Vapor−Liquid Equilibria and Excess Enthalpies for Binary Mixtures of Acrylonitrile with Hexane, Cyclohexane, Benzene, Toluene, 2-Butanone, and Acetonitrile

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Isobaric vapor−liquid equilibrium data have been measured for binary mixtures of acrylonitrile with hexane, cyclohexane, benzene, toluene, and 2-butanone. For each of these five binary mixtures and the acrylonitrile + acetonitrile system excess enthalpies have been determined at 50 °C. Azeotropic compositions have been measured for the systems acrylonitrile + hexane, +cyclohexane, +benzene, and +2-butanone. The experimental data were fitted to the NRTL model using linear temperature dependent interaction parameters.

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1. Introduction

In the literature only a few experimental vapor−liquid and no excess enthalpy data are available for binary mixtures of acrylonitrile with hexane, cyclohexane, benzene, toluene, 2-butanone, and acetonitrile. Activity coefficients have been reported for the acrylonitrile + benzene system at 50 and 60 °C by Balashova et al. (1989). Laevskaya et al. (1977) reported some isobaric vapor−liquid equilibrium data for the acrylonitrile + toluene system at 6.67 kPa (50 mmHg). For the system acrylonitrile + acetonitrile vapor−liquid equilibrium data have been reported by various authors (Gmehling and Onken, 1977) of which the only consistent data sets were obtained by Sokolov et al. (1967).

In this paper isobaric vapor−liquid equilibrium data are reported for binary mixtures of acrylonitrile with hexane, cyclohexane, benzene, toluene, and 2-butanone. For each of these five binary mixtures and the acrylonitrile + acetonitrile system excess enthalpies were measured at 50 °C. Azeotropic compositions have been determined for the binary systems acrylonitrile + hexane, +cyclohexane, +benzene, and +2-butanone. The experimental data were correlated by the temperature dependent five-parameter NRTL model.

2. Experimental Section

2.1. Materials and Purification. All materials were 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 99.9%.

2.2. Vapor−Liquid Equilibria. The equilibrium compositions and boiling points were measured using a modified Swietoslawski ebulliometer. A detailed description of the apparatus is given by Rogalski et al. (1977) and Rogalski and Malanowski (1980). The pressure was controlled by a Vacuubrand vacuum controller CVC 24 and a Wallace & Tiernan mercury gauge with a resolution of ±0.01 kPa. Equilibrium temperatures were measured using a calibrated thermometer with an accuracy of ±0.05 K.

Table 1. Vapor−Liquid Equilibrium Data, Temperature t, Liquid-Phase Mole Fraction x_1, Vapor-Phase Mole Fraction y_1, Calculated Difference in Temperature Δt, and Calculated Difference in Vapor-Phase Mole Fraction Δy_1, for the System Acrylonitrile (1) + Hexane (2) at 30.04 kPa

x_1	y_1	t/°C	$\Delta y_1{}^a$	Δt^b/°C
0.000	0.000	34.50	0.000	0.00
0.015	0.113	32.10	0.037	−0.65
0.040	0.220	29.20	−0.024	0.25
0.068	0.285	26.90	−0.039	0.52
0.132	0.339	25.80	0.004	0.59
0.193	0.365	25.30	0.001	−0.05
0.360	0.383	25.20	0.005	−0.14
0.519	0.377	25.20	−0.014	0.01
0.819	0.456	26.60	−0.000	−0.41
0.857	0.474	27.55	−0.004	−0.41
0.892	0.527	9.00	−0.010	−0.23
0.920	0.578	30.95	−0.007	−0.19
0.939	0.625	32.50	−0.001	−0.42
0.951	0.676	34.10	0.000	−0.42
0.961	0.717	35.45	0.003	−0.47
0.983	0.835	38.80	0.005	−0.36
1.000	1.000	42.95	0.000	0.00

[a] $\Delta y_1 = y_1 - y_{1,\mathrm{calc}}$. [b] $\Delta t = t - t_{\mathrm{calc}}$.

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 enthalpy 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%.

