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Vapor−Liquid Equilibria of the Binary *n*-Butane (HC-600) + Difluoromethane (HFC-32), + Pentafluoroethane (HFC-125), + 1,1,1,2-Tetrafluoroethane (HFC-134a) Systems

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Binary vapor−liquid equilibrium data were measured for the *n*-butane (HC-600) + difluoromethane (HFC-32), + pentafluoroethane (HFC-125), 1,1,1,2-tetrafluoroethane (HFC-134a) systems at temperatures from 313.15 K to 333.15 K. These experiments were carried out with a circulating-type apparatus with on-line gas chromatography. The experimental data were correlated well with the Peng−Robinson equation of state using the Wong−Sandler mixing rules.

Introduction

As a consequence of the Montreal Protocol of 1987, the production and use of refrigerants with high ozone depletion potentials (ODPs) such as the chlorofluorocarbons (CFCs) were regulated. Hydrofluorocarbons (HFCs) are proposed as promising alternative refrigerants to substitute for CFCs because of their zero ODPs and low global warming potentials (GWPs). However, HFCs are expensive and immiscible with the oils in the refrigeration cycle. This problem is solved by mixing the HFCs and hydrocarbons (HCs) widely used as refrigerants in Europe. Adding HCs to HFCs improves their miscibility with oils. Moreover, the ODPs of the HCs are zero, their GWPs are very low, and they are cheap. Thus, mixtures of HFCs and HCs are important as alternative refrigerants.[1−5]

Thermodynamic properties of the mixtures such as vapor−liquid equilibria (VLE) are important in determining the optimal compositions of the mixtures and in evaluating the performance of the refrigeration cycles. In this work, we measured the isothermal VLE data for the three binary systems, HFC-32 + HC-600, HFC125 + HC-600, and HFC134a + HC-600, at temperatures from 313.15 K to 333.15 K at 10 K intervals. The experimental data were correlated by the Peng−Robinson equation[7] of state with the Wong−Sandler mixing rules.[8,9]

Experimental Section

Chemicals. *n*-Butane of 99.5% purity was supplied by Korean industrial gases, and HFC-32, HFC-125, and HFC-134a, each with a purity of 99.8 mass %, were obtained from Ulsan Chemical Co. All components were used without further purification in these experiments.

Apparatus. Details of this apparatus were given in our previous studies.[6] The measurement of the VLE data was conducted in a circulation-type apparatus. The equilibrium cell was made of 316 stainless steel with an inner volume of about 320 mL. It was equipped with two windows and two magnetic pumps. Because two magnetic pumps were forcing the vapor and liquid phases to circulate separately, equilibrium was quickly reached. The temperature in the cell was measured with a 100-Ω platinum resistance thermometer (Hart Scientific Co., model 5627) and a digital indicator (Hart Scientific Co., model 1502A) with an accuracy of 0.05 K. The temperature of the system was maintained within ±0.02 K by controlling the air bath. The pressure of the cell was measured with a pressure transducer (Sensotec Co., model Super TJE, (0 to 1500) psia) connected a digital indicator (Laurel, model L20010WM1). The accuracy of the digital pressure gauge is 0.05%. To trap the liquid and vapor samples, we used two sampling valves (Rheodyne Instruments, model 7413 with a 0.5-μL sampling loop and model 7010 with a 10-μL sampling loop). A gas chromatograph, which used helium gas as a carrier, was connected on-line to the equilibrium apparatus with a thermal conductivity detector (TCD) and equipped with a packed column whose packing material was Porapak-Q (1.83-m long, 3.18-mm diameter; mesh range 80/100). To calibrate the samples, we used a 500-μL Hamilton gastight syringe.

