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Vapor−Liquid Equilibrium Studies for Systems Containing *n*-Butylisocyanate at Temperatures between 323.15 K and 371.15 K

Suhel Ahmad, Ralf Giesen, and Klaus Lucas*
Institute of Technical Thermodynamics, Aachen University of Technology (RWTH Aachen),
Schinkelstr. 8, D-52056 Aachen, Germany

Vapor−liquid equilibria for the binary systems *n*-butylisocyanate (1-isocynato-butane) + *n*-butanol, *n*-nonane + *n*-butanol, *n*-nonane + *n*-butylisocyanate, and *N*-butylcarbamicacidbutylester (urethane) + *n*-nonane and for the ternary systems *N*-butylcarbamicacidbutylester + *n*-butanol + *n*-nonane and *n*-butylisocyanate + *n*-butanol + *N*-butylcarbamicacidbutylester have been studied at (323.15, 338.15, 353.15, and 371.15) K. The measurements have been performed using a specially designed reactor and attenuated total reflection Fourier transform infrared based spectroscopic technique. The vapor pressure and the composition of the liquid phase have been measured simultaneously. The experimental results have been correlated using UNIQUAC and NRTL models, and the binary interaction parameters have been determined. By use of the binary interaction parameters, the VLE of ternary systems is predicted.

Introduction

Isocyanates are compounds that contain at least one nitrogen−carbon−oxygen (−N=C=O) group. They constitute a group of highly reactive substances which readily undergo a chemical reaction with alcohols to form urethanes. Urethanes (or their polymer polyurethanes) have applications in various industries, e.g., plastics, paints, agriculture, rubber, foams, etc. *N*-Butylisocyanate has a particularly high industrial demand and is used in the production of pesticides, pharmaceuticals, cosmetics, and agricultural products.[1] However, despite its technological potential, thermodynamic data for isocyanates and their mixtures are scarce. Other than the measurements from our institute,[2,3] few published data are available.[4−6]

For this reason, the systems *n*-nonane + *n*-butanol, *n*-nonane + *n*-butylisocyanate, urethane + *n*-nonane, urethane + *n*-butanol, urethane + *n*-butylisocyanate, *n*-butylisocyanate + *n*-butanol + urethane, and urethane + *n*-butanol + *n*-nonane were studied and reported in this work.

Because of high reactivity and toxicity of the isocyanate group, a very high level of safety is maintained while experimenting with this group of chemicals. A reactor has been designed to measure the composition of the liquid phase and the vapor pressure simultaneously. This is necessary to study the reactive systems such as *n*-butylisocyanate + *n*-butanol where the composition of the liquid phase changes as the reaction proceeds forward with time.

As far as we are aware, only a few VLE measurements for the abovementioned systems in a restricted temperature range have been reported in the past. Heintz et al.[7] published the VLE for *n*-nonane + *n*-butanol at 323.15 K, and Hamann[8] reported the VLE for *n*-butylisocynate + *n*-nonane at 50 kPa.

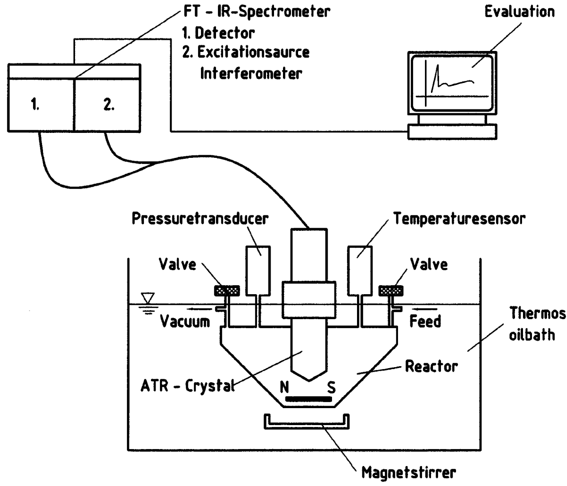

Figure 1. Schematic diagram of vapor−liquid equilibrium apparatus.

