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Liquid-Phase Thermodynamic Properties of New Refrigerants: Pentafluoroethyl Methyl Ether and Heptafluoropropyl Methyl Ether[†]

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In the present paper, we report an experimental study on the liquid-phase thermodynamic properties of the new-generation alternative refrigerants, pentafluoroethyl methyl ether, $CF_3CF_2OCH_3$ (245cbE$\beta\gamma$), and heptafluoropropyl methyl ether, $CF_3CF_2CF_2OCH_3$ (347sE$\gamma\delta$). The measurements of vapor pressures and saturated- and compressed-liquid densities were performed by means of a magnetic densimeter coupled with a variable-volume cell mounted with a metallic bellows for temperatures from 260 to 370 K (for $CF_3CF_2OCH_3$) and from 250 to 370 K (for $CF_3CF_2CF_2OCH_3$) and pressures up to 3 MPa. The experimental uncertainties of the temperature, pressure, and density measurements were estimated to be within ± 8 mK, ± 1.9 kPa (single-phase measurements) and ± 2.5 kPa (two-phase vapor pressure measurements), and ± 2 kg·m^{-3}, respectively. The purities of the samples used throughout the measurements were 99.99 mass % for $CF_3CF_2CF_2OCH_3$ and 99.9 mass % for $CF_3CF_2CF_2OCH_3$. On the basis of these measurements and the available data reported by other investigators, the thermodynamic behaviors with respect to vapor pressures and saturated- and compressed-liquid densities are discussed in terms of vapor pressure and saturated liquid density correlations optimized and a liquid-phase equation of state. By examining the thermodynamic behavior of the derived properties such as the specific isochoric and isobaric heat capacities, speeds of sound, and Joule−Thomson coefficients, the range of validity for the optimized simple correlations and their physical soundness will be discussed.

Introduction

In accord with the increasing concerns about the global warming impact of released hydrofluorocarbon (HFC) refrigerants, several hydrofluoroether (HFE) refrigerants recently developed are considered as promising long-term replacements for some HFCs. HFE refrigerants, pentafluoroethyl methyl ether, $CF_3CF_2OCH_3$ (245cbE$\beta\gamma$), and heptafluoropropyl methyl ether, $CF_3CF_2CF_2OCH_3$ (347sE$\gamma\delta$), have vapor pressures similar to those of dichlorotetrafluoroethane (R-114) and trichlorofluoromethane (R-11), respectively. These HFE refrigerants have lower global warming potential than R-114 and R-11, respectively. So they are expected as a promising alternative to replace R-114 and R-11. The present study, therefore, aims to investigate the vapor pressures and the saturated- and compressed-liquid densities of $CF_3CF_2OCH_3$ and $CF_3CF_2CF_2OCH_3$.

Experimental Section

All experimental measurements in the present work were performed with a magnetic densimeter coupled with a variable volume cell, which enabled measurements either at the saturated-liquid or compressed-liquid conditions. The experimental apparatus is shown in Figure 1. The magnetic densimeter (A) consists of a buoy equipped with a permanent magnet, a sample cell, an electromagnetic coil, and a search coil. The sample density was measured by the

