Compressed Liquid Densities, Saturated Liquid Densities, and Vapor Pressures of 1,1-Difluoroethane

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The compressed liquid densities and vapor pressures of 1,1-difluoroethane (HFC-152a) have been measured, correlated, and compared with other data. The liquid densities were measured with a combined standard uncertainty of $\pm 0.05\%$ using a vibrating tube densimeter over a temperature range of 243 K to 371 K and at pressures from near the saturated vapor pressure to 6500 kPa; thus the data extend nearly to the critical point ($T_c = 386.41$ K and $P_c = 4514.7$ kPa). The vapor pressures were measured with a combined standard uncertainty of $\pm 0.02\%$ using a stainless steel ebulliometer in the temperature range from 280 K to 335 K. Saturated liquid densities were calculated by extrapolating the compressed liquid isotherms to the saturation pressure.

Introduction

We have measured the compressed liquid density and the vapor pressure of 1,1-difluoroethane (HFC-152a). We report measurements in the compressed liquid region along 29 isotherms between 243 K and 371 K, with pressure in the range 700 kPa to 6500 kPa. The apparatus used to determine density was a vibrating tube densimeter. The densimeter had been calibrated in the range of 273 K to 373 K when under vacuum (\approx 1 Pa) and when filled with distilled, degassed water. Propane was used as the calibrating fluid in the range 243 K to 273 K. Details of the calibration can be found in reference Defibaugh and Morrison (1992). The combined standard uncertainty of the compressed liquid densities is estimated to be 0.05% in density. The data representing compressed liquid states $(P \rho T)$ for HFC-152a have been correlated with a 14-term equation. The correlation represents the compressed liquid density of this work to within $\pm 0.02\%$ except in the nearcritical temperature region.

The saturation pressures for HFC-152a were determined using a stainless steel ebulliometer. The vapor pressures ranged from 350 kPa to 1550 kPa, which correspond to temperatures from 280 K to 335 K. The vapor pressures are compared with other laboratories through a vapor pressure correlation for HFC-152a developed recently by Silva and Weber (1993). The vapor pressures of this work agree with other labs to within $\pm 0.15\%$ in pressure. Compressed liquid isotherms are extrapolated to the saturation pressure to determine the saturation densities. The saturation densities are correlated and then compared with other reference data. The combined standard uncertainty of the saturation densities is $\pm 0.1\%$.

Apparatus and Procedure

1,1-Difluoroethane (HFC-152a) was obtained from PCR Inc., Gainesville, FL, and was received from the supplier with a stated purity of 99.9%. The purity of the sample was verified by gas chromatography; no further purity analysis or attempts to purify were considered. Brand names and commercial sources of materials and instruments, when noted, are given for scientific completeness. Such information does not constitute a recommendation

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by the National Institute of Standards and Technology nor does it suggest that these products or instruments are the best for the described application.

Densimeter. A stainless steel vibrating tube densimeter designed by Anton PAAR was used to measure compressed liquid densities (see Figure 1). The temperature of the apparatus was regulated by a thermostated bath. The bath circulated a water/ethylene glycol mixture through a steel heat exchanger surrounding the vibrating tube. The steel heat exchanger was in an air-filled well-insulated enclosure. A platinum resistance thermometer (PRT) was used to monitor the temperature of the water/ethylene glycol mixture exiting the heat exchanger. The manifold outside the thermostated enclosure consisted of mercury reservoirs and a mercury-filled manometer/separator. The manometer separated the refrigerant sample from the pressurizing argon gas. A glass capillary in the manometer/separator allowed us to locate the mercury level. The pressure of the system was always maintained approximately 100 kPa above the vapor pressure of HFC-152a at any given temperature. When the vibrating tube was below room temperature, the low-pressure limit of the density measurements was determined by the vapor pressure at the temperature of the external manifold (room temperature). The pressure of the argon was monitored with a quartz pressure transducer with a standard uncertainty of ± 0.5 kPa.

Before the apparatus was filled, it was rinsed with ethanol and then acetone to remove residue from previous experiments. The apparatus was then evacuated and cooled to 273 K. Mercury was drawn into the reservoirs and the manometer/separator. The valve connecting the two sides of the separator was closed. Finally, refrigerant sample was condensed into the vibrating tube and separator manifold. Once the sample was loaded, the argon pressure was raised, and the valve separating the sides of the manifold was opened while monitoring the location of the mercury level to ensure that the apparatus was indeed completely filled with liquid. Once the apparatus was filled, compressed liquid densities for HFC-152a were measured.

