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ARTICLE

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Compressed Liquid Density Measurements for 2,3,3,3-Tetrafluoroprop-1-ene (R1234yf)

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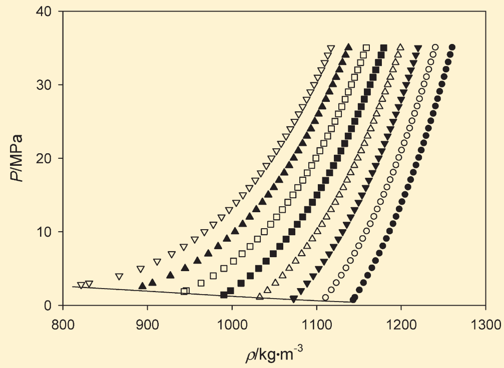
Supporting Information

ABSTRACT: A total of 13 796 compressed liquid density measurements for 2,3,3,3-tetrafluoroprop-1-ene (R1234yf) for eight isotherms evenly spaced from (283.15 to 353.15) K for pressures from near saturation to 35 MPa are presented. In addition, a saturated liquid density correlation and a Tait correlation are developed.



INTRODUCTION

In recent years, a large effort has been expended on studying refrigerants with low global warming potential (GWP), especially due to several agreements and laws at the national and international levels. Recently, the European Union's F-Gas regulations[1,2] have limited the global warming potential (GWP) < 150 for refrigerants for automotive systems, beginning January 1, 2011 for new car models and from January 1, 2017 for new vehicles.

Among others, 2,3,3,3-tetrafluoroprop-1-ene (R1234yf) is a new alternative refrigerant with low GWP (4 compared to CO_2 over a 100 year time horizon) and saturation pressure quite similar to 1,1,1,2-tetrafluoroethane (R134a) and thus deemed by many to be suitable for automotive applications.[3] This fluid has been extensively studied in the automotive industry over the past few years; however, only few thermodynamic (especially regarding $P\rho T$ behavior) and transport property data, and associated equations of state (EoS), have been published in the open literature. For this reason, a research project on this fluid has been undertaken at our institute,[4,5] and a set of compressed liquid density data is presented here.

The density along eight isotherms between (283.15 and 333.15) K and up to 35 MPa was measured by means of a vibrating tube densimeter (Anton Paar DMA 512), with an estimated expanded uncertainty of approximately 0.05 %. Saturated liquid densities were extrapolated from the measured data. The results of the

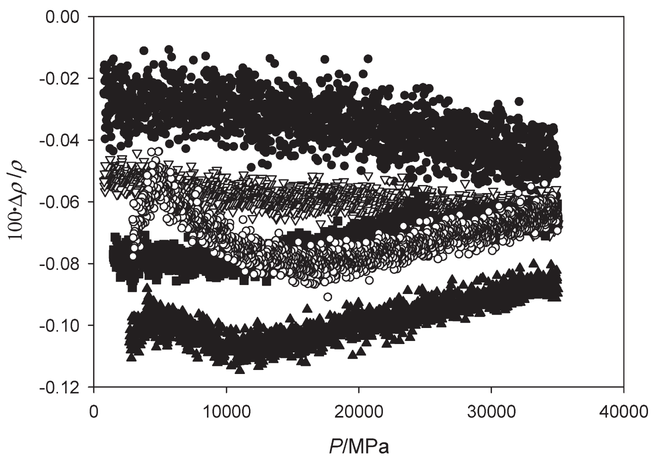


Figure 1. Relative differences $\Delta\rho/\rho = (\rho_{exp} - \rho_{calc})/\rho_{calc}$ of the experimental data of this work (ρ_{exp}), from the value obtained from Tillner-Roth and Baher[10] (ρ_{calc}) for R134a. ●, 283.15 K; △, 303.15 K; ■, 323.15 K; ▲, 343.15 K; ○, 353.15 K.

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Table 1. Selected Compressed Liquid Density Measurements for R1234yf by the Authors for Eight Isotherms at (283.15, 293.15, 303.15, 313.15, 323.15, 333.15, 343.15, and 353.15) K for P < 35 MPa[a]

