Binary Liquid−Liquid Equilibrium (LLE) for Methyl *tert*-Amyl Ether (TAME) + Water from (288.15 to 313.15) K and Ternary LLE for Systems of TAME + C_1−C_4 Alcohols + Water at 298.15 K

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The binary liquid−liquid equilibrium data for the system methyl *tert*-amyl ether + water from (288.15 to 313.15) K and the ternary liquid−liquid equilibria for the systems methyl *tert*-amyl ether + C_1−C_4 alcohols (methanol, ethanol, 1-propanol, 2-propanol, 1-butanol, and 2-butanol) + water at 298.15 K were analytically determined at atmospheric pressure by the use of stirred and thermo-regulated cells. The experimental binary and ternary liquid−liquid equilibrium data were correlated with the NRTL and UNIQUAC activity coefficient models. Bachman−Brown, Hand, and Othmer−Tobias correlations were used to ascertain the reliability of the experimental data for each system. In addition, the distribution and selectivity of C_1−C_4 alcohols as solvents were analyzed.

Introduction

Recently, there has been considerable interest in the use of various fuel additives as antiknock agents to improve gasoline performance and to reduce air pollution. Methyl *tert*-butyl ether (MTBE) is tertiary ether and is mostly used so far because of its low vapor pressure and the availability of ethanol feedstock from renewable resources. However, MTBE has drawbacks of easily dissolving in water and contaminating groundwater. Methyl *tert*-amyl ether (TAME) is considered to be a suitable and alternative candidate for gasoline antiknock agents with diisopropyl ether (DIPE) and ethyl *tert*-butyl ether (ETBE). The phase equilibria and mixture properties for some antiknock agents were systematically studied because accurate design data are strongly related to the processing of the compounds and the application of the group contribution model.[1]

In the present work, we report binary liquid−liquid equilibrium (LLE) data for the system TAME (1) + water (2) in the temperature range of (288.15 to 313.15) K. The measurements were carried out at 5 K temperature intervals at atmospheric pressure. The six ternary liquid−liquid equilibrium phase diagrams of TAME (1) + C_1−C_4 alcohols (2) + water (3) mixtures at 298.15 K and at atmospheric pressure were also measured. The six measured systems are TAME + water with methanol, ethanol, 1-propanol, 2-propanol, 1-butanol, or 2-butanol. The experimented LLE data of the binary and ternary systems were correlated using two activity coefficient models: the NRTL and the UNIQUAC models. Bachman−Brown, Hand, and Othmer−Tobias correlations were used to check the reliability of the experimental data for each system. In addition, the distribution and selectivity of C_1−C_4 alcohols as solvents were analyzed.

Table 1. Densities, Purities, and UNIQUAC Parameters of Chemicals Used in This Work

	ρ/g·cm^3 at 298.15 K		GC analysis	UNIQUAC	
chemicals	this work	ref[a]	(wt %)	*r* value[a]	*q* value[a]
methyl *tert*-amyl ether	0.76568	0.76570	> 99.9	4.7422	4.1720
methanol	0.78664	0.78660	> 99.9	1.4311	1.4320
ethanol	0.78529	0.78500	> 99.9	2.1055	1.9720
1-propanol	0.79989	0.79970	> 99.9	2.7799	2.5120
2-propanol	0.78134	0.78130	> 99.9	2.7791	2.5080
1-butanol	0.80587	0.80600	> 99.9	3.4543	3.0520
2-butanol	0.80273	0.80260	> 99.9	3.4535	3.0480
water	0.99717	0.99700	> 99.9	0.9200	1.4000

[a] Ref 2.

Experimental Section

Materials. The commercial grade chemicals were used in this investigation without further purification. Ethanol was provided by J. T. Baker Chemical. TAME, methanol, 1-propanol, 2-propanol, 1-butanol, and 2-butanol were supplied by Aldrich. Water was distilled twice in the laboratory. All chemicals were dried by the use of molecular sieves of pore diameter (0.4 or 0.3) nm. The water content of the chemicals, determined by Karl Fischer titration (Metrohm 684 KF coulometer), was less than $7 \cdot 10^{-5}$ g·g^{-1}. The purity of the chemicals was checked by gas chromatography and a comparison of the density with values reported in the literature.[2] The measured densities and purities of the chemicals are listed in Table 1 with the literature values.

