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# Vapor−Liquid Equilibria of the 1,1,1,2,3,3,3-Heptafluoropropane + Isobutene System

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In this study, isothermal vapor−liquid equilibrium data were measured for the 1,1,1,2,3,3,3-heptafluoropropane (HFC-227ea) + isobutene system in the temperature range from (288.15 to 363.15) K at 15 K intervals. The experiments were carried out with a circulating-type apparatus with online gas chromatography. The VLE data were correlated by the Peng−Robinson equation of state with the Wong−Sandler mixing rules involving the NRTL model. The calculated results show a good agreement with the measured data, and this system presents an azeotrope at each temperature.

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## Introduction

Environmental problems have become a serious global issue because the influence of the problems is widespread and critical. After the discovery of an ozone hole over Antarctica was reported,[1] stratospheric ozone depletion, especially, has led to great concerns about the global environmental change. Since the chlorine of chlorofluorocarbons (CFCs) acts as a catalytic agent in ozone depletion, the production and use of CFCs were regulated under the 1987 Montreal Protocol.[2] CFCs have been widely used in a variety of industrial, commercial, and household applications because of their nontoxic, nonflammable, and stable thermodynamic properties. Although hydrofluorocarbons (HFCs) have been considered as another option for the replacement of CFCs, some of them have been banned by Kyoto Protocol,[3] an agreement for reducing greenhouse gas emission and global warming. Because of zero ozone depletion potentials[4] (ODPs) and near zero global warming potentials[4] (GWPs), hydrocarbons (HCs), such as propane, *n*-butane, and propylene, have also been used as alternatives. However, HCs are flammable, and this characteristic has a possibility of the risk of fires in case of leakage. The mixture of the two possible alternatives, HFCs and HCs, shows the advanced properties from environmental and safe points of view. Therefore, many studies of mixtures of HFCs and HCs have been investigated.[5−10]

Vapor−liquid equilibrium (VLE) data for HFCs and HCs are essential to decide the optimal composition of the mixtures and to design refrigeration units. Although many VLE experiments for the mixture have been carried out, more VLE data are still needed to replace CFCs and to investigate the behavior of the alternative refrigerants. In this paper, we present isothermal VLE data of the HFC-227ea + isobutene system in the temperature range from (288.15 to 363.15) K at 15 K intervals. The experimental data were correlated by the Peng−Robinson equation of state[11] with the Wong−Sandler mixing rules[12,13] involving the NRTL model.[14]

## Experimental Section

***Chemicals.*** HFC-227ea was purchased from Ulsan Chemical Co. with a guaranteed purity higher than 99.9 %. Isobutene was supplied by Korean Industrial Gases with a guaranteed purity higher than 99.5 %. The purities of the chemicals were analyzed by gas chromatography, and the measured purities were higher than the certified purities. All components were used without further purification in these experiments.

**Table 1. Critical Properties and Acentric Factors of Pure Components (REFPROP 8.0[16])**

| | HFC-227ea | isobutene |
|---|---|---|
| $T_c$/K | 374.9 | 418.09 |
| $P_c$/MPa | 2.925 | 4.010 |
| $\omega$ | 0.357 | 0.193 |

**Table 2. Comparison between the Measured Vapor Pressures and the Values from REFPROP 8.0[16]**

| $T$/K | $P_{\mathrm{exp}}$/MPa | $P_{\mathrm{cal}}{}^{16}$/Mpa | $\Delta P^a$/MPa | $\lvert\Delta P/P_{\mathrm{exp}}\rvert$ |
|---|---|---|---|---|
| | | Isobutene | | |
| 288.15 | 0.223 | 0.223 | −0.001 | 0.004 |
| 303.15 | 0.352 | 0.354 | 0.002 | 0.005 |
| 318.15 | 0.536 | 0.536 | 0.000 | 0.000 |
| 333.15 | 0.785 | 0.781 | −0.004 | 0.005 |
| 348.15 | 1.107 | 1.100 | −0.007 | 0.006 |
| 363.15 | 1.515 | 1.505 | −0.010 | 0.007 |
| | | HFC-227ea | | |
| 288.15 | 0.335 | 0.331 | −0.004 | 0.012 |
| 303.15 | 0.529 | 0.528 | −0.001 | 0.002 |
| 318.15 | 0.804 | 0.804 | 0.000 | 0.000 |
| 333.15 | 1.176 | 1.176 | 0.000 | 0.000 |
| 348.15 | 1.664 | 1.665 | 0.001 | 0.001 |
| 363.15 | 2.294 | 2.298 | 0.004 | 0.002 |

[a] $\Delta P = P_{\mathrm{cal}} - P_{\mathrm{exp}}$.

