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PVT Properties for 1,1,1-Trifluoroethane (R-143a)

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Densities of 1,1,1-trifluoroethane (R-143a) have been measured using a vibrating-tube densitometer over a density range from 41 kg·m^{-3} to 1113 kg·m^{-3} along 15 isotherms between 263 K and 403 K and at pressures between 1.5 MPa and 15 MPa. The experimental uncertainties of the measurements are estimated to be ±5 kPa in pressure, ±11 mK in temperature, and ±0.3% to ±2% in density.

Introduction

Binary mixtures of pentafluoroethane (R-125) and 1,1,1-trifluoroethane (R-143a) and ternary mixtures of R-125, 1,1,1,2-tetrafluoroethane (R-134a), and R-143a have been considered as alternatives to R-502. *PVT* properties of R-143a have been reported by Giuliani et al. (1995) and Zhang et al. (1995). The data range is for densities between 4 kg·m^{-3} and 620 kg·m^{-3}, temperatures between 270 K and 380 K, and pressures between 0.1 MPa and 6.1 MPa. There are no values published over a range of density greater than 620 kg·m^{-3}. We have measured the *PVT* properties of R-143a over a density range from 41 to 1113 kg·m^{-3} along 15 isotherms between 263 K and 403 K and at pressures between 1.5 MPa and 15 MPa.

Theory

A vibrating U-shaped tube was completely filled with a sample. The density of the sample was determined on the basis of measurements of the frequency of oscillation of the tube. The following relation exists between the frequency and the density:

$$\rho = A/f^2 + B \tag{1}$$

where

$$A = c/4\pi^2 V \tag{2}$$

$$B = M/V \tag{3}$$

c is the spring constant of the tube, f is the frequency, M is the weight of the tube, V is the inner volume of the tube, and ρ is the density of the sample.

The constants A and B are the apparatus constants, and the values were determined by calibrations with two fluids of known density: nitrogen and R-134a.

Experimental Apparatus and Procedure

A schematic diagram of the apparatus is shown in Figure 1. A stainless steel vibrating U-tube densitometer (A; Anton Paar DMA 512) was connected to a quartz pressure transducer (B; Paroscientific Digiquartz 4000 series), a metallic bellows variable-volume vessel (C), a circulating pump (D), and a hand pressure controller (E). The U-tube with inner volume of approximately 1 cm^3 was housed in a brass housing enclosed in a steel heat exchanger. Two tubes connected the heat exchanger with an external temperature-controlled circulating bath (F). Approximately 50 L of silicon oil was contained in the bath. The oil was circulated through the heat exchanger from the bath. The inner volumes of the bellows and the pressure controller were variable from 85 cm^3 to 130 cm^3 and up to 20 cm^3, respectively. Valves, V1 and V2, were closed to measure the frequency of the U-tube; V3 and V4, equipped with the bellows, served as an inlet and an outlet, respectively; V5 was closed to circulate the samples, R-134a and R-143a; and V6 was opened to evacuate the U-tube or introduce nitrogen. The densitometer, the pressure transducer, the valves, V1 and V2, and pipes connecting the instruments with the valves were encased in foam insulation.

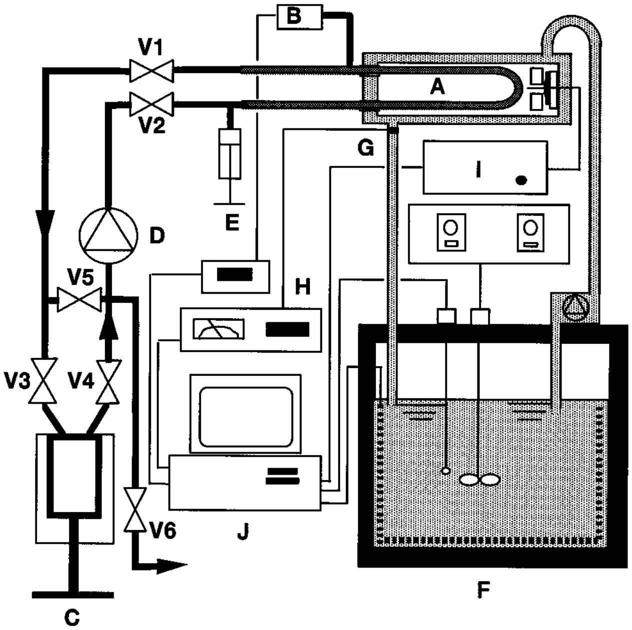

Figure 1. Schematic diagram of the experimental apparatus: (A) vibrating densitometer, (B) pressure transducer, (C) bellows vessel, (D) circulating pump, (E) pressure controller, (F) thermostatic bath, (G) platinum resistance thermometer, (H) resistance bridge, (I) frequency counter, (J) personal computer, (V) valve.