Table 1. Critical Properties and Acentric Factors of Pure Components (REFPROP[11])

	HFC-32	HFC-125	HFC-134a	*n*-butane
T_c/K	351.26	339.33	374.21	425.16
P_c/bar	57.82	36.29	40.59	37.96
ω	0.2768	0.3035	0.3268	0.1995

Experimental Procedure. The whole system was evacuated with a vacuum pump to remove all impurities. A certain amount of *n*-butane was then injected into the cell, which was then charged with the proper amount of HFCs. Two magnetic pumps were started to circulate the vapor and liquid phases. When equilibrium was reached, the vapor and liquid samples were taken into the gas chromatograph, and the compositions were measured im-

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Table 2. Vapor−Liquid Equilibrium Measurements for the HFC-32 (1) + HC-600 (2) System

P_{exptl}/bar	$x_{1,\mathrm{exptl}}$	$y_{1,\mathrm{exptl}}$	P_{calcd}/bar	$y_{1,\mathrm{calcd}}$	P/bar[a]	y_1[b]
$T = 313.15$ K						
3.78	0.000	0.000	3.78	0.000	0.00	0.000
4.57	0.010	0.137	4.49	0.149	−0.08	0.012
5.24	0.020	0.241	5.17	0.256	−0.07	0.015
6.68	0.041	0.401	6.52	0.404	−0.16	0.003
9.22	0.083	0.546	8.92	0.560	−0.30	0.014
12.56	0.169	0.664	12.84	0.693	0.28	0.029
15.34	0.239	0.730	15.23	0.743	−0.11	0.013
19.04	0.391	0.793	18.80	0.800	−0.24	0.007
21.02	0.533	0.823	20.90	0.830	−0.12	0.007
22.33	0.646	0.847	22.13	0.851	−0.20	0.004
23.38	0.763	0.873	23.22	0.876	−0.16	0.003
24.05	0.849	0.901	23.97	0.903	−0.08	0.002
24.34	0.885	0.915	24.25	0.918	−0.09	0.003
24.59	0.925	0.937	24.52	0.940	−0.07	0.003
24.69	0.922	0.938	24.51	0.938	−0.18	0.000
24.73	0.951	0.954	24.65	0.957	−0.08	0.003
24.79	0.965	0.966	24.70	0.968	−0.09	0.002
24.76	1.000	1.000	24.73	1.000	−0.03	0.000
$T = 323.15$ K						
4.96	0.000	0.000	4.95	0.000	−0.01	0.000
6.24	0.016	0.214	6.23	0.191	−0.01	−0.023
7.15	0.027	0.301	7.07	0.281	−0.08	−0.020
8.13	0.041	0.372	8.09	0.366	−0.04	−0.006
12.55	0.120	0.572	13.04	0.596	0.49	0.024
16.06	0.188	0.676	16.38	0.676	0.32	0.000
20.69	0.306	0.753	20.69	0.746	0.00	−0.007
25.37	0.502	0.812	25.28	0.804	−0.09	−0.008
28.28	0.694	0.854	28.28	0.848	0.00	−0.006
30.26	0.844	0.901	30.25	0.896	−0.01	−0.005
30.44	0.863	0.904	30.47	0.904	0.03	0.000
30.68	0.891	0.917	30.77	0.918	0.09	0.001
30.79	0.897	0.922	30.83	0.922	0.04	0.000
31.15	0.928	0.939	31.11	0.941	−0.04	0.002
31.27	0.940	0.948	31.20	0.949	−0.07	0.001
31.38	0.965	0.966	31.34	0.968	−0.04	0.002
31.37	1.000	1.000	31.41	1.000	0.04	0.000
$T = 333.15$ K						
6.38	0.000	0.000	6.38	0.000	0.00	0.000
6.99	0.007	0.093	7.01	0.082	0.02	−0.011
8.68	0.027	0.249	8.76	0.250	0.08	0.001
11.30	0.059	0.404	11.34	0.407	0.04	0.003
15.15	0.112	0.541	15.09	0.544	−0.06	0.003
18.69	0.181	0.614	19.15	0.634	0.46	0.020
22.76	0.265	0.699	23.11	0.695	0.35	−0.004
27.03	0.375	0.751	27.10	0.742	0.07	−0.009
29.92	0.470	0.782	29.82	0.771	−0.10	−0.011
33.17	0.606	0.815	33.02	0.806	−0.15	−0.009
35.84	0.732	0.851	35.62	0.843	−0.22	−0.008
36.69	0.778	0.868	36.51	0.860	−0.18	−0.008
37.62	0.831	0.888	37.49	0.883	−0.13	−0.005
38.09	0.865	0.904	38.07	0.900	−0.02	−0.004
38.93	0.927	0.939	38.93	0.938	0.00	−0.001
39.11	0.950	0.955	39.16	0.956	0.05	0.001
39.23	0.965	0.967	39.27	0.968	0.04	0.001
39.33	1.000	1.000	39.39	1.000	0.06	0.000

[a] $\Delta P = P_{\mathrm{calcd}} - P_{\mathrm{exptl}}$. [b] $\Delta y_1 = y_{\mathrm{calcd}} - y_{\mathrm{exptl}}$.

