

Critical Parameters and Vapor Pressure Measurements for 1,1,1-Trifluoroethane (R-143a)

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An apparatus to measure the critical pressure, density, and temperature by visual observation of the meniscus in an optical cell is presented. The critical parameters and vapor pressures for 1,1,1,2-tetrafluoroethane (R-134a) were measured to confirm the reliability of the measurements with the present apparatus. The critical pressure, density, and temperature of 1,1,1-trifluoroethane (R-143a) were determined to be 3.7639 MPa, 434 kg·m⁻³, and 345.861 K, respectively. Thirteen vapor pressures of R-143a were obtained at temperatures from 263 K to the critical temperature. The uncertainties of pressure, density, and temperature measurements are estimated to be within ±2.6 kPa, ±2 kg·m⁻³, and ±6 mK, respectively.

Introduction

The critical parameters are important to understand the thermodynamic state surface of fluids. They are also important to predict various thermodynamic properties using the principle of corresponding states.

The critical temperature and density of fluids have been mainly determined by visual observation of the appearance and/or disappearance of the meniscus at the vapor–liquid interface of the sample confined in an optical cell. The critical pressure has been found by extrapolating to the critical temperature using a vapor pressure equation fitted to vapor pressure data.

Vapor pressures are also required to understand the thermodynamic state surface of fluids and to derive enthalpies of vaporization using the Clausius–Clapeyron equation.

R-502, an azeotropic mixture of 48.8 mass % of chlorodifluoromethane (R-22) and 51.2 mass % of chloropentafluoroethane (R-115), has been widely used as a refrigerant in low-temperature refrigerators. This substance has not been supplied since 1996 according to the Montreal Protocol. Ternary mixtures of pentafluoroethane (R-125), 1,1,1-trifluoroethane (R-143a), and 1,1,1,2-tetrafluoroethane (R-134a) and binary mixtures of R-125 and R-143a have been considered as alternatives to R-502. R-143a is a component of these candidates to R-502. A literature survey indicates that the thermodynamic property data for R-143a are less accurately known than those for R-125 and R-134a.

We have constructed an apparatus to measure the critical pressure directly as well as the critical temperature and density by observing the disappearance of the vapor–liquid interface. The critical parameters and vapor pressures for R-134a were measured to confirm the uncertainty of the measurements obtained with the present apparatus. After that, we measured those for R-143a.

Experimental Apparatus and Procedure

A schematic diagram of the experimental apparatus is shown in Figure 1. The main portion of the apparatus is

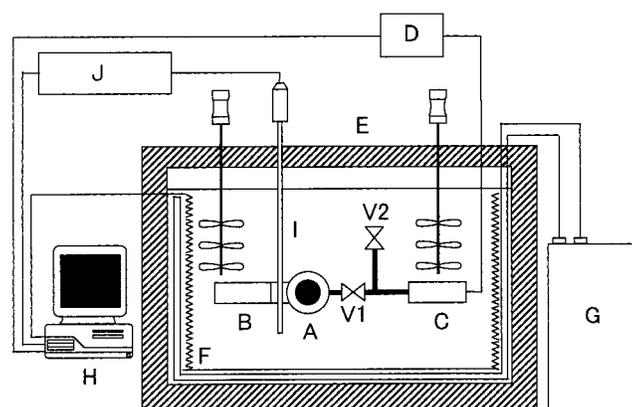


Figure 1. Schematic diagram of the experimental apparatus: A, optical cell; B, supplying vessel; C, pressure transducer; D, computer; E, thermostatic bath; F, heater; G, cooler; H, personal computer; I, thermometer; V, valve.

composed of an optical cell (A), a supplying vessel (B), and a pressure transducer (C, Paroscientific, Inc., Model 43K-101). The pressure measured with the transducer (C) is displayed on a computer (D, Paroscientific, Model Series 700).

The optical cell (A) is a cylindrical vessel made of 304 stainless steel (26 mm long and 20 mm inner diameter, about 8 cm³ in inner volume) with two synthetic sapphire windows (12 mm in thickness). The cell is welded to the supplying vessel (B) and is connected to the pressure gauge (C) by a valve V1. Each central axis of these vessels and gauge is placed in the same level.

