Vapor-Liquid Equilibria for the 1,1-Difluoroethane (HFC-152a) + Propane (R-290) System

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Isothermal vapor-liquid equilibrium data of the binary system 1,1-difluoroethane (HFC-152a) + propane (R-290) were measured in the temperature range from 273.15 K to 313.15 K at 10 K intervals and in the composition range from 0.2 to 0.8 mole fraction of propane. The temperature, pressure, and compositions of the liquid and vapor phases were measured with a circulation-type apparatus. The experimental data were correlated with the Carnahan–Starling–De Santis, Peng–Robinson, and Redlich–Kwong–Soave equations of state. Azeotropic behavior was revealed near 0.73 mole fraction of propane. The vapor pressure of the azeotropic system was compared with those of chlorodifluoroethane (HCFC-22) and the zeotropic mixture of difluoroethane (HFC-32) + pentafluoroethane (HFC-125) (50–50) (alternative refrigerant blend R-410A), the zeotropic mixture of HFC-32 + HFC-125 + 1,1,1,2-tetrafluoroethane (HFC-134a) (23–25–52) (alternative refrigerant blend R-407C), dichlorodifluoromethane (HCFC-22. This azeotropic system has good potential to replace the use of HCFC-22 as a refrigerant.

Introduction

The regulation of CFCs and HCFCs has led to investigations of pure HFCs and HCs and mixtures of these fluids as alternative refrigerants. Particularly, hydrocarbons have been considered as alternative refrigerants because of their zero ozone depletion potential, low toxicity, and chemical stability and because the flammability of hydrocarbons can be reduced by mixing with nonflammable alternative refrigerants. The possibility of using these mixtures as alternative refrigerants should be examined on the basis of accurate and reliable data of their thermophysical properties. Vapor—liquid equilibrium data are one of the most important properties for evaluating the thermophysical properties of these mixtures and for determining their optimal composition.

In this study, isothermal vapor-liquid equilibrium data of the binary system 1,1-difluoroethane (HFC-152a) + propane (R-290) were measured in the temperature range from 273.15 to 313.15 K and correlated with the Carnahan-Starling-DeSantis (CSD),¹ Peng-Robinson (PR),² and Soave-Redlich-Kwong (SRK)³ equations of state. On the basis of the measured vapor-liquid equilibrium data, azeotropic behavior of the binary system and the possibility of replacing the use of HCFC-22 with the azeotropic system will be discussed.

Experimental Section

Chemicals. The sample of pure 1,1-difluoroethane (HFC-152a) was obtained from SDCh Co. Ltd., and propane (R-290) was obtained from MG Industries. The manufacturers stated that the purity of the chemicals was 99.9 %. The chemicals were used without further purification. Typical properties of 1,1-difluoroethane (HFC-152a) and propane (R-290) are listed in Table 1.

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Table 1.	Thermophysical	Properties	of HFC-152a	(1) and Propane
$(2)^9$				

	R-290 (propane)	HFC-152a (1,1-difluoroethane)
chemical formula	C ₃ H ₈	$C_2H_4F_2$
М	44.10	66.05
$T_{\rm b}/{ m K}$	231.06	249.13
$T_{\rm c}/{\rm K}$	369.85	386.41
$P_{\rm c}/{\rm kPa}$	4248	4517
ω	0.1524	0.2752

Experimental Apparatus and Procedures. The measurement of the vapor-liquid equilibrium was conducted in a circulationtype apparatus. This apparatus is shown schematically in Figure 1. It is described only briefly here because the apparatus and procedures have already been presented in more detail.^{4–6} The apparatus consisted of an equilibrium unit, a sampling and analysis unit, and a supply unit. The equilibrium cell was made of type-316 stainless steel with an inner volume of about 128 cm³. The temperature of the isothermal bath and equilibrium cell was maintained at a set point within \pm 0.02 K. A magnetic pump circulated the vapor phase into the liquid phase to ensure proper mixing, thus enabling equilibrium to be quickly reached. The bath and cell temperatures were measured with a 100 Ω platinum resistance sensor immersed in the cell and a precision thermometer (Yokogawa, 7563) with a precision of 0.01 K. The sensor and thermometer were calibrated on the ITS-90. The uncertainty in the temperature measurements was estimated to be \pm 0.02 K. The pressure was measured with a pressure transducer (Druck PDCR 922) with a precision of 0.1 kPa. The pressure was read when the cell pressure was constant within 1 kPa to 2 kPa. The pressure transducer was calibrated against a quartz crystal pressure transducer after each series of experiments. The uncertainty of the pressure measurement was estimated to be 0.4 %.