Ebulliometer. A stainless steel ebulliometer was used to measure vapor pressures (see Figure 2). The apparatus consisted of a boiler, a condenser, and a cold trap. Liquid in the boiler was heated by passing electrical current

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Figure 1. Schematic diagram of vibrating tube densimeter.



Figure 2. Schematic diagram of ebulliometer apparatus.

through a heating ribbon wrapped around the bottom exterior of the boiler. As the liquid boiled, heated vapor rose up from the boiler and entered the condenser. Thermocouples in various locations around the apparatus help monitor the temperature of the apparatus and its approach to thermal equilibrium. A PRT located between the boiler and condenser measured the condensing temperature of the fluid within the apparatus. Cool water was circulated through the jacket of the condenser so that heated vapor coming from the boiler condensed and fell back into the boiler. A cold trap on top of the apparatus was filled with a dry ice/methanol slurry and acted as a safety mechanism to ensure the condensation of any vapor which might have passed through the condenser. A pressure controller located beyond the cold trap monitors and controls the helium pressure above the condensing HFC-152a.

Before the ebulliometer was loaded, it was evacuated, the cold trap was loaded with dry ice and methanol, and the water-cooled condenser was cooled to 273 K. Approximately 18 cm³ of liquid sample was condensed into the boiler, and finally the system was pressurized with helium. Heat was applied to the boiler, and the temper-



Figure 3. Compressed liquid density data (\bigcirc) and extrapolated saturated liquid densities (\square) of this work.

ature of each thermocouple was monitored. Once the temperature of the three thermocouples located in the boiler agreed to within 2 K of the PRT, the apparatus was considered to be in equilibrium and a condensation temperature and pressure were recorded. The pressure of the system was then increased and the process of monitoring the thermocouples for equilibrium was repeated.

Results

Compressed Liquid Density. Compressed-liquiddensity measurements were made along isotherms ranging from 243 K to 371 K at pressures from 700 kPa to 6500 kPa. The densities, shown in Figure 3 and listed in Table 1, span the range from 9.7 mol·dm⁻³ to 15.67 mol·dm⁻³ (the molecular weight of HFC-152a is 66.051). The temperatures (ITS-90) have a standard uncertainty of ± 0.01 K; however, temperature gradients across the densimeter may have exceeded this at the highest and lowest temperatures. The pressures have a standard uncertainty of ± 0.5 kPa.

The compressed liquid densities were correlated with the following equation:

$$P = RT\rho + \frac{b_1\rho^6}{T} + e^{-(\rho/\rho_c)^2} \sum_{n=2}^7 a_n(T)\rho^{2n-1}$$
(1)

where *P* is in kPa, *T* is in Kelvin, and ρ is in mol·dm⁻³. The temperature dependence of the coefficients is given by:

$b_1 = 0.948602920789893$
$b_2 = -988500945.479363$
$b_3 = 24675727183.2342$
$b_4 = 2173873.88796000$
$b_5 = -794490707710.284$
$b_6 = -207002.729161860$
$b_7 = -41660911.4218338$
$b_8 = 1824.90878720446$
$b_9 = 201024904.188244$
$b_{10} = 0.0260101097956443$
$b_{11}^{10} = -6558.55820796058$
$b_{12}^{11} = 0.0195808782129117$
$b_{13}^{12} = 11.1725470419356$
$b_{14}^{10} = -1248.56261373225$
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R = 0.0831445 J/(kmol·K)
$ ho_{ m c}=5.571~{ m mol}\cdot{ m dm}^{-3}$

(Higashi et al., 1987)