Apparatus and Procedure. We carried out LLE measurements by measuring the end points of the tie lines for all of the ternary systems. The self-designed LLE measuring system, which is similar to that of Horstmann et al.,[3] consisted of three parts: an equilibrium glass vessel jacket, a thermostat (Lauda MD 20 with DLK15 cooler) with a precision temperature measuring system (AΣA F250), and a Corning PC-320 magnetic stirrer. The equilibrium vessel temperature was controlled by a thermostat within ± 0.02 K. When the system temperature was

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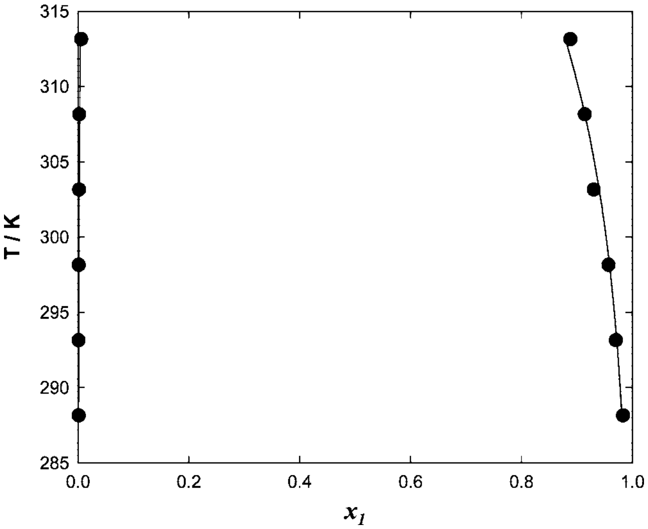

Figure 1. LLE for the binary system TAME (1) + water (2): ●, experimental value. Solid curves were calculated from the NRTL equation.

Table 2. Experimental LLE Data for the Binary System TAME (1) + Water (2)

	organic phase	aqueous phase
T/K	x_1	x_1
288.15	0.9833	0.0014
293.15	0.9706	0.0013
298.15	0.9573	0.0016
303.15	0.9306	0.0021
308.15	0.9144	0.0024
313.15	0.8884	0.0060

Table 3. Correlated G^E Model Parameters and Mean Deviation for the Binary System DBE (1) + Water (2)

	NRTL		UNIQUAC	
DBE (1) + water (2)	$ij = 12$	$ij = 21$	$ij = 12$	$ij = 21$
A_{ij} $(K)^a$	6129.14	− 18.98	3797.04	− 10.57
B_{ij}	1611.85	− 0.21	683.71	− 1.96
α	0.20			
RMSD	0.0023		0.0026	

[a] Parameters (K): $a_{ij} = A_{ij} + B_{ij}T$.

reached, the sample mixture was rigorously stirred in a closed equilibrium vessel for about 6 h. It was then allowed to settle for about 12 h at constant system temperature, allowing the mixture into equilibrium. After sufficient settling of both phases, sampling was carefully carried out from the top for the upper phase and from the bottom for the lower liquid phase without cross contamination of both phases during the sampling procedure. The analysis was carried out using gas chromatography (HP 5890N) with an HP-FFAP (polyethylene glycol TPA, (25 m)·(0.20 mm)·(0.30 μm)) capillary column and a thermal conductivity detector. We estimate the uncertainty of the calculated mole fractions to be less than ca. $\pm 1 \cdot 10^{-3}$. The procedure is described in detail elsewhere.[4]

Results and Discussion

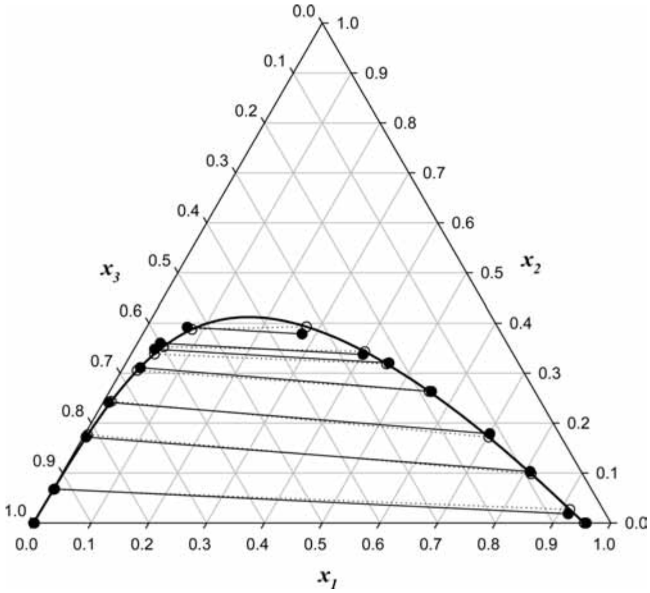

Figure 2. LLE for the ternary system TAME (1) + methanol (2) + water (3) at 298.15 K: ●, experimental value; ○, calculated value. Dashed lines were calculated from the UNIQUAC equation.