Table 3. Vapor−Liquid Equilibrium Measurements for the HFC-125 (1) + HC-600 (2) System

P_{exptl}/bar	$x_{1,\mathrm{exptl}}$	$y_{1,\mathrm{exptl}}$	P_{calcd}/bar	$y_{1,\mathrm{calcd}}$	P/bar[a]	y_1[b]
$T = 313.15$ K						
3.78	0.000	0.000	3.78	0.000	0.00	0.000
4.65	0.017	0.177	4.61	0.175	−0.04	−0.002
6.05	0.047	0.355	5.96	0.357	−0.08	0.002
7.88	0.094	0.498	7.79	0.506	−0.09	0.008
9.90	0.160	0.599	9.86	0.611	−0.04	0.012
11.97	0.253	0.672	12.03	0.687	0.06	0.015
13.96	0.361	0.729	13.83	0.736	−0.13	0.007
15.85	0.518	0.778	15.73	0.784	−0.12	0.006
17.42	0.676	0.826	17.37	0.831	−0.05	0.005
19.13	0.867	0.905	19.32	0.911	0.19	0.006
19.59	0.928	0.943	19.90	0.947	0.31	0.004
19.88	1.000	1.000	20.49	1.000	0.61	0.000
$T = 323.15$ K						
4.96	0.000	0.000	4.95	0.000	−0.01	0.000
5.86	0.016	0.138	5.86	0.147	0.00	0.009
6.98	0.035	0.263	6.87	0.267	−0.11	0.004
9.00	0.079	0.418	8.96	0.433	−0.04	0.015
11.70	0.152	0.547	11.79	0.567	0.09	0.020
14.17	0.240	0.627	14.38	0.647	0.21	0.020
17.27	0.392	0.703	17.50	0.720	0.23	0.017
20.05	0.515	0.768	19.39	0.761	−0.66	−0.007
21.63	0.669	0.812	21.53	0.813	−0.10	0.001
24.09	0.866	0.904	24.24	0.905	0.15	0.001
24.69	0.928	0.941	25.02	0.945	0.33	0.004
25.05	1.000	1.000	25.80	1.000	0.75	0.000
$T = 333.15$ K						
6.38	0.000	0.000	6.38	0.000	0.00	0.000
7.66	0.021	0.148	7.70	0.161	0.04	0.013
9.28	0.049	0.283	9.34	0.300	0.06	0.017
11.52	0.091	0.409	11.53	0.426	0.01	0.017
14.11	0.154	0.503	14.30	0.532	0.19	0.029
16.44	0.222	0.578	16.73	0.599	0.29	0.021
19.30	0.314	0.645	19.34	0.656	0.04	0.011
21.93	0.426	0.696	21.84	0.703	−0.09	0.007
25.13	0.591	0.757	24.93	0.761	−0.20	0.004
28.60	0.786	0.844	28.48	0.846	−0.12	0.002
29.43	0.837	0.874	29.42	0.876	−0.01	0.002
30.09	0.870	0.904	30.01	0.897	−0.08	−0.007
30.82	0.924	0.939	30.92	0.936	0.10	−0.003
31.57	1.000	1.000	31.96	1.000	0.39	0.000

[a] $\Delta P = P_{\mathrm{calcd}} - P_{\mathrm{exptl}}$. [b] $\Delta y_1 = y_{\mathrm{calcd}} - y_{\mathrm{exptl}}$.

mediately. The equilibrium compositions of each phase were measured at least three times in succession to obtain reliable average values. Deviations of the equilibrium composition were <±0.001 in mole fraction for both the vapor and liquid phases.