Table 1.	Compressed	Liquid	Densities	for	HFC-152a

Table 1. Compressed Liquid Densities for HFC-152a								
<i>T</i> /K	<i>P</i> /kPa	$ ho/mol \cdot dm^{-3}$	<i>T</i> /K	<i>P</i> /kPa	$ ho/mol\cdot dm^{-3}$	<i>T</i> /K	<i>P</i> /kPa	$ ho/mol \cdot dm^{-3}$
243.088	699.8	15.527	243.079	1000.2	15.536	243.082	1500.6	15.549
243.1	2000.5	15.561	243.093	2501.4	15.574	243.099	3002.4	15.586
243.104	3502.8	15.599	243.113	4002.7	15.611	243.106	4503.9	15.623
243.107	5004.8 6506 5	15.030	243.095	5505.3 699.9	15.048	243.093	10003.4	15.00
246.678	1500.7	15.436	246.665	2000.6	15.449	246.657	2501.6	15.463
246.68	3002.5	15.476	246.67	3503	15.489	246.667	4002.9	15.502
246.662	4504	15.515	246.664	5004.9	15.528	246.657	5505.5	15.541
246.653	6005.5	15.553	246.659	6506.5	15.566	249.407	699.8	15.326
249.411	1000.3	15.335	249.421	1500.6	15.348	249.427	2000.6 3502 Q	15.362
249.429	4002.9	15.416	249.442	4503.9	15.429	249.434	5004.9	15.443
249.449	5505.4	15.455	249.457	6005.4	15.468	249.459	6506.5	15.481
254.078	699.8	15.175	254.081	1000.2	15.184	254.08	1500.6	15.199
253.964	2000.5	15.217	253.933	2501.5	15.233	253.922	3002.4	15.247
253.924	3302.9 5004 9	15.202	253.91	4002.9	15.270	200.92 253 933	4505.9	15.29
253.946	6506.4	15.344	259.631	699.7	14.993	259.647	1000.2	15.002
259.65	1500.6	15.018	259.639	2000.5	15.034	259.639	2501.4	15.05
259.645	3002.3	15.065	259.649	3502.8	15.08	259.649	4002.8	15.095
259.652	4503.8	15.11	259.643	5004.7	15.125	259.638	5505.2	15.14
259.639	6005.2	15.154	259.647	6506.2 1500 G	15.168	264.407	699.7 2000 4	14.834
264.423	2501.3	14.893	264.449	3002.2	14.80	264.441	2000.4	14.926
264.452	4002.7	14.942	264.462	4503.8	14.957	264.462	5004.7	14.973
264.478	5505.2	14.988	264.473	6005.3	15.004	264.475	6506.2	15.019
269.254	699.8	14.671	269.263	1000.2	14.682	269.264	1500.5	14.7
269.267	2000.6	14.718	269.258	2501.5	14.735	269.258	3002.4	14.753
269.263	5004.9	14.821	269.263	4002.8 5505.4	14.837	269.251	4303.9	14.804
269.248	6506.5	14.87	272.58	699.8	14.558	272.592	1000.2	14.569
272.592	1500.6	14.588	272.59	2000.6	14.606	272.594	2501.5	14.625
272.59	3002.4	14.643	272.596	3502.9	14.661	272.612	4002.9	14.678
272.616	4503.9	14.695	272.616	5004.9 6506 5	14.713	272.637	5505.4 600.6	14.729
274.135	1000.1	14.516	274.131	1500.5	14.536	274.127	2000.3	14.505
274.135	2501.3	14.573	274.129	3002.2	14.592	274.129	3502.7	14.61
274.127	4002.8	14.628	274.124	4503.8	14.646	274.125	5004.8	14.664
274.125	5505.3	14.682	274.119	6005.3	14.699	274.119	6506.3	14.716
278.899	699.8 2000 5	14.339	278.902	1000.2 2501.5	14.331	278.898	1500.7	14.371
278.893	3502.9	14.352	278.891	4002.8	14.471	278.885	4503.8	14.432
278.882	5004.8	14.509	278.88	5505.3	14.527	278.878	6005.3	14.546
278.881	6506.3	14.564	283.726	699.8	14.166	283.729	1000.2	14.179
283.729	1500.6	14.201	283.728	2000.5	14.223	283.729	2501.4	14.245
283.73	3002.4	14.200	283.73	3502.8	14.280	283.727	4002.8 5505-3	14.307
283.723	6005.3	14.386	283.73	6506.3	14.406	288.618	699.7	13.987
288.629	1000.2	14.001	288.645	1500.7	14.024	288.647	2000.6	14.048
288.646	2501.4	14.071	288.65	3002.3	14.093	288.651	3502.8	14.115
288.649	4002.8	14.138	288.651	4503.8	14.159	288.651	5004.8	14.181
293 58	699 8	13 801	293 582	1000.3	13.816	293 585	1500.3	14.243
293.588	2000.5	13.867	293.588	2501.4	13.892	293.587	3002.3	13.916
293.59	3502.8	13.94	293.591	4002.8	13.964	293.588	4503.8	13.987
293.589	5004.7	14.01	293.588	5505.2	14.032	293.585	6005.2	14.055
293.591	6506.3	14.076	298.544	699.7 1500 5	13.609	298.544	1000.2	13.625
298.544	2501.4	13.708	298.538	3002.3	13.734	298.543	2000.4	13.76
298.543	4002.7	13.785	298.543	4503.8	13.81	298.54	5004.7	13.835
298.541	5505.2	13.859	298.542	6005.2	13.883	298.543	6506.3	13.906
303.506	799.9	13.416	303.5	1000.1	13.428	303.513	1500.6	13.458
303.513	2000.5	13.488	303.515	2501.4	13.517	303.514	3002.3	13.546
303.515	5002.8 5004 8	13.655	303.513	5505 2	13.681	303.510	4005.8 6005.2	13.706
303.515	6506.3	13.732	308.396	900.1	13.22	308.396	1000.1	13.227
308.399	1500.5	13.26	308.396	2000.4	13.293	308.398	2501.4	13.324
308.4	3002.3	13.356	308.401	3502.8	13.386	308.403	4002.7	13.416
308.398	4503.8	13.445	308.403	5004.8	13.473	308.399	5505.2	13.501
313 285	1500 G	13.529	308.404 313 984	2000.5	13.000	313.283	2501 4	13.018
313.289	3002.3	13.159	313.292	3502.8	13.192	313.291	4002.8	13.224
313.29	4503.8	13.256	313.29	5004.7	13.287	313.291	5505.2	13.317
313.293	6005.3	13.346	313.288	6506.3	13.376	318.319	1500.6	12.833
318.321	2000.5	12.873	318.321	2501.4	12.911	318.321	3002.3	12.949
318.322	5004.7	13.089	318.323	5505.1	13.122	318.321	6005.2	13.154