Experimental Section

Apparatus. For simultaneous measurement of the vapor pressure and the composition of the liquid phase and considering the hazardous nature of *n*-butylisocynate, a special reactor of stainless steel has been constructed. The apparatus is shown schematically in Figure 1. The reactor consists of two parts: The lower part is conical, while the upper part is a circular disk. Two valves (for filling and for evacuation), a pressure transducer, a Pt 100 thermometer, and an attenuated total reflection (ATR) immersion probe are introduced through the disk. The ATR immersion probe has been placed in the center. Both the upper and lower parts are fixed together with the help of an O ring. Further, to maintain the homogeneity in the mixture, a magnet fish driven from outside has been placed in the reactor. A precalibrated and certified pressure transducer (±2 mbar in the range of 0−2000 mbar) is used for measuring the vapor pressure. The Pt 100 thermometer is calibrated with the help of a standard quartz crystal thermometer and a water triple-point cell. The uncertainties in the temperature measurements are expected to be ±0.01 K. The signal of pressure and temperature measurements are digitized by an analog-to-digital converter and can be read on the display. The ATR immersion probe, which is made of ZnSe, is connected to a FTIR spectrometer (Vector 22, Firm Bruker, Germany). The reactor is placed in an oil thermostat. The temperature of the thermostat

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* To whom correspondence may be addressed. E-mail: lucas@ltt.rwth-aachen.de.

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Table 1. Measured Vapor Pressures of *n*-Nonane, *n*-Butanol, *n*-Butylisocyanate, and Urethane

n-nonane		*n*-butanol		*n*-butylisocyanate		urethane	
T/K	P/kPa	T/K	P/kPa	T/K	P/kPa	T/K	P/kPa
322.59	2.34	314.19	2.70	294.15	2.48	322.60	0.880
338.33	4.99	323.20	4.49	323.24	9.52	337.20	1.287
351.53	9.00	338.21	10.38	338.02	17.88	342.30	1.494
371.37	19.81	342.17	12.89	353.02	32.10	352.50	1.957
		352.81	21.44	362.65	44.82	362.60	2.381
		361.24	31.52	372.60	61.84	371.70	2.953
		371.94	49.26	387.87	99.08		

Table 2. Antoine Parameters

		Antoine coefficients			
substance	temperature range T/K	A	B	C	data source
n-nonane	323.15−373.15	6.93442	1429.459	201.82	ref 10
n-butanol	314.19−373.15	7.838	1558.19	196.881	ref 10
n-butylisocyanate	293.12−388.15	7.721795	1723.782294	250.147212	this work and refs 8 and 9
urethane	323.15−373.15	4.540852	994.331432	225.438225	this work

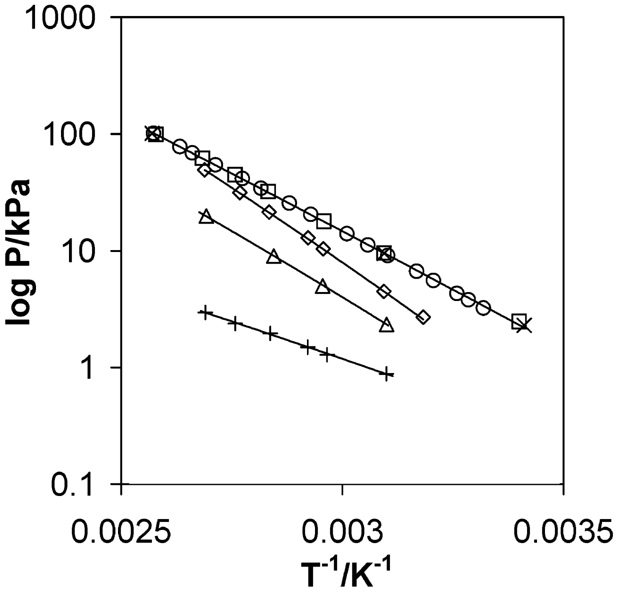

Figure 2. Vapor pressures of pure substances: □, experimental data for *n*-butylisocyanate this work; ○, Hamann;[8] ×, Merck Schuchardt;[9] solid line, fit this work; solid line, *n*-butanol this work; solid line, fit Gmehling et al.;[10] △, *n*-nonane this work; solid line, fit Gmehling et al.;[10] +, urethane this work; solid line, fit this work.

Table 3. Experimental $P-T-x$ Data for the System *n*-Nonane (1) + *n*-Butanol (2)

	P/kPa			
x_2	323.15 K	338.15 K	353.15 K	371.15 K
0.042		8.38	15.60	29.43
0.072		9.45	17.85	34.30
0.129	4.96	10.18	19.58	38.62
0.313	5.32	11.08	22.39	46.29
0.449	5.38	11.35	23.03	48.15
0.458	5.43	11.28	23.45	48.33
0.595	5.61	11.63	23.58	49.73
0.669	5.73	11.74	23.96	50.31
0.832	5.55	11.72	23.45	49.99
0.910	5.32	11.02	22.84	49.53

can be adjusted from 273 K to 420 K and does not vary by more than ±0.5 K.