2.4. Azeotropic Compositions. For the measurements of the azeotropic compositions, a commercially available micro spinning band column, with an electronically controlled reflux ratio, from NORMAG GmbH was used. With this column, described in detail previously (Gmehling, 1996), homogeneous as well as heterogeneous pressure maximum azeotropes can be measured, realizing up to 50

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Table 2. Vapor−Liquid Equilibrium Data, Temperature *t*, Liquid-Phase Mole Fraction x_1, Vapor-Phase Mole Fraction y_1, Calculated Difference in Temperature Δt, and Calculated Difference in Vapor-Phase Mole Fraction Δy_1, for the System Acrylonitrile (1) + Cyclohexane (2) at 30.03 kPa

x_1	y_1	t/°C	Δy_1[a]	Δt[b]/°C
0.000	0.000	45.10	0.000	0.00
0.013	0.187	40.05	0.041	−1.33
0.028	0.247	38.20	−0.005	−0.21
0.041	0.297	36.55	−0.019	0.08
0.060	0.355	34.85	−0.019	0.26
0.080	0.389	33.55	−0.022	0.20
0.125	0.435	32.50	−0.019	0.55
0.162	0.457	31.80	−0.013	0.32
0.237	0.468	31.40	−0.011	0.21
0.320	0.482	31.20	0.000	0.07
0.428	0.477	31.15	−0.008	0.05
0.537	0.496	31.15	0.003	0.05
0.623	0.507	31.15	0.001	−0.05
0.696	0.525	31.30	0.002	−0.15
0.709	0.534	31.30	0.007	−0.22
0.779	0.544	31.70	−0.012	−0.41
0.835	0.580	32.50	−0.014	−0.50
0.885	0.642	33.85	−0.006	−0.54
0.919	0.695	35.45	−0.008	−0.38
0.949	0.774	37.40	−0.004	−0.32
0.970	0.851	39.25	0.003	−0.20
0.991	0.946	41.70	0.001	−0.02
1.000	1.000	42.95	0.000	0.00

[a] $\Delta y_1 = y_1 - y_{1,\text{calc}}$. [b] $\Delta t = t - t_{\text{calc}}$.

Table 3. Vapor−Liquid Equilibrium Data, Temperature *t*, Liquid-Phase Mole Fraction x_1, Vapor-Phase Mole Fraction y_1, Calculated Difference in Temperature Δt, and Calculated Difference in Vapor-Phase Mole Fraction Δy_1, for the System Acrylonitrile (1) + Benzene (2) at 30.03 kPa

x_1	y_1	t/°C	Δy_1[a]	Δt[b]/°C
0.000	0.000	45.15	0.000	0.00
0.086	0.154	43.05	0.037	−0.65
0.158	0.230	42.15	−0.024	0.25
0.240	0.303	41.45	−0.039	0.52
0.329	0.389	40.80	0.004	0.59
0.430	0.459	40.50	0.001	−0.05
0.531	0.532	40.40	0.005	−0.14
0.619	0.603	40.50	−0.014	0.01
0.653	0.629	40.50	−0.000	−0.41
0.709	0.671	40.60	−0.004	−0.41
0.713	0.683	40.60	−0.010	−0.23
0.781	0.741	40.90	−0.007	−0.19
0.844	0.800	41.25	−0.001	−0.42
0.892	0.852	41.65	0.000	−0.42
0.931	0.897	42.00	0.003	−0.47
0.959	0.938	42.40	0.005	−0.36
0.977	0.965	42.65	0.005	−0.35
0.987	0.980	42.75	0.003	−0.17
0.994	0.991	42.85	0.002	−0.07
1.000	1.000	42.95	0.000	0.00

[a] $\Delta y_1 = y_1 - y_{1,\text{calc}}$. [b] $\Delta t = t - t_{\text{calc}}$.

theoretical stages at a low pressure drop. The temperature was determined with a resistance thermometer with an accuracy of ±0.1 deg and the pressure, by means of a sensor (Druck Limited, type PDCR) with an accuracy of ±0.05 kPa.

2.5. Sample Analysis. Liquid- and vapor-phase samples were analyzed using an HP 5890 gas chromatograph, equipped with a polar capillary column (CP-Wax 52 CB) and a flame ionization detector. Hydrogen was used as the carrier gas. For each binary system first a calibration curve was constructed that was subsequently used to determine the liquid and vapor sample mole fractions with an accuracy of ±0.002.

