Vapor−Liquid Equilibria and Excess Enthalpies for Octane + *N*-Methylacetamide, Cyclooctane + *N*-Methylacetamide, and Octane + Acetic Anhydride 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 octane + *N*-methylacetamide, cyclooctane + *N*-methylacetamide, and octane + acetic anhydride. For each binary system linear temperature dependent interaction parameters were fitted to experimental data using the NRTL model. Activity coefficients at infinite dilution were derived from the $P-x$ data at low concentrations using a flexible Legendre polynomial.

1. Introduction

At present the application of group contribution methods such as UNIFAC (Fredenslund et al., 1977; Hansen et al., 1991), modified UNIFAC (Dortmund, Lyngby) (Gmehling et al., 1993; Larsen et al., 1987; Weidlich and Gmehling, 1987), and ASOG (Kojima and Tochigi, 1979; Tochigi et al., 1990) for the prediction of phase equilibria and excess enthalpies in amide- or anhydride-containing systems is inhibited by the unavailability of reliable interaction parameters between the various groups. The main reason for this situation is the lack of experimental data for these kinds of systems (Christensen et al., 1984; Gmehling and Onken, 1977; Rasmussen et al., 1979; Tiegs and Gmehling, 1986).

In this paper we have focused on binary mixtures of alkanes and cycloalkanes with amides and anhydrides. Up to now the only available data for binary mixtures of alkanes and cycloalkanes with amides have been reported by Shorokhodova and Kogan (1973) for the cyclohexane + ε-caprolactam system. Schmelzer and Pusch (1995) showed that these data are unreliable and presented additional pressure−composition $P-x$ data for various amide + alkane systems, including octane + *N*-methylacetamide at 90 and 110 °C. However, it appeared that most of his $P-x$ data have been measured in the two-phase region. For binary mixtures of alkanes and cycloalkanes with anhydrides, liquid−liquid equilibrium data have been reported for heptane + acetic anhydride by Nagarajan et al. (1980) and for cyclohexane + acetic anhydride by Jones et al. (1928). Furthermore, Jones (1962) has reported some vapor−liquid equilibrium data for cyclohexane + acetic anhydride.

This overview illustrates that only limited experimental data are available for binary mixtures of alkanes and cycloalkanes with amides and anhydrides. Therefore, we have measured pressure−composition $P-x$, activity coeffient at infinite dilution γ^∞, and excess enthalpy H^E data for binary mixtures of octane + *N*-methylacetamide, cyclooctane + *N*-methylacetamide, and octane + acetic anhydride at approximately 125 °C. The results of these measurements are presented in this paper and have been used to determine the interaction parameters of the NRTL model (Gmehling and Kolbe, 1992). In the future we intend to use these data for the extension of the UNIFAC interaction parameter matrix.

2. Experimental Section

2.1. Degassing and Purification. The octane, cyclooctane, *N*-methylacetamide, and acetic anhydride were degasssed 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). A schematic diagram of the apparatus is shown in Figure 1. The degassed, pure components were filled into the 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, $t = 25.0-40.0$ °C). To check for complete degassing, the vapor pressure was measured again after a second injection of the pure component. As shown previously (Fischer and Gmehling, 1994; Kolbe and Gmehling, 1985) the accuracy of the respective measurements 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 and 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.

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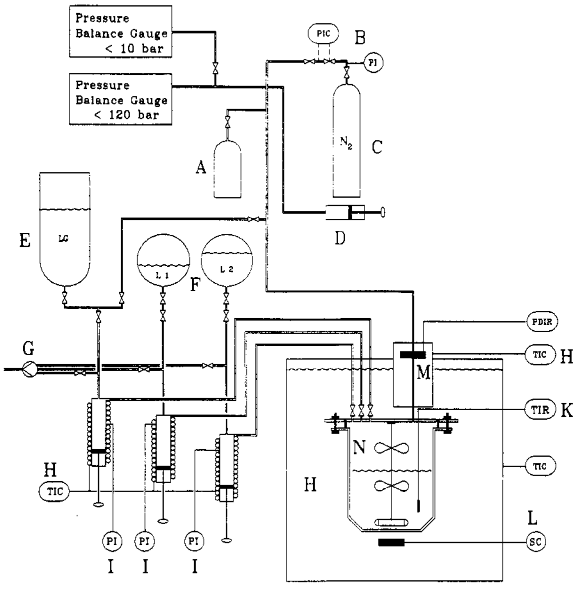

