

Critical Parameters and *PVT* Properties for R-404A

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The critical parameters of R-404A, a ternary mixture of 44 mass % pentafluoroethane (R-125), 52 mass % 1,1,1-trifluoroethane (R-143a), and 4 mass % 1,1,1,2-tetrafluoroethane (R-134a), were measured by visual observation of the meniscus in an optical cell. The critical pressure, density, and temperature were determined to be (3.726 ± 0.002) MPa, (490 ± 3) kg·m⁻³, and (345.15 ± 0.01) K, respectively. Forty single-phase *PVT* properties have been measured with a constant-volume method along three isochores in a range of densities from 487 kg·m⁻³ to 493 kg·m⁻³, temperatures from 345 K to 393 K, and pressures from 3.7 MPa to 7.8 MPa. One hundred and nineteen densities of R-404A have been measured with a vibrating-tube densitometer over a density range from 47 kg·m⁻³ to 1247 kg·m⁻³ along eight isotherms between 263 K and 403 K and at pressures between 1.5 MPa and 15 MPa. An 18-term modified Benedict–Webb–Rubin equation of state for R-404A was developed using the *PVT* property data.

Introduction

R-502, an azeotropic mixture of 48.8 mass % chlorodifluoromethane (R-22) and 51.2 mass % chloropentafluoroethane (R-115), has been widely used as a refrigerant in low-temperature refrigerators. This substance has not been supplied since 1996 in accordance with the Montreal Protocol. R-404A, a ternary mixture of 44 mass % pentafluoroethane (R-125), 52 mass % 1,1,1-trifluoroethane (R-143a), and 4 mass % 1,1,1,2-tetrafluoroethane (R-134a), is one of the candidates to replace to R-502.

Bouchot and Richon (1995) reported the critical parameters for R-404A. They also reported the *PVT* and saturation properties at pressures from 0.1 MPa to 19 MPa, densities from 4 kg·m⁻³ to 1279 kg·m⁻³, and temperatures from 253 K to 333 K.

We measured the critical parameters for R-404A by observing the disappearance of the vapor–liquid interface. We also measured its *PVT* properties with a constant-volume method and a vibrating-tube densitometer. On the basis of the *PVT* properties of Bouchot and Richon (1995) and this work, we developed an 18-term modified Benedict–Webb–Rubin equation of state for R-404A.

Experimental Apparatus and Procedure

Sample. The purity values of nitrogen, R-125, R-134a, and R-143a used were 99.995 mass %, 99.99 mass %, 99.99 mass %, and 99.99 mass %, respectively. R-125, R-134a, and R-143a were obtained from our company. The samples were used without further purification.

For making the ternary mixtures of 44 mass % R-125, 52 mass % R-143a, and 4 mass % R-134a with prescribed densities, each component was charged to a separate stainless steel 304 cylindrical vessel to the required mass. The masses were measured with an analytical balance. Whenever an overestimation of the mass of a sample was made, portions of the sample were discharged to the necessary extent. The combined standard uncertainty of the mass measurements for each component was calculated to be 2 mg from the accuracy of the analytical balance, ± 1 mg, and the fluctuation, ± 2 mg. The combined standard

uncertainty of the mass measurement for the mixture was calculated to be 3 mg from the uncertainties for the components. The uncertainty of the mass fraction, δw_i , caused by that of the mass measurement, δm , is calculated from the following equation:

$$(\delta w_i)^2 = \left(\frac{\partial w_i}{\partial m_1}\right)^2 (\delta m_1)^2 + \left(\frac{\partial w_i}{\partial m_2}\right)^2 (\delta m_2)^2 + \left(\frac{\partial w_i}{\partial m_3}\right)^2 (\delta m_3)^2 \quad (1)$$

Subscripts 1, 2, and 3 indicate R-125, R-143a, and R-134a, respectively.

Constant Volume Apparatus with an Optical Cell.