Table 4. Vapor−Liquid Equilibrium Data, Temperature *t*, Liquid-Phase Mole Fraction x_1, Vapor-Phase Mole Fraction y_1, Calculated Difference in Temperature Δt, and Calculated Difference in Vapor-Phase Mole Fraction Δy_1, for the System Acrylonitrile (1) + Toluene (2) at 26.94 kPa

x_1	y_1	t/°C	Δy_1[a]	Δt[b]/°C
0.000	0.000	69.80	0.000	−0.00
0.025	0.183	65.60	0.037	−0.64
0.067	0.283	62.10	−0.024	0.27
0.119	0.391	58.60	−0.039	0.54
0.182	0.529	55.50	0.004	0.60
0.229	0.576	53.10	0.001	−0.04
0.305	0.642	50.80	0.005	−0.15
0.345	0.650	50.00	−0.014	0.01
0.401	0.697	48.40	−0.000	−0.42
0.466	0.728	47.20	−0.004	−0.43
0.544	0.761	46.10	−0.010	−0.25
0.620	0.801	45.00	−0.007	−0.21
0.625	0.809	44.70	−0.001	−0.44
0.696	0.844	43.70	0.000	−0.45
0.761	0.878	42.80	0.003	−0.49
0.809	0.904	42.30	0.005	−0.37
0.859	0.929	41.70	0.005	-0.35
0.907	0.952	41.30	0.003	-0.17
0.950	0.974	40.90	0.002	-0.07
0.975	0.986	40.70	-0.000	0.02
0.992	0.995	40.50	-0.001	0.01
1.000	1.000	40.40	0.000	0.00

[a] $\Delta y_1 = y_1 - y_{1,\text{calc}}$. [b] $\Delta t = t - t_{\text{calc}}$.

Table 5. Vapor−Liquid Equilibrium Data, Temperature *t*, Liquid-Phase Mole Fraction x_1, Vapor-Phase Mole Fraction y_1, Calculated Difference in Temperature Δt, and Calculated Difference in Vapor-Phase Mole Fraction Δy_1, for the System Acrylonitrile (1) + 2-Butanone(2) at 20.11 kPa

x_1	y_1	t/°C	Δy_1[a]	Δt[b]/°C
0.000	0.000	36.40	0.000	0.00
0.022	0.021	36.45	0.001	0.01
0.051	0.047	36.45	−0.001	−0.03
0.078	0.077	36.50	0.002	−0.01
0.106	0.107	36.50	0.004	−0.04
0.157	0.156	36.50	−0.000	−0.06
0.190	0.192	36.45	0.001	−0.11
0.240	0.249	36.40	0.003	−0.14
0.289	0.305	36.30	0.005	−0.18
0.407	0.436	36.00	0.005	−0.25
0.526	0.559	35.70	−0.004	−0.17
0.651	0.687	35.20	−0.006	−0.15
0.735	0.769	34.80	−0.006	−0.14
0.801	0.834	34.50	−0.002	−0.08
0.846	0.875	34.20	−0.001	−0.13
0.893	0.915	34.00	−0.001	−0.05
0.907	0.927	33.90	−0.000	−0.07
0.939	0.952	33.75	−0.001	−0.03
0.941	0.956	33.75	0.001	−0.01
0.972	0.979	33.55	0.000	−0.02
0.990	0.993	33.45	0.001	−0.01
1.000	1.000	33.40	0.000	0.00

[a] $\Delta y_1 = y_1 - y_{1,\text{calc}}$. [b] $\Delta t = t - t_{\text{calc}}$.

