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Isothermal Vapor−Liquid Equilibria for the Binary System of Carbon Dioxide (CO_2) + 1,1,1,2,3,3,3-Heptafluoropropane (R-227ea)

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Isothermal vapor−liquid equilibrium (VLE) data for the binary mixture of carbon dioxide (CO_2) + 1,1,1,2,3,3,3-heptafluoropropane (HFC-227ea) were measured at six evenly spaced temperatures of (278.15, 288.15, 298.15, 308.15, 318.15, and 328.15) K. The data in the two-phase region were measured by using a circulation-type equilibrium apparatus in which both vapor and liquid phases are continuously recirculated. The experimental data were correlated with the Peng−Robinson equation of state (PR-EoS) using the Wong−Sandler (W−S) mixing rules combined with the nonrandom two-liquid (NRTL) excess Gibbs free energy model. The calculated values with the PR-EoS using the W−S mixing rules show good agreement with the experimental data.

Introduction

For many years, chlorofluorocarbons (CFCs) were extensively used as refrigerants due to many advantages such as nontoxicity, nonflammability, thermodynamic properties, and so on. However, the use of CFCs has been restricted by the Montreal Protocol (1989) because of their harmfulness to the ozone layer.[1] So, many researchers investigated CFC alternatives such as hydrochlorofluorocarbons (HCFCs) and hydrofluorocarbons (HFCs). However, the use of HCFCs has been banned by the Kyoto Protocol (2005) because of their high global warming potential (GWP). HFCs are synthetic refrigerants which consist of hydrogen, fluorine, and carbon. They are considered as alternative refrigerants for CFCs and HCFCs because they have zero ozone depletion potential (ODP).[2] HFC-227ea (1,1,1,2,3,3,3-heptafluoropropane, C_3HF_7) belongs to the halogenated aliphatic group. It has no ozone depletion effect and, in addition, leaves no residue on valuable equipment after discharge.[3] Unlike CFCs used as refrigerants, it has no chlorine atoms and, therefore, is not a potent ozone-depleting chemical.[1,3] On the other hand, carbon dioxide (CO_2) is a representative compound of the carbon cycle. It is natural refrigerant and often used in air conditioners and other cooling processes. To develop optimal alternative refrigerants for CFCs and HCFCs, many mixtures have been investigated. These mixtures satisfy the properties of original refrigerants conditions such as flammability and toxicity, but with less environmental impact (ozone depletion and global warming). Vapor−liquid equilibrium (VLE) data are necessary to design appropriate refrigeration process and determine optimal process conditions;[4,5] however, there are few experimental data reported in the literature for the CO_2 (1) + HFC-227ea system.[6]

In this work, isothermal VLE data for the binary mixture of CO_2 + HFC-227ea at six equally spaced temperatures from (278.15 to 328.15) K were measured by using a circulation-type equilibrium apparatus in which both phases were continuously recirculated. The experimental data have been correlated with the Peng−Robinson equation of state (PR-EoS)[7] using the Wong−Sandler (W−S) mixing rule[8] combined with the nonrandom two-liquid (NRTL) excess Gibbs free energy model. In the range of experimental temperature, the average absolute deviations of pressure and vapor phase compositions between experimental and calculated values were determined, and the relevant parameters are presented.

Experimental Section

Chemicals. High-grade chemicals of carbon dioxide and HFC-227ea were used for VLE measurement. Carbon dioxide was purchased from Dong-A Special Gas Co. (Korea) with 99.9 % mass purity. HFC-227ea was supplied by DuPont (U.S.) with a mass purity higher than 99.8 %. They were used without any further purification.

Apparatus. The experimental apparatus used in this study was a circulation type equilibrium apparatus, where both liquid and vapor phases were recirculated continuously. This apparatus was described well in our previous works,[9−12] so the explanation of this is omitted here. The temperatures of all equilibrium states were 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 Beamex (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 0.0001 MPa, and measurement uncertainty 0.0001 MPa. Therefore, total uncertainty of the pressure measurement is estimated to be within 0.001 MPa.