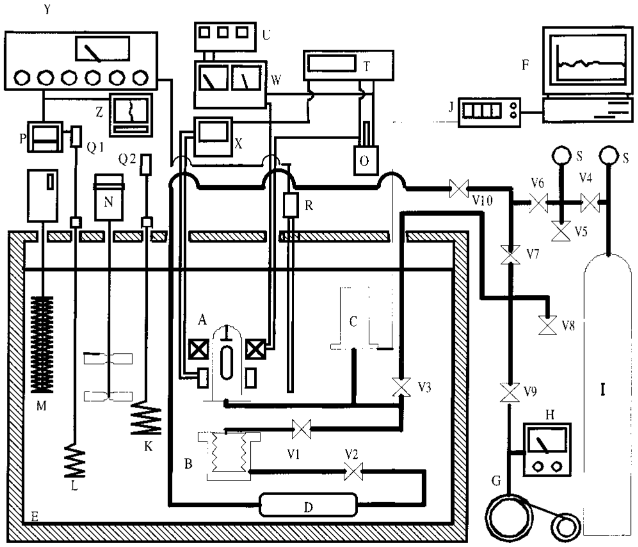

Figure 1. Experimental apparatus: (A) magnetic densimeter; (B) variable volume cell; (C) digital quartz pressure transducer; (D) damper; (E) thermostated fluid bath; (F) personal computer; (G) vacuum pump; (H) vacuum gauge; (I) nitrogen gas; (J) digital quartz pressure computer; (K) main heater; (L) subheater; (M) cooler; (N) stirrer; (O) standard resistor; (P) PID controller; (Q1, 2) thyristor transformer; (R) 25 Ω standard platinum resistance thermometer; (S) pressure gauge; (T) digital multimeter; (U) current controller; (V1−10) valves; (W) DC power supply; (X) galvanometer; (Y) thermometer bridge; (Z) pen recorder.

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magnetic densimeter immersed in a thermostated fluid bath (E). The sample pressure was directly measured by a digital quartz pressure transducer (Paroscientific, Model 31K-101) (C) with the aid of a digital quartz pressure computer (J). The temperature was measured by means of a 25 Ω standard platinum resistance thermometer (R) placed in the vicinity of the magnetic densimeter. By controlling the pressure of the nitrogen gas in the outer space of the metallic bellows within a variable-volume cell (B), it is possible to create a saturated- and/or a compressed-liquid condition. The saturated-liquid condition was determined from careful visual observation of the appearance and disappearance of a bubble in the liquid sample.

The experimental apparatus used for the present measurements was originally constructed by Maezawa et al.[1] and modified by Widiatmo et al.[2] Since then, we have completed a series of similar measurements with respect to HCFC and HFC refrigerants and their mixtures.[3,4]

The sample purities, as analyzed by the chemical manufacturer, are 99.99 mass % for $CF_3CF_2OCH_3$ and 99.9 mass % for $CF_3CF_2CF_2OCH_3$. These samples were provided by the Research Institute of Innovative Technology for the Earth (RITE), Tsukuba, Japan.

We estimated the uncertainties of the present measurements according to the ISO recommendation[5] in terms of the expanded uncertainties with a coverage factor of 2. The experimental uncertainties of temperature, pressure, and density measurements were estimated to be within ±8 mK, ±1.9 kPa (single-phase measurements) and ±2.5 kPa (two-phase vapor pressure measurements), and $\pm 2.2\ \mathrm{kg\cdot m^{-3}}$, respectively.

Results and Discussion

Fifteen vapor pressures and saturated-liquid densities, and 49 compressed-liquid densities of $CF_3CF_2OCH_3$ have been measured at temperatures from 260 to 370 K in 10 K intervals and pressures up to 3 MPa. The numerical experimental results are summarized in Table 1. For $CF_3CF_2CF_2OCH_3$, 22 vapor pressures and saturated-liquid densities, and 80 compressed-liquid densities have been measured for temperatures from 250 to 370 K and pressures up to 3 MPa. The obtained data are tabulated in Table 2.

Widiatmo and Watanabe[6] correlated the vapor pressure correlations for both $CF_3CF_2OCH_3$ and $CF_3CF_2CF_2OCH_3$ by using the present data added with the data by Tsuge et al.[7,8] and Uchimura et al.,[9] in a functional form given in eq 1.

$$\ln P_r = \frac{T_c}{T}(a_1\tau + a_2\tau^{1.5} + a_3\tau^3 + a_4\tau^6) \qquad (1)$$

where P_r and τ are defined as P/P_c and $1 - (T/T_c)$, respectively. The critical temperature, T_c, used in eq 1 for $CF_3CF_2OCH_3$ is that reported by Yoshii et al.,[10] while that of $CF_3CF_2CF_2OCH_3$ is that reported by Sako et al.[11] The critical pressures, P_c, in eq 1 are those determined by Widiatmo and Watanabe[6] for both compounds. The critical temperatures and pressures are given in Table 3, while coefficients a_1 to a_4 are given in Table 4.

