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Vapor Pressure and Gas Phase PVT Data and Correlation for 1,1,1-Trifluoroethane (R143a)

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A constant volume apparatus has been used to measure the vapor pressure and single phase densities (PVT) for 1,1,1-trifluoroethane (R143a), an ozone friendly refrigerant. The sample has a purity greater than 99.91% by mass. The measurements were done from 244 to 365 K at pressures up to 4.4 MPa and at densities ranging from 5% to 99% of the critical density. The uncertainties involved in the measurement of temperature, pressure, mass, volume, and specific volume are explicitly given. Thirty-three values for the vapor pressure and sixty-three densities along six isochores are presented and have been compared with previously published data and correlations. A Wagner type correlation for the vapor pressure and a Martin–Hou equation of state have been used to fit the measured data, which have been analyzed using the average absolute deviation, bias, and standard deviation. The gas phase densities have been used to determine the second virial coefficients.

Introduction

The Montreal Protocol of 1987 and the deliberations later at London in 1989 and at Copenhagen in 1992 envisage the rapid phase-out of ozone-depleting substances used in refrigeration and other allied applications. Consequently, the search for alternative refrigerants continues, and as it is clear that no ideal refrigerant exists and no single substance can easily meet all favorable property requirements, binary and tertiary mixtures are also being considered as suitable replacements.

The thermodynamic properties of the replacement refrigerants and their harmful effects on the environment should be known before they can be considered for large scale applications. Of the many alternative refrigerants proposed, 1,1,1-trifluoroethane (R143a) shows promise, in spite of its flammability, as a constituent of binary or tertiary mixtures with difluoromethane (R32), pentafluoroethane (R125), and 1,1,1,2-tetrafluoroethane (R134a). These mixtures are expected to replace chlorodifluoromethane (R22) and the R22/R115 azeotropic mixture better known as R502. However, accurate and reliable data on the vapor pressure and gas phase PVT behavior of R143a are few, as compared to, for example, R134a.

This paper reports the vapor pressure and densities for R143a, along with the uncertainties involved in the measurements. A Wagner type correlation for vapor pressure has been proposed, and the present measurements have been compared with earlier values. The gas phase densities along six isochores have been verified for thermodynamic consistency through the virial equation of state. The measured densities were well represented by a Martin–Hou type equation of state and the second virial coefficients estimated.

Experimental Apparatus and Procedure

A constant volume apparatus of about 252 cm^3 was used to measure the vapor pressure and densities. The details of the apparatus and the verification of the experimental procedure are given in detail elsewhere (Giuliani et al., 1995), and only a brief summary will be presented here. This apparatus was earlier used to measure the thermodynamic properties of R134a (Baroncini et al., 1990), R123 and R124 (Baroncini et al., 1991), R125 (Baroncini et al., 1992), R32 (Baroncini et al., 1993), R134a and R22 (Giuliani et al., 1995), and R143a (Giuliani et al., 1994).

Figure 1 shows the schematic of the experimental setup used. A stainless steel (AISI 304) spherical cell (A) of internal diameter about 80 mm contains the refrigerant sample and is connected to a diaphragm type differential pressure transducer (B; Ruska Model 2413), which is coupled to an electronic null indicator (C; Ruska Model 2416). The pressure is regulated by a precision pressure controller (D; Ruska Model 3981). A gas-lubricated piston cylinder dead weight gauge (E; Ruska Model 2465, full scale 7 MPa) and a vibrating cylinder pressure gauge (F; Ruska Model 6220, full scale 130 kPa) are used to measure pressures. The vibrating cylinder pressure gauge is used to measure the ambient pressure and the sample pressure when it is lower than 130 kPa. Nitrogen gas is used as the pressure transmitting fluid, and the nitrogen circuit consists of a reservoir (G), expansion vessels (H), and pressure regulating systems (I). The cell and the pressure transducer are immersed in a principal thermostatic bath (J) containing a mixture of ethylene glycol and water and controlled by a PID device (K). An auxiliary bath (L) aids in maintaining a constant bath temperature between 243 and 363 K. A 100 Ω platinum resistance thermometer (M; Minco Products Model S7929 S/N 1476) is immersed close to the cell and is connected to a digital indicator (N; Franco Corradi Model RP 7000).

