

Liquid Densities of HCFC 225ca, HCFC 225cb, and HCFC 141b

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Liquid densities of HCFC 225ca (1,1-dichloro-2,2,3,3,3-pentafluoropropane), HCFC 225cb (1,3-dichloro-1,2,2,3,3-pentafluoropropane), and HCFC 141b (1,1-dichloro-1-fluoroethane) have been measured at temperatures from 298 to 348 K by a vibrating-tube method. Pressure ranges of the measurements are up to 5 MPa for HCFC 225ca and HCFC 225cb and up to 100 MPa for HCFC 141b. The uncertainty of the densities obtained is estimated to be within $\pm 0.1\%$. Simple correlation equations were developed to represent the saturated liquid densities as a function of temperature. Comparisons of the volumetric properties for the present possible alternatives and those of restricted substances CFC 113 (1,1,2-trichloro-1,2,2-trifluoroethane) and CFC 11 (trichlorofluoromethane) are given.

Introduction

In previous studies, we have reported volumetric properties and viscosities of aqueous solutions of three fluoroalcohols, i.e., 2,2,2-trifluoroethanol, 2,2,3,3-tetrafluoropropanol, and 2,2,3,3,3-pentafluoropropanol (Matsuo et al., 1993; Matsuo et al., 1994). These aqueous solutions appear to be favorable working fluids for waste-heat recovery and possible alternatives for the restricted substance CFC 113 (1,1,2-trichloro-1,2,2-trifluoroethane) which has been widely used as a cleaning agent. In the present work, the liquid densities of some other possible alternatives have been measured, i.e., HCFC 225ca (1,1-dichloro-2,2,3,3,3-pentafluoropropane) and HCFC 225cb (1,3-dichloro-1,2,2,3,3-pentafluoropropane) for CFC 113 and HCFC 141b (1,1-dichloro-1-fluoroethane) for CFC 11 (trichlorofluoromethane) which has been used as a blowing agent for insulating form. There exist very few reliable measurements on thermophysical properties for these fluids (Kumagai and Takahashi, 1993; Defibaugh et al., 1993; Takagi and Hongo, 1993), and a rapid accumulation of accurate data over wide temperature and pressure ranges is required. In the present investigation, density measurements were performed by a vibrating-tube method in the temperature range from 298 to 323 K. Pressure ranges are largely different among the substances, i.e., up to 5 MPa for HCFC 225ca and HCFC 225cb and up to 100 MPa for HCFC 141b. This paper also reports the saturated liquid densities which were determined by the extrapolation of experimental compressed liquid densities along the isotherms to their saturation lines. Viscosity and dielectric constant measurements for HCFC 225ca and HCFC 225cb have also been performed (Tanaka et al., 1994).

Experimental Section

Measurements were performed with two vibrating-tube densitometers, i.e., a commercial one (Paar DMA602) with a pressure limit of 5 MPa and a newly constructed one for measurements in the higher pressure range. The former instrument was mainly used for the purpose of determining the saturated liquid densities and the latter one for the determination of the pressure effects on volumetric properties for HCFC 141b. The principles of measurement for

the two densitometers are basically the same, and the apparatus and experimental procedure are described in detail elsewhere (Matsuo and Makita, 1989; Matsuo et al., 1994). Each densitometer cell is jacketed and maintained isothermally to ± 10 mK by circulating water, and the temperature was measured with a precise thermistor (TECNOL SEVEN E640) to within ± 10 mK. Pressure lower than 10 MPa was measured with a precise pressure calibrator (VAISALA PM210) whose precision should be within $\pm 6 \times 10^{-4}$ MPa at pressures lower than 1 MPa and within $\pm 1 \times 10^{-2}$ MPa in the range from 1 to 10 MPa. Pressure higher than 10 MPa was measured with a digitized Bourdon gauge (HEISE 901B) to within ± 0.2 MPa.

The densitometers were used on a relative basis, and the instrument constants for the Paar DMA602 were calibrated against the recommended densities of pure water (Kestin and Sengers, 1986) and pure nitrogen (Jacobsen and Stewart, 1973), while those for the newly constructed one were calibrated against the recommended densities of pure water (Kestin and Sengers, 1986) and pure heavy water (Chen and Millero, 1981) over the entire range of temperature and pressure.