3. Results and Correlation

The vapor−liquid equilibrium data obtained for binary mixtures of acrylonitrile with hexane, cyclohexane, benzene, toluene and 2-butanone are presented in Tables 1−5. Table 6 lists the excess enthalpies measured at 50 °C for these five binary mixtures and the acrylonitrile + acetonitrile system. The azeotropic compositions measured for the binary systems acrylonitrile + hexane, +cyclohexane, +benzene, and +2-butanone are given in Table 7. For each binary system, the interaction parameters of the NRTL model have been fitted simultaneously to the measured vapor−liquid equilibrium data and excess enthalpies using

Table 6. Measured Excess Enthalpies H^E vs Liquid Mole Fraction x_1 at 50 °C

x_1	H^E/J mol^{-1}	x_1	H^E/J mol^{-1}	x_1	H^E/J mol^{-1}
		Acrylonitrile (1) + Hexane (2)			
0.0486	465.7	0.5178	1682.6	0.8231	1003.5
0.0950	788.5	0.5707	1642.8	0.8568	847.0
0.1814	1217.9	0.6200	1577.4	0.8886	692.2
0.2603	1473.1	0.6660	1528.6	0.9187	535.2
0.3327	1622.5	0.7091	1398.7	0.9472	358.9
0.3993	1663.4	0.7495	1280.4	0.9743	182.5
0.4608	1690.8	0.7874	1147.8	0.9873	92.6
		Acrylonitrile (1) + Cyclohexane (2)			
0.0406	440.5	0.4702	1730.8	0.8318	954.7
0.0798	761.9	0.5236	1696.8	0.8683	786.3
0.1548	1140.5	0.5742	1657.9	0.9033	608.1
0.2254	1426.5	0.6683	1475.4	0.9369	402.0
0.2919	1568.1	0.7120	1383.2	0.9691	202.3
0.3546	1680.9	0.7538	1262.0		
0.4140	1715.9	0.7937	1111.3		
		Acrylonitrile (1) + Benzene (2)			
0.0336	31.0	0.4220	197.1	0.7598	174.8
0.0666	56.2	0.4747	205.5	0.8026	155.9
0.1309	94.6	0.5259	210.1	0.8443	132.0
0.1931	124.3	0.5755	210.6	0.8848	104.2
0.2531	148.5	0.6236	208.5	0.9242	73.1
0.3112	168.2	0.6703	201.6	0.9626	38.2
0.3675	183.8	0.7157	190.2	0.9814	18.9
		Acrylonitrile (1) + Toluene (2)			
0.0399	47.2	0.4659	251.5	0.7908	190.8
0.0786	85.1	0.5192	255.4	0.8294	166.2
0.1526	140.2	0.5700	253.8	0.8663	140.2
0.2223	178.5	0.6183	249.8	0.9018	108.7
0.2883	207.4	0.6644	241.4	0.9358	74.3
0.3507	228.1	0.7085	228.1	0.9685	38.9
0.4098	242.1	0.7505	212.2	0.9844	19.2
		Acrylonitrile (1) + 2-Butanone (2)			
0.0338	−34.7	0.4239	−220.7	0.7612	−139.1
0.0671	−64.9	0.4767	−220.3	0.8039	−116.5
0.1318	−114.6	0.5278	−215.4	0.8453	−94.1
0.1943	−153.6	0.5774	−208.0	0.8856	−70.4
0.2546	−182.9	0.6255	−194.1	0.9248	−46.5
0.3129	−202.6	0.6721	−178.2	0.9629	−23.7
0.3693	−215.1	0.7173	−159.4		
		Acrylonitrile (1) + Acetonitrile (2)			
0.0201	2.49	0.3014	24.56	0.7062	22.58
0.0405	4.74	0.3482	26.31	0.7622	19.56
0.0818	9.22	0.3960	27.52	0.8195	15.48
0.1239	13.06	0.4948	28.34	0.8782	11.34
0.1669	16.58	0.5459	27.86	0.9384	5.96
0.2108	19.62	0.5981	26.71	0.9690	2.90
0.2557	22.43	0.6516	25.04		

Table 7. Experimental and Calculated Azeotropic Compositions x_1, at Pressure P and Temperature t.