The R143a refrigerant samples with a stated purity of 99.91% by mass were obtained from Ausimont SpA and were degassed to remove noncondensible gases. Gas chromatographic studies conducted in our laboratory confirmed the sample purity to be greater than 99.91% by mass. The sample mass in the cell was determined by weighing it with an electronic balance (Sartorius MC1). After filling the cell, the pressure was measured at different temperatures along the saturation curve and in the gas

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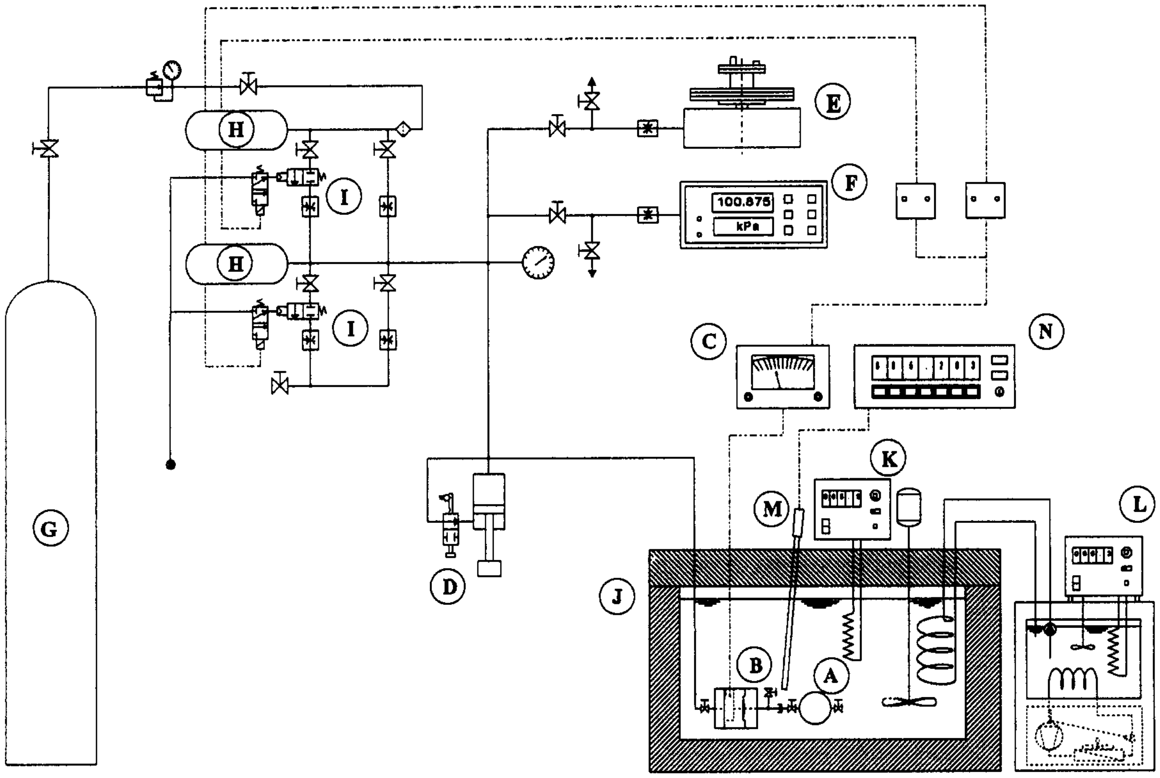

Figure 1. Schematic of the experimental apparatus: (A) constant volume spherical cell, (B) differential pressure transducer, (C) electronic null indicator, (D) precision pressure controller, (E) gas-lubricated dead weight pressure gauge, (F) vibrating cylinder pressure gauge, (G) nitrogen reservoir, (H) expansion chambers, (I) automatic pressure control system, (J) thermostatic bath, (K) PID temperature controller, (L) auxiliary bath, (M) platinum resistance thermometer, (N) digital temperature indicator.

phase. When sufficient measurements had been completed with one filling, a new sample was added and the measurements were continued.

