GENERAL RESEARCH

Application of the Perturbed-Chain SAFT Equation of State to Associating Systems

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The perturbed-chain SAFT (PC−SAFT) equation of state is applied to pure associating components as well as to vapor−liquid and liquid−liquid equilibria of binary mixtures of associating substances. For these substances, the PC−SAFT equation of state requires five pure-component parameters, two of which characterize the association. The pure-component parameters were identified for 18 associating substances by correlating vapor pressure and liquid density data. A comparison to an earlier version of SAFT confirms the good results for pure substances. When only one associating compound is present in a mixture, the PC−SAFT equation of state does not require mixing rules for the association term. Using one binary interaction parameter k_{ij} for the dispersion term only, the model was applied to azeotropic and nonazeotropic vapor−liquid equilibria at low and at high pressures, as well as to liquid−liquid equilibria. Simple mixing and combining rules were adopted for mixtures with more than one associating compound, introducing no additional binary interaction parameter. The simultaneous description of liquid−liquid and vapor−liquid equilibrium was also possible with a single k_{ij} parameter.

1. Introduction

Modeling the phase equilibrium and thermodynamic properties of systems in which molecules exhibit associating interactions remains a challenging problem in chemical industry. The large number of recent studies on equations of state applied to associating systems is an indication of this circumstance. Detailed reviews are given, for example, by Müller and Gubbins,[1] Economou and Donohue,[2] and Wei and Sadus.[3]

It is beyond dispute that associating interactions that contribute significantly to the overall intermolecular forces have to be explicitly taken into account in molecular theories and, further, that physically based equations of state have the potential for correlating and someday even predicting the behavior of such systems. Progress toward this goal has been achieved by applying principles of statistical mechanics. One step along this road was the development of the statistical association fluid theory (SAFT) by Chapman et al.[4,5] The perturbation theory of Wertheim[6−9] is suitable for highly directional interactions and provided the basis for the development of a fairly simple association term.[10−12] Numerous subsequent investigations have aimed at

numerical aspects of solving the appropriate equations or at applications of the theory to real substances (see ref 1).

Huang and Radosz applied the SAFT framework developing a modified SAFT equation of state and determined pure-component parameters for an array of components of industrial relevance.[13,14] This particular version was subsequently often used for academic and for industry applications.[1−3]

Many SAFT models account for the chainlike shape of the molecules only in the repulsive contribution of the equation of state (hard-chain contribution). In contrast, Gross and Sadowski developed the perturbed-chain SAFT (PC−SAFT) equation of state, where the chain-length dependence of the attractive (dispersive) interactions is also taken into account. The PC−SAFT model was shown to be suitable for pure components and mixtures of solvents and gases,[15] as well as for liquid−liquid and vapor−liquid equilibria of polymer systems.[16] The PC−SAFT equation of state is implemented in a common process simulation tool (by Aspen-Tech Inc.). It is further available as coded subroutines[17] and is incorporated in the engineering software PE (Phase Equilibria, developed and maintained by Pfohl, Petkov, and Brunner[18]), which is freely available for scientific and industrial applications.

In this work, the PC−SAFT equation of state is applied to systems in which molecular association prevails. Pure components and mixtures are investigated. As in our earlier studies, we seek an evaluation of the PC−SAFT equation of state by comparing it to

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Table 1. Pure-Component Parameters of the Perturbed-Chain SAFT Equation of State for Associating Components