The experimental apparatus and procedures have been described previously, and measurements were reported on the critical parameters and vapor pressures for R-134a and R-143a (Fujiwara et al., 1998). The main portion of the apparatus, composed of an optical cell, a supplying vessel, and a pressure transducer, is located in a thermostatic bath. The temperature measurements are made with a platinum resistance thermometer mounted in the vicinity of the optical cell. The inner volume of the optical cell, including tubes and valves as well as the supplying vessel, is about 95 cm³ at room temperature and atmospheric pressure. It was calculated using equations we have developed considering the thermal expansion and the pressure deformation. These equations were calibrated by charging a known mass of nitrogen and measuring temperatures and pressures ranging from 263 K to 393 K and from 0.5 MPa to 10 MPa. The combined standard uncertainties of the measurements are shown in Table 1. The sources and their values of the uncertainties in inner volume and temperature were shown in our previous paper (Fujiwara et al., 1998). The uncertainty of density was calculated from the uncertainties of mass and volume. The uncertainty of mass fraction was calculated from the uncertainty calculated from eq 1 and the difference between the sample composition and the composition of R-404A. The sources of the uncertainty of pressure include the uncertainties of the temperature and composition measurements. The uncertainty of temperature was empirically estimated to be equivalent to ± 0.3 kPa in pres-

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Table 1. Uncertainty Analysis in Measurements with Constant Volume Apparatus

(a)	pressure	
	accuracy of transducer	2 kPa
	uncertainty in temperature measurements	0.3 kPa
	uncertainty in composition measurements	1 kPa
	total	2 kPa
(b)	inner volume	0.3 cm ³
(c)	density	2 kg·m ⁻³
(d)	temperature	3 mK
(e)	composition of R-125	
	uncertainty in mass measurements	0.004 mass %
	difference with R-404A	0.002 mass %
	total	0.004 mass %
(f)	composition of R-143a	
	uncertainty in mass measurements	0.004 mass %
	difference with R-404A	0.015 mass %
	total	0.02 mass %
(g)	composition of R-134a	
	uncertainty in mass measurements	0.004 mass %
	difference with R-404A	0.013 mass %
	total	0.01 mass %

sure; the uncertainty in composition was estimated to be equivalent to ± 1 kPa in pressure with a database written by Huber et al. (1996). In this database pressures of R-404A were calculated with the Carnahan–Starling–DeSantis equation of state and the interaction parameters.

Vibrating-Tube Densitometer. The experimental apparatus and procedures were described in detail in our previous publication (Nakamura et al., 1997). Using the apparatus, we have measured the densities for R-143a. A stainless steel vibrating U-tube densitometer (about 1 cm³) is connected to a quartz pressure transducer, a metallic bellows variable-volume vessel, a circulating pump, and a hand pressure controller. The U-tube is housed in a brass housing enclosed in a steel heat exchanger. Silicone oil is circulated through the heat exchanger from an external temperature-controlled bath. The temperature measurements were made with a platinum resistance thermometer installed near the outlet of the heat exchanger. Each component charged in the stainless steel 304 cylindrical vessel was transferred successively into the bellows vessel, which had been evacuated in advance to around 0.4 mPa and cooled by liquid nitrogen. The circulating pump was used in order to obtain a homogeneous composition. The densities of the samples were calculated from an equation where the density is given as a function of frequency, pressure, and temperature. Coefficients of this equation

were determined on the basis of the measurements for nitrogen and R-134a over a temperature range of 263 K to 403 K and at pressures from 0.5 MPa to 15 MPa. The combined standard uncertainties of the measurement are shown in Table 2. The sources and their values of the uncertainties in pressure, temperature, frequency, and the density equation were shown in our previous paper (Nakamura et al., 1997). The uncertainty of mass fraction was calculated from the value calculated from eq 1 and a difference between the sample composition and the composition of R-404A. The uncertainty of the measurements in pressure, temperature, and frequency were estimated from the density equation to be equivalent to ± 0.0007 kg·m⁻³, ± 0.02 kg·m⁻³, and ± 0.06 kg·m⁻³ in density. The density equivalent of the uncertainty of the composition measurement was estimated to be ± 0.1 kg·m⁻³ with the database written by Huber et al. (1996) using the same equation of state and mixing parameters as mentioned in the previous section.