A: Buffer Volume
B: Pressure Regulator
C: Nitrogen Flask
D: Variable Volume
E: Container for Liquified Gas
F: Container for Degassed Liquids
G: Vacuum Pump
H: Constant Temperature Bath
I: Bourdon Pressure Gauge
K: Quartz Thermometer
L: Rotating Magnetic Field
M: Differential Pressure Null Indicator
N: Equilibrium Cell

Figure 1. Schematic of the static apparatus used for the $P-x$ measurements (Fischer and Gmehling, 1994; Kolbe and Gmehling, 1985).

Table 1. Vapor–Liquid Equilibrium Data, Pressure P, Liquid Phase Mole Fraction x_1, Calculated Vapor Mole Fraction $y_{1,\text{calcd}}$, and Calculated Difference in Pressure ΔP, for the System Octane (1) + *N*-Methylacetamide (2) at 124.9 °C

experimental		NRTL	
x_1	P/kPa	ΔP^a/kPa	$y_{1,\text{calcd}}$
0.0000	6.16	0.00	0.0000
0.0206	31.92	0.57	0.8074
0.0307	42.03	0.72	0.8551
0.0399	49.83	0.58	0.8794
0.0484	56.56	0.80	0.8941
0.0552	61.12	0.66	0.9028
0.0654	67.36	0.61	0.9126
0.0779	73.85	0.46	0.9211
0.0984	82.23	0.06	0.9304
0.1385	93.11	−0.75	0.9402
0.1755	98.91	−1.49	0.9448
0.2092	102.95	−1.20	0.9473
0.2399^b	103.10	−3.26	0.9487
0.9383^b	101.73	1.51	0.9351
0.9468^b	101.78	1.59	0.9379
0.9552	101.69	1.58	0.9417
0.9649	101.47	1.54	0.9478
0.9730	101.28	1.62	0.9548
0.9853	100.52	1.59	0.9701
0.9881	99.88	1.19	0.9746
0.9912	99.42	1.04	0.9802
0.9934	99.06	0.92	0.9846
0.9980	98.17	0.63	0.9949
1.0000	97.91	0.68	1.0000

^{a} $\Delta P = P_{\text{exptl}} - P_{\text{calcd}}$. ^{b} Two liquid phases.

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%.

Table 2. Vapor–Liquid Equilibrium Data, Pressure P, Liquid Phase Mole Fraction x_1, Calculated Vapor Mole Fraction $y_{1,\text{calcd}}$, and Calculated Difference in Pressure ΔP, for the System Cyclooctane (1) + *N*-Methylacetamide (2) at 124.9 °C

