J. Chem. Eng. Data 2008, 53, 189–193

Vapor–Liquid Equilibria for 1,1,1,2,3,3,3-Heptafluoropropane (HFC-227ea) + Butane (R600) at Various Temperatures

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Isothermal vapor–liquid equilibrium data for the binary mixture of 1,1,1,2,3,3,3-heptafluoropropane (HFC-227ea) + butane (R600) were measured at (273.15, 283.15, 293.15, 303.15, 313.15, and 323.15) K, respectively. The experiments were carried out using a continuous circulation-type equilibrium apparatus to measure temperature, pressure, and the compositions of the liquid and vapor phases. The experimental data were correlated with the Peng–Robinson equation of state (PR EOS) using the Wong–Sandler mixing rule. Calculated results showed good agreement with experimental data. It was found that this system has very strong positive azeotropes for all the temperature ranges studied here.

Introduction

After having announced the restriction of using CFCs, many countries which are mostly developed countries started to find replacements for CFCs. The movement of studies for new refrigerants was quick in action, so researchers have reported many candidate refrigerants. Although a lot of studies for the replacement were announced before, the development of new refrigerants is continuously performed by many scientists and engineers all over the world because the most optimal replacement material is still needed.

Vapor–liquid equilibrium (VLE) data are the most important data requested to evaluate the performance of refrigeration cycles and to determine their optimal compositions. Azeotropic mixtures have merit because their behaviors are similar to pure compounds. However, very few experimental data for the binary system of HFC-227ea + butane have been reported previously in the literature.

In this work, isothermal VLE data for the binary mixture of HFC-227ea + butane at six equally spaced temperatures between (273.15 and 323.15) K were measured using a circulation-type equilibrium apparatus in which both phases were continuously recirculated. The experimental data were correlated with the Peng–Robinson equation of state (PR EOS)[1] using the Wong–Sandler mixing rule[2] combined with the NRTL excess Gibbs free energy model. In the range of experimental temperatures, the average absolute deviations of pressure and vapor phase compositions between experimental and calculated values were determined and the relevant parameters were presented. All the calculated values with this model give good agreement with the experimental data, and the system exhibits an azeotrope.

Experimental

Chemicals. High-quality chemicals of HFC-227ea and butane were used for VLE measurement. HFC-227ea was supplied by DuPont (U.S.A.) with a guaranteed purity in mass fraction higher than 0.998, and R600 (butane) was supplied by MG Industry (U.K.) with a guaranteed purity in mass fraction higher than 0.995. Furthermore, we also analyzed these two pure components with a gas chromatograph. The resulted mass fraction purities of HFC-227ea and R600 were higher than 0.999 and 0.9985, respectively. So, they were used without any further purification.

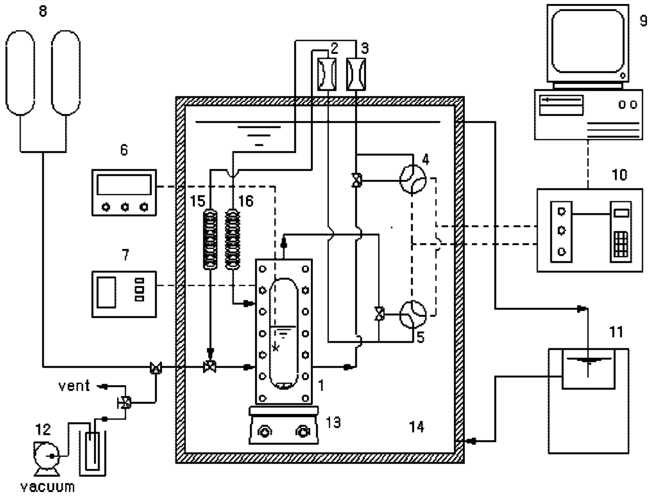

Figure 1. Schematic diagram of the experimental apparatus: 1, equilibrium cell; 2, vapor circulation pump; 3, liquid circulation pump; 4, liquid sample valve; 5, vapor sample valve; 6, temperature indicator; 7, pressure indicator; 8, sample reservoir; 9, computer; 10, gas chromatograph; 11, circulator;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

