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Vapor−Liquid Equilibrium Data for the Binary Mixture Difluoroethane (HFC-32) + Pentafluoroethane (HFC-125) of an Alternative Refrigerant

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Isothermal vapor−liquid equilibrium data for the binary system difluoroethane (HFC-32) + pentafluoroethane (HFC-125) were measured at temperatures from (265.15 to 303.15) K by the circulating method in this work. The vapor−liquid equilibrium data were correlated by the Peng−Robinson equation of state combined with a linear combination of the Vidal and Michelsen mixing rule and nonrandom two-liquid model. The results reveal that the correlated data are in a good agreement with the experimental values.

Introduction

It is very important to have accurate and reliable information about the thermophysical properties on the working fluids for research and development of energy-efficient, cost-effective, and environment-friendly industrial equipment. Over the past several decades among various hydrochlorofluorocarbon (HCFC) refrigerants, chlorodifluoroethane (HCFC-22) has been the most widely used working fluid in refrigeration and air conditioning systems because of its excellent thermal properties. However, much evidence has shown the destruction of the ozone layer and the global warming problems related to HCFC-22. This popular refrigerant will be phased out before 2020 because the chlorine it contains has an ozone depletion potential (ODP) of 0.055, and it has a comparatively high global warming potential of 1500 based on the time horizons of 100 years.[1−2] As a result, it has become an urgent subject to develop a substitute for HCFC-22 in recent years. The technical committee for the Alternative Refrigerant Evaluation Program (AREP) has proposed an updated list of the potential alternatives to HCFC-22. Some of the alternative on the AREP's list are R410A, R410B, R407C, and R507. Among these alternatives, refrigerant R410A is the widely used zero ODP refrigerant and is currently recognized as the main replacement to HCFC-22. It is a mixture with mass fraction of 50 % of difluoroethane (HFC-32) and pentafluoro-ethane (HFC-125). Because of its current and future importance, accurate knowledge of its thermodynamic, thermophysical, and heat transfer properties is essential to design economic refrigeration cycles.[3] Details of heat transfer for refrigerant R410A has been reported in the open literatures.[4−5] However, other thermodynamic parameters, such as enthalpy, entropy, and specific heat, can be calculated by means of the vapor−liquid equilibrium (*TPxy*) data for the binary mixture (HFC-32 + HFC-125). The literatures[6−20] mentioned the vapor−liquid equilibrium and other thermophysical data of the system (HFC-32 + HFC-125) at different temperatures. For example, Horstmann et al.[7] gave few data about $TPxH^E$ at 308.15 K, Weber[9] gave the *PVT* data from (294.68 to 333.77) K, Lee at el.[12] measured the vapor−liquid equilibrium data (*TPxy*) at temperatures of (303.15 and 323.15) K, Kato et al.[14] measured the critical vapor−liquid equilibrium data (*TPxy*) from (318.15 to 349.15) K, Higashi[15] measured the vapor−liquid equilibrium data (*TPxy*) from (283.05 to 313.04) K, Benmansour et al.[16−17] measured large numbers of data ρ at variable P and constant x_1 and T.

Table 1. Vapor−Liquid Equilibrium (T, p, x_1, y_1) Data for the System HFC-32 (1) + HFC-125 (2) at Temperatures of (265.15 and 303.15) K