The vapor pressure deviation of $CF_3CF_2OCH_3$ from eq 1, as illustrated in Figure 2, shows that the present measured vapor pressures agree satisfactorily with the data by Tsuge et al.[7,8] within our claimed uncertainty. On the other hand, the vapor pressures calculated from the vapor pressure correlation developed by Sako et al.[14] show a significant deviation from both the present data and the data by Tsuge et al.[7,8] This may reflect the existence of a systematic error in the development of the vapor pressure correlation by Sako et al.[14]

Table 1. Measured $P-\rho-T$ Properties for $CF_3CF_2OCH_3$

T/K	P/kPa	$\rho/(\mathrm{kg\cdot m^{-3}})$
259.984	43.5	1373.8[a]
269.986	69.0	1347.7[a]
269.986	69.0	1346.3[a]
279.989	104.8	1319.5[a]
279.989	104.7	1319.1[a]
279.989	502.4	1320.5
279.989	1003.9	1322.5
279.989	1501.8	1324.4
279.989	1998.7	1326.3
279.989	2504.2	1328.3
279.989	3007.4	1330.0
289.988	153.3	1290.9[a]
289.988	504.7	1292.3
289.988	996.1	1294.6
289.988	1496.4	1296.8
289.988	1996.3	1299.0
289.988	2503.8	1300.9
289.988	2997.7	1303.0
299.987	217.3	1260.8[a]
299.987	218.2	1260.9[a]
299.987	501.2	1262.4
299.987	1001.5	1264.9
299.987	1501.1	1267.3
299.987	2000.5	1269.5
299.987	2501.2	1272.2
299.987	2998.4	1274.2
309.986	300.1	1229.8[a]
309.986	999.8	1233.7
309.986	1498.7	1236.7
309.986	2000.1	1239.6
309.986	2501.9	1242.3
309.986	2998.2	1244.7
319.986	404.5	1196.6[a]
319.986	1006.4	1201.0
319.986	1502.3	1205.9
319.986	2002.1	1207.7
319.986	2499.1	1210.7
319.986	3001.6	1213.7
329.985	535.1	1163.0[a]
329.985	997.0	1167.1
329.985	1496.8	1170.9
329.985	1993.8	1174.7
329.985	2503.2	1178.7
329.985	3005.5	1182.4
339.985	694.9	1126.2[a]
339.985	1007.3	1129.6
339.985	1498.9	1134.3
339.985	1999.3	1139.2
339.985	2501.2	1143.6
339.985	2994.4	1148.3
349.984	888.0	1086.4[a]
349.984	1494.3	1094.3
349.984	1999.3	1100.3
349.984	2496.3	1105.8
349.984	2998.4	1111.2
359.984	1119.0	1042.4[a]
359.984	1502.0	1048.8
359.984	1997.0	1056.8
359.984	2500.0	1064.0
359.984	2996.3	1070.9
369.983	1393.0	992.9[a]
369.983	1993.2	1006.5
369.983	2498.6	1016.4
369.983	2996.4	1025.5

[a] Observed values at two-phase conditions.

Figure 3 depicts the vapor pressure deviation of $CF_3CF_2CF_2OCH_3$ from eq 1. As also seen in Figure 2, the vapor pressures calculated from the vapor pressure correlation developed by Sako et al.[14] deviate systematically to greater values with increasing temperature. On the contrary, the present vapor pressure data and those by Tsuge et al.[7,8] are in agreement within ±2 kPa. From Figures 2 and 3, it can be concluded that those vapor pressure correlations by Sako et al.[14] were developed on the basis of less reliable vapor pressure data. Since there is no numerical data available in their report, detailed discussion about the measurements by Sako et al.[14] cannnot be done.

For the purpose of data comparison, the saturated-liquid density correlation optimized by Widiatmo and Watanabe[6] and given in eq 2 is referred to in the present study.

$$\rho' = \rho_c[1 + B(1 - T_r)^{\beta} + \sum_{i=1}^{3} B_i(1 - T_r)^{b_i/3}] \qquad (2)$$

The critical density, ρ_c, used in eq 2 for $CF_3CF_2OCH_3$ is that reported by Yoshii et al.,[10] while that for $CF_3CF_2CF_2OCH_3$ is reported by Sako et al.[11] The numerical values of critical densities are given in Table 3, while the coefficients