***Apparatus.*** The measurement of the VLE data was conducted in a circulation-type apparatus. The details of this apparatus were given in our previous studies.[15] The equilibrium cell was made of 316-stainless steel and equipped with two windows for observing the inside of the cell. The inner volume of the cell is about 200 mL. Since two magnetic pumps made both phases repeatedly circulated, equilibrium was quickly reached. To trap the liquid and vapor sample, we used two sampling valves (Rheodyne Instruments, model 7413 with a 1 $\mu$L sampling loop for liquid, model 7010 with a 10 $\mu$L sampling loop for vapor). A gas chromatograph, which was equipped with a thermal conductivity detector (TCD) and Porapak-Q column (1.83 m

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**Table 3. Experimental VLE Data for the Binary System HFC-227ea (1) + Isobutene (2)**

| $P_{exp}$/MPa | $x_{1,exp}$ | $y_{1,exp}$ | $P_{cal}$/MPa | $y_{1,cal}$ | $\Delta P^a$/MPa | $\Delta y_1{}^b$ |
|---|---|---|---|---|---|---|
| $T$ = 288.15 K | | | | | | |
| 0.223 | 0.0000 | 0.0000 | 0.222 | 0.0000 | −0.002 | 0.0000 |
| 0.264 | 0.0544 | 0.1727 | 0.264 | 0.1876 | 0.000 | 0.0149 |
| 0.276 | 0.0764 | 0.2242 | 0.276 | 0.2324 | 0.000 | 0.0082 |
| 0.298 | 0.1343 | 0.3146 | 0.300 | 0.3145 | 0.002 | −0.0002 |
| 0.321 | 0.2121 | 0.3895 | 0.321 | 0.3866 | 0.000 | −0.0029 |
| 0.349 | 0.3714 | 0.5117 | 0.348 | 0.4935 | −0.001 | −0.0183 |
| 0.363 | 0.5154 | 0.5971 | 0.363 | 0.5805 | 0.000 | −0.0166 |
| 0.369 | 0.6823 | 0.6839 | 0.369 | 0.6846 | 0.000 | 0.0007 |
| 0.369 | 0.7503 | 0.7371 | 0.368 | 0.7306 | −0.001 | −0.0065 |
| 0.363 | 0.8234 | 0.7975 | 0.364 | 0.7855 | 0.001 | −0.0120 |
| 0.335 | 1.0000 | 1.0000 | 0.330 | 1.0000 | −0.006 | 0.0000 |
| $T$ = 303.15 K | | | | | | |
| 0.352 | 0.0000 | 0.0000 | 0.352 | 0.0000 | 0.000 | 0.0000 |
| 0.383 | 0.0205 | 0.0680 | 0.378 | 0.0796 | −0.005 | 0.0116 |
| 0.418 | 0.0611 | 0.1714 | 0.418 | 0.1848 | 0.000 | 0.0134 |
| 0.461 | 0.1282 | 0.2772 | 0.462 | 0.2878 | 0.001 | 0.0106 |
| 0.512 | 0.2570 | 0.4038 | 0.512 | 0.4027 | 0.000 | −0.0011 |
| 0.552 | 0.4375 | 0.5260 | 0.553 | 0.5225 | 0.001 | −0.0035 |
| 0.569 | 0.5832 | 0.6154 | 0.571 | 0.6178 | 0.002 | 0.0024 |
| 0.573 | 0.6661 | 0.6736 | 0.575 | 0.6749 | 0.002 | 0.0012 |
| 0.574 | 0.7380 | 0.7277 | 0.575 | 0.7274 | 0.001 | −0.0004 |
| 0.570 | 0.8265 | 0.8032 | 0.568 | 0.7989 | −0.002 | −0.0043 |
| 0.560 | 0.9035 | 0.8791 | 0.556 | 0.8729 | −0.004 | −0.0062 |
| 0.529 | 1.0000 | 1.0000 | 0.527 | 1.0000 | −0.003 | 0.0000 |
| $T$ = 318.15 K | | | | | | |
| 0.536 | 0.0000 | 0.0000 | 0.534 | 0.0000 | −0.002 | 0.0000 |
| 0.622 | 0.0600 | 0.1548 | 0.623 | 0.1672 | 0.001 | 0.0124 |
| 0.660 | 0.0948 | 0.2131 | 0.660 | 0.2264 | 0.000 | 0.0133 |
| 0.705 | 0.1500 | 0.2829 | 0.705 | 0.2949 | 0.000 | 0.0120 |
| 0.776 | 0.2776 | 0.3939 | 0.774 | 0.4035 | −0.002 | 0.0096 |
| 0.817 | 0.3939 | 0.4926 | 0.816 | 0.4844 | −0.001 | −0.0082 |