Table 4. Experimental LLE Data of the Ternary Systems TAME (1) + C_1−C_4 Alcohols (2) + Water (3) at 298.15 K

systems	organic phase		aqueous phase	
	x_1	x_2	x_1	x_2
TAME (1) + methanol (2) + water (3)	0.9169	0.0185	0.0028	0.0680
	0.8086	0.1030	0.0052	0.1722
	0.7009	0.1788	0.0096	0.2415
	0.5577	0.2632	0.0297	0.3111
	0.4555	0.3204	0.0360	0.3479
	0.4021	0.3375	0.0398	0.3601
	0.2760	0.3787	0.0705	0.3921
TAME (1) + ethanol (2) + water (3)	0.8480	0.0683	0.0027	0.0492
	0.6206	0.1971	0.0045	0.1024
	0.4266	0.2910	0.0106	0.1549
	0.1194	0.2823	0.0525	0.2291
TAME (1) + 1-propanol (2) + water (3)	0.8091	0.1028	0.0021	0.0151
	0.5630	0.2296	0.0069	0.0289
	0.4305	0.2937	0.0032	0.0332
	0.2459	0.3526	0.0026	0.0428
	0.1559	0.3568	0.0027	0.0501
TAME (1) + 2-propanol (2) + water (3)	0.8530	0.0590	0.0022	0.0170
	0.6652	0.1787	0.0033	0.0383
	0.4195	0.2758	0.0031	0.0572
	0.2848	0.3178	0.0043	0.0691
	0.2030	0.3220	0.0049	0.0776
	0.1385	0.2991	0.0094	0.0972
	0.0976	0.2685	0.0118	0.1112
TAME (1) + 1-butanol (2) + water (3)	0.7861	0.1084	0.0023	0.0032
	0.5958	0.2226	0.0018	0.0056
	0.4431	0.3081	0.0016	0.0077
	0.3449	0.3599	0.0015	0.0092
	0.2124	0.4294	0.0012	0.0113
	0.1164	0.4672	0.0038	0.0260
	0.0000	0.5015	0.0000	0.0198
TAME (1) + 2-butanol (2) + water (3)	0.8128	0.0818	0.0048	0.0058
	0.5974	0.2069	0.0020	0.0115
	0.4355	0.3126	0.0049	0.0190
	0.2679	0.3870	0.0018	0.0220
	0.1685	0.4165	0.0013	0.0240
	0.0964	0.4200	0.0016	0.0320
	0.0000	0.3228	0.0000	0.0524

Liquid−Liquid Equilibrium Data and Data Correlation. The measured LLE data for the binary system TAME + water at (288.15, 293.15, 298.15, 303.15, 308.15, and 313.15) K and at atmospheric pressure are listed in Table 2 and plotted in Figure 1. The solubility of TAME in the aqueous phase was less than 0.6 mol %, whereas the solubility of water in the TAME phase increased with increasing temperature. The highest solubility of water in the TAME phase was more than 2.7 mol % in the investigated temperature range. The adjustable binary parameters of the NRTL and the UNIQUAC models are listed in Table 3, along with the mean deviations between the experimental values and values that were recalculated using these two model parameters. In this work, we used the DDBST software package[2] for the regression of

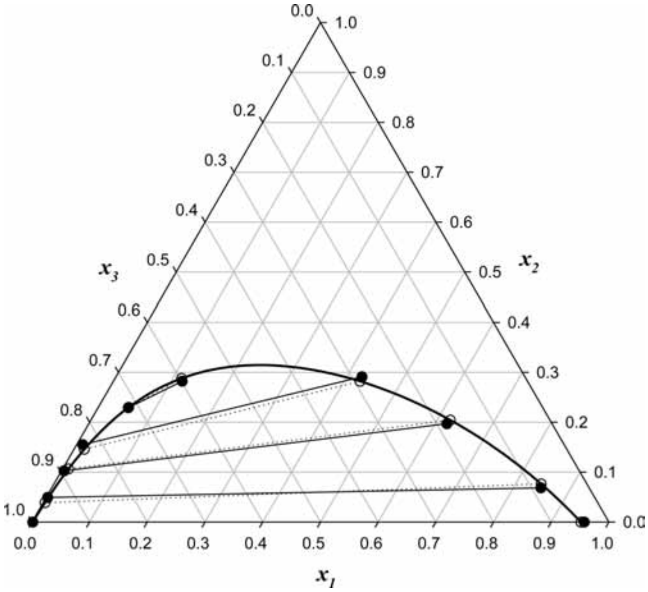

Figure 3. LLE for the ternary system TAME (1) + ethanol (2) + water (3) at 298.15 K: ●, experimental value; ○, calculated value. Dashed lines were calculated from the NRTL equation.