Table 2. Measured $P-\rho-T$ Properties for $CF_3CF_2CF_2OCH_3$

T/K	P/kPa	ρ/(kg·m^{-3})	T/K	P/kPa	ρ/(kg·m^{-3})
249.981	4.8	1528.3[a]	319.986	155.8	1341.2[a]
			319.986	746.8	1344.1
259.984	10.2	1504.0[a]	319.986	1001.3	1345.4
			319.986	1503.0	1347.8
269.986	18.9	1478.9[a]	319.986	2084.5	1351.2
			319.986	2450.5	1353.0
279.989	31.7	1452.7[a]	319.986	2993.6	1355.6
279.988	31.5	1452.9[a]			
279.988	500.4	1454.3	329.985	214.6	1311.7[a]
279.988	1005.3	1456.0	329.985	213.8	1311.5[a]
279.988	1506.5	1457.9	329.985	213.9	1312.4[a]
279.988	2003.5	1459.5	329.985	503.4	1313.3
279.988	2499.6	1461.2	329.985	999.6	1316.4
279.988	3004.2	1462.6	329.985	1497.8	1319.3
			329.985	1999.1	1322.3
289.989	49.8	1425.8[a]	329.985	2499.9	1325.4
289.989	550.1	1428.7			
289.989	1011.5	1429.9	329.985	2995.6	1327.9
289.989	1502.9	1432.2	329.985	504.5	1314.7
289.989	2007.0	1433.6	329.985	998.8	1317.4
289.989	2504.8	1435.5	329.985	1500.2	1320.3
289.989	2998.9	1437.2	329.985	2010.3	1323.7
			329.985	2505.8	1327.0
299.987	75.5	1399.6[a]	329.985	2989.8	1329.0
299.987	534.7	1400.4			
299.987	996.3	1403.3	339.985	288.7	1281.0[a]
299.987	1504.4	1404.6	339.985	288.1	1280.6[a]
299.987	2007.0	1407.0	339.985	288.2	1280.6[a]
299.987	2499.9	1408.6	339.985	508.3	1281.9
299.987	2992.0	1410.3	339.985	1002.7	1285.5
			339.985	1503.7	1288.9
309.986	110.2	1370.5[a]	339.985	2000.1	1292.4
309.986	557.3	1372.3	339.985	2503.7	1295.7
309.986	1008.3	1374.1	339.985	3000.6	1298.9
309.986	1501.4	1376.8	339.985	522.7	1282.3
309.986	2000.3	1379.7	339.985	998.4	1285.7
309.986	2502.3	1381.6	339.985	1493.5	1288.9
309.986	2992.0	1383.6	339.985	2007.9	1292.5
			339.985	2505.4	1296.0
			339.985	3006.3	1299.2
349.985	381.9	1247.8[a]	359.984	494.7	1211.4
349.985	380.4	1247.2[a]	359.984	493.7	1212.1
349.985	380.7	1246.2[a]	359.984	493.9	1210.6
349.985	501.8	1248.2	359.984	1010.0	1217.3
349.985	1000.6	1252.4	359.984	1505.6	1222.2
349.985	1502.5	1256.4	359.984	2002.4	1226.7
349.985	2009.8	1260.4	359.984	2506.1	1231.2
349.985	2500.6	1264.2	359.984	2996.1	1235.2
349.985	2998.5	1268.7	359.984	1000.9	1215.6
349.985	998.3	1251.4	359.984	1505.3	1220.8
349.985	1504.6	1255.8	359.984	2001.7	1225.7
349.985	2001.2	1259.3	359.984	2507.6	1230.4
349.985	2507.7	1263.5	359.984	3002.6	1234.3
349.985	2998.2	1267.1			
			369.983	631.3	1173.6[a]
			369.983	1003.2	1178.2
			369.983	1498.7	1183.9
			369.983	2004.0	1189.9
			369.983	2501.7	1194.9
			369.983	2993.0	1200.2

[a] Observed values at two-phase conditions.

Table 3. Critical Parameters for $CF_3CF_2OCH_3$ and $CF_3CF_2CF_2OCH_3$

	$CF_3CF_2OCH_3$	$CF_3CF_2CF_2OCH_3$
P_c/kPa	2887[a]	2476[b]
T_c/K	406.83[a]	437.7[c]
ρ_c/(kg·m^{-3})	509[a]	530[c]

[a] Yoshii et al.[10] [b] Uchimura et al.[9] [c] Sako et al.[11]

B and B_1 through B_3 together with the respective exponents are given in Table 4.