The main portion of the apparatus is set in a thermostatic bath (E, 500 × 280 × 275 mm in inner dimensions) with two glass windows (45 mm diameter). The temperature in the bath is controlled by electric power to heaters (F) and a cooler (G) and is monitored continuously. The electric power to the heaters (F) was controlled with a personal computer (H), and the temperature of a fluid of the cooler was controlled with the internal controller. In the present study the bath fluid used is a silicon oil.

The temperature measurements are made with a 25-Ω platinum resistance thermometer (I, Chino, Model R800-

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Table 1. Uncertainty Analysis in Measurements

(a) pressure	
accuracy of transducer	±2 kPa
uncertainty in temperature measurements	±0.6 kPa
total	±2.6 kPa
(b) mass	
accuracy of analytical balance	±1 mg
fluctuation	±2 mg
total	±3 mg
(c) inner volume	
uncertainty of equation of state for nitrogen	±0.1 cm ³
uncertainty of equation for inner volume	±0.3 cm ³
total	±0.4 cm ³
(d) density	±2 kg·m ⁻³
(e) temperature	
accuracy of thermometer	±1 mK
accuracy in bridge	±1 mK
control	±2 mK
distribution	±2 mK
total	±6 mK

2) with the aid of a Mueller-type bridge (J, Automatic Systems Laboratories Limited, Model F17A). The thermometer (I) is calibrated with an accuracy of ±1 mK on ITS-90 at the National Research Laboratory of Metrology, Ibaraki, Japan. The thermometer (I) is mounted in the vicinity of the optical cell (A) in the thermostatic bath because the sample temperature is not measured directly. The fluctuations in the temperature are within ±2 mK.

Corrections were made to the inner volume of the optical cell (A), including the tubes and the valves as well as the supplying vessel (B), as a result of the thermal expansion and the pressure deformation at the experimental temperatures and pressures. We developed equations giving the inner volume as a function of temperature and pressure and calibrated them by charging known masses of nitrogen and measuring temperatures and pressures. An equation of state proposed by Jacobsen and Stewart (1973) was used to calculate the nitrogen densities.

For charging samples with prescribed densities, additional cylindrical stainless steel 304 vessels were used to fill each of them with the respective samples. To prepare the prescribed density of the sample, we used a precision analytical balance (Mettler, Model PM5003 Comparator) with an uncertainty of ±1 mg. Whenever an overestimation of the mass of a sample was made, portions of the sample were discharged to the necessary extent. Each sample was then transferred successively into the supplying vessel (B), which had been evacuated in advance up to around 0.4 mPa and cooled by liquid nitrogen. Prior to the measurement, the main portion of the cell was evacuated with the valve V1 closed to 0.4 mPa through a valve V2. Pressures and temperatures of the sample were measured with some temperature increments. For obtaining the critical parameters, the meniscus observation, pressure and temperature measurements were made after the sample temperature was maintained for about 1 day. Vapor pressures were measured after temperature equilibrium had been reached. The personal computer (H) was used in measuring temperature and pressure, processing the raw data, and storing the information.

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

As shown in Table 1, the uncertainties of the measurement is estimated to be within ±2.6 kPa in pressure, ±2 kg·m⁻³ in density, and ±6 mK in temperature. The uncertainty of the density measurement was estimated

Table 2. Measured Critical Parameters for R-134a and R-143a

R-134a			R-143a		
<i>P</i> /MPa	ρ /kg m ⁻³	<i>T</i> /K	<i>P</i> /MPa	ρ /kg m ⁻³	<i>T</i> /K
4.0481	507	374.070	3.7639 ^a	434 ^a	345.861 ^a
4.0482 ^a	509 ^a	374.074 ^a			
4.0478	511	374.070			

^a Value is considered the best critical property from this work.

from the uncertainties of the mass measurement and inner volume.

Results

Critical Parameters. Three sets of *PVT* properties in the saturated state were measured for R-134a. One set was measured for R-143a. The numerical values of the results are given in Table 2. The critical opalescence has been observed at all the measurements. With respect to R-143a, we considered that the density of 434 kg·m⁻³ was close to the critical density in consideration of the meniscus behavior, and so additional measurements were not made.

The critical point of fluids is defined as the state where all the thermodynamic properties of coexisting vapor and liquid phases become identical. That is, the meniscus disappears at the center of the optical cell, and the critical opalescence is observed most intensely and equally in both vapor and liquid phases. On the basis of our measurements, we determined the critical parameters in consideration of the meniscus disappearing level as well as the intensity of the critical opalescence.