The uncertainty in the temperature measurement is due to the thermometer and the bath instability. The Pt resistance thermometer was calibrated according to ITS-90 at the Istituto Metrologico G. Colonnetti (IMGC), Turin, and has an uncertainty of ±10 mK for temperatures above 248 K and ±20 mK for temperatures below 248 K. Preliminary experiments were conducted to understand the thermostatic bath instability so as to estimate the total uncertainty in the temperature measurement (δT); this is estimated to be ±15 mK for temperature measurements above 268 K and ±30 mK for temperature measurements below 268 K.

The uncertainty in the pressure measurement is due to the transducer uncertainty and null indicator system (±0.3 kPa) and uncertainties in pressure gauges. The vibrating cylinder gauge has an uncertainty of ±26 Pa, while the dead weight gauge has an uncertainty of ±0.01% of the reading. The constant term in the pressure uncertainty (δP_c) is given by

$$\delta P_c/\text{kPa} = \pm[0.3^2 + 0.026^2 + 0.0001P^2/\text{kPa}]^{0.5} \qquad (1)$$

where P is the measured pressure.

The uncertainty in the measurement of the sample mass (δm) is estimated to be within ±0.04 g. The volume (V) of the cell, the piping, and the cavity of the pressure transducer was measured using distilled and deaerated water at 295 K and is estimated as 252.2 cm^3 with an uncertainty (δV) of ±0.3 cm^3. The variations in cell volume due to temperature and pressure have been taken into account.

The total uncertainty associated with the calculation of specific volume (δv) was estimated from the following equation:

$$\delta v = \pm\left[\left(\frac{\delta V}{m}\right)^2 + \left(\frac{V\,\delta m}{m^2}\right)^2\right]^{0.5} \qquad (2)$$

For a sample mass of 5 g, δv is estimated to be about 0.06 dm^3·kg^{-1}, while it is 0.002 dm^3·kg^{-1} for a sample of 150 g.

The total uncertainty in pressure in the vapor pressure measurement (δP_{vp}) was estimated as the sum of error propagation as

$$\delta P_{\text{vp}} = \pm[\delta P_c^{\,2} + \delta P_{\text{bath}}^{\,2}]^{0.5} \qquad (3)$$

with the uncertainty due to the bath instability calculated from

$$\delta P_{\text{bath}} = \pm(\mathrm{d}P/\mathrm{d}T)\delta T \qquad (4)$$

where the derivative $\mathrm{d}P/\mathrm{d}T$ of the vapor pressure curve is calculated using the Wagner equation for the vapor pressure of R143a presented later and δT is the uncertainty in the temperature measurement.

Similarly, the total uncertainty in the pressure measurement in the gas phase (δP_{PVT}) is also influenced by temperature fluctuations due to bath temperature instability and by specific volume uncertainty, and was estimated from

$$\delta P_{\text{PVT}} = \pm[\delta P_c^{\,2} + \delta P_T^{\,2} + \delta P_v^{\,2}]^{0.5} \qquad (5)$$

where

$$\delta P_T = \pm(\partial P/\partial T)_v\, \partial T \qquad (6)$$

and

$$\delta P_v = \pm(\partial P/\partial v)_T\, \delta v \qquad (7)$$

respectively, where the partial derivatives are calculated from the proposed Martin–Hou equation of state.

Table 1. Summary of experimental vapor pressure data for R143a

ref	T/K range	sample purity/%	uncertainty	no. of exptl. points
Russell et al. (1944)	173−225	99.95	±0.013 kPa	9
Fukushima (1993)	275−345	99.98	±5 kPa	18
Widiatmo et al. (1994)	280−340	99.9	±10 kPa	12
Higashi (1994)	283−338		±0.5%	13
Giuliani et al. (1994)	244−345	99.8 (mol)	±0.5 kPa	62
this work	244−345	99.91	as given	33

