J. Chem. Eng. Data **1999,** *44,* 303−308

Vapor−Liquid Equilibria at 453.25 K and Excess Enthalpies at 363.15 K and 413.15 K for Mixtures of Benzene, Toluene, Phenol, Benzaldehyde, and Benzyl Alcohol with Benzyl Benzoate

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Isothermal $P-x$ data at 453.25 K and excess enthalpies at 363.15 K and 413.15 K are presented for binary systems of benzyl benzoate together with benzene, toluene, phenol, benzaldehyde, and benzyl alcohol. As pointed out previously (Nienhaus, B.; Limbeck, U.; Bölts, R.; de Haan, A. B.; Niemann, S. H.; Gmehling, J. *J. Chem. Eng. Data* **1998**, *43*, 941−948), information about the real behavior of systems including these components is very important, especially for the production of phenol by toluene oxidation. In addition, these data will be used for the introduction of a new aromatic ester group (AC−COO−) into modified UNIFAC (Dortmund), which is part of the revision, extension, and further development of this group contribution method. The vapor−liquid equilibrium (VLE) data of four systems were measured using a static apparatus. The corresponding experimental excess enthalpy (H^E) data were determined with the help of an isothermal flow calorimeter. For the simultaneous description of VLE and H^E data the NRTL model was used.

Introduction

There is a lack of reliable published experimental data for binary systems containing aromatic components, and there are only very few systems investigated in a satisfying manner, especially at higher temperatures (Gmehling et al., 1977). Accurate information about the real behavior of these systems, particularly at higher temperatures, is very important for synthesis and design of chemical processes, so efforts have been made to overcome this lack of data. In a first series of selected measurements, vapor−liquid equilibria (VLE) and excess enthalpies (H^E) of systems with benzaldehyde and phenol have been investigated (Nienhaus et al., 1998). Both components are important intermediates in bulk chemistry, as they are byproducts of the large-scale toluene oxidation process (Dow Chemical Co., 1955). Furthermore, they serve as model components for the extension and revision of the group contribution method modified UNIFAC (Dortmund) (Weidlich and Gmehling, 1987; Gmehling et al., 1993; Gmehling et al., 1998; Lohmann et al., 1998), in particular for the revision of the phenol group (AC−OH) and for the extension of the model by the new aromatic aldehyde group (AC−CHO).

In this work, which is presenting a second series of measurements, selected aromatic systems with benzyl benzoate have been investigated at higher temperatures. Benzyl benzoate is not only an important byproduct of the above-mentioned phenol process but also serves as a model component for the modified UNIFAC (Dortmund) aromatic ester group (AC−COO−), which is also planned to be introduced into the model soon.

To obtain information about the real behavior of the binary systems with an acceptable experimental effort, the precise measurement of one $P-x$ data set together with two excess enthalpy data sets covering the temperature range of interest has proved to be a good solution. Therefore, for all of the five systems benzene + benzyl benzoate, toluene + benzyl benzoate, benzaldehyde + benzyl benzoate, phenol + benzyl benzoate, and benzyl alcohol + benzyl benzoate, isothermal $P-x$ data at 453.25 K as well as excess enthalpy data at 363.15 K and 413.15 K have been measured.

Experimental Section

(a) Degassing and Purification. Benzene, toluene, benzyl alcohol, and benzyl benzoate were obtained from commercial sources (Fluka, Merck, Aldrich); benzaldehyde and phenol were delivered by DSM Research. All components were purified and degassed according to the vacuum rectification method described by Fischer and Gmehling (1994). The final purity of the components determined by gas chromatography and Karl Fischer titration was better than 99.5%.

(b) VLE Measurements. Isothermal $P-x$ data were obtained with a static apparatus, which is described in detail by Fischer and Gmehling (1994) and Kolbe and Gmehling (1985), as well as in the first paper about the measurements of aromatic systems (Nienhaus et al., 1998).

The vapor−liquid equilibria of the five systems investigated in this work have been measured at 453.25 K (according to the temperature range of interest for the phenol process and for the support of the planned modified UNIFAC (Dortmund) parameters), which is at the temperature limit of the static apparatus. To obtain reliable γ_i^∞ values, all of the systems in this work have been measured in half-isotherms.

