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Phase Equilibria of HFC Mixtures: Binary Mixtures of Trifluoromethane + 1,1-Difluoroethane and Trifluoromethane + 1,1,1-Trifluoroethane at 283.15 and 293.15 K

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Isothermal vapor−liquid equilibria for the binary mixtures of trifluoromethane (R-23) + 1,1-difluoroethane (HFC-152a) and trifluoromethane + 1,1,1-trifluoroethane (HFC-143a) were measured at 283.15 and 293.15 K. The experiments were carried out with a circulation-type equilibrium apparatus with the measurement of temperature, pressure, and the compositions of the liquid and vapor phases. The experimental data were correlated with the Peng−Robinson equation of state using the Wong and Sandler mixing rules and the Carnahan−Starling−Desantis (CSD) equation of state. Calculated results with these equations showed good agreement with the experimental data, but the Peng−Robinson equation of state gave more accurate predictions for the binary systems. Azeotropic behavior has not been found in either of these mixtures.

Introduction

Chlorofluorocarbons (CFCs) have been widely used as refrigerants, blowing agents, propellants, or cleaning agents because of their outstanding properties. However, production and use of CFCs are currently being phased out under the international agreement Montreal Protocol because of global environment concerns. HFC mixtures have been considered as promising candidates for the replacement of CFC compounds, since their ozone depletion potentials are zero. Vapor−liquid equilibrium data are required to evaluate the performance of refrigeration cycles and to determine their optimal compositions. In this study, we measured VLE data for two binary systems of trifluoromethane (R-23) + 1,1-difluoroethane (HFC-152a) and + 1,1,1-trifluoroethane (HFC-143a) at 283.15 and 293.15 K. Experimental data were correlated with the Peng−Robinson equation of state[1] using the Wong and Sandler mixing rules[2] and the Carnahan−Starling−Desantis (CSD) equation of state.[3]

Experimental Section

Chemicals. HFC-23, HFC-152a, and HFC-143a were supplied by DuPont with a purity of 99.9 mass %. The purities of the chemicals were guaranteed by the manufacturer, and they were used without any further purification.

Apparatus. Vapor−liquid equilibria were measured in a circulation-type apparatus in which both vapor and liquid phases were continuously circulated. The experimental apparatus used in this work is the same as that used in our previous work.[4−9] The schematic diagram of the apparatus is shown in Figure 1. The equilibrium cell is a type-316 stainless steel vessel with an inner volume of about 85 cm^3. In its middle part, a pair of Pyrex glass windows of 20-mm thickness was installed so that the liquid level, mixing and circulating behaviors, and critical phenomena could be observed during operation. A stirrer, rotated at variable speeds by an external magnet, was installed to accelerate the attainment of the equilibrium state and reduce any concentration gradients. The temperature of the equilibrium cell in the water bath was maintained constant by a circulator from Jeio Tech (Korea) with a stability of ±0.01 K. The temperature in the cell was measured with a platinum resistance sensor and digital temperature indicator model F250MkII precision thermometer from Automatic Systems Laboratories Ltd. (U.K.). They were calibrated by a NAMAS accredited calibration laboratory. Its uncertainty was estimated to be ±0.01 K. The total uncertainty in temperature measurements is estimated to be 0.05 K, including the precision of the standard thermometer and the fluctuation of the temperature controller. The pressure was measured with a pressure transducer model XPM60 and digital pressure calibrator model PC 106 from Beamax, Finland. Pressure calibrations are traceable to National Standards (Finland, Center for Metrology and Accreditation certification no. M-95P077, 14.11.1995, M-M 730, 16.11.1995, and M-95P078, 16.11.1995), and the uncertainty was estimated to be ±5 kPa. The vapor and liquid phases in the equilibrium cell were continuously recirculated by a dual-head circulation pump (Milton Roy). When equilibrium was reached, the circulation pump was stopped and the pressure was measured. Then, the vapor and liquid samples were withdrawn from the recycling loop and injected on-line into a Gow-Mac model 550P gas chromatograph. The gas chromatograph was equipped with a thermal conductivity detector and a Porapak Q column (Alltech). The experimental data at the equilibrium state were measured at least five times in order to ensure the repeatability. The composition uncertainty of the liquid and vapor phase composition is estimated to be within ±0.2 mol %, including the margin of error, the reproducibility of the GC, and the uncertainty of the calibration curve.