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Procedure. Experiments to measure VLE data for the binary system of CO_2 (1) + HFC-227ea (2) were performed using the following procedures. The system was first evacuated to remove all inert gases. A certain amount of HFC-227ea 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 CO_2 was introduced into the cell from a sample reservoir. Both the vapor and the liquid phases were recirculated by the dual-head circulation pump until an equilibrium state was established. It was observed that 2 h are 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 gas chromatograph (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 the mole fraction for both the liquid and the vapor phases.

Correlation

The experimental VLE data are correlated with the PR-EoS[7] using the W−S mixing rule.[8]

PR-EoS.

$$P = \frac{R(\mathrm{J \cdot mol^{-1} \cdot K^{-1}})T(\mathrm{K})}{\upsilon_M(\mathrm{m^3 \cdot mol^{-1}}) - b(\mathrm{m^3 \cdot mol^{-1}})} - \frac{a(T)(\mathrm{J \cdot mol^{-2} \cdot m^3})}{\upsilon_M(\upsilon_M + b) + b(\upsilon_M - b)} \quad (1)$$

with

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

$$b = 0.077796 \frac{RT_c}{P_c} \quad (3)$$

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

$$k = 0.37464 + 1.54226\omega - 0.26992\omega^2 \quad (5)$$

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, and υ_M is the molar volume of the mixture.

The W−S mixing rules[8] 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

Table 1. Thermodynamic Properties of Components[15]

chemical	chemical formula	T_c/K	P_c/MPa	ω
carbon dioxide (1)	CO_2	304.21	7.384	0.2239
HFC-227ea (2)	C_3HF_7	375.95	2.980	0.3632

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

with

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

and

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

where C is a numerical constant equal to $\ln(2^{1/2} - 1)/2^{1/2}$ for the PR-EoS used in this work. The single adjustable parameter (k_{ij}) for each binary pair is referred to as the W−S 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.[13] In this study, we used the NRTL model[14] given by

$$\frac{A_\infty^E}{RT} = \sum_i x_i \frac{\sum_j x_j G_{ji} \tau_{ji}}{\sum_r x_r G_{ri}} \quad (9)$$

with

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

The critical temperature (T_c), critical pressure (P_c), and acentric factor (ω) for both carbon dioxide and HFC-227ea 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 this system. The parameters of the PR-EoS were obtained by minimizing the following objective function.

$$\text{objective function} = \frac{1}{N} \sum_j^N \left[\left(\frac{P_{j,\exp} - P_{j,\mathrm{cal}}}{P_{j,\exp}}\right) \cdot 100\right]^2 \quad (11)$$

Results and Discussion

In this work, the equilibrium compositions for the CO_2 (1) + HFC-227ea (2) binary systems were measured at six equally spaced temperatures from (278.15 to 328.15) K. To verify the

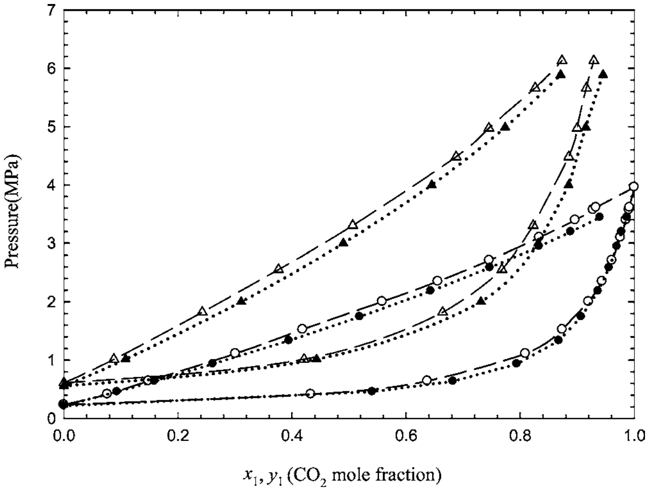

Figure 1. Comparison of our experimental data with literature data for the CO_2 (1) + HFC-227ea (2) system. Experimental data of this work: ○, 278.15 K; △, 308.15 K; and Valtz et al:[6] ●, 276.01 K; ▲, 305.17 K.