During a measurement, the system pressure is determined as a function of liquid composition. Since the liquid composition is not directly analyzed, it is determined from

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10.1021/je9802524 CCC: $18.00

Table 1. Experimental $P-x$ Data for the System Benzene (1) + Benzyl Benzoate (2) at 453.23 K

x_1	P/kPa	x_1	P/kPa	x_1	P/kPa
0.0000	1.6	0.4662	429.16	0.8534	854.34
0.0189	17.735	0.5566	524.08	0.9007	910.04
0.0620	53.014	0.5833	555.35	0.9279	942.67
0.1526	132.30	0.6323	606.36	0.9565	976.31
0.2275	199.32	0.6870	669.09	0.9812	1008.4
0.2941	260.77	0.7676	757.60	1.0000	1032.3
0.3814	344.36				

Table 2. Experimental $P-x$ Data for the System Toluene (1) + Benzyl Benzoate (2) at 453.25 K

x_1	P/kPa	x_1	P/kPa	x_1	P/kPa
0.0000	1.6	0.4105	209.28	0.8498	436.12
0.0211	12.410	0.5056	258.62	0.8947	460.46
0.0415	22.363	0.5706	290.19	0.9343	482.79
0.0890	45.959	0.6174	313.96	0.9633	499.25
0.1525	77.642	0.6735	343.44	0.9838	511.21
0.2275	115.52	0.7403	378.12	0.9976	519.12
0.3151	160.25	0.7984	408.79	1.0000	519.80

Table 3. Experimental $P-x$ Data for the System Phenol (1) + Benzyl Benzoate (2) at 453.26 K

x_1	P/kPa	x_1	P/kPa	x_1	P/kPa
0.0000	1.6	0.4051	37.332	0.7213	69.553
0.0437	6.655	0.4662	43.388	0.8111	79.317
0.0952	10.526	0.5252	49.533	0.8893	87.782
0.1889	17.907	0.5432	50.920	0.9420	93.447
0.2711	24.882	0.5912	55.754	0.9778	96.876
0.3457	31.636	0.6505	61.892	1.0000	98.664

Table 4. Experimental $P-x$ Data for the System Benzaldehyde (1) + Benzyl Benzoate (2) at 453.25 K

x_1	P/kPa	x_1	P/kPa	x_1	P/kPa
0.0000	1.6	0.4248	45.359	0.8093	86.597
0.0133	3.271	0.5213	55.695	0.8829	94.599
0.0563	7.593	0.5543	59.954	0.9416	100.652
0.0989	11.828	0.5897	63.173	0.9750	104.166
0.1529	17.303	0.6018	64.861	0.9957	106.247
0.2318	25.303	0.6566	70.440	1.0000	106.193
0.3298	35.376	0.7252	77.703		

the volumes of the injected compounds. As decribed before, these volumes are obtained from precise piston injectors, taking into account the small composition changes caused by evaporation (Fischer and Gmehling, 1994). In this work, a new piston injector was used for the dosage of phenol, which was thermostated at 333.15 K, to keep phenol liquid. This new piston injector is thermostatable up to 473.15 K, so that even the measurement of systems with high-melting components is possible now. Throughout all of the measurements, the temperature fluctuation was within a range of ±10 mK, uncertainties in pressure were estimated to be about 0.1 kPa, and an overall accuracy of ±0.5% concerning the mole fractions was assumed.

(c) Excess Enthalpy Measurements. For the measurement of the excess enthalpies an isothermal flow calorimeter (Hart Scientific, Model 7501) was used. The experimental setup has been described in detail previously (Gmehling, 1993). The temperature was kept constant within a range of ±5 mK; the error in mole fraction was less than 0.0005. The accuracy in H^E was estimated to be smaller than 1%.