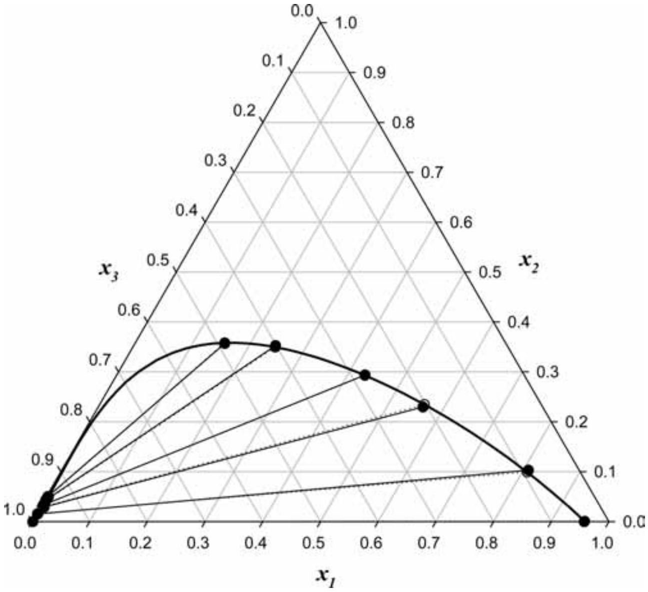

Figure 4. LLE for the ternary system TAME (1) + 1-propanol (2) + water (3) at 298.15 K: ●, experimental value; ○, calculated value. Dashed lines were calculated from the UNIQUAC equation.

our binary and ternary data, in which NRTL parameter α was fixed at 0.2. The NRTL model gave a slightly better correlation result with a mean deviation of ca. 0.3 mol % of the TAME mole fraction. The solid lines in Figure 1 represent the calculated values by the NRTL model.

The experimental ternary LLE data for six ternary systems of TAME (1) + C_1-C_4 alcohols (2) + water (3) at 298.15 K are given in Table 4. The ternary LLE data for each system are plotted in the form of Gibbs triangles in Figures 2, 3, 4, 5, 6, and 7. Ternary LLE data were also correlated using NRTL and UNIQUAC models. We determined each constituent binary parameter of NRTL and UNIQUAC models by minimizing the differences between the experimental and calculated mole fractions for each component over all of the measured LLE data of the ternary systems. The objective function (OF) used was

$$\mathrm{OF} = \min \sum_i \sum_j \sum_k (x_{ijk} - x^{\mathrm{c}}_{ijk})_2 \qquad (1)$$

where x and x^{c} are the experimental and calculated mole fractions, respectively. The subscripts i, j, and k denote component, phase, and tie line, respectively. The parameters can be optimized according to the mean deviation in the compositions of coexisting phases.[5] The binary parameters optimized are listed in Table 5 with the root-mean-square deviation (RMSD) values between experimental and calculated data defined as

$$\mathrm{RMSD} = \left[\frac{\sum_i \sum_j \sum_k (x_{ijk} - x^{\mathrm{c}}_{ijk})^2}{6N} \right]^{1/2} \qquad (2)$$

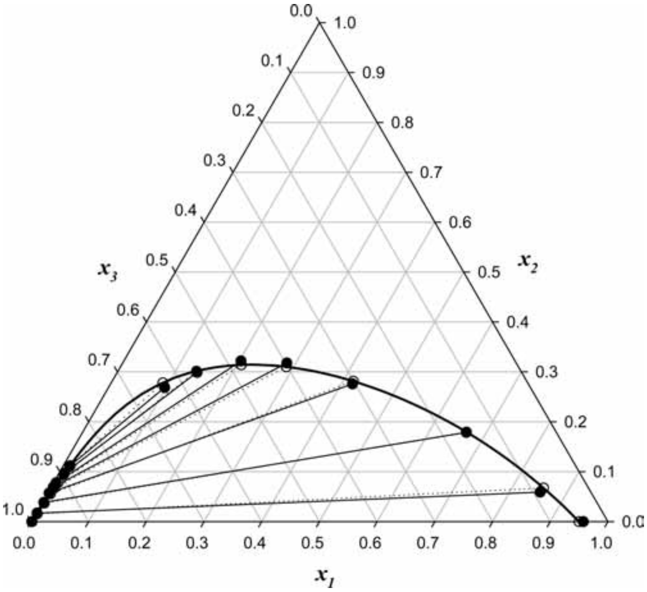

Figure 5. LLE for the ternary system TAME (1) + 2-propanol (2) + water (3) at 298.15 K: ●, experimental value; ○, calculated value. Dashed lines were calculated from the NRTL equation.