Table 2. Comparison of the Measured Pure Component Vapor Pressures with the Database REFPROP 6.01[15]

component	T/K	P_{exp}/MPa	P_{ref}/MPa	ΔP	$\lvert\Delta P\rvert/P_{exp}$
CO_2 (1)	278.15	3.960	3.965	−0.005	0.0013
	288.15	5.080	5.081	−0.001	0.0002
	298.15	6.425	6.425	0.000	0.0000
					avg. 0.0005
HFC-227ea (2)	278.15	0.234	0.232	0.002	0.0085
	288.15	0.327	0.330	−0.003	0.0092
	298.15	0.451	0.455	−0.004	0.0089
	308.15	0.611	0.613	−0.002	0.0033
	318.15	0.805	0.808	−0.003	0.0037
	328.15	1.040	1.046	−0.006	0.0058
					avg. 0.0066

Table 3. VLE Measurements for the CO_2 (1) + HFC-227ea (2) System

experimental data			PR-EoS			
P_{exp}/MPa	$x_{1,exp}$	$y_{1,exp}$	P_{cal}/MPa	$y_{1,cal}$	ΔP^a/MPa	$\Delta y_1{}^b$
			T/K = 278.15			
0.234	0.000	0.000	0.229	0.000	0.005	0.000
0.414	0.076	0.433	0.421	0.464	−0.007	−0.031
0.643	0.148	0.638	0.619	0.643	0.024	−0.005
1.112	0.301	0.810	1.087	0.810	0.023	0.000
1.527	0.419	0.874	1.489	0.870	0.038	0.004
2.003	0.558	0.920	2.001	0.914	0.003	0.006
2.351	0.656	0.944	2.378	0.936	−0.027	0.008
2.710	0.746	0.961	2.745	0.953	−0.035	0.008
3.099	0.834	0.976	3.129	0.969	−0.030	0.007
3.396	0.897	0.986	3.426	0.980	−0.030	0.006
3.572	0.928	0.991	3.581	0.986	−0.009	0.005
3.613	0.933	0.992	3.610	0.987	0.003	0.005
3.960	1.000	1.000	3.965	1.000	−0.005	0.000
			T/K = 288.15			
0.327	0.000	0.000	0.324	0.000	0.003	0.000
0.765	0.115	0.538	0.765	0.571	0.000	−0.033
1.310	0.263	0.748	1.325	0.757	−0.015	−0.009
1.910	0.414	0.847	1.910	0.839	0.000	0.008
2.351	0.520	0.886	2.341	0.877	0.010	0.009
2.758	0.609	0.911	2.737	0.903	0.021	0.008
3.103	0.686	0.930	3.104	0.922	−0.001	0.008
3.468	0.757	0.947	3.477	0.940	−0.009	0.007
3.785	0.813	0.959	3.799	0.953	−0.014	0.006
4.206	0.883	0.973	4.280	0.970	−0.074	0.003
4.557	0.929	0.984	4.556	0.981	0.001	0.003
5.080	1.000	1.000	5.094	1.000	−0.014	0.000
			T/K = 298.15			
0.451	0.000	0.000	0.447	0.000	0.004	0.000
0.945	0.110	0.470	0.923	0.504	−0.001	−0.034
1.448	0.227	0.683	1.449	0.684	−0.049	−0.001
1.889	0.326	0.780	1.903	0.760	0.036	0.020
2.282	0.407	0.829	2.282	0.803	0.019	0.026
2.930	0.537	0.882	2.927	0.856	0.023	0.026
3.544	0.649	0.914	3.540	0.892	0.040	0.022
4.102	0.741	0.931	4.115	0.919	−0.034	0.012
4.737	0.830	0.947	4.768	0.944	−0.062	−0.003
5.357	0.905	0.967	5.422	0.966	−0.085	0.007
5.785	0.948	0.979	5.858	0.980	−0.078	0.014
6.425	1.000	1.000	6.447	1.000	−0.022	0.016
			T/K = 308.15			
0.611	0.000	0.000	0.602	0.000	0.009	0.000
1.014	0.088	0.421	1.019	0.393	−0.005	0.028
1.820	0.242	0.665	1.806	0.651	0.014	0.014
2.551	0.376	0.768	2.535	0.751	0.016	0.017
3.303	0.506	0.823	3.303	0.812	0.000	0.011
4.482	0.687	0.885	4.5288	0.872	−0.047	0.013
4.971	0.745	0.900	4.983	0.890	−0.012	0.010
5.661	0.826	0.916	5.708	0.915	−0.047	0.001
6.129	0.873	0.929	6.193	0.930	−0.064	−0.001
			T/K = 318.15			
0.805	0.000	0.000	0.795	0.000	0.010	0.000
1.372	0.108	0.393	1.376	0.399	−0.004	−0.006
2.013	0.214	0.589	1.991	0.577	0.022	0.012
2.772	0.336	0.707	2.770	0.691	0.022	0.016
3.344	0.420	0.754	3.344	0.743	0.000	0.011
3.820	0.487	0.784	3.824	0.774	−0.004	0.010
4.799	0.617	0.824	4.829	0.821	−0.030	0.003
5.454	0.693	0.848	5.472	0.843	−0.018	0.005
6.095	0.764	0.863	6.114	0.861	−0.019	0.002
			T/K = 328.15			
1.040	0.000	0.000	1.033	0.000	0.007	0.000
1.655	0.105	0.345	1.660	0.348	−0.006	−0.002
2.399	0.216	0.542	2.391	0.531	0.009	0.011
3.103	0.314	0.648	3.083	0.627	0.020	0.021
3.799	0.406	0.716	3.799	0.690	0.000	0.026
4.385	0.481	0.745	4.405	0.727	−0.020	0.019
4.930	0.545	0.760	4.949	0.751	−0.020	0.008
5.661	0.631	0.774	5.693	0.776	−0.033	−0.002
6.281	0.699	0.780	6.289	0.787	−0.007	−0.006

