

# Binary Liquid–Liquid Equilibrium (LLE) for *N*-Methylformamide (NMF) + Hexadecane between (288.15 and 318.15) K and Ternary LLE for Systems of NMF + Heterocyclic Nitrogen Compounds + Hexadecane at 298.15 K

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The binary liquid–liquid equilibrium (LLE) data for the system *N*-methylformamide (NMF) + *n*-hexadecane from (288.15 to 318.15) K and the ternary LLE for the systems NMF + heterocyclic nitrogen compounds (quinoline, pyridine, pyrrole, aniline, indole) + *n*-hexadecane at 298.15 K were analytically determined at atmospheric pressure by the use of stirred and thermoregulated cells. Experimental data were correlated with the NRTL and UNIQUAC activity coefficient models. The reliability of the measured LLE data were tested using the Bachman–Brown, Hand, and Othmer–Tobias equations. In addition, the extraction capability of NMF for heterocyclic nitrogen compounds was investigated with respect to distribution and selectivity.

## Introduction

Air pollution by nitrogen compounds (NO<sub>x</sub>) and particulate matter (PM) of diesel exhaust is now a serious common problem in the whole world. In the very early 2000s, most governmental regulatory limits for highway vehicle diesel were within the mass fraction range of (300 to 500)·10<sup>-6</sup> by weight of total sulfur. However, the U.S. Environmental Protection Agency (EPA) recently passed rules requiring the use of ultra-low sulfur diesel (ULSD) for diesel vehicles, which allows a mass fraction of only 15·10<sup>-6</sup> of sulfur. This ULSD oil is expected to reduce NO<sub>x</sub> and PM dramatically. So far, the hydrodesulfurization (HDS) process is most commonly used in the oil industry to reduce sulfur content in diesel fuel by converting sulfur compounds to hydrogen sulfide.<sup>1–4</sup> For the operation of the HDS process, heterocyclic nitrogen compounds act as inhibitors of desulfurization. This is the reason why they should be eliminated in the HDS process, and *N*-methylformamide (NMF) could be a good solvent for extracting nitrogen compounds from diesel oil.

Therefore, in the present study, we report the binary liquid–liquid equilibrium (LLE) data for the system of NMF + hexadecane at several temperatures from (288.15 to 318.15) K and five ternary LLE data for NMF + heterocyclic nitrogen compounds (quinoline, pyridine, pyrrole, aniline, indole) + hexadecane (model compound of diesel oil) mixtures at 298.15 K and atmospheric pressure. The experimental binary and ternary LLE data were correlated by the use of two activity coefficient models: NRTL and UNIQUAC. The Bachman–Brown, Hand and Othmer–Tobias correlations were used to ascertain the reliability of the experimental data for each system. In addition, the extraction capabilities of NMF for heterocyclic nitrogen compounds were investigated with respect to distribution and selectivity.

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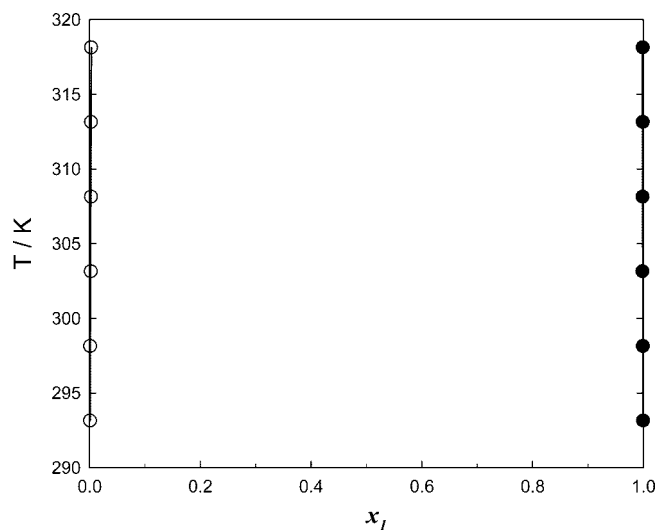
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**Table 1.** Densities of Chemicals Used in These Experiments