T/K	x_1	y_1	P/kPa	T/K	x_1	y_1	P/kPa
265.15	0.0000	0.0000	517.0	293.15	0.0000	0.0000	1205.0
	0.0937	0.1179	537.3		0.1736	0.2288	1299.1
	0.1708	0.2282	553.3		0.2948	0.3500	1340.8
	0.2427	0.3077	561.8		0.3244	0.3875	1348.4
	0.2583	0.3109	562.8		0.3986	0.4571	1373.6
	0.2909	0.3441	567.8		0.4780	0.5382	1398.6
	0.4442	0.5035	591.4		0.5569	0.6027	1422.6
	0.5468	0.5937	603.9		0.6075	0.6477	1437.3
	0.6350	0.6779	613.4		0.6706	0.6968	1450.3
	0.6897	0.7162	619.6		0.7338	0.7562	1460.7
	0.7736	0.7783	624.8		0.7972	0.8103	1470.4
	0.8931	0.8916	628.1		1.0000	1.0000	1475.0
	0.9108	0.9062	628.6				
	1.0000	1.0000	624.1				
275.15	0.0000	0.0000	714.3	303.15	0.0000	0.0000	1567.0
	0.0723	0.1004	732.8		0.1640	0.2086	1683.5
	0.1015	0.1309	740.4		0.2714	0.3140	1736.6
	0.2289	0.2876	776.8		0.3798	0.4321	1781.7
	0.2833	0.3427	788.6		0.4716	0.5132	1816.0
	0.3125	0.3811	802.6		0.5539	0.5908	1839.4
	0.4286	0.4952	824.6		0.6219	0.6589	1869.6
	0.4827	0.5434	837.5		0.6722	0.6918	1887.0
	0.6170	0.6666	859.1		0.7394	0.7588	1893.4
	0.7747	0.7938	867.1		0.7912	0.7982	1905.9
	0.8395	0.8486	872.5		1.0000	1.0000	1925.0
	0.8657	0.8665	881.1				
	0.9274	0.9021	878.9				
	1.0000	1.0000	866.5				
283.15	0.0000	0.0000	908.8				
	0.0956	0.1209	926.8				
	0.1926	0.2344	962.1				
	0.2293	0.2815	980.2				
	0.3124	0.3718	1007.0				
	0.3255	0.3865	1013.0				
	0.3669	0.4261	1026.6				
	0.5021	0.5556	1059.2				
	0.6271	0.6761	1083.8				
	0.6458	0.6870	1089.2				
	0.7211	0.7483	1096.5				
	0.7936	0.8076	1106.6				
	0.8548	0.8690	1109.4				
	0.9104	0.9156	1112.3				
	1.0000	1.0000	1107.0				

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Table 2. Critical Parameters and Acentric Factors

substance	formula	$M/(\mathrm{g{\cdot}mol^{-1}})$	T_c/K	P_c/MPa	$V_c/(\mathrm{cm^3{\cdot}mol^{-1}})$	Z_c	ω
HFC-32	CH_2F_2	52.02	351.6	5.83	122.688	0.242	0.271
HFC-125	C_2HF_5	120.03	339.20	3.59	210.099	0.270	0.303

Table 3. Correlated Results of the Vapor−Liquid Equilibrium Data for the Binary Mixture HFC-32 (1) + HFC-125 (2) at Different Temperatures Using PR + LCVM Model

		PR + LCVM					
T/K	Np	Δy	$100\ \delta P$	$\Delta y_{\max}$	$100\ \delta P_{\max}$	τ_{12}	τ_{21}
265.15	14	0.0067	0.8887	0.0161	1.8073	0.8474	−0.4986
275.15	14	0.0077	1.3053	0.0125	2.2426	1.4461	−0.8320
283.15	14	0.0068	1.4027	0.0150	2.1716	1.0295	−0.6641
293.15	12	0.0094	0.4804	0.0200	0.9237	0.3489	−0.1371
303.15	11	0.0072	0.9062	0.0123	1.0692	−0.8344	1.3778

All, especially Benmansour et al.,[16−17] gave some important vapor−liquid equilibrium data ($P\rho Tx$) for (HFC-32 + HFC-125), but the vapor−liquid equilibrium ($TPxy$) data from (265.15 to 313.15) K are still scarce in actual application and theoretical study from the existing literatures. To complement previous studies for the binary mixture (HFC-32 + HFC-125), vapor−liquid equilibrium data at temperatures from (265.15 to 303.15) K were measured in this work, and the experimental data were correlated with the Peng−Robinson equation of state combined with the linear combination of the Vidal and Michelsen (LCVM) mixing rule and the nonrandom two-liquid (NRTL) excess free energy model. Over the range of experimental temperatures, the average absolute deviations of pressures and vapor-phase compositions between experimental and calculated values were determined, and the relevant parameters are presented.

Experiment

Materials. Chemicals of difluoroethane and pentafluoroethane were supplied by the Zhejiang Chemical Engineering Research Institute, China, with its mass fraction purity of 99.98 %, respectively. The purities of the chemicals were certified by the manufacturer. The samples were used without further purification.