The pressure was measured using the pressure transducer. The transducer was calibrated using an oil-operated dead weight gauge.

A platinum resistance thermometer (G; Minco Products Model S1059PJ5X20) was installed near the outlet of the heat exchanger. The temperature was measured using this thermometer and a thermometer bridge (H; Automatic System Laboratories Model F700). The thermometer was calibrated at fixed points at the National Institute of Standards and Technology (NIST). The resistance of water at the triple point was measured to establish the resistance at 273.16 K. The temperatures were given using the ITS-

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Table 1. Uncertainty Analysis in Measurements

(a) Uncertainty of Pressure Measurements in kPa		
accuracy of transducer	±4	
variation	±1	
total	±5	
(b) Uncertainty of Temperature Measurements in mK		
accuracy of thermometer	±0.5	
accuracy in bridge	±0.5	
control	±5	
distribution	±5	
total	±11	
(c) Uncertainty of Frequency Measurements in Hz		
accuracy in counter	±0.0001	
variation	±0.001	
total	±0.0011	
(d) Uncertainty of Eq 6 in kg·m^{-3}		
density range in kg·m^{-3}	$4 < \rho \le 194$	$194 < \rho < 1370$
uncertainty of equation of state	±0.2	±2.7
representation of eq 6	±0.5	±1.0
total	±0.7	±3.7
(e) Uncertainty of Density for R-143a		
density range in kg·m^{-3}	$41 < \rho \le 194$	$194 < \rho < 1113$
uncertainty in pressure measurements	±0.0008	
uncertainty in temperature measurements	±0.03	
uncertainty in frequency measurements	±0.07	
uncertainty of eq 6	±0.7	±3.7
total	±0.80008	±3.8008

90 temperature scale. The temperature glides of the inlet and outlet in the heat exchanger were obtained to be within ±5 mK at room temperature or so; therefore, the temperature distribution in the U-tube was estimated to be within ±5 mK and the sample temperature was regarded as the outlet temperature.

The frequency of oscillation of the U-tube was measured using a frequency counter (I).

The pressure, temperature, and frequency values were recorded automatically with a personal computer (J) through the interface. The temperature data inputted into the personal computer were used to control the oil temperature.

The accuracy values of the pressure transducer, thermometer, bridge, and frequency counter are shown in Table 1a−c.

Before the sample was introduced into the U-tube, the tube was evacuated close to 10^{-3} Pa. Nitrogen was introduced directly into the tube. In the case of R-134a and R-143a, the sample was charged in the bellows vessel and then introduced into the U-tube. The circulating pump was used so that the sample was smoothly introduced into the U-tube. In the case where the sample was not in the compressed liquid state, it was compressed using the bellows vessel and/or the pressure controller. After thermal equilibration, the frequency of the U-tube was measured using the counter at the desired pressure. The pressure was adjusted with the pressure controller and varied within ±1 kPa during the measurement. The oil temperature was maintained at ±5 mK. The frequency varied within ±0.001 Hz during the measurement at a given temperature and pressure.

Results

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

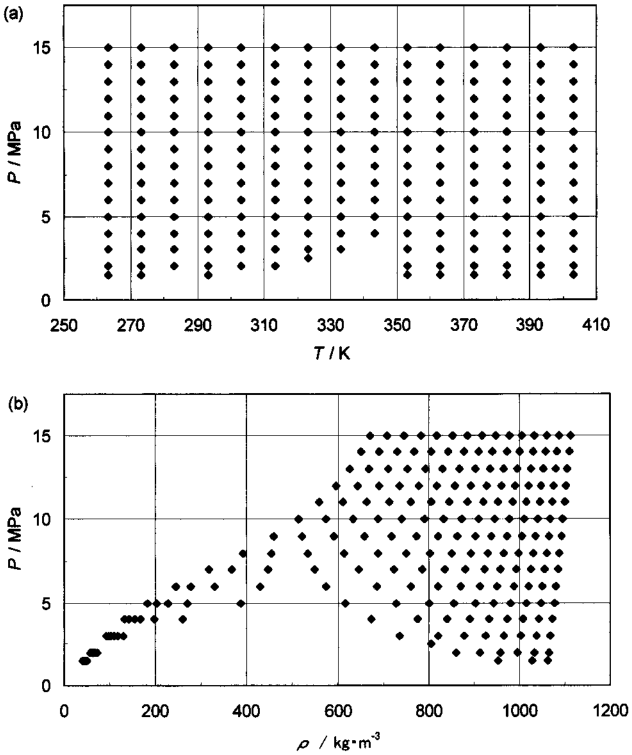

Figure 2. Experimental range for R-143a measurements.