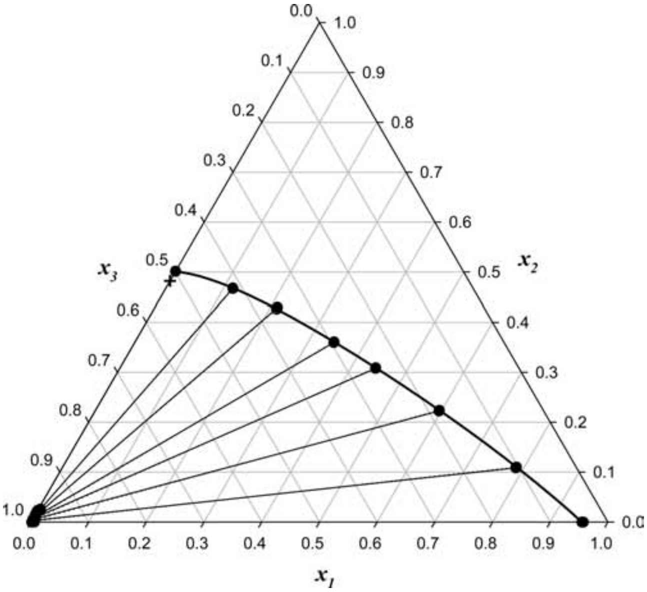

Figure 6. LLE for the ternary system TAME (1) + 1-butanol (2) + water (3) at 298.15 K: ●, experimental value; ○, calculated value; +, 1-butanol (1) + water (3) by Petritis.[10] Dashed lines were calculated from the NRTL equation.

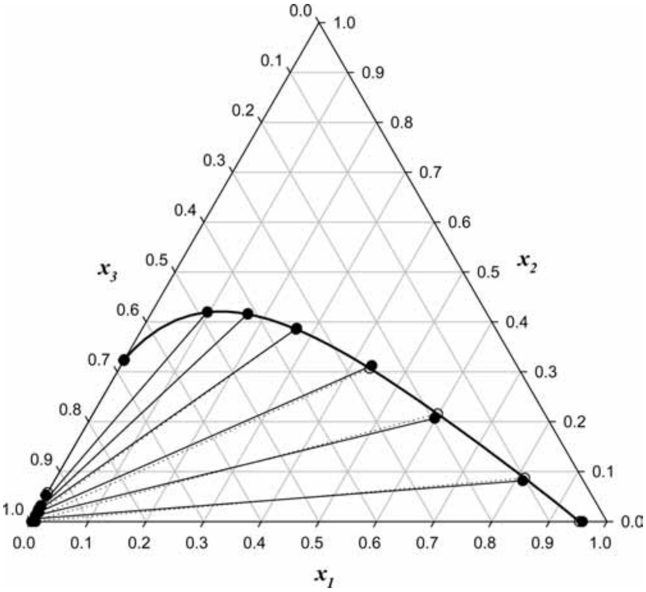

Figure 7. LLE for the ternary system TAME (1) + 2-butanol (2) + water (3) at 298.15 K: ●, experimental value; ○, calculated value; +, 2-butanol (2) + water (3) by Bozdag.[11] Dashed lines were calculated from the UNIQUAC equation.

Table 5. Correlated NRTL and UNIQUAC Model Parameters for the Ternary Systems TAME (1) + C_1−C_4 Alcohols (2) + Water (3) at 298.15 K

		NRTL parameters (K)			
component	$i-j$	$(g_{ij} - g_{ii})/R$	$(g_{ji} - g_{jj})/R$	α_{ij}	RMSD
methanol	1−2	− 312.75	404.43		0.0063
	2−3	− 703.45	553.70	0.20	
	1−3	501.67	2394.00		
etnanol	1−2	549.14	− 196.90		0.0058
	2−3	426.59	1624.00	0.20	
	1−3	− 568.73	1143.40		
1-propanol	1−2	951.41	− 396.62		0.0034
	2−3	− 423.85	1310.9	0.20	
	1−3	422.62	1565.9		
2-propanol	1−2	651.34	− 323.87		0.0040
	2−3	− 485.82	1266.90	0.20	
	1−3	416.49	1486.20		
1-butanol	1−2	− 772.81	481.32		0.0058
	2−3	− 302.81	1491.20	0.20	
	1−3	420.33	1064.80		
2-butanol	1−2	− 589.06	25.05		0.0072
	2−3	− 389.97	1382.20	0.20	
	1−3	378.12	1273.80		
		UNIQUAC parameters (K)			
component 2	$i-j$	$(u_{ij} - u_{ii})/R$	$(u_{ji} - u_{jj})/R$		RMSD
methanol	1−2	472.71	− 156.92		0.0055
	2−3	− 438.71	803.07		
	1−3	527.08	234.05		
etnanol	1−2	− 755.26	263.19		0.0059
	2−3	− 332.55	− 332.82		
	1−3	519.62	258.85		
1-propanol	1−2	330.22	− 116.73		0.0019
	2−3	− 90.60	309.22		
	1−3	653.42	87.41		
2-propanol	1−2	320.20	350.18		0.0042
	2−3	− 165.98	377.02		
	1−3	618.71	124.11		
1-butanol	1−2	36.35	− 57.87		0.0029
	2−3	23.89	222.49		
	1−3	717.84	12.29		
2-butanol	1−2	45.33	− 196.24		0.0050
	2−3	− 14.89	197.73		
	1−3	734.37	− 8.58		

where N is the number of tie lines. The digit number 6 is the number of binary interaction parameters adjusted for a ternary system.