Vapor−Liquid Equilibrium Apparatus. Temperature, pressure, liquid, and vapor compositions (T, P, x, y) were obtained from experiments with a recirculating still.[21] The experimental apparatus consists of a stainless steel equilibrium cell with its volume of 80 mL fitted with a motor stirrer coupled to an external drive. A visual observation of the solution in the cell was made to observe the phase behavior. The cell was thermostated in a bath, fitted with glass windows, and insulated with polyurethane foam. The bath temperature stability has been determined as ± 0.01 K at temperatures in the range (243.15 to 313.15) K. The temperature is measured with a platinum resistance thermometer (model WZP-II) with a given uncertainty of ± 0.01 K. The pressure was measured with a pressure transducer (model PMP4010, DRUCK, England) and differential pressure null transducer (model 1151DP, from Xi'an Instrument Factory, China). The total uncertainty in pressure measurement was estimated to be ± 0.0014 MPa.

A circulation loop from vapor to liquid phase was used to accelerate the equilibration by a vapor pump. The compositions of the samples were analyzed by immediately injecting them into the gas chromatograph (GC) equipped with a flame ionization detector (FID) (model GC112A, China), 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. The experimental data at the equilibrium state were measured at least three times to ensure repeatability. Considering the margin of error and the reproducibility of the GC, the overall uncertainty in the measurement of the composition was estimated to be within ± 0.002 mole fraction for both the liquid and vapor phases.

Experimental Procedures. The experiment for the binary system was performed by the following procedure: (1) the system was first evacuated; (2) (HFC-32 + HFC-125) was introduced into cell, and then the temperature of the entire system was maintained by the thermostatic bath; (3) the vapor in the cell was circulated continuously with the magnetic-activated circulation pump until an equilibrium state was established; (4) about 1 h was sufficient to obtain thermal equilibrium between the cell fluid and the thermostatic bath; (5) after the desired temperature was attained, the pressure in the equilibrium cell was measured and the (vapor + liquid) samples were withdrawn from the recycling lines for analysis; and (6) the compositions of the samples were measured by immediately injecting them into the GC. The experimental data at the equilibrium state were measured at least three times to ensure good reproducibility.

Chromatogram Analysis. The experimental conditions of the GC were as follows: (1) an FID; (2) the chromatograph column was 4 m stainless steel with an inner diameter 3 mm with GDX101 packing; (3) the rate of flow for carrier gas (nitrogen) was 37.1 mL/min, rate of flow for air was 252.1 mL/min, and rate of flow for hydrogen was 43.9 mL/min; (4) column temperature was 363.15 K, injection temperature was 393.15 K, and detector temperature was 393.15 K; (5) liquid sample was 2 μL and vapor sample was 15 μL.

Vapor−Liquid Equilibrium Data for HFC-32 (1) + HFC-125 (2). Vapor−liquid equilibrium (T, P, x_1, y_1) data for the binary mixture HFC-32 (1) + HFC-125 (2) at the temperatures from (265.15 to 303.15 K) were measured and listed in Table 1.

Model

Peng−Robinson (PR) Equation of State. The PR equation-of-state[22] given by

$$P = \frac{RT}{v - b} - \frac{a}{v^2 + 2vb - b^2} \tag{1}$$

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

$$b = 0.07780RT_c/P_c \tag{3}$$

where P is the pressure, v is the molar volume, T is the absolute temperature, R is the universal gas constant, a and b are adjustable parameters, T_c is the critical temperature, P_c is the critical pressure, and ω is the acentric factor. The values of T_c, P_c, and ω for each pure substance are listed in Table 2, and these were obtained from refs 23 and 24.