Materials. The chemicals *n*-butylisocyanate, *n*-butanol, and *n*-nonane with stated purities of better than 98, 99, and 99.5 mol %, respectively, were obtained from Fluka AG, Germany. They were used after a degassing process. Urethane was prepared by mixing *n*-butylisocyanate and *n*-butanol in exactly equimolar proportion via a complete chemical reaction.

Procedures. In the beginning of the experiment, the pure substances were degassed to free them from low boiling trace substances. The desired mixture was prepared by weighing appropriate amounts of the components. A well-known composition was fed into an evacuated reactor. The reactor with the desired composition of the mixture was then placed into the thermostat that had already been brought to a preset temperature. After an equilibration time, a constant vapor pressure was noted. At the same time, the spectrum of the liquid phase was taken. The temperature of the thermostat was then raised to the next higher temperature, and the vapor pressure and the spectrum were recorded. The same process was repeated at four different temperatures for each of the binary systems.

Before performing the spectroscopic measurement, a calibration was made by taking the spectra of the liquid mixtures of known composition. The spectra scanned over a wide wavelength range (500−4000) cm^{-1} were subjected to the well-known partial least-squares method to establish a calibration model at a definite temperature. This model was subsequently used to obtain the composition of the known liquid mixtures with sufficient accuracy. For this purpose, a commercially available software “QUANT II” (Firm Bruker, Germany) was used. The overall uncertainty in the composition of the liquid phase is estimated to be ±0.02.

Results

To check the apparatus and the accuracy of the method, the vapor pressures of the pure substances *n*-nonane, *n*-butanol, *n*-butylisocyanate, and urethane were measured at different temperatures and compared with the values from the available literature.[8−10] These are given in Table 1 and shown in Figure 2. The average deviation between the experimental value and the literature value vary from 0.7% for *n*-nonane,[10] 1.2% for *n*-butanol,[10] and 1.5% for *n*-butylisocyanate.[8,9] The experimental vapor pressures at different temperatures have been fitted to an Antoine-type equation in the form

$$\log(P/\text{mbar}) = A - \frac{B}{t/{}^\circ\text{C} + C} \qquad (1)$$

and the coefficients A, B, and C determined. These are listed in Table 2.

The measured isothermal $P-T-x$ data for the binary systems *n*-nonane + *n*-butanol, *n*-nonane + *n*-butylisocyanate, urethane + *n*-nonane, urethane + *n*-butanol, and urethane + *n*-butylisocyanate are given in Tables 3−7 and shown in Figures 3−7.

Table 4. Experimental $P-T-x$ Data for the System *n*-Nonane (1) + *n*-Butylisocyanate (2)

	P/kPa			
x_2	323.15 K	338.15 K	353.15 K	371.15 K
0.112	4.45	8.83	15.78	30.04
0.155	5.00	9.37	16.61	30.89
0.371	6.65	12.72	22.28	41.64
0.574	7.65	14.51	25.56	48.03
0.614	7.94	14.96	26.64	49.46
0.729	8.44	15.71	27.94	51.98
0.877	8.96	16.98	30.12	55.76

Table 5. Experimental $P-T-x$ Data for the System *n*-Nonane (1) + Urethane (2)

	P/kPa			
x_2	323.15 K	338.15 K	353.15 K	371.15 K
0.088	2.37	5.09	9.69	19.23
0.278	2.26	4.95	9.22	18.33
0.362	2.16	4.92	8.78	17.19
0.465	2.07	4.62	7.94	15.56
0.551	1.98	4.32	7.40	14.02
0.729	1.76	3.44	5.34	9.69
0.930	1.27	2.05	2.78	4.56

Table 6. Experimental $P-T-x$ Data for the System *n*-Butanol (1) + Urethane (2)

	P/kPa			
x_2	323.15 K	338.15 K	353.15 K	371.15 K
0.212	3.92	8.95	18.42	39.73
0.330	3.63	8.21	16.95	35.50
0.393	3.55	7.91	16.04	33.77
0.429	3.40	7.68	15.26	33.04
0.528	3.23	7.01	13.84	30.02
0.638	2.84	6.09	12.24	26.20
0.817	2.17	4.54	8.55	18.51

Table 7. Experimental $P-T-x$ Data for the System *n*-Butylisocyanate (1) + Urethane (2)

