volume, surface area, and interaction parameters used in this work are shown in Table 5 [23].

The experimental LLE data and tie lines were compared with predictions obtained by the UNIFAC. The RMSD (Root Mean Square Deviation), deviation of the estimate by experimental and calculated compositions, was obtained from Eq. (3) for the ternary system and from Eq. (4) for the quaternary system.

$$\text{RMSD}_3 = 100 \left[\sum_{i=1}^{N-3} \sum_{j=1}^3 \sum_{k=1}^2 (X_{jk}^{\text{exp}}(i) - X_{jk}^{\text{cal}}(i))^2 / 6N \right]^{1/2} \quad (3)$$

$$\text{RMSD}_4 = 100 \left[\sum_{i=1}^{N-4} \sum_{j=1}^4 \sum_{k=1}^2 (X_{jk}^{\text{exp}}(i) - X_{jk}^{\text{cal}}(i))^2 / 8N \right]^{1/2} \quad (4)$$

where, N is the number of tie line data.

For the ternary systems, the predictions for *o*-xylene(1)+water(2)+butyric acid(3) and water(2)+butyric acid(3)+ethyl acetate(4) composed of type 1 were shown within an average RMSD of 4.35 mol%, and the prediction for *o*-xylene(1)+water(2)+ethyl acetate(4) composed of type 2 was shown within 1.77 mol%. For the quaternary system, an average RMSD for the system in ratios of 75/25, 50/50, and 25/75 in mass percent as mixed solvents are 4.07, 6.44, and 5.17 mol%, respectively.

CONCLUSION

Liquid-liquid equilibrium data were determined for the quater-

nary system of *o*-xylene(1)+water(2)+butyric acid(3)+ethyl acetate(4) at 25 °C and atmospheric pressure. The tie-line data for all

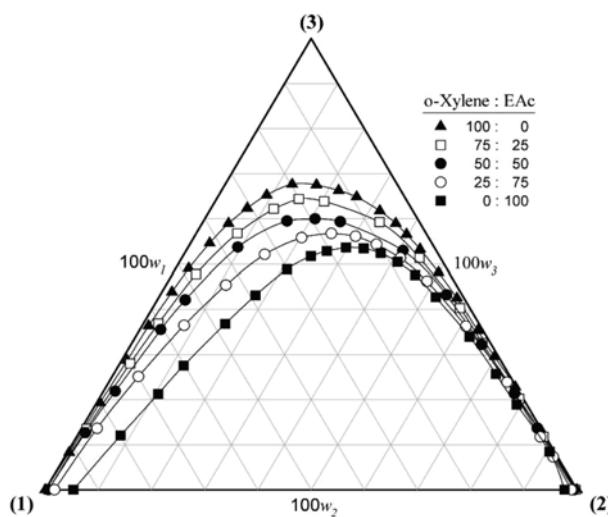


Fig. 7. The effect of the mixing ratio on binodal curves of the *o*-xylene(1)+water(2)+butyric acid(3)+ethyl acetate(4) quaternary system at 25 °C.

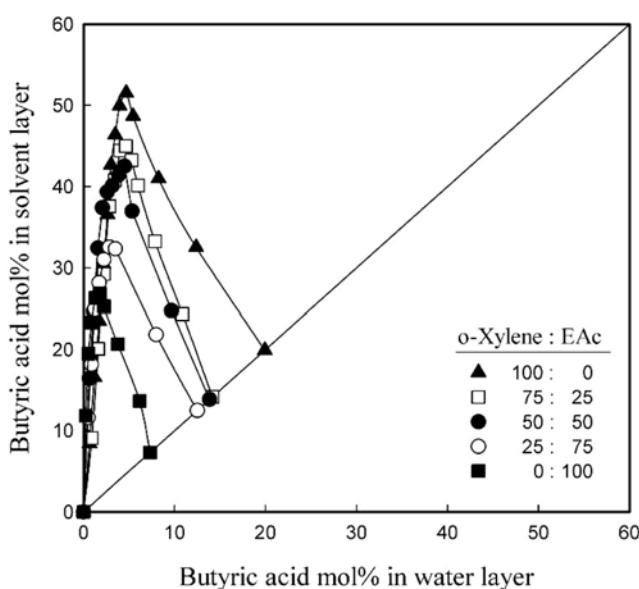


Fig. 8. Distribution of butyric acid between aqueous and organic phases at 25 °C.

systems were satisfactorily correlated by the methods of Othmer-Tobias and Hand. The usage of mixed solvents in the ratios of 75/25, 50/50, and 25/75 to improve the extraction of butyric acid would be advisable only when both solvents show some favorable properties, such as the distribution coefficient and/or selectivity, in order to take advantage of the synergistic effect of the mixture. It is apparent from the distribution and selectivity data that the separation of butyric acid from water by extraction with *o*-xylene is feasible. In addition, these experimental tie line data were also compared with the values predicted by the UNIFAC model. It was found that the UNIFAC method goodness was not satisfactory for the experimental data.