After equilibrium was reached, the vapor and liquid samples for analyzing compositions were trapped in a space between

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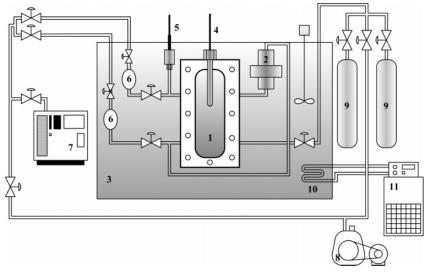


Figure 1. Schematic diagram of the vapor-liquid equilibrium apparatus for refrigerant mixtures: 1, equilibrium cell; 2, magnetic pump; 3, isothermal bath; 4, platinum resistance thermometer; 5, pressure transducer; 6, sample cylinder; 7, gas chromatograph; 8, vacuum pump; 9, refrigerant vessel; 10, heat exchanger; 11, circulator.

Table 2. Vapor-Liquid Equilibrium Data for the HFC-152a (1) + Propane (2) System

T/K	P/kPa	<i>x</i> ₂	<i>y</i> ₂
273.15	265.8	0.0000	0.0000
	319.1	0.0728	0.1449
	412.6	0.2331	0.4219
	504.9	0.5771	0.6408
	514.2	0.5851	0.6661
	507.2	0.7011	0.7215
	506.5	0.7565	0.7568
	512.9	0.8178	0.8053
	472.5	1.0000	1.0000
283.15	372.8	0.0000	0.0000
	437.4	0.0848	0.1652
	553.3	0.2452	0.3967
	669.9	0.5600	0.6286
	674.5	0.6176	0.6479
	677.9	0.7061	0.7202
	676.1	0.7592	0.7547
	671.3	0.8343	0.8087
	633.9	1.0000	1.0000
293.15	513.3	0.0000	0.0000
	587.6	0.0829	0.1629
	726.9	0.2333	0.3734
	876.3	0.5542	0.6265
	902.7	0.6254	0.6641
	901.4	0.6979	0.7129
	894.2	0.7542	0.7488
	904.2	0.8312	0.7982
	832.94	1.0000	1.0000
303.15	689.9	0.0000	0.0000
	775.1	0.0624	0.1449
	940.7	0.2326	0.3489
	1146.5	0.5473	0.6135
	1166.7	0.6436	0.6562
	1161.0	0.7054	0.7069
	1154.4	0.7652	0.7484
	1162.3	0.8297	0.7974
	1077	1.0000	1.0000
313.15	909.4	0.0000	0.0000
	1005.3	0.0656	0.1238
	1196.4	0.2226	0.3138
	1467.8	0.5844	0.6008
	1501.8	0.6102	0.6447
	1479.5	0.7065	0.7041
	1464.1	0.7633	0.7448
	1479.4	0.8282	0.8035
	1370	1.0000	1.0000

two valves and introduced into evacuated sample cylinders. The compositions of the vapor and liquid samples were determined

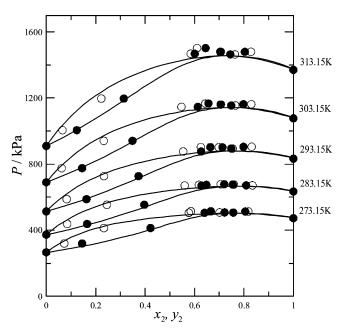


Figure 2. Vapor-liquid equilibrium data for the HFC-152a (1) + propane (2) system: \bigcirc , x_2 ; \bigcirc , y_2 , experimental data; -, Carnahan-Starling-De Santis (CSD) equation of state.