Experimental Procedure. Experiments for a binary system were performed by the following procedure. The whole system was evacuated by a vacuum pump to remove the inert gases. A target amount of HFC-152a (or HFC-

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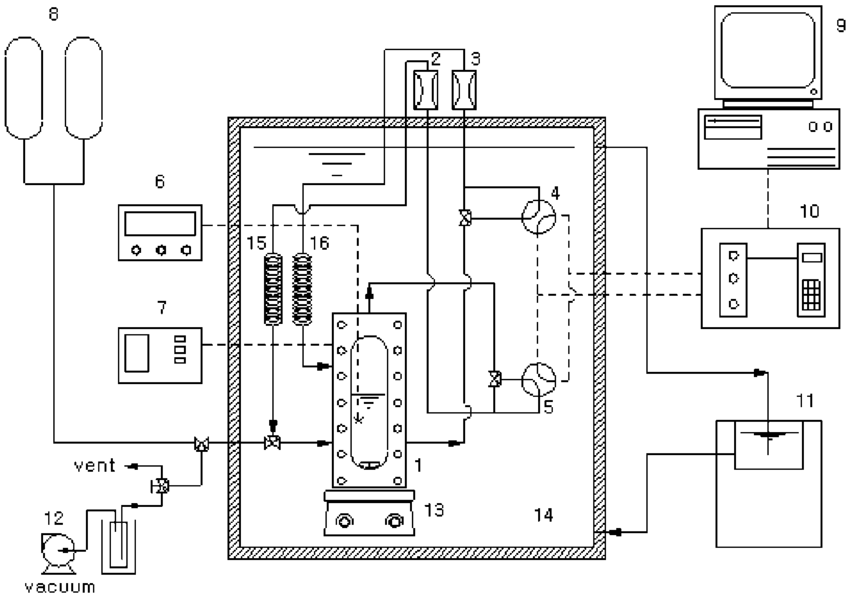

Figure 1. Schematic diagram of the experimental apparatus: 1, equilibrium cell; 2, vapor circulation pump; 3, liquid circulation pump; 4, liquid sampling valve; 5, vapor sampling valve; 6, temperature indicator; 7, pressure indicator; 8, sample reservoir; 9, computer; 10, gas chromatograph; 11, circulator; 12, vacuum pump; 13, magnetic stirrer; 14, constant temperature water bath; 15, vapor-phase heat exchanger; 16, liquid-phase heat exchanger.

Table 1. Characteristic Properties of the Chemicals

component	T_c/K	P_c/MPa	ω	source
HFC-23	299.07	4.836	0.2634	REFPROP 6.01[14]
HFC-152a	386.41	4.517	0.2752	
HFC-143a	346.04	3.776	0.2611	

143a) was introduced into the cell, and then the temperature of the entire system was held constant by controlling the temperature of the water bath. After the desired temperature was attained, the pressure of the pure component was measured. Then, an amount of HFC-23 was supplied to the cell from a charging cylinder. The mixture in the cell was stirred continuously by a magnetic stirrer for over 1 h. Both vapor and liquid phases were circulated by the high-pressure pump until an equilibrium state was established. It was believed that 1 h or more was sufficient to obtain thermal equilibrium between the cell fluid and the thermostatic bath. After equilibration, the pressure was measured and then vapor and liquid samples were taken from the recycling lines by the vapor and liquid sampling valves. The sample compositions were measured by immediate injection into the gas chromatograph which was connected on-line to both the vapor and liquid sampling valves. The gas chromatograph had been calibrated with pure components of known purity and with standard mixtures of known composition that were prepared gravimetrically.

Table 3. Comparison of the Measured Pure Component Vapor Pressures with the Calculated Values from the Database REFPROP 6.01[14]

		P/MPa			
component	*T*/K	exp	cal	ΔP^a/MPa	$\Delta P/P^b$/(%)
HFC-23	283.15	3.275	3.274	0.001	0.015
	293.15	4.203	4.202	0.001	0.021
HFC-152a	283.15	0.374	0.373	0.001	0.214
	293.15	0.515	0.513	0.002	0.543
HFC-143a	283.15	0.838	0.836	0.002	0.239
	293.15	1.104	1.105	0.001	0.091

[a] $\Delta P = |P_{\rm exp} - P_{\rm cal}|$. [b] $\Delta P/P = |(P_{\rm exp} - P_{\rm cal})/P_{\rm exp}| \times 100$.