Results

Tables 1−5 present the results of the isothermal $P-x$ measurements of the systems benzene + benzyl benzoate, toluene + benzyl benzoate, phenol + benzyl benzoate, benzaldehyde + benzyl benzoate, and benzyl alcohol + benzyl benzoate. The experimentally obtained pressures P are given as a function of liquid composition at 453.25 K. The corresponding graphical representations are shown in Figures 1−5. From the $P-x$ data it can be seen that all systems show nearly ideal behavior, tending to a slightly negative deviation from Raoult's law, except for the system benzyl alcohol + benzyl benzoate (Figure 5), showing a moderately positive deviation from Raoult's law. Since all systems investigated are more or less low-pressure systems, the vapor phase can be expected to show ideal behavior. When the virial equation is used for the descrip-

Table 5. Experimental $P-x$ Data for the System Benzyl Alcohol (1) + Benzyl Benzoate (2) at 453.23 K

x_1	P/kPa	x_1	P/kPa	x_1	P/kPa
0.0000	1.6	0.3973	23.065	0.7509	40.061
0.0473	4.224	0.4456	25.371	0.8009	41.991
0.0977	7.117	0.4978	28.188	0.8513	43.771
0.1506	10.091	0.5483	30.688	0.9018	46.076
0.1952	12.554	0.5984	33.276	0.9520	48.080
0.2445	15.177	0.6478	35.622	1.0000	49.955
0.2944	17.771	0.6501	36.118		
0.3468	20.557	0.7009	38.170		

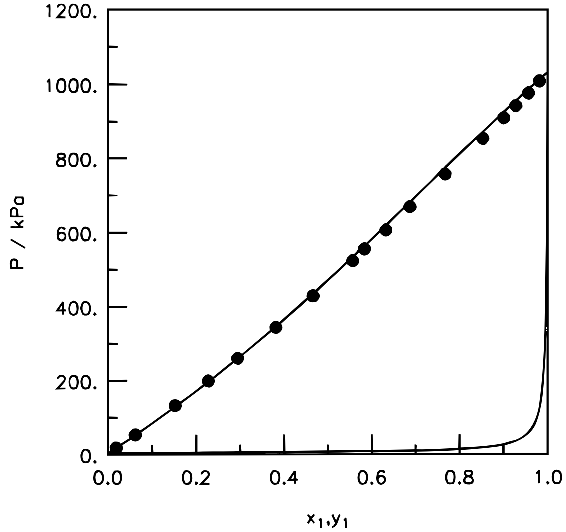

Figure 1. Experimental $P-x$ data (●) and $P-x-y$ behavior calculated by NRTL (—) for the system benzene (1) + benzyl benzoate (2) at 453.23 K.

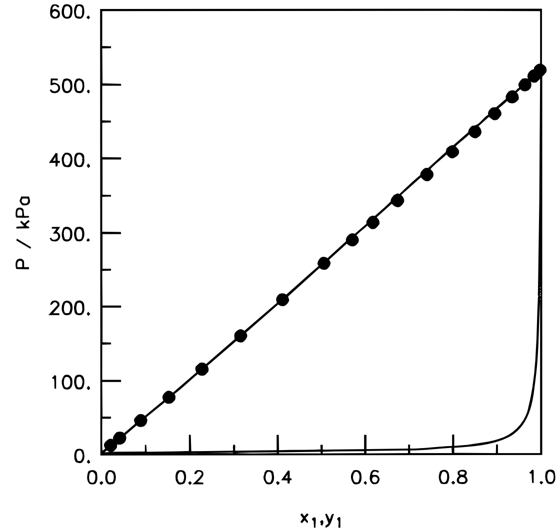

Figure 2. Experimental $P-x$ data (●) and $P-x-y$ behavior calculated by NRTL (—) for the system toluene (1) + benzyl benzoate (2) at 453.25 K.

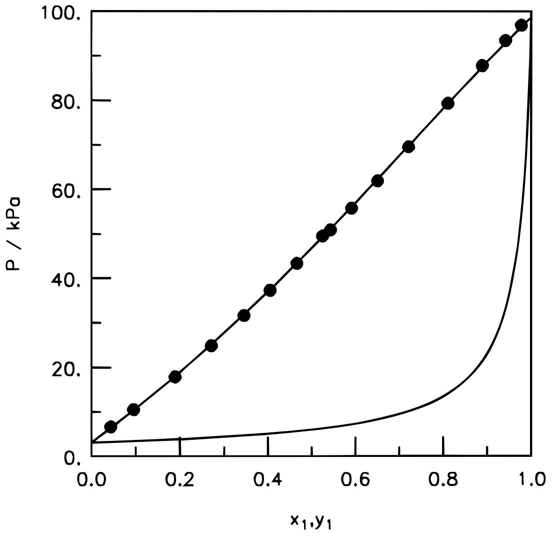

Figure 3. Experimental $P-x$ data (●) and $P-x-y$ behavior calculated by NRTL (—) for the system phenol (1) + benzyl benzoate (2) at 453.26 K.