Table 3. Coefficients of the CSD Equation of State Used for Data Correlation $(REFPROP)^{8}$

,	
HFC-152a	propane
3.1986257·10 ³	$2.988277 \cdot 10^{3}$
$2.9613439 \cdot 10^{-3}$	$-2.629019 \cdot 10^{-3}$
3.2189735 • 10 ⁻⁷	$-1.097062 \cdot 10^{-6}$
$1.3326433 \cdot 10^{-1}$	$1.429625 \cdot 10^{-1}$
$-2.0363288 \cdot 10^{-4}$	$-1.7651912 \cdot 10^{-4}$
$7.7725097 \cdot 10^{-8}$	$-5.785137 \cdot 10^{-8}$
	$\begin{array}{c} 3.1986257{\cdot}10^3\\ 2.9613439{\cdot}10^{-3}\\ 3.2189735{\cdot}10^{-7}\\ 1.3326433{\cdot}10^{-1}\\ -2.0363288{\cdot}10^{-4} \end{array}$

using a gas chromatograph (HP 5890II) equipped with a Porapak-Q column (1.83 m long; 3.18 mm diameter; mesh range, 80/100) and a thermal conductivity detector. The compositions at one condition were measured at least three times to ensure accuracy and repeatability. The maximum variance of mole fraction among three measurements was < 0.1 %. The gas chromatograph was calibrated with the pure components of known purity and with mixtures of known composition that

had been prepared gravimetrically. The uncertainty of the mole fraction was estimated to be \pm 0.3 %.

Results and Discussion

Vapor-liquid equilibrium data for the binary system of HFC-152a + propane were measured in a temperature range from 273.15 K to 313.15 K at 10 K intervals. These data are presented in Table 2 and in Figure 2. The experimental data were correlated with the Carnahan-Starling-De Santis (CSD),¹ Peng-Robinson (PR),² and Redlich-Kwong-Soave (SRK)³ equations of state using the van der Waals mixing rules.⁷ The CSD equation of state is given by

$$P = \frac{RT}{v} \left[\frac{1 + y + y^2 - y^3}{(1 - y^3)} \right] - \frac{a}{v(v + b)}$$
(1)

where

$$y = \frac{b}{4v} \tag{2}$$

P is the pressure; v is the molar volume; *T* is the temperature; and *R* is the universal gas constant.

The van der Waals mixing rules⁷ were used for three equations of state

$$a = \sum_{i} \sum_{j} x_{i} y_{i} a_{ij}$$
(3)

$$b = \sum_{i} \sum_{j} \mathbf{x}_{i} \mathbf{y}_{i} b_{ij} \tag{4}$$

where x_i is the mole fraction of the *i*th component and

$$a_{ii} = (1 - k_{ii})(a_{ii}a_{ji})^{1/2}$$
(5)

$$b_{ij} = \frac{(b_{ii}^{1/3} + b_{jj}^{1/3})^3}{8} \tag{6}$$

where k_{ij} is the binary interaction parameter. The critical properties and acentric factors are given in Table 1. The *a* and *b* of the CSD equation of state were cited from REFPROP 5.0.⁸ They are listed in Table 3.

The binary interaction parameter k_{ij} was determined by minimizing the following objective function

$$obj = \sum_{i=1}^{N} \left(\frac{P_{calcd,i} - P_{exptl,i}}{P_{exptl,i}} \right)$$
(7)

where N is the number of experimental points; obj is the objective function; and the subscripts calcd and exptl denote calculated and experimental results, respectively.

The binary interaction parameters were determined at (273.15, 283.15, 293.15, 303.15, and 313.15) K. They are presented in Figure 3. The binary interaction parameters for SRK and PR equations of state decrease slightly as the temperature increases, but the CSD equation of state increases slightly as the temperature increases. The binary interaction parameters for three equations of state were correlated with a linear function of temperature as shown in Table 4. However, the temperature effect is almost negligible.

The relative deviations between the experimentally measured data for the pressure and vapor-phase mole fraction of propane and the calculated results from equations of state are given in Table 5. The average deviation between the measured pressures

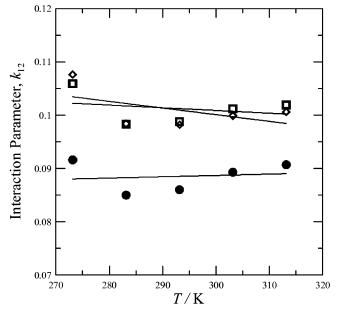


Figure 3. Binary interaction parameters in Carnahan–Starling–De Santis, Peng–Robinsion (PR), and Redlich–Kwong–Soave (RKS) equations of state for the HFC-152a (1) + propane (2) system at five temperature conditions: \bullet , CSD; \Box , PR; \diamond , RKS.

