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Vapor−Liquid Equilibria and Excess Enthalpies for Binary Mixtures of *N*-Methylacetamide with 1-Octene, Dipropylamine, Triethylamine, Pyridine, and Methanol at 125 °C

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Isothermal $P-x$ data and excess enthalpies have been measured at approximately 125 °C for the binary mixtures of *N*-methylacetamide with 1-octene, dipropylamine, triethylamine, pyridine, and methanol. For each binary system the vapor−liquid equilibrium and excess enthalpy data were correlated simultaneously by the temperature dependent five-parameter NRTL model.

1. Introduction

In previous papers we have presented experimental results on vapor−liquid equilibria and excess enthalpies for various *N*-methylacetamide-containing binary systems (Aim and De Haan, 1996; De Haan et al., 1995, De Haan and Gmehling, 1996). This systematic study on vapor−liquid equilibria and excess enthalpies in amide-containing systems is continued in the present paper with pressure−composition $P-x$ and excess enthalpy H^E data for binary mixtures of *N*-methylacetamide with 1-octene, dipropylamine, triethylamine, pyridine, and methanol at 125 °C. For all four systems no experimental data are available in the literature (Gmehling and Onken, 1977).

2. Experimental Section

2.1. Degassing and Purification. 1-Octene, dipropylamine, triethylamine, pyridine, methanol, and *N*-methylacetamide were degassed and purified according to the vacuum rectification procedure described by Fischer and Gmehling (1994). The final purity of the components, determined by gas chromatography and Karl-Fischer titration, was better than 99.99%.

2.2. $P-x$ Measurements. The $P-x$ results were obtained with the static apparatus developed and described in detail by Fischer and Gmehling (1994) and Kolbe and Gmehling (1985). The degassed, pure components were filled into evacuated piston injectors. Then one of the pure components was introduced into the evacuated thermostated equilibrium cell, and the pressure and the temperature were measured after equilibrium was reached. Before and after every injection, the liquid volume left in the piston injectors was determined at the chosen conditions (P = 50 bar, t = 25.0 to 40.0 °C). To check for complete degassing, the vapor pressure was measured again after a second injection of the pure component. As shown previously (Fisher and Gmehling, 1994; Kolbe and Gmehling, 1985), the accuracy of the respective measure-

Table 1. Vapor−Liquid Equilibrium Data, Pressure P, Liquid Phase Mole Fraction x_1, Calculated Vapor Mole Fraction y_1, and Calculated Difference in Pressure ΔP, for the System 1-Octene (1) + *N*-Methylacetamide (2) at 125.4 °C

experimental		NRTL	
x_1	P/kPa	ΔP/kPa[a]	$y_{1,\text{calc}}$
0.0000	6.55	0.00	0.0000
0.0009	7.50	−0.08	0.1367
0.0017	8.58	0.09	0.2295
0.0025	9.34	−0.04	0.3039
0.0065	13.74	−0.02	0.5272
0.0115	18.94	−0.03	0.6587
0.0168	24.33	0.13	0.7336
0.0211	28.29	0.07	0.7725
0.0251	31.87	0.07	0.7988
0.0328	38.48	0.21	0.8340
0.0409	45.00	0.47	0.8583
0.0514	52.35	0.46	0.8794
0.0615	58.78	0.53	0.8934
0.0742	65.87	0.50	0.9059
0.0886	72.95	0.55	0.9159
0.1034	79.12	0.46	0.9233
0.1297	88.01	0.27	0.9323
0.1690	97.50	−0.17	0.9404
0.2310	106.45	−1.02	0.9472
0.2619	109.24	−1.36	0.9492
0.3230	112.93	−1.72	0.9518
0.3370	113.10	−1.90	0.9520
0.3563	113.76	−1.22	0.9520
0.4234	114.92	−0.07	0.9520
0.4936	115.45	0.46	0.9520
0.5933	129.17	0.85	0.9520
0.7172	115.96	0.97	0.9520
0.8400	116.24	1.25	0.9520
0.9040	116.61	1.63	0.9520
0.9194	116.57	1.53	0.9525
0.9351	116.80	1.55	0.9557
0.9457	116.79	1.40	0.9588
0.9554	116.84	1.34	0.9625
0.9646	116.82	1.65	0.9670
0.9733	116.57	0.98	0.9724
0.9789	116.31	0.76	0.9767
0.9836	116.01	0.51	0.9807
0.9878	116.01	0.60	0.9849
0.9915	115.78	0.47	0.9889
0.9941	115.26	0.04	0.9920
0.9961	115.19	0.05	0.9946
0.9973	115.01	−0.07	0.9962
1.0000	114.95	0.00	1.0000

[a] $\Delta P = P_{\text{exp}} - P_{\text{calc}}$.