chemicals	$\rho/\text{g}\cdot\text{cm}^3$ at 298.15 K		GC analysis (wt %)	UNIQUAC	
	this work	ref <sup>a</sup>		<i>r</i> value <sup>a</sup>	<i>q</i> value <sup>a</sup>
NMF	0.78532	0.78500	> 99.7	2.4317	2.1920
hexadecane	0.77319	0.77330	> 99.8	11.2438	9.2560
quinoline	1.08945	1.09290 <sup>b</sup>	> 99.6	4.7923	3.1530
pyridine	0.97825	0.97820	> 99.5	2.9993	2.1130
indole	1.21668	1.21950 <sup>b</sup>	> 99.0	4.2820	2.6920
pyrrole	0.96590	0.96980 <sup>b</sup>	> 97.0	2.5734	1.8240
aniline	1.01742	1.01750	> 99.6	3.7165	2.8160

<sup>a</sup> Ref 12. <sup>b</sup> At 293.15 K.



**Figure 1.** LLE for the binary system: ●, NMF (1) + hexadecane (2). Solid curves were calculated from the UNIQUAC equation.

## Experimental Section

**Materials.** Commercial grade NMF, *n*-hexadecane, pyridine, and aniline were obtained from Aldrich. Pyrrole and quinoline were obtained from Fluka and Junsei, respectively. All chemicals were dried using molecular sieves with a pore diameter of 0.4 nm. The purity of the chemicals was examined by gas

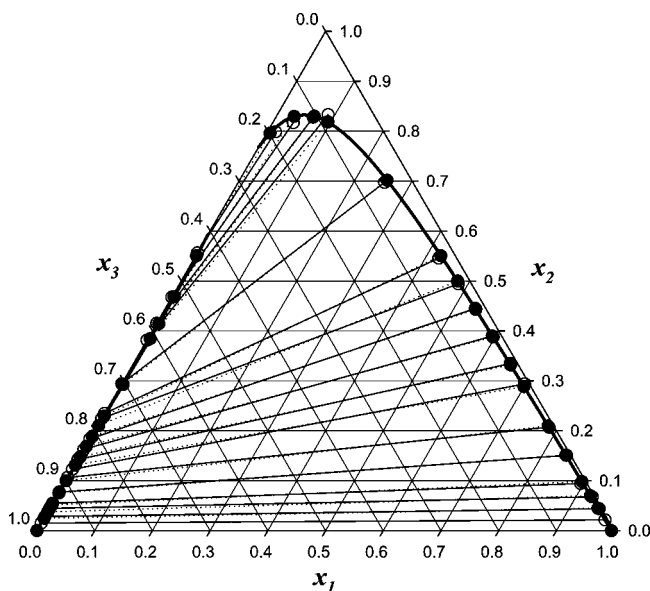
**Table 2. Experimental Data,  $G^E$  Model Parameters, and Mean Deviation between the Calculated and Experimental Values for the Binary System of NMF (1) + Hexadecane (2)**

Experimental Data				
T/K	NMF-rich phase		hexadecane-rich phase	
	$x_1$	$x_1$	$x_1$	$x_1$
293.15	0.9993		0.0009	
298.15	0.9990		0.0011	
303.15	0.9984		0.0021	
308.15	0.9985		0.0026	
313.15	0.9988		0.0027	
318.15	0.9987		0.0029	

$G^E$ Model				
	NRTL		UNIQUAC	
	$ij = 12$	$ij = 21$	$ij = 12$	$ij = 21$
$A_{ij}$ (K) <sup>a</sup>	1773.2552	4452.1926	134.5659	1890.8760
$B_{ij}$	-0.8516	-10.0943	-0.4634	-2.9627
$\alpha$	0.20			
RMSD	0.0027		0.0014	

<sup>a</sup> Parameters (K):  $a_{ij} = A_{ij} + B_{ij}T$ .



**Figure 2.** LLE for the ternary system of NMF (1) + quinoline (2) + hexadecane (3) at 298.15 K: O, experimental value; ●, calculated value. Dashed lines were calculated from the UNIQUAC equation.

chromatography and by comparing the densities with values reported in the literature. Their impurities were less than 0.5 %, except for pyrrole (< 3.0 %) and indole (< 1.0 %). The measured densities and purities of the samples are summarized in Table 1.