Results and Correlation

Isothermal vapor−liquid equilibria were measured for the binary systems HFC-23 + HFC-152a and HFC-23 + HFC-143a at 283.15 and 293.15 K. The VLE data were correlated with the Peng−Robinson equation of state[1] and the CSD equation of state.[3] The Peng−Robinson equation of state is expressed as follows:

$$P = \frac{RT}{v-b} - \frac{a(T)}{v(v+b)+b(v-b)} \tag{1}$$

with

$$a(T) = (0.457232R^2T_c^{\,2}/P_c)\alpha(T) \tag{2}$$

$$b = 0.077796RT_c/P_c \tag{3}$$

$$\alpha(T) = [1 + \kappa(1 - T_r^{\,0.5})]^2 \tag{4}$$

$$\kappa = 0.37464 + 1.54226\omega - 0.26992\omega^2 \tag{5}$$

where the parameter *a* is a function of temperature, *b* is constant, κ is a constant characteristic of each substance, ω is the acentric factor, *P*/MPa is pressure, P_c/MPa is the critical pressure, *T*/K is absolute temperature, T_c/K is the

Table 2. Pure Fluid Coefficients of the CSD Equation of State

component	$\alpha_0{}^a$	$10^3\alpha_1{}^b$	$10^6\alpha_2{}^c$	$\beta_0{}^d$	$10^4\beta_1{}^e$	$10^7\beta_2{}^f$	source
HFC-23	1743.890	−3.525 950	−1.127 738 8	0.090 205 5	−1.256 023 7	−0.506 749 8	REFPROP 5.0[13]
HFC-152a	3198.626	−2.961 344	−0.321 897 35	0.133 264 3	−2.036 328 8	0.777 250 9	
HFC-143a	2763.909	−2.509 056	−1.797 107 7	0.133 152 6	−1.589 537 9	−0.583 310 5	

[a] kJ m^3 kg^{-1} mol^{-2}. [b] kJ m^3 kg^{-1} mol^{-2} K^{-1}. [c] kJ m^3 kg^{-1} mol^{-2} K^{-2}. [d] m^3 kg^{-1} mol^{-1}. [e] m^3 kg^{-1} mol^{-1} K^{-1}. [f] m^3 kg^{-1} mol^{-1} K^{-2}.

Table 4. Vapor−Liquid Equilibrium Measurements for the HFC-23 (1) + HFC-152a (2) System

experimental data			Peng−Robinson equation of state				CSD equation of state			
P_{exp}/MPa	x_1	$y_{1,exp}$	P_{cal}/MPa	$y_{1,cal}$	ΔP^a/MPa	$\Delta y_1{}^b$	P_{cal}/MPa	$y_{1,cal}$	ΔP^a/MPa	$\Delta y_1{}^b$
					T/K = 283.15					
0.374	0.000	0.000	0.372	0.000	0.001	0.000	0.373	0.000	0.001	0.000
0.427	0.024	0.111	0.421	0.127	0.006	−0.015	0.423	0.131	0.003	−0.019
0.540	0.082	0.321	0.540	0.341	0.000	−0.020	0.546	0.347	−0.006	−0.026
0.632	0.124	0.430	0.631	0.450	0.001	−0.020	0.638	0.455	−0.007	−0.025
0.867	0.230	0.598	0.872	0.631	−0.005	−0.033	0.880	0.632	−0.013	−0.034
1.249	0.384	0.749	1.256	0.776	−0.007	−0.027	1.255	0.774	−0.006	−0.025
1.667	0.525	0.838	1.646	0.855	0.021	−0.017	1.632	0.853	0.035	−0.016
2.330	0.754	0.931	2.351	0.935	−0.021	−0.003	2.314	0.935	0.016	−0.004
2.695	0.851	0.960	2.686	0.960	0.009	0.000	2.639	0.962	0.056	−0.001
3.275	1.000	1.000	3.291	1.000	−0.016	0.000	3.211	1.000	0.063	0.000
					T/K = 293.15					
0.515	0.000	0.000	0.513	0.000	0.002	0.000	0.512	0.000	0.003	0.000
0.618	0.045	0.173	0.630	0.200	−0.012	−0.027	0.628	0.200	−0.010	−0.027
0.728	0.083	0.297	0.731	0.321	−0.003	−0.023	0.728	0.322	0.000	−0.025
0.890	0.140	0.434	0.885	0.453	0.005	−0.018	0.881	0.457	0.009	−0.023
1.195	0.244	0.597	1.178	0.608	0.017	−0.011	1.174	0.618	0.021	−0.021
1.570	0.377	0.726	1.574	0.728	−0.004	−0.002	1.575	0.743	−0.006	−0.017
2.106	0.550	0.835	2.131	0.826	−0.026	0.009	2.148	0.844	−0.042	−0.009
2.505	0.654	0.882	2.502	0.870	0.003	0.012	2.524	0.888	−0.019	−0.006
3.379	0.851	0.952	3.337	0.941	0.042	0.010	3.390	0.945	−0.011	0.006
4.203	1.000	1.000	4.210	1.000	−0.007	0.000	4.078	1.000	0.125	0.000