| 0.841 | 0.4949 | 0.5479 | 0.842 | 0.5533 | 0.001 | 0.0054 |
| 0.856 | 0.5920 | 0.6009 | 0.859 | 0.6216 | 0.003 | 0.0207 |
| 0.862 | 0.6500 | 0.6607 | 0.864 | 0.6639 | 0.002 | 0.0032 |
| 0.864 | 0.7142 | 0.7078 | 0.866 | 0.7128 | 0.002 | 0.0049 |
| 0.861 | 0.8086 | 0.7960 | 0.860 | 0.7900 | −0.001 | −0.0060 |
| 0.848 | 0.8990 | 0.8798 | 0.842 | 0.8754 | −0.006 | −0.0044 |
| 0.804 | 1.0000 | 1.0000 | 0.803 | 1.0000 | −0.001 | 0.0000 |
| $T$ = 333.15 K | | | | | | |
| 0.785 | 0.0000 | 0.0000 | 0.779 | 0.0000 | −0.006 | 0.0000 |
| 0.857 | 0.0354 | 0.0742 | 0.854 | 0.1008 | −0.003 | 0.0266 |
| 0.923 | 0.0787 | 0.1656 | 0.925 | 0.1845 | 0.002 | 0.0189 |
| 0.989 | 0.1310 | 0.2590 | 0.990 | 0.2553 | 0.001 | −0.0037 |
| 1.105 | 0.2666 | 0.3932 | 1.103 | 0.3786 | −0.002 | −0.0146 |
| 1.178 | 0.4046 | 0.4972 | 1.178 | 0.4794 | 0.000 | −0.0178 |
| 1.212 | 0.4995 | 0.5597 | 1.214 | 0.5484 | 0.002 | −0.0113 |
| 1.233 | 0.5793 | 0.6148 | 1.236 | 0.6084 | 0.003 | −0.0064 |
| 1.248 | 0.6747 | 0.6822 | 1.250 | 0.6833 | 0.002 | 0.0011 |
| 1.250 | 0.7354 | 0.7267 | 1.251 | 0.7331 | 0.001 | 0.0064 |
| 1.248 | 0.7809 | 0.7663 | 1.249 | 0.7720 | 0.001 | 0.0057 |
| 1.232 | 0.8961 | 0.8795 | 1.225 | 0.8797 | −0.007 | 0.0002 |
| 1.176 | 1.0000 | 1.0000 | 1.178 | 1.0000 | 0.001 | 0.0000 |
| $T$ = 348.15 K | | | | | | |
| 1.107 | 0.0000 | 0.0000 | 1.099 | 0.0000 | −0.007 | 0.0000 |
| 1.215 | 0.0433 | 0.1057 | 1.211 | 0.1055 | −0.004 | −0.0002 |
| 1.310 | 0.0941 | 0.1920 | 1.312 | 0.1892 | 0.002 | −0.0027 |
| 1.432 | 0.1785 | 0.2965 | 1.434 | 0.2842 | 0.002 | −0.0123 |
| 1.596 | 0.3436 | 0.4391 | 1.593 | 0.4186 | −0.003 | −0.0205 |
| 1.667 | 0.4496 | 0.5167 | 1.665 | 0.4991 | −0.002 | −0.0176 |
| 1.720 | 0.5707 | 0.6010 | 1.722 | 0.5955 | 0.002 | −0.0055 |
| 1.744 | 0.6552 | 0.6661 | 1.745 | 0.6664 | 0.001 | 0.0003 |
| 1.750 | 0.7090 | 0.7053 | 1.752 | 0.7128 | 0.002 | 0.0076 |
| 1.751 | 0.7642 | 0.7512 | 1.753 | 0.7617 | 0.002 | 0.0105 |
| 1.747 | 0.8203 | 0.7983 | 1.746 | 0.8128 | −0.001 | 0.0145 |
| 1.735 | 0.8749 | 0.8492 | 1.733 | 0.8649 | −0.002 | 0.0157 |
| 1.721 | 0.9192 | 0.8923 | 1.716 | 0.9095 | −0.005 | 0.0172 |
| 1.664 | 1.0000 | 1.0000 | 1.672 | 1.0000 | 0.008 | 0.0000 |
| $T$ = 363.15 K | | | | | | |
| 1.515 | 0.0000 | 0.0000 | 1.509 | 0.0000 | −0.007 | 0.0000 |
| 1.619 | 0.0315 | 0.0773 | 1.612 | 0.0715 | −0.007 | −0.0057 |
| 1.799 | 0.1069 | 0.1991 | 1.804 | 0.1887 | 0.005 | −0.0104 |
| 1.925 | 0.1712 | 0.2761 | 1.926 | 0.2581 | 0.001 | −0.0180 |
| 2.123 | 0.3119 | 0.4001 | 2.123 | 0.3758 | 0.000 | −0.0243 |
| 2.276 | 0.4591 | 0.5146 | 2.271 | 0.4908 | −0.005 | −0.0237 |
| 2.348 | 0.5637 | 0.5942 | 2.345 | 0.5786 | −0.003 | −0.0156 |
| 2.389 | 0.6702 | 0.6764 | 2.390 | 0.6752 | 0.001 | −0.0012 |
| 2.401 | 0.7639 | 0.7549 | 2.402 | 0.7636 | 0.001 | 0.0087 |
| 2.395 | 0.8278 | 0.8094 | 2.394 | 0.8245 | −0.001 | 0.0151 |
| 2.378 | 0.8792 | 0.8551 | 2.379 | 0.8743 | 0.001 | 0.0193 |
| 2.294 | 1.0000 | 1.0000 | 2.309 | 1.0000 | 0.015 | 0.0000 |