Results and Discussion

Vapor−liquid equilibrium data for binary systems HFC-32 (1) + HC-600 (2), HFC-125 (1) + HC-600 (2), and HFC-134a(1) + HC-600 (2) were measured in the temperature range from 313.15 K to 333.15 K. Experimental data for each system are given in Tables 2 to 4. Experimental data were correlated with the Peng−Robinson[7] equation of state (PR EOS) using the Wong−Sandler mixing rules.[8,9] The PR EOS and the Wong−Sandler mixing rules are expressed as follows

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

$$a(T) = 0.45724\frac{R^2{T_c}^2}{P_c}\alpha(T) \tag{2}$$

$$b(T_c) = 0.07780\frac{RT_c}{P_c} \tag{3}$$

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

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

where T_c is the critical temperature, P_c is the critical pressure, T_r is the reduced temperature, and ω is the acentric factor. The critical properties and acentric factors are listed in Table 1.

Table 4. Vapor−Liquid Equilibrium Measurements for the HFC-134a + HC-600 System

P_{exptl}/bar	$x_{1,\mathrm{exptl}}$	$y_{1,\mathrm{exptl}}$	P_{calcd}/bar	$y_{1,\mathrm{calcd}}$	P^a/bar	$y_1{}^b$
			$T = 313.15$ K			
3.78	0.000	0.000	3.78	0.000	0.00	0.000
4.59	0.026	0.167	4.59	0.177	0.00	0.010
5.27	0.050	0.268	5.24	0.283	−0.03	0.015
5.66	0.070	0.315	5.72	0.347	0.06	0.032
6.73	0.115	0.422	6.65	0.446	−0.08	0.024
7.46	0.160	0.496	7.38	0.509	−0.08	0.013
9.07	0.315	0.613	8.98	0.622	−0.09	0.009
10.43	0.619	0.728	10.36	0.734	−0.07	0.006
10.57	0.701	0.754	10.56	0.765	−0.01	0.011
10.71	0.764	0.790	10.66	0.793	−0.05	0.003
10.72	0.833	0.827	10.71	0.832	−0.01	0.005
10.68	0.885	0.865	10.66	0.869	−0.02	0.004
10.46	0.956	0.936	10.44	0.938	−0.02	0.002
10.36	0.971	0.956	10.36	0.957	0.00	0.001
10.13	1.000	1.000	10.15	1.000	0.02	0.000
			$T = 323.15$ K			
4.96	0.000	0.000	4.95	0.000	−0.01	0.000
5.60	0.017	0.104	5.59	0.113	−0.01	0.009
6.17	0.034	0.177	6.18	0.198	0.01	0.021
7.32	0.071	0.299	7.30	0.325	−0.02	0.026
8.48	0.117	0.393	8.45	0.423	−0.03	0.030
9.66	0.179	0.467	9.65	0.504	−0.01	0.037
10.91	0.267	0.544	10.87	0.575	−0.04	0.031
12.64	0.479	0.664	12.61	0.673	−0.03	0.009
12.83	0.514	0.677	12.80	0.686	−0.03	0.009
13.41	0.651	0.733	13.42	0.740	0.01	0.007
13.75	0.792	0.795	13.78	0.809	0.03	0.014
13.76	0.849	0.838	13.81	0.845	0.05	0.007
13.74	0.884	0.870	13.77	0.872	0.03	0.002
13.63	0.918	0.899	13.69	0.901	0.06	0.002
13.27	0.976	0.965	13.38	0.966	0.11	0.001
13.63	0.918	0.899	13.69	0.901	0.06	0.002
13.08	1.000	1.000	13.18	1.000	0.10	0.000
			$T = 333.15$ K			
7.14	0.018	0.104	7.16	0.107	0.02	0.003
7.98	0.040	0.193	8.04	0.204	0.06	0.011
9.25	0.075	0.302	9.27	0.311	0.02	0.009
10.72	0.128	0.402	10.81	0.414	0.09	0.012
13.32	0.259	0.540	13.41	0.546	0.09	0.006
14.96	0.384	0.615	14.96	0.615	0.00	0.000
16.21	0.532	0.680	16.24	0.680	0.03	0.000
16.89	0.647	0.733	16.96	0.732	0.07	−0.001
17.35	0.779	0.800	17.48	0.801	0.13	0.001
17.40	0.894	0.882	17.50	0.883	0.10	0.001
17.24	0.938	0.922	17.33	0.925	0.09	0.003
17.11	0.958	0.946	17.21	0.947	0.10	0.001
17.40	0.791	0.808	17.50	0.809	0.10	0.001
17.45	0.849	0.846	17.55	0.848	0.10	0.002
16.71	1.000	1.000	16.84	1.000	0.13	0.000