experimental		NRTL	
x_1	P/kPa	ΔP^a/kPa	$y_{1,\text{calcd}}$
0.0000	6.06	−0.10	0.0000
0.0013	7.50	0.75	0.0889
0.0020	7.93	0.87	0.1301
0.0029	8.48	1.02	0.1777
0.0050	9.52	1.13	0.2698
0.0137	13.26	1.25	0.4943
0.0273	17.37	0.29	0.6487
0.0387	20.81	−0.01	0.7148
0.0522	24.23	−0.52	0.7627
0.0727	28.84	−0.97	0.8061
0.0969	33.42	−1.22	0.8359
0.1276	38.03	−1.35	0.8584
0.1589	41.79	−1.22	0.8726
0.1842	44.40	−0.87	0.8805
0.2078	46.14	−0.83	0.8860
0.2311	47.57	−0.75	0.8902
0.2551	48.74	−0.70	0.8936
0.2770	49.63	−0.64	0.8961
0.2994	50.25	−0.71	0.8982
0.3127	50.63	−0.67	0.8992
0.3307	51.04	−0.66	0.9004
0.3335	51.48	−0.27	0.9006
0.3533	51.46	−0.64	0.9016
0.3553	51.91	−0.22	0.9017
0.3738	51.80	−0.58	0.9025
0.3759	52.09	−0.31	0.9026
0.4120	52.32	−0.42	0.9037
0.4746	52.45	−0.52	0.9045
0.5595^b	52.40	−0.41	0.9038
0.6796^b	52.23	0.13	0.8994
0.8586^b	52.01	0.67	0.8890
0.9027^b	52.03	0.69	0.8917
0.9366	51.92	0.73	0.9022
0.9581	51.74	0.92	0.9177
0.9732	51.29	1.01	0.9361
0.9834	50.54	0.85	0.9544
0.9925	49.62	0.67	0.9763
0.9963	48.61	0.05	0.9876
0.9982	48.35	0.00	0.9938
1.0000	48.13	0.00	1.0000

^{a} $\Delta P = P_{\text{exptl}} - P_{\text{calcd}}$. ^{b} Two liquid phases.

3. Results

3.1. $P-x$ 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 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

Table 3. Vapor–Liquid Equilibrium Data, Pressure P, Liquid Phase Mole Fraction x_1, Calculated Vapor Mole Fraction $y_{1,\mathrm{calcd}}$, and Calculated Difference in Pressure ΔP, for the System Octane (1) + Acetic Anhydride (2) at 124.5 °C

experimental		NRTL	
x_1	P/kPa	ΔP^a/kPa	$y_{1,\mathrm{calcd}}$
0.0000	61.94	0.00	0.0000
0.0237	78.73	1.69	0.2142
0.0331	83.69	1.56	0.2691
0.0391	86.77	1.63	0.2986
0.0448	89.35	1.51	0.3234
0.0499	91.47	1.34	0.3434
0.0564	93.93	1.05	0.3662
0.0756	100.82	0.77	0.4203
0.1019	107.50	−0.35	0.4720
0.1320	113.01	−1.52	0.5120
0.1620	117.23	−2.14	0.5391
0.1950	120.19	−2.93	0.5597
0.2275	122.14	−3.50	0.5737
0.2513	123.45	−4.50	0.5811
0.2738	124.55	−3.30	0.5865
0.2898	125.47	−2.87	0.5895
0.3093	126.22	−2.59	0.5925
0.3305	127.08	−2.09	0.5950
0.3517	127.88	−1.55	0.5969
0.6245	129.78	−0.17	0.6065
0.6328	129.75	−0.19	0.6073
0.6417	129.74	−0.19	0.6083
0.6487	129.80	−0.11	0.6091
0.6591	129.76	−0.12	0.6105
0.6687	129.60	−0.24	0.6119
0.6766	129.53	−0.26	0.6132
0.6820	129.47	−0.29	0.6141
0.6988	129.36	−0.27	0.6174
0.7154	128.92	−0.52	0.6212
0.7402	128.62	−0.42	0.6282
0.7620	127.99	−0.55	0.6358
0.7836	127.18	−0.68	0.6450
0.8059	126.44	−0.48	0.6566
0.8431	124.30	−0.37	0.6819
0.8701	121.90	−0.43	0.7066
0.8952	119.41	−0.06	0.7360
0.9170	116.54	0.22	0.7682
0.9323	114.04	0.37	0.7956
0.9478	111.07	0.50	0.8286
0.9588	108.61	0.53	0.8558
0.9676	106.23	0.34	0.8804
0.9772	103.56	0.26	0.9105
0.9827	101.74	0.04	0.9296
0.9873	100.11	−0.20	0.9467
0.9911	98.91	−0.20	0.9616
0.9948	97.56	−0.35	0.9770
0.9981	96.53	−0.27	0.9914
1.0000	95.48	−0.67	1.0000

[a] $\Delta P = P_{\mathrm{exptl}} - P_{\mathrm{calcd}}$.

in Tables 1–3. Since we cannot observe the mixture in the equilibrium cell optically, liquid–liquid immiscibility in binary mixtures can only be recognized from the pressure constancy in the P–x diagram. The experimental points in Tables 1–3 within the miscibility gap are marked.