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

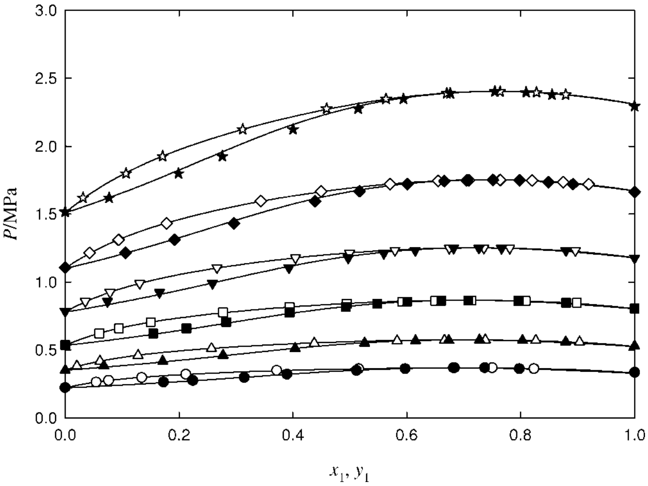

**Figure 1.** Vapor−liquid equilibria of the HFC-227ea (1) + isobutene (2) system. −, the PR EOS using the Wong−Sandler mixing rules; ●, experimental data at 288.15 K; ▲, 303.15 K; ■, 318.15 K; ▼, 333.15 K; ♦, 348.15 K; ★, 363.15 K.