T	P	ρ	e_{Tait}	e_{ref}	T	P	ρ	e_{Tait}	e_{ref}
K	MPa	kg·m^{-3}	%	%	K	MPa	kg·m^{-3}	%	%
283.14	35.000	1260.9	−0.02	−0.08	323.15	35.001	1179.4	0.00	−0.07
283.14	34.014	1258.5	−0.01	−0.08	323.15	34.017	1176.4	−0.02	−0.07
283.14	33.002	1256.2	−0.01	−0.08	323.15	33.013	1173.2	−0.01	−0.08
283.14	32.019	1253.7	−0.01	−0.09	323.15	32.014	1170.0	−0.01	−0.08
283.14	31.007	1251.3	−0.01	−0.08	323.15	31.002	1166.6	−0.01	−0.08
283.14	30.011	1248.8	−0.01	−0.09	323.15	30.004	1163.2	−0.01	−0.08
283.14	29.001	1246.2	0.00	−0.09	323.15	29.010	1159.8	−0.00	−0.08
283.14	28.001	1243.6	0.00	−0.10	323.15	28.019	1156.3	0.00	−0.09
283.14	27.006	1241.1	−0.01	−0.09	323.15	27.014	1152.7	−0.00	−0.08
283.14	26.011	1238.4	−0.01	−0.09	323.15	26.000	1148.9	0.01	−0.09
283.14	25.003	1235.7	0.00	−0.09	323.15	25.009	1145.2	0.01	−0.09
283.14	24.002	1232.8	0.01	−0.11	323.15	24.013	1141.3	0.01	−0.09
283.14	23.004	1230.0	0.01	−0.11	323.15	23.016	1137.3	0.01	−0.09
283.14	22.005	1227.2	0.00	−0.10	323.15	22.014	1133.2	0.01	−0.09
283.15	21.019	1224.4	−0.01	−0.10	323.15	21.001	1128.9	0.01	−0.10
283.14	20.019	1221.3	0.00	−0.10	323.15	20.007	1124.6	0.01	−0.09
283.14	19.001	1218.2	0.00	−0.11	323.15	19.001	1119.9	0.02	−0.10
283.14	18.011	1215.1	0.01	−0.12	323.15	18.014	1115.4	0.01	−0.10
283.14	17.014	1211.8	0.01	−0.12	323.15	17.001	1110.5	0.01	−0.10
283.14	16.010	1208.6	0.00	−0.11	323.15	16.017	1105.6	0.01	−0.10
283.14	15.003	1205.1	0.01	−0.12	323.15	15.010	1100.3	0.01	−0.10
283.14	14.013	1201.6	0.02	−0.13	323.15	14.017	1095.0	0.01	−0.10
283.14	13.020	1198.3	0.00	−0.12	323.15	13.018	1089.3	0.01	−0.10
283.14	12.011	1194.5	0.01	−0.13	323.15	12.018	1083.4	0.00	−0.11
283.14	11.022	1191.0	−0.01	−0.12	323.15	11.008	1077.1	0.00	−0.11
283.13	10.013	1186.9	0.01	−0.13	323.15	10.006	1070.7	−0.01	−0.10
283.14	9.009	1183.0	0.00	−0.13	323.15	9.0147	1063.8	0.00	−0.11
283.14	8.001	1178.9	0.01	−0.13	323.15	8.021	1056.6	−0.01	−0.11
283.14	7.021	1174.8	0.00	−0.13	323.15	7.021	1048.8	−0.01	−0.11
283.13	6.000	1170.3	0.01	−0.13	323.15	6.007	1040.4	−0.02	−0.11
283.13	5.021	1166.0	−0.01	−0.12	323.15	5.020	1031.4	−0.01	−0.11
283.14	4.001	1161.0	0.01	−0.14	323.15	4.019	1021.6	−0.01	−0.11
283.14	3.004	1156.0	0.01	−0.14	323.15	3.011	1010.6	0.00	−0.11
283.13	2.013	1151.2	0.00	−0.12	323.15	2.007	998.2	0.03	−0.13
283.13	1.022	1145.8	0.00	−0.12	323.15	1.421	990.3	0.05	−0.12
283.13	0.684	1143.8	0.01	−0.13					
293.15	35.002	1240.8	−0.06	−0.05	333.15	35.014	1158.6	−0.01	−0.11
293.15	34.017	1238.2	−0.05	−0.07	333.15	34.014	1155.2	0.00	−0.11
293.15	33.006	1235.7	−0.05	−0.07	333.15	33.001	1151.9	−0.01	−0.11
293.15	32.007	1233.2	−0.05	−0.06	333.15	32.015	1148.4	0.00	−0.11
293.15	31.003	1230.4	−0.04	−0.07	333.15	31.003	1144.8	0.00	−0.11
293.15	30.015	1227.6	−0.03	−0.08	333.15	30.004	1141.2	0.00	−0.12
293.15	29.013	1225.0	−0.04	−0.07	333.15	29.001	1137.5	0.00	−0.12
293.15	28.009	1222.3	−0.04	−0.07	333.15	28.003	1133.7	0.00	−0.12
293.15	27.012	1219.4	−0.04	−0.07	333.15	27.016	1129.8	0.00	−0.12
293.15	26.012	1216.6	−0.04	−0.07	333.16	26.013	1125.7	0.01	−0.12
293.15	25.002	1213.5	−0.03	−0.08	333.15	25.016	1121.6	0.00	−0.12
293.15	24.020	1210.7	−0.05	−0.07	333.15	24.004	1117.4	0.00	−0.12
293.16	23.008	1207.5	−0.04	−0.08	333.15	23.008	1113.0	0.00	−0.12
293.15	22.003	1204.3	−0.03	−0.09	333.16	22.013	1108.5	0.00	−0.13
293.15	21.020	1201.1	−0.03	−0.09	333.15	21.007	1103.8	0.01	−0.13