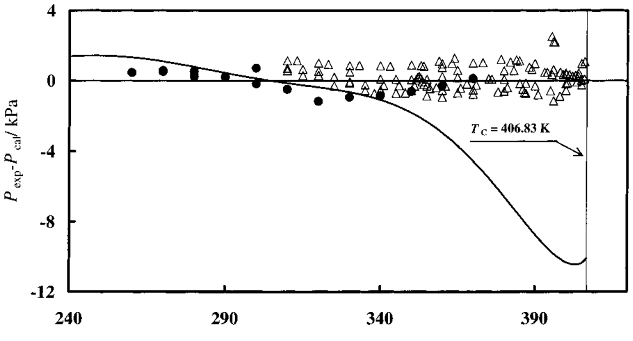

Figure 2. Vapor pressure deviation for $CF_3CF_2OCH_3$: ●, this work; △, Tsuge et al.;[7,8] —, Sako et al.[14]

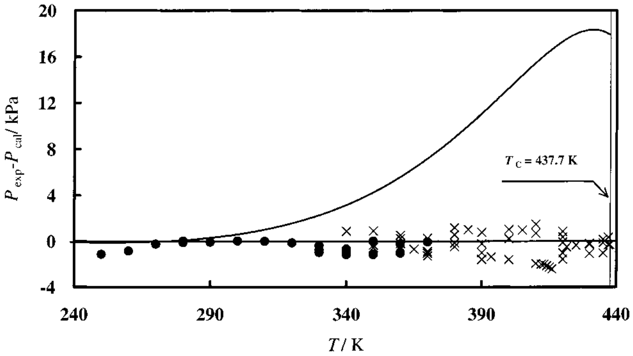

Figure 3. Vapor pressure deviation for $CF_3CF_2CF_2OCH_3$: ●, this work; ×, Uchimura et al.;[9] —, Sako et al.[14]

Table 4. Coefficients and Exponents in Eqs 1−3 for $CF_3CF_2OCH_3$ and $CF_3CF_2CF_2OCH_3$

	$CF_3CF_2OCH_3$	$CF_3CF_2CF_2OCH_3$
a_1	−7.73986	−7.95132
a_2	1.52151	1.50989
a_3	−4.05631	−4.48124
a_4	−11.0921	−20.8350
B	1.43926	1.81014
B_1	1.69075	0.98763
B_2	−1.54018	
B_3	1.57395	
β	0.322	0.325
b_1	2	2.36
b_2	4	
b_3	6	
c_0	0.084716	0.074730
c_1	−0.17288	−0.11568
d_0	−0.99575	−0.99907
d_1	12.306	11.368
d_2	69.742	106.35
d_3	−359.41	−456.44
d_4	545.71	570.32
e_0	0.56963	0.56658
e_1	−0.69563	−0.71530
e_2	0.49193	0.73236
e_3	0.28421	−0.43312
e_4	−0.71301	0.098989

Figure 4 illustrates the saturated-liquid density deviation of $CF_3CF_2OCH_3$ from eq 2. Earlier measurements by Tsuge et al.[7,8] are also represented in Figure 4. As shown in Figure 4, the present measurements agree well with the data by Tsuge et al.[7,8] Concerning $CF_3CF_2CF_2OCH_3$, as given in Figure 5, the present measurements also show an excellent agreement with the data by Yoshii et al.[10] and a smooth extension to those by Uchimura et al.[9]

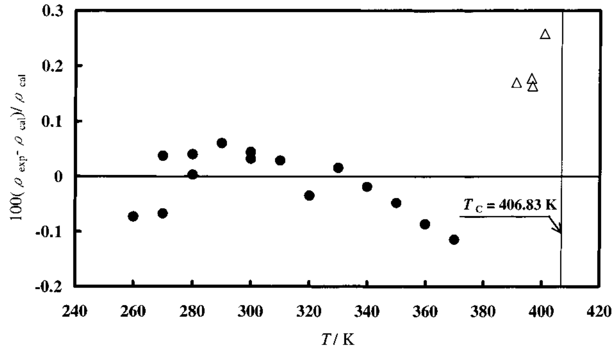

Figure 4. Saturated-liquid density deviation for $CF_3CF_2OCH_3$: ●, this work; △, Tsuge et al.[7,8]