Table 2. Experimental Vapor Pressure of R143a

T_{90}/K	P/kPa	δP_{vp}/kPa (eq 3)	T_{90}/K	P/kPa	δP_{vp}/kPa (eq 3)
244.42	226.2	±0.4	304.06	1469.3	±0.7
245.07	231.9	±0.4	308.23	1628.1	±0.8
246.55	245.9	±0.4	313.21	1837.6	±0.8
248.02	260.2	±0.5	318.24	2066.0	±0.9
253.03	314.2	±0.5	323.29	2317.7	±1.0
256.99	361.8	±0.5	323.32	2318.8	±1.0
263.79	457.6	±0.6	326.29	2478.2	±1.0
268.07	527.2	±0.7	328.18	2585.9	±1.1
268.16	528.3	±0.7	328.22	2588.2	±1.1
273.37	624.7	±0.5	333.31	2890.4	±1.1
278.95	739.6	±0.5	336.34	3083.1	±1.2
283.21	838.6	±0.5	338.13	3202.8	±1.2
286.59	922.4	±0.6	343.11	3557.2	±1.3
294.54	1148.3	±0.6	343.20	3563.2	±1.3
296.64	1213.9	±0.7	345.21	3717.3	±1.3
298.22	1265.1	±0.7	345.21	3717.3	±1.3
298.22	1264.8	±0.7			

Results and Discussion

The results were compared with previously published data using the statistical parameters, namely, the absolute average deviation (AAD), the bias, and the standard deviation (σ) defined as

$$\mathrm{AAD} = \frac{1}{N}\sum_{i=1}^{N} 100\left|\frac{(P - P_{\mathrm{calc}})}{P}\right| \tag{8}$$

$$\mathrm{bias} = \frac{1}{N}\sum_{i=1}^{N} 100\left(\frac{P - P_{\mathrm{calc}}}{P}\right) \tag{9}$$

$$\sigma = \left[\frac{1}{N-1}\sum_{i=1}^{N}(P - P_{\mathrm{calc}})^2\right]^{0.5} \tag{10}$$

Vapor Pressure. Table 1 summarizes the status of previous vapor pressure measurements along with the uncertainties in the pressure measurements.

Thirty three vapor pressure values from 244 K to the critical temperature are given in Table 2 along with the uncertainties calculated using eq 3. A change in the pressure uncertainty values at 268.15 K is due to the influence of the uncertainty contributed by the bath instability, and the maximum uncertainty is 1.3 kPa at a pressure of about 3.7 MPa.

The results presented in Table 2 were used along with the critical temperature from Higashi (1994) to fit a three-coefficient Wagner type equation as:

$$\ln P_r = (1/T_r)[a_1(1 - T_r) + a_2(1 - T_r)^{1.5} + a_3(1 - T_r)^{2.5}] \tag{11}$$

where $P_r = P/P_c$ and $T_r = T/T_c$. The critical pressure was obtained from eq 11 as P_c = 3.7697 MPa with T_c = 345.88 K and a_1 = −7.385 168 51, a_2 = 1.818 473 62, a_3 = −2.380 153 13.

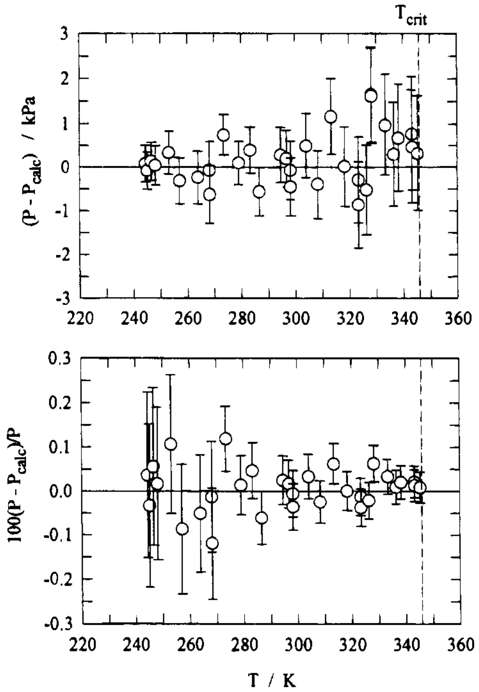

Figure 2. Vapor pressure deviations of R143a data from eq 11 with uncertainties.