component i[a]	M_i (g/mol)	m_i	σ_i (Å)	ϵ_i/k (K)	$\kappa^{A_iB_i}$	$\epsilon^{A_iB_i}/k$ (K)	AAD% P^{sat}	AAD% v	T range (K)	ref[b]
					alkanols					
methanol	32.042	1.5255	3.2300	188.90	0.035176	2899.5	2.36	2.01	200−512	1, 2
ethanol	46.069	2.3827	3.1771	198.24	0.032384	2653.4	0.99	0.79	230−516	1, 2
1-propanol	60.096	2.9997	3.2522	233.40	0.015268	2276.8	0.85	1.71	240−537	2
1-butanol	74.123	2.7515	3.6139	259.59	0.006692	2544.6	1.78	1.63	184−563	1, 2
1-pentanol	88.15	3.6260	3.4508	247.28	0.010319	2252.1	0.5	0.52	250−588	2
1-hexanol	102.177	3.5146	3.6735	262.32	0.005747	2538.9	0.58	0.71	228−610	2
1-heptanol	116.203	4.3985	3.5450	253.46	0.001155	2878.5	1.47	1.19	239−632	2
1-octanol	130.23	4.3555	3.7145	262.74	0.002197	2754.8	1.28	0.92	258−652	2
1-nonanol	144.257	4.6839	3.7292	263.64	0.001427	2941.9	0.86	1.23	268−670	2
2-propanol	60.096	3.0929	3.2085	208.42	0.024675	2253.9	0.7	1.25	185−508	2
2-methyl-2-butanol	88.15	2.5487	3.9053	266.01	0.001863	2618.8	0.19	0.44	264−545	2
					water					
water	18.015	1.0656	3.0007	366.51	0.034868	2500.7	1.88	6.83	273−647	1
					amines					
methylamine	31.06	2.3967	2.8906	214.94	0.095103	684.3	0.3	0.53	179−430	3
ethylamine	45.09	2.7046	3.1343	221.53	0.017275	854.7	0.41	1.15	210−456	3
1-propylamine	59.11	2.4539	3.5347	250.52	0.022674	1028.1	0.32	0.7	220−497	3
2-propylamine	59.11	2.5908	3.4777	231.80	0.021340	932.2	0.24	0.22	210−471	3
aniline	93.13	2.6607	3.7021	335.47	0.074883	1351.6	0.79	1.09	267−699	3
					acetic acid					
acetic acid	60.053	1.3403	3.8582	211.59	0.075550	3044.4	2.12	1.36	302−592	1, 2

[a] Two association sites ($N^{site} = 2$) are assumed for all substances. [b] References: (1) *VDI−Wärmeatlas*; VDI−Gesellschaft Verfahrenstechnik und Chemieingenieurwesen (GVC): Düsseldorf, Germany, 1994. (2) Daubert, T. E.; Danner, R. P.; Sibul, H. M.; Stebbins, C. C. *Physical and Thermodynamic Properties of Pure Chemicals: Data Compilation*; Taylor & Francis: Washington, DC, 1989. (3) Chao, J.; Gadalla, N. A. M.; Gammon, B. E. Marsh, K. N.; Rodgers, A. S.; Somayajulu, G. R.; Wilhoit, R. C. *J. Phys. Chem. Ref. Data*, **1990**, *19* (6), 1547−1569.

Table 2. Binary Interaction Parameters k_{ij} Correcting the Cross-Dispersive Interactions

binary system	k_{ij}[a] PC−SAFT	k_{ij}[a] SAFT
methanol−isobutanol	0.05	0.03
methanol−cyclohexane	0.051	0.044
methanol−1-octanol	0.020	0.034
ethanol−n-butane	0.028	0.021
1-propanol−benzene	0.020	0.005
1-propanol−ethylbenzene	0.023	0.011
2-propanol−benzene	0.021	0.007
1-butanol−n-butane	0.015	0.025
1-pentanol−benzene	0.0135	0.0065
water−1-pentanol	0.016	−0.069

[a] $k_{ij} = k_{ji}$, nonzero for $i \neq j$.

the SAFT version of Huang and Radosz[13,14] (hereafter called SAFT for short).

2. Perturbed-Chain SAFT Equation of State

The PC−SAFT equation of state was derived and described in detail by Gross and Sadowski.[15] In terms of the compressibility factor Z, the equation of state is given as the sum of the ideal gas contribution ($Z^{id} = 1$), the hard-chain term (hc), the dispersive part (disp), and the contribution due to association (assoc) according to

$$Z = 1 + Z^{hc} + Z^{disp} + Z^{assoc} \qquad (1)$$

where Z^{assoc} denotes the contribution of the associating interactions to the compressibility factor (details given in refs 5 and 14). The effect of multipole interactions (such as dipole−dipole forces) is not separately taken into account in eq 1. This contribution will be the subject of further investigations.