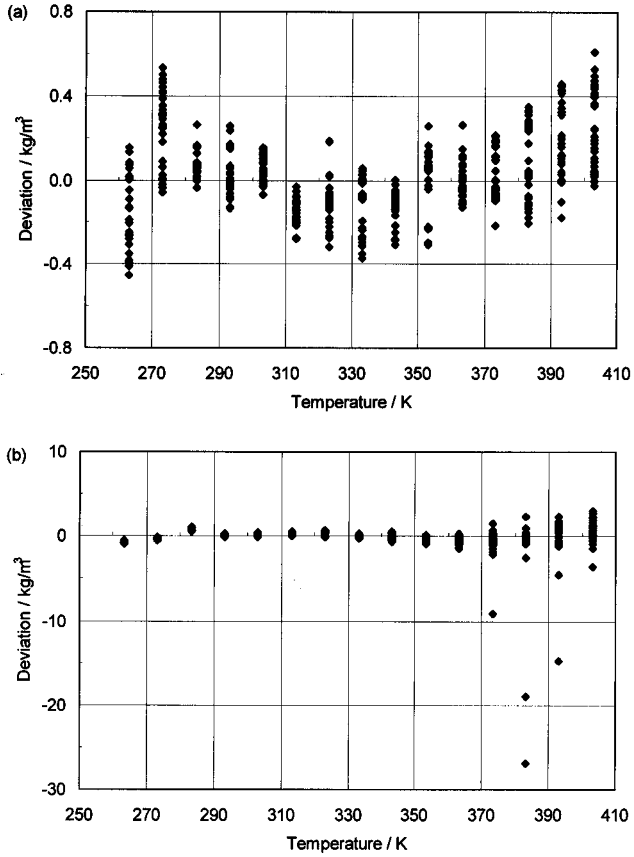

Figure 3. Comparison of nitrogen (a) and R-134a (b) density values calculated from eq 6 with equations of state (EOS); $\delta\rho = \rho_{\text{EOS}} - \rho_{\text{eq 6}}$.

The uncertainties of the measurements in pressure, temperature, and frequency are shown in Table 1a−c, respectively.

The frequencies of nitrogen and R-134a were measured along 15 isotherms between 263 K and 403 K and 30