Results

Critical Parameters. PVT properties for R-404A in the two-phase region were measured by visual observation of the meniscus disappearance in the optical cell at three densities: 487 kg·m⁻³, 490 kg·m⁻³, and 493 kg·m⁻³. The numerical values of the results are asterisked in Table 3. At all the densities, the level at which the meniscus disappeared was near the center of the cell and the critical opalescence was observed. At 487 kg·m⁻³ the critical opalescence in the liquid phase was more intense than that in the vapor phase; at 490 kg·m⁻³ the liquid phase was as intense as the vapor phase; at 493 kg·m⁻³ the vapor phase was more intense than the liquid phase.

In consideration of the meniscus behavior with respect to the three densities mentioned above, the critical density, ρ_c , of R-404A was determined to be

$$\rho_c = (490 \pm 3) \text{ kg}\cdot\text{m}^{-3}$$

We considered that the critical density should be between 487 kg·m⁻³ and 493 kg·m⁻³ and estimated the uncertainty of the critical density to be within ± 3 kg·m⁻³. The critical temperature, T_c , and pressure, P_c , of R-404A were determined using the values at the density of 490 kg·m⁻³ as follows:

Table 2. Uncertainty Analysis in Measurements with Densitometer

(a)	pressure	4 kPa
(b)	temperature	7 mK
(c)	frequency	0.001 Hz
(d)	density	
	density range in kg·m ⁻³	47 < ρ ≤ 194, 194 < ρ < 1247
	uncertainty in pressure measurements	0.0007 kg·m ⁻³
	uncertainty in temperature measurements	0.02 kg·m ⁻³
	uncertainty in frequency measurements	0.06 kg·m ⁻³
	uncertainty in composition measurements	0.1 kg·m ⁻³
	uncertainty of density equation	0.5 kg·m ⁻³ , 3 kg·m ⁻³
	total	0.5 kg·m ⁻³ , 3 kg·m ⁻³
(e)	composition of R-125	
	uncertainty in mass measurements	0.0011 mass %
	difference to R-404A	0.033 mass %
	total	0.03 mass %
(f)	composition of R-143a	
	uncertainty in mass measurements	0.0015 mass %
	different to R-404A	0.032 mass %
	total	0.03 mass %
(g)	composition of R-134a	
	uncertainty in mass measurements	0.0001 mass %
	difference to R-404A	0.001 mass %
	total	0.001 mass %

Table 3. Experimental PVT Properties with Constant Volume Apparatus

$w_1 = 44.998 \text{ mass } \%$ $w_2 = 52.003 \text{ mass } \%$ $w_3 = 3.999 \text{ mass } \%$			$w_1 = 43.998 \text{ mass } \%$ $w_2 = 52.015 \text{ mass } \%$ $w_3 = 3.987 \text{ mass } \%$			$w_1 = 43.999 \text{ mass } \%$ $w_2 = 52.009 \text{ mass } \%$ $w_3 = 3.992 \text{ mass } \%$		
P/MPa	$\rho/\text{kg}\cdot\text{m}^{-3}$	T/K	P/MPa	$\rho/\text{kg}\cdot\text{m}^{-3}$	T/K	P/MPa	$\rho/\text{kg}\cdot\text{m}^{-3}$	T/K
3.7237 ^a	487.0 ^a	345.159 ^a	3.7261 ^a	490.0 ^a	345.150 ^a	3.7235 ^a	492.9 ^a	345.160 ^a
3.7245	487.0	345.169	3.7267	490.0	345.159	3.7240	492.9	345.169
3.7391	487.0	345.350	3.7276	490.0	345.170	3.7249	492.9	345.180
3.7548	487.0	345.549	3.7284	490.0	345.179	3.9661	492.8	348.149
3.7712	487.0	345.752	3.8061	490.0	346.150	4.3813	492.7	353.150
3.9649	486.9	348.149	3.9685	489.9	348.149	4.8014	492.6	358.149
4.3745	486.8	353.150	4.3808	489.8	353.150	5.2244	492.5	363.149
4.7881	486.7	358.150	4.7976	489.7	358.151	5.6500	492.4	368.149
5.2046	486.5	363.150	5.2168	489.6	363.149	6.0770	492.2	373.150
5.6229	486.4	368.151	5.6387	489.5	368.150	6.5051	492.1	378.149
6.0428	486.3	373.150	6.0616	489.4	373.148	6.9346	492.0	383.150
6.4634	486.2	378.149	6.4859	489.2	378.150	7.3642	491.9	388.149
6.8853	486.1	383.150	6.9113	489.1	383.151	7.7943	491.8	393.150
7.3073	486.0	388.150	7.3368	489.0	388.150			
7.7297	485.8	393.150	7.7628	488.9	393.150			