3.2. H^E and γ^∞ Data. The excess enthalpies measured at 125.0 °C for the three binary systems are listed in Tables 4–6. In general, modern G^E models, such as NRTL and UNIQUAC, cannot describe the big changes in the slope dP/dx that occur at high dilution and near the miscibility gap with the required accuracy (Fischer and Gmehling, 1994). Therefore, a flexible Legendre polynomial with a variable number of parameters was used to obtain reliable γ^∞ values by fitting the parameters to a few data points (about 5–10) at high dilution. Finally the activity coefficients at infinite dilution were calculated from the slope of the P–x curve at $x = 0$ or $x = 1$. The values are listed in Table 7.

Table 4. Excess Enthalpies H^E vs Liquid Mole Fraction x_1 for the System Octane (1) + N-Methylacetamide (2) at 125.0 °C

x_1	H^E/(J mol^{-1})	x_1	H^E/(J mol^{-1})
0.0118	64.6	0.4110^a	827.1
0.0491	254.5	0.5205^a	804.6
0.0491	259.5	0.6504^a	783.9
0.1040	498.0	0.8072^a	756.8
0.1042	502.6	0.8072^a	756.4
0.1662	711.8	0.9478	701.4
0.2367^a	858.2	0.9735	509.6
0.3175^a	848.0	0.9866	315.6

[a] Two liquid phases.

Table 5. Excess Enthalpies H^E vs Liquid Mole Fraction x_1 for the System Cyclooctane (1) + N-Methylacetamide (2) at 125.0 °C

x_1	H^E/(J mol^{-1})	x_1	H^E/(J mol^{-1})
0.0144	66.6	0.4612	1171.3
0.0292	136.5	0.5145^a	1194.0
0.0596	269.5	0.5711^a	1195.8
0.0596	268.2	0.6007^a	1176.6
0.0915	397.4	0.6313^a	1152.5
0.0915	395.1	0.6628^a	1124.1
0.1249	519.4	0.6954^a	1100.1
0.1249	518.5	0.7290^a	1071.7
0.1598	632.6	0.7638^a	1049.2
0.1965	739.2	0.7998^a	1011.3
0.2350	839.1	0.8370^a	970.0
0.2756	929.2	0.8756^a	887.8
0.3183	1008.0	0.9156	779.0
0.3633	1078.7	0.9570	596.7
0.3633	1078.3	0.9783	435.2
0.4109	1131.6		

[a] Two liquid phases.

Table 6. Excess Enthalpies H^E vs Liquid Mole Fraction x_1 for the System Octane (1) + Acetic Anhydride (2) at 125.0 °C

x_1	H^E/(J mol^{-1})	x_1	H^E/(J mol^{-1})
0.0146	167.8	0.5741	2647.9
0.0603	685.3	0.6980	2385.1
0.1262	1319.3	0.7660	2091.6
0.1985	1812.8	0.8387	1633.8
0.2781	2228.7	0.9165	974.3
0.3662	2477.7	0.9575	542.0
0.4643	2661.2		

Table 7. Quasi Experimental and Calculated Activity Coefficients at Infinite Dilution, γ_i^∞, at Temperature t

		quasi experimental		NRTL	
system (1 + 2)	t/°C	γ_1^∞	γ_2^∞	γ_1^∞	γ_2^∞
octane + N-methylacetamide	124.9	14.5	37.0	14.6	41.7
cyclooctane + N-methylacetamide	125.0	10.8	42.5	9.7	27.9
octane + acetic anhydride	124.5	9.3	7.9	8.1	7.2

3.3. 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 (Christensen et al., 1984; 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

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

Table 8. 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})
octane + *N*-methylacetamide	0.3738	18.033	6.850	−21.774	−3.116
cyclooctane + *N*-methylacetamide	0.3871	12.229	5.622	−9.610	−3.179
octane + acetic anhydride	0.2047	8.945	5.270	−14.374	−2.539