 Table 4. Binary Interaction Parameters in CSD, PR, and SRK

 Equations of State for the HFC-152a (1) + Propane (2) System^a

equation of state	a_1	a_2
CSD	0.08113755	0.000023
PR	0.11617065	-0.000051
SRK	0.13758375	-0.000125

 $a_{k_{12}} = a_1 + a_2 T (T/K).$

Table 5. Binary Interaction Parameters for the HFC-152a (1) +Propane (2) System and Deviation of Pressure and Vapor MoleFraction of Propane from Equations of State

T/K	k_{12}	RMS P/%	RMS y ₂ /%			
Carnahan-Starling-De Santis (CSD)						
273.15	0.0916	2.6	3.9			
283.15	0.0850	2.5	3.2			
293.15	0.0860	2.8	2.4			
303.15	0.0893	2.1	1.9			
313.15	0.0907	2.3	2.3			
average	0.0899	2.5	2.7			
	Peng-I	Robinson (PR)				
273.15	0.1059	2.1	3.9			
283.15	0.0983	2.1	3.3			
293.15	0.0988	2.7	3.1			
303.15	0.1012	2.1	2.7			
313.15	0.1019	2.4	3.1			
average	0.1012	2.3	3.2			
Relich-Kwong-Soave (SRK)						
273.15	0.1076	2.1	3.9			
283.15	0.0984	2.1	3.2			
293.15	0.0982	2.7	2.9			
303.15	0.0999	2.2	2.6			
313.15	0.1006	2.5	2.9			
average	0.1009	2.3	3.1			

and calculated results from the CSD equation of state is about 2.5 %; that from the PR equation of state is 2.3 %; and that from the SRK equation of state is about 2.3 %. It can be concluded that, on the basis of these results, the CSD equation of state correlated the experimental data as well as the PR and SRK equations of state.

Figures 4 and 5 show comparisons of the relative pressure and vapor-phase mole fraction of propane between the measured data and the calculated results from the CSD equation of state.

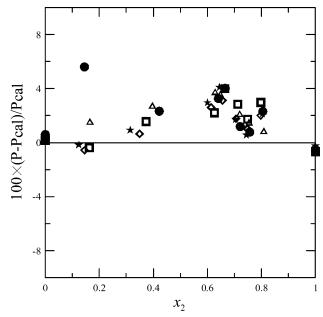


Figure 4. Deviation of the bubble point pressure for the HFC-152a (1) + propane (2) system between the measured data and the calculated results from the Carnahan–Starling–De Santis equation of state. Percent deviation = $100 \cdot (P - P_{calcd})/P_{calcd}$: •, 273.15 K; \triangle , 283.15 K; \square , 293.15 K; \diamondsuit , 303.15 K; \star , 313.15 K.

Table 6. Coefficients of Equilibrium Pressure Correlation for the System HFC-152a (1) + Propane $(2)^a$

<i>T</i> /K	а	b	С	d	е
273.15	267.726	265.117	406.752	-312.36	-156.251
283.15	374.58	115.061	1761.624	-2466.985	848.015
293.15	514.31	182.57	1876.614	-2385.018	643.752
303.15	692.422	178.548	2887.077	-4130.086	1447.88
313.15	913.741	241.416	3941.23	-6144.085	2416.752