[a] $\Delta P = P_{\mathrm{calcd}} - P_{\mathrm{exptl}}$. [b] $\Delta y_1 = y_{\mathrm{calcd}} - y_{\mathrm{exptl}}$.

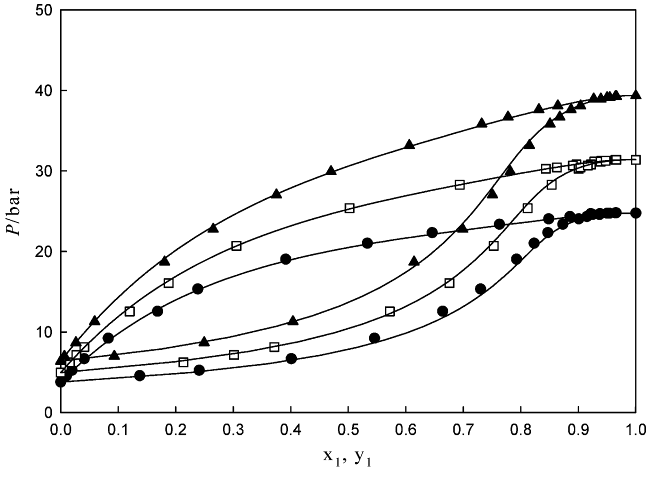

Figure 1. Vapor−liquid equilibria of the HFC-32 (1) + HC-600 (2) system: −, PR EOS using the Wong−Sandler mixing rules; ▲, experimental data at 333.15 K; □, experimental data at 323.15 K; ●, experimental data at 313.15 K.

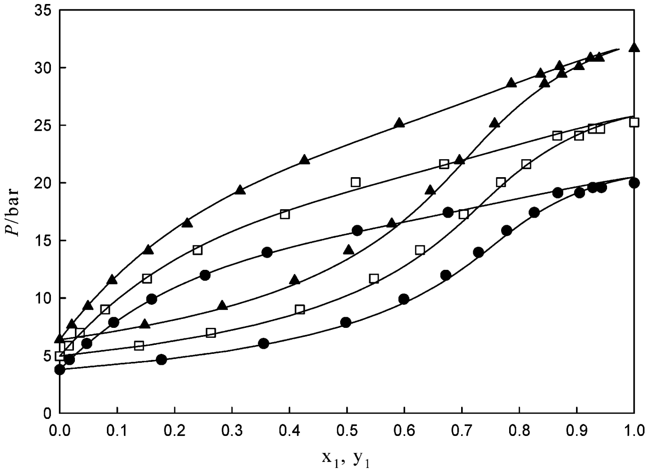

Figure 2. Vapor−liquid equilibria of the HFC-125 (1) + HC-600 (2) system: −, PR EOS using the Wong−Sandler mixing rules; ▲, experimental data at 333.15 K; □, experimental data at 323.15 K; ●, experimental data at 313.15 K.