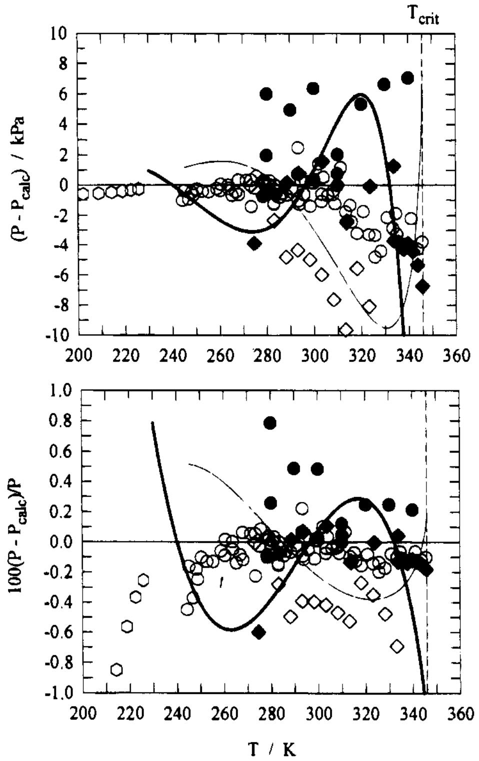

Figure 3. Vapor pressure deviations of R143a from eq 11: (○) Giuliani et al. (1994), (●) Widiatmo et al. (1994), (◇) Higashi (1994), (◆) Fukushima (1993), (○) Russel et al. (1944), (thick line) Arnaud et al. (1991), (thin line) Mears et al. (1955).

Figure 2 shows the deviation of the present results from eq 11 with the uncertainties of the pressure shown as error bars. The goodness of fit between the data and eq 11 was calculated in terms of σ, AAD, and bias as 0.6285 kPa, 0.0388, and −0.008 32, respectively, indicating the closeness of fit.

Figure 3 shows the deviation between eq 11 and the results of Giuliani et al. (1994), Higashi (1994), Widiatmo et al. (1994), Fukushima (1993), and Russell et al. (1944)

Table 3. Experimental PVT Properties of R143a

ϱ/ (kg·m^{-3})	T_{90}/K	P/kPa	δP_{PVT}/ kPa (eq 5)	ϱ/ (kg·m^{-3})	T_{90}/K	P/kPa	δP_{PVT}/ kPa (eq 5)
	$\varrho/\varrho_c = 0.051$				$\varrho/\varrho_c = 0.319$		
22.00	267.96	502.6	±3.1	137.45	330.08	2594.9	±1.9
21.99	273.12	516.1	±3.2	137.43	332.73	2656.1	±2.0
21.99	278.14	529.3	±3.4	137.42	335.22	2710.5	±2.1
21.98	283.09	542.1	±3.5	137.40	338.02	2771.4	±2.3
21.98	288.23	555.1	±3.6	137.38	340.43	2821.9	±2.4
21.97	293.05	567.2	±3.7	137.36	343.29	2883.1	±2.5
21.96	298.16	579.9	±3.8	137.33	346.48	2950.9	±2.6
21.95	308.24	604.9	±4.0	137.31	350.33	3031.1	±2.8
21.94	318.19	629.2	±4.2	137.28	354.51	3117.3	±3.0
21.93	328.29	653.7	±4.4	137.25	358.42	3196.9	±3.1
21.92	338.29	677.5	±4.6	137.22	363.03	3291.6	±3.3
	$\varrho/\varrho_c = 0.196$				$\varrho/\varrho_c = 0.361$		
84.64	313.22	1779.2	±2.3	155.66	335.71	2865.9	±1.8
84.62	318.22	1841.5	±2.5	155.65	336.96	2898.9	±1.9
84.59	323.27	1902.6	±2.7	155.64	338.61	2941.3	±2.0
84.57	328.30	1964.5	±2.9	155.60	342.70	3044.1	±2.2
84.55	333.29	2021.5	±3.0	155.59	344.74	3094.4	±2.3
84.53	338.35	2082.5	±3.2	155.56	347.73	3168.6	±2.4
84.50	343.43	2141.1	±3.4	155.56	347.76	3169.3	±2.4
84.48	348.39	2197.5	±3.5	155.54	350.69	3241.3	±2.5
84.46	353.15	2250.0	±3.6	155.51	353.80	3316.1	±2.7
84.46	353.47	2254.4	±3.7	155.49	356.79	3387.7	±2.8
84.44	358.65	2311.2	±3.8	155.44	362.84	3532.6	±3.1
84.42	363.59	2368.3	±4.0				
	$\varrho/\varrho_c = 0.261$				$\varrho/\varrho_c = 0.996$		
112.61	323.36	2239.6	±2.1	429.18	345.92	3768.3	±4.4
112.59	326.39	2291.8	±2.2	429.18	346.01	3775.8	±4.4
112.57	329.91	2352.8	±2.4	429.18	346.11	3783.4	±4.4
112.55	333.38	2412.1	±2.5	429.18	346.14	3786.2	±4.4
112.52	338.31	2494.5	±2.7	429.18	346.29	3797.7	±4.4
112.49	343.31	2576.5	±2.9	429.13	348.25	3956.7	±4.4
112.47	347.42	2643.8	±3.0	429.07	350.47	4136.3	±4.7
112.43	353.39	2740.0	±3.3				
112.41	358.45	2821.5	±3.4				
112.38	363.32	2897.9	±3.6				