**Apparatus and Procedure.** The self-designed LLE measuring vessel, which is similar to that of S. Horstmann et al.,<sup>5</sup> was used. The equilibrium vessel was thermostatted in a thermostat (Lauda MD 20 with DLK 15 cooler) within  $\pm 0.02$  K. The sample mixture was stirred in an equilibrium vessel for about 8 h and was then allowed to settle for more than 16 h at constant system temperature. After both phases settled, sampling was carefully carried out from the upper and lower phases without contamination of both phases. The analysis was carried out using a gas chromatograph (HP 6890N) with an HP-5 ((30 m)•(0.32 mm)•(0.25  $\mu$ m)) capillary column and a thermal conductivity detector. We estimate the uncertainty of the calculated mole fraction to be less than ca.  $\pm 1 \cdot 10^{-4}$ . The procedure is described in detail elsewhere.<sup>6</sup>

**Table 3. Experimental LLE Data of the System NMF (1) + Nitrogen-Containing Compounds (2) + Hexadecane (3) at 298.15 K**

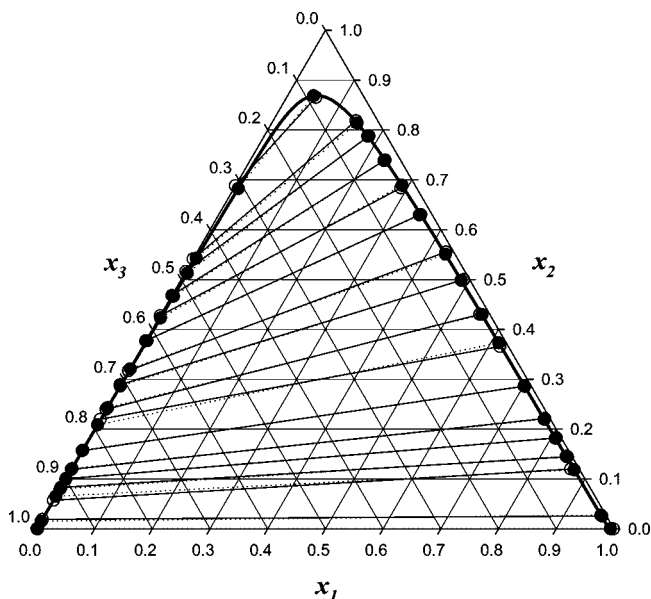
nitrogen-containing compds	NMF-rich phase		hexadecane-rich phase	
	$x_{11}$	$x_{21}$	$x_{13}$	$x_{23}$
quinoline	0.9523	0.0439	0.0000	0.0283
	0.9283	0.0674	0.0000	0.0444
	0.8947	0.0946	0.0000	0.0548
	0.8421	0.1509	0.0000	0.0773
	0.7836	0.2071	0.0000	0.1064
	0.6980	0.2938	0.0000	0.1229
	0.6529	0.3347	0.0000	0.1422
	0.5941	0.3896	0.0000	0.1629
	0.5371	0.4449	0.0000	0.1837
	0.4829	0.4942	0.0000	0.2244
	0.4235	0.5456	0.0000	0.2345
	0.2536	0.6974	0.0000	0.2942
	0.0882	0.8329	0.0000	0.3816
	0.0648	0.8279	0.0000	0.4154
	0.0358	0.8169	0.0000	0.4675
	0.0141	0.7984	0.0000	0.5564
	0.9664	0.0258	0.0000	0.0188
	0.8649	0.1201	0.0000	0.0571
	0.8474	0.1444	0.0000	0.0831
	0.8088	0.1825	0.0000	0.1005