Experimental Apparatus. A schematic diagram of the experimental apparatus for measuring the VLE is given in Figure 1. Because the vapor–liquid equilibrium apparatus used in this work was the same as that used in our previous work,[3–7] only a brief description is given here. It was a circulation-type in

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10.1021/je7004837 CCC: $40.75

Table 1. Characteristic Properties of the Chemicals (Critical Temperature T_c, Critical Pressure P_c, Acentric Factor *w*) from database REFPROP 6.01[10]

component	chemical formula	molecular weight	T_c / K	P_c / MPa	ω
HFC-227ea (1)	CF_3CHCF_3	170.03	375.95	2.980	0.3632
butane (2)	C_4H_{10}	58.12	425.16	3.796	0.1995

Table 2. Comparison of the Measured Pure Component Vapor Pressures *P*, with Reference Vapor Pressures P_{REF} from the Database REFPROP 6.01[10]

T / K	P / MPa	P_{REF} / MPa	ΔP^a / MPa	$^b\lvert\Delta P/P\rvert$
butane				
273.15	0.104	0.103	0.001	0.010
283.15	0.150	0.149	0.001	0.007
293.15	0.208	0.208	0.000	0.000
303.15	0.284	0.284	0.000	0.000
313.15	0.378	0.379	−0.001	0.003
323.15	0.494	0.497	−0.003	0.006
average value				**0.004**
HFC-227ea				
273.15	0.194	0.193	0.001	0.005
283.15	0.280	0.278	0.002	0.007
293.15	0.389	0.389	0.000	0.000
303.15	0.527	0.530	−0.003	0.006
313.15	0.704	0.706	−0.002	0.003
323.15	0.920	0.922	−0.002	0.002
average value				**0.004**

[a] $\Delta P = P - P_{REF}$. [b] $\lvert\Delta P/P\rvert = \lvert P - P_{calcd}\rvert/P$.

Table 3. Vapor–Liquid Equilibrium Measurements for the HFC-227ea (1) + Butane (2) System at (273.15 to 323.15) K

P / MPa	x_1	y_1	P / MPa	x_1	y_1
Experimental Data					
T = 273.15 K			T = 283.15 K		
0.104	0.000	0.000	0.150	0.000	0.000
0.174	0.095	0.358	0.221	0.067	0.275
0.188	0.137	0.421	0.257	0.128	0.393
0.210	0.261	0.517	0.285	0.216	0.481
0.225	0.468	0.593	0.293	0.258	0.519
0.227	0.561	0.628	0.306	0.360	0.557
0.230	0.606	0.659	0.315	0.457	0.592
0.230	0.692	0.690	0.322	0.546	0.627
0.230	0.710	0.690	0.324	0.608	0.651
0.229	0.732	0.702	0.326	0.693	0.694
0.229	0.779	0.726	0.324	0.734	0.715
0.227	0.845	0.776	0.321	0.811	0.762
0.219	0.913	0.848	0.313	0.892	0.834
0.194	1.000	1.000	0.280	1.000	1.000
T = 293.15 K			T = 303.15 K		
0.208	0.000	0.000	0.284	0.000	0.000
0.277	0.049	0.226	0.387	0.061	0.253
0.327	0.103	0.360	0.452	0.129	0.374
0.367	0.181	0.448	0.511	0.228	0.474
0.393	0.255	0.505	0.552	0.355	0.548
0.420	0.412	0.588	0.567	0.429	0.577
0.435	0.538	0.631	0.582	0.537	0.627
0.439	0.632	0.670	0.589	0.626	0.667
0.443	0.690	0.697	0.591	0.695	0.702
0.443	0.747	0.728	0.591	0.726	0.719
0.435	0.833	0.785	0.586	0.813	0.777
0.424	0.901	0.849	0.575	0.880	0.835
0.389	1.000	1.000	0.527	1.000	1.000
T = 313.15 K			T = 323.15 K		
0.378	0.000	0.000	0.494	0.000	0.000
0.480	0.046	0.197	0.618	0.046	0.184
0.560	0.101	0.312	0.732	0.112	0.321
0.625	0.169	0.415	0.805	0.178	0.409
0.670	0.242	0.483	0.868	0.257	0.477
0.718	0.357	0.544	0.919	0.354	0.536
0.739	0.439	0.589	0.954	0.451	0.586
0.759	0.547	0.640	0.979	0.550	0.636
0.768	0.634	0.675	0.991	0.627	0.674
0.772	0.691	0.706	0.994	0.694	0.713
0.772	0.742	0.737	0.994	0.751	0.747
0.765	0.822	0.794	0.987	0.817	0.796
0.747	0.905	0.871	0.961	0.912	0.882
0.704	1.000	1.000	0.920	1.000	1.000