(above 200 K). A difference between the present results and the earlier values (Giuliani et al., 1994) is in the purity of the sample. The two sets agree well, and the differences are larger only at lower temperatures (below 250 K) and near the critical temperature. Fukushima (1993) reports an uncertainty of ±5 kPa, and except one value (close to the critical temperature), all are within his uncertainty and his critical temperature is higher. The results of Widiatmo et al. (1994) are higher than the present results but within the ±10 kPa uncertainty reported by them, while the results of Higashi (1994) are lower than the present correlation but within his uncertainty of ±0.5%. Extrapolation of eq 11 to temperatures between 200 and 225 K and comparison with the results of Russell et al. (1944) (above 200 K) show a maximum deviation of less than 1 kPa (±0.6%), while it is higher at lower temperatures. The correlations of Arnaud et al. (1991) and Mears et al. (1955) have also been drawn, which also show good agreement when the uncertainties and the deviation between their equation and their data are considered. Equation 11 is, therefore, expected to be valid from 200 K to the critical temperature.

PVT. Sixty-three gas phase densities for R143a along six isochores are presented in Table 3 with the critical density taken from Higashi (1994) as 431 kg·m^{-3}. The uncertainty in the pressure from eq 5 for each triplet is also given. The range of the density is from 21 to 429 kg·m^{-3}, temperature is from 268 to 363 K, and pressure is from 0.50 to 4.13 MPa.

The gas phase data have been used to fit the Martin–Hou equation of state (Martin and Hou, 1955) given below:

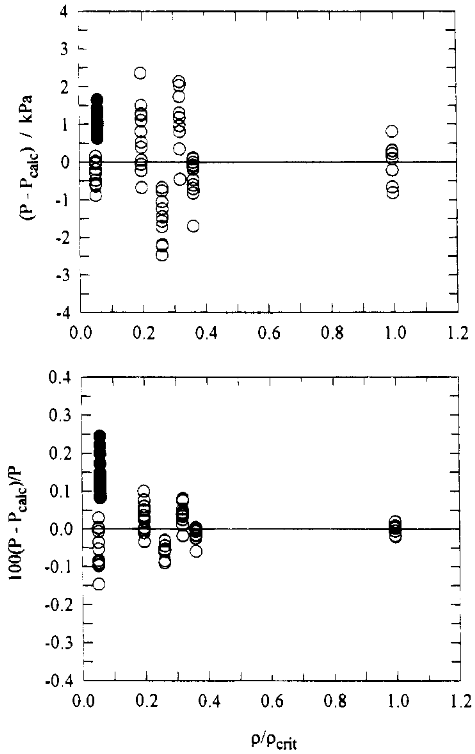

Figure 4. Pressure deviations of R143a from the Martin–Hou equation of state (eq 12): (○) this work, (●) Giuliani et al. (1994).

$$P/\text{kPa} = \frac{RT}{(v-b)} + \frac{A_2 + B_2T + C_2e^{-kT_r}}{(v-b)^2} + \frac{A_3 + B_3T + C_3e^{-kT_r}}{(v-b)^3} + \frac{A_4}{(v-b)^4} + \frac{A_5 + B_5T + C_5e^{-kT_r}}{(v-b)^5} \tag{12}$$

with $T_r = T/T_c$, R = 0.098 932 6 kPa·m^3·kg^{-1} · K^{-1}, b = 0.000 404 6 m^3·kg^{-1}, and k = 5.475.