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Table 2. Vapor−Liquid Equilibrium Data, Pressure P, Liquid Phase Mole Fraction x_1, Calculated Vapor Mole Fraction y_1, and Calculated Difference in Pressure ΔP, for the System Dipropylamine (1) + N-Methylacetamide (2) at 125.4 °C

experimental		NRTL	
x_1	P/kPa	ΔP/kPa[a]	$y_{1,\mathrm{calc}}$
0.0000	6.29	0.00	0.0000
0.0071	10.98	−0.02	0.4320
0.0137	15.18	−0.05	0.5923
0.0187	18.29	−0.06	0.6631
0.0246	21.98	0.06	0.7197
0.0312	25.98	0.19	0.7633
0.0371	29.40	0.25	0.7918
0.0439	33.18	0.28	0.8167
0.0520	37.57	0.35	0.8391
0.0600	41.65	0.34	0.8562
0.0692	46.19	0.36	0.8714
0.0794	50.99	0.37	0.8846
0.0969	58.54	0.24	0.9014
0.1108	64.14	0.17	0.9112
0.1333	72.35	−0.05	0.9230
0.1581	80.29	−0.45	0.9323
0.1953	90.68	−0.93	0.9421
0.2206	96.75	−1.28	0.9469
0.2793	108.40	−2.04	0.9548
0.3552	120.66	−1.73	0.9612
0.4100	126.65	−2.17	0.9644
0.4694	132.08	−2.14	0.9670
0.5138	135.33	−2.03	0.9685
0.5400	137.03	−1.91	0.9692
0.5759	139.16	−1.61	0.9702
0.6094	140.96	−1.26	0.9710
0.6442	142.54	−0.97	0.9717
0.6750	143.92	−0.57	0.9723
0.7112	145.51	−0.01	0.9730
0.7523	147.11	0.51	0.9738
0.7934	148.79	1.08	0.9747
0.8254	149.93	1.27	0.9756
0.8672	151.53	1.35	0.9775
0.9020	152.89	1.09	0.9799
0.9366	154.45	0.62	0.9838
0.9636	156.29	0.51	0.9887
0.9789	157.20	0.17	0.9926
0.9944	158.28	−0.10	0.9978
1.0000	158.89	0.00	1.0000

[a] $\Delta P = P_{\mathrm{exp}} - P_{\mathrm{calc}}$.

Table 3. Vapor−Liquid Equilibrium Data, Pressure P, Liquid Phase Mole Fraction x_1, Calculated Vapor Mole Fraction y_1, and Calculated Difference in Pressure ΔP, for the System Triethylamine (1) + N-Methylacetamide (2) at 125.4 °C

experimental		NRTL	
x_1	P/kPa	ΔP/kPa[a]	$y_{1,\mathrm{calc}}$
0.0000	6.33	0.00	0.0000
0.0060	16.45	−0.21	0.6226
0.0127	27.40	−0.24	0.7740
0.0193	37.63	−0.28	0.8362
0.0266	48.42	−0.25	0.8733
0.0338	58.37	−0.34	0.8957
0.0417	68.76	−0.36	0.9120
0.0497	78.85	−0.20	0.9236
0.0584	89.19	0.01	0.9328
0.0677	100.01	0.67	0.9401
0.0781	110.77	0.88	0.9463
0.0894	121.49	1.00	0.9515
0.1128	141.43	1.49	0.9589
0.1297	153.75	1.61	0.9627
0.1541	169.29	1.84	0.9667
0.1848	185.37	1.86	0.9702
0.2125	197.16	1.66	0.9724
0.2386	206.18	1.17	0.9741
0.2830	218.68	0.62	0.9762
0.3492	230.83	−1.11	0.9783
0.3550	231.26	−1.65	0.9784
0.4052	237.25	−2.72	0.9794
0.4707	242.75	−3.47	0.9804
0.5155	245.47	−3.53	0.9808
0.5695	247.70	−3.40	0.9811
0.6397	250.03	−1.73	0.9813
0.6923	251.42	−0.35	0.9813
0.7544	252.65	0.87	0.9813
0.8027	253.67	1.90	0.9813
0.8331	254.46	2.69	0.9813
0.8788	255.49	3.35	0.9816
0.9165	256.82	3.06	0.9829
0.9382	257.87	2.62	0.9846
0.9530	258.54	1.98	0.9865
0.9678	259.58	1.49	0.9891
0.9755	260.22	1.25	0.9910
0.9843	261.06	1.02	0.9936
0.9907	261.29	0.44	0.9959
0.9960	261.81	0.27	0.9981
1.0000	262.06	0.00	1.0000