which both liquid and vapor phases were recirculated continuously. The equilibrium cell was a type-316 stainless steel vessel within an inner volume of about 85 cm.[3] In its middle part, two Pyrex glass windows of 20 mm thickness were installed in front and behind so that the liquid level, mixing and circulating behaviors, and critical phenomena could be observed by a back light during operation. A stirrer, rotated at variable speeds by an external magnet, was used to accelerate the attainment of the equilibrium state and to reduce concentration gradients in both phases.

The temperature of the equilibrium cell in the water bath was maintained by a circulator from Jeio Tech, Korea. The temperature in the cell was measured with a platinum-resistance sensor and a digital temperature indicator (model F250MkII) precision thermometer from Automatic Systems Laboratories, Ltd., United Kingdom. They were calibrated by the National Measurement Accreditation Service accredited calibration laboratory. The total uncertainty in temperature measurements is estimated to be within 0.01 K, including sensor uncertainty, 0.001 K, temperature resolution, 0.001 K, and measurement uncertainty, 0.001 K. The pressure was measured with a pressure transducer (model XPM60) and a digital pressure calibrator (model PC106), from Beamax, Finland. Pressure calibrations are traceable to National Standards (Center for Metrology and Accreditation Cert. No. M-95P077, 14.11.1995, M-M 730, 16.11.1995, and M-95P078, 16.11.1995), and the calibrator uncertainty was 0.0005 MPa. The sensor uncertainty was 0.0001 MPa, and the measurement uncertainty was 0.0001 MPa. Therefore, the total uncertainty of the pressure measurement is estimated to be within 0.001 MPa.

The vapor and liquid phases in the equilibrium cell were continuously recirculated by a dual-head circulation pump (Milton Roy Company) to reach the equilibrium state rapidly in the cell. The composition of the phases was determined by means of a gas chromatograph (Gow-Mac model 550P) connected online to the VLE cell. The response of the thermal conductivity detector (TCD) was carefully calibrated using the mixture prepared gravimetrically and the gas chromatographs (GC) with a Porapak Q column from Alltech Company. Data derived from gas chromatography were treated with a computer program (Autochro-WIN from Young-Lin Instrument Co., Ltd.). The uncertainty for the GC measurement and calibration is estimated to be within 0.001 in area fraction.

Experimental Procedures. Experiments to measure VLE data for the binary system of HFC-227ea (1) + butane (2) at various temperatures were performed by the following procedures. The system was first evacuated to remove all inert gases. A certain amount of butane was supplied to 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. A certain amount of HFC-227ea was introduced into the cell from a sample reservoir. The mixture in the cell was stirred continuously with the magnetic stirrer for over 1 h. Both the