^a Indicates the saturation properties.

$$T_c = (345.15 \pm 0.01) \text{ K}$$

$$P_c = (3.726 \pm 0.002) \text{ MPa}$$

The meniscus behavior was observed at an interval of 10 mK, and so we estimated the uncertainty of the critical temperature to be within ± 10 mK. Considering the uncertainties of pressure measurement and the critical temperature, we estimated the uncertainty of the critical pressure to be ± 2 kPa.

Single-Phase PVT Properties. Forty single-phase PVT properties of R-404A listed in Table 3 were measured with the constant volume apparatus along three isochores in a range of densities from $487 \text{ kg}\cdot\text{m}^{-3}$ to $493 \text{ kg}\cdot\text{m}^{-3}$, temperatures from 345 K to 393 K, and pressures from 3.7 MPa to 7.8 MPa. One hundred and nineteen PVT properties of R-404A listed in Table 4 were measured with the densitometer in a range of pressures from 1.5 MPa to 15 MPa, densities from $47 \text{ kg}\cdot\text{m}^{-3}$ to $1247 \text{ kg}\cdot\text{m}^{-3}$, and temperatures from 263 K to 403 K.

Discussion

Critical Parameters. A comparison of the critical parameters for R-404A is shown in Table 5. Bouchot and Richon (1995) correlated their saturation PVT data by considering R-404A as a pseudo pure compound and gave the critical temperature and density as parameters of the equations. The critical density and temperature of this work are $2.6 \text{ kg}\cdot\text{m}^{-3}$ and 0.45 K greater than those of Bouchot and Richon (1995), respectively. Regarding the critical density, the difference between Bouchot and Richon (1995) and this work is within the uncertainty of our critical density; regarding the critical temperature, the difference is 45 times as great as the uncertainty of our critical temperature. Our saturation densities and temperatures are consistent with those of Bouchot and Richon (1995) as shown in Figure 1; our saturation pressures are consistent with those of Bouchot and Richon (1995) as shown in Figure 2.

PVT Properties. The pressure glides between the saturated liquid and saturated vapor states are small as shown in Figure 2. We have developed an equation of state for R-404A by correlating measurements for the mixture as a pseudo pure fluid. This equation of state is used primarily for the single-phase PVT properties. We have considered only data at the composition of R-404A.

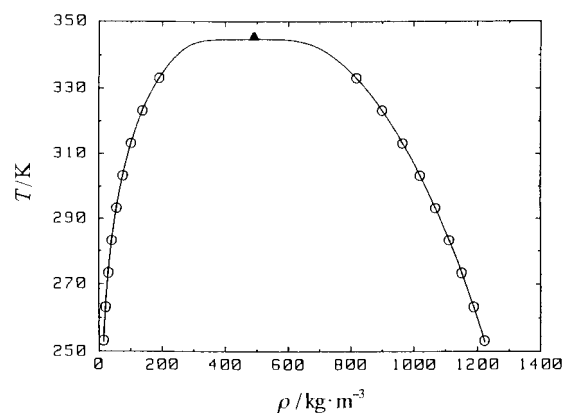


Figure 1. Vapor-liquid coexistence curve of R-404A: (○) Bouchot and Richon (1995); (△) this work; (—) calculated values from equations of Bouchot and Richon (1995).