[a] $\Delta P = P_{exp} - P_{cal}$. [b] $\Delta y_1 = y_{exp} - y_{cal}$.

Table 5. Vapor−Liquid Equilibrium Measurements for the HFC-23 (1) + HFC-143a (2) System

experimental data			Peng−Robinson equation of state				CSD equation of state			
P_{exp}/MPa	x_1	$y_{1,exp}$	P_{cal}/MPa	$y_{1,cal}$	ΔP^a/MPa	$\Delta y_1{}^b$	P_{exp}/MPa	$y_{1,exp}$	ΔP^a/MPa	$\Delta y_1{}^b$
					T/K = 283.15					
0.838	0.000	0.000	0.839	0.000	−0.001	0.000	0.838	0.000	0.001	0.000
0.968	0.070	0.165	0.964	0.165	0.005	0.000	0.978	0.177	−0.010	−0.011
1.108	0.146	0.298	1.110	0.313	−0.001	−0.016	1.134	0.323	−0.025	−0.026
1.341	0.255	0.466	1.335	0.479	0.006	−0.013	1.362	0.482	−0.021	−0.015
1.593	0.375	0.595	1.605	0.617	−0.012	−0.022	1.622	0.611	−0.029	−0.017
1.862	0.485	0.699	1.867	0.713	−0.005	−0.014	1.896	0.704	−0.035	−0.005
2.106	0.580	0.771	2.101	0.779	0.005	−0.008	2.091	0.772	0.015	−0.001
2.385	0.690	0.839	2.385	0.844	0.000	−0.005	2.359	0.840	0.026	−0.001
2.655	0.784	0.892	2.634	0.891	0.021	0.001	2.600	0.892	0.055	0.001
3.275	1.000	1.000	3.291	1.000	−0.016	0.000	3.211	1.000	0.063	0.000
					T/K = 293.15					
1.104	0.000	0.000	1.111	0.000	0.007	0.000	1.107	0.000	0.003	0.000
1.331	0.103	0.214	1.327	0.212	−0.004	−0.003	1.357	0.227	0.026	0.013
1.506	0.178	0.335	1.503	0.340	−0.003	0.005	1.544	0.352	0.038	0.016
1.801	0.296	0.491	1.802	0.501	0.002	0.011	1.847	0.503	0.047	0.013
2.100	0.402	0.606	2.101	0.616	0.001	0.010	2.133	0.611	0.034	0.005
2.522	0.550	0.727	2.554	0.738	0.031	0.012	2.553	0.730	0.031	0.003
2.909	0.664	0.806	2.916	0.811	0.007	0.004	2.895	0.806	−0.014	0.000
3.286	0.759	0.866	3.238	0.864	−0.047	−0.002	3.200	0.864	−0.085	−0.001
3.804	0.898	0.942	3.746	0.936	−0.058	−0.006	3.681	0.943	−0.123	0.001
4.203	1.000	1.000	4.210	1.000	0.007	0.000	4.078	1.000	−0.125	0.000

[a] $\Delta P = P_{exp} - P_{cal}$. [b] $\Delta y_1 = y_{exp} - y_{cal}$.