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

consistency of our experimental data, we compared our experimental data at (278.15 and 308.15) K with literature data already reported by Valtz et al.[6] at similar temperatures of (276.01 and 305.17) K. As can be seen from Figure 1, although the exact comparison could not be made because of the temperature differences, the tendency of our experimental data agreed well with the literature data reported by Valtz et al.[6]

Table 2 shows the comparison of measured vapor pressures of pure CO_2 and HFC-227ea with those calculated from the database REFPROP 6.01,[15] which are considered to be reliable for the pure compounds considered and consistent with literature data. The absolute average relative deviation ($\sum|\Delta P/P|/N$) between measured and calculated values from the data of REFPROP 6.01[15] is 0.0005 for CO_2 and 0.0066 for HFC-227ea. The experimental VLE data and the results of the correlation are reported in Table 3. This table lists the measured mole fractions of the liquid and vapor phases, the pressures and temperatures in equilibrium, and the deviations between measured and calculated pressures (ΔP) and vapor compositions (Δy).

Figure 2 shows the comparison of measured and calculated values with the PR-EOS for the binary system of the CO_2 (1) + HFC-227ea (2) at six equally spaced temperatures of (278.15, 288.15, 298.15, 308.15, 318.15, and 328.15) K. As can be seen, the CO_2 + HFC-227ea system showed slightly negative deviations from the Raoult's law, and no azeotropes for all the temperature ranges studied here were observed. Also, the calculated values were well-matched with experimental data. The interaction parameters of the binary mixtures for each isotherm, the binary parameters k_{ij}, and the absolute average