Table 1. Continued

T	P	ρ	e_{Tait}	e_{ref}	T	P	ρ	e_{Tait}	e_{ref}
K	MPa	$kg \cdot m^{-3}$	%	%	K	MPa	$kg \cdot m^{-3}$	%	%
293.15	20.015	1197.7	−0.03	−0.09	333.15	20.009	1099.0	0.01	−0.13
293.15	19.006	1194.3	−0.02	−0.10	333.16	19.016	1094.0	0.01	−0.13
293.16	18.014	1191.1	−0.04	−0.08	333.15	18.015	1088.9	0.01	−0.14
293.15	17.011	1187.6	−0.04	−0.08	333.15	17.014	1083.5	0.01	−0.14
293.15	16.004	1183.8	−0.03	−0.09	333.15	16.012	1077.9	0.01	−0.14
293.15	15.012	1180.1	−0.03	−0.10	333.15	15.006	1072.0	0.01	−0.14
293.15	14.016	1176.2	−0.02	−0.10	333.15	14.018	1066.0	0.01	−0.14
293.15	13.007	1172.2	−0.03	−0.10	333.15	13.007	1059.5	0.01	−0.14
293.15	12.001	1168.3	−0.04	−0.09	333.15	12.004	1052.7	0.01	−0.15
293.15	11.016	1164.1	−0.04	−0.09	333.15	11.013	1045.7	0.00	−0.14
293.15	10.025	1159.7	−0.03	−0.11	333.15	10.009	1038.0	0.01	−0.15
293.15	9.011	1155.1	−0.03	−0.11	333.16	9.009	1030.0	0.00	−0.15
293.15	8.016	1150.5	−0.04	−0.11	333.15	8.003	1021.2	0.01	−0.16
293.15	7.009	1145.6	−0.04	−0.11	333.15	7.009	1011.9	0.01	−0.16
293.15	6.022	1140.7	−0.04	−0.10	333.15	6.010	1001.6	0.01	−0.16
293.15	5.007	1135.3	−0.04	−0.10	333.15	5.010	990.3	0.02	−0.17
293.15	4.022	1129.8	−0.04	−0.10	333.15	4.001	977.6	0.03	−0.17
293.15	3.003	1123.9	−0.03	−0.11	333.15	3.010	963.3	0.06	−0.17
293.15	2.024	1117.9	−0.03	−0.11	333.14	2.003	945.8	0.16	−0.20
293.15	0.990	1111.2	−0.02	−0.11	333.14	1.866	943.2	0.18	−0.20
303.15	35.017	1220.0	−0.02	−0.09	343.16	35.000	1137.8	0.03	−0.13
303.15	34.018	1217.4	−0.02	−0.09	343.15	34.014	1134.3	0.03	−0.14
303.15	33.001	1214.6	−0.02	−0.09	343.15	33.007	1130.6	0.03	−0.14
303.15	32.004	1211.6	0.00	−0.12	343.15	32.001	1126.9	0.02	−0.14
303.15	31.010	1208.9	−0.01	−0.10	343.15	31.008	1123.1	0.02	−0.14
303.15	30.013	1205.9	0.00	−0.11	343.15	30.012	1119.1	0.02	−0.15
303.15	29.016	1202.9	0.00	−0.12	343.15	29.007	1115.0	0.02	−0.16
303.15	28.002	1200.0	0.00	−0.11	343.15	28.014	1111.0	0.02	−0.16
303.15	27.010	1196.9	0.00	−0.11	343.16	27.005	1106.7	0.02	−0.16
303.15	26.005	1193.8	0.00	−0.11	343.16	26.009	1102.4	0.01	−0.16
303.15	25.007	1190.5	0.01	−0.12	343.16	25.003	1097.8	0.01	−0.16
303.15	24.019	1187.3	0.00	−0.12	343.15	24.011	1093.3	0.01	−0.16
303.15	23.008	1183.9	0.01	−0.12	343.15	23.013	1088.5	0.00	−0.16
303.15	22.000	1180.5	0.00	−0.12	343.16	22.004	1083.4	0.01	−0.17
303.14	21.015	1177.0	0.01	−0.12	343.16	21.001	1078.3	0.00	−0.17
303.14	20.019	1173.4	0.01	−0.13	343.16	20.005	1073.0	0.00	−0.17
303.14	19.011	1169.8	−0.01	−0.11	343.16	19.002	1067.5	−0.01	−0.17
303.14	18.026	1166.0	0.00	−0.12	343.16	18.011	1061.8	−0.01	−0.18
303.14	17.003	1161.8	0.02	−0.14	343.15	17.005	1055.7	0.00	−0.18
303.14	16.021	1158.0	0.00	−0.12	343.16	16.014	1049.5	−0.01	−0.19
303.15	15.003	1153.7	0.01	−0.13	343.16	15.013	1042.9	−0.01	−0.19
303.14	14.016	1149.6	0.00	−0.12	343.16	14.013	1035.9	−0.01	−0.19
303.14	13.017	1144.9	0.01	−0.14	343.16	13.015	1028.6	−0.01	−0.20
303.14	12.001	1140.3	0.01	−0.14	343.16	12.010	1020.8	−0.02	−0.20
303.14	11.006	1135.7	0.00	−0.14	343.17	11.012	1012.5	−0.02	−0.20
303.15	10.010	1130.7	0.00	−0.14	343.17	10.010	1003.6	−0.02	−0.21
303.14	9.017	1125.6	0.00	−0.14	343.17	9.005	993.9	−0.02	−0.21
303.15	8.010	1120.1	0.00	−0.15	343.16	8.008	983.5	−0.02	−0.22
303.14	7.016	1114.7	−0.01	−0.13	343.16	7.002	972.0	−0.02	−0.22
303.14	6.003	1108.7	−0.01	−0.14	343.17	6.004	959.0	0.00	−0.24
303.14	5.014	1102.5	−0.01	−0.14	343.17	5.013	944.4	0.03	−0.26
303.14	4.016	1095.9	0.00	−0.15	343.17	4.013	927.2	0.09	−0.28