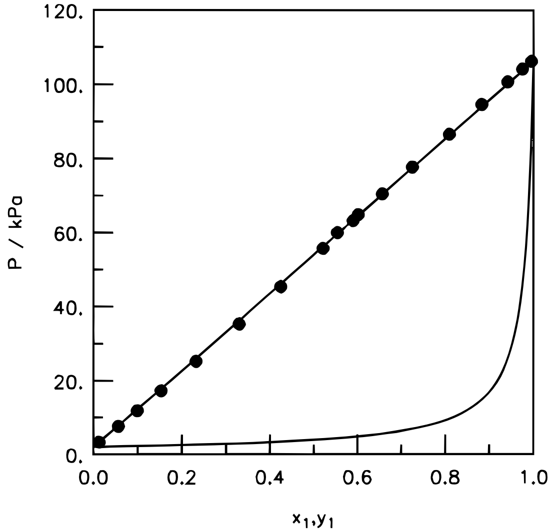

Figure 4. Experimental $P-x$ data (●) and $P-x-y$ behavior calculated by NRTL (—) for the system benzaldehyde (1) + benzyl benzoate (2) at 453.25 K.

tion of the real behavior of the vapor phase, no significant change can be observed during the calculation of the $P-x$ data.

In Tables 6−10 the experimental excess enthalpy (H^E) data at 363.15 K and 413.15 K are given as a function of liquid composition for the systems benzene + benzyl benzoate, toluene + benzyl benzoate, phenol + benzyl benzoate, benzaldehyde + benzyl benzoate, and benzyl alcohol + benzyl benzoate. The corresponding plots of these data are given in Figures 6−10.

Using the $P-x$ and H^E data measured within this work, temperature-dependent parameters for the NRTL model (Renon and Prausnitz, 1968) have been fitted. During the fitting procedure, ideal behavior of the vapor phase was assumed and the following objective function F has been chosen:

$$F = w_1\sum_{i=1}^{n_1}\left|\frac{P_{i,\text{expt}} - P_{i,\text{calc}}}{P_{i,\text{expt}}}\right| + w_2\sum_{i=1}^{n_2}\left|\frac{H^E_{i,\text{expt}} - H^E_{i,\text{calc}}}{|H^E_{i,\text{max}}|}\right| \qquad (1)$$

where w_1 and w_2 are variable weighting factors, n_1 and n_2 are the number of experimental data sets, $P_{i,\text{expt}} - P_{i,\text{calc}}$ is the difference between experimental and calculated pressures, $H^E_{i,\text{expt}} - H^E_{i,\text{calc}}$ is the difference between experi-

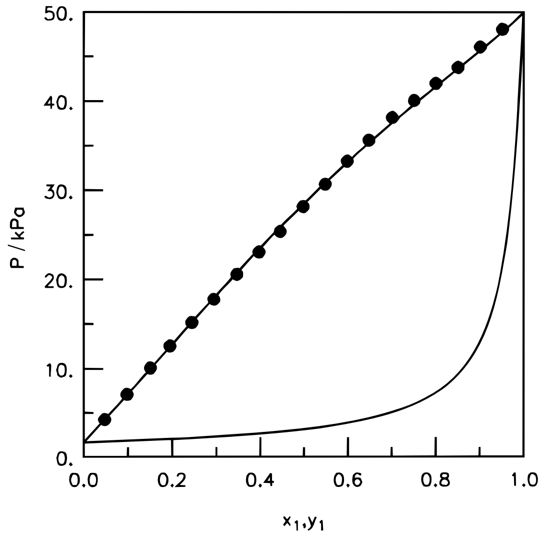

Figure 5. Experimental $P-x$ data (●) and $P-x-y$ behavior calculated by NRTL (—) for the system benzyl alcohol (1) + benzyl benzoate (2) at 453.23 K.