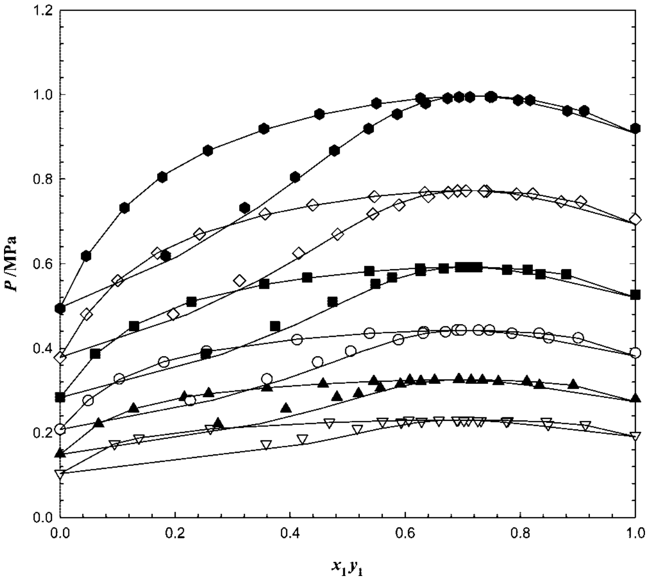

Figure 2. *P*–*x*–*y* diagram for HFC-227ea (1) + butane (2). Experimental data at: ∇, 273.15 K; ▲, 283.15 K; ○, 293.15 K; ■, 303.15 K; ◇, 313.15 K; and ⬢, 323.15 K calculated with the PR EOS using W–S mixing, —.

Table 4. Optimal Values of the Binary Parameter k_{12}, Adjustable Parameters for the NRTL Model τ_{21} and τ_{12}, and Absolute Average Relative Deviations of *P* and Absolute Average Deviation of *y*

T	PR EOS				
K	$^a k_{12}$	$^b \tau_{21}$	$^b \tau_{12}$	^{c}AARD-*P*	^{d}AAD-*y*
273.15	−0.1746	2.1719	2.2959	0.003	0.020
283.15	−0.1115	1.8911	2.0366	0.002	0.022
293.15	−0.0758	1.7036	1.7476	0.002	0.011
303.15	0.0016	1.5014	1.3967	0.002	0.010
313.15	0.0201	1.5439	1.1194	0.002	0.009
323.15	0.0870	1.4653	0.7790	0.002	0.008
	average values			**0.002**	**0.013**

[a] k_{12} is an interaction parameter. [b] τ_{ij} are adjustable parameters for the NRTL model (dimensionless). [c] AARD-$P = 1/N \sum |(P_{\text{exptl}} - P_{\text{calcd}})/P_{\text{exptl}}|$. [d] AAD-$y = 1/N \sum |y_{\text{exptl}} - y_{\text{calcd}}|$.

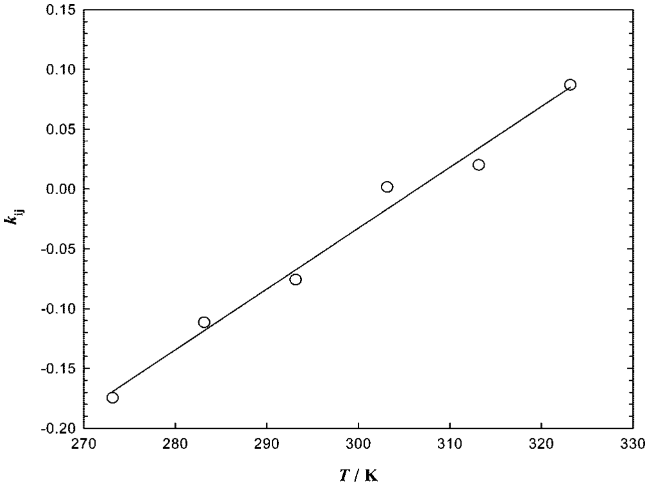

Figure 3. Parameter k_{ij} obtained by using the PR EOS with the W–S mixing rule. The equation of the fitting line is $k_{ij} = 0.0051T/\text{K} - 1.5583$(323.15 K ≥ *T* ≥ 273.15 K).