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Table 1. Continued

T	P	ρ	e_{Tait}	e_{ref}	T	P	ρ	e_{Tait}	e_{ref}
K	MPa	$kg \cdot m^{-3}$	%	%	K	MPa	$kg \cdot m^{-3}$	%	%
303.14	3.007	1088.8	0.01	−0.16	343.17	3.006	905.9	0.22	−0.32
303.14	2.001	1081.4	0.00	−0.14	343.17	2.542	894.0	0.33	−0.34
303.14	1.009	1073.4	0.01	−0.15					
303.14	0.905	1072.8	0.00	−0.13					
313.16	35.001	1199.1	0.04	−0.14	353.15	35.003	1116.9	1.75	−0.17
313.16	34.013	1196.2	0.04	−0.14	353.15	34.007	1113.2	1.73	−0.18
313.16	33.002	1193.2	0.04	−0.14	353.15	33.000	1109.5	1.68	−0.16
313.15	32.013	1190.3	0.04	−0.14	353.15	32.004	1105.5	1.66	−0.16
313.15	31.001	1187.1	0.05	−0.15	353.15	31.012	1101.2	1.66	−0.18
313.15	30.002	1184.1	0.05	−0.14	353.15	30.011	1096.9	1.64	−0.19
313.15	29.001	1180.9	0.05	−0.14	353.16	29.008	1092.6	1.62	−0.19
313.15	28.010	1177.6	0.06	−0.15	353.16	28.007	1088.2	1.59	−0.19
313.15	27.005	1174.3	0.06	−0.15	353.15	27.004	1083.5	1.57	−0.20
313.15	26.001	1170.9	0.06	−0.15	353.16	26.007	1078.8	1.54	−0.20
313.15	25.022	1167.4	0.06	−0.15	353.16	25.010	1073.9	1.52	−0.20
313.15	24.011	1164.0	0.05	−0.14	353.16	24.004	1068.9	1.48	−0.20
313.15	23.014	1160.2	0.06	−0.15	353.16	23.011	1063.8	1.45	−0.19
313.15	22.006	1156.4	0.06	−0.15	353.17	22.000	1058.0	1.45	−0.22
313.15	21.008	1152.6	0.05	−0.15	353.17	21.001	1052.4	1.41	−0.22
313.15	20.005	1148.5	0.07	−0.16	353.17	20.007	1046.6	1.39	−0.22
313.15	19.011	1144.6	0.06	−0.16	353.17	19.007	1040.5	1.35	−0.22
313.15	18.012	1140.3	0.06	−0.16	353.17	18.002	1034.1	1.32	−0.22
313.15	17.014	1136.0	0.06	−0.16	353.18	17.015	1027.4	1.29	−0.23
313.15	16.011	1131.5	0.06	−0.16	353.18	16.004	1020.4	1.24	−0.21
313.15	15.006	1126.8	0.06	−0.16	353.18	15.017	1012.9	1.22	−0.24
313.15	14.007	1122.1	0.05	−0.16	353.18	14.000	1005.1	1.16	−0.22
313.15	13.004	1116.9	0.06	−0.17	353.17	13.014	996.6	1.14	−0.25
313.15	12.000	1111.7	0.06	−0.17	353.17	12.011	987.5	1.10	−0.25
313.15	11.001	1106.3	0.06	−0.17	353.18	11.013	977.9	1.05	−0.26
313.15	10.017	1100.9	0.04	−0.17	353.17	10.004	967.2	0.99	−0.26
313.15	9.019	1094.9	0.05	−0.17	353.17	9.012	955.8	0.93	−0.27
313.15	8.002	1088.6	0.04	−0.17	353.18	8.001	942.9	0.87	−0.27
313.15	7.018	1082.2	0.03	−0.17	353.18	7.015	928.6	0.80	−0.28
313.14	6.004	1075.1	0.03	−0.17	353.17	6.010	911.5	0.75	−0.34
313.15	5.009	1067.6	0.05	−0.18	353.17	5.009	891.8	0.63	−0.33
313.15	4.006	1059.8	0.03	−0.17	353.17	4.012	866.6	0.56	−0.38
313.14	3.006	1051.2	0.03	−0.17	353.17	3.010	830.9	0.58	−0.45
313.14	2.008	1041.9	0.04	−0.17	353.2	2.813	821.5	0.64	−0.42
313.14	1.118	1032.7	0.06	−0.18					

[a] Relative differences $e = 100 \cdot \Delta\rho/\rho = (\rho_{calc} - \rho_{exp})/\rho_{exp}$ of the values calculated (ρ_{calc}) from eq 3 (Tait) or Refprop[11] (Ref), from the experimental values of this work (ρ_{exp}).

compressed liquid density measurements and the extrapolated values of saturated liquid density were correlated with fitting equations and compared to existing experimental data sets.

EXPERIMENTAL SECTION

Materials. R1234yf (2,3,3,3-tetrafluoroprop-1-ene, $CF_3CF{=}CH_2$, CAS No. 754-12-1) was provided by DuPont with a declared purity higher than 0.995 in mass fraction. R134a (1,1,1,2-tetrafluoroethane, CF_3CH_2F, CAS No. 811-97-2) was supplied by Nevada with a purity higher than 0.995 in mass fraction. Bidistilled water was from Carlo Erba (H_2O, CAS No. 7732-18-5), with a declared contaminants concentration lower than 10^{-6} in mass fraction.

To eliminate noncondensable gases, each sample underwent several cycles of freezing with liquid nitrogen, evacuation, thawing, and ultrasound agitation. The samples were then used with no further purification.

Because of the possibility of polymerization of R1234yf under conditions of high pressure and high temperature,[6] a study was conducted to ensure that polymerization does not occur under the experimental conditions considered in this paper. In particular, mass spectrometric analyses were performed on two R1234yf samples, one taken before being subjected to extreme conditions and the other one after having been subjected to conditions of 35 MPa and 353 K (the extreme conditions in the present experimental range) for two hours, by means of a MD800 GC-MS system, operating under electron ionization (EI) conditions, and equipped with a capillary gas chromatographic column. The GC/MS analyses for both samples showed the presence of only one chromatographic peak at room temperature, 240 s: the related EI mass spectra are in agreement with the molecular structure of R1234yf, the ion M^+ being seen at m/z 114. Moreover, considering the experimental procedure used to measure the density along each isotherm, the vibrating tube frequency monotonically decreased with time (and pressure), indicating no polymer was formed inside the densimeter.

Experimental Apparatus. Compressed liquid density measurements were performed by means of an apparatus based on a stainless steel vibrating tube densimeter (Anton Paar DMA 512). The experimental apparatus and procedure are described in previous papers.[7,8] Here, only the main features of the experimental apparatus are described. The measurement principle is based on the relationship between the period of oscillation of the vibrating U-tube densimeter, that is, a hollow resonating stainless steel tube, and the density of the sample, determined through calibration by varying temperature T and pressure P. Pressure was measured by means of a piezo-resistive pressure gauge (Druck DPI 145) with a scale up to 35 MPa, with an estimated experimental uncertainty of approximately 10 kPa. The temperature was controlled with a stability of about ± 0.003 K by means of an external water thermostatic bath surrounding the vibrating tube, through a heat exchanger surrounding the U-tube. Moreover, an electrical resistance heater, controlled by a variable transformer, allowed fine local temperature control around the circuit connection. This ensured temperature and density uniformity inside the vibrating tube. Temperature measurements were obtained by means of a Pt 100 Ω resistance thermometer, with an estimated uncertainty of 0.05 K.

The densimeter was connected to an electronic evaluation unit for the measurement of the oscillation period (Anton Paar mPDS 2000) and filled with the sample through a circuit of stainless steel tubes connecting the cell and the refrigerant bottle. The fluid was pressurized directly by a syringe pump (Isco Pump, model 260D) connected to the circuit.