0.7696	0.2213	0.0000	0.1194	
0.7014	0.2862	0.0000	0.1567	
0.6195	0.3657	0.0000	0.2199	
0.5524	0.4303	0.0000	0.2397	
0.4895	0.4999	0.0000	0.2890	
0.4310	0.5555	0.0000	0.3172	
0.3480	0.6298	0.0000	0.3770	
0.2886	0.6840	0.0000	0.4275	
0.2323	0.7388	0.0000	0.4680	
0.1814	0.7882	0.0000	0.5163	
0.1436	0.8186	0.0000	0.5413	
0.0506	0.8655	0.0000	0.6881	
0.9185	0.0759	0.0000	0.0000	
0.8412	0.1464	0.0000	0.0104	
0.7780	0.2157	0.0000	0.0146	
0.6466	0.3453	0.0000	0.0195	
0.5853	0.4082	0.0000	0.0333	
0.4875	0.5033	0.0000	0.0389	
0.4043	0.5912	0.0000	0.0683	
0.3275	0.6654	0.0000	0.0654	
0.2563	0.7359	0.0000	0.0761	
0.1832	0.8105	0.0000	0.1179	
0.1594	0.8319	0.0000	0.1206	
0.1145	0.8729	0.0000	0.1369	
0.0738	0.9125	0.0000	0.1484	
0.0000	0.9807	0.0000	0.1722	
0.0000	0.9874	0.0000	0.1735	
0.9635	0.0356	0.0000	0.0000	
0.9295	0.0694	0.0000	0.0000	
0.8782	0.1199	0.0000	0.0000	
0.8389	0.1599	0.0000	0.0141	
0.7888	0.2096	0.0000	0.0181	
0.7398	0.2586	0.0000	0.0234	
0.6915	0.3069	0.0000	0.0299	
0.6315	0.3667	0.0000	0.0348	
0.6040	0.3943	0.0000	0.0406	
0.5500	0.4464	0.0000	0.0469	
0.5086	0.4846	0.0000	0.0545	
0.4566	0.5410	0.0000	0.0591	
0.4367	0.5610	0.0000	0.0639	
0.3835	0.6147	0.0000	0.0712	
0.3422	0.6535	0.0000	0.0773	
0.2869	0.7106	0.0000	0.0870	
0.2369	0.7575	0.0000	0.0895	
0.1841	0.8123	0.0000	0.1035	
0.1274	0.8679	0.0000	0.1064	
0.0719	0.9206	0.0000	0.1062	
0.0000	0.9948	0.0000	0.1289	
0.9288	0.0695	0.0000	0.0000	
0.8588	0.1384	0.0000	0.0000	
0.7982	0.1992	0.0000	0.0000	
0.7328	0.2637	0.0000	0.0000	
0.6725	0.3223	0.0000	0.0122	
0.6068	0.3876	0.0000	0.0150	
0.5366	0.4567	0.0000	0.0276	
0.4715	0.5209	0.0000	0.0339	
0.4089	0.5817	0.0000	0.0386	
0.3498	0.6403	0.0000	0.0452	
0.3029	0.6845	0.0000	0.0497	
0.2783	0.7103	0.0000	0.0523	