	P/kPa			
x_2	323.15 K	338.15 K	353.15 K	371.15 K
0.174	8.50	15.82	27.83	51.19
0.294	7.81	14.67	24.90	44.74
0.499	7.09	12.81	20.65	36.68
0.745	6.20	10.67	16.40	25.90
0.916	4.00	6.25	9.41	13.38

The results at (323.15, 338.15, 353.15, and 371.15) K were correlated simultaneously using UNIQUAC[11] and NRTL[12] models. The vapor phase was taken as an ideal gas. The binary interaction parameters are treated as temperature dependent. The temperature dependence of the binary interaction parameters is given by

$$\Delta a_{ij}/\mathrm{K} = a_{ij}/\mathrm{K} + b_{ij}(T/\mathrm{K}) \tag{2}$$

or

$$g_{ij}/(\mathrm{J{\cdot}mol^{-1}}) = a_{ij}/(\mathrm{J{\cdot}mol^{-1}}) + [b_{ij}/(\mathrm{J{\cdot}mol^{-1}{\cdot}K^{-1}})](T/\mathrm{K}) \tag{3}$$

for the UNIQUAC or NRTL model, respectively.

The parameters a_{ij} and b_{ij} have been determined through minimization of the objective function F

$$F = \sum_{j=1}^{N} \left(\frac{P_j^{\mathrm{cal}} - p_j^{\mathrm{exp}}}{P_j^{\mathrm{exp}}} \right)^2 \tag{4}$$

where N is the total number of the data points.

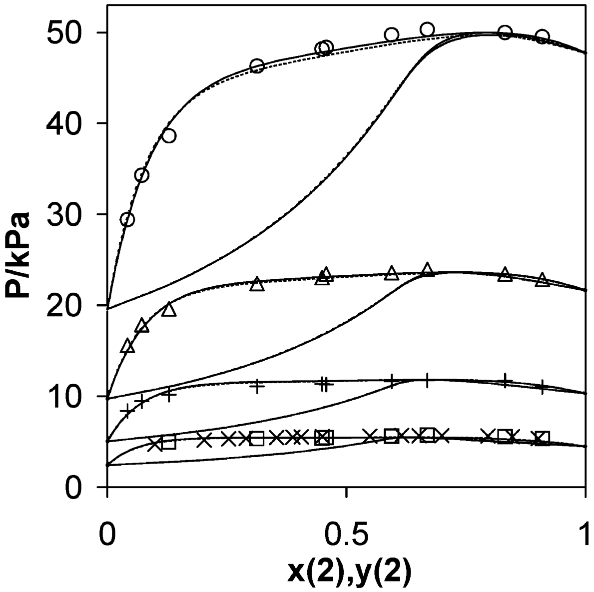

Figure 3. Experimental and calculated $P-x(y)$ for *n*-nonane (1) + *n*-butanol (2): □, experimental data this work at 323.15 K; ×, Heintz et al.[7] at 323.15 K; +, this work at 338.15 K; △, this work at 353.15 K; ○, this work at 371.15 K; solid line, UNIQUAC; dashed line, NRTL.

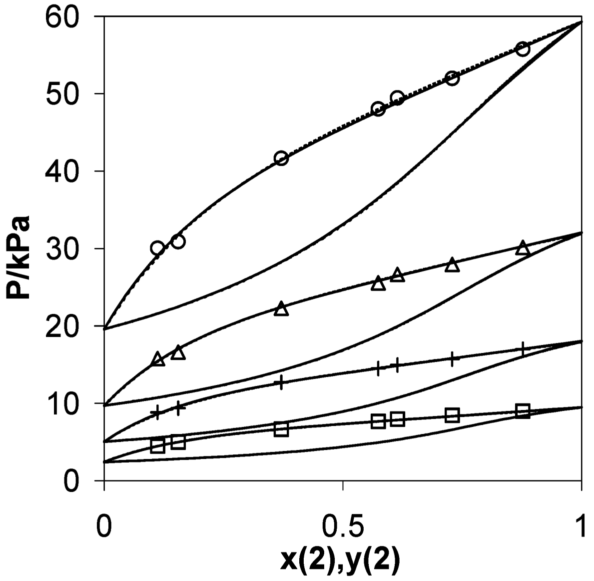

Figure 4. Experimental and calculated $P-x(y)$ for *n*-nonane (1) + *n*-butylisocyanate (2): □, experimental data at 323.15 K; +, at 338.15 K; △, at 353.15 K; ○, at 371.15 K; solid line, UNIQUAC; dashed line, NRTL.