To obtain the saturated liquid density by the extrapolation method, the compressed liquid densities were measured along isotherms at temperatures from 298 to 348 K and at pressures up to 0.8 MPa. Temperature and pressure intervals of the measurements were 5 K and 0.1 MPa, and 92, 92, and 82 values were obtained for HCFC 225ca, HCFC 225cb, and HCFC 141b, respectively. Additional measurements were performed at pressures up to 5 MPa for HCFC 225ca and HCFC 225cb and up to 100 MPa for HCFC 141b for the purpose of examining the pressure dependence of the volumetric properties. These supplementary measurements were performed at three representative temperatures of 298, 323, and 348 K, and 32, 32, and 40 values were obtained for HCFC 225ca, HCFC 225cb, and HCFC 141b, respectively.

HCFC 225ca and HCFC 225cb with guaranteed purity of more than 99.95 mass % were obtained from Asahi Glass Co. Ltd., while HCFC 141b with guaranteed purity of more than 99.9 mass % was obtained from Daikin Industries, Ltd., and they were used without any purification. Judging from the precision of the instruments, the purity of the samples, and the reproducibility of the measurements, we estimate the uncertainties of the densities obtained to be within $\pm 0.1\%$.

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Table 1. Density of Liquid HCFC 225ca at Pressures up to 0.8 MPa

P/MPa	$\rho/(\text{kg}\cdot\text{m}^{-3})$					
	298.15 K	303.15 K	308.15 K	313.15 K	318.15 K	323.15 K
	1550.25 (0.0616) ^a	1538.02 (0.0643) ^a	1525.61 (0.0684) ^a	1513.17 (0.0772) ^a	1500.52 (0.0888) ^a	
0.1000	1550.39	1538.14	1525.72	1513.25	1500.56	1487.75
0.2000	1550.71	1538.47	1526.06	1513.61	1500.94	1488.16
0.3000	1551.02	1538.79	1526.40	1513.97	1501.31	1488.56
0.4000	1551.33	1539.11	1526.74	1514.33	1501.69	1488.95
0.5000	1551.64	1539.44	1527.09	1514.68	1502.07	1489.36
0.6000	1551.95	1539.76	1527.42	1515.04	1502.45	1489.76
0.7000	1552.26	1540.08	1527.76	1515.40	1502.82	1490.14
0.8000	1552.56	1540.41	1528.10	1515.75	1503.19	1490.55

P/MPa	$\rho/(\text{kg}\cdot\text{m}^{-3})$				
	328.15 K	333.15 K	338.15 K	343.15 K	348.15 K
	1474.86 (0.1252) ^a	1461.69 (0.1409) ^a	1448.26 (0.1618) ^a	1434.68 (0.1841) ^a	
0.2000	1475.17	1461.94	1448.44	1434.76	1420.80
0.3000	1475.59	1462.38	1448.91	1435.26	1421.33
0.4000	1476.01	1462.83	1449.38	1435.77	1421.86
0.5000	1476.43	1463.27	1449.85	1436.26	1422.36
0.6000	1476.85	1463.70	1450.32	1436.74	1422.89
0.7000	1477.26	1464.13	1450.79	1437.22	1423.39
0.8000	1477.67	1464.58	1451.24	1437.71	1423.90

^a Pressure in megapascals.**Table 2. Density of Liquid HCFC 225cb at Pressures up to 0.8 MPa**

P/MPa	$\rho/(\text{kg}\cdot\text{m}^{-3})$					
	298.15 K	303.15 K	308.15 K	313.15 K	318.15 K	323.15 K
	1555.98 (0.0618) ^a	1544.16 (0.0648) ^a	1532.18 (0.0666) ^a	1520.14 (0.0718) ^a	1507.93 (0.0827) ^a	
0.1000	1556.10	1544.27	1532.28	1520.23	1507.99	1495.59
0.2000	1556.39	1544.58	1532.60	1520.57	1508.35	1495.96
0.3000	1556.68	1544.88	1532.92	1520.91	1508.70	1496.32
0.4000	1556.98	1545.19	1533.24	1521.24	1509.05	1496.70
0.5000	1557.27	1545.50	1533.56	1521.57	1509.40	1497.05
0.6000	1557.57	1545.81	1533.88	1521.91	1509.74	1497.43
0.7000	1557.86	1546.11	1534.20	1522.22	1510.10	1497.79
0.8000	1558.14	1546.40	1534.51	1522.57	1510.44	1498.15