pyridine

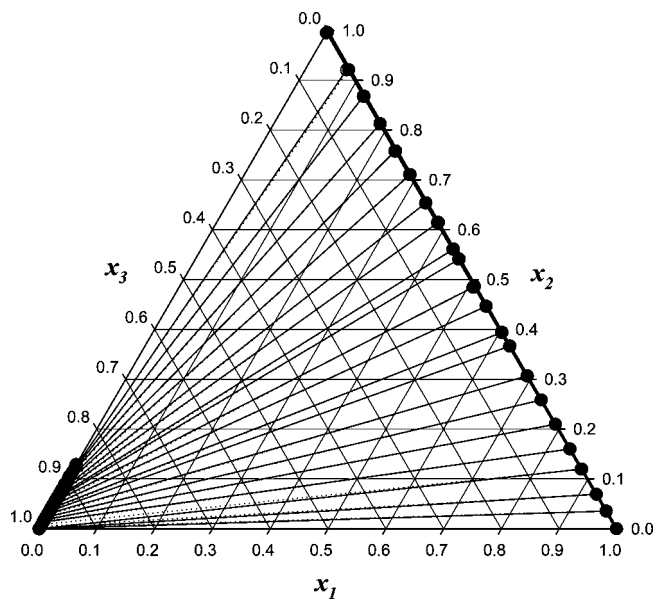
pyrrole

aniline

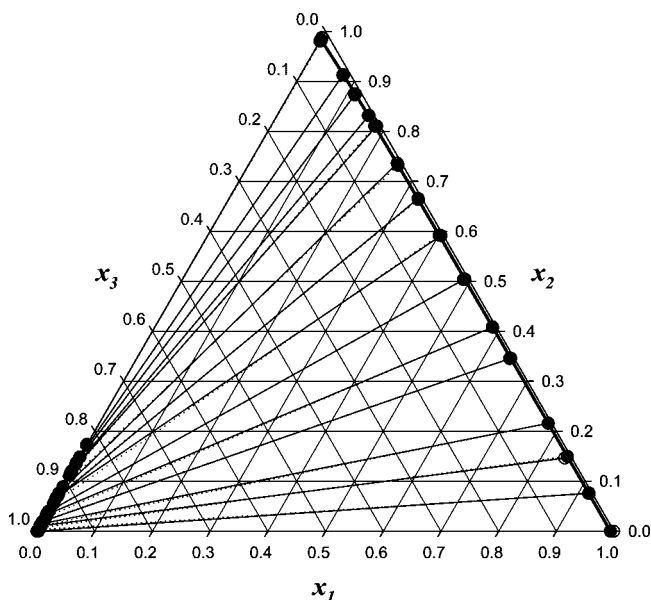
indole



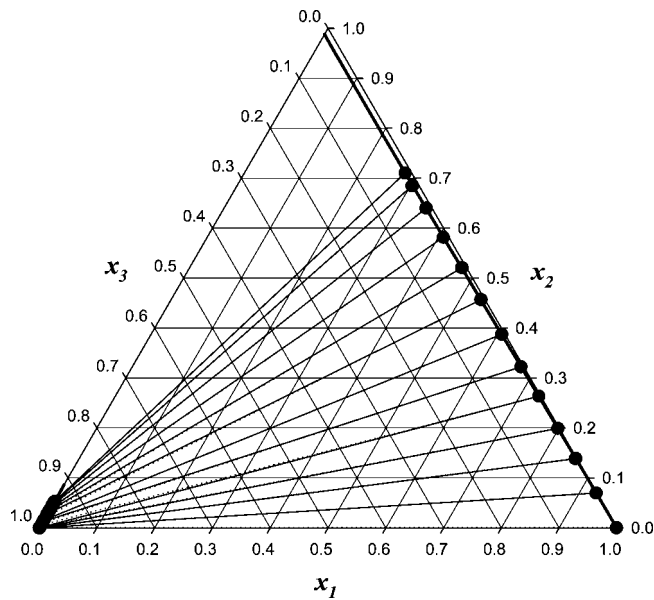
**Figure 3.** LLE for the ternary system of NMF (1) + pyridine (2) + hexadecane (3) at 298.15 K; ○, experimental value; ●, calculated value. Dashed lines were calculated from UNIQUAC equation.



**Figure 5.** LLE for the ternary system of NMF (1) + aniline (2) + hexadecane (3) at 298.15 K; ○, experimental value; ●, calculated value. Dashed lines were calculated from the UNIQUAC equation.



**Figure 4.** LLE for the ternary system of NMF (1) + pyrrole (2) + hexadecane (3) at 298.15 K; ○, experimental value; ●, calculated value. Dashed lines were calculated from the NRTL equation.



**Figure 6.** LLE for the ternary system of NMF (1) + indole (2) + hexadecane (3) at 298.15 K; ○, experimental value; ●, calculated value. Dashed lines were calculated from the UNIQUAC equation.

## Results and Discussion

**Tie Lines and Data Correlation Using NRTL and UNIQUAC.** The LLE data for the binary system NMF + *n*-hexadecane at (293.15, 298.15, 303.15, 308.15, 313.15, and 318.15) K and at atmospheric pressure are listed in Table 2 and plotted in Figure 1. The binary LLE data were correlated with the NRTL and the UNIQUAC models. The adjustable binary parameters of the NRTL and the UNIQUAC models are listed in Table 2, along with the root-mean-square deviations (RMSDs) between the experimental values and values recalculated using these two models. The UNIQUAC model gave slightly better correlation results with a 0.14 % mean deviation of the NMF mole fraction. The dashed lines in Figure 1 represent the values calculated using the UNIQUAC model.