In the case of R-134a, at the density of 507 kg·m⁻³, the level at which the meniscus disappeared was slightly below the center. The critical opalescence in the liquid phase was observed to be more intense than in the vapor phase. On the basis of this observation, we considered that the density of 507 kg·m⁻³ was slightly lower than the critical density. On the other hand, at the density of 511 kg·m⁻³, the level at which the meniscus disappeared was slightly higher than the center. The critical opalescence in the vapor phase was observed to be more intense than in the liquid phase. Therefore we concluded that the density of 511 kg·m⁻³ was slightly higher than the critical density. In the case of 509 kg·m⁻³, the critical opalescence in the vapor phase was as intense as in the liquid phase. Moreover, the level at which the meniscus disappeared was the center of the cell. Therefore we considered that the density of 509 kg·m⁻³ was the density closest to the critical density among these measurements.

In consideration of the meniscus behavior with respect to the three densities mentioned above, the critical density, ρ_c , of R-134a was determined, finally, as

$$\rho_c = (509 \pm 2) \text{ kg}\cdot\text{m}^{-3}$$

The uncertainty of the critical density was estimated to be within that of the density measurement. The critical temperature, T_c , and pressure, P_c , of R-134a were determined by adopting the experimental temperature and pressure values at the density of 509 kg·m⁻³ as follows:

$$T_c = (374.074 \pm 0.006) \text{ K}$$

$$P_c = (4.0482 \pm 0.0026) \text{ MPa}$$

The uncertainties of the critical temperature and pressure

Table 3. Experimental Vapor Pressures of R-134a and R-143a

<i>T</i> /K	<i>P</i> /MPa	<i>T</i> /K	<i>P</i> /MPa	<i>T</i> /K	<i>P</i> /MPa
R-134a					
263.148	0.2003	303.152	0.7700	343.149	2.1167
273.149	0.2929	314.034	1.0409	353.150	2.6332
283.151	0.4143	323.151	1.3179	363.150	3.2444
293.150	0.5715	333.150	1.6817	373.150	3.9726
R-143a					
263.154	0.4473	314.067	1.8727	345.648	3.7465
273.149	0.6194	323.151	2.3087	345.749	3.7544
283.001	0.8357	333.150	2.8763	345.852	3.7627
293.149	1.1048	343.150	3.5552		
303.150	1.4341	345.149	3.7073		

were estimated to be within those of the temperature and pressure measurements, respectively.

In the case of R-143a, at the density of $434 \text{ kg}\cdot\text{m}^{-3}$, the critical opalescence in the vapor phase was as intense as in the liquid phase. The meniscus disappearance occurred at the center of the cell. Therefore we considered that the density of $434 \text{ kg}\cdot\text{m}^{-3}$ was close to the critical density.

In consideration of the meniscus behavior, the critical density of R-143a was determined as

$$\rho_c = (434 \pm 2) \text{ kg}\cdot\text{m}^{-3}$$

The critical temperature and pressure were taken as the experimental temperature and pressure values at the density of $434 \text{ kg}\cdot\text{m}^{-3}$:

$$T_c = (345.861 \pm 0.006) \text{ K}$$

$$P_c = (3.7639 \pm 0.0026) \text{ MPa}$$

Judging from the uncertainties of the critical parameters for R-134a, those for R-143a were estimated to be within the uncertainties of the measurements.

Vapor Pressures. Twelve vapor pressures were measured at temperatures from 263 K to the critical temperature for R-134a. Thirteen vapor pressures were also measured at temperatures from 263 K to the critical

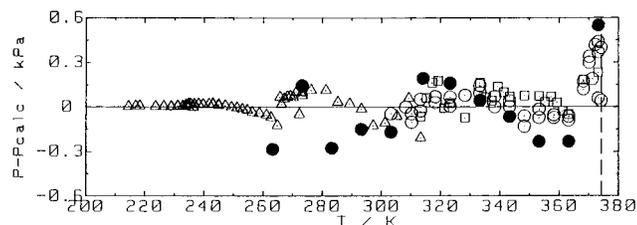


Figure 2. Comparison of R-134a vapor pressure values calculated from an equation by McLinden et al. (1993) with experimental data: (○), Baehr and Tillner-Roth (1991); (△), Goodwin et al. (1992); (□), Weber (1989); (●), this work.

temperature for R-143a. The numerical values are given in Table 3.