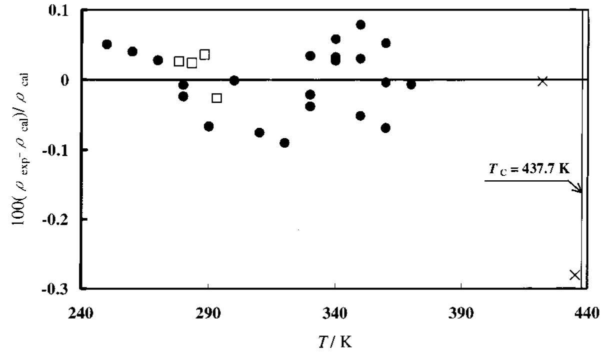

Figure 5. Saturated-liquid density deviation for $CF_3CF_2CF_2OCH_3$: ●, this work; ×, Uchimura et al.;[9] □, Nakazawa et al.[13]

Regarding the representation of thermodynamic properties of fluorinated ethers of present interest in their liquid phases including the saturated liquids, we have employed eq 3, which was successfully applied to similar representation by Fujimine et al.[12] for binary and ternary mixtures of difluoromethane (R-32), pentafluoroethane (R-125), and 1,1,1-trifluoroethane (R-143a).

$$\rho_r = (P_r + D)^C/E \tag{3}$$

where $\rho_r = \rho/\rho_c$ and $P_r = P/P_c$. The exponent C and the coefficients D and E are correlated using functional forms given in eqs 4−6.

$$C = c_0 + c_1\tau \tag{4}$$

$$D = d_0 + d_1\tau + d_2\tau^2 + d_3\tau^3 + d_4\tau^4 \tag{5}$$

$$E = e_0 + e_1\tau + e_2\tau^2 + e_3\tau^3 + e_4\tau^4 \tag{6}$$

The numerical constants in eqs 4−6 are given in Table 4. The data used to determine the numerical constants in eq 3 are the present measurements and unpublished data by Tsuge et al.[7,8] for $CF_3CF_2OCH_3$, and the present measurements and unpublished data by Uchimura et al.[9] for $CF_3CF_2CF_2OCH_3$, respectively.

The range of validity of eq 3, therefore, covers the reduced range of temperatures 0.639 to 0.999, pressures up to 1.03, and the range of densities 1.34 to 2.70 for $CF_3CF_2OCH_3$, whereas it covers 0.571 to 0.998 for T_r, up to 1.21 for P_r, and 1.35 to 2.88 for ρ_r for $CF_3CF_2CF_2OCH_3$.

It should be noted that the range of validity mentioned above was concluded on the basis of $P\rho T$ properties reported, excluding the very vicinity of the critical point of both compounds. The liquid density deviation from eq 3 is plotted in Figures 6 and 7 for $CF_3CF_2OCH_3$, and in Figures 8 and 9 for $CF_3CF_2CF_2OCH_3$, respectively. As shown in those figures, eq 3 represents the present compressed-liquid densities satisfactorily within ±0.2%.

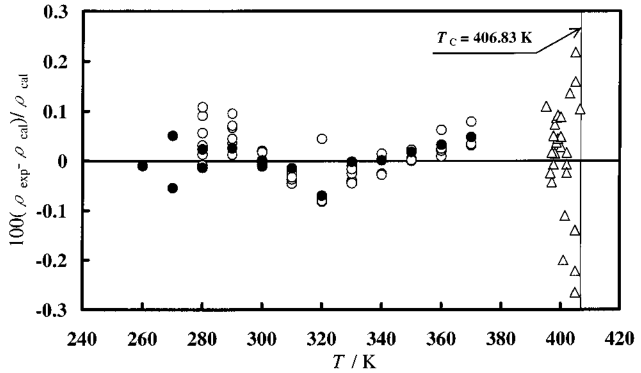

Figure 6. Liquid-density deviation for $CF_3CF_2OCH_3$ at different temperatures: ●, saturated-liquid densities from this work; ○, compressed-liquid densities from this work; △, Tsuge et al.[7,8]