LCVM Mixing Rule. The mixing rules used to correlate the vapor−liquid equilibrium data are the LCVM mixing rule[25]

$$\frac{a_\mathrm{m}}{b_\mathrm{m}RT} = \lambda\alpha_v + (1-\lambda)\alpha_M = \left(\frac{\lambda}{C^\mathrm{HV}} + \frac{1-\lambda}{C^\mathrm{MHV1}}\right)\frac{G_\gamma^\mathrm{E}}{RT} + \frac{1-\lambda}{C^\mathrm{MHV1}}\sum_i x_i \ln\left(\frac{b_\mathrm{m}}{b_i}\right) + \sum_i x_i \frac{a_i}{b_iRT} \tag{4}$$

$$b_\mathrm{m} = \sum_i x_ib_i \tag{5}$$

where G is the Gibbs energy, $\alpha = a/(bRT)$, subscript m indicates a mixture, superscripts E, HV, and MHV1 indicate excess property, Huron−Vidal mixing rule, the first Modified Huron−Vidal mixing rule, respectively, x_i is liquid mole fraction, and λ is a parameter that determines the relative contributions of MHV1 and HV models and is obtained by fitting bubble-point and dew-point pressure for a variety of binary mixtures at high and low pressures. The empirical value of parameter λ is set equal to 0.36.[25]

Fugacity Coefficient. With the mixing rule for *a* and *b*, the fugacity coefficient of component *i* is

$$\ln\phi_i = b_i/b_\mathrm{m}(Z-1) - \ln(Z-B) - \frac{1}{2\sqrt{2}}\bar{\alpha}\ln\left(\frac{Z+(1+\sqrt{2})B}{Z+(1-\sqrt{2})B}\right) \tag{6}$$

where

$$A = a_\mathrm{m}P_\mathrm{c}/(RT_\mathrm{c})^2; \qquad B = b_\mathrm{m}P_\mathrm{c}/RT_\mathrm{c};$$

$$\bar{\alpha} = 1/RT\,(a_i/b_i - RT(\lambda/C^\mathrm{HV} + (1-\lambda)/C^\mathrm{MHV1})\ln\gamma_i - (1-\lambda)/C^\mathrm{MHV1}(\ln b_\mathrm{m}/b_i + b_i/b_\mathrm{m} - 1))$$

γ_i is the activity coefficient of component *i* and can be calculated from the NRTL model.

Results and Discussion

Through the use of the LCVM with PR equation-of-state model, vapor−liquid equilibrium data for the binary mixture (HFC-32 + HFC-125) were correlated in which the NRTL activity coefficient model[26] was used to calculate the excess Gibbs energy (α_{12} was recommended to 0.3). The correlation was made by minimizing the following objective function:

$$\mathrm{OF} = \frac{1}{Np}\sum_i^{Np}((y_1 - y_{1,\mathrm{cal}})_i^2 + (y_{2,} - y_{2,\mathrm{cal}})_i^2) \tag{7}$$

where *N*p is the number of data points, and y_i and $y_{i,\mathrm{cal}}$ are experimental and calculated vapor mole fractions.

For the correlation of the experimental data, a computer program had been developed applying the least-squares method. The correlated results are shown in Table 3 and Figure 1. The deviations, δP and Δy, listed in the tables were defined by

$$\delta p = 1/Np\sum_i |(P_\mathrm{cal} - P)/P|_i \tag{8}$$

$$\Delta y = 1/Np\sum_i |y_\mathrm{cal} - y|_i \tag{9}$$

where *N*p is the number of data points, $P_{i,\mathrm{cal}}$ and P_i are calculated pressure and experimental pressures, respectively.

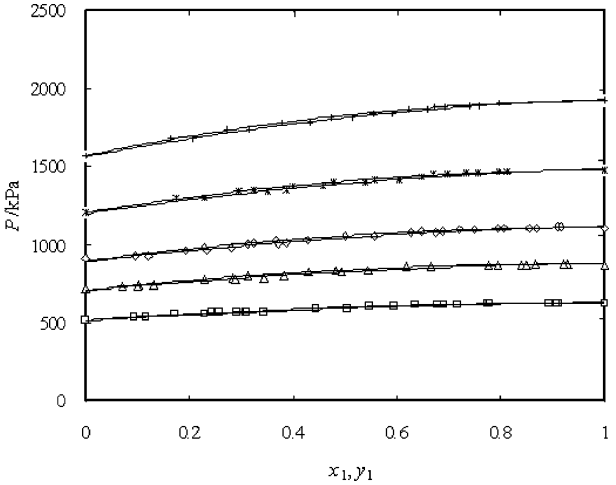

Figure 1. Pressure-composition phase diagram for the binary mixture HFC-32 (1) + HFC-125 (2). Experimental data in this work: □, 265.15 K; △, 275.15 K; ◇, 283.15 K; ×, 293.15 K; +, 303.15 K; −, calculated with PR + LCVM.