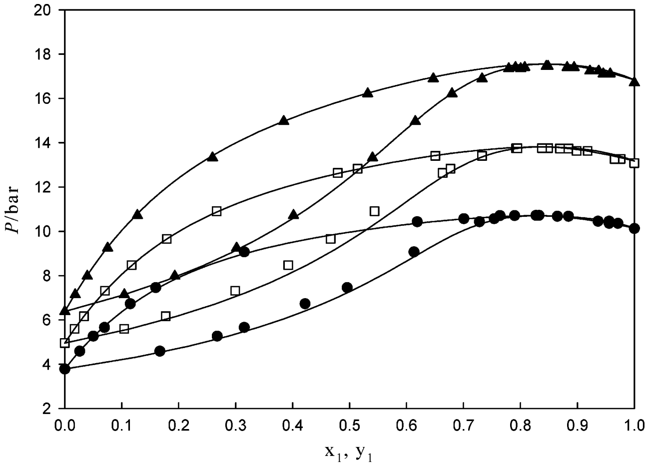

Figure 3. Vapor−liquid equilibria of the HFC-134a (1) + HC-600 (2) system: −, PR EOS using Wong−Sandler mixing rules; ▲, experimental data at 333.15 K; □, experimental data at 323.15 K; ●, experimental data at 313.15 K.

The Wong−Sandler mixing rule is represented as

$$b_{\mathrm{m}} = \frac{\sum_i \sum_j x_i x_j \left(b - \frac{a}{RT}\right)_{ij}}{1 - \sum_i x_i \frac{a_i}{b_i RT} - \frac{A^{\mathrm{E}}_{\infty}}{CRT}} \qquad (6)$$

$$a_{\mathrm{m}} = b_{\mathrm{m}} \left(\sum_i x_i \frac{a_i}{b_i} + \frac{A^{\mathrm{E}}_{\infty}}{C} \right) \qquad (7)$$

$$\left(b - \frac{a}{RT}\right)_{ij} = \frac{\left(b_i - \frac{a_i}{RT}\right) + \left(b_j - \frac{a_j}{RT}\right)}{2}(1 - k_{ij}) \qquad (8)$$

where $C = \ln(\sqrt{2} - 1)/\sqrt{2}$ for the PR EOS. Because the excess Helmholtz free energy of mixing at infinite pressure is assumed to be equal to the excess Gibbs free energy (G^{E})

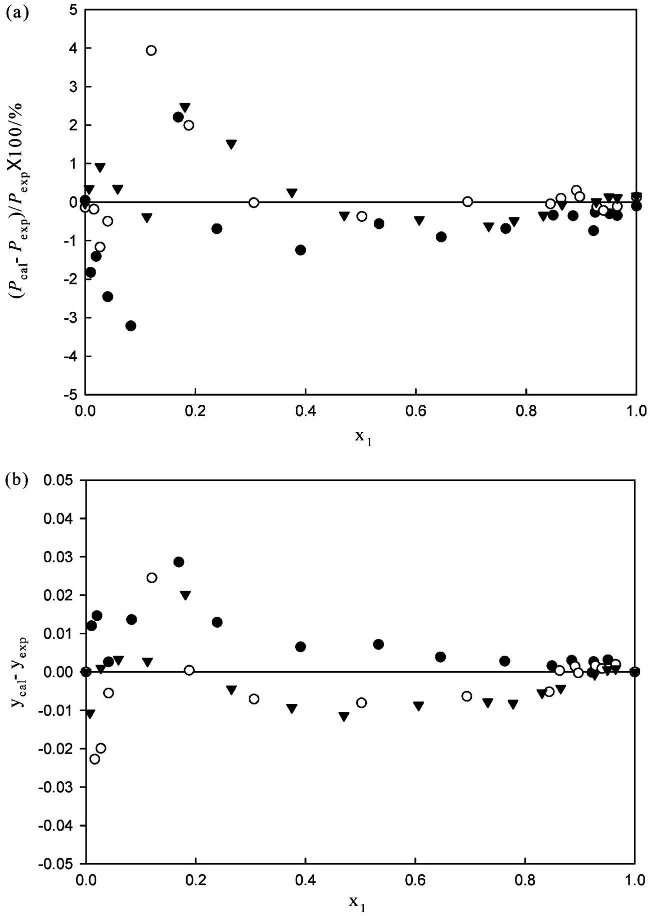

Figure 4. Deviation of the HFC-32 (1) + HC-600 (2) system: (a) deviation of pressure and (b) deviation of vapor pressure; ▼, experimental data at 333.15 K; ○, experimental data at 323.15 K; ●, experimental data at 313.15 K.