 $^{a}P = a + bx + cx^{2} + dx^{3} + ex^{4}.$

 Table 7. Composition and Pressures of the Azeotrope (HFC-152a

 (1) + Propane (2)) at Each Temperature

T/K		273.15	283.15	293.15	303.15	313.15
experiment	χ_{exptl}	0.77	0.75	0.76	0.74	0.73
<u>^</u>	P _{exptl} /MPa	0.5141	0.679	0.905	1.166	1.487
CSD	X _{az,calcd}	0.763	0.775	0.764	0.745	0.733
	Paz,calcd/MPa	0.5027	0.6669	0.8793	1.1428	1.456
	Δx	0.007	-0.025	-0.004	-0.005	-0.003
	$\Delta P/MPa$	0.011	0.012	0.026	0.023	0.031
PR	Xaz,calcd	0.757	0.767	0.755	0.74	0.728
	Paz,calcd/MPa	0.5053	0.6699	0.8833	1.1476	1.4629
	Δx	0.013	-0.017	0.005	0	0.002
	$\Delta P/MPa$	0.009	0.009	0.022	0.018	0.024
SRK	Xaz,calcd	0.764	0.779	0.769	0.755	0.743
	Paz,calcd/MPa	0.5061	0.671	0.8848	1.1485	1.4636
	Δx	0.006	-0.029	-0.009	-0.015	-0.013
	$\Delta P/MPa$	0.008	0.008	0.02	0.018	0.023

As was shown in Figure 2, azeotropic behavior was found at a mole fraction near 0.76 mole fraction of propane. An azeotrope behaves like a pure fluid and then its components cannot be separated by simple distillation. Mathematically, an azeotrope corresponds to an extremum of temperature or pressure, and at an azeotropic composition the mole fractions of liquid and vapor phases are equal to each other.

This can be expressed as follows¹⁰

$$\left(\frac{\partial P}{\partial x_i}\right)_T = 0 \quad \text{for } i = 1 \text{ to } N - 1$$
 (8)

$$x_i = y_i$$
 for $i = 1$ to N and at constant temperature (9)

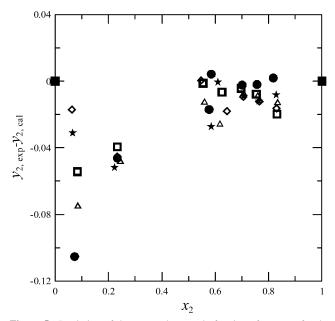


Figure 5. Deviation of the vapor-phase mole fraction of propane for the HFC-152a (1) + propane (2) system between the measured data and the calculated results from the Carnahan–Starling–De Santis equation of state. Deviation = $y_{exptl} - y_{calcd}$: •, 273.15 K; \triangle , 283.15 K; \Box , 293.15 K; \diamond , 303.15 K; \star , 313.15 K.

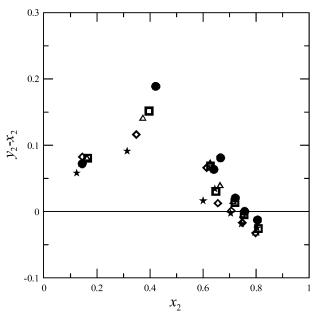


Figure 6. Difference between vapor composition and liquid composition $(y_2 - x_2)$ against liquid composition x_2 for the HFC-152a (1) + propane (2) system: •, 273.15 K; \Box , 283.15 K; \triangle , 293.15 K; \diamondsuit , 303.15 K; \bigstar , 313.15 K.

To obtain experimental azeotropic points, experimental equilibrium pressures were fitted with a polynomial of liquid mole fraction of propane as follows.

$$P = a + bx + cx^2 + dx^3 + ex^4$$
(10)

Coefficients of eq 10 were presented in Table 6. Experimental azeotropic points were calculated from eqs 8-10 over the temperature range from 273.15 K to 313.15 K (Figure 6). The experimental azeotropic points were found to be in the range from 0.73 to 0.76 mole fraction of propane. Azeotropic points

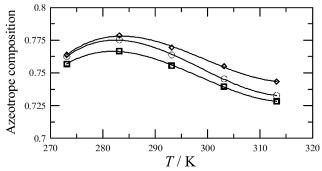


Figure 7. Calculated azeotropic mole fraction of propane as a function of temperature: \bigcirc , CSD; \square , PR; \diamondsuit , SRK.