The measured ternary LLE data for NMF + nitrogen-containing compounds (quinoline, pyridine, pyrrole, aniline,

indole) + *n*-hexadecane mixtures at 298.15 K are given in Table 3. The ternary LLE data for each system are plotted in the form of Gibbs triangles in Figures 2, 3, 4, 5, and 6. The experimental LLE data points were correlated using NRTL and UNIQUAC models. We determined the constituent binary parameters of both 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

$$\text{OF} = \min \sum_i \sum_j \sum_k (x_{ijk} - x_{ijk}^c)^2 \quad (1)$$

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 composi-

**Table 4. NRTL and UNIQUAC Model Parameters for the Ternary Systems of NMF (1) + Nitrogen-Containing Compounds (2) + Hexadecane (3) at 298.15 K**

nitrogen-containing compds	NRTL parameters ( <i>K</i> )				RMSD
	<i>i</i> – <i>j</i>	( <i>g<sub>ij</sub></i> – <i>g<sub>ii</sub></i> )/ <i>R</i>	( <i>g<sub>ji</sub></i> – <i>g<sub>jj</sub></i> )/ <i>R</i>	<i>α<sub>ij</sub></i>	
quinoline	1–2	– 49.718	365.390	0.20	0.0054
	2–3	782.170	– 23.063	0.20	
	1–3	1064.200	1872.200	0.20	
pyridine	1–2	– 445.860	638.340	0.20	0.0040
	2–3	1172.900	– 364.060	0.20	
	1–3	906.160	1880.800	0.20	
pyrrole	1–2	– 618.730	330.660	0.20	0.0035
	2–3	1160.800	112.650	0.20	
	1–3	989.450	1305.600	0.20	
aniline	1–2	156.160	– 187.620	0.20	0.0025
	2–3	1441.700	159.310	0.20	
	1–3	1314.500	1450.200	0.20	
indole	1–2	143.990	– 585.340	0.20	0.0021
	2–3	970.100	300.160	0.20	
	1–3	1263.900	1347.600	0.20	

nitrogen-containing compds	UNIQUAC parameters ( <i>K</i> )			RMSD
	<i>i</i> – <i>j</i>	( <i>u<sub>ij</sub></i> – <i>u<sub>ii</sub></i> )/ <i>R</i>	( <i>u<sub>ji</sub></i> – <i>u<sub>jj</sub></i> )/ <i>R</i>	
quinoline	1–2	–162.400	339.520	0.0048
	2–3	–111.810	265.960	
	1–3	–59.186	1352.200	
pyridine	1–2	–218.150	401.680	0.0036
	2–3	–65.550	266.710	
	1–3	–78.160	1419.500	
pyrrole	1–2	–223.110	28.442	0.0035
	2–3	–59.734	522.640	
	1–3	–50.972	1063.200	
aniline	1–2	193.220	–190.470	0.0022
	2–3	35.937	254.130	
	1–3	9.682	1095.800	
indole	1–2	278.280	–488.140	0.0016
	2–3	–105.520	412.100	
	1–3	22.297	976.790	

**Table 5. Bachman–Brown, Hand, and Othmer–Tobias Correlation of LLE Data for the Ternary Systems of NMF (1) + Nitrogen-Containing Compounds (2) + Hexadecane (3) at 298.15 K**

correlation	parameter, linearity	quinoline	pyridine	pyrrole	aniline	indole
		Bachman–Brown	<i>A</i> = 0.806	0.681	0.991	0.981
	<i>B</i> = 0.228	0.342	0.020	0.030	0.023	
	<i>r</i> = 0.996	0.993	0.999	0.999	0.999	
Hand	<i>A</i> = 1.445	1.278	1.313	1.260	0.780	
	<i>B</i> = 0.826	0.552	1.830	1.582	0.921	
	<i>r</i> = 0.994	0.997	0.996	0.991	0.999	
Othmer–Tobias	<i>A</i> = 0.295	0.394	0.073	0.085	0.049	
	<i>B</i> = 1.418	1.225	1.047	1.825	1.894	
	<i>r</i> = 0.989	0.994	0.991	0.989	0.989	

tions of coexisting phases.<sup>7</sup> The binary parameters optimized are listed in Table 4 with the RMSD values between experimental and calculated data defined as