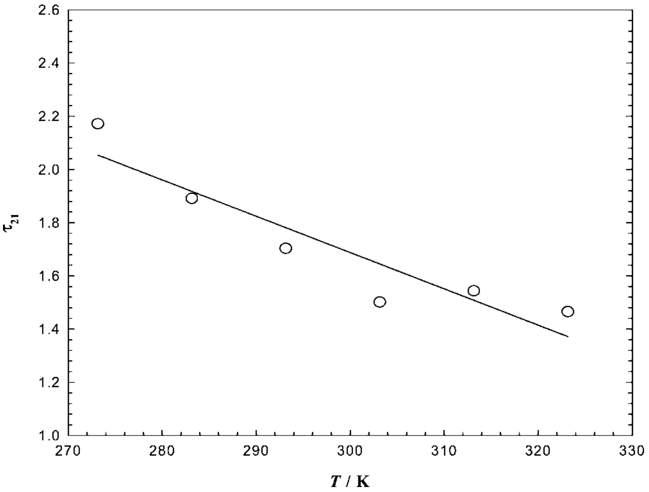

Figure 4. NRTL parameter τ_{21} obtained by using the PR EOS with the W–S mixing rule. The equation of the fitting line is $\tau_{21} = -0.0136T/\text{K} + 5.7820$ (323.15 K ≥ *T* ≥ 273.15 K).

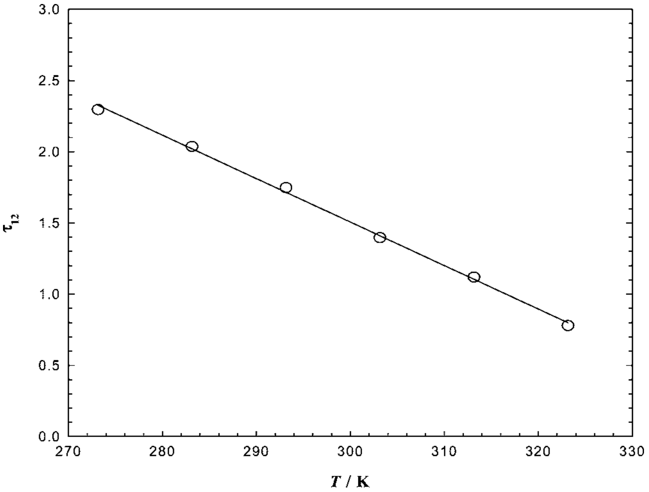

Figure 5. NRTL parameter τ_{12} obtained by using the PR EOS with the W–S mixing rule. The equation of the fitting line is $\tau_{12} = -0.0468T/\text{K} + 14.965$ (323.15 K ≥ *T* ≥ 273.15 K).

vapor and liquid phases were recirculated by the dual-head circulation pump until an equilibrium state was established. According to our experimental experiences, 1 h or more was sufficient to obtain thermal equilibrium between the cell fluid and the thermostatic bath as well as the vapor and liquid phases. After equilibration, the pressure in the equilibrium cell was measured, and then vapor and liquid samples were withdrawn from the recycling lines by the vapor and liquid sampling valves, respectively. The compositions of the samples were measured by immediately injecting them into the GC, which was connected online to the vapor and liquid sampling valves. The GC was calibrated with pure components of known purity and with mixtures of known composition that were prepared gravimetrically. At least five analyses were performed for each phase, and the average values were considered to correspond to the equilibrium values. Considering the margin of error and the reproducibility of the GC, we generally estimated an overall uncertainty in the measurements of the composition of 0.002 in mole fraction for both the liquid and the vapor phases.