Table 1 (Continued)								
<i>T</i> /K	<i>P</i> /kPa	ρ/mol∙dm ^{−3}	<i>T</i> /K	<i>P</i> /kPa	ρ/mol∙dm ^{−3}	<i>T</i> /K	P/kPa	$ ho/mol \cdot dm^{-3}$
318.321	6506.2	13.186	323.189	1600.6	12.617	323.191	2000.4	12.652
323.192	2501.4	12.695	323.194	3002.3	12.737	323.185	3502.7	12.777
323.184	4002.7	12.816	323.186	4503.8	12.854	323.187	5004.7	12.891
323.185	5505.2	12.927	323.185	6005.2	12.962	323.185	6506.2	12.996
328.051	1700.6	12.392	328.055	2000.5	12.421	328.058	2501.4	12.469
328.058	3002.3	12.515	328.057	3502.7	12.56	328.056	4002.7	12.603
328.058	4503.8	12.645	328.056	5004.7	12.685	328.057	5505.2	12.725
328.055	6005.2	12.763	328.055	6506.2	12.8	332.892	1800.6	12.155
332.894	2000.5	12.178	332.896	2501.4	12.232	332.897	3002.3	12.284
332.897	3502.8	12.334	332.899	4002.7	12.382	332.897	4503.7	12.428
332.9	5004.7	12.473	332.897	5505.2	12.516	332.897	6005.2	12.558
332.9	6506.2	12.599	337.894	1900.5	11.896	337.902	2000.5	11.908
337.905	2501.4	11.97	337.902	3002.3	12.03	337.903	3502.8	12.087
337.903	4002.8	12.141	337.903	4503.8	12.193	337.905	5004.7	12.243
337.905	5505.2	12.291	337.905	6005.2	12.337	337.903	6506.2	12.383
342.87	2000.5	11.618	342.872	2501.4	11.692	342.874	3002.4	11.761
342.877	3502.7	11.826	342.868	4002.7	11.888	342.863	4503.7	11.948
342.863	5004.7	12.004	342.864	5505.2	12.058	342.864	6005.1	12.11
342.865	6506.2	12.159	347.793	2200.7	11.339	347.794	2501.4	11.391
347.799	3002.4	11.473	347.801	3502.7	11.549	347.798	4002.6	11.621
347.803	4503.7	11.688	347.803	5004.6	11.753	347.803	5505.1	11.814
347.804	6005	11.872	347.801	6506.1	11.928	352,694	2501.5	11.058
352.695	3002.3	11.158	352.698	3502.7	11.25	352.698	4002.6	11.334
352.688	4503.6	11.414	352.692	5004.6	11.488	352.693	5505.1	11.557
352.692	6005	11.623	352.693	6506.1	11.686	357.563	2701.8	10.733
357.566	3002.3	10.807	357.567	3502.7	10.921	357.568	4002.6	11.024
357.568	4503.6	11.118	357.568	5004.6	11.204	357.573	5505	11.285
357.572	6005	11.361	357.57	6506.1	11.432	362.405	3002.2	10.403
362.411	3502.7	10.551	362.41	4002.6	10.68	362.407	4503.7	10.795
362.407	5004.7	10.9	362.411	5505.2	10.995	362.41	6005.2	11.083
362.408	6506.2	11.165	367.275	3202.7	9.988	367.279	3502.8	10.11
367.28	4002.8	10.284	367.281	4503.8	10.431	367.284	5004.8	10.56
367.281	5505.2	10.676	367.282	6005.3	10.78	367.283	6506.3	10.877
371.331	3602.9	9.704	371.333	4002.8	9.892	371.336	4503.8	10.084
371.338	5004.8	10.245	371.336	5505.2	10.384	371.334	6005.2	10.507
371.334	6506.3	10.619	0.1000		10.001	0.1.001		10.007