critical temperature, T_r is the reduced temperature, and v/cm^3·mol^{-1} is the molar volume. The Wong−Sandler mixing rules[2] were used to obtain equation of state parameters for a mixture from those of the pure components. These mixing rules for a cubic equation of state can be written as

$$b_m = \frac{\sum_i \sum_j x_i x_j (b - a/RT)_{ij}}{(1 - A_\infty^E/CRT - \sum_i x_i a_i/RTb_i)} \tag{6}$$

with

$$(b - a/RT)_{ij} = \frac{1}{2}[(b - a/RT)_i + (b - a/RT)_j](1 - k_{ij}) \tag{7}$$

and

$$\frac{a_m}{b_m} = \sum_i x_i \frac{a_i}{b_i} + \frac{A_\infty^E}{C} \tag{8}$$

and where C is a numerical constant equal to $\ln(\sqrt{2} - 1)/\sqrt{2}$ for the Peng−Robinson equation used in this work. Also, A_∞^E is an excess Helmholtz free energy model at infinite pressure which can be equated to a low-pressure excess Gibbs free energy model;[10] in this study we use the NRTL model[11] given by

$$\frac{A_\infty^E}{RT} = \sum_i x_i \frac{\sum_j x_j G_{ji} \tau_{ji}}{\sum_k x_k G_{ki}} \tag{9}$$

with

$$G_{ij} = \exp(-\alpha_{ij}\tau_{ji}) \quad \text{and} \quad \tau_{ij} = A_{ij}/(RT) \tag{10}$$

Table 6. Values of Binary Parameters and Average Deviations of *P* and *y*

	Peng−Robinson EOS[a]			CSD EOS		
T/K	binary parameters	δP^b/%	δy^c	binary parameters	δP^b/%	δy^c
	HFC-23/HFC-152a					
283.15	$K_{ij} = 0.086$					
	$A_{ij} = -1.889$	0.597	0.014	$f_{ij} = -0.0179$	1.196	0.015
	$A_{ji} = 1.513$					
293.15	$K_{ij} = -0.061$					
	$A_{ij} = -1.929$	0.773	0.011	$f_{ij} = -0.0170$	1.142	0.013
	$A_{ji} = 3.489$					
	HFC-23/HFC-143a					
283.15	$K_{ij} = 0.212$					
	$A_{ij} = -1.465$	0.373	0.008	$f_{ij} = 0.0007$	1.435	0.008
	$A_{ji} = 0.412$					
293.15	$K_{ij} = 0.200$					
	$A_{ij} = -1.145$	0.592	0.005	$f_{ij} = -0.0001$	1.946	0.005
	$A_{ji} = -0.091$					

[a] The units of A_{ij} and A_{ji} are kJ·mol^{-1}. [b] $\delta P = 1/N\sum|(P_{exp} - P_{cal})/P_{exp}| \times 100$. [c] $\delta y = 1/N\sum|y_{exp} - y_{cal}|$.

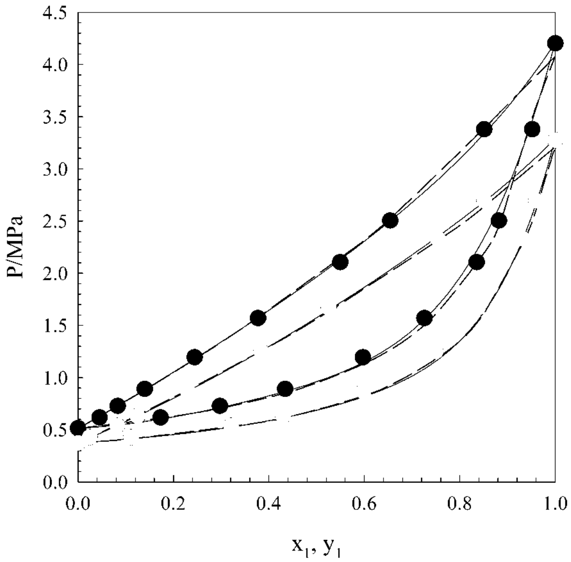

Figure 2. $P-x-y$ diagram for the HFC-23 (1) + HFC-152a (2) system at 283.15 and 293.15 K: ○, experimental at 283.15 K; ●, experimental at 293.15 K; —, calculated with the PR EOS; - - -, calculated with the CSD EOS.