P/MPa	$\rho/(\text{kg}\cdot\text{m}^{-3})$				
	328.15 K	333.15 K	338.15 K	343.15 K	348.15 K
	1483.20 (0.1200) ^a	1470.51 (0.1401) ^a	1457.58 (0.1589) ^a	1444.49 (0.1801) ^a	
0.2000	1483.51	1470.78	1457.77	1444.64	1431.22
0.3000	1483.91	1471.18	1458.21	1445.10	1431.71
0.4000	1484.29	1471.59	1458.64	1445.55	1432.19
0.5000	1484.68	1471.97	1459.06	1446.00	1432.67
0.6000	1485.07	1472.40	1459.48	1446.44	1433.16
0.7000	1485.46	1472.79	1459.91	1446.88	1433.63
0.8000	1485.83	1473.18	1460.31	1447.33	1434.10

^a Pressure in megapascals.

Results and Discussion

The densities obtained with the Paar DMA602 densitometer for determining the saturated liquid densities are given in Tables 1–3. For each substance, the density at a given temperature increases monotonically with increasing pressure and can be represented by the following polynomial equation:

$$\rho = a_1 + a_2P + a_3P^2 \quad (1)$$

The maximum deviations ($\delta\rho/\rho$) for HCFC 225ca, HCFC 225cb, and HCFC 141b are 0.73×10^{-5} , 2.18×10^{-5} , and 1.27×10^{-5} , respectively, and are well within the experimental uncertainties.

In terms of eq 1, the saturated liquid densities were determined as the extrapolated values of the experimental

Table 3. Density of Liquid HCFC 141b at Pressures up to 0.8 MPa

P/MPa	$\rho/(\text{kg}\cdot\text{m}^{-3})$					
	298.15 K	303.15 K	308.15 K	313.15 K	318.15 K	323.15 K
	1233.88 (0.0891) ^a	1224.10 (0.1043) ^a	1214.23 (0.1217) ^a	1204.29 (0.1417) ^a	1194.13 (0.1644) ^a	1184.01 (0.1901) ^a
0.1000	1233.90					
0.2000	1234.15	1224.33	1214.44	1204.44	1194.23	1184.04
0.3000	1234.38	1224.59	1214.69	1204.70	1194.50	1184.33
0.4000	1234.61	1224.83	1214.95	1204.98	1194.79	1184.64
0.5000	1234.85	1225.07	1215.21	1205.26	1195.08	1184.93
0.6000	1235.08	1225.31	1215.47	1205.51	1195.36	1185.22
0.7000	1235.30	1225.56	1215.72	1205.77	1195.63	1185.50
0.8000	1235.55	1225.79	1215.97	1206.03	1195.91	1185.78

P/MPa	$\rho/(\text{kg}\cdot\text{m}^{-3})$				
	328.15 K	333.15 K	338.15 K	343.15 K	348.15 K
	1173.66 (0.2192) ^a	1163.14 (0.2519) ^a	1152.52 (0.2885) ^a	1141.69 (0.3293) ^a	1130.70 (0.3746) ^a
0.3000	1173.91	1163.29	1152.57		
0.4000	1174.22	1163.62	1152.91	1141.92	1130.80
0.5000	1174.53	1163.95	1153.24	1142.29	1131.20
0.6000	1174.84	1164.27	1153.60	1142.65	1131.61
0.7000	1175.14	1164.58	1153.93	1143.01	1131.97
0.8000	1175.44	1164.86	1154.26	1143.34	1132.34