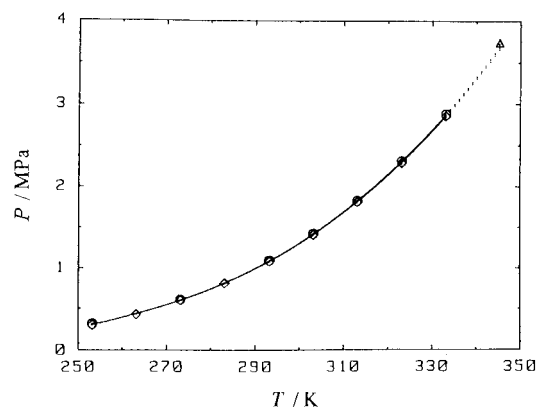


Figure 2. Saturation pressures of R-404A: (○) bubble-point pressure of Bouchot and Richon (1995); (◇) dew-point pressure of Bouchot and Richon (1995); (△) this work; (—) calculated values from equations of Bouchot and Richon (1995); (⋯) extrapolated values of equations of Bouchot and Richon (1995).

The equation of state in this study is an 18-term modified Benedict-Webb-Rubin equation of state developed by Piao et al. (1994). The equation is given as

$$\frac{P}{P_c} = \frac{\delta}{\tau Z_c} + \sum_{i=1}^{14} a_i \rho^{d_i} \tau^{t_i} + \delta^3 e^{-\delta^2} \sum_{i=15}^{18} a_i \rho^{d_i} \tau^{t_i} \quad (2)$$

where $\delta = \rho/\rho_c$, $\tau = T_c/T$, and $Z_c = P_c M/(R \rho_c T_c)$.

Table 4. Experimental PVT Properties with Densitometer ($w_1 = 43.967$ mass %, $w_2 = 52.032$ mass %, $w_3 = 4.001$ mass %)