For the NRTL model, the value of α (nonrandomness parameter) is fixed at 0.3 for all the systems. The binary parameters a_{ij} and b_{ij} for the UNIQUAC and NRTL models are listed in Table 8. The mean relative deviation between the calculated and the experimental pressures, σ, at various temperatures are also given. The pressures calculated with these parameters are shown as drawn lines in Figures 3−7.

In Figure 3, the available experimental data of Heintz et al.[7] for the *n*-nonane + *n*-butanol system at 323.15 are also shown. They are in good agreement with our values. It is noted that NRTL describes this system somewhat better than UNIQUAC.

For the system *n*-nonane + *n*-butylisocyanate, both models represent the experimental data quite well (see Figure 4 and Table 4). For this system, further experimen-

Table 8. Temperature-Dependent UNIQUAC and NRTL Interaction Paramaters and Mean Relative Deviation in *P*(σ) for: (a) *n*-Nonane + *n*-Butanol; (b) *n*-Nonane + *n*-Butylisocyanate; (c) Urethane + *n*-Nonane; (d) Urethane + *n*-Butanol; (e) Urethane + *n*-Butylisocyanate

UNIQUAC	*a*	*b*	*c*	*d*	*e*
a_{ij}/K	−12.3057	119.2915	1127.8574	258.9404	502.3778
a_{ji}/K	403.0158	13.5589	−2131.9517	−134.9977	37.1379
b_{ij}	−0.3559	−0.7266	−3.7180		−0.4619
b_{ji}	−0.0946	0.6961	7.0489		−0.5573
σ at 323.15 K	0.0316	0.0052	0.0437	0.0109	0.0055
σ at 338.15 K	0.0258	0.0102	0.0122	0.0149	0.0144
σ at 353.15 K	0.0141	0.0119	0.0567	0.0107	0.0176
σ at 371.15 K	0.0088	0.013	0.0163	0.0123	0.0085

NRTL	*a*	*b*	*c*	*d*	*e*
a_{ij}/[J/mol]	6582.2573	2480.8887	3450.4779	2087.3622	9659.7765
a_{ji}/[J/mol]	4838.3255	4374.0357	−3732.3827	643.2066	8220.1356
b_{ij}/[J/(mol K)]	−15.5691	−6.936	−17.0377		−17.2676
b_{ji}/[J/(mol K)]	1.1360	−5.4729	35.3880		−22.7441
σ at 323.15 K	0.021	0.0055	0.032	0.0212	0.0075
σ at 338.15 K	0.021	0.0080	0.0207	0.0208	0.0107
σ at 353.15 K	0.0121	0.0109	0.032	0.0145	0.0184
σ at 371.15 K	0.0116	0.0116	0.0092	0.0137	0.0087

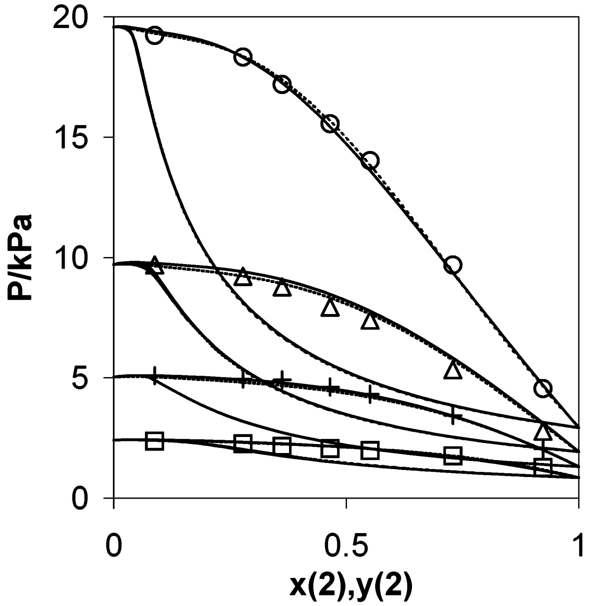

Figure 5. Experimental and calculated *P*−*x*(*y*) for *n*-nonane (1) + urethane (2): □, experimental data at 323.15 K; +, at 338.15 K; △, at 353.15 K; ○, at 371.15 K; solid line, UNIQUAC; dashed line, NRTL.