^a Pressure in megapascals.**Table 4. Extrapolated Saturated Liquid Densities**

T/K	$\rho_{\text{sat}}/(\text{kg}\cdot\text{m}^{-3})$	T/K	$\rho_{\text{sat}}/(\text{kg}\cdot\text{m}^{-3})$	T/K	$\rho_{\text{sat}}/(\text{kg}\cdot\text{m}^{-3})$
HCFC 225ca					
298.15	1550.19	318.15	1500.50	338.15	1448.25
303.15	1537.97	323.15	1487.75	343.15	1434.69
308.15	1525.58	328.15	1474.82	348.15	1420.88
313.15	1513.15	333.15	1461.67		
HCFC 225cb					
298.15	1555.91	318.15	1507.91	338.15	1457.51
303.15	1544.10	323.15	1495.55	343.15	1444.45
308.15	1532.13	328.15	1483.14	348.15	1431.17
313.15	1520.11	333.15	1470.45		
HCFC 141b					
298.15	1233.85	318.15	1194.10	338.15	1152.50
303.15	1224.07	323.15	1183.98	343.15	1141.66
308.15	1214.20	328.15	1173.63	348.15	1130.67
313.15	1204.25	333.15	1163.11		

Table 5. Coefficients in Eq 2

	HCFC 225ca	HCFC 225 cb	HCFC 141b
T_c/K	478	484.85	477.3
$\rho_c/(\text{kg}\cdot\text{m}^{-3})$	558	557	461
d_1	2.174 267	2.182 532	2.034 890
d_2	-0.281 99	-0.285 21	-0.319 28
d_3	0.944 43	0.925 60	0.998 25

compressed liquid densities along the isotherms to the saturated vapor pressures. In this extrapolating procedure, the vapor pressure equations given by Widiatmo et al. (1992) were used for both HCFC 225ca and HCFC 225cb, while that given by Defibaugh et al. (1993) was used for HCFC 141b. The saturated liquid densities obtained are listed in Table 4 and are correlated with temperature with the following equation:

$$\rho_{\text{sat}}/\rho_c = 1 + d_1\tau^{1/3} + d_2\tau^{2/3} + d_3\tau \quad (2)$$

$$\tau = 1 - T/T_c$$

where ρ_{sat} and ρ_c are the saturated liquid density and critical density and T and T_c are the temperature and critical temperature, respectively. The coefficients of eq 2 are given in Table 5 together with the critical properties adopted in this correlation. The critical densities and temperatures reported by Fukushima and Watanabe (1992)

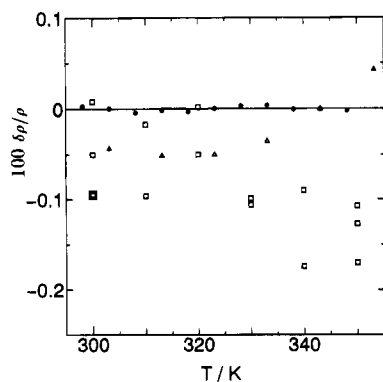


Figure 1. Deviations of saturated liquid densities of HCFC 225ca from eq 2: ●, this work; △, Kumagai; □, Widiatmo.

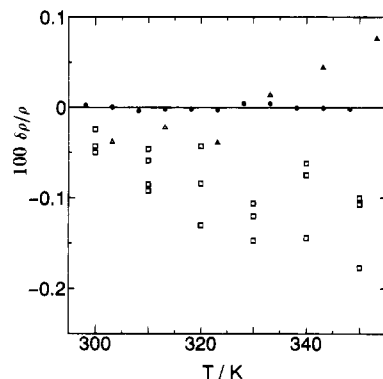


Figure 2. Deviations of saturated liquid densities of HCFC 225cb from eq 2: ●, this work; △, Kumagai; □, Widiatmo.