[a] $\Delta P = P_{\mathrm{exp}} - P_{\mathrm{calc}}$.

ments in the equilibrium cell was ±0.0001 for the mixture mole fraction, ±0.02 K for the temperature, and ±0.1 kPa for the pressure. After the pure component vapor pressure had been determined, the second component was injected. The equilibrium was established after approximately 30 min, and the pressure was measured for the given composition. This procedure was continued until approximately 60 vol % of the second component was attained. Then the equilibrium cell was emptied, evacuated, and the same procedure followed, starting with the pure component 2. After the second series, the whole concentration range is covered and the two independent parts of the isotherm should overlap.

2.3. Excess Enthalpies. A commercially available flow calorimeter from Hart Scientific (model 7501) was used to measure the excess enthalpies at isothermal conditions by compensating for the heat of mixing effect. The calorimeter consists of two solvent pumps, an adiabatic flow cell in a constant temperature bath, and a back-pressure regulator to prevent evaporation. A detailed description of the calorimeter and the experimental procedures has been given by Gmehling (1993). The uncertainty in H^{E} was estimated to be less than ±1%, as shown previously by Gmehling (1993).

3. Results

3.1. P−x and H^{E} Data. The concentration of the liquid phase was obtained from the feed composition by an iterative flash calculation. The volume of the liquid mixture was calculated from the pure component volumes at the pressure and temperature of the measurement, neglecting the excess volume. The difference of the known volume of the equilibrium cell and the volume of the liquid mixture is used to calculate the number of moles in the vapor phase assuming ideal behavior of the vapor phase. At low pressures this assumption causes only negligible errors. The raw results (isothermal $P-x$ data) are then used to fit the parameters of a Legendre polynomial, and finally, the composition of the liquid phase is recalculated using the calculated composition of the vapor phase.

This iterative procedure converges after a few cycles. For our measurements, the corrections to obtain the liquid mole

Table 4. Vapor−Liquid Equilibrium Data, Pressure *P*, Liquid Phase Mole Fraction x_1, Calculated Vapor Mole Fraction y_1, and Calculated Difference in Pressure ΔP, for the System Pyridine (1) + *N*-Methylacetamide (2) at 125.4 °C

experimental		NRTL	
x_1	P/kPa	ΔP/kPa[a]	$y_{1,\mathrm{calc}}$
0.0000	6.38	0.00	0.0000
0.0094	8.25	0.11	0.2240
0.0181	9.91	0.15	0.3588
0.0256	11.36	0.21	0.4432
0.0354	13.23	0.25	0.5259
0.0470	15.38	0.27	0.5977
0.0578	17.36	0.27	0.6482
0.0721	19.98	0.29	0.6992
0.0837	22.10	0.32	0.7314
0.0982	24.63	0.26	0.7637
0.1116	27.00	0.25	0.7878
0.1260	29.43	0.16	0.8092
0.1452	32.74	0.13	0.8323
0.1713	37.00	−0.05	0.8567
0.1993	41.48	−0.24	0.8768
0.2315	46.51	−0.45	0.8946
0.2956	55.93	−0.98	0.9196
0.3541	64.02	−1.42	0.9351
0.4282	73.59	−1.85	0.9491
0.4645	77.98	−2.03	0.9544
0.4887	80.99	−1.95	0.9576
0.5618	89.21	−2.06	0.9656
0.6184	95.23	−1.97	0.9708
0.6838	101.94	−1.67	0.9759
0.7322	106.70	−1.42	0.9793
0.7901	112.45	−0.93	0.9833
0.8460	117.81	−0.63	0.9871
0.8820	121.39	−0.41	0.9897
0.9126	124.43	−0.34	0.9920
0.9389	127.14	−0.28	0.9942
0.9611	129.57	−0.21	0.9961
0.9702	130.39	−0.40	0.9970
0.9784	131.33	−0.37	0.9978
0.9856	132.35	−0.17	0.9985
0.9917	133.15	−0.13	0.9991
0.9969	133.86	0.01	0.9997
0.9990	133.86	−0.24	0.9999
1.0000	134.22	0.00	1.0000