Table 9. Experimental *P*−*T*−*x* Data for the Reactive System Urethane (1) + *n*-Butanol (2) + *n*-Butylisocyanate (3), Experiment 1

time/min	x_1	x_2	x_3	*T*/K	*P*/kPa
4	0.186	0.417	0.397	332.19	7.91
6	0.276	0.379	0.345	334.59	8.61
8	0.438	0.295	0.267	337.48	8.59
9	0.532	0.254	0.214	338.42	8.20
10	0.579	0.234	0.187	338.03	7.86
11	0.610	0.218	0.172	336.80	7.26
12	0.641	0.201	0.159	335.71	6.88
13	0.674	0.178	0.148	334.87	6.47
15	0.704	0.166	0.130	332.60	5.89
17	0.723	0.155	0.122	331.31	5.81
20	0.752	0.141	0.107	329.67	5.85
25	0.780	0.129	0.090	327.42	5.02
40	0.818	0.116	0.066	326.34	4.10
60	0.839	0.104	0.057	325.03	3.54

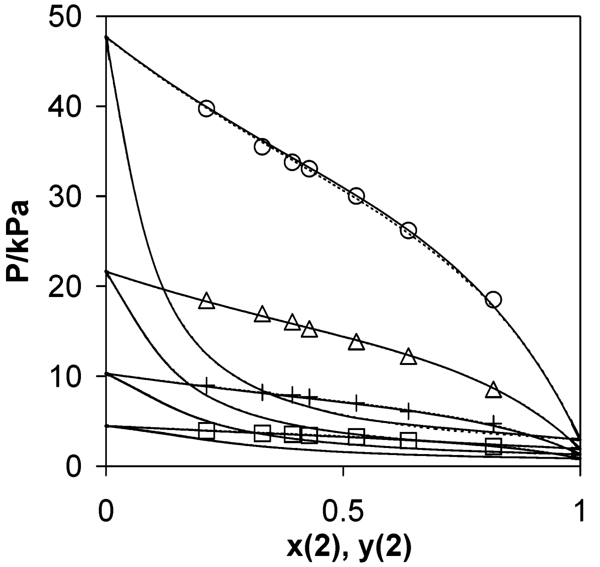

Figure 6. Experimental and calculated *P*−*x*(*y*) for *n*-butanol (1) + urethane (2): □, experimental data at 323.15 K; +, at 338.15 K; △, at 353.15 K; ○, at 371.15 K; solid line, UNIQUAC; dashed line, NRTL.

Table 10. Experimental *P*−*T*−*x* Data for the Reactive System Urethane (1) + *n*-Butanol (2) + *n*-Butylisocyanate (3), Experiment 2

time/min	x_1	x_2	x_3	*T*/K	*P*/kPa
6	0.779	0.105	0.116	339.39	7.75
7	0.818	0.087	0.094	338.35	6.99
10	0.830	0.086	0.084	338.00	6.67
12	0.838	0.082	0.080	337.90	6.31
15	0.879	0.053	0.069	337.74	5.81
18	0.878	0.058	0.065	337.78	5.76
21	0.902	0.031	0.067	337.75	5.80
25	0.892	0.049	0.058	337.77	5.45
30	0.906	0.033	0.062	337.74	5.65
35	0.908	0.036	0.056	337.74	5.26
40	0.920	0.025	0.055	337.75	5.27
45	0.913	0.027	0.060	337.72	5.38
50	0.916	0.026	0.058	337.74	5.35
55	0.917	0.030	0.053	337.71	5.39
60	0.927	0.023	0.050	337.74	5.07

tal *T*−*x* data exist at 50 kPa.[8] These literature values at constant pressure are compared with the calculated values from UNIQUAC and NRTL parameters obtained from our *P*−*x* data (see Table 8). The agreement is very good as shown in Figure 8.

The results for the system urethane + *n*-nonane are given in Figure 5 and Table 5. It is observed that the

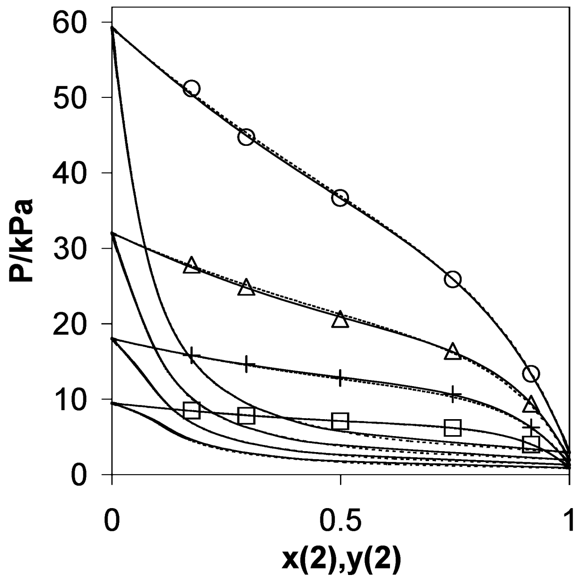

Figure 7. Experimental and calculated $P-x(y)$ for *n*-butylisocyanate (1) + urethane (2): □, experimental data at 323.15 K; +, at 338.15 K; △, at 353.15 K; ○, at 371.15 K; solid line, UNIQUAC; dashed line, NRTL.