$A_2 = -0.1812$ $\quad B_2 = 1.9587 \times 10^{-4}$ $\quad C_2 = -3.7592$
$A_3 = 3.6953 \times 10^{-4}$ $\quad B_3 = -3.6648 \times 10^{-7}$ $\quad C_3 = 7.7033 \times 10^{-3}$
$A_4 = -3.7831 \times 10^{-7}$
$A_5 = 1.8594 \times 10^{-10}$ $\quad B_5 = 3.1761 \times 10^{-13}$ $\quad C_5 = -8.4211 \times 10^{-9}$

Equation 12 is based on data from $0.05\varrho_c$, to $0.99\varrho_c$ and the fit between the data and the equation is good as shown by the following statistical parameters: AAD = 0.0389; bias = −0.007 47, and σ = 1.077 kPa. Figures 4 and 5 show the deviation in pressure between the results and eq 12 as a function of reduced density and temperature. Out of 63 data covering a range from $0.05\varrho_c$ to nearly the critical density, the maximum deviation is 0.15%. Arnaud et al. (1991) have given a Martin–Hou equation of state based on data having a density range between $0.06\varrho_c$ and $0.26\varrho_c$, and a comparison between their correlation and eq 12 shows that the maximum deviation is 2.3%. Considering their uncertainties and the difference between their data and their correlation, the agreement with the present study is good. Mears et al. (1955) made measurements between $0.11\varrho_c$ and $0.49\varrho_c$. Their correlation shows a systematic deviation on the order of 3–4% when compared with eq 12, but their uncertainties in pressure, temperature, and volume measurements are thought to be higher. Arnaud et al. (1991) also show higher deviation when comparing their data with those of Mears et al. (1955). Besides, the limited data used to fit the equation of state are also probably the cause for such large deviations. This systematic difference is also evident when a comparison with the

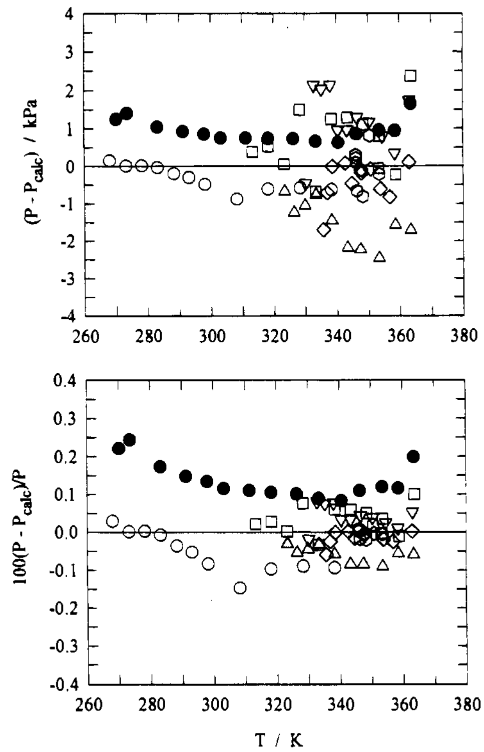

Figure 5. Pressure deviations of R143a from the Martin–Hou equation of state (eq 12): (○) $\varrho/\varrho_c = 0.051$, (□) $\varrho/\varrho_c = 0.196$, (△) $\varrho/\varrho_c = 0.261$, (▽) $\varrho/\varrho_c = 0.319$, (◇) $\varrho/\varrho_c = 0.361$, (○) $\varrho/\varrho_c = 0.996$, (●) Giuliani et al. (1994) ($\varrho/\varrho_c = 0.057$).

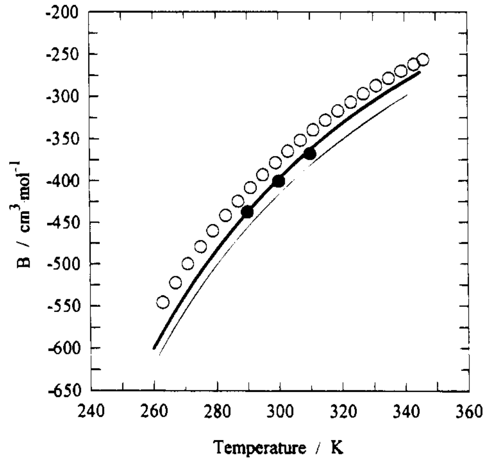

Figure 6. Second virial coefficients of R143a: (●) Bignell and Dunlop (1993), (○) Smith and Srivastava (1986), (thin line) Weber (1994), (thick line) this work.

second virial coefficient is made, as shown later. A comparison with one data series of Giuliani et al. (1994) shows a maximum deviation of 0.24%.