		experimental		calculated	
system	P/kPa	x_1	t/°C	x_1	t/°C
acrylonitrile (1) + hexane (2)	37.06	0.406	30.30	0.395	30.20
	64.21	0.406	44.70	0.396	44.75
	101.05	0.405	57.75	0.396	58.16
acrylonitrile (1) + cyclohexane (2)	30.25	0.481	31.30	0.489	31.24
	60.56	0.483	49.40	0.487	49.63
	101.94	0.484	64.60	0.486	65.39
acrylonitrile (1) + benzene (2)	30.19	0.548	40.60	0.551	40.43
	57.17	0.554	57.30	0.526	57.58
	101.46	0.557	74.30	0.507	74.94
acrylonitrile (1) + 2-butanone (2)	20.11	0.120	36.50	0.057	36.41
	20.09	0.116	36.60	0.057	36.38

the following linear temperature dependency for the interaction parameters, g_{ij}

$$g_{ij} = g^{o}_{ij} + g^{1}_{ij}T/\mathrm{K} \qquad (1)$$

Table 8. Fitted NRTL Nonrandomness Parameter, α_{ij}, and Interaction Parameters, $g_{ij} = g^0_{ij} + g^1_{ij}$ T/(K) for Binary Mixtures

acrylonitrile (1) +	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}
hexane (2)	0.424	6.587	8.966	−10.301	−14.512
cyclohexane (2)	0.429	6.911	9.525	−12.772	−14.405
benzene (2)	0.300	1.627	−0.955	−5.076	8.332
toluene (2)	0.300	1.464	−0.719	−6.019	11.474
2-butanone (2)	0.300	−0.581	−0.202	−4.506	7.319
acetonitrile (2)	0.300	0.170	−0.107	−4.862	6.452

Table 9. Constants for the Antoine Vapor Pressure Equation

$$\log(P/\mathrm{kPa}) = A - B/(t/°\mathrm{C} + C)$$

component	A^a	B	C
acrylonitrile	6.0572	1255.9	231.30
hexane	6.0034	1171.5	224.37
cyclohexane	5.9764	1206.5	223.14
benzene	6.0054	1196.8	219.16
toluene	6.0752	1342.3	219.19
2-butanone	6.1854	1261.3	221.97
acetonitrile	6.4648	1482.3	250.52

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

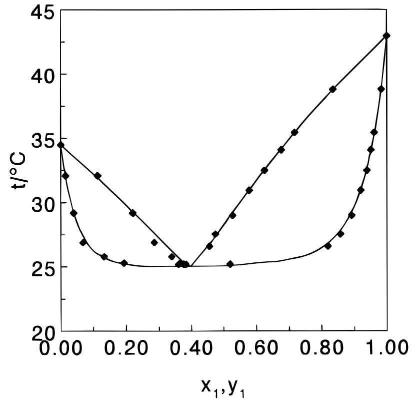

Figure 1. Experimental $t-x-y$ data (♦) and NRTL fit for acrylonitrile (1) + hexane (2) at 30.04 kPa.

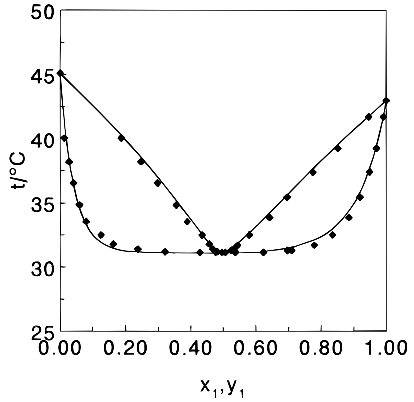

Figure 2. Experimental $t-x-y$ data (♦) and NRTL fit for acrylonitrile (1) + cyclohexane (2) at 30.03 kPa.

In the case of the acrylonitrile + acetonitrile system the vapor−liquid equilibrium data reported by Sokolov et al. (1967) were used. The parameters were fitted with the help of the Simplex−Nelder−Mead method (Nelder and Mead, 1965), by minimization of the following objective function, F

$$F = \sum\{(\gamma_i - \gamma_{i,\mathrm{calc}})/\gamma_i\}^2 + \sum\{(H^E - H^E_{\mathrm{calc}})/H^E\}^2 \quad (2)$$

where γ_i is the activity coefficient of component i, H^E is

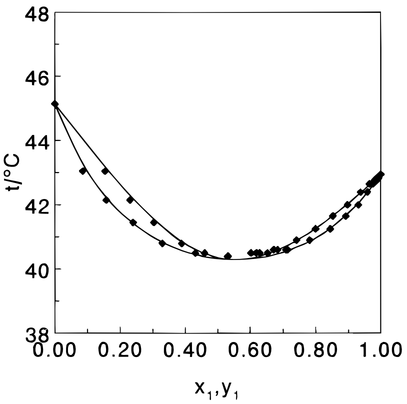

Figure 3. Experimental *t*−*x*−*y* data (♦) and NRTL fit for acrylonitrile (1) + benzene (2) at 30.03 kPa.