Table 9. Constants for the Antoine Vapor Pressure Equation $\log(P/\mathrm{kPa}) = A - B/(t/°\mathrm{C} + C)$

component	A^a	B	C
octane	6.0468	1358.8	209.86
cyclooctane	5.9767	1438.7	210.13
N-methylacetamide	5.6901	1348.3	150.22
acetic anhydride	6.8111	1781.3	230.40

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

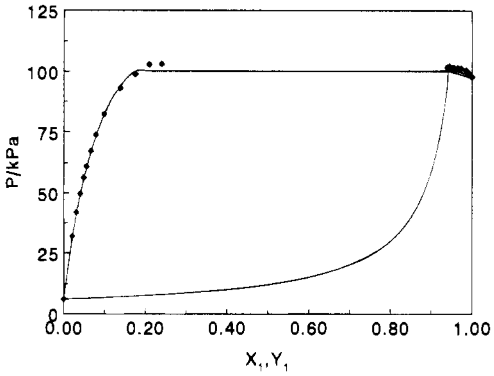

Figure 2. Experimental $P-x$ data (◆) and $P-x-y$ behavior calculated with the NRTL model for the system octane + *N*-methylacetamide at 124.9 °C.

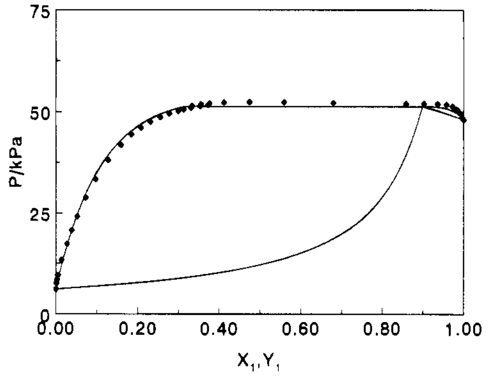

Figure 3. Experimental $P-x$ data (◆) and $P-x-y$ behavior calculated with the NRTL model for the cyclooctane + *N*-methylacetamide at 124.9 °C.

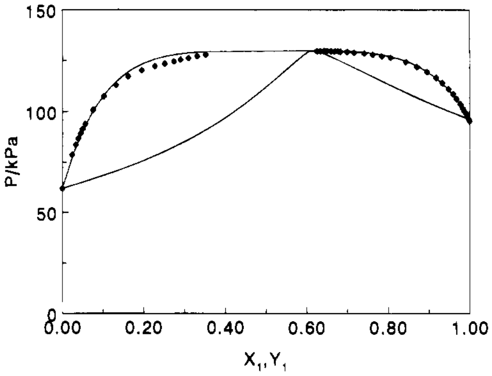

Figure 4. Experimental $P-x$ data (◆) and $P-x-y$ behavior calculated with the NRTL model for the system octane + acetic anhydride at 124.5 °C.

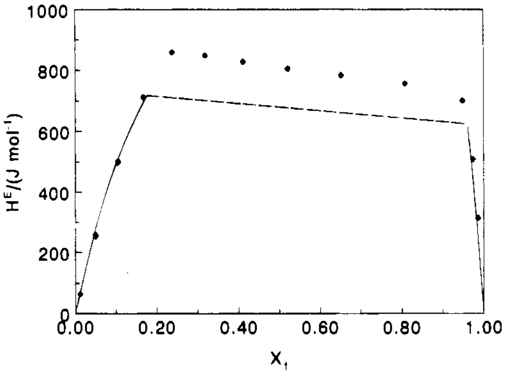

Figure 5. Experimental H^E data (◆) and excess enthalpy curves calculated with the NRTL model for the system octane + *N*-methylacetamide at 125.0 °C.