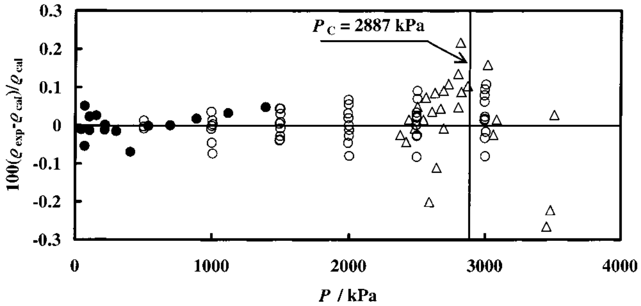

Figure 7. Liquid-density deviation for $CF_3CF_2OCH_3$ at different pressures: ●, saturated-liquid densities from this work; ○, compressed-liquid densities from this work; △, Tsuge et al.[7,8]

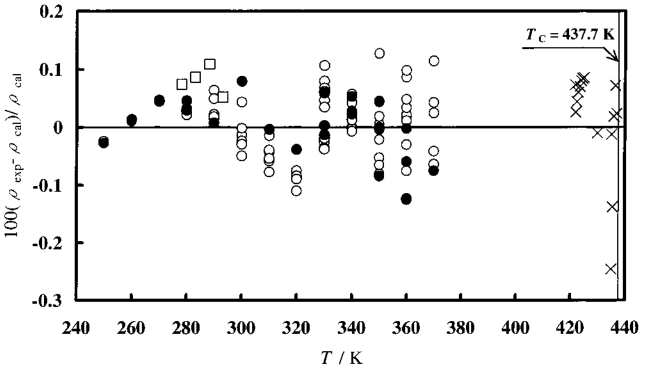

Figure 8. Liquid-density deviation for $CF_3CF_2CF_2OCH_3$ at different temperatures: ●, saturated-liquid densities from this work; ○, compressed-liquid densities from this work; ×, Uchimura et al.;[9] □, Nakazawa et al.[13]

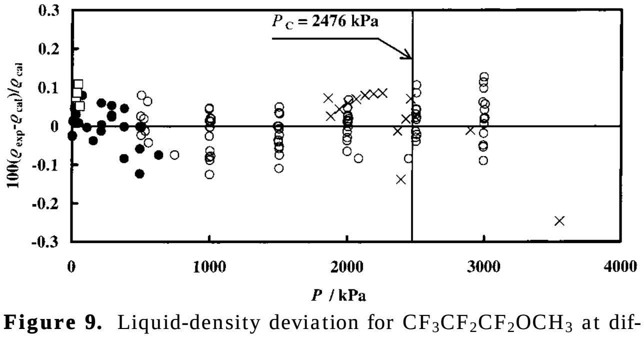

Figure 9. Liquid-density deviation for $CF_3CF_2CF_2OCH_3$ at different pressures: ●, saturated-liquid densities from this work; ○, compressed-liquid densities from this work; ×, Uchimura et al.;[9] □, Nakazawa et al.[13]

It should be noted that the present representation, eq 3, was confirmed to provide physically sound behaviors with respect to essential thermodynamic properties such as isothermal compressibility, specific isobaric and isochoric heat capacities, speed of sound, and the Joule–Thomson coefficient for the range of pressures up to 10 MPa and temperatures up to 390 K for $CF_3CF_2OCH_3$, whereas it covers up to 10 MPa and up to 420 K for $CF_3CF_2CF_2OCH_3$.

Conclusions

Forty-nine compressed-liquid densities and fifteen vapor-pressures and saturated-liquid densities of $CF_3CF_2OCH_3$ have been measured over the range of temperatures 260 to 370 K and pressures up to 3 MPa. Eighty compressed-liquid densities and twenty-two vapor pressures and saturated-liquid densities of $CF_3CF_2CF_2OCH_3$ have also been obtained over the range of temperatures 250 to 370 K and pressures up to 3 MPa.

On the basis of the present measurements, a simple thermodynamic formulation to represent the compressed-liquid densities has also been developed for $CF_3CF_2OCH_3$ and $CF_3CF_2CF_2OCH_3$, which enabled us to reproduce the measured data within ±0.2% in density.

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