Two pure-component parameters determine the associating interactions between the association site A_i and B_i of a pure component i: the association energy

$\epsilon^{A_iB_i}/k$ and the effective association volume $\kappa^{A_iB_i}$. It is generally far from trivial to obtain cross-association parameters between two different associating substances i and j. For many systems, however, approximate values for cross-association parameters can be determined from pure-component association parameters. Simple combining rules for cross-association were suggested by Wolbach and Sandler,[19] from a consideration of gas-phase association in the low-pressure limit, as

$$\epsilon^{A_iB_j} = \frac{1}{2}(\epsilon^{A_iB_i} + \epsilon^{A_jB_j}) \qquad (2)$$

$$\kappa^{A_iB_jA} = \sqrt{\kappa^{A_iB_i}\kappa^{A_jB_j}}\left(\frac{\sqrt{\sigma_{ii}\sigma_{jj}}}{{}^1\!/_2(\sigma_{ii} + \sigma_{jj})}\right)^3 \qquad (3)$$

These rules will be used in this work. Within the scope of this investigation, no binary correction parameters are introduced in these combining rules. Hence, only one binary interaction parameter, k_{ij}, is permitted, which corrects the dispersive interactions[15].

3. Pure-Component Parameters for Associating Compounds

All associating components are assigned two association sites (often referred to as the 2B model[13]). Although this is a reasonable assumption for some species (such as alcohols), it is a considerable simplification for other compounds—in particular for water. A study of Economou and Tsonopoulos[20] indicates that water is best represented with a four-site treatment, whereas Suresh and Elliott[21] found the two-site model to perform at least as well. For simplicity at this point, we follow the latter study. (Calculation results from the SAFT version of Huang and Radosz, which are shown later in this work
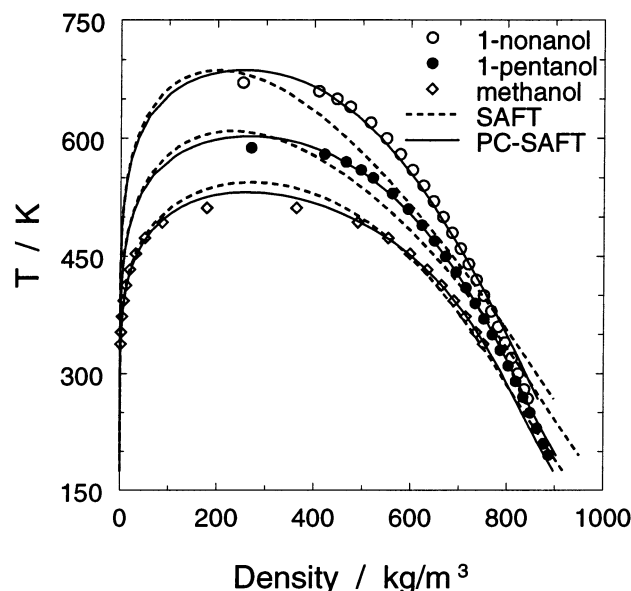
Figure 1. Saturated liquid and vapor densities for methanol, 1-pentanol, and 1-nonanol. Comparison of PC−SAFT (solid lines) and SAFT (dashed lines) to experimental data.
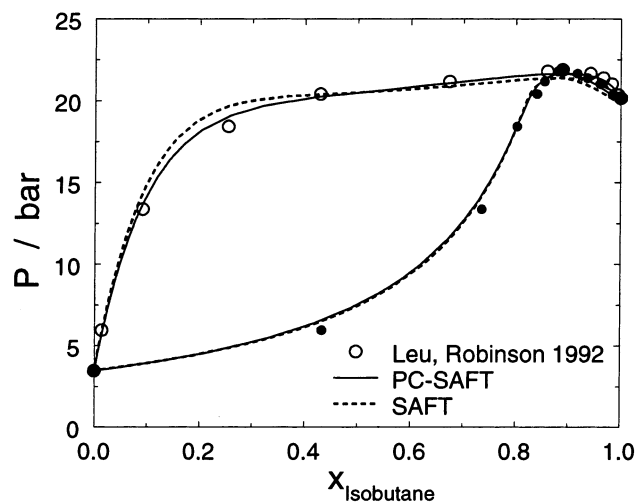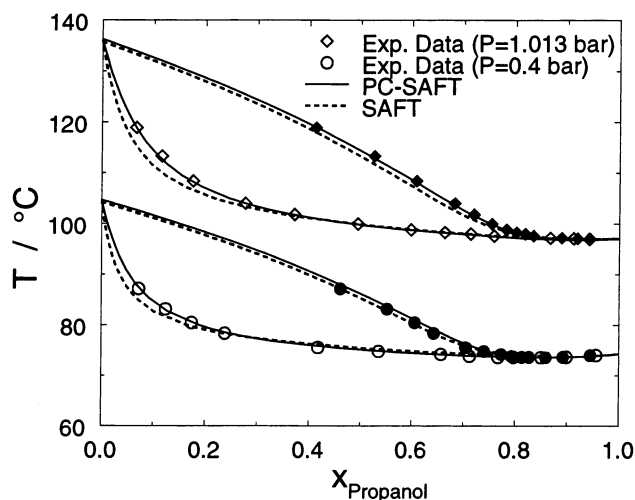

