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P−*V*−*T* Behavior of 2,3,3,3-Tetrafluoroprop-1-ene (HFO-1234yf) in the Vapor Phase from (243 to 373) K

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The *P*−*V*−*T* properties of 2,3,3,3-tetrafluoroprop-1-ene ($CF_3CF{=}CH_2$, HFO-1234yf), an environmentally friendly refrigerant, were measured using a constant volume apparatus. Measurements were carried out at temperatures from (243 to 373) K and at pressures from (84 to 3716) kPa. A total of 136 experimental points, taken along 12 isochores, were obtained. Our experimental results were compared with a preliminary equation of state. The measurements were also regressed to the Martin−Hou equation of state. No other data on this fluid were found in the literature for the superheated region.

Introduction

HydroFluoroCarbons (HFCs) have been chosen as new refrigerants with no ozone depletion, but they have relatively large values of Global Warming Potential (GWP). As a result, the European Union decided to ban refrigerants with Global Warming Potential (GWP) over 150 in mobile air conditioning. This sector thus needs to find alternatives to the currently used fluid, R-134a (GWP = 1430 for a 100 years time horizon). During the past decade, several refrigerants have been evaluated as possible options in automobile air conditioning, either natural refrigerants (i.e., R-744, carbon dioxide) or "synthetic" refrigerants.[1,2]

A new refrigerant to replace R-134a, a HydroFluoroOlefin called HFO-1234yf ($CF_3CF{=}CH_2$, 2,3,3,3-tetrafluoroprop-1-ene), was recently proposed by the chemical industry. It features thermophysical properties and offers cooling performance similar to R-134a and for this reason requires minimum equipment changes. ASHRAE, in their more recent version of standard 34,[3] designated it as R-1234yf.

This fluid is mildly flammable, featuring a small gap between lower and upper flammability limits, a high