[a] $\Delta P = P_{\mathrm{exp}} - P_{\mathrm{calc}}$.

Table 5. Vapor−Liquid Equilibrium Data, Pressure *P*, Liquid Phase Mole Fraction x_1, Calculated Vapor Mole Fraction y_1, and Calculated Difference in Pressure ΔP, for the System Methanol (1) + *N*-Methylacetamide (2) at 125.4 °C

experimental		NRTL	
x_1	P/kPa	ΔP/kPa[a]	$y_{1,\mathrm{calc}}$
0.0000	6.65	0.00	0.0000
0.0242	17.89	0.18	0.6335
0.0454	28.14	0.47	0.7706
0.0653	37.60	0.35	0.8333
0.0847	47.22	0.40	0.8702
0.1002	55.16	0.53	0.8909
0.1217	66.24	0.55	0.9115
0.1425	77.05	0.37	0.9261
0.1652	89.10	0.12	0.9382
0.1859	100.42	−0.08	0.9467
0.2074	112.71	−0.03	0.9539
0.2275	124.22	−0.27	0.9595
0.2481	136.07	−0.73	0.9642
0.2707	150.90	−0.55	0.9687
0.2918	163.19	−0.72	0.9722
0.3559	204.28	−1.83	0.9802
0.4158	245.14	−3.11	0.9855
0.5079	312.73	−5.36	0.9909
0.5667	359.13	−6.62	0.9933
0.6517	430.11	−8.24	0.9958
0.7159	497.15	1.59	0.9971
0.7650	540.20	0.27	0.9980
0.8122	581.15	−1.66	0.9986
0.8313	597.64	−2.45	0.9988
0.8523	615.94	−3.02	0.9990
0.8737	635.24	−2.75	0.9992
0.8891	648.28	−3.24	0.9993
0.9058	662.31	−3.70	0.9995
0.9230	677.58	−3.09	0.9996
0.9319	684.88	−3.27	0.9996
0.9408	692.30	−3.24	0.9997
0.9540	703.70	−2.63	0.9998
0.9737	720.77	−1.25	0.9999
0.9910	735.05	−2.69	1.0000
0.9949	738.81	0.56	1.0000
0.9966	739.83	0.32	1.0000
0.9983	740.94	0.16	1.0000
1.0000	742.03	0.00	1.0000

[a] $\Delta P = P_{\mathrm{exp}} - P_{\mathrm{calc}}$.

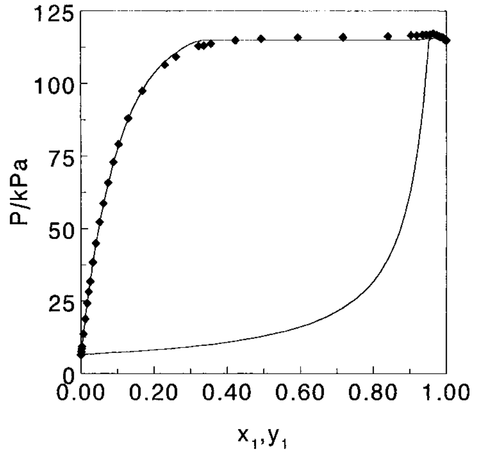

Figure 1. Experimental $P-x$ data (◆) and NRTL fit for 1-octene (1) + *N*-methylacetamide (2) at 125.4 °C.

fraction from the total composition are smaller than 0.0003, which is of the magnitude of the accuracy of the total composition. The experimental $P-x$ values are listed in Tables 1−5. The excess enthalpies measured at 125.0 °C for the five binary systems are listed in Table 6.