Figure 2. Vapor−liquid equilibria of methanol−isobutane at $T = 100$ °C. Comparison of experimental data[23] to calculation results of PC−SAFT ($k_{ij} = 0.05$) and SAFT ($k_{ij} = 0.03$).

Figure 3. Vapor−liquid equilibria of 1-propanol−ethylbenzene at two pressures. Comparison of experimental data[24] to calculation results of PC−SAFT ($k_{ij} = 0.023$) and SAFT ($k_{ij} = 0.011$).
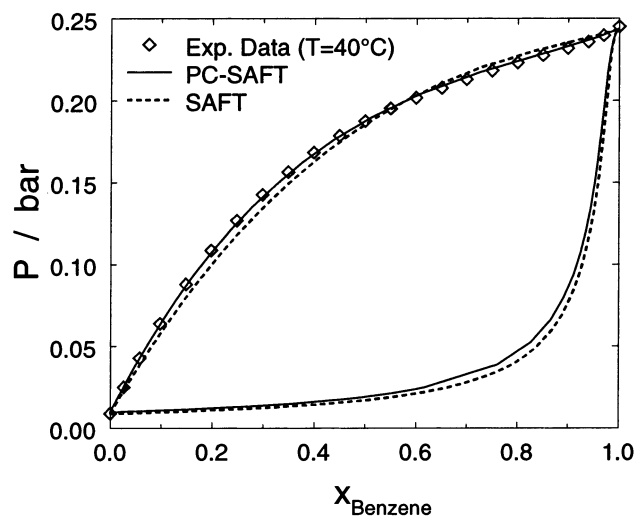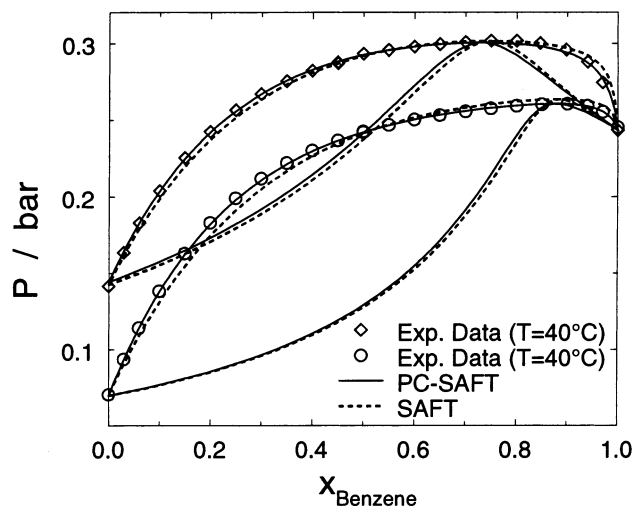

Figure 4. Vapor−liquid equilibria of 1-pentanol−benzene at $T = 40$ °C. Comparison of experimental data[25] to calculation results of PC−SAFT ($k_{ij} = 0.0135$) and SAFT ($k_{ij} = 0.0065$).

for comparison, were obtained using three association sites for water.[13])

The pure-component parameters of associating substances were identified by simultaneously fitting vapor pressure data and liquid density data. A total of five parameters must be adjusted for any associating component i, namely, the segment diameter σ_i, the segment number m_i, the segment energy parameter ϵ_i/k, the association energy $\epsilon^{A_iB_i}/k$, and the effective association volume $\kappa^{A_iB_i}$. Table 1 gives these parameters for some alkanols, amines, acetic acid, and water. Dimensionless absolute average deviations (AAD%) of vapor pressures and liquid densities, along with the temperature range covered by the literature data, are also listed.