A dedicated LabView based data acquisition system was used to control the experimental apparatus and to record the experimental variables.

Considering the complete experimental method, the expanded uncertainty of the density measurements is estimated to be approximately 0.8 $kg \cdot m^{-3}$.

Experimental Procedure. After purging and evacuating the system, refrigerant in the liquid phase was charged into the vibrating tube and associated measurement circuit. After pressure and temperature stabilization, a controlled pressure bleeding of about (15 to 20) $kPa \cdot s^{-1}$ was performed expanding the volume inside the syringe pump, while the values of the vibrating tube period of oscillation were continuously acquired. The measurements for a single isotherm took approximately 14 400 s, including time for temperature change and stabilization.

Density Calibration. The period of oscillation of the U-tube under vacuum conditions and filled with water was acquired at different pressures (up to 35 MPa) and temperatures [(283.15 to 353.15) K] to determine the relationship between the period of oscillation and the fluid density. Water was chosen as the calibration fluid due to the availability of the high accuracy equation of state of Wagner and Pruss.[9] The calibration correlations are given in Fedele et al.[8]

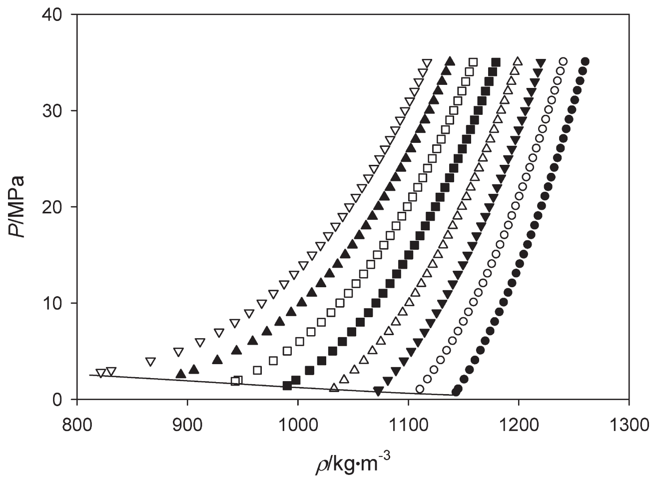

Figure 2. Measured compressed liquid densities of this work for R1234yf. ●, 283.15 K; ○, 293.15 K; ▼, 303.15 K; △, 313.15 K; ■, 323.15 K; □, 333.15 K; ▲, 343.15 K; ▽, 353.15 K; —, eq 2.

Table 2. Constants[5] for Equation 1

A_1	A_2	A_3	A_4
−7.34421	1.49565	−1.77264	−4.92486

Table 3. Constants for Equation 2

B_1	B_2	B_3	B_4
1.83126	0.79835	−0.61911	0.87149

■ RESULTS AND DISCUSSION

Experimental Compressed Liquid Density. To confirm the reliability of the experimental techniques and equipment, compressed liquid density data for R134a were measured for five isotherms (283.15, 303.15, 323.15, 343.15, and 353.15) K for $P < 35$ MPa. The relative differences between these data and the calculations of Tillner-Roth and Baehr[10] (whose declared uncertainty is 0.05 %) are shown in Figure 1. The relative differences, $(0 < 100\Delta\rho/\rho < -0.12)$ %, are comparable with the experimental uncertainties, validating the reliability of the experimental measurements.

A total of 13 796 new experimental compressed liquid density data points for R1234yf are provided for eight isotherms at (283.15, 293.15, 303.15, 313.15, 323.15, 333.15, 343.15, and 353.15) K for $P < 35$ MPa. A selection of 280 data points is summarized in Table 1 and shown in Figure 2. All of the measured data are available in the Supporting Information as described at the end of the paper.

The experimental data are used to develop a saturated liquid density correlation, which is compared to the equation of Richter et al.[6] as implemented in Refprop 9.0.[11] They are also used to develop a compressed liquid density correlation, which is compared to the equation in Refprop 9.0, the data of Table 1, and literature data.[12−14] However, before discussing the development

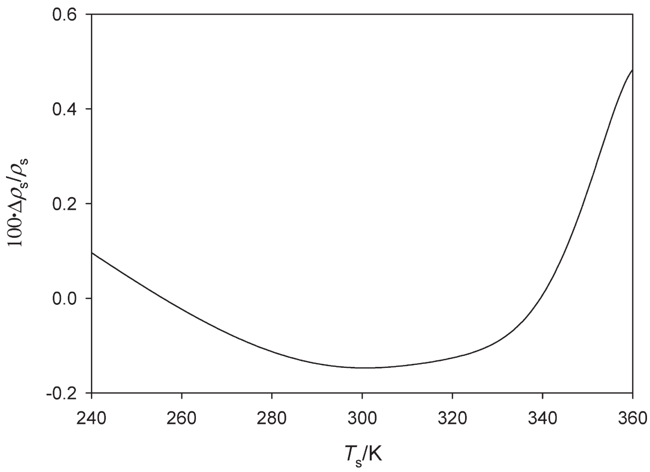

Figure 3. Relative differences $\Delta\rho/\rho = (\rho_{calc1} - \rho_{calc2})/\rho_{calc2}$ of the value obtained from eq 2 (ρ_{calc1}) from the value obtained from Richter et al.[6] (ρ_{calc2}) for R1234yf.