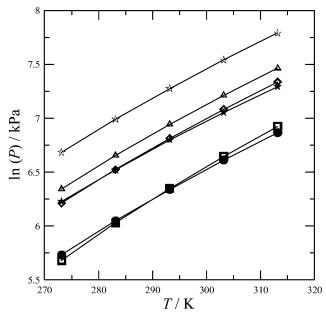


Figure 8. Vapor pressure for HCFC-12,⁹ HFC-134a,⁹ HCFC-22,⁹ R-407C,⁹ and R-410A⁹ and calculated vapor pressure of the azeotropic mixture HFC-152a (1) + propane (2): •, HCFC-12; \Box , HFC-134a; \diamond , HCFC-22; \triangle , R-407C; \Leftrightarrow , R-410A; \bigstar , azeotropic mixture of the HFC-152a (1) + propane (2) system.

Table 8. Coefficients of the Composition Polynomial and the VaporPressure Correlations of the Azeotrope (HFC-152a (1) + Propane(2))

coefficients	CSD	PR	SRK
	$x_{\rm az} = a + bT +$	$-cT^2+dT^3$ (T/K)	
а	$-6.5247 \cdot 10$	$-5.5419 \cdot 10$	$-5.8868 \cdot 10$
b	$6.6695 \cdot 10^{-1}$	$5.6815 \cdot 10^{-1}$	$6.0093 \cdot 10^{-1}$
с	$-2.2398 \cdot 10^{-3}$	$-1.9098 \cdot 10^{-3}$	$-2.0131 \cdot 10^{-3}$
d	$2.5 \cdot 10^{-6}$	$2.1333 \cdot 10^{-6}$	$2.2417 \cdot 10^{-6}$
	$\ln P_{\rm az} (P_{\rm az}/{\rm MPa}) =$	A + BT + C/T (T/H)	K)
Α	1.0538.10	1.03.10	1.0521.10
В	$6.8769 \cdot 10^{-3}$	$7.2852 \cdot 10^{-3}$	$6.895 \cdot 10^{-3}$
С	$-1.6929 \cdot 10^{3}$	$-1.657 \cdot 10^{3}$	$-1.6878 \cdot 10^{3}$

were also numerically calculated using CSD, PR, and SRK equations of state. The calculated azeotropic points were found to be in the range from 0.73 to 0.78 mole fraction of propane. The experimental and calculated azeotropic points were compared and presented in Table 7. In Figure 7, the calculated azeotropic compositions are fitted into a polynomial expression of temperature.

 $x_{az} = a + bT + cT^2 + dT^3$

temperature

$$\ln P_{az} = A + BT + C/T \tag{12}$$

Table 8 presents the coefficients of eqs 11 and 12. In Figure 8, the vapor pressures of chlorodifluoromethane (HCFC-22),⁹ the azeotropic mixture of 1,1-difluoroethane (HFC-152a) + propane, the zeotropic mixture of difluoroethane (HFC-32) + pentafluoroethane (HFC-125) $(50-50)^9$ (alternative refrigerant blend R-410A), the zeotropic mixture of HFC-32 + HFC-125 + 1,1,1,2-tetrafluoroethane (HFC-134a)⁹ (23-25-52) (alternative refrigerant blend R-407C), dichlorodifluoromethane (HCFC-12),⁹ and HFC-134a⁹ were compared. The calculated vapor pressure of the azeotropic system HFC-152a + propane was lower than that of R-410A and R-407C and very close to that of HCFC-22 from below. In terms of vapor pressure, the azeotropic system HFC-152a + propane may therefore be a suitable alternative refrigerant to replace HCFC-22.

Conclusions

Isothermal vapor-liquid equilibrium data for the binary system of HFC-152a + propane were measured, and 45 data points along five isotherms in the temperature range from 273.15 K to 313.15 K were obtained, including the vapor pressure data for pure components. The experimental data were correlated with the CSD, PR, and SRK equations of state. The binary interaction parameters for the CSD, PR, and SRK equations of state were calculated for five isotherms for the HFC-152a + propane system.

The present data were in good agreement with the calculated results from the three equations of state, and the deviations were less than 2.5%.

It was found that this mixture exhibited azeotropic behavior at a mole fraction near 0.76 for propane. It was found that the vapor pressures of the azeotropic system HFC-152a + propane were very close to that of HCFC-22 from below and lower than those of R-407C and R-410A. In terms of vapor pressure, this azeotropic mixture is better than R-407C and R-410A and will be a potential refrigerant to replace the use of HCFC-22.

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