P/MPa	$\rho/\text{kg}\cdot\text{m}^{-3}$	T/K	P/MPa	$\rho/\text{kg}\cdot\text{m}^{-3}$	T/K	P/MPa	$\rho/\text{kg}\cdot\text{m}^{-3}$	T/K
15.000	1247.0	263.15	15.000	1188.5	283.15	15.000	1127.5	303.15
14.000	1243.6	263.15	14.000	1184.3	283.15	14.000	1122.0	303.15
13.000	1240.1	263.15	13.000	1179.9	283.15	13.000	1116.2	303.15
12.000	1236.5	263.15	12.000	1175.3	283.15	12.000	1110.3	303.15
11.000	1232.9	263.15	11.000	1170.8	283.15	11.000	1104.1	303.15
10.000	1229.2	263.15	10.000	1165.9	283.15	10.000	1097.5	303.15
9.000	1225.4	263.15	9.000	1160.8	283.15	9.000	1090.5	303.15
8.000	1221.4	263.15	8.000	1155.5	283.15	8.000	1083.2	303.15
7.000	1217.3	263.15	7.000	1149.9	283.15	7.000	1075.4	303.15
6.000	1213.1	263.15	6.000	1144.2	283.15	6.000	1067.2	303.15
5.000	1208.6	263.15	5.000	1138.2	283.15	5.000	1058.2	303.15
4.000	1204.2	263.15	4.000	1131.9	283.15	4.000	1048.3	303.15
3.000	1199.4	263.15	3.000	1125.2	283.15	3.000	1037.5	303.15
2.000	1194.6	263.15	2.000	1118.0	283.15	2.000	1025.5	303.15
1.500	1192.1	263.15	1.500	1114.4	283.15	1.500	1018.9	303.15
15.000	1062.8	323.15	15.000	992.8	343.15	15.000	917.3	363.15
14.000	1055.4	323.15	14.000	983.1	343.15	14.000	903.8	363.15
13.000	1047.8	323.15	13.000	972.5	343.15	13.000	888.9	363.15
12.000	1039.8	323.15	12.000	961.0	343.15	12.000	872.2	363.15
11.000	1031.1	323.15	11.000	948.4	343.15	11.000	853.4	363.15
10.000	1021.7	323.15	10.000	934.6	343.15	10.000	831.2	363.15
9.000	1011.7	323.15	9.000	918.7	343.15	9.000	804.4	363.15
8.000	1000.8	323.15	8.000	900.8	343.15	8.000	770.3	363.15
7.000	988.7	323.15	7.000	879.4	343.15	7.000	722.5	363.15
6.000	975.2	323.15	6.000	852.5	343.15	6.000	640.4	363.15
5.000	959.7	323.15	5.000	815.6	343.15	5.000	435.2	363.15
4.000	941.4	323.15	4.000	749.7	343.15	4.000	221.8	363.15
3.000	918.6	323.15	3.000	172.7	343.15	3.000	136.3	363.15
2.500	904.5	323.15	2.000	89.2	343.15	2.000	78.6	363.15
			1.500	61.7	343.15	1.500	55.7	363.15
15.000	836.4	383.15	15.000	752.0	403.15			
14.000	817.9	383.15	14.000	727.7	403.15			
13.000	797.1	383.15	13.000	699.7	403.15			
12.000	772.8	383.15	12.000	666.5	403.15			
11.000	743.8	383.15	11.000	626.9	403.15			
10.000	708.1	383.15	10.000	577.6	403.15			
9.000	661.5	383.15	9.000	515.8	403.15			
8.000	596.6	383.15	8.000	440.8	403.15			
7.000	499.4	383.15	7.000	357.6	403.15			
6.000	372.8	383.15	6.000	278.4	403.15			
5.000	259.5	383.15	5.000	209.8	403.15			
4.000	177.7	383.15	4.000	152.8	403.15			
3.000	117.7	383.15	3.000	105.3	403.15			
2.000	71.0	383.15	2.000	65.0	403.15			
1.500	50.9	383.15	1.500	47.0	403.15			

Table 5. Comparison of Critical Parameters of R-404A

author	year	purity	P_c/MPa	$\rho_c/\text{kg}\cdot\text{m}^{-3}$	T_c/K	uncertainty		
						P_c/kPa	$\rho_c/\text{kg}\cdot\text{m}^{-3}$	T_c/mK
Bouchot and Richon	1995	na ^a	na	487.4	344.7	na	na	na
this work	1998	99.99 mass %	3.726	490	345.15	2	3	10

^a na, not available.

The critical parameters were taken from this work. The gas constant is $R = 8.314\,471\text{ J}\cdot\text{mol}^{-1}\cdot\text{K}^{-1}$, and the molecular weight is $M = 97.604\text{ g}\cdot\text{mol}^{-1}$. The coefficients and exponents are given in Table 6.

This equation of state has been developed on the basis of the single-phase PVT data of both Bouchot and Richon (1995) and this work, saturation data of Bouchot and Richon (1995), and thermodynamic restrictions at the critical point, $(\partial P/\partial \rho)_{T_c} = (\partial^2 P/\partial^2 \rho^2)_{T_c} = 0$, using least-squares fitting.

Figures 3–5 show comparisons of the single-phase PVT values calculated from eq 2 with the experimental data. Information about these experimental data is listed in Table 7. Figure 3a,b shows comparisons of density values calculated from eq 2 with the experimental data of Bouchot and Richon (1995). In Figure 3a,b deviations at pressures

up to 1 MPa exceed 0.3%; however, these deviations are less than $0.4\text{ kg}\cdot\text{m}^{-3}$. In Figure 3b a maximum deviation is 0.8% in density; the deviation of this point is -0.3% in pressure because the slope of the isotherm at 333 K is small at 2.8 MPa. Figure 4 shows a comparison of pressure values calculated from eq 2 with our data obtained with the constant volume apparatus. All the deviations are within $\pm 0.1\%$, and a root-mean-square of the deviations is 0.05%. Figure 5a,b shows comparisons of density values calculated from eq 2 with our data obtained with the vibrating-tube densitometer. In Figure 5a a maximum deviation is 0.7% because the isotherm at 343 K largely curves around 4 MPa. In Figure 5b three points exceed 1% because the slope of the isotherm at 363 K is small at 5 MPa and the isotherm largely curves around 4 MPa and 6 MPa.