Correlation. In this work, the experimental VLE data were correlated with the Peng–Robinson equation of state (PR EOS).[1]

PR EOS.

$$P = \frac{RT}{v_{\text{M}} - b} - \frac{a(T)}{v_{\text{M}}(v_{\text{M}} + b) + b(v_{\text{M}} - b)} \qquad (1)$$

with

$$a(T) = \left(0.457235\frac{R^2T_c^2}{P_c}\right)\alpha(T) \tag{2}$$

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

$$\alpha(T) = \left[1 + k\left(1 - \sqrt{T/T_c}\right)\right]^2 \tag{4}$$

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

where the parameter a is a function of temperature; b is constant; k is a constant characteristic of each substance; ω is the acentric factor; P (MPa) is the pressure; P_c (MPa) is the critical pressure; T (K) is the absolute temperature; T_c (K) is the critical temperature; T_r is the reduced temperature; and v_M is the molar volume.

The Wong–Sandler mixing rules[2] were used in this work to obtain EOS parameters for a mixture from those of the pure components. These mixing rules for a cubic equation of state can be written as

$$b_m = \frac{\sum_i\sum_j x_ix_j(b - a/RT)_{ij}}{\left(1 - A_\infty^E/CRT - \sum_i x_ia_i/RTb_i\right)} \tag{6}$$

with

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

and

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

where C is a numerical constant equal to $\ln(\sqrt{2}-1)/\sqrt{2}$ for the PR EOS used in this work. The single adjustable parameter (k_{ij}) for each binary pair is referred to as the Wong–Sandler mixing rule parameter. Also, A_∞^E is an excess Helmholtz free energy model at infinite pressure that can be equated to a low-pressure excess Gibbs energy model.[8] In this study, we used the nonrandom two-liquid (NRTL) model[9] given by

$$\frac{A_\infty^E}{RT} = \sum_i x_i\frac{\sum_j x_jG_{ji}\tau_{ji}}{\sum_r x_rG_{ri}} \tag{9}$$

with

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

where

$$A_{ij} = (g_{ij} - g_{ij}) \tag{11}$$

The critical temperature (T_c), critical pressure (P_c), and acentric factor (ω) for both HFC-227ea and butane that were used to calculate the parameters for the PR EOS are provided in Table 1. We have set the nonrandomness parameter, α_{ij}, equal to a fixed value of 0.3 for all of the binary mixtures studied here. The parameter of the PR EOS was obtained by minimizing the following objective function

$$\text{objective function} = \frac{1}{N}\sum_j^N\left[\left(\frac{P_{j,\text{exptl}} - P_{j,\text{calcd}}}{P_{j,\text{exptl}}}\right)\right]^2 \tag{12}$$

Results and Discussion

Table 2 shows the comparison of measured vapor pressures of pure HFC-227ea + butane with those calculated from the database REFPROP 6.01,[10] which is considered to be reliable for the pure compounds considered and consistent with other literature data. As can be seen in this table, the deviations of vapor pressure (ΔP) between measured and calculated values from REFPROP 6.01[10] are within 0.003 MPa. The average values of absolute relative deviations of $P(|\Delta P/P|)$ between measured and calculated values were 0.004 for HFC-227ea and 0.004 for butane. The experimental data of isothermal VLE for the binary system of HFC-227ea (1) + butane (2) are listed in Table 3. These tables list 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.

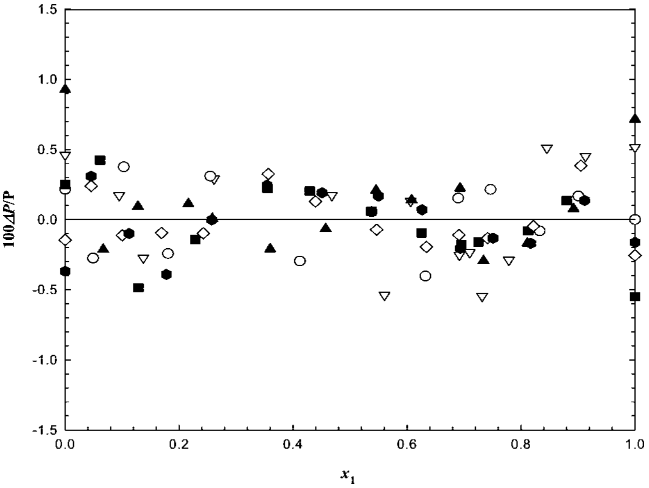

Figure 6. Deviation of pressure for the system HFC-227ea (1) + butane (2) from the PR EOS using the W–S mixing rule at: ∇, 273.15 K; ▲, 283.15 K; ○, 293.15 K; ■, 303.15 K; ◇, 313.15 K; and ⬢, 323.15 K.