**Table 4. Binary Parameters and Average Absolute Deviations (AAD)**

| $T$/K | $k_{ij}$ | $A_{ij}$/K | $A_{ji}$/K | AAD-$P^a$/% | AAD-$y^b$ |
|---|---|---|---|---|---|
| 288.15 | −0.5166 | 618.38 | 705.60 | 0.192 | 0.0089 |
| 303.15 | −0.4400 | 529.87 | 675.36 | 0.346 | 0.0055 |
| 318.15 | −0.3655 | 482.64 | 637.24 | 0.213 | 0.0083 |
| 333.15 | −0.2973 | 423.58 | 606.86 | 0.197 | 0.0102 |
| 348.15 | −0.2449 | 379.19 | 575.81 | 0.153 | 0.0104 |
| 363.15 | −0.1575 | 308.54 | 520.52 | 0.135 | 0.0142 |

[a] AAD-$P = (100/N)\sum_i^N(|P_{i,cal} - P_{i,exp}|/P_{i,exp})$. [b] AAD-$y = (1/N)\sum_i^N|y_{i,cal} - y_{i,exp}|$.

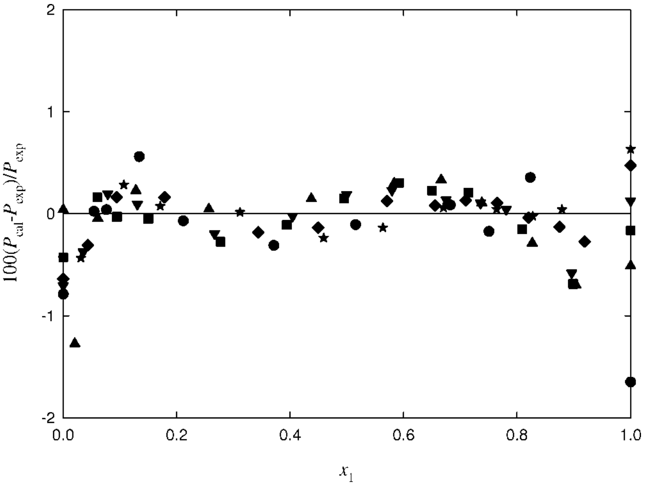

**Figure 2.** Deviation between calculated and experimental VLE pressure for the binary system HFC-227ea (1) + isobutene (2): ●, 288.15 K; ▲, 303.15 K; ■, 318.15 K; ▼, 333.15 K; ♦, 348.15 K; ★, 363.15 K.

long; 3.18 mm diameter; mesh range, 80/100), was connected online to the equilibrium apparatus. Helium was used as a carrier gas. The temperature in the cell was measured with a 100 Ω platinum resistance thermometer (Hart Scientific Co., model 5614) and a digital indicator (Hart Scientific Co., model 1502A) with an accuracy of ± 0.05 K. The pressure of the cell was measured with a pressure transducer (Sensotec Co., model Super TJE, (0 to 1000) psia) connected to a digital indicator (Laurel, model L20001WM1). An accuracy of the digital pressure gauge is 0.05 % of full scale. Calibration of the pressure transducer and thermometer was done by Korea Testing Laboratory (KTL), the national calibration laboratory.

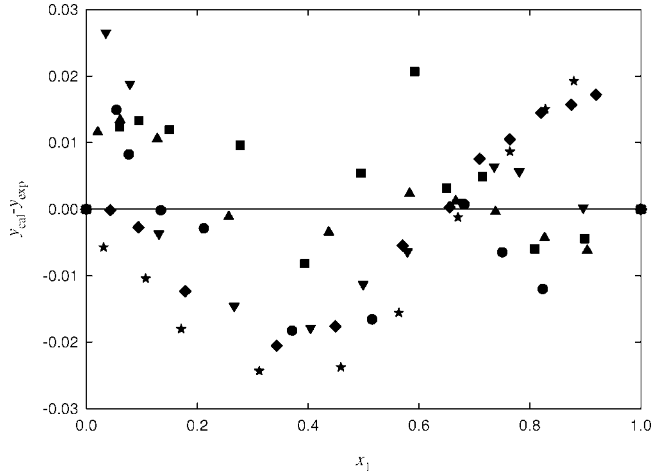

**Figure 3.** Deviation of vapor mole fraction for the HFC-227ea (1) + isobutene (2) system: ●, 288.15 K; ▲, 303.15 K; ■, 318.15 K; ▼, 333.15 K; ♦, 348.15 K; ★, 363.15 K.