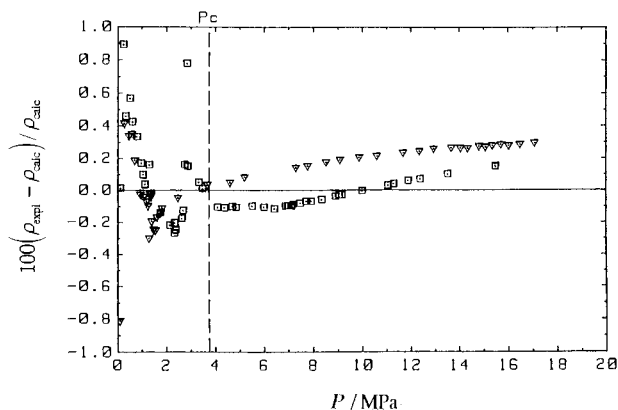
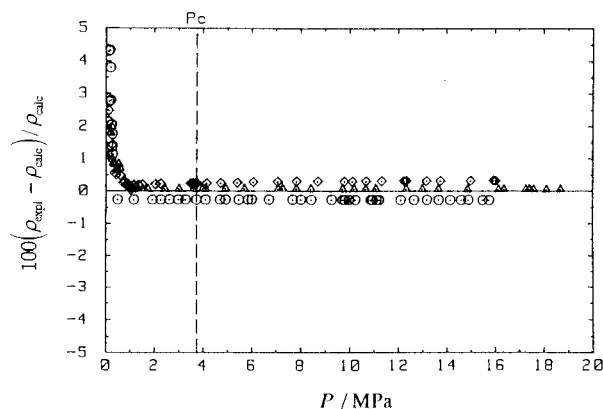


Figure 3. Comparison of R-404A density values calculated from eq 2 with experimental data of Bouchot and Richon (1995). (a, top) Isotherms: (○) 253 K; (△) 273 K; (◇) 293 K. (b, bottom) Isotherms: (▽) 313 K; (□) 333 K.

Table 6. Coefficients and Exponents for Eq 2

i	a_i	d_i	t_i
1	1.754 150 671 49	2	-1
2	-1.894 190 157 93	2	0
3	-6.345 694 554 31	2	2
4	1.268 875 638 11	2	3
5	1.069 774 986 72	3	-1
6	-3.996 422 729 33	3	0
7	9.111 708 249 43	3	2
8	8.103 526 274 30	4	0
9	-3.094 759 127 39	4	2
10	-8.103 372 985 91	5	0
11	-2.464 257 402 13	5	1
12	4.024 364 681 48	6	0
13	-9.234 772 347 31 × 10 ⁻¹	6	1
14	-6.972 707 730 57 × 10 ⁻¹	7	0
15	-1.193 797 610 14	0	0
16	-3.640 298 794 19	0	1
17	1.103 540 104 44	2	0
18	-3.139 708 039 22	2	1

Conclusion

The *PVT* properties of R-404A were measured with a constant volume apparatus with an optical cell in the critical region, and the critical parameters were deter-

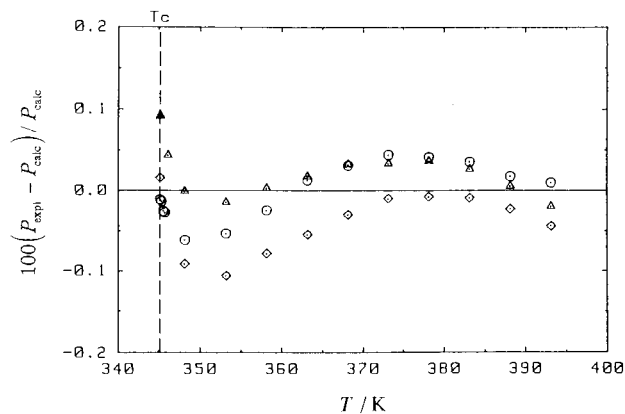


Figure 4. Comparison of R-404A pressure values calculated from eq 2 with our experimental data with the constant volume apparatus. Isochores: (○) 487 kg·m⁻³; (△) 490 kg·m⁻³; (◇) 493 kg·m⁻³.