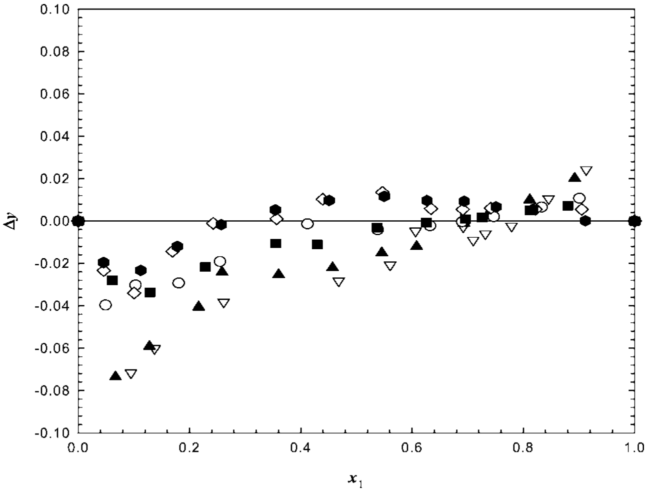

Figure 7. Deviation of vapor composition for the system HFC-227ea (1) + butane (2) from the PR EOS using the W–S mixing rule at: ∇, 273.15 K; ▲, 283.15 K; ○, 293.15 K; ■, 303.15 K; ◇, 313.15 K; and ⬢, 323.15 K.

Figure 2 shows the comparison of measured and calculated values with the PR EOS for the binary system of HFC-227ea (1) + butane (2) at various temperatures. The experimental VLE data at (273.15, 283.15, 293.15, 303.15, 313.15, and 323.15) K are shown as reverse triangles, triangles, circles, squares, diamonds, and hexagons, respectively. The black solid lines represent the calculated data by the PR EOS.

The interaction parameters of the binary mixtures for each isotherm, the binary parameters of the NRTL model with the Wong–Sandler mixing rules, and the average absolute relative

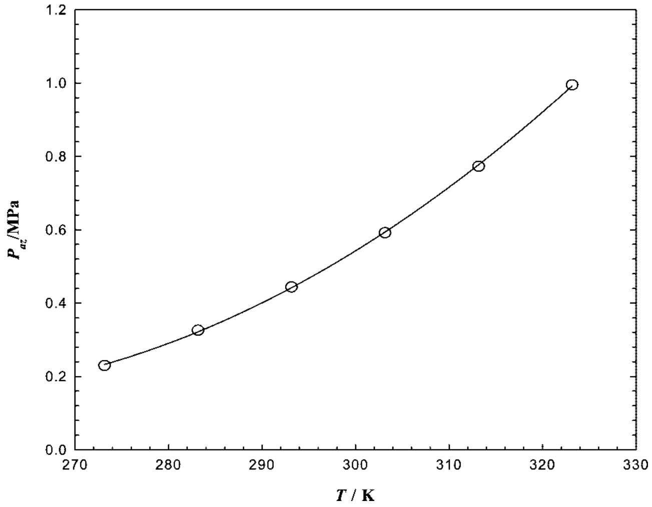

Figure 8. Variation of the azeotropic pressure with temperature for HFC-227ea (1) + butane (2). The equation of the fitting line is $P_{az} = 0.2 \cdot 10^{-3} (T/K)^2 - 0.0787(T/K) + 9.988$ (323.15 K ≥ T ≥ 273.15 K).