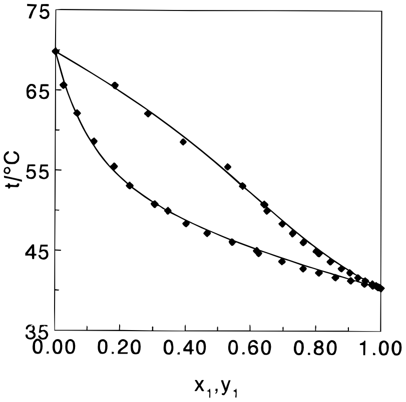

Figure 4. Experimental *t*−*x*−*y* data (♦) and NRTL fit for acrylonitrile (1) + toluene (2) at 26.94 kPa.

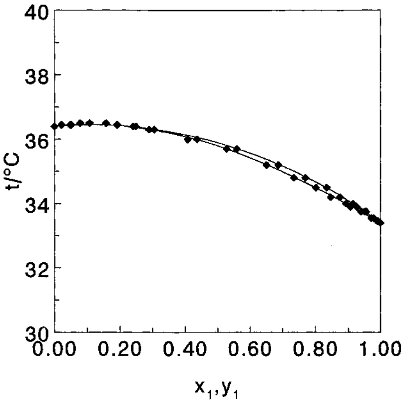

Figure 5. Experimental *t*−*x*−*y* data (♦) and NRTL fit for acrylonitrile (1) + 2-butanone (2) at 20.11 kPa.

the excess enthalpy, and the index "calc" denotes the calculated values. 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. Table 8 lists the obtained parameters, that were used to calculate vapor composition, temperature, and excess enthalpies. In Tables 1−5, the deviations in calculated vapor composition and temperature are given. The calculated and measured azeotropic compositions are compared in Table 7. Table 9 gives the Antoine constants used for the calculation of the pure component vapor pressures.

4. Discussion and Conclusions

Figure 1 shows a comparison between the experimental *t*−*y* data and the *t*−*x*−*y* curves calculated by using the fitted NRTL parameters for the acrylonitrile + hexane system. The same comparison is made in Figure 2 for acrylonitrile + cyclohexane, Figure 3 for acrylonitrile + benzene, Figure 4 for acrylonitrile + toluene, and Figure 5 for acrylonitrile + 2-butanone. For all five systems good agreement is obtained between the experimental *t*−*x*−*y* data and the calculated curves. Except for the acrylontrile + toluene system the other four systems exhibit a homogeneous pressure maximum azeotrope. Table 7 illustrates that for the acrylonitrile + hexane and acrylonitrile + cyclohexane systems the calculated azeotropic compositions are within 0.01 mole fraction of the experimental values. Due to the much smaller differences in vapor- and liquid-phase compositions, the difference between the experimental and calculated azeotropic compositions are significantly larger, up to 0.06 mole fraction, in the acrylonitrile +

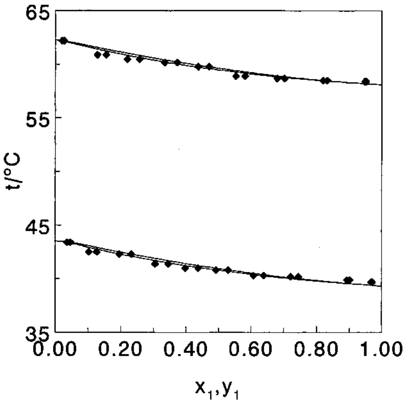

Figure 6. Experimental *t*−*x*−*y* data (♦) obtained by Sokolov et al., 1967, and NRTL fit for acrylonitrile (1) + acetonitrile (2) at 26.66 and 53.33 kPa.