Table 5. Binary Parameters and AAD (%)

T/K	binary parameters[a]	AAD-P[b]/%	AAD-y[c]
	HFC-32 + HC-600		
313.15	$k_{ij} = 0.316$ $A_{ij} = 548.04$ $A_{ji} = 266.41$	0.667	0.0067
323.15	$k_{ij} = 0.456$ $A_{ij} = 127.27$ $A_{ji} = 364.57$	0.658	0.0083
333.15	$k_{ij} = 0.399$ $A_{ij} = 308.16$ $A_{ji} = 277.43$	0.471	0.0037
	HFC-125 + HC-600		
313.15	$k_{ij} = 0.229$ $A_{ij} = 194.57$ $A_{ji} = 285.66$	0.788	0.0091
323.15	$k_{ij} = 0.368$ $A_{ij} = -186.20$ $A_{ji} = 574.95$	0.887	0.0197
333.15	$k_{ij} = 0.211$ $A_{ij} = 290.48$ $A_{ji} = 204.04$	0.439	0.0104
	HFC-134a + HC-600		
313.15	$k_{ij} = 0.205$ $A_{ij} = 379.81$ $A_{ji} = 280.76$	0.293	0.0114
323.15	$k_{ij} = 0.188$ $A_{ij} = 371.39$ $A_{ji} = 298.83$	0.238	0.0147
333.15	$k_{ij} = 0.205$ $A_{ij} = 337.19$ $A_{ji} = 284.90$	0.234	0.0046

[a] The unit of A_{ij} and A_{ji} is K. [b] $\text{AAD} - P = (100/N)\sum_i^N(|P_{i,\text{calcd}} - P_{i,\text{exptl}}|/P_{i,\text{exptl}})$. [c] $\text{AAD} - y = (1/N)\sum_i^N|y_{i,\text{calcd}} - y_{i,\text{exptl}}|$.

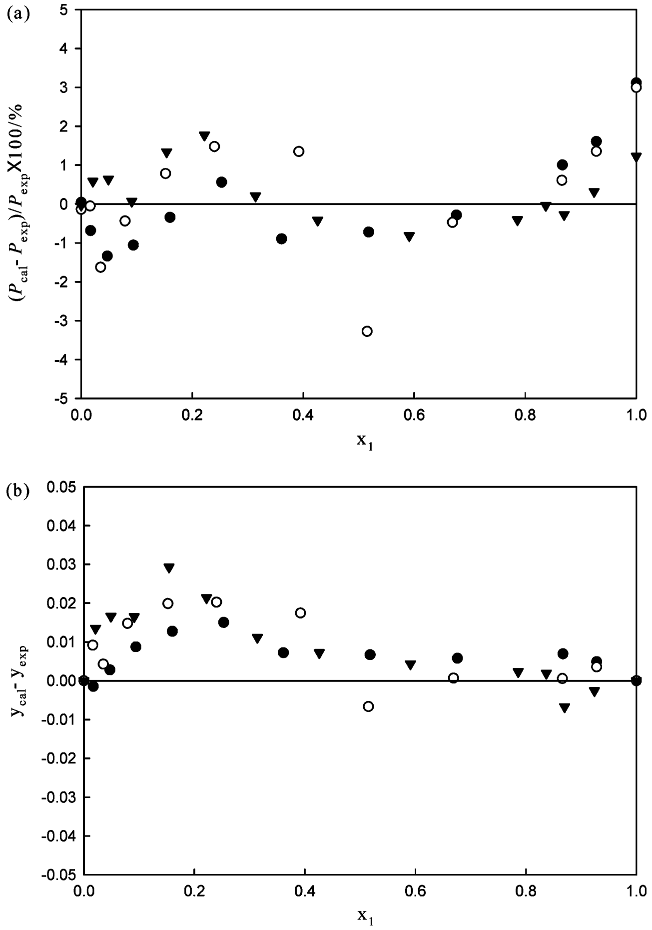

Figure 5. Deviation of the HFC-125 (1) + HC-600 (2) system: (a) deviation of pressure and (b) deviation of vapor pressure; ▼, experimental data at 333.15 K; ○, experimental data at 323.15 K; ●, experimental data at 313.15 K.

at low pressure, the G^E model is used in place of A^E_∞. We used the NRTL model[10] as a G^E model in this study.