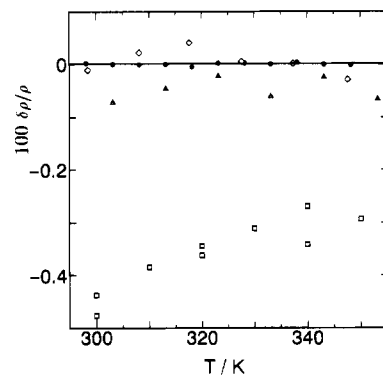


Figure 3. Deviations of saturated liquid densities of HCFC 141b from eq 2: ●, this work; △, Kumagai; ◇, Defibaugh; □, Maezawa.

and those by Chae et al. (1990) were adopted for HCFC 225cb and for HCFC 141b, respectively. Because of the absence of reliable experimental critical properties for HCFC 225ca, those estimated by Widiatmo et al. (1992) were used. The saturated vapor and saturated liquid densities of HCFC 225ca, which were measured near the critical point by Fukushima (1994), confirm the accuracy of these estimated values. The deviations of experimental saturated liquid densities from the correlation equation (2) are shown in Figures 1–3. In these figures, eq 2 reproduces the present results satisfactorily, and the maximum deviations ($\delta\rho/\rho$) are only 4.2×10^{-5} for HCFC 225ca, 4.6×10^{-5} for HCFC 225cb, and 5.2×10^{-5} for HCFC 141b.

Comparisons to literature values are also shown in these figures. In Figures 1 and 2, the results by Kumagai and Takahashi (1993) are in excellent agreement with the present results, within $\pm 0.05\%$ for HCFC 225ca and $\pm 0.08\%$ for HCFC 225cb. For both HCFC 225ca and HCFC 225cb, the values by Widiatmo et al. (1992) are about 0.1%

Table 6. Densities of Liquid HCFC 225ca and HCFC 225cb in the Single Phase

P/MPa	$\rho/(\text{kg}\cdot\text{m}^{-3})$					
	HCFC 225ca			HCFC 225cb		
	298.15 K	323.15 K	348.15 K	298.15 K	323.15 K	348.15 K
0.10	1550.3	1487.7		1556.0	1495.5	
0.50	1551.6	1489.3	1422.3	1557.2	1497.0	1432.6
1.00	1553.1	1491.2	1424.9	1558.6	1498.8	1434.9
1.50	1554.5	1493.0	1427.3	1560.0	1500.5	1437.1
2.00	1556.0	1494.9	1429.8	1561.3	1502.2	1439.4
2.50	1557.4	1496.7	1432.2	1562.7	1503.9	1441.6
3.00	1558.8	1498.5	1434.5	1564.0	1505.6	1443.7
3.50	1560.2	1500.2	1436.8	1565.3	1507.2	1445.8
4.00	1561.6	1502.0	1439.0	1566.6	1508.8	1447.9
4.50	1562.9	1503.7	1441.3	1567.9	1510.5	1450.0
5.00	1564.3	1505.4	1443.5	1569.1	1512.1	1452.0

Table 7. Density of Liquid HCFC 141b in the Single Phase

P/MPa	$\rho/(\text{kg}\cdot\text{m}^{-3})$	P/MPa	$\rho/(\text{kg}\cdot\text{m}^{-3})$	P/MPa	$\rho/(\text{kg}\cdot\text{m}^{-3})$
298.15 K		323.15 K		348.15 K	
1.19	1236.2	1.02	1185.9	1.06	1132.8
4.76	1243.4	2.46	1189.8	2.42	1137.5
10.37	1254.3	4.89	1195.8	5.25	1146.1
19.49	1270.6	10.09	1207.8	7.55	1153.4
30.29	1286.8	19.91	1227.5	9.94	1159.3
40.21	1300.3	30.12	1246.0	20.31	1185.1
49.50	1312.0	40.16	1261.7	30.03	1205.5
59.94	1324.1	49.95	1275.5	39.92	1224.2
70.35	1335.5	60.05	1289.0	50.42	1240.9
78.26	1343.5	70.12	1301.2	60.20	1255.1
82.22	1347.3	80.14	1312.7	70.17	1268.7
93.75	1358.1	89.89	1322.9	80.20	1282.0
101.25	1365.2	99.81	1333.2	89.85	1292.7
				100.01	1304.1