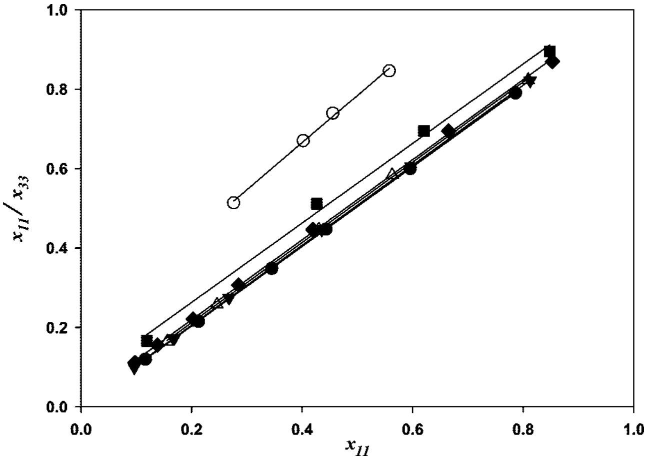

Figure 8. Bachman−Brown correlation for the ternary systems TAME (1) + C_1−C_4 alcohols (2) + water (3) at 298.15 K: ○, methanol; ■, ethanol; △, 1-propanol; ◆, 2-propanol; ●, 1-butanol; ▼, 2-butanol.

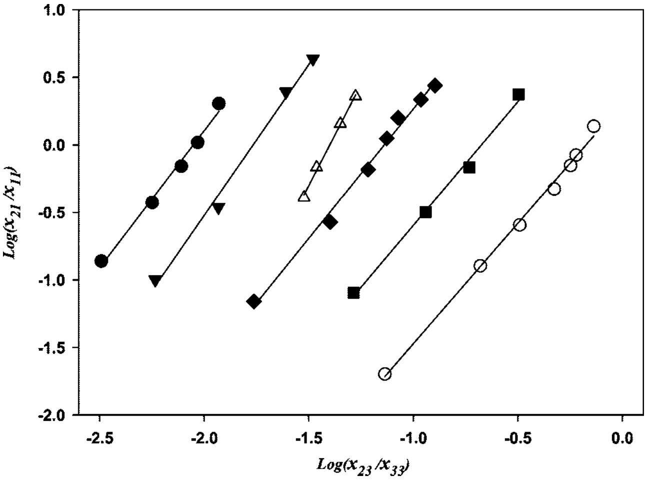

Figure 9. Hand correlation for ternary systems TAME (1) + C_1−C_4 alcohols (2) + water (3) at 298.15 K: ○, methanol; ■, ethanol; Δ, 1-propanol; ◆, 2-propanol; ●, 1-butanol; ▼, 2-butanol.

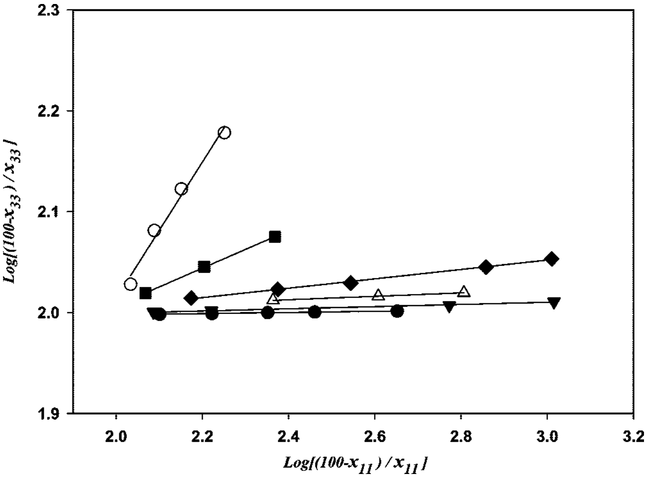

Figure 10. Othmer−Tobias correlation for ternary systems TAME (1) + C_1−C_4 alcohols (2) + water (3) at 298.15 K: ○, methanol; ■, ethanol; Δ, 1-propanol; ◆, 2-propanol; ●, 1-butanol; ▼, 2-butanol.

The ternary LLE of each system at 298.15 K are plotted in the form of Gibbs triangles in Figures 2, 3, 4, 5, 6, and 7. In the case of methanol, ethanol, and 1- and 2-propanol, the hydrogen bonding in water is stronger than that in the alcohol because these compounds are completely miscible with water.