Discussion

Critical Parameters. A comparison of the critical parameters for R-134a is shown in Table 4. The present critical density is in agreement with the data of Basu and Wilson (1989), Kabata et al. (1989), Fukushima et al. (1990a), Nagel and Bier (1995), and Aoyama et al. (1996) within the respective uncertainties. With respect to the critical temperature, our result agrees with the data of Basu and Wilson (1989), Nishiumi et al. (1993), Nagel and Bier (1995), and Aoyama et al. (1996) within the respective uncertainties. The critical pressure is depend on the critical temperature, so it is impossible to compare the values directly. However, the present critical pressure agrees with the values of McLinden et al. (1989), Fukushima et al. (1990b), and Nagel and Bier (1995) within the respective uncertainties.

A comparison of the critical parameters for R-143a is shown in Table 5. The present critical density agrees with the data of Mears et al. (1955), Fukushima et al. (1993), and Aoyama et al. (1996) within the respective uncertainties. With respect to the critical temperature, our result agrees with the data of Mears et al. (1955) and Aoyama et al. (1996) within the respective uncertainties. The present critical pressure agrees with the value of Mears et al. (1955) within the respective uncertainties.

Table 4. Comparison of Critical Parameters of R-134a

author	year	purity	$\rho_c/\text{kg}\cdot\text{m}^{-3}$	T_c/K	P_c/MPa	uncertainty		
						$\rho_c/\text{kg}\cdot\text{m}^{-3}$	T_c/K	P_c/kPa
Basu and Wilson	1989	99.95 mass %	512.2	374.23 ^a	4.067	±5	±0.15	±2.7
Kabata et al.	1989	99.8 mass %	508	374.27 ^a		±3	±0.01	
Kubota et al.	1989			374.24 ^a	4.065			
McLinden et al.	1989	99.94 mass %	515.3	374.179 ^a	4.056	±1	±0.010	±10
Fukushima et al.	1990a	99.99%	507	374.15 ^a		±5	±0.03	
Fukushima et al.	1990b	99.99%		374.15 ^a	4.055		±0.03	±6
Morrison and Ward	1991	99.94%	515.2	374.229 ^a	4.0680	±1.5	±0.010	±5
Nishiumi et al.	1993	99.8%		374.04 ^a	4.035		±0.05	±6
Nagel and Bier	1995	99.9%	514	374.10	4.051	±10	±0.06	±6
Aoyama et al.	1996	99.95%	509	374.083		±1	±0.010	
this work	1997	99.995 mass %	509	374.074	4.0482	±2	±0.006	±2.6

^a Critical temperature value converted from IPTS-68 to ITS-90.

Table 5. Comparison of Critical Parameters of R-143a

author	year	purity	$\rho_c/\text{kg}\cdot\text{m}^{-3}$	T_c/K	P_c/MPa	uncertainty		
						$\rho_c/\text{kg}\cdot\text{m}^{-3}$	T_c/K	P_c/kPa
Mears et al.	1955		434	346.2 ^a	3.758	±10	±0.5	±69
Fukushima	1993	99.98%	429	345.97	3.769	±3	±0.03	
Wang et al.	1993	99.95 mass %	442	346.18	3.780	±4	±0.05	±6
Aoyama et al.	1996	99.95 mol %	434	345.860		±1	±0.010	
this work	1997	99.99 mass %	434	345.861	3.7639	±2	±0.006	±2.6

^a Critical temperature value converted from IPTS-68 to ITS-90.

Table 6. Reported Vapor Pressure Measurements for R-134a

author	year	purity	no. pts	range		uncertainty	
				<i>T</i> /K	<i>P</i> /MPa	<i>T</i> /mK	<i>P</i> /kPa
Weber	1989	99.96%	22	313–373	1.02–3.97	±1	±0.1
Baehr and Tillner-Roth	1991	99.97 mol %	37	303–374	0.77–4.05	±5	±0.47
Goodwin et al.	1992	99.954 mol %	57	214–313	0.02–1.02	±1	±0.1
this work	1997	99.995 mass %	12	263–373	0.20–3.97	±6	±2.6