lower than the present results in the temperature range of these measurements. And their deviations from eq 2 tend to become larger with increasing temperature, but they are still within the combined uncertainty of the measurements, $\pm 0.3\%$. For HCFC 225cb, the correlation equation by Fukushima and Watanabe (1992), which was developed using both their own experimental data and the data by Kumagai and Takahashi (1993), shows a slightly different temperature dependence, and the deviation from eq 2 changes from -0.14% to $+0.31\%$ with increasing temperature. In Figure 3, the results by Defibaugh et al. (1993) and those by Kumagai and Takahashi (1993) show excellent agreement with the present results, within $\pm 0.04\%$ and 0.07% , respectively. The values by Maezawa et al. (1991) are lower than the present results, and their deviations are slightly larger than the combined uncertainties of the measurements, 0.3% .

In comparison to the corresponding restricted substances, it is found that the saturated liquid densities of HCFC 225ca and HCFC 225cb are very close to those of CFC 113 reported by Harada (1982) in the range of the present measurements, i.e., only $1.0 \pm 0.1\%$ and $0.5 \pm 0.1\%$ lower than the densities of CFC 113, respectively. On the other hand, the saturated liquid densities of HCFC 141b are found to be $16.3 \pm 0.1\%$ lower than those of CFC 11 reported by ASHRAE (1969).

The densities which were measured at higher pressures aimed at the determination of the isothermal compressibility are given in Tables 6 and 7. For each substance, the density increases monotonically with increasing pressure at each temperature and the specific volume, $v (= \rho^{-1})$, can be satisfactorily represented by the following Tait equation:

$$v_P = v_{\text{sat}} \left(1 - C \ln \frac{B + P}{B + P_{\text{sat}}} \right)^{-1} \quad (3)$$

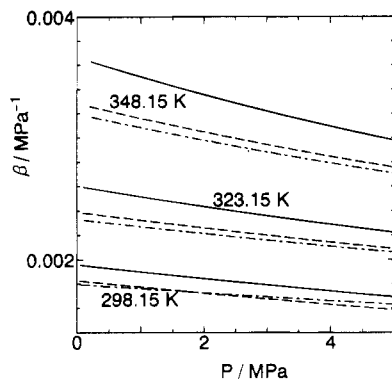


Figure 4. Isothermal compressibilities of HCFC 225ca and HCFC 225cb as a function of pressure: —, HCFC 225ca; - - -, HCFC 225cb; - · - ·, CFC 113.

where v_P and v_{sat} are the specific volumes at pressure P and at saturated vapor pressure P_{sat} . In this fitting procedure, the values of P_{sat} were calculated by the vapor pressure equations by Widiatmo et al. (1992) for HCFC 225ca and HCFC 225cb and by Defibaugh et al. for HCFC 141b. The values of v_{sat} are calculated by the use of eq 2. The maximum deviations ($\delta v_P/v_P$) of eq 3 are 4.6×10^{-5} for HCFC 225ca, 5.8×10^{-5} for HCFC 225cb, and 40.7×10^{-5} for HCFC 141b.

The isothermal compressibility of the present substances, β_T , which is defined by the following equation, is also determined in terms of the parameters of eq 3:

$$\beta_T = \frac{-1}{v} \left(\frac{\partial v}{\partial P} \right)_T = \left(1 - C \ln \frac{B+P}{B+P_{\text{sat}}} \right)^{-1} \frac{C}{B+P} \quad (4)$$

The results for HCFC 225ca and HCFC 225cb are given in Figure 4 which includes comparisons of CFC 113 reported by Harada (1982). As is illustrated in this figure, HCFC 225ca shows significantly higher values of β_T than HCFC 225cb, i.e., $7 \pm 1\%$ at 298 K, $8 \pm 1\%$ at 323 K, and $9 \pm 1\%$ at 348 K. It is also found that the isothermal compressibility of CFC 113 is close to that of HCFC 225cb over the entire range of the present measurement. The isothermal compressibilities of HCFC 141b are found to be lower than those of CFC 113, which were calculated using the equation of state by Rivkin and Kremnevckaya (1971), over the pressure range in which Rivkin's equation is valid, i.e., $14 \pm 2\%$ around their saturated vapor pressure and $5 \pm 2\%$ at the highest pressure, 20 MPa.

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