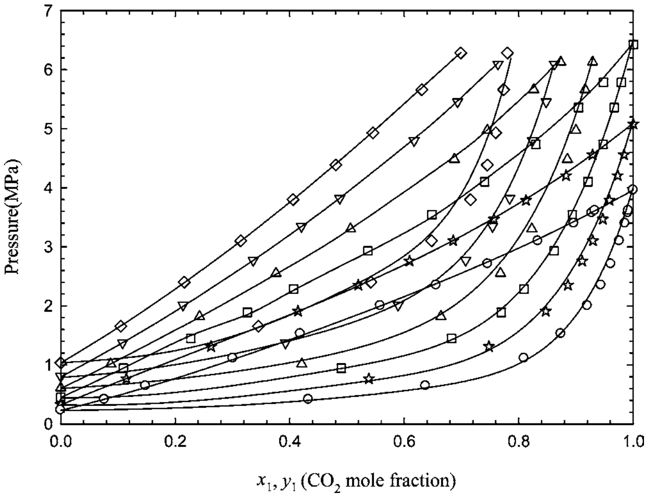

Figure 2. *P*−*x*−*y* diagram for the CO_2 (1) + HFC-227ea (2) system. Experimental data at various temperatures: ○, 287.15 K; ☆, 288.15 K; □, 298.15 K; △, 308.15 K; ▽, 318.15 K; ◇, 328.15 K; —, calculated with the PR-EoS using W−S mixing.

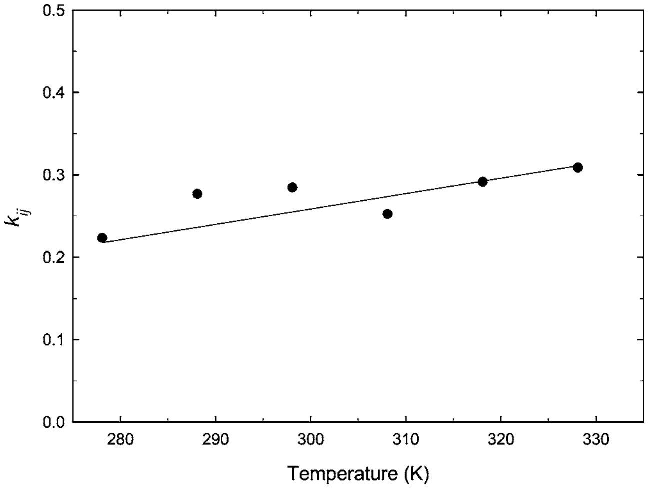

Figure 3. Parameter k_{ij} obtained for the PR-EoS with the W−S mixing rule. The equation of the fitting line is $k_{ij} = 0.0019\ T/K - 0.3027$ (328.15 K ≥ *T* ≥ 278.15 K).

deviations of pressure (AAD-*P* (%)) and the absolute average deviation of vapor-phase composition (AAD-*y*) between measured and calculated values are reported in Table 4. Figure 3 plots the binary parameters, k_{ij}, versus temperature. As can be seen in this figure and Table 4, the binary parameters, k_{ij} shows the temperature dependency. They can be fitted with a linear function of temperature given by eq 12.

$$k_{ij} = 0.0019T/\mathrm{K} - 0.3027 \qquad (12)$$

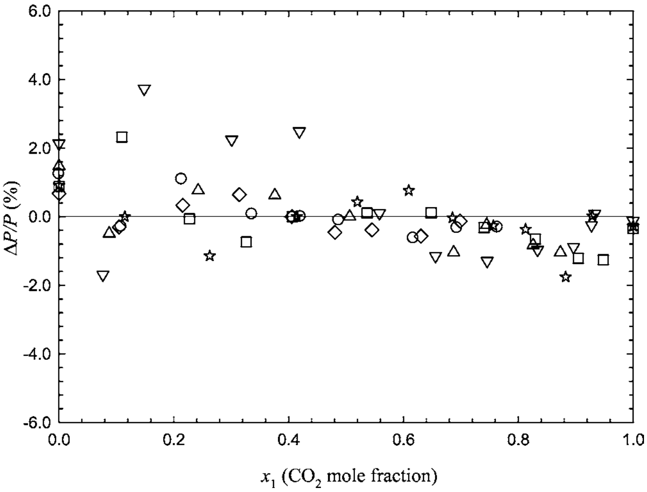

Figure 4. Deviation of pressure for the system CO_2 (1) + HFC-227ea (2) from the PR-EoS using the W−S mixing rule: ○, 287.15 K; ☆, 288.15 K; □, 298.15 K; △, 308.15 K; ▽, 318.15 K; ◇, 328.15 K.