3.2. Modeling. For each binary system, the interaction parameters have been fitted simultaneously to the measured $P-x$ data and excess enthalpies for the NRTL model (Gmehling and Kolbe, 1992) with the help of the Simplex−Nelder−Mead method (Nelder and Mead, 1965) using the following linear temperature dependency for the interaction parameters, g_{ij}

$$g_{ij} = g^{0}_{ij} + g^{1}_{ij}T/\mathrm{K} \tag{1}$$

and the following objective function:

$$F = \sum((P - P_{\mathrm{calcd}})/P)^2 + \sum((H^{\mathrm{E}} - H^{\mathrm{E}}_{\mathrm{calcd}})/H^{\mathrm{E}})^2 \tag{2}$$

The obtained parameters, which were used to calculate the pressure, vapor composition, and excess enthalpies, are listed in Table 7. The initial value of 0.300 for the nonrandomness parameter α_{ij} was not optimized unless the value of the objective function, F, decreased significantly. In Tables 1−5, the deviations $P - P_{\mathrm{calcd}}$ and the calculated vapor compositions are given. The Antoine constants used for the calculation of the pure component vapor pressures are listed in Table 8.

Table 6. Excess Enthalpies H^E and Liquid Mole Fraction x_1 for Binary Mixtures with N-Methylacetamide at 125 °C

x_1	H^E/(J mol^{-1})	x_1	H^E/(J mol^{-1})	x_1	H^E/(J mol^{-1})
		1-Octene (1) + N-Methylacetamide (2)			
0.0123	40.8	0.2078	575.6	0.5319	931.8
0.0250	88.8	0.2451	646.5	0.5937	942.9
0.0513	175.1	0.2849	711.0	0.6608	941.2
0.0791	259.2	0.3275	779.0	0.7340	923.4
0.1085	342.8	0.3732	825.3	0.8142	864.7
0.1397	421.6	0.4222	871.8	0.9025	717.2
0.1727	500.5	0.4749	909.1	0.9500	524.5
		Dipropylamine (1) + N-Methylacetamide (2)			
0.0141	−10.7	0.2315	−38.3	0.5662	149.5
0.0286	−17.2	0.2716	−28.7	0.6266	204.4
0.0585	−26.4	0.3140	−12.2	0.6911	260.8
0.0898	−34.4	0.3587	3.9	0.7602	315.9
0.1227	−38.8	0.4061	31.3	0.8343	359.7
0.1572	−43.1	0.4562	65.0	0.9140	339.6
0.1934	−41.9	0.5095	105.4	0.9562	243.0
		Triethylamine (1) + N-Methylacetamide (2)			
0.0139	24.3	0.2283	371.6	0.5617	695.1
0.0281	49.2	0.2680	425.9	0.6223	722.6
0.0575	102.6	0.3101	474.5	0.6872	739.6
0.0884	156.5	0.3545	524.6	0.7569	740.7
0.1207	210.6	0.4017	570.2	0.8318	710.7
0.1548	262.6	0.4517	613.9	0.9126	604.6
0.1906	318.6	0.5050	656.7	0.9554	457.2
		Pyridine (1) + N-Methylacetamide (2)			
0.0238	13.2	0.3385	157.4	0.6892	240.1
0.0476	24.6	0.3879	175.9	0.7403	237.0
0.0955	47.8	0.4375	193.2	0.7917	223.9
0.1436	70.3	0.4873	208.2	0.8434	200.2
0.1920	94.7	0.5374	222.9	0.8953	158.3
0.2406	114.3	0.5877	231.4	0.9475	96.9
0.2894	135.6	0.6384	239.1	0.9737	52.2
		Methanol (1) + N-Methylacetamide (2)			
0.0461	−39.3	0.5037	−271.2	0.8147	−206.4
0.0902	−74.8	0.5568	−275.6	0.8497	−181.4
0.1731	−134.0	0.6066	−275.4	0.8829	−152.7
0.2496	−181.4	0.6533	−271.7	0.9144	−121.8
0.3203	−216.6	0.6973	−260.2	0.9443	−83.5
0.3858	−242.7	0.7387	−246.3	0.9728	−44.2
0.4468	−260.3	0.7778	−228.5	0.9866	−22.4