4. Results

This section presents correlation results for pure components as well as for vapor−liquid and liquid−liquid equilibria of binary mixtures of associating substances obtained with the perturbed-chain SAFT

equation of state. Mixtures containing one associating compound (self-association) are investigated in section 4.2; mixtures of two associating substances (cross-association) are discussed in section 4.3. Table 1 gives the kij parameters of all mixtures investigated.

4.1. Pure Associating Substances. The absolute average deviations (AAD%) of the PC−SAFT equation of state from experimental vapor pressure and density data in the majority of cases range below 2% (Table 1). Water is an exception here. Average deviations of 6.8% in liquid density and 1.88% in vapor pressure were obtained. Various assumptions inherent in the molecular model adopted here are specifically severe for water. Associating interactions of water lead to cyclic chainlike molecular topologies, which are currently not taken into account in the association term.[2] Furthermore, the dipole−dipole interactions, are not separately taken into account in the present form of the PC−SAFT equation of state. These directional interactions are known to have a significant impact on the thermophysical behavior of water.[22]

Figure 1 shows the vapor−liquid equilibrium of three alkanols in a $T-\rho$ diagram. The densities of the coexisting phases are well-described by the PC−SAFT model.

Figure 5. Vapor−liquid equilibria of 2-propanol−benzene (◇) and 1-propanol−benzene (○) at T = 40 °C. Comparison of experimental data[25] to calculation results of PC−SAFT (2-propanol−benzene, k_{ij} = 0.021; 1-propanol−benzene, k_{ij} = 0.020) and SAFT (2-propanol−benzene, k_{ij} = 0.007; 1-propanol−benzene, k_{ij} = 0.005).
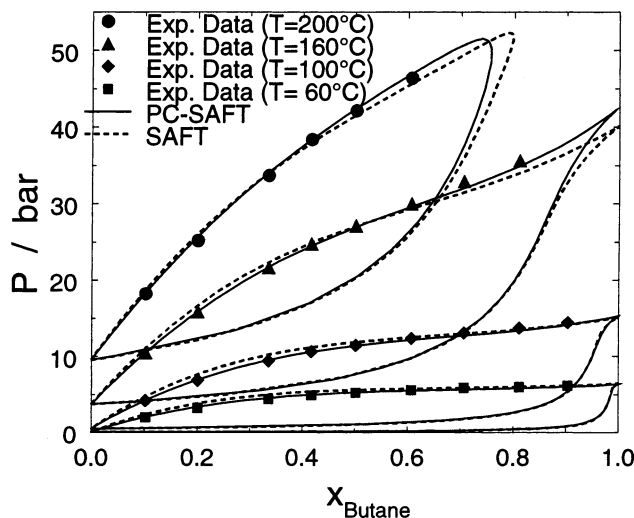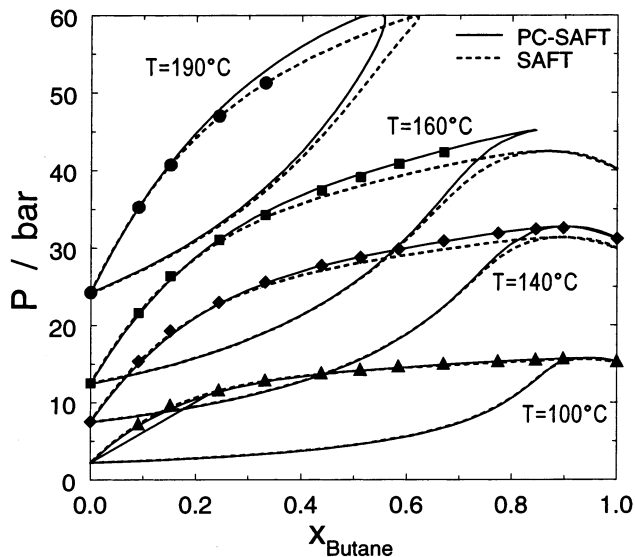

Figure 6. Vapor−liquid equilibria of 1-butanol−n-butane at four temperatures. Comparison of experimental data[26] to calculation results of PC−SAFT (k_{ij} = 0.015) and SAFT (k_{ij} = 0.025).

Results of the SAFT equation of state obtained with pure-component parameters determined by Huang and Radosz[13] are given for comparison. It becomes apparent from Figure 1 that an improved description of dispersive interactions also leads to better results for associating compounds.