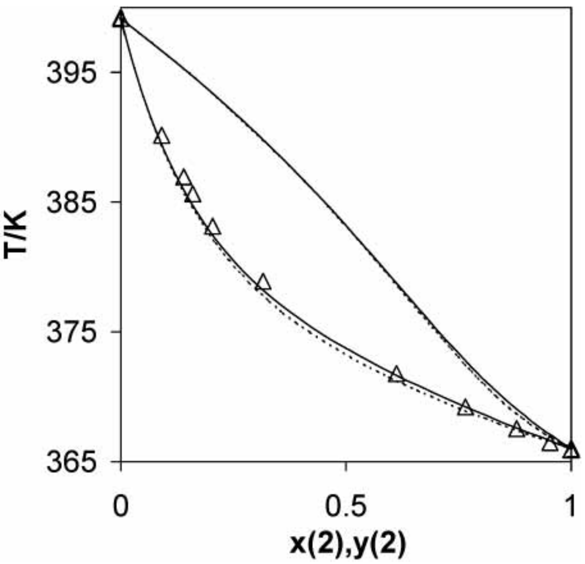

Figure 8. $T-x$ data for the system *n*-nonane (1) + *n*-butylisocyanate (2) at 50 kPa: △, experimental data Hamann[8]; solid line, UNIQUAC; dashed line, NRTL.

Table 11. Experimental *P*−*x* Data for the Reactive System Urethane (1) + *n*-Butanol (2) + *n*-Butylisocyanate (3), Experiment 3

time/min	x_1	x_2	x_3	T/K	P/kPa
9	0.854	0.109	0.048	371.34	17.39
12	0.883	0.074	0.038	371.23	13.09
15	0.899	0.078	0.027	371.27	13.10
20	0.915	0.058	0.020	371.29	10.23
25	0.936	0.039	0.010	371.66	8.13
40	0.947	0.028	0.011	371.62	7.28

results of the NRTL model are a little bit better than those of the UNIQUAC model.

For the system urethane + *n*-butanol, both the UNIQUAC and NRTL models are able to reproduce the experimental data quite well with only two parameters (see Figure 6 and Table 6).

The urethane + *n*-butylisocyanate system shown in Figure 7 and Table 7 can also be represented with UNIQUAC and NRTL models quite well.

Table 12. Experimental and Predicted (UNIQUAC and NRTL) *P*−*T*−*x* Data for the System Urethane (1) + *n*-Butanol (2) + *n*-Nonane (3)

			P/kPa			
x_1	x_2	x_3	323.15 K	338.15 K	353.15 K	371.15 K
			Experimental Data			
0.221	0.179	0.600	4.04	8.36	15.69	31.33
0.142	0.471	0.387	4.71	9.98	19.58	40.55
0.123	0.410	0.467	4.78	10.10	19.75	40.60
			UNIQUAC-Predicted Data			
0.221	0.179	0.600	4.07	8.20	15.31	29.83
0.142	0.471	0.387	4.53	9.73	19.30	40.41
0.123	0.410	0.467	4.62	9.84	19.37	40.17
			NRTL-Predicted Data			
0.221	0.179	0.600	4.14	8.58	16.46	33.03
0.142	0.471	0.387	4.89	10.42	20.54	42.48
0.123	0.410	0.467	4.92	10.45	20.50	42.24

Table 13. Experimental and Calculated Concentration of *n*-Butylisocyanate in the Urethane (1) + *n*-Butanol (2) + *n*-Butylisocyanate (3) System, Experiment 1

time/min	x_3 (exp)	x_3 (calc)	absolute deviation
4	0.397	0.393	0.004
6	0.345	0.346	−0.002
8	0.267	0.264	0.003
9	0.214	0.215	−0.001
10	0.187	0.192	−0.004
11	0.172	0.176	−0.004
12	0.159	0.160	−0.001
13	0.148	0.143	0.005
15	0.130	0.128	0.002
17	0.122	0.118	0.004
20	0.107	0.103	0.004
25	0.090	0.088	0.002
40	0.066	0.069	−0.003
60	0.057	0.058	−0.002