Table 6. Bachman−Brown, Hand, and Othmer−Tobias Correlation Parameters for the Ternary Systems TAME (1) + C_1−C_4 Alcohols (2) + Water (3) at 298.15 K

correlation		methanol	ethanol	1-propanol	2-propanol	1-butanol	2-butanol
Bachman−Brown	$A =$	1.187	1.001	1.011	1.008	1.002	1.008
	$B =$	0.190	0.063	0.010	0.017	0.003	0.004
	$r =$	0.998	0.996	0.999	0.999	0.999	0.999
Hand	$C =$	1.783	1.831	3.000	1.915	2.028	2.232
	$D =$	0.311	1.235	4.197	2.181	4.161	3.941
	$r =$	0.996	0.994	0.997	0.992	0.990	0.993
Othmer−Tobias	$E =$	0.676	0.186	0.017	0.047	0.006	0.011
	$F =$	0.661	1.634	1.972	1.912	1.986	1.978
	$r =$	0.989	0.999	0.999	0.996	0.990	0.990

Table 7. Calculated Distribution Values, *D*, and Selectivity Values, *S*, for Each Tie Line

system	x_{21}	D	S
TAME (1) + methanol (2) + water (3)	0.0185	0.2721	3.2647
	0.1030	0.5981	3.1088
	0.1788	0.7404	2.4516
	0.2632	0.8460	1.8560
	0.3204	0.9210	1.6305
	0.3375	0.9372	1.5052
	0.3787	0.9658	1.2400
TAME (1) + ethanol (2) + water (3)	0.0683	1.3882	9.1083
	0.1971	1.9248	5.0505
	0.2910	1.8786	3.2416
	0.2823	1.2322	1.3258
TAME (1) + 1-propanol (2) + water (3)	0.1028	6.8079	35.5875
	0.2296	7.9446	18.0545
	0.2937	8.8464	15.4839
	0.3526	8.2383	10.8963
	0.3568	7.1218	8.4143
TAME (1) + 2-propanol (2) + water (3)	0.0590	3.4706	23.5575
	0.1787	4.6658	13.8901
	0.2758	4.8217	8.2803
	0.3178	4.5991	6.4029
	0.3220	4.1495	5.1809
	0.2991	3.0772	3.5383
	0.2685	2.4146	2.6441
TAME (1) + 1-butanol (2) + water (3)	0.1084	33.8750	158.0041
	0.2226	39.7500	98.1654
	0.3081	40.0130	71.7345
	0.3599	39.1196	59.6258
	0.4294	38.0000	48.1899
	0.4672	17.9692	20.2591
TAME (1) + 2-butanol (2) + water (3)	0.0818	14.1034	74.9773
	0.2069	17.9913	44.5984
	0.3126	16.4526	29.0027
	0.3870	17.5909	23.9848
	0.4165	17.3542	20.8438
	0.4200	13.1250	14.5020

However, 1- and 2-butanol have a miscibility gap with water because the hydrocarbon part of the 1- or 2-butanol molecule hardly interacts with water even though water molecules form strong polar and hydrogen bonds with the hydroxyl group. Therefore, the systems TAME + methanol or ethanol or 1-propanol or 2-propanol + water have one partially miscible binary and are classified as Treybal's type I.[6] However, the system TAME + 1- or 2-butanol + water is therefore classified as Treybal's type II[6] because they have two partially miscible binaries. The dashed lines are tie lines calculated using the NRTL or the UNIQUAC model. The experimental and calculated LLE data agreed relatively well, as shown in the Figures. The slopes of the tie lines presented in the Figures show that methanol is more soluble in water than in TAME. Meanwhile, ethanol, 1-propanol, 2-propanol, 1-butanol, and 2-butanol are more soluble in TAME than in water. The correlated parameters of the NRTL and the UNIQUAC models for ternary systems and mean deviations of comparison results are given in Table 5. The experimental ternary LLE data are well correlated with both the NRTL and the UNIQUAC equations, with mean deviations of less than 1.0 mol % for all of the systems. The plait points of type 1 systems were calculated using best-fit model parameters: $x_1 = 0.1755$ and $x_2 = 0.4123$ for the system TAME + methanol + water, $x_1 = 0.2307$ and $x_2 = 0.3146$ for the system TAME + ethanol + water, $x_1 = 0.1750$ and $x_2 = 0.3583$ for the system TAME + 1-propanol + water, and $x_1 = 0.2254$ and $x_2 = 0.3145$ for the system TAME + 2-propanol + water.

Additionally, Bachman−Brown,[7] Hand,[8] and Othmer−Tobias[9] correlations were used to ascertain the reliability of the experimental data. The correlations are given in Figures 8, 9, and 10, and the constants of the correlations are also given in Table 6. The

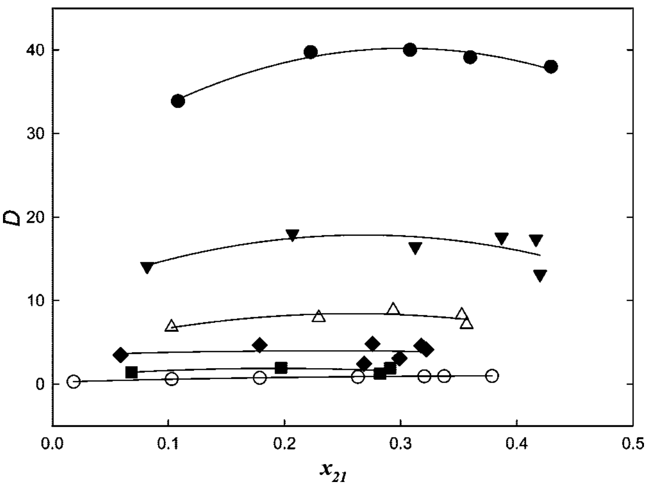

Figure 11. Distribution coefficient (D) against x_{21} for the ternary systems TAME (1) + C_1−C_4 alcohols (2) + water (3) at 298.15 K: ○, methanol; ■, ethanol; △, 1-propanol; ◆, 2-propanol; ●, 1-butanol; ▼, 2-butanol.