Second Virial Coefficients. The virial equation of state truncated to the third term was fitted to the observed density results. The temperature dependence of the second virial coefficient can be described (Tillner-Roth and Baehr, 1992) by

$$\frac{B(\tau)}{B_0} = b_1\tau + b_2\tau^{1.5} + b_3\tau^{2.5} \qquad (13)$$

where $B_0 = 1$ cm^3·mol^{-1} and $\tau = T_c/T$ with $b_1 = -6.6991 \times 10^{-4}$, $b_2 = 1.0381 \times 10^{-3}$, $b_3 = -6.3778 \times 10^{-4}$. and shown in Figure 6. Three measured second virial coefficients by Bignell and Dunlop (1993) at 290, 300, and 310 K have also been given. The differences between eq 13 and the measured values are 0.16, 0.75, and 1.4%, respectively, indicating the thermodynamic consistency of our measured data. The same Figure 6 shows the comparison with derived second virial coefficients due to Smith and Srivastava (1986) (who used data from Mears et al. (1955)) and to Weber (1994).

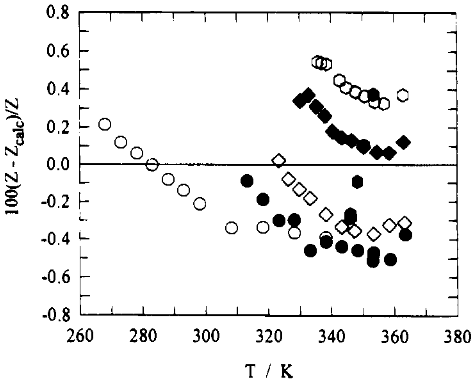

Figure 7. Deviations of Z from eq 14: (○) $\varrho/\varrho_c = 0.051$, (●) $\varrho/\varrho_c = 0.196$, (◇) $\varrho/\varrho_c = 0.261$, (◆) $\varrho/\varrho_c = 0.319$, (○) $\varrho/\varrho_c = 0.361$, («) $\varrho/\varrho_c = 0.996$.

The truncated virial equation of state has the form

$$Z = 1 + F_1(\tau)\delta + F_2(\tau)\delta^2 \qquad (14)$$

with $Z = (PM)/(\varrho RT)$, where $M = 0.084\,041$ kg·mol^{-1}, $\delta = \varrho/\varrho_0$ with $\varrho_0 = 1$ kg·m^{-3}, and $R = 0.008\,314\,471$ kJ·K^{-1}·mol^{-1}. $F_1(\tau)$ and $F_2(\tau)$ are functions of temperature given by

$$F_1(\tau) = B(\tau)\varrho_0/M \qquad (15)$$

and

$$F_2(\tau) = C(\tau)\varrho_0/M \qquad (16)$$

with $B(\tau)$ from eq 13 and $C(\tau)$ from eq 17 as

$$C(\tau) = c_1\tau^3 + c_2\tau^5 \qquad (17)$$

with $c_1 = 2.6244 \times 10^{-7}$ and $c_2 = 2.668\,8 \times 10^{-8}$.

The deviations of Z between the data and eq 14 have been drawn in Figure 7. The absolute average deviation is 0.28%, and the maximum deviation is 0.56%; in terms of the compression factor Z, the average deviation is $0.0018Z$ and the greatest deviations are $+0.0031Z$ and $-0.0039Z$.

Conclusions

New accurate data on the vapor pressure and gas phase PVT of refrigerant R143a have been presented with a Wagner type correlation for vapor pressure and a Martin–Hou type of equation of state for PVT behavior. The data and correlations have been compared with existing data in the literature. Consistency checks on the gas phase PVT data have been done by comparison with the second virial coefficients.

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