4.2. Mixtures Containing One Associating Substance. Mixtures with only one associating substance (self-association) do not require mixing rules for the association term. Calculations for such mixtures are suitable for investigating whether the association term contributes to the total Helmholtz energy (in eq 1) at the right order of magnitude.

Figures 2−5 show vapor−liquid equilibria of binary, self-associating mixtures in which both compounds are below their critical point. Most of the systems displayed show azeotropic behavior. Mixtures in which one compound is above its critical point are given in Figures 6 and 7. Some of the improvement of the PC−SAFT equation of state over the SAFT model is actually due to an improved description of pure components. Figure 7, for example, shows deficiencies of the SAFT equation

Figure 7. Vapor−liquid equilibria of ethanol−n-butane at four temperatures. Comparison of experimental data[26] to calculation results of PC−SAFT (k_{ij} = 0.028) and SAFT (k_{ij} = 0.021).
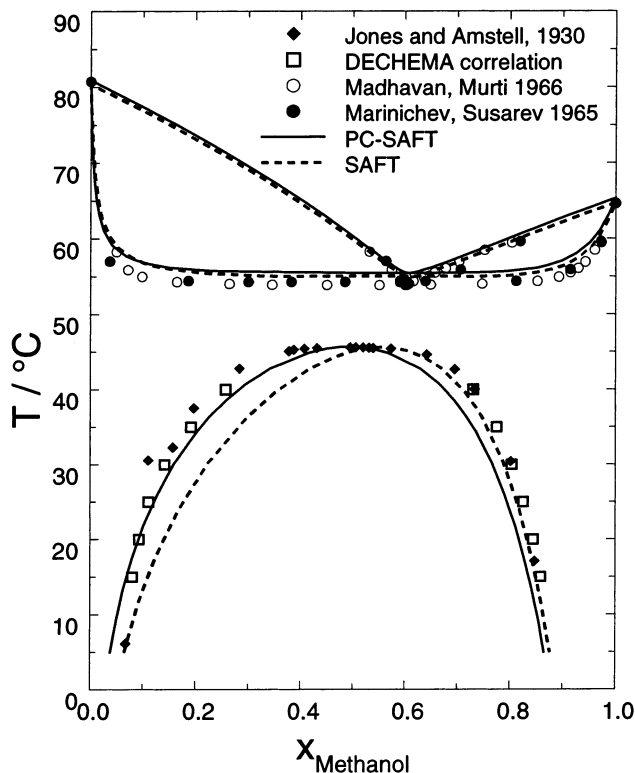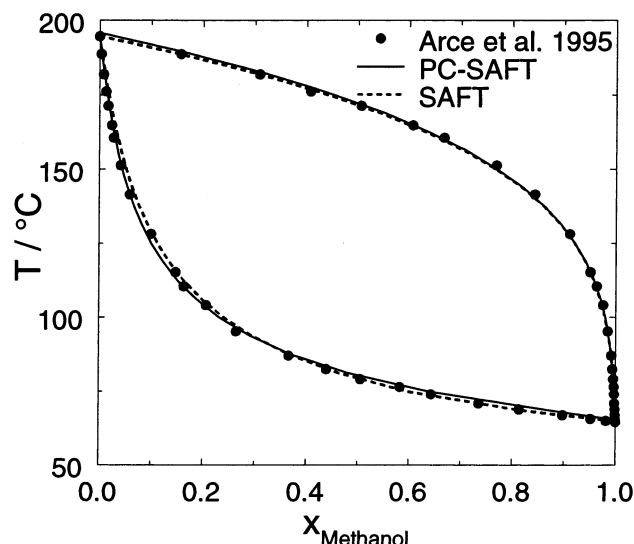

Figure 8. Isobaric vapor−liquid and liquid−liquid equilibria of methanol−cyclohexane at P = 1.013 bar. Comparison of experimental data[27−30] to calculation results of PC−SAFT (k_{ij} = 0.051) and SAFT (k_{ij} = 0.044).

of state in reproducing the vapor pressure of butane at higher temperatures (T = 140 °C). Furthermore, a subcritical system is predicted at 160 °C, although the critical point of butane is about 8 K lower.