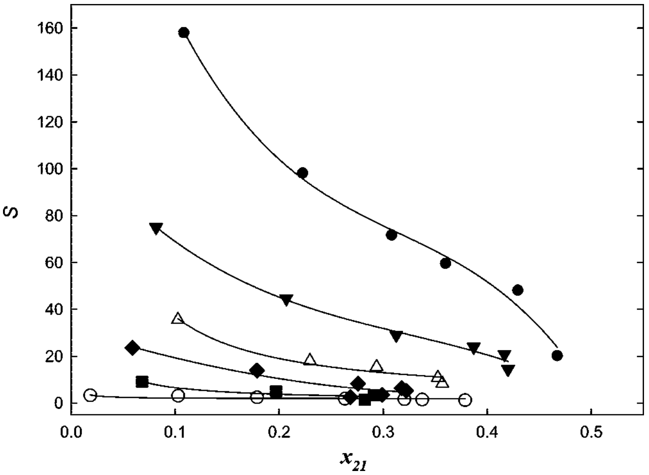

Figure 12. Selectivity (S) against x_{21} for the ternary systems TAME (1) + nitrogen-containing compounds (2) + hexadecane (3) at 298.15 K: ○, methanol; ■, ethanol; Δ, 1-propanol; ◆, 2-propanol; ●, 1-butanol; ▼, 2-butanol.

Bachman−Brown equation gave the best correlation result. The Bachman−Brown correlation is

$$x_{11}/x_{33} = Ax_{11} + B \tag{3}$$

The Hand correlation is

$$\log(x_{21}/x_{11}) = C\log(x_{23}/x_{33}) + D \tag{4}$$

The Othmer−Tobias correlation is

$$\log[(100 - x_{11})/x_{11}] = E\log[(100 - x_{33})/x_{33}] + F \tag{5}$$

Distribution Coefficient and Selectivity. The distribution coefficient (D) of C_1-C_4 alcohols over the two liquid phases in the equilibrium is defined as

$$D = \frac{x_{21}}{x_{23}} \tag{6}$$

and the effectiveness of extraction of water by C_1-C_4 alcohols could be given by its selectivity (S), which is a measure of the suitability of C_1-C_4 alcohols as separating agents of water from the TAME.

$$S = \frac{x_{21}/(x_{21} + x_{31})}{x_{23}/(x_{23} + x_{33})} \tag{7}$$

x_{21} is the mole fraction of alcohol in TAME-rich phase (organic phase), x_{23} is the mole fraction of alcohol in water-rich phase (aqueous phase), and x_{31} is the mole fraction of water in the TAME-rich phase. Calculated D and S values of each alcohol are presented in Table 7 and plotted in Figures 11 and 12. As shown in Figures 11 and 12, the D values for 1-butanol are larger than those of methanol, ethanol, 1-propanol, 2-propanol, and 2-butanol in the measured systems. The S value decreases when going through the tie-line end compositions from low to high concentrations of C_1-C_4 alcohols.

Conclusions

Binary LLE data for the system TAME + water were measured in the temperature range of (288.15 to 313.15) K. Ternary tie-line data for six different systems of TAME + C_1-C_4 alcohols + water were measured at 298.15 K. The binary LLE has no critical solution temperature under our experimental conditions. The mutual solubility of TAME and water increases with increasing temperature. The ternary system TAME + C_1-C_3 alcohols + water is Treybal's type I and has a plait point, whereas TAME + 1- or 2-butanol + water are ternary mixtures of type II and have two partially miscible binaries. Methanol is more soluble in water than in TAME, whereas ethanol, 1-propanol, 2-propanol, 1-butanol, and 2-butanol are more soluble in TAME than in water. Binary and ternary LLE data were well correlated with the NRTL and UNIQUAC models, with less than 1.0 mol % of RMSD. The Bachman−Brown, Hand, and Othmer−Tobias correlations were also used to ascertain the reliability of the experimental data, and the Bachman−Brown equation gave the best correlation result. The distribution coefficient of 1-butanol is larger than those of other alcohols. Selectivity decreased when going through the tie-line end compositions from low concentrations to high concentrations of C_1-C_4 alcohols.

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