Table 6. Experimental Excess Enthalpies for the System Benzene (1) + Benzyl Benzoate (2) at 363.15 K (1342 kPa) and 413.15 K (1342 kPa)

T/K	x_1	H^E/(J mol^{-1})	x_1	H^E/(J mol^{-1})
363.15	0.1015	11.55	0.7241	5.02
	0.1926	18.24	0.7630	1.29
	0.2748	22.74	0.7995	−1.50
	0.3493	25.87	0.8336	−3.33
	0.4171	25.47	0.8656	−4.46
	0.4792	22.65	0.8957	−4.70
	0.5362	20.05	0.9240	−4.00
	0.5887	16.29	0.9508	−2.91
	0.6372	12.32	0.9761	−1.75
	0.6822	9.29		
413.15	0.1015	−16.08	0.7241	−85.04
	0.1926	−28.69	0.7630	−83.65
	0.2748	−40.48	0.7995	−81.17
	0.3493	−52.75	0.8336	−77.11
	0.4171	−63.28	0.8656	−69.44
	0.4792	−71.47	0.8957	−56.14
	0.5362	−79.74	0.9240	−44.21
	0.5887	−82.87	0.9508	−31.46
	0.6372	−84.59	0.9761	−16.03
	0.6822	−85.81		

Table 7. Experimental Excess Enthalpies for the System Toluene (1) + Benzyl Benzoate (2) at 363.15 K (1342 kPa) and 413.15 K (1410 kPa)

T/K	x_1	H^E/(J mol^{-1})	x_1	H^E/(J mol^{-1})
363.15	0.0440	13.69	0.6870	75.85
	0.0864	24.30	0.7293	70.86
	0.1664	41.98	0.7694	65.43
	0.2407	55.95	0.8074	57.87
	0.3099	66.65	0.8435	49.82
	0.3745	74.25	0.8778	41.33
	0.4350	80.94	0.9105	30.51
	0.4917	84.53	0.9417	20.02
	0.5449	84.78	0.9715	9.56
	0.5951	82.49	0.9859	5.28
	0.6424	79.49		
413.15	0.1069	8.41	0.7496	21.38
	0.2042	16.46	0.8435	14.41
	0.3745	27.85	0.9263	7.36
	0.5187	31.82	0.9642	3.72
	0.6424	29.90		

Table 8. Experimental Excess Enthalpies for the System Phenol (1) + Benzyl Benzoate (2) at 363.15 K (1479 kPa) and 413.15 K (1342 kPa)

T/K	x_1	H^E/(J mol^{-1})	x_1	H^E/(J mol^{-1})
363.15	0.1019	−241.8	0.7250	−760.7
	0.1933	−451.6	0.7639	−684.1
	0.3139	−693.9	0.8175	−555.2
	0.4182	−839.8	0.8661	−420.9
	0.5094	−910.7	0.9105	−285.7
	0.5898	−919.7	0.9510	−155.8
	0.6383	−897.2	0.9762	−73.23
	0.6832	−847.9		
413.15	0.0019	−242.1	0.7250	−559.0
	0.1933	−426.1	0.7639	−484.9
	0.3139	−617.0	0.8175	−392.5
	0.4182	−693.2	0.8661	−291.2
	0.5094	−711.0	0.9105	−191.5
	0.5898	−706.9	0.9510	−103.8
	0.6832	−621.1	0.9762	−47.88

Table 9. Experimental Excess Enthalpies for the System Benzaldehyde (1) + Benzyl Benzoate (2) at 363.15 K (1204 kPa) and 413.15 K (1410 kPa)

T/K	x_1	H^E/(J mol^{-1})	x_1	H^E/(J mol^{-1})
363.15	0.0116	−21.25	0.3830	52.17
	0.0230	−22.20	0.5006	56.95
	0.0456	−18.25	0.6506	57.12
	0.0892	−7.76	0.7757	47.87
	0.1104	−7.58	0.8482	38.78
	0.1515	−2.41	0.8977	27.36
	0.1714	14.48	0.9437	16.03
	0.2831	37.90	0.9725	7.84
413.15	0.0901	16.93	0.7384	46.79
	0.2118	36.68	0.8145	36.89
	0.3199	48.86	0.8827	26.12
	0.4464	54.18	0.9294	16.31
	0.5564	55.30	0.9728	5.53
	0.6529	52.19		

Table 10. Experimental Excess Enthalpies for the System Benzyl Alcohol (1) + Benzyl Benzoate (2) at 363.15 K (997 kPa) and 413.15 K (1893 kPa)