where G_{ij} is the local composition factor for the NRTL model, τ_{ij} is the NRTL model binary interaction parameter, $A_{ij} = (g_{ij} - g_{jj})$ and g_{ij} is an interaction energy parameter of the $i-j$ interaction, α_{ij} is a nonrandomness parameter, and R is the universal gas constant (8.314 J·K^{-1}·mol^{-1}). The critical properties (T_c, P_c) and acentric factors (ω) of HFC-23, HFC-152a, and HFC-143a used to calculate the parameters for the Peng−Robinson equation of state are given in Table 1. We have set the nonrandomness parameter, α_{ij}, equal to 0.3 for all the binary mixtures studied here. The Carnahan−Starling−Desantis equation of state[3] is expressed as follows:

$$\frac{PV}{RT} = \frac{1 + y + y^2 - y^3}{(1 - y)^3} - \frac{a}{RT(V + b)} \tag{11}$$

with

$$y = \frac{b}{4V} \tag{12}$$

$$a = \alpha_0 \exp(\alpha_1 T + \alpha_2 T^2) \tag{13}$$

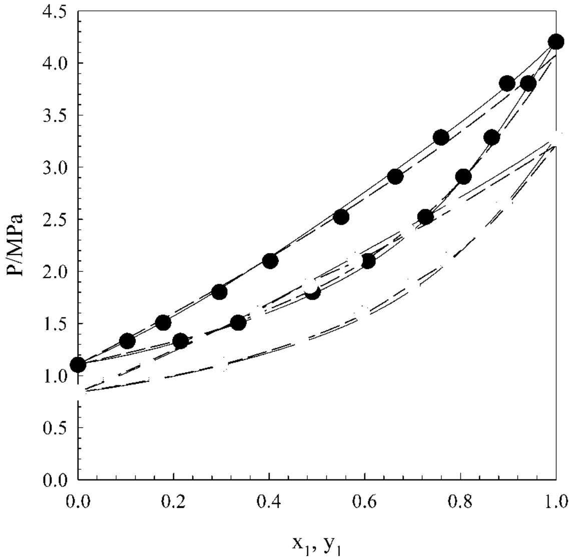

Figure 3. $P-x-y$ diagram for the HFC-23 (1) + HFC-143a (2) system at 283.15 and 293.15 K: ○, experimental at 283.15 K; ●, experimental at 293.15 K; —, calculated with the PR EOS; - - -, calculated with the CSD EOS.

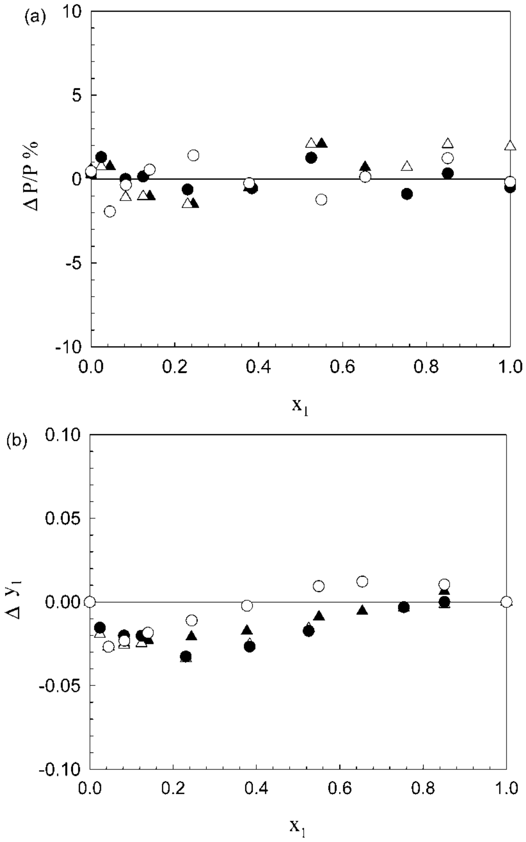

Figure 4. Deviation of the HFC-23 (1) + HFC-152a (2) binary system: ○, PR EOS at 283.15 K; ●, P−R EOS at 293.15 K; △, CSD EOS at 283.15 K; ▲, CSD EOS at 293.15 K; a, $\Delta P/P$ % versus x_1; b, Δy_1 versus x_1.

$$b = \beta_0 + \beta_1 T + \beta_2 T^2 \tag{14}$$

where a is an attraction parameter and b is a molecular volume. The following mixing rules[12] were used to obtain equation of state parameters for a mixture.