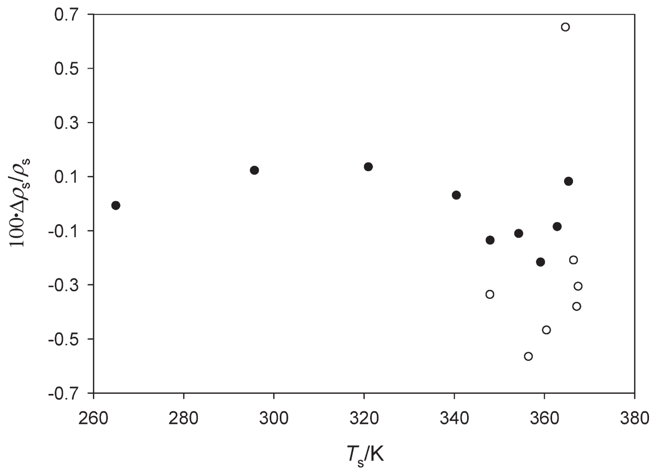

Figure 4. Relative differences $\Delta\rho/\rho = (\rho_{calc} - \rho_{exp})/\rho_{exp}$ of the value obtained from eq 2 (ρ_{calc}), from the experimental data from the literature[12,13] (ρ_{exp}) for R1234yf. ●, Hulse et al.;[12] ○, Tanaka and Higashi.[13]

of these correlations, the critical state properties and a vapor pressure correlation will be briefly discussed, since these are necessary for both density correlations.

Critical State Properties. The critical state properties[11] are critical temperature (T_c) = 367.85 K, critical pressure (P_c) = 3382.2 kPa, and critical density (ρ_c) = 475.55 kg·m^{-3}.

Vapor Pressure Correlations. Fedele et al.[5] provide a Wagner-type vapor pressure correlation given in eq 1, which fitted all publicly available experimental data at the time of its publication with an absolute mean differences $\Sigma_{i=1}^{n} 100 \cdot |\Delta\rho_i/\rho_i|/n < 0.075$.

$$T_r \ln(P_r) = A_1\tau + A_2\tau^{1.5} + A_3\tau^{2.5} + A_4\tau^5 \quad (1)$$

with the constants provided in Table 2 and where the reduced temperature is $T_r = T_s/T_c$, the reduced pressure $P_r = P_s/P_c$, $\tau = 1 - T_r$, T_s is the saturation temperature, and P_s is the saturation pressure.

Saturated Liquid Density Correlation. The compressed liquid density data for T = (283.15 to 343.15) K were used to develop the saturated liquid density (ρ_s) correlation given in eq 2.

$$\rho_s = \rho_c(1 + B_1\tau^{1/3} + B_2\tau^{2/3} + B_3\tau + B_4\tau^{4/3}) \quad (2)$$

with the constants provided in Table 3.

Equation 2 was developed by first fitting the data for each isotherm for P < 5 MPa with a third-order polynomial. These polynomials were coupled with eq 1 to provide estimates for ρ_s for T_s = (283.15 to 343.15) K, which were then used to develop eq 2.

Figure 3 shows relative differences ($\Delta\rho/\rho$) between eq 2 and the equation of Richter et al. for T_s = (240 to 360) K. For T_r = (0.65 to 0.95) for eq 2 relative to the equation of Richter et al., $100\Delta\rho/\rho = (\pm 0.2)$ % and for T_r = (0.95 to 0.98), $100\Delta\rho/\rho < 0.5$ %. Note: the data of 353.15 K were not used in the development of eq 2 since the slope of the ρ—T curve for the 353.15 K isotherm, as calculated with the equation of Richter et al.,[6] changes more dramatically as it approaches P_s than do the slopes for the other isotherms. If the 353.15 K isotherm were to be included, $\Delta\rho/\rho$ for T_r = (0.65 to 0.95) would be considerably higher at $100\Delta\rho/\rho = (-1.4$ to $0.5)$ %. Figure 4 shows relative differences ($\Delta\rho/\rho$) between eq 2 and literature data.[12,13] For

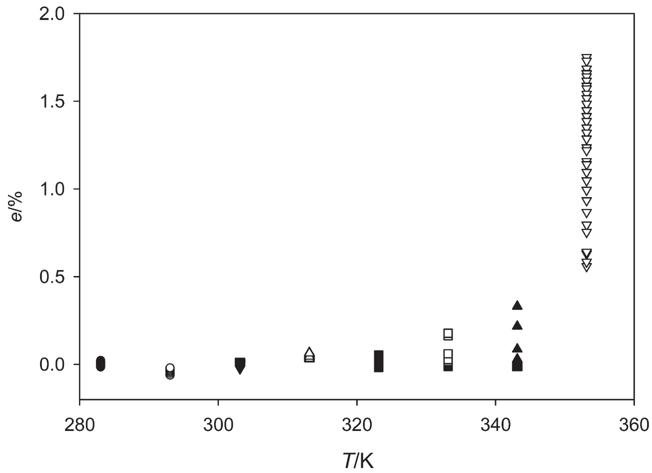

Figure 5. Relative differences $\Delta\rho/\rho = (\rho_{calc} - \rho_{exp})/\rho_{exp}$ of the value obtained from eq 3 (ρ_{calc}), from the experimental data of this work (ρ_{exp}) for R1234yf. ●, 283.15 K; ○, 293.15 K; ▼, 303.15 K; △, 313.15 K; ■, 323.15 K; □, 333.15 K; ▲, 343.15 K; ▽, 353.15 K.

T_r = (0.72 to 0.999) the deviations between eq 2 and to literature data[12,13] are $100\Delta\rho/\rho = (\pm 0.65)$ %. Note: Tanaka and Higashi[13] provide four additional data that are not shown in Figure 5 for $T_r > 0.999$ since $\Delta\rho/\rho$ varies up to 8 % because of the nearness to the critical point.

Compressed Liquid Density Correlation. The compressed liquid density data were used to develop a Tait correlation given in eq 3. Similar to what was done for the saturated liquid density correlation, the data for the 353.15 K isotherm were not used in the development of eq 3.

$$\rho^{-1} = \rho_s^{-1}\left[1 - C\ln\left(\frac{\beta + P}{\beta + P_s}\right)\right] \quad (3)$$

with ρ_s given by eq 2, β given by eq 4,

$$\beta = P_c(-1 + a\tau^{1/3} + b\tau^{2/3} + d\tau + e\tau^{4/3}) \quad (4)$$

and the constants are provided in Table 4.