The phase behavior of a mixture of methanol and cyclohexane at normal pressure is displayed in Figure 8. This system exhibits an azeotropic vapor−liquid equilibrium at higher temperatures and shows a liquid−liquid equilibrium at lower temperatures. The k_{ij} parameters of the PC−SAFT equation of state and of the SAFT model were adjusted to the highest available liquid−liquid demixing temperature (close to the critical

Figure 9. Isobaric vapor−liquid equilibria of methanol−1-octanol at $P = 1.013$ bar. Comparison of experimental[31] data to calculation results of PC−SAFT ($k_{ij} = 0.020$) and SAFT ($k_{ij} = 0.034$).
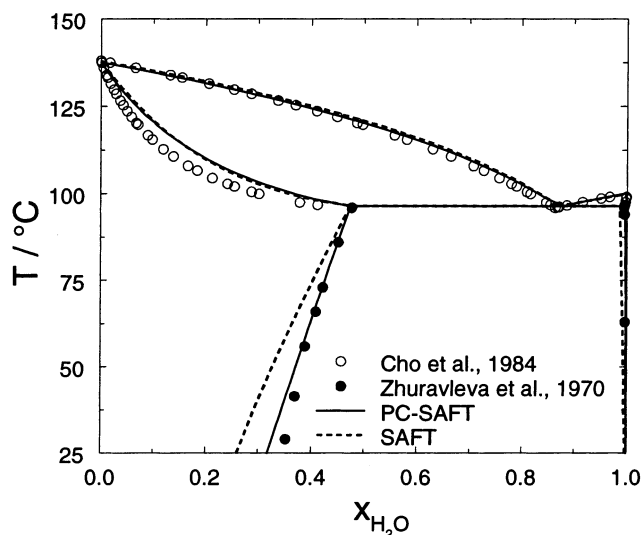
Figure 10. Isobaric, heteroazeotropic vapor−liquid and liquid−liquid equilibria of water−1-pentanol at $P = 1.013$ bar. Comparison of experimental data[32,33] to calculation results of PC−SAFT ($k_{ij} = 0.016$) and SAFT ($k_{ij} = -0.069$).

point of the liquid−liquid equilibrium at the given pressure). The vapor−liquid equilibrium is well-described using this k_{ij} parameter. Figure 8 indicates that the association term of SAFT-type equations of state is a powerful expression that enables also the PC−SAFT model to correlate associating mixtures with only one binary interaction parameter covering wide temperature ranges.

4.3. Mixtures with Two Associating Substances. The modeling of mixtures in which associating interactions occur between different substances is particularly demanding for molecular theories. Also, the calculation results for cross-associating systems depend significantly on the combining rules (here, eqs 2 and 3).

A $T−x$ diagram of methanol and 1-octanol at $P = 1.013$ bar is displayed in Figure 9. The two models perform equally well for this system.

The phase behavior of water and 1-pentanol at $P = 1.013$ bar is given in Figure 10 in a $T−x$ diagram. The system shows a liquid−liquid equilibrium at lower temperatures and a heteroazeotropic vapor−liquid equi-

librium. The k_{ij} parameters of both the PC−SAFT equation of state and the SAFT model were fitted to the 1-pentanol-rich liquid concentration of the vapor−liquid−liquid equilibrium. The results of both models show some deviations from the vapor−liquid data. The PC−SAFT equation of state is found to agree well with the liquid−liquid data.

5. Conclusions

The perturbed-chain SAFT equation of state was used to model phase equilibria of associating components. The pure-component parameters of 18 associating substances were determined by simultaneously fitting vapor pressure and liquid density data. Good correlation results were obtained for alcohols, amines, and acetic acid, as a comparison to an earlier SAFT version proposed by Huang and Radosz confirms. Deviations of 6.83% in liquid density and 1.88% in vapor pressure were observed for water. Mixtures with one associating compound do not require mixing rules in the association term of the PC−SAFT equation of state. Both vapor−liquid and liquid−liquid equilibria were calculated for mixtures of this type using one constant k_{ij} parameter that corrects the dispersive interactions. Liquid−liquid and vapor liquid equilibria were also calculated for mixtures with two associating substances. Simple combination rules were adopted for the cross-associating interactions in this study; no additional binary interaction parameter was thereby introduced. A constant k_{ij} parameter correcting the contribution due to dispersion was used in the correlation of vapor−liquid and liquid−liquid demixing.

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