T/K	x_1	H^E/(J mol^{-1})	x_1	H^E/(J mol^{-1})
363.15	0.0452	227.9	0.6930	1041.4
	0.0886	416.5	0.7348	952.1
	0.1703	733.3	0.7743	858.4
	0.2458	964.0	0.8117	752.4
	0.3159	1117.4	0.8471	640.0
	0.3811	1210.1	0.8808	520.3
	0.4419	1251.5	0.9128	399.1
	0.4987	1259.4	0.9433	263.9
	0.5519	1228.1	0.9723	127.2
	0.6018	1176.7	0.9863	57.63
	0.6488	1122.2		
413.15	0.0452	138.1	0.6931	942.2
	0.0886	271.0	0.7348	869.5
	0.1703	503.5	0.7743	786.4
	0.2459	690.5	0.8117	689.5
	0.3159	837.6	0.8472	582.6
	0.3811	947.7	0.8808	475.8
	0.4419	1018.4	0.9128	357.9
	0.4987	1053.0	0.9433	238.5
	0.5519	1057.9	0.9723	124.0
	0.6018	1037.9	0.9863	64.99
	0.6488	999.6		

mental and calculated excess enthalpies, and $H^E_{i,\max}$ is the maximum excess enthalpy value of the corresponding data set. The NRTL parameters were fitted simultaneously with emphasis on the accurate description of the VLE data. The obtained binary parameters for the NRTL model Δg_{ij} as well as the nonrandomness parameters α_{ij} are given in Table 11. The corresponding expression for the temperature dependence is given by the following equation:

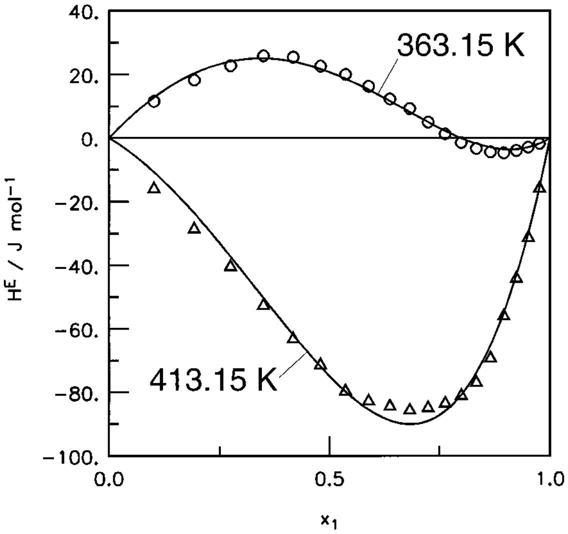

Figure 6. Experimental H^E data at 363.15 K (○) and 413.15 K (△) for the system benzene (1) + benzyl benzoate (2) together with the results of the NRTL model.

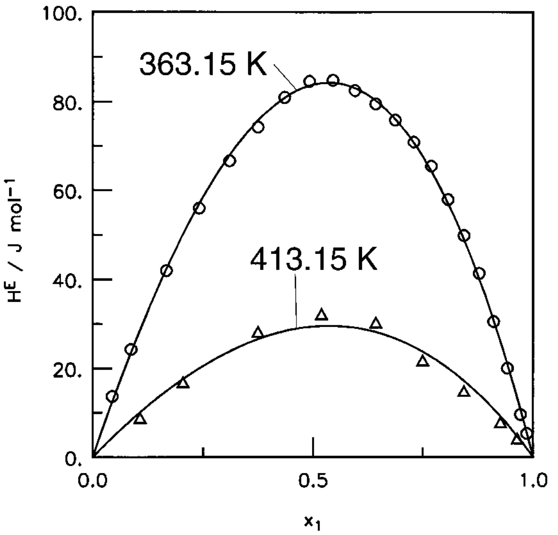

Figure 7. Experimental H^E data at 363.15 K (○) and 413.15 K (△) for the system toluene (1) + benzyl benzoate (2) together with the results of the NRTL model.

$$\Delta g_{ij} = \Delta g^0_{ij} + \Delta g^1_{ij}T + \Delta g^2_{ij}T^2 \qquad (2)$$

for binary systems containing the components i and j.