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Table 4. Constants for Equations 3 and 4

C	*a*	*b*	*d*	*e*
0.08621	−15.2557	109.6546	−254.1543	232.1123

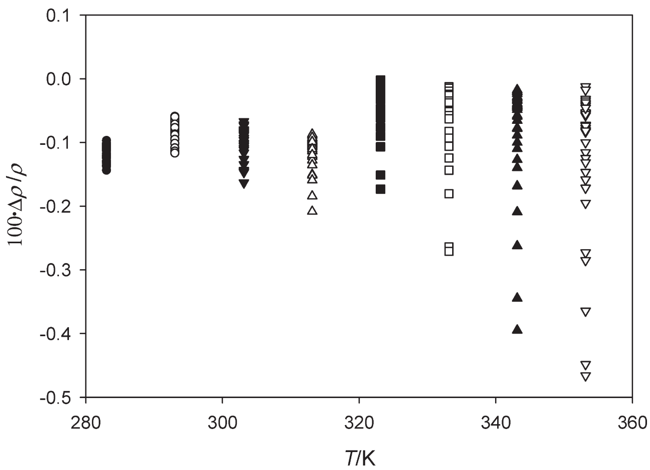

Figure 6. Relative differences $\Delta\rho/\rho = (\rho_{exp} - \rho_{calc})/\rho_{calc}$ of the experimental data of this work (ρ_{exp}), from the value obtained from Richter et al.[6] (ρ_{calc}) for R1234yf. ●, 283.15 K; ○, 293.15 K; ▼, 303.15 K; △, 313.15 K; ■, 323.15 K; □, 333.15 K; ▲, 343.15 K; ▽, 353.15 K.

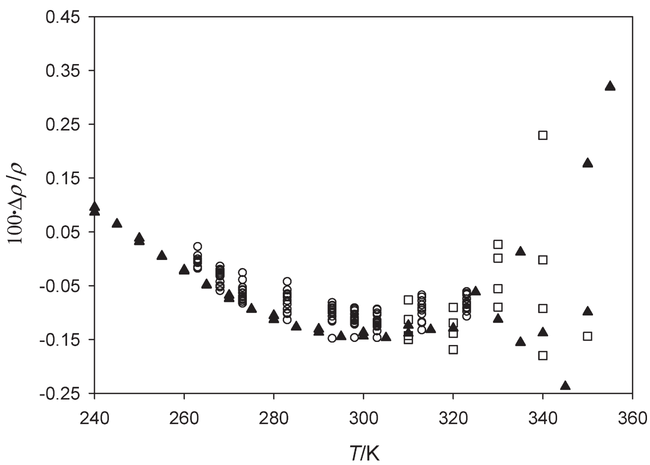

Figure 7. Relative differences $\Delta\rho/\rho = (\rho_{calc} - \rho_{exp})/\rho_{exp}$ of the value obtained from eq 3 (ρ_{calc}), from the experimental data from the literature[6,14,15] (ρ_{exp}) for R1234yf. ▲, Richter et al.;[6] ○, Yoshitake et al.;[14] □, Tanaka et al.[15]

Figure 5 shows relative differences ($\Delta\rho/\rho$) between eq 3 and the data of Table 1. For the isotherms T = (283.15, 293.15, 303.15, 313.15, and 323.15) K, $100\Delta\rho/\rho$ = (−0.08 to 0.08) %; for the isotherms T = (333.15 and 343.15) K, $100\Delta\rho/\rho$ = (−0.08 to 0.35) %; and for the isotherm T = 353.15 K, $100\Delta\rho/\rho$ = (−0.08 to 1.8) %.

Figure 6 shows relative differences ($\Delta\rho/\rho$) between the data of Table 1 and the equation of Richter et al. For this equation, the declared liquid density uncertainty is 0.1 % from 240 to 320 K and pressures up to 10 MPa, while it increases outside of this region up to 0.5 %. For the isotherms T = (283.15, 293.15, 303.15, 313.15, and 323.15) K, $100\Delta\rho/\rho$ = (0.0 to −0.17) %; for the isotherms T = (333.15 and 343.15) K, $100\Delta\rho/\rho$ = (−0.01 to −0.39) %; and for the isotherm T = 353.15 K, $100\Delta\rho/\rho$ = (−0.01 to −0.47) %. Note: In addition to providing T, P, and ρ, Table 1 shows relative differences for the Tait correlation (eq 3) and Refprop 9.0 calculations compared to the measured data.

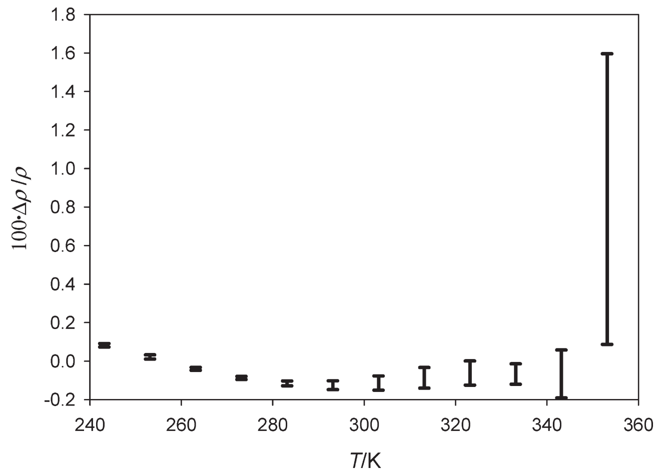

Figure 8. Relative differences $\Delta\rho/\rho = (\rho_{calc1} - \rho_{calc2})/\rho_{calc2}$ of the value obtained from eq 3 (ρ_{calc1}), from the value obtained from Richter et al.[6] (ρ_{calc2}) for R1234yf.

Figure 7 shows relative differences ($\Delta\rho/\rho$) between eq 3 and literature data.[6,14] For T = (250 to 355) K, $100\Delta\rho/\rho$ = (−0.25 to 0.32) %. Tanaka et al.[15] provide three data points for the isotherm T = 360 K, which are not shown in Figure 8 since the errors are up to 33 %. Also, one data point for the isotherm T = 310 K with $100\Delta\rho/\rho$ = −1 % is not shown because: (1) the data point is not consistent with their other data, and (2) it would require a rescaling of the ordinate axis, thus losing resolution in the figure.