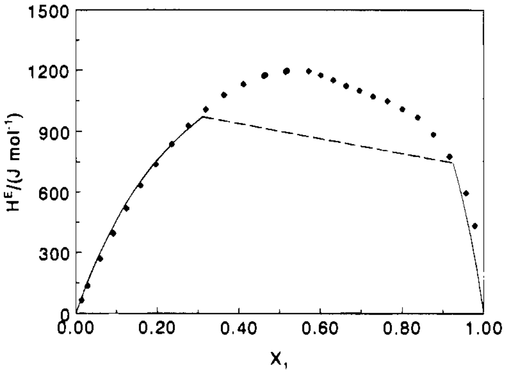

Figure 6. Experimental H^E data (◆) and excess enthalpy curves calculated with the NRTL model for the system cyclooctane + *N*-methylacetamide at 125.0 °C.

The obtained parameters, which were used to calculate the pressure, vapor composition, and excess enthalpies, are listed in Table 8. In Tables 1−3, the deviations $P - P_{\mathrm{calcd}}$ and the calculated vapor compositions are given. Furthermore, in Table 7 the calculated γ_i^∞ values are compared with the quasi experimental values. The Antoine constants used for the calculation of the pure component vapor pressures are listed in Table 9.

4. Discussion

Figure 2 shows a comparison between the experimental $P-x$ data and the $P-x-y$ curves calculated using the fitted NRTL parameters for the octane + *N*-methylacetamide system. The same comparison is made in Figure 3 for the cyclooctane + *N*-methylacetamide system and Figure 4 for the octane + acetic anhydride system. In general good agreement is obtained between the experimental $P-x$ data and the calculated $P-x-y$ curves for all three systems. However, close to the miscibility gap in the octane + *N*-methylacetamide and cyclooctane + *N*-methylacetamide systems the NRTL model clearly fails in describing the big changes in the slope dP/dx and the resulting equilibrium compositions accurately. This is more clearly illustrated by Figures 5 and 6, in which the experimental H^E results at 125 °C are compared with the excess enthalpies calculated using the fitted NRTL parameters for the octane + *N*-methylacetamide and the cyclooctane + *N*-methylacetamide systems. Figure 7 shows that, for the completely miscible octane + acetic anhydride system, the large positive excess enthalpies are well described over the whole

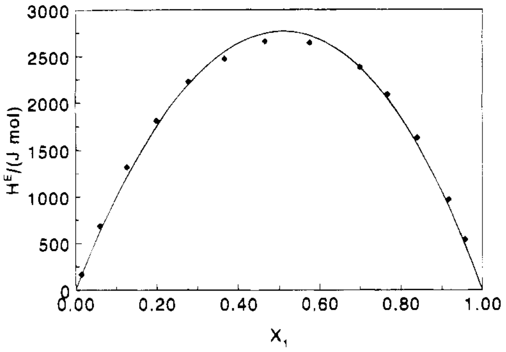

Figure 7. Experimental H^E data (◆) and excess enthalpy curves calculated with the NRTL model for the system octane + acetic anhydride at 125.0 °C.

concentration range by the NRTL model using linear temperature dependent parameters.

Furthermore, it can be seen from Figures 2 and 3 that the octane + *N*-methylacetamide and cyclooctane + *N*-methylacetamide systems show a heterogeneous pressure maximum azeotrope. Figure 4 illustrates that the octane + acetic anhydride system forms a homogeneous pressure maximum azeotrope.

Finally Table 7 illustrates that for all three systems reasonable agreement is obtained between the quasi experimental and the calculated activity coefficients at infinite dilution.

5. Conclusions

Vapor–liquid equilibria and excess enthalpies have been measured at a temperature of about 125 °C for binary mixtures of octane + *N*-methylacetamide, cyclooctane + *N*-methylacetamide, and octane + acetic anhydride. For each binary system the experimental data have been fitted to the NRTL model using linear temperature dependent interaction parameters. Finally the measured $P-x$ data at low concentrations were used to determine the activity coefficients at infinite dilution using a flexible Legendre polynomial. For all three systems good agreement was obtained between the experimental data and the $P-x-y$ and excess enthalpy curves calculated using the fitted NRTL parameters.

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