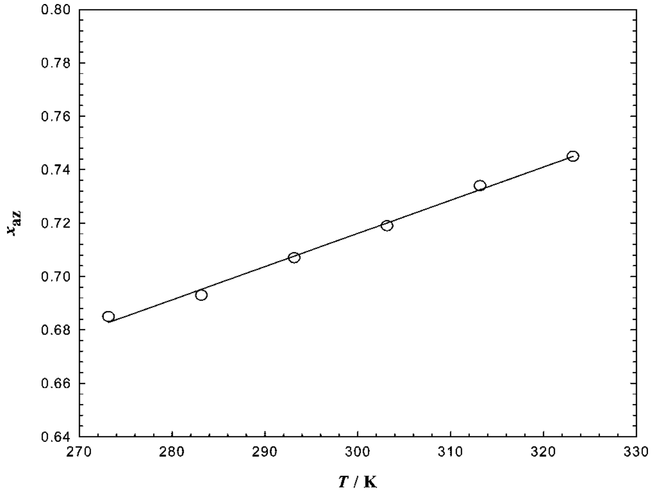

Figure 9. Variation of the azeotropic composition with temperature for HFC-227ea (1) + butane (2). The equation of the fitting line is $x_{az} = 0.8 \cdot 10^{-3}\ T/K + 0.6036$ (323.15 K ≥ T ≥ 273.15 K).

deviations of pressure (AARD-*P*) and average absolute deviation of vapor-phase composition (AAD-*y*) between measured and calculated values are reported in Table 4. They can be fitted with first-order polynomial equations for the temperature. The temperature dependence of the W–S mixing rule's binary parameter and the NRTL parameters are plotted in Figures 3, 4, and 5. The average values of AARD-*P* and AAD-*y* for all the six temperatures were 0.002 and 0.013 for the PR EOS. The results of error estimations with the PR EOS for *P* and *y* were shown in Figures 6, 7, 8, and 9, respectively.

This mixture exhibited an azeotrope. Azeotropic data of this mixture have been determined at the composition between 0.68 and 0.75 in mole fraction and at pressures between (0.23 and 1.00) MPa in the temperature range from (273.15 to 323.15) K. Azeotropic compositions and pressures are shown in Table 5 and Figures 5 and 6. As can be seen in these figures, azeotropic compositions and pressures are temperature dependent. In the range of experimental temperatures, the azeotropic compositions were correlated by the empirical equation $x_{az} = 0.8 \cdot 10^{-3}(T/K) + 0.6036$, and azeotropic pressure was correlated by the empirical equation $P_{az}/MPa = 0.2 \cdot 10^{-3}(T/K)^2 - 0.0787(T/K) + 9.988$. These equations were fitted within the temperature range $T \geq 273.15$ K.

Table 5. Azeotropic Pressure P_{az} and Azeotropic Liquid Mole Fraction x_{az} for HFC-227ea (1) + Butane (2) as a Function of Temperature

T / K	azeotropic pressure P_{az} / MPa	azeotropic composition x_{az} / (HFC-227ea mole fraction)
273.15	0.230	0.685
283.15	0.326	0.693
293.15	0.444	0.707
303.15	0.592	0.719
313.15	0.773	0.734
323.15	0.995	0.745

Conclusions

VLE data for binary systems of HFC-227ea (1) + butane (2) were measured at five equally spaced temperatures between (273.15 and 323.15) K using a circulation-type equilibrium apparatus. The experimental data were correlated with the PR EOS using the Wong–Sandler mixing rules. Calculated results with these equations have given satisfactory results in comparison with the experimental data. This system shows strong positive azeotropes for the full temperature range studied here. These results indicate that the Peng–Robinson equation of state with Wong–Sandler mixing rules can be used to estimate pressures and compositions for the mixture of HFC-227ea + butane in the range of experimental temperatures and can be applied to other ranges. However, this may need additional experiments to confirm it.

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Received for review August 26, 2007. Accepted September 26, 2007. We gratefully acknowledge financial support from the Korea Ministry of Commerce, Industry & Energy, and the Korea Energy Management Corporation and Sogang University Research Grant in 2007.

JE7004837