NOMENCLATURE

- a_{ij} : interaction parameter for i-j interaction [dimensionless]
- Q_k : surface area parameter, for group k [dimensionless]
- R_k : volume parameter, for group k [dimensionless]
- T : absolute temperature [K]
- x_i : mole fraction for component i [%]
- γ_i : activity coefficient for component i [dimensionless]

REFERENCES

1. M. Bilgin, S. İ. Kırbaşular, Ö. Özcan and U. Dramur, *J. Chem. Eng.*

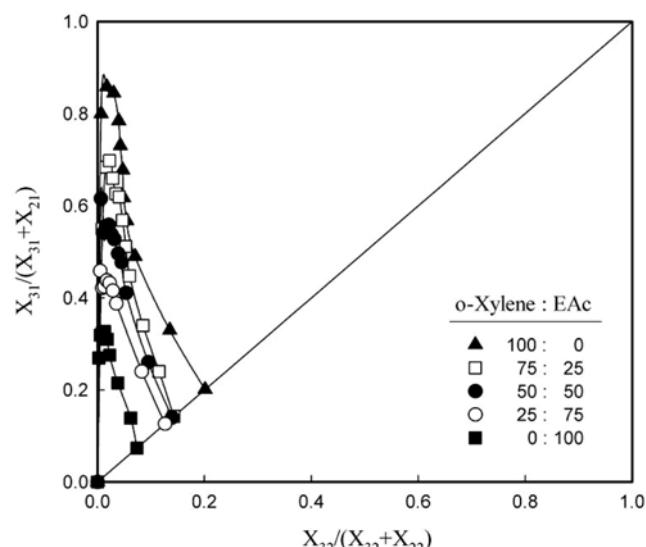


Fig. 9. Selectivity of butyric acid between aqueous and organic phases at 25 °C.

Table 5. The UNIFAC group volume (R_k), surface area (Q_k), and interaction parameters (a_{nm})

	CH ₃	CH ₂	ACH	ACCH ₃	H ₂ O	COOH	CH ₃ COO	R _k	Q _k
CH ₃	0	0	-114.8	-115.7	1,300.0	139.4	972.4	0.9011	0.8480
CH ₂	0	0	-114.8	-115.7	1,300.0	139.4	972.4	0.6744	0.5400
ACH	156.5	156.5	0	167.0	859.4	461.8	6.0	0.5313	0.4000
ACCH ₃	104.4	104.4	-146.8	0	5,695.0	339.1	5,688.0	1.2663	0.9680
H ₂ O	342.4	342.4	372.8	203.7	0	-465.7	-6.32	0.9200	1.4000
COOH	1,744.0	1,744.0	75.49	147.3	652.3	0	-117.6	1.3013	1.2240
CH ₃ COO	-320.10	-320.10	114.8	-170.0	385.9	1,417.0	0	1.9031	1.7280

- Data*, **51**(5), 1546 (2006).
2. P. K. Roychoudhury, A. Srivastava and V. Sahai, in: Fiechter (Ed.), A., *Downstream processing biosurfactants/carotenoids*, Springer, Berlin, 61 (1995).
 3. J. Hartl and R. Marr, *Separ. Sci. Technol.*, **28**, 805 (1993).
 4. U. Dramur and B. Tatli, *J. Chem. Eng. Data*, **38**, 23 (1993).
 5. J. A. Briones, J. C. Mullins and M. C. Thies, *Ind. Eng. Chem. Res.*, **33**, 151 (1994).
 6. A. Arce, A. Blanco, P. Souza and I. Vidal, *J. Chem. Eng. Data*, **40**, 225 (1995).
 7. M. A. Fahim and A. A. Al-Muhtaseb, *J. Chem. Eng. Data*, **41**, 1311 (1996).
 8. M. A. Fahim, A. A. Al-Muhtaseb and I. M. Al-Nashef, *J. Chem. Eng. Data*, **42**, 183 (1997).
 9. T. Hano, M. Matsumoto, T. Ohtake, K. Sasaki, F. Hori and Y. Kawano, *J. Chem. Eng. Japan*, **23**(6), 734 (1990).
 10. J. D. Dziedzak, *Food Technol.*, **40**, 8 (1986).
 11. F. H. J. Sharpell, In *Comprehensive biotechnology*, Pergamon Press, Moo-Young M. editor, Microbial flavors and fragrances, New York, 965 (1985).
 12. D. Vandak, J. Zigova, E. Sturdik and S. Schlosser, *Proc. Biochem.*, **32**, 245 (1997).
 13. J. Zigova, E. Sturdik, D. Vandak and S. Schlosser, *Proc. Biochem.*, **34**, 835 (1999).
 14. A. Fredenslund, *Vapor-liquid equilibria using UNIFAC*, Elsevier, Amsterdam (1977).
 15. P. A. Gupte and R. P. Danner, *Ind. Eng. Chem. Res.*, **26**, 2036 (1987).
 16. B. L. Larsen, P. Rasmussen and A. Fredenslund, *Ind. Eng. Chem. Res.*, **26**, 2274 (1987).
 17. Y. S. Jang and D. W. Park, *J. Korean Ind. Eng. Chem.*, **11**, 254 (2000).
 18. J. M. Prausnitz, T. F. Anderson, E. A. Grens, C. A. Eckert, R. Hsieh and J. P. O'Connell, *Computer calculations for multicomponent vapor-liquid and liquid-liquid equilibria*, Prentice Hall, Englewood Cliffs, NJ, 145 (1980).
 19. J. M. Sørensen and W. Arlt, *Liquid-liquid equilibrium data collection*, DECHEMA, Frankfurt am Main, Vol. V, parts 1-3 (1980).
 20. D. F. Othmer, R. E. White and E. Trueger, *Ind. Eng. Chem.*, **33**, 1240 (1941).
 21. D. F. Othmer and P. E. Tobias, *Ind. Eng. Chem.*, **34**(6), 690 (1942).
 22. D. B. Hand, *J. Phys. Chem.*, **34**, 9, 1961 (1930).
 23. T. Magnussen, P. Rasmussen and A. Fredenslund, *Ind. Eng. Process Des. Dev.*, **20**, 331 (1981).