Figure 4. Compressed liquid density deviations (%) from eq 1: this work (\bigcirc) ; Tillner-Roth and Baehr, 1993 (\Box) ; Blanke and Weiss, 1992 (\triangle) .

Equation 1 is a correlation of only the compressed liquid data. The terms in eq 1 are a subset of the terms in the widely used MBWR equation of state used by Jacobsen and Stewart (1973). The deviations of the densities from eq 1 are shown in Figure 4. Equation 1 can reproduce the density of this work to $\pm 0.02\%$ except at the highest temperature where the isotherm begins to curve due to the proximity of the critical point, $T_c = 386.41$, Higashi et al. (1987). A deviation greater than 0.02% at 371 K is attributed to two factors: the inability of eq 1 to model the very curved isotherms near the critical point, and the high sensitivity of the density to uncertainties in the temperature and pressure measurements close to the critical point.

Table 2. Vapor Pressures for HFC-152a

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_	<i>T</i> /K	<i>P</i> /kPa	<i>T</i> /K	<i>P</i> /kPa
	281.442	351.91	318.650	1049.98
	288.967	449.93	322.221	1149.99
	295.452	550.01	325.565	1249.99
	301.090	649.99	328.714	1349.96
	306.113	750.01	331.689	1449.66
	310.652	850.03	334.518	1549.62
	314.808	950.00		

The data of Blanke and Weiss (1992) agree with this work to 0.02% above 260 K. The data of Tillner-Roth and Baehr (1993) also agree very well with this work with an average deviation of 0.045% above 260 K. At temperatures below 260 K deviations increase to 0.1%.

Vapor Pressures. Vapor pressure measurements were made between 280 K and 335 K and are listed in Table 2. The data fall within a region where there are few published data. The vapor pressures were compared to a Wagner type vapor pressure correlation, eq 2, established by Silva and Weber (1993).

$$\ln \frac{P}{P_{c}} = \frac{T_{c}}{T} [A_{1}\tau + A_{2}\tau^{1.5} + A_{3}\tau^{2.5} + A_{4}\tau^{5}]$$
(2)
$$A_{1} = -7.3943 \qquad T_{c} = 386.41 \text{ K}$$

$$A_{2} = 1.6466 \qquad P_{c} = 4514.73 \text{ kPa}$$

$$A_{3} = -2.0461 \qquad \tau = 1 - T/T_{c}$$

$$A_{4} = -2.8248$$

Figure 5 shows that the results from this work agree with eq 2 to $\pm 0.015\%$ in pressure and smoothly bridge the



Figure 5. Vapor pressure difference (%) from eq 2 proposed by Silva and Weber, 1993: this work (\bigcirc); Silva and Weber, 1993 (\square); Blanke and Weiss, 1992 (\triangle); Higashi et al., 1987 (*); Baehr and Tillner-Roth, 1991 (+); Tamatsu et al., 1992 (\diamondsuit).