In Figures 1−10, the experimental VLE and H^E data are compared with the behavior calculated with the help of the NRTL model using the simultaneously fitted parameters. As can be seen in Figures 1−5, the isothermal $P-x$ data are well-described by the NRTL equation, which is true for all of the five systems investigated.

Regarding the H^E data, the systems toluene + benzyl benzoate (Figure 7), benzaldehyde + benzyl benzoate (Figure 9) and benzyl alcohol + benzyl benzoate (Figure 10) show mainly endothermal mixing behavior, while for the systems benzene + benzyl benzoate (Figure 6) and phenol + benzyl benzoate (Figure 8) mainly exothermal excess enthalpies are observed. A change of sign can be noted for the system benzene + benzyl benzoate as well as for the system benzaldehyde + benzyl benzoate at 363.15 K, which in both cases disappears at 413.15 K. While the system benzene + benzyl benzoate at 363.15 K can be well-described by the NRTL model, the course of the experi-

Table 11. NRTL Interaction Parameters Fitted Simultaneously to VLE and H^E Data

system comp (1) + comp (2)	Δg_{12}^0/ (J mol^{-1})	Δg_{12}^1/ (J mol^{-1} K^{-1})	Δg_{12}^2/ (J mol^{-1} K^{-2})	Δg_{21}^0/ (J mol^{-1})	Δg_{21}^1/ (J mol^{-1} K^{-1})	Δg_{21}^2/ (J mol^{-1} K^{-2})	α_{12}
benzene (1) + benzyl benzoate (2)	9.9377×10^3	-3.8466×10	4.2403×10^{-2}	-4.0039×10^3	9.6428	-1.1829×10^{-2}	0.4689
toluene (1) + benzyl benzoate (2)	6.3898×10^2	-5.3966×10^{-1}	3.4995×10^{-3}	4.5904×10^2	-4.6489	2.2727×10^{-3}	0.4636
phenol (1) + benzyl benzoate (2)	-4.7643×10^3	5.8138×10	-8.9818×10^{-2}	1.3128×10^3	-2.7944×10	4.0426×10^{-2}	0.4682
benzaldehyde (1) + benzyl benzoate (2)	1.2205×10^4	-2.9762×10	2.1203×10^{-3}	-1.2208×10^3	-2.1692×10	5.8067×10^{-2}	0.2664
benzyl alcohol (1) + benzyl benzoate (2)	-1.5771×10^4	8.5559×10	-8.3195×10^{-2}	1.5629×10^4	-7.2209×10	6.7915×10^{-2}	0.3142

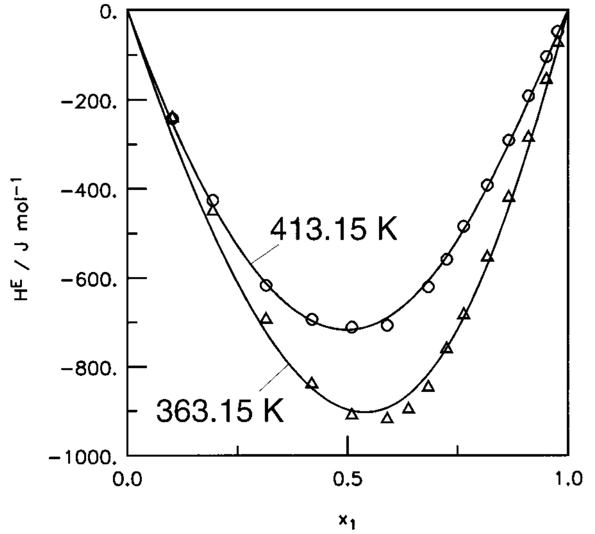

Figure 8. Experimental H^E data at 363.15 K (△) and 413.15 K (○) for the system phenol (1) + benzyl benzoate (2) together with the results of the NRTL model.