Figure 8 shows relative differences of eq 3 relative to Refprop 9.0. For T (240 to 343.15) K, $100\Delta\rho/\rho$ = (−0.20 to 0.10) % and for T = 353.15 K, $100\Delta\rho/\rho$ = (0.10 to 1.6) %.

■ CONCLUSIONS

A total of 13 796 compressed liquid density measurements of R1234yf (2,3,3,3-tetrafluoroprop-1-ene; $CF_3CF{=}CH_2$) for reduced temperatures from 0.77 to 0.96 for reduced pressures less than 10.34 are presented, in a broader experimental pressure range than the available literature data. The data were used to develop a saturated liquid density correlation and a compressed liquid density correlation (Tait equation), which were compared to the experimental points presented herein, 332 data points from three different research groups, and the equation of Richter et al. as implemented in Refprop 9.0. The saturated liquid density correlation provided in eq 2 has deviations for T_r = (0.65 to 0.95) relative to the equation of Richter et al. of $\Delta\rho/\rho$ = ± 0.2 % and for T_r = (0.95 to 0.98) of $100\Delta\rho/\rho$ < (0.5) %. The compressed liquid density correlation provided in eq 3 has deviations for T_r = (0.65 to 0.95) relative to the experimental data of Table 1, the experimental data of three research groups,[6,14,15] and Refprop 9.0 of $100\Delta\rho/\rho$ = ± 0.5 %.

■ ASSOCIATED CONTENT

Supporting Information. All experimental data for R134a (file 1) and R1234yf (file 2). This material is available free of charge via the Internet at http://pubs.acs.org.

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REFERENCES

(1) Regulation (EC) No 842/2006 of The European Parliament and of the Council of 17 May 2006 on Certain Fluorinated Greenhouse Gases. *Official J. European Union* **2006**, *161*, 1−11.

(2) Directive 2006/40/EC of The European Parliament & of the Council of 17 May 2006 Relating to Emissions from Air-Conditioning Systems in Motor Vehicles & Amending Council Directive 70/156/EC. *Official J. European Union* **2006**, *161*, 12−18.

(3) SAE. Industry Evaluation of Low Global Warming Potential Refrigerant HFO-1234yf, 2009. Retrieved online at: http://www.sae.org/standardsdev/tsb/cooperative/crp1234-3.pdf (Accessed Dec 11, 2010).

(4) Bobbo, S.; Groppo, F.; Scattolini, M.; Fedele, L. *R1234yf as a Substitute of R134a in Automotive Air Conditioning. Solubility Measurements in Commercial PAG*, presented at "The 23rd IIR International Congress of Refrigeration", August 21−26, 2011, Prague, Czech Republic.

(5) Fedele, L.; Bobbo, S.; Groppo, F.; Brown, J. S.; Zilio, C. Saturated Pressure Measurements of 2,3,3,3-Tetrafluoroprop-1-ene (R1234yf) for Reduced Temperatures Ranging from 0.67 to 0.93. *J. Chem. Eng. Data* **2011**, *56*, 2608–2612.

(6) Richter, M.; McLinden, M. O.; Lemmon, E. W. Thermodynamic Properties of 2,3,3,3-Tetrafluoroprop-1-ene (R1234yf): Vapor Pressure and *p*-*ρ*-*T* Measurements and an Equation of State. *J. Chem. Eng. Data* **2011**, *56*, 3254–3264.

(7) Fedele, L.; Marinetti, S.; Bobbo, S; Scattolini, M. *PρT* Experimental Measurements and Data Correlation of Pentaerythritol esters. *J. Chem. Eng. Data* **2007**, *52*, 108–115.

(8) Fedele, L.; Pernechele, F.; Bobbo, S.; Scattolini, M. Compressed Liquid Density Measurements for 1,1,1,2,3,3,3-Heptafluoropropane (R227ea). *J. Chem. Eng. Data* **2007**, *52*, 1955–1959.

(9) Wagner, W.; Pruss, A. The IAPWS Formulation 1995 for the Thermodynamic Properties of Ordinary Water Substance for General and Scientific Use. *J. Phys. Chem. Ref. Data* **2002**, *31*, 387–535.

(10) Tillner-Roth, R.; Baehr, H. D. An International Standard Formulation of the Thermodynamic Properties of 1,1,1,2-Tetrafluoroethane (HFC-134a) for Temperatures from 170 to 455 K at Pressures up to 70 MPa. *J. Phys. Chem. Ref. Data* **1994**, *23*, 657–729.

(11) Lemmon, E. W.; Huber, M. L.; McLinden, M. O. *NIST Standard Reference Database 23, Reference Fluid Thermodynamic and Transport Properties (REFPROP)*, version 9.0; National Institute of Standards and Technology: Gaithersburg, MD, 2010 (R134a.fld file dated November 10, 2010 and R1234yf.fld file dated December 22, 2010).

(12) Hulse, R.; Singh, R.; Pham, H. *Physical Properties of HFO-1234yf*, Proceedings of the 3rd IIR Conference on Thermophysical Properties and Transfer Processes of Refrigerants, Boulder, CO, June 21−26, 2009; paper IIR-178.

(13) Tanaka, K.; Higashi, Y. Thermodynamic Properties of HFO-1234yf (2,3,3,3-Tetrafluoropropene). *Int. J. Refrig.* **2010**, *33*, 474–479.

(14) Yoshitake, M.; Matsuo, S.; Sotani, T. *Density and Speed of Sound Measurements of HFO-1234yf*, Proceedings of the 30th Japan Symposium on Thermophysical Properties, Yonezawa, Japan, 2009 (in Japanese).

(15) Tanaka, K.; Higashi, Y.; Akasaka, R. Measurements of the Isobaric Specific Heat Capacity and Density for HFO-1234yf in the Liquid State. *J. Chem. Eng. Data* **2010**, *55*, 901–903.