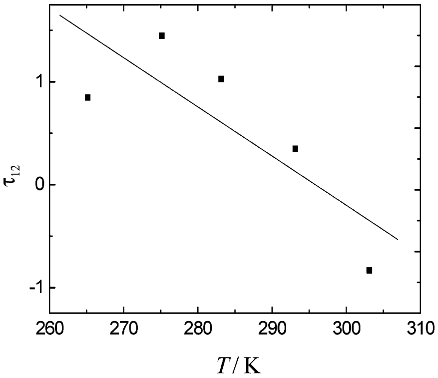

Figure 2. τ_{12} NRTL binary parameter as a function of temperature. ◆, fitted on isothermal data; −, solid line calculated through eq 10.

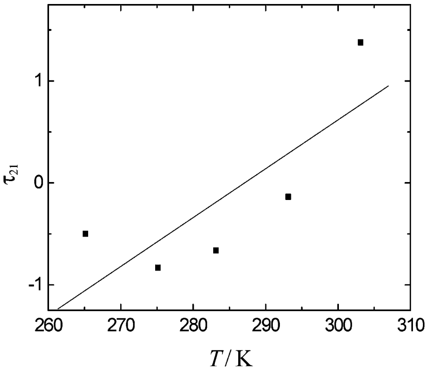

Figure 3. τ_{21} NRTL binary parameter as a function of temperature. ◆, fitted on isothermal data; −, solid line calculated through eq 11.

In the calculations, we have adjusted the two NRTL parameters (τ_{12}, τ_{21}). They appear slightly temperature dependent as shown in Figures 2−3. Linear relationships are convenient for their representations:

$$\tau_{12} = -0.0478T/\mathrm{K} + 14.14 \tag{10}$$

$$\tau_{21} = 0.0479T/\mathrm{K} - 13.76 \tag{11}$$

From the results listed in Table 3 and Figure 1, we can see that over a wide range of temperatures and pressures the PR + LCVM model has a good agreement with existing experimental data, that is, the data show small deviations from the model, and average deviations for both vapor composition and pressure are 0.007 and 0.99 % respectively.

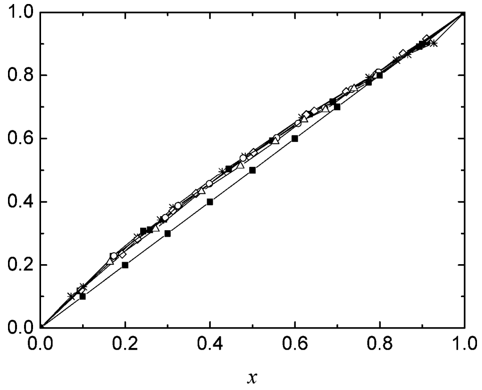

Figure 4. The curves $y = f(x)$ at different temperature cross the line $y =$ x: −◆−, 265.15 K; −◇−, 283.15 K; −○−, 293.15 K; −△−, 303.15 K; −*−, 275.15 K; −■−, $y = x$.

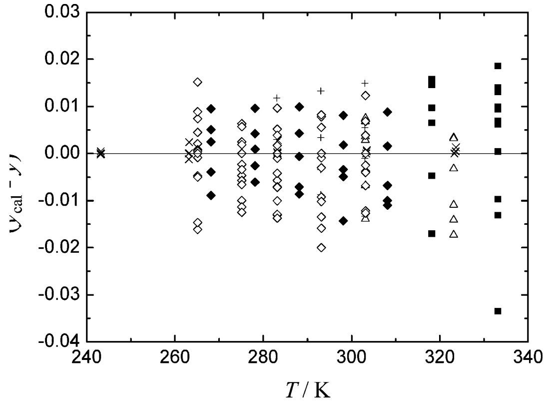

Figure 5. Deviations of vapor composition for the system HFC-32 (1) + HFC-125 (2) from the PR + LCVM model for the experimental data of literatures: ×, Nagel et al.;[6] ◆, Jung et al.;[8] +, Higashi;[11] △, Lee et al.;[12] ■, Kato et al.;[14] ◇, this work.