Table 2. Experimental Results of R-143a

P/MPa	ρ/kg·m^{-3}	T/K	P/MPa	ρ/kg·m^{-3}	T/K
15.000	1112.6	263.15	7.000	1085.2	263.15
14.000	1109.5	263.15	6.000	1081.4	263.15
13.000	1106.2	263.15	5.000	1077.4	263.15
12.000	1103.0	263.15	4.000	1073.1	263.15
11.000	1099.6	263.15	3.000	1068.9	263.15
10.000	1096.2	263.15	2.000	1064.3	263.15
9.000	1092.7	263.15	1.500	1062.1	263.15
8.000	1089.0	263.15			
15.000	1086.9	273.15	7.000	1056.0	273.15
14.000	1083.4	273.15	6.000	1051.5	273.15
13.000	1079.7	273.15	5.000	1046.9	273.15
12.000	1076.1	273.15	4.000	1042.1	273.15
11.000	1072.3	273.15	3.000	1037.0	273.15
10.000	1068.4	273.15	2.000	1031.8	273.15
9.000	1064.5	273.15	1.500	1028.9	273.15
8.000	1060.4	273.15			
15.000	1060.8	283.15	8.000	1030.3	283.15
14.000	1056.7	283.15	7.000	1025.3	283.15
13.000	1052.7	283.15	6.000	1020.1	283.15
12.000	1048.5	283.15	5.000	1014.6	283.15
11.000	1044.2	283.15	4.000	1008.8	283.15
10.000	1039.8	283.15	3.000	1002.7	283.15
9.000	1035.1	283.15	2.000	996.2	283.15
15.000	1033.6	293.15	7.000	992.9	293.15
14.000	1029.1	293.15	6.000	986.8	293.15
13.000	1024.6	293.15	5.000	980.1	293.15
12.000	1019.8	293.15	4.000	973.1	293.15
11.000	1015.0	293.15	3.000	965.6	293.15
10.000	1009.9	293.15	2.000	957.4	293.15
9.000	1004.5	293.15	1.500	953.0	293.15
8.000	998.8	293.15			
15.000	1006.0	303.15	8.000	965.8	303.15
14.000	1001.0	303.15	7.000	958.8	303.15
13.000	995.8	303.15	6.000	951.2	303.15
12.000	990.4	303.15	5.000	943.0	303.15
11.000	984.8	303.15	4.000	934.2	303.15
10.000	978.8	303.15	3.000	924.4	303.15
9.000	972.5	303.15	2.000	913.6	303.15
15.000	977.6	313.15	8.000	930.5	313.15
14.000	972.0	313.15	7.000	921.8	313.15
13.000	966.0	313.15	6.000	912.4	313.15
12.000	959.7	313.15	5.000	902.1	313.15
11.000	953.1	313.15	4.000	890.5	313.15
10.000	946.0	313.15	3.000	877.2	313.15
9.000	938.6	313.15	2.000	861.4	313.15
15.000	948.2	323.15	8.000	892.5	323.15
14.000	941.4	323.15	7.000	881.8	323.15
13.000	935.0	323.15	6.000	869.7	323.15
12.000	927.7	323.15	5.000	855.9	323.15
11.000	919.8	323.15	4.000	839.5	323.15
10.000	911.4	323.15	3.000	819.6	323.15
9.000	902.4	323.15	2.500	806.3	323.15
15.000	918.0	333.15	8.000	850.8	333.15
14.000	910.5	333.15	7.000	836.7	333.15
13.000	902.4	333.15	6.000	820.3	333.15
12.000	893.7	333.15	5.000	800.4	333.15
11.000	884.5	333.15	4.000	775.0	333.15
10.000	974.3	333.15	3.000	735.1	333.15
9.000	863.2	333.15			
15.000	886.4	343.15	9.000	820.1	343.15
14.000	877.6	343.15	8.000	804.0	343.15
13.000	868.1	343.15	7.000	784.9	343.15
12.000	857.8	343.15	6.000	761.1	343.15
11.000	846.6	343.15	5.000	728.6	343.15
10.000	834.1	343.15	4.000	673.0	343.15
15.000	853.4	353.15	7.000	723.5	353.15
14.000	843.1	353.15	6.000	685.4	353.15
13.000	831.9	353.15	5.000	615.1	353.15
12.000	819.5	353.15	4.000	260.0	353.15
11.000	805.8	353.15	3.000	130.5	353.15
10.000	790.3	353.15	2.000	72.2	353.15
9.000	772.4	353.15	1.500	50.5	353.15
8.000	750.6	353.16			

Table 2 (Continued)

P/MPa	ρ/kg·m^{-3}	T/K	P/MPa	ρ/kg·m^{-3}	T/K
15.000	819.0	363.15	7.000	646.7	363.15
14.000	806.9	363.15	6.000	574.0	363.15
13.000	793.5	363.15	5.000	386.8	363.15
12.000	778.5	363.15	4.000	197.4	363.15
11.000	761.7	363.15	3.000	118.4	363.15
10.000	742.2	363.15	2.000	68.1	363.15
9.000	718.5	363.15	1.500	48.2	363.15
8.000	688.5	363.15			
15.000	783.5	373.15	7.000	548.8	373.15
14.000	769.4	373.15	6.000	428.3	373.15
13.000	753.5	373.15	5.000	269.6	373.15
12.000	735.6	373.15	4.000	166.9	373.15
11.000	714.7	373.15	3.000	109.4	373.15
10.000	689.5	373.15	2.000	64.6	373.15
9.000	657.8	373.15	1.500	46.1	373.15
15.000	747.0	383.15	7.000	445.6	383.15
14.000	730.6	383.15	6.000	328.4	383.15
13.000	711.9	383.15	5.000	226.8	383.15
12.000	690.3	383.15	4.000	154.7	383.15
11.000	664.5	383.15	3.000	102.3	383.15
10.000	632.6	383.15	2.000	61.6	383.15
9.000	591.3	383.15	1.500	44.1	383.15
8.000	533.7	383.15			
15.000	709.6	393.15	7.000	367.0	393.15
14.000	690.5	393.15	6.000	276.4	393.15
13.000	668.6	393.15	5.000	201.0	393.15
12.000	642.8	393.15	4.000	142.5	393.15
11.000	611.9	393.15	3.000	96.4	393.15
10.000	573.0	393.15	2.000	58.8	393.15
9.000	522.5	393.15	1.500	42.3	393.15
8.000	454.0	393.15			
15.000	671.8	403.15	7.000	315.8	403.15
14.000	650.1	403.15	6.000	244.2	403.15
13.000	625.0	403.15	5.000	183.2	403.15
12.000	595.3	403.15	4.000	133.0	403.15
11.000	559.6	403.15	3.000	91.5	403.15
10.000	515.1	403.15	2.000	56.3	403.15
9.000	459.5	403.15	1.500	40.5	403.15
8.000	391.2	403.15			

isobars between 0.5 MPa and 15 MPa. The density range for nitrogen was from 4 kg·m^{-3} to 194 kg·m^{-3}; that for R-134a was from 16 kg·m^{-3} to 1370 kg·m^{-3}.