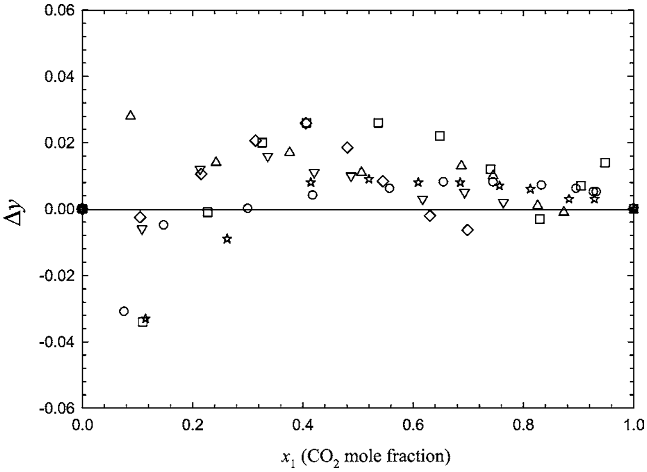

Figure 5. Deviation of vapor composition for the system CO_2 (1) + HFC-227ea (2) from the PR-EoS using the W−S mixing rule: ○, 287.15 K; ☆, 288.15 K; □, 298.15 K; △, 308.15 K; ▽, 318.15 K; ◇, 328.15 K.

In Figures 4 and 5, the deviations of pressure ($\Delta P/P_{\mathrm{exp}} \cdot 100$ (%)) and the vapor phase compositions (Δy_1) were plotted with the liquid phase compositions (x_1), point by point. The overall average values of AAD-*P* (%) and AAD-*y* through the temperature range from (278.15 to 328.15) K were 0.677 % and 0.0105, respectively. All values are small and acceptable. From these figures and the low average deviations of *P* and *y*, we conclude that the calculated values with the PR-EOS using the W−S mixing rules give good agreements with the experimental data.

Conclusions

The isothermal VLE data for the binary systems of CO_2 (1) + HFC-227ea (2) were measured at six equally spaced temperatures of (278.15, 288.15, 298.15, 308.15, 318.15, and 328.15) K using a circulation-type equilibrium apparatus. The

Table 4. Values of Binary Parameters and Average Absolute Deviations of *P* and *y*

	T/K		278.15	288.15	298.15	308.15	318.15	328.15
PR-EoS	binary parameter	k_{12}[a]	0.2225	0.2758	0.2837	0.2515	0.2905	0.3078
		τ_{21}[a]	−0.9741	−0.1619	0.2510	−0.6501	−0.8035	−0.8732
		τ_{12}[a]	2.8512	1.0382	0.4224	1.9342	2.0206	2.1188
	AAD-*P* (%)[b]		1.37	0.41	0.77	0.73	0.44	0.34
	AAD-*y*[c]		0.008	0.009	0.015	0.012	0.008	0.011

[a] k_{12}, τ_{21}, and τ_{12} are dimensionless. [b] AAD-*P* (%) = $(1/N)\sum|(P_{\mathrm{exp}} - P_{\mathrm{cal}})/P_{\mathrm{exp}}| \cdot 100$. [c] AAD-*y* = $(1/N)\sum|(y_{\mathrm{exp}} - y_{\mathrm{cal}})|$.

experimental VLE data were correlated with the PR-EoS using the W−S mixing rules. Calculated results with these equations show good agreement with our experimental data.

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