Table 7. Fitted NRTL Nonrandomness Parameter α_{ij} and Interaction Parameters $g_{ij} = g^0_{ij} + g^1_{ij}T$/K

system (1) + (2)	α_{12}	g^0_{12}/(kJ mol^{-1})	g^0_{21}/(kJ mol^{-1})	g^1_{12}/(J mol^{-1} K^{-1})	g^1_{21}/(J mol^{-1} K^{-1})
1-octene + N-methylacetamide	0.398	9.622	4.323	−3.763	−0.669
dipropylamine + N-methylacetamide	0.470	5.253	−1.015	2.396	8.194
triethylamine + N-methylacetamide	0.432	8.979	2.422	−2.898	2.724
pyridine + N-methylacetamide	0.300	2.425	−0.407	6.627	−3.935
methanol + N-methylacetamide	0.300	0.058	0.845	7.389	−7.643

Table 8. Constants for the Antoine Vapor Pressure Equation, log(P/kPa) = A − B/(t/°C + C)

component	A^a	B	C
1-octene	6.0667	1355.8	213.02
dipropylamine	6.3211	1415.4	218.00
triethylamine	6.0217	1251.8	222.00
pyridine	6.1639	1373.8	214.98
methanol	7.2039	1582.3	239.73
N-methylacetamide	5.7008	1348.3	150.22

[a] Adjusted to the measured pure component vapor pressures.

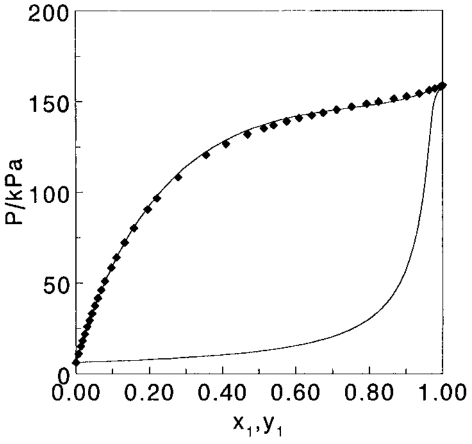

Figure 2. Experimental P−x data (◆) and NRTL fit for dipropylamine (1) + N-methylacetamide (2) at 125.4 °C.

4. Discussion and Conclusions

Figure 1 compares the experimental P−x data and the P−x−y curves calculated using the fitted NRTL parameters for the 1-octene + N-methylacetamide system, which shows a pressure maximum azeotrope. The same comparison is made in Figures 2−5 for the dipropylamine, triethylamine, pyridine, and methanol-containing systems. In general, good agreement is obtained between the experimental P−x data and the calculated P−x−y curves for all five systems. However, in the 1-octene- and triethylamine-containing systems, the NRTL model predicts liquid−liquid immiscibility, which is not observed experimentally. For both

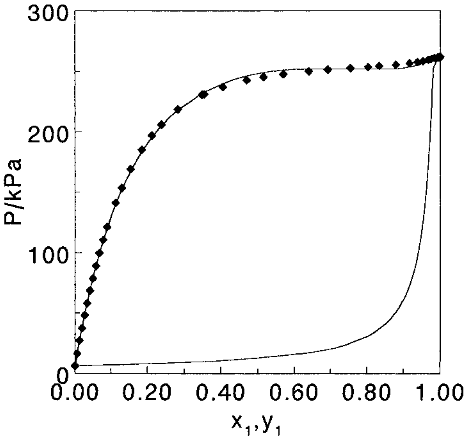

Figure 3. Experimental $P-x$ data (◆) and NRTL fit for triethylamine (1) + *N*-methylacetamide (2) at 125.4 °C.