**Table 5. Calculated Azeotropic Compositions and Pressures for the HFC-227ea + Isobutene System**

| $T$/K | $x_1$ | $P$/MPa |
|---|---|---|
| 288.15 | 0.689 | 0.369 |
| 303.15 | 0.697 | 0.575 |
| 318.15 | 0.708 | 0.866 |
| 333.15 | 0.722 | 1.251 |
| 348.15 | 0.741 | 1.753 |
| 363.15 | 0.757 | 2.402 |

***Experimental Procedure.*** First of all, the whole system was evacuated with a vacuum pump to remove all impurities. The cell was loaded with a less volatile component, isobutene, and then a proper amount of HFC-227ea was charged. To reduce the time required to reach equilibrium, the vapor and liquid phases of the mixture were circulated separately by two magnetic pumps. After the equilibrium was reached, each sample of the binary mixture was taken into the gas chromatography, and the peak area ratio was obtained. The calibration was carried out to convert the peak area ratio to the composition of the sample. The square correlation coefficients ($R^2$) of the calibration data were better than 0.999. To obtain reliable data for each equilibrium state, at least five samples of vapor and liquid were measured. Deviations of the equilibrium composition were less than 0.001 in the mole fraction.

## Results and Discussion

Isothermal vapor−liquid equilibrium data for the binary system HFC-227ea (1) + isobutene (2) were measured in a temperature range from (288.15 to 363.15) K. Experimental data were correlated with the Peng−Robinson equation of state[11] (PR EOS) using the Wong−Sandler mixing rules.[12,13] The PR EOS and the Wong−Sandler mixing rules are expressed as follows

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

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

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

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

$$\kappa = 0.37464 + 1.54226\omega - 0.26992\omega^2 \qquad (5)$$

where $T_c$ is the critical temperature; $P_c$ is the critical pressure; $T_r$ is the reduced temperature; and $\omega$ is the acentric factor. The critical properties and acentric factors are listed in Table 1.

The Wong−Sandler mixing rule is represented as

$$b_m = \frac{\sum_i\sum_j x_i x_j\left(b - \frac{a}{RT}\right)_{ij}}{1 - \sum_i x_i\frac{a_i}{b_iRT} - \frac{A_\infty^E}{CRT}} \qquad (6)$$

$$a_m = b_m\left(\sum_i x_i\frac{a_i}{b_i} + \frac{A_\infty^E}{C}\right) \qquad (7)$$

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

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 equal to the excess Gibbs free energy ($G^E$) at low pressure, the $G^E$ model is used in place of $A_\infty^E$. We used the NRTL model[14] as a $G^E$ model in this study

$$\frac{G^E}{RT} = \sum_i x_i\frac{\sum_i x_i\tau_{ji}G_{ji}}{\sum_k x_kG_{ki}} \qquad (9)$$

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

$$\tau_{ij} = A_{ij}/T \qquad (11)$$

where $\tau_{ij}$ and $\tau_{ji}$ are the interaction parameters and $\alpha_{ij}$ is the nonrandomness parameter. In this study, the nonrandomness parameter $\alpha_{ij}$ was taken as 0.3. The Simplex algorithm was used to minimize the following objective function

$$F = \frac{1}{N}\sum_i^N\left(\frac{P_{i,\mathrm{exp}} - P_{i,\mathrm{cal}}}{P_{i,\mathrm{exp}}}\right)^2 \qquad (12)$$

where $N$ is the number of experimental data points and $P_{\mathrm{exp}}$ and $P_{\mathrm{cal}}$ are the experimental and the calculated pressures, respectively.

The comparison of the measured vapor pressures of pure components with the values calculated from the database REFPROP 8.0[16] is provided in Table 2, and experimental and calculated data for binary system of HFC-227ea (1) + isobutene (2) are given in Table 3. As can be seen in Figure 1, the calculated results are in good agreement with the measured data at each temperature. Binary parameters, the average absolute deviation (AAD) of pressure, and AAD of vapor composition are listed in Table 4. The parameters are linear with temperature, and the AAD-$P$ and AAD-$y$ were less than 0.346 % and 0.0142, respectively. Figures 2 and 3 illustrate the deviations of $P$ and $y$ between the experimental data and the calculated values. This mixture shows azeotropes in the measured temperature range. The calculated azeotropic compositions and pressures are provided in Table 5.

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