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

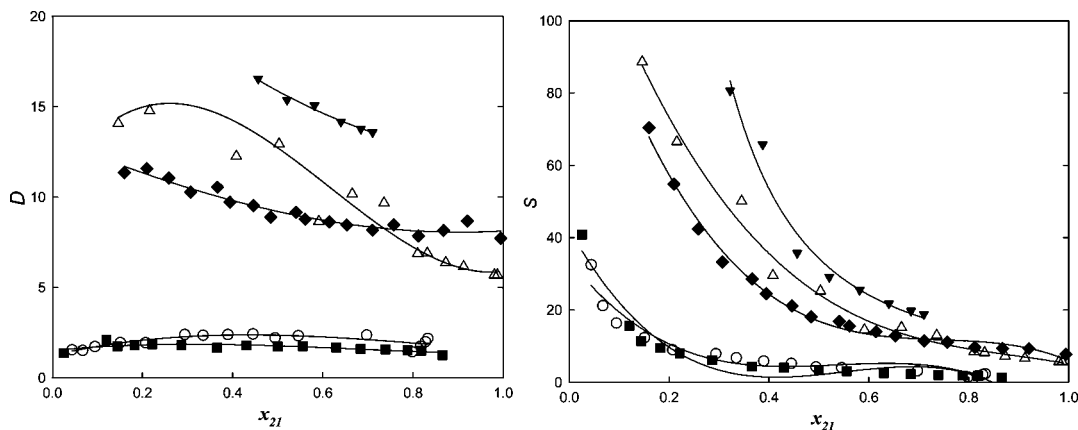
where *N* is the number of tie lines. The digit number 6 is the number of binary interaction parameters adjusted for a ternary system. The ternary LLE of each system at 298.15 K are plotted in Figures 2, 3, 4, 5, and 6. The dashed lines are calculated tie lines. The experimental and calculated LLE data agreed relatively well, as shown in the Figures. The slopes of the tie lines presented in the Figures show that quinoline, pyridine, pyrrole, aniline, and indole are more soluble in NMF than in hexadecane. All ternary systems are therefore classified as Treybal's type II<sup>8</sup> because they have two partially miscible binaries. The correlated parameters of the NRTL and UNIQUAC models for ternary systems and mean deviations of comparison

**Table 6. Experimental Distribution Values, *D*, and Selectivity Values, *S*, for Each Tie Line**

nitrogen-containing compds	<i>x</i> <sub>21</sub>	<i>D</i>	<i>S</i>
quinoline	0.0439	1.5512	32.5207
	0.0674	1.5180	21.1718
	0.0946	1.7263	16.3939
	0.1509	1.9521	12.3631
	0.2071	1.9464	8.9946
	0.2938	2.3906	7.9158
	0.3347	2.3537	6.7811
	0.3896	2.3917	5.8922
	0.4449	2.4219	5.2320
	0.4942	2.2023	4.2590
	0.5456	2.3267	4.0358
	0.6974	2.3705	3.1759
	0.8329	2.1827	2.3938
	0.8279	1.9930	2.1311
	0.8169	1.7474	1.8123
pyridine	0.7984	1.4349	1.4555
	0.0258	1.3723	40.8435
	0.1201	2.1033	15.5687
	0.1444	1.7377	11.3871
	0.1825	1.8159	9.4975
	0.2213	1.8534	8.0444
	0.2862	1.8264	6.1166
	0.3657	1.6630	4.3706
	0.4303	1.7952	4.0106
	0.4999	1.7298	3.3884
	0.5555	1.7513	3.0778
	0.6298	1.6706	2.5622
	0.6840	1.6000	2.2491
	0.7388	1.5786	2.0563
	0.7882	1.5266	1.8649
pyrrole	0.8186	1.5123	1.7659
	0.8655	1.2578	1.3248
	0.1464	14.0769	88.6456
	0.2157	14.7740	66.5494
	0.3453	17.7077	50.1067
	0.4082	12.2583	29.5593
	0.5033	12.9383	25.2455
	0.5912	8.6559	14.5307
	0.6654	10.1743	15.1291
	0.7359	9.6702	13.0028
	0.8105	6.8745	8.4163
	0.8319	6.8980	8.2061
	0.8729	6.3762	7.2007
	0.9125	6.1489	6.6389
	0.9807	5.6951	5.6951
0.9874	5.6911	5.6911	
aniline	0.1599	11.3404	70.3937
	0.2096	11.5801	54.8301
	0.2586	11.0513	42.4723
	0.3069	10.2642	33.2714
	0.3667	10.5374	28.5953
	0.3943	9.7118	24.5248
	0.4464	9.5181	21.1514
	0.4846	8.8917	18.0947
	0.5410	9.1540	16.8457
	0.5610	8.7793	15.5856
	0.6147	8.6334	14.0039
	0.6535	8.4541	12.8520
	0.7106	8.1678	11.4540
	0.7575	8.4637	11.0912
	0.8123	7.8483	9.6192
0.8679	8.1570	9.3479	
0.9206	8.6685	9.3401	
0.9948	7.7176	7.7176	
indole	0.3223	26.4180	80.6657
	0.3876	25.8400	65.7172
	0.4567	16.5471	35.7080
	0.5209	15.3658	29.0743
	0.5817	15.0699	25.4948
	0.6403	14.1659	21.7870
	0.6845	13.7726	19.7570
	0.7103	13.5813	18.8184