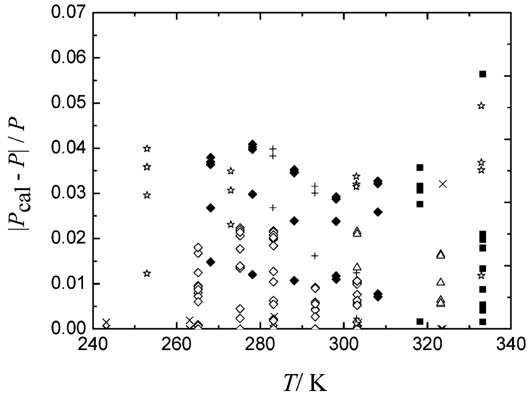

Figure 6. Deviations of pressure for the system HFC-32 (1) + HFC-125 (2) from the PR + LCVM model for the experimental data of literatures: ×, Nagel et al.;[6] ◆, Jung et al.;[8] +, Higashi;[11] △, Lee et al.;[12] ■, Kato et al.;[14] ☆, Benmansour et al.;[17] ◇, this work.

The system exhibits a positive deviation from the Raoult's law with the presence of an azeotrope from Figure 1. To clearly see the azeotropic behavior, the curves $y = f(x)$ at the different temperatures and line $y =$ x are shown Figure 4. From Figure 4, we can see that the curves $y = f(x)$ cross the line $y =$ x, that is, azeotropic behavior is displayed. Azeotropic data of the system (HFC-32 + HFC-125) have been determined at the composition between (085 and 0.93) in mole fraction and pressure between (630 kPa and 1.95 MPa) in the temperature ranges from (265.15 to 303.15) K.

In the system, bubble-point pressure line is similar to the dew-point pressure line, and there is a small variation in the vaporation temperature of 0.1 K, which means that the binary mixture (HFC-32 + HFC-125) exhibits near-azeotropic behavior in other compositions.

Comparsion with Literature Data

Many vapor−liquid equilibrium data of the system (HFC-32 + HFC-125) have been reported in the literatures.[6−20] In the paper, the vapor−liquid equilibrium data (*PTxy*) are chosen to conduct the comparison for the composition and pressure.

A comparison with literature data has been performed by means of the PR + LCVM model. The data of Kato et al.[14] show big deviations from the model for both vapor composition (up to 0.035) and pressure (up to 5.8 %). A better agreement for the vapor composition is reached from the data of Nagel et al.,[6] Jung et al.,[8] and Lee et al.;[12] their average vapor composition deviations are 0.0006, 0.0062, and 0.0078, respectively. The vapor composition data of Higashi[11] show an acceptable agreement (within 0.015). Data from Benmansour et al.[17] have no vapor composition, so no comparison was done. For pressure, the data from Nagel et al.[6] and Lee et al.[12] have a better agreement, and average deviations are 0.21 % and 1.07 %, respectively; the data from Jung et al.,[8] Higashi,[11] and Benmansour et al.[17] have an acceptable agreement with the average deviations being 2.65 %, 2.27 %, and 2.96 %, respectively, and the data in this work have a better agreement with the average deviation of pressure being 0.99 %. The plot of the deviations between the experimental and the calculated data are given in Figures 5−6.

Conclusions

Isothermal vapor−liquid equilibrium data for the binary mixture (HFC-32 + HFC-125) were reported. The range of temperatures were explored from (265.15 to 303.15) K at pressures up to 2 MPa. The experimental data were correlated with the PR equation of state combined with the LCVM mixing rule and NRTL excess free energy model. Almost all of the calculated values with the model gave a good agreement with the experimental data of this work, and average deviations of vapor composition and pressure are 0.007, 0.99 %, respectively; the parameters in NRTL model were given at different temperatures.

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