$$\frac{G^E}{RT} = \sum_i x_i \frac{\sum_i x_i \tau_{ji} G_{ji}}{\sum_k x_k G_{ki}} \tag{9}$$

$$G_{ij} = \exp(-\alpha_{ij}\tau_{ij}) \qquad \alpha_{ij} = \alpha_{ji} \tag{10}$$

$$\tau_{ij} = \frac{A_{ij}}{T} \tag{11}$$

where τ_{ij} and τ_{ji} are the interaction parameters and α_{ij} is the nonrandomness parameter. We fixed the nonrandomness parameter α_{ij} to 0.3 in this study. The Marquardt algorithm was used to minimize the following objective function:

$$\text{OF} = \sum_i^N \left|\frac{P_{\text{exptl}} - P_{\text{calcd}}}{P_{\text{exptl}}}\right| \tag{12}$$

where N is the number of experimental data points and P_{exptl} and P_{calcd} are the experimental and the calculated pressures, respectively.

As shown in Figures 1, 2, and 3, the calculated results are in good agreement with the experimental data at each temperature. The overall deviations of pressure and vapor

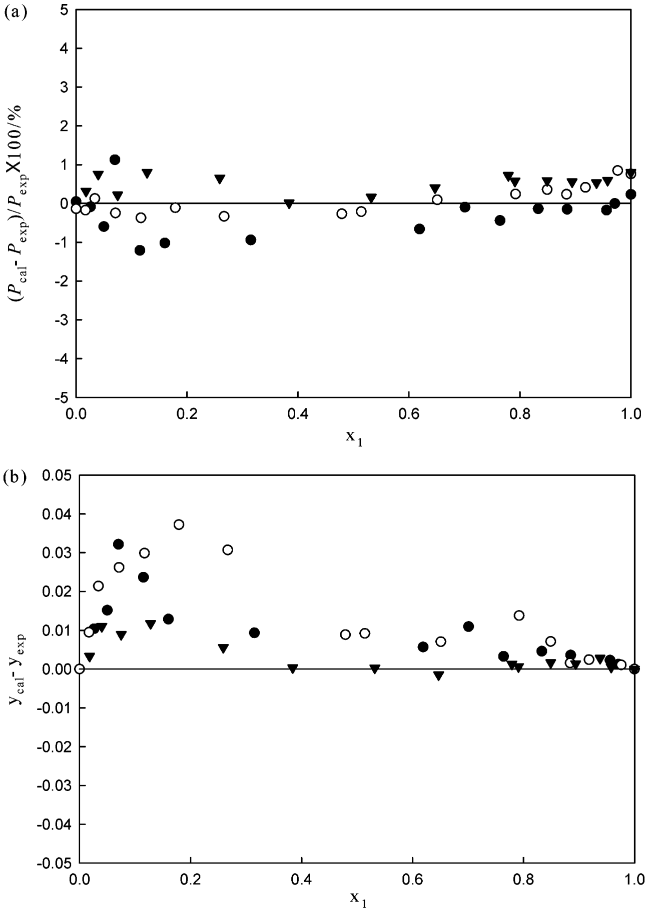

Figure 6. Deviation of the HFC-134a (1) + HC-600 (2) system: (a) deviation of pressure and (b) deviation of vapor pressure; ▼, experimental data at 333.15 K; ○, experimental data at 323.15 K; ●, experimental data at 313.15 K.

composition were 0.598% and 0.0062 for the HFC-32 + HC-600 system, 0.704% and 0.0130 for the HFC-125 + HC-600 system, and 0.255% and 0.0102 for the HFC-134a + HC-600 system, respectively. Figures 4, 5, and 6 show the deviations of *P* and *y* of each system between the measured data and the calculated values. Binary parameters and the average absolute deviations of *P* and *y* are given in Table 5. In the HFC-134a + HC-600 system, azeotropic behaviors were found, and the calculated azeotropic compositions and pressures are listed in Table 6.

Table 6. Calculated Azeotropic Compositions and Pressures for the HFC-134a + HC-600 System

T/K	x_1	*P*/bar
313.15	0.830	10.71
323.15	0.839	13.81
333.15	0.847	17.55

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