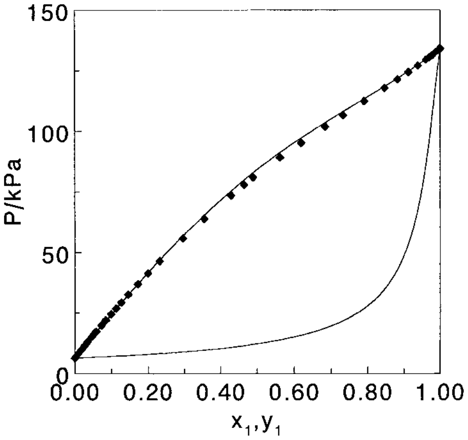

Figure 4. Experimental $P-x$ data (◆) and NRTL fit for pyridine (1) + *N*-methylacetamide (2) at 125.4 °C.

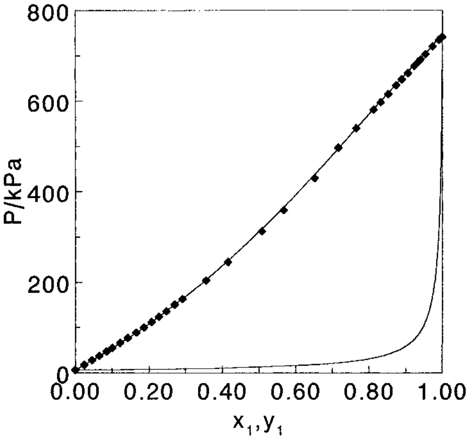

Figure 5. Experimental $P-x$ data (◆) and NRTL fit for methanol (1) + *N*-methylacetamide (2) at 124.4 °C.

systems this is more clearly illustrated by Figure 6, in which the experimental H^E results at 125 °C are compared with the excess enthalpies calculated using the fitted NRTL parameters. Figures 6 and 7 show that for the pyridine, dipropylamine, and methanol-containing systems, the excess enthalpies are well represented over the whole concentration range by the NRTL model using linear temperature dependent parameters. The methanol + *N*-methylacetamide system shows negative excess enthalpies due to hydrogen bonding, which is confirmed by the negative deviation from Raoults law observed in Figure 5.

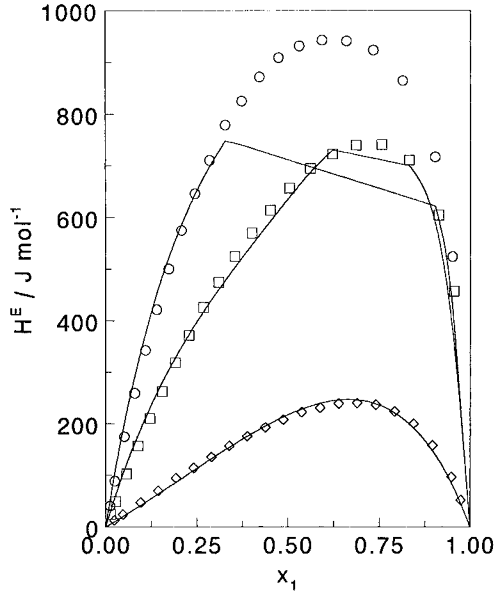

Figure 6. Experimental H^E data and NRTL fit for *N*-methylacetamide (2) with 1-octene (1) (○), triethylamine (1) (□), and pyridine (1) (◇) at 125.0 °C.

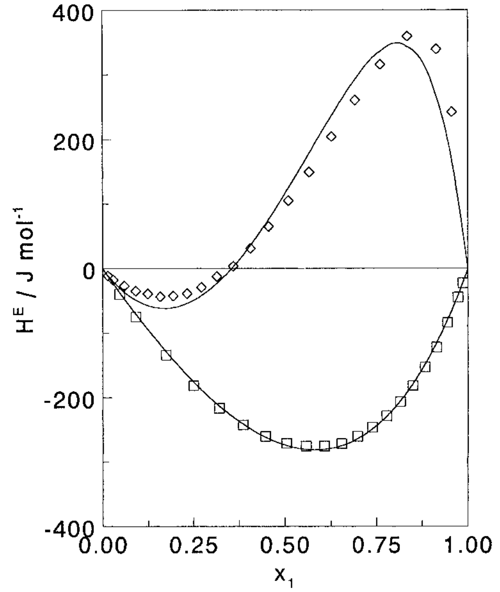

Figure 7. Experimental H^E data and NRTL fit for *N*-methylacetamide (2) with dipropylamine (1) (◇) and methanol (1) (□) at 125.0 °C.

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