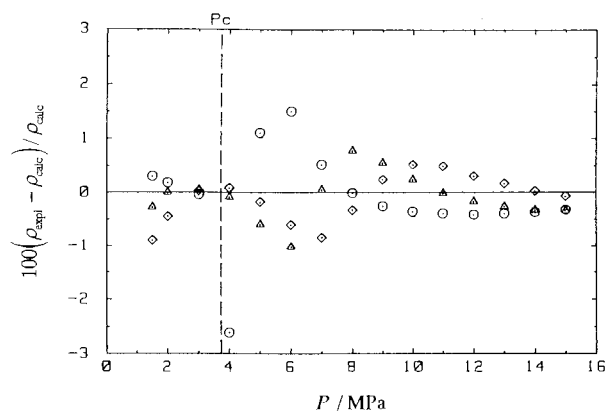
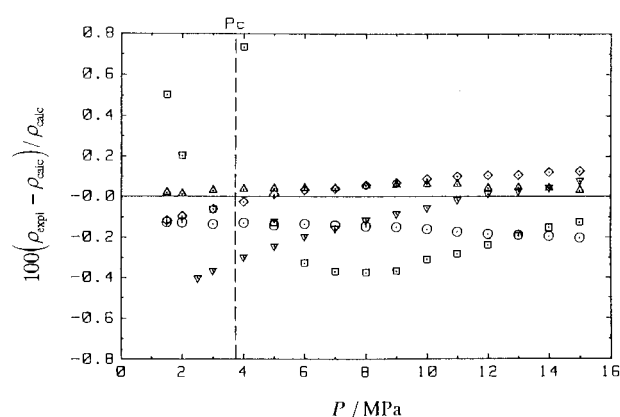


Figure 5. Comparison of R-404A density values calculated from eq 2 with our experimental data with the densitometer. (a, top) Isotherms: (○) 263 K; (△) 283 K; (◇) 303 K; (▽) 323 K; (□) 343 K. (b, bottom) Isotherms: (○) 363 K; (△) 383 K; (◇) 403 K.

mined. The *PVT* properties in the single phase were measured with the vibrating-tube densitometer. Our saturation pressures and densities of this work are consistent with the saturation pressures and the vapor-liquid

Table 7. Experimental Studies of Single-Phase *PVT* Data for R-404A

author	year	purity	method ^b	no. pts	range of data			uncertainty		
					<i>P</i> /MPa	ρ /kg·m ⁻³	<i>T</i> /K	<i>P</i> /kPa	ρ /kg·m ⁻³	<i>T</i> /mK
Bouchot and Richon	1995	na ^a	VTD	214	0.1–18.7	4–1279	253–333	±3	±0.3	±20
this work	1998	99.99 mass %	CVM	40	3.7–7.8	487–493	345–393	2	2	3
this work	1998	99.99 mass %	VTD	119	1.5–15.0	47–1247	263–403	4	0.5–3	7

^a na, not available. ^b CVM, constant volume method; VTD, vibrating tube densitometer.

coexistence curve of Bouchot and Richon (1995). The *PVT* properties of Bouchot and Richon (1995) and of this work were correlated with an 18-term modified Benedict–Webb–Rubin (MBWR) equation of state. These experimental data covered pressures 0.1 MPa to 19 MPa, densities from $4 \text{ kg}\cdot\text{m}^{-3}$ to $1279 \text{ kg}\cdot\text{m}^{-3}$, and temperatures from 253 K to 403 K. The MBWR equation of state represented these data very well.

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