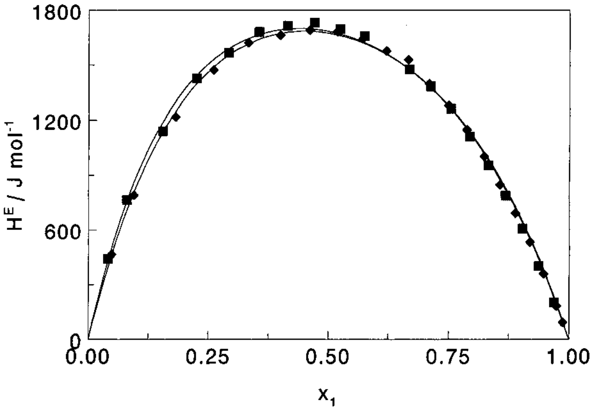

Figure 7. Experimental H^E data and NRTL fit for acrylonitrile (1) with hexane (2) (♦) and cyclohexane (2) (■) at 50 °C.

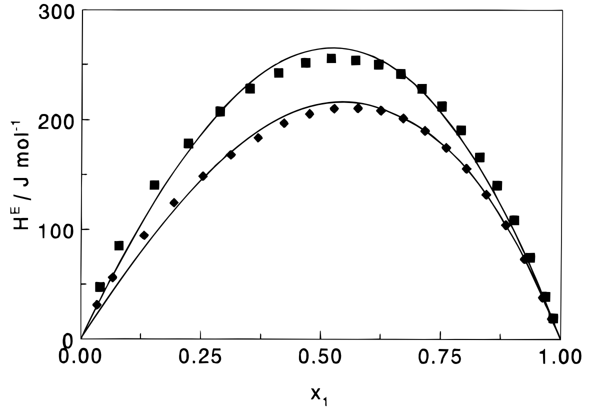

Figure 8. Experimental H^E data and NRTL fit for acrylonitrile (1) with benzene (2) (♦) and toluene (2) (■) at 50 °C.

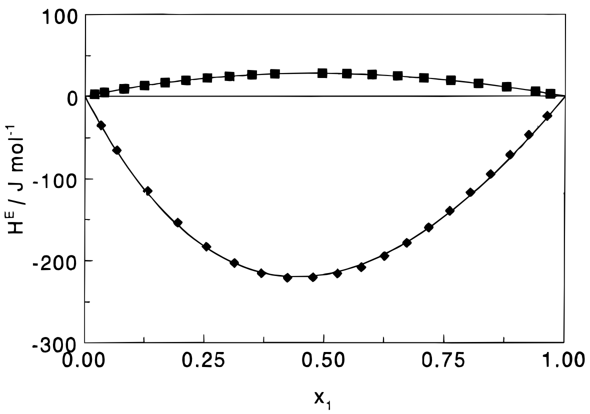

Figure 9. Experimental H^E data and NRTL fit for acrylonitrile (1) with 2-butanone (2) (♦) and acetonitrile (2) (■) at 50 °C.

benzene and acrylonitrile + 2-butanone systems. Finally, Figure 6 illustrates that also for the acrylonitrile + acetonitrile system acceptable agreement is obtained between the calculated $t-x-y$ curves and the experimental $t-x-y$ data determined at 26.66 and 53.33 kPa by Sokolov et al., 1967.

A comparison between the experimental results at 50 °C and the excess enthalpies calculated using the fitted NRTL parameters is presented in Figure 7 for the acrylonitrile + hexane and acrylonitrile + cyclohexane systems, Figure 8 for the acrylonitrile + benzene and acrylonitrile + toluene systems, and Figure 9 for the acrylonitrile + 2-butanone and acrylonitrile + acetonitrile systems. Except for the acrylonitrile + 2-butanone system, that exhibits large negative excess enthalpies, the other five systems show small to very large positive excess enthalpies. Although the agreement for the binary mixtures with benzene and toluene is somewhat less in comparison to the other four systems, it can be concluded that for all six systems acceptable agreement between the experimental data and calculated curves is obtained.

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