Table 3. Saturated Liquid Densities of HFC-152a

<i>T</i> /K	P/kPa	$ ho/{ m mol}{\cdot}{ m dm}^{-3}$	<i>T</i> /K	₽⁄kPa	$ ho/mol \cdot dm^{-3}$
243.106	77.0	15.511	308.381	799.1	13.214
246.676	90.7	15.397	313.280	912.6	13.011
249.455	102.7	15.307	318.308	1041.4	12.795
253.944	124.7	15.162	323.167	1178.3	12.579
259.653	157.9	14.975	328.036	1328.6	12.353
264.452	190.8	14.816	332.874	1491.7	12.119
269.271	229.0	14.653	337.873	1675.6	11.865
272.600	258.7	14.540	342.842	1874.7	11.599
274.130	273.3	14.487	347.771	2089.3	11.318
278.889	322.8	14.322	352.669	2320.6	11.020
283.727	379.9	14.151	357.548	2569.9	10.698
288.636	445.4	13.974	362.377	2836.4	10.350
293.575	519.6	13.791	367.256	3127.1	9.956
298.534	603.2	13.603	371.305	3385.9	9.586
303.502	696.9	13.409			

low-temperature data of Silva and Weber (1993) and the higher temperature data of Baehr and Tillner-Roth (1991).

Saturated Liquid Densities. Saturated liquid densities were deduced by evaluating eq 1 at the vapor pressures determined from eq 2 at temperatures where isotherms of compressed liquid were studied. The 29 saturated liquid densities are listed in Table 3. They were correlated with the following function:

$$\rho = B_1 + B_2 \tau^{1/3} + B_3 \tau^{2/3} + B_4 \tau + B_5 \tau^{4/3} \tag{3}$$

$$B_1 = 5.518774 \text{ mol·dm}^{-3} \qquad T_c = 386.41 \text{ K}$$

$$B_2 = 10.725938 \qquad \tau = 1 - T/T_c$$

$$B_3 = 3.6452398$$

$$B_4 = -0.8747102$$

$$B_5 = 2.7166140$$

Equation 3 reproduces the saturation densities of this work with a standard deviation of $\pm 0.02\%$; see Figure 6. The deviations below 260 K are consistent in sign and magnitude with the deviations seen in Figure 4 for the compressed liquid. The data of Tillner-Roth and Baehr (1993), Blanke and Weiss (1992), and Holcomb et al. (1993) all agree with the smoothed data of this work to within $\pm 0.04\%$ in density throughout most of the temperature range. The data of Sato et al. (1987) show slightly more scatter. This level of agreement between data taken in different laboratories using different techniques is remarkably good.



Figure 6. Saturated liquid density difference (%) from eq 3: this work (\bullet); Tillner-Roth and Baehr, 1993 (\Box); Blanke and Weiss, 1992 (\triangle); Holcomb et al., 1993 (\bigcirc); Sato et al., 1987 (\diamondsuit).

Equation 3 extrapolates to a critical density of 5.519 mol·dm⁻³. Higashi et. al. (1987) report a critical density of 5.571 mol·dm⁻³. Holcomb et al. (1993) report 5.584 mol·dm⁻³ for the critical density of HFC-152a. We consider this to be reasonable agreement given that our extrapolated critical density does not rely on measurements of the density of the saturated vapor.

Summary

We have measured the compressed liquid density and saturation pressure for HFC-152a. The compressed liquid surface ($P\rho T$) has been correlated. The vapor pressures were measured between 280 K and 335 K and agree very well with the vapor pressure correlation developed recently by Silva and Weber (1993). The saturation pressures at 29 different temperatures were used in the compressed liquid density correlation to extrapolate saturated liquid densities. Finally, the saturated liquid densities were correlated. The equations presented in this paper have been chosen only for convenience in correlating data sets. The functional forms of some of the equations traditionally do not perform well at or near the critical point. Caution should be used when evaluating any equation outside the temperature and/or pressure range of the underlying data.

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