results are given in Table 4. The experimental ternary LLE data are well correlated with both models. Their RMSDs between measured and calculated mole fractions are less than 0.5 % for almost all systems. Additionally, Bachman–Brown,<sup>9</sup> Hand<sup>10</sup> and Othmer–Tobias<sup>11</sup> correlations were used to ascertain





**Figure 7.** Distribution coefficient ( $D$ ) and selectivity ( $S$ ) against  $x_{21}$  for the ternary systems of NMF (1) + nitrogen-containing compounds (2) + hexadecane (3) at 298.15 K: ○, quinoline; ■, pyridine; △, pyrrole; ◆, aniline; ▼, indole.

the reliability of the experimental data for each system. The constants of the correlations are given in Table 5. The Bachman–Brown correlation gave the best results for almost all of the systems. The Bachman–Brown correlation is

$$x_{11}/x_{33} = Ax_{11} + B \quad (3)$$

The Hand correlation is

$$\log(x_{21}/x_{11}) = A \log(x_{23}/x_{33}) + B \quad (4)$$

The Othmer–Tobias correlation is

$$\log[(100 - x_{11})/x_{11}] = A \log[(100 - x_{33})/x_{33}] + B \quad (5)$$

where  $A$  and  $B$  are each model parameters and  $r$  is a calculated linearity.

**Distribution and Selectivity.** The distribution coefficient ( $D$ ) of the solute (nitrogen-containing compounds) over the two liquid phases in the equilibrium is defined as

$$D = \frac{x_{21}}{x_{23}} \quad (6)$$

where  $x_{21}$  is the mole fraction of solute in the NMF-rich phase and  $x_{23}$  is the mole fraction of solute in the hexadecane-rich phase. Calculated  $D$  values of each solute are presented in Table 6 and plotted in Figure 7. As shown in Figure 7, the  $D$  values of indole, pyrrole, and aniline are greater than those of quinoline and pyridine in the measured systems. The effectiveness of the extraction of nitrogen-containing compounds by NMF could be given by its selectivity ( $S$ )

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

$x_{21}$  is the mole fraction of solute in the NMF-rich phase,  $x_{23}$  is the mole fraction of solute in the hexadecane-rich phase, and  $x_{31}$  is the mole fraction of hexadecane in the NMF-rich phase.

The experimental  $S$  values are also listed in Table 6 and plotted in Figure 7. As shown in Figure 7, the  $S$  value decreases when going through the tie-line end compositions from low to high concentrations of nitrogen-containing compounds.

## Conclusions

Binary LLE data for the NMF +  $n$ -hexadecane were measured in the temperature ranging of (293.15 to 318.15) K, and ternary tie-line data for five different NMF + nitrogen-containing compounds + hexadecane systems were measured at 298.15 K. The

UNIQUAC model gave a slightly better correlation result with 0.14 % mean deviation of NMF mole fraction for the binary system. All of the ternary systems are classified as Treybal's type II because they have two partially miscible binaries. They were also well correlated with the NRTL and the UNIQUAC models, and their RMSDs between the calculated and measured values were mostly less than 0.6 mol %. The Bachman–Brown correlation gave the best linearity. The  $D$  values of indole, pyrrole, and aniline are greater than those of quinoline and pyridine in the measured systems. Selectivity,  $S$ , decreases when going through the tie-line end compositions from low to high concentrations of nitrogen-containing compounds.

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