Table 7. Reported Vapor Pressure Measurements for R-134a

author	year	purity	no. pts	range		uncertainty	
				<i>T</i> /K	<i>P</i> /MPa	<i>T</i> /mK	<i>P</i> /kPa
Kubota et al.	1990	99.9 mol %	4	273–338	0.62–3.19	±50	±8
Fukushima	1993	99.98%	18	275–346	0.64–3.75	±20	±5
Kubota and Matsumoto	1993	99.5 mol %	5	278–333	0.72–2.88	±50	±2
Wang et al.	1993	99.95 mass %	30	312–346	1.78–3.78	±28	±1.4
Widiatmo et al.	1994	99.0 mol %	12	280–340	0.77–3.34	±15	±10
Giuliani et al.	1995	99.91 mass %	33	244–345	0.23–3.72	±30	±1.3
Zhang et al.	1995	99.95 mol %	11	295–342	1.16–3.47	±8	±0.8
this work	1997	99.99 mass %	13	263–346	0.45–3.76	±6	±2.6

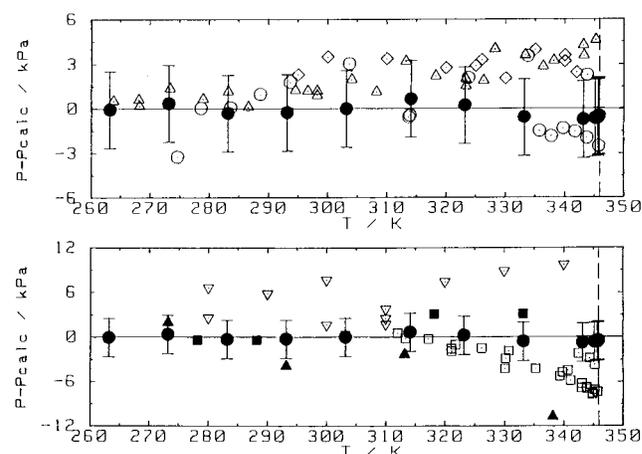


Figure 3. Comparison of R-134a vapor pressure values calculated from eq 1 with experimental data: (○), Fukushima (1993); (△), Giuliani et al. (1995); (▲), Kubota et al. (1990); (■), Kubota and Matsumoto (1993); (□), Wang et al. (1993); (▽), Widiatomo et al. (1994); (◇), Zhang et al. (1995); (●), this work.

Vapor Pressures. Figure 2 shows a comparison of the R-134a vapor pressure values calculated from a equation developed by McLinden et al. (1993) with the experimental results. Information about these experimental data is listed in Table 6. This equation represents the results of Baehr and Tillner-Roth (1991) within ±0.4 kPa, Goodwin et al. (1992) within ±0.2 kPa, Weber (1989) within ±0.2 kPa, and our results within ±0.6 kPa. Our results agree with those of Baehr and Tillner-Roth (1991), Goodwin et al. (1992), and Weber (1989) within our estimated uncertainty.

The present vapor pressure results for R-134a were correlated by the following equation using our reported critical pressure and temperature.

$$\ln\left(\frac{P}{P_c}\right) = \frac{1}{1-\tau}(-7.327\ 17\tau + 1.501\ 11\tau^{1.5} - 3.211\ 43\tau^3 + 0.275\ 322\tau^6) \quad (1)$$

where $\tau = 1 - T/T_c$, $P_c = 3.7639$ MPa, and $T_c = 345.861$ K. This equation represents the present results within ±0.7 kPa. Figure 3 shows a comparison of the R-134a vapor pressure values calculated from eq 1 with the experimental results. Information about these experimental data is listed in Table 7. Equation 1 represents the data of Fukushima (1993) within ±3.5 kPa and Kubota and Matsumoto (1993) within ±3.1 kPa. The deviations of

Giuliani et al. (1995), Widiatomo et al. (1994), and Zhang et al. (1995) are positive; most deviations of Kubota et al. (1990) and Wang et al. (1993) are negative. The data of Fukushima (1993), Kubota et al. (1990), Kubota and Matsumoto (1993), and Widiatomo et al. (1994) agree with our results within the respective uncertainties. Most of the results of Giuliani et al. (1995), Wang et al. (1993), and Zhang et al. (1995) agree with our results within the respective uncertainties.

Conclusion

The present critical parameters for R-134a agree well with the reliable results within their respective uncertainties. Our measured vapor pressures of R-134a agree well with other reliable results within our estimated uncertainty. Therefore, we believe that the critical parameters and vapor pressures obtained with the present apparatus are reliable.

The critical parameters and vapor pressures for R-134a were measured. Using the critical temperature and pressure obtained, the present vapor pressures were correlated by an equation over the range from 263 K to the critical temperature. This work will be useful to provide reliable information about the critical parameters and vapor pressures of R-134a.

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