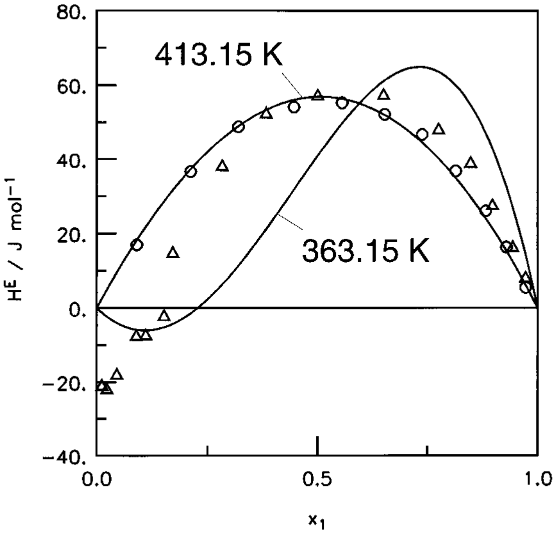

Figure 9. Experimental H^E data at 363.15 K (△) and 413.15 K (○) for the system benzaldehyde (1) + benzyl benzoate (2) together with the results of the NRTL model.

mental data points for the system benzaldehyde + benzyl benzoate at 363.15 K shows an unusual shape in the dilute region. This effect can only be reproduced qualitatively by the NRTL model. A similar effect had been observed previously for the excess enthalpies of the systems benzene + benzaldehyde and toluene + benzaldehyde, together, however, with a reversed temperature dependence.

It has to be noted that for the systems benzene + benzyl benzoate, benzaldehyde + benzyl benzoate, and, in addition, the system toluene + benzyl benzoate the total excess enthalpy values are very small compared to the other systems investigated ($|H^E_{\max}| < 100$ J mol^{-1}). Moreover, H^E values mainly decrease with temperature, as can be seen

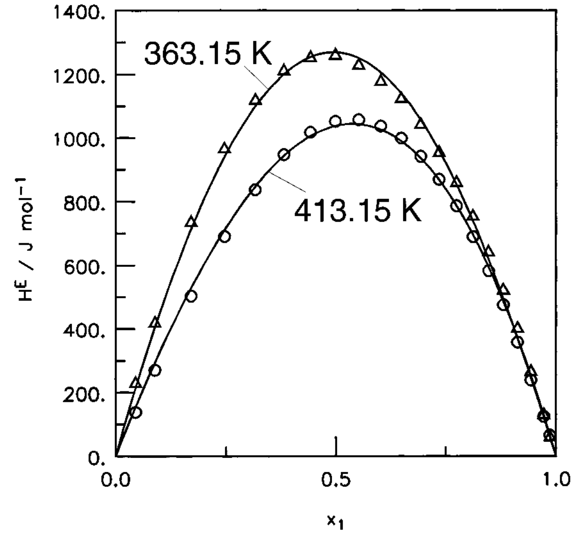

Figure 10. Experimental H^E data at 363.15 K (△) and 413.15 K (○) for the system benzyl alcohol (1) + benzyl benzoate (2) together with the results of the NRTL model.

for the systems toluene + benzyl benzoate, phenol + benzyl benzoate, and benzyl alcohol + benzyl benzoate.

Except for the system benzaldehyde + benzyl benzoate the experimental data is in any case very well described by the temperature-dependent NRTL parameters.

Conclusion

The thermodynamic behavior of five binary aromatic mixtures containing benzyl benzoate has been investigated at high temperatures by measuring reliable VLE and H^E data. The experimental data have been fitted using temperature-dependent NRTL parameters, which allow the simultaneous description of the vapor–liquid equilibrium and excess enthalpy behavior for all of the five systems. Together with reliable experimental data determined previously for aromatic systems with benzaldehyde and phenol, these data form a solid basis for the description of systems containing aromatic components at high temperatures.

The measurement of the required reliable experimental data for the introduction of the aromatic ester group (AC–COO–) into the modified UNIFAC (Dortmund) model has been completed with this work. This will be another important contribution to the revision and extension of this model, besides the revision of the phenol group (AC–OH) and the extension of the model by the aromatic aldehyde group (AC–CHO).

During the following parts of this project, further phase equilibrium and excess enthalpy data of aromatic systems will be measured and reported in future papers. Furthermore, it is scheduled to define more aromatic structural groups, e.g. aromatic acids (AC–COOH) or ethers (AC–O–), for the group contribution method modified UNIFAC (Dortmund).

Acknowledgment

The authors would like to thank B. Werner for technical assistance as well as Dr. K. Fischer and Dr. P. Guilbot, École de Mines, Fontainebleau, France, for technical advice.

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Received for review October 15, 1998. Accepted December 17, 1998.

JE9802524