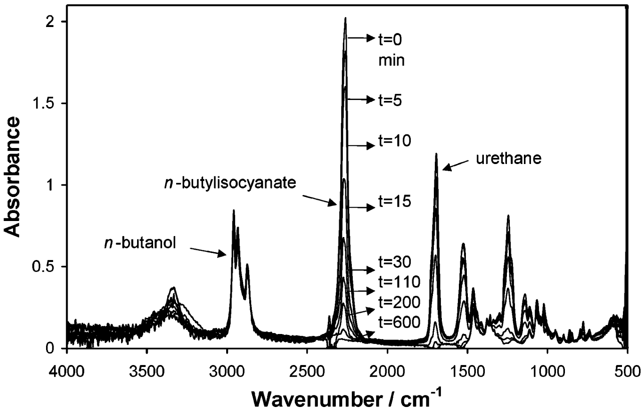

Figure 9. IR spectra of the reactive mixture urethane (1) + *n*-butanol (2) + *n*-butylisocyanate (3) at various time intervals.

The system *n*-butylisocyanate + *n*-butanol is a reactive system. Both components react to form the additive compound urethane. The spectroscopic measurements in the liquid phase (see Figure 9) show that for an almost equimolar mixture at 323.15 K the composition changes even after 4 h. It is expected that even when the reaction in the liquid phase has not been completed there exists an equilibrium between vapor and liquid phases in the particular span of time and hence a VLE may be considered in which all the three components *n*-butylisocyanate, *n*-butanol, and urethane are present. The $P-T-x$ data for such ternary systems are given in Tables 10−12. The results have been correlated with UNIQUAC and NRTL models and the binary interaction parameters for *n*-butylisocyanate + *n*-butanol determined as the binary

Table 14. Experimental and Calculated Concentration of *n*-Butylisocyanate in the Urethane (1) + *n*-Butanol (2) + *n*-Butylisocyanate (3) System, Experiment 2

time/min	x_3 (exp)	x_3 (calc)	absolute deviation
6	0.116	0.119	−0.003
7	0.094	0.100	−0.006
10	0.084	0.094	−0.010
12	0.080	0.090	−0.010
15	0.069	0.070	−0.001
18	0.065	0.071	−0.006
21	0.067	0.059	0.008
25	0.058	0.063	−0.005
30	0.062	0.057	0.005
35	0.056	0.056	0.001
40	0.055	0.050	0.005
45	0.060	0.053	0.007
50	0.058	0.051	0.006
55	0.053	0.051	0.002
60	0.050	0.046	0.004

Table 15. Experimental and Calculated Concentration of *n*-Butylisocyanate in the Urethane (1) + *n*-Butanol (2) + *n*-Butylisocyanate (3) System, Experiment 3

time/min	x_3 (exp)	x_3 (calc)	absolute deviation
9	0.048	0.055	−0.007
12	0.038	0.036	0.002
15	0.027	0.031	−0.003
20	0.020	0.020	0.000
25	0.010	0.007	0.003
40	0.011	0.002	0.009

interaction parameters for *n*-butanol + urethane and butylisocyanate + urethane are already available (see Table 8). The calculated vapor pressures are also shown in Tables 9−11. By use of the reaction equation

$$C_4H_9{-}N{=}C{=}O + C_4H_9{-}OH \rightleftharpoons C_4H_9{-}(N{-}H){-}(C{=}O){-}O{-}C_4H_9 \qquad (5)$$

and the experimental concentration of the product urethane at any time, the degree of reaction (ξ) has been calculated. The concentration of *n*-butylisocyanate at various times calculated with this model is compared with the experimental values at 323.15 K, 338.15 K, and 371.15 K in Tables 13−15, respectively.

To check the accuracy of the prediction of the vapor−liquid equilibrium of a ternary system from the binary parameters, the vapor pressures of the ternary system urethane + *n*-butanol + *n*-nonane were measured at (323.15, 338.15, 353.15, and 371.15) K and are shown in Table 12. The vapor pressures of this ternary system have been predicted using the binary interaction parameter for urethane + *n*-butanol, urethane + *n*-nonane, and *n*-nonane + *n*-butanol (from Table 8). The predicted values are compared from the experimental values are also listed in parts a, b, and c of Table 12. The agreement between the experimental and predicted vapor pressure is very good.

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