The A and B values were calculated on the basis of the frequency measurements and densities calculated from equations of state. The equations of state have been proposed by Jacobsen and Stewart (1973) for nitrogen and by Piao et al. (1994) for R-134a. The A and B values tended to depend on temperature and pressure; so we set the A and B values to correlate with the A and B values in the following equations:

$$A = a_1 + a_2T + a_3T^2 + (a_4 + a_5T + a_6T^2)P \qquad (4)$$

$$B = b_1 + b_2T + b_3T^2 + (b_4 + b_5T + b_6T^2)P \qquad (5)$$

Equations 4 and 5 represented the A and B values within ±0.3%.

Substituting eqs 4 and 5 for A and B in eq 1, the following equation can be obtained:

$$\rho = [a_1 + a_2T + a_3T^2 + (a_4 + a_5T + a_6T^2)P]/f^2 + [b_1 + b_2T + b_3T^2 + (b_4 + b_5T + b_6T^2)P] \qquad (6)$$

We calculated the densities of the samples from eq 6, which was useful for calculating densities at desired temperatures and pressures.

A total of 216 points of densities for R-143a were measured over a density range from 41 kg·m^{-3} to 1113 kg·m^{-3} along 15 isotherms between 263 K and 403 K and pressures between 1.5 MPa and 15 MPa. These data are tabulated in Table 2. Figure 2 shows the distribution of the PVT data.

Discussion

Table 1d shows the uncertainty of eq 6, which includes the uncertainty of the equation of state and representation of the density values from the equation of state. In the density range from 14 kg·m^{-3} to 194 kg·m^{-3}, the uncertainty of eq 6 was estimated depending on the nitrogen results, and in the range from 194 kg·m^{-3} to 1370 kg·m^{-3}, it was estimated depending on the R-134a results. The R-134a results in the supercritical regions were not taken into account because the equation of state was not as accurate in this region as in the other regions: vapor and liquid phases. Jacobsen and Stewart (1973) stated that the uncertainty of the equation of state was estimated to be within ±0.1%, equivalent to ±0.2 kg·m^{-3}. According to the paper by Piao et al. (1994), the uncertainties of the equation of state for R134a were estimated to be within ±0.5% in the vapor phase and within ±0.2% in the liquid phase, equivalent to ±2.7 kg·m^{-3}. Parts a and b of Figure 3 show the comparisons of the density values calculated from eq 6 and from the equation of states for nitrogen and R-134a, respectively. Equation 6 represented the nitrogen values calculated from the equation of state within ±0.5 kg·m^{-3}. It represented the R-134a values within ±1 kg·m^{-3} except for the data in the supercritical region. Therefore, the uncertainty of eq 6 was calculated to be within ±0.7 kg·m^{-3} in the density range from 14 kg·m^{-3}

to 194 kg·m^{-3} and to ±3.7 kg·m^{-3} from 194 kg·m^{-3} to 1370 kg·m^{-3}.

Table 1e shows the uncertainty of R-143a density values, which include the uncertainties derived from the measurements in pressure, temperature, and frequency. It also includes the uncertainty of eq 6. The density uncertainties derived from the measurements in pressure, temperature, and frequency were estimated to be ±0.0008 kg·m^{-3}, ±0.03 kg·m^{-3}, and ±0.07 kg·m^{-3}. Therefore, the uncertainties of the density values were calculated to be within ±0.8 kg·m^{-3} in the density range up to 194 kg·m^{-3} and ±3.8 kg·m^{-3} up to 1113 kg·m^{-3}. Those were equivalent to ±0.3% to ±2% in the density range from 41 kg·m^{-3} to 1113 kg·m^{-3}.

Conclusions

A total of 216 densities for R-143a were measured using a vibrating U-tube densitometer over a density range from 41 kg·m^{-3} to 1113 kg·m^{-3} along 15 isotherms between 263 K and 403 K and at pressures between 1.5 MPa and 15 MPa. The uncertainty of the density measurement was estimated to be ±0.3% to ±2%.

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