$$a_{12} = (1 - f_{12})(a_1 a_2)^{1/2} \tag{15}$$

$$b_{12} = \frac{1}{8}(b_1^{1/3} + b_2^{1/3})^3 \qquad (16)$$

where f_{12} is an interaction parameter and a_i and b_i are the properties of the pure components. The coefficients α_i and β_i for pure components were cited from REFPROP 5.0[13] and listed in Table 2. The parameters of these equations were obtained by minimizing the objective function

$$\text{objective function} = \frac{1}{N}\sum_j^n\left[\left(\frac{P_{j,\text{exp}} - P_{j,\text{cal}}}{P_{j,\text{exp}}}\right) \times 100\right]^2 \qquad (17)$$

Table 3 is a comparison of measured vapor pressures of pure HFC-23, HFC-152a, and HFC-143a with those calculated from the database REFPROP 6.01[14] which is considered to be reliable for the pure compounds considered and consistent with literature data. The average deviation ($|\Delta P|/P$) between measured and calculated values from the data of REFPROP 6.01[14] is 0.0184% for HFC-23, 0.3788% for HFC-152a, and 0.1646% for HFC-143a. The experimental vapor pressure data for the pure components and isothermal vapor liquid equilibria for the HFC-23 + HFC-152a and HFC-23 + HFC-143a mixtures are shown in Tables 4 and 5. Each table lists the measured mole fraction of the liquid and vapor phases, pressures and temperatures in equilibrium, and the deviations between measured and calculated pressures (ΔP) and vapor compositions (Δy), point by point. Table 6 lists the interaction parameters of binary mixtures for each isotherm, the binary parameters of the NRTL model with the Wong−Sandler mixing rules, and the average deviations of pressure (δP) and vapor-phase composition (δy) between measured and calculated values. Figures 2 and 3 show the comparison of measured and calculated values for HFC-23 + HFC-152a and HFC-23 + HFC-143a at 283.15 and 293.15 K. Figures 4 and 5 show the comparison of the relative pressure and vapor-phase mole fraction of HFC-23 between the measured data and the calculated values. From these figures and the low average deviations of P and y in Table 6, we concluded that the calculated values using the Peng−Robinson equation of state and the CSD equation of state give good agreement with the experimental data, but the Peng−Robinson equation of state yielded more accurate predictions for these binary systems. Azeotropic behavior has not been found in either of these mixtures.

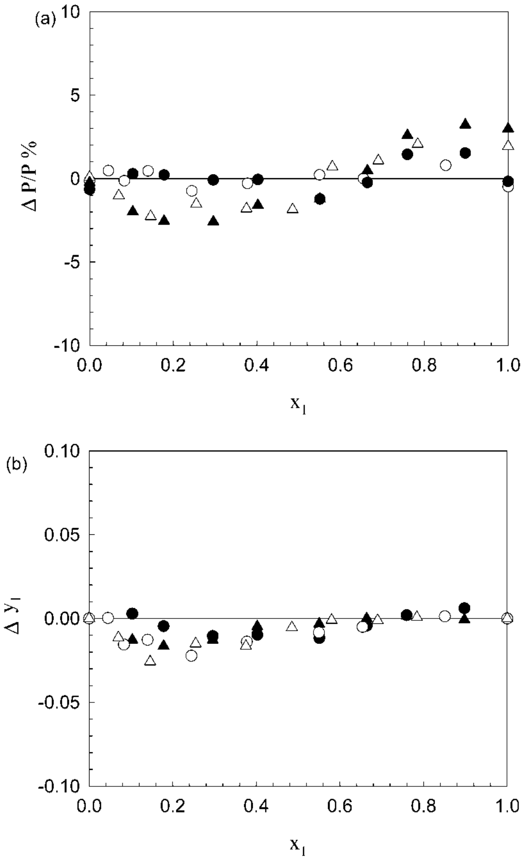

Figure 5. Deviation of the HFC-23 (1) + HFC-143a (2) binary system: ○, PR EOS at 283.15 K; ●, P−R EOS at 293.15 K; △, CSD EOS at 283.15 K; ▲, CSD EOS at 293.15 K; a, $\Delta P/P$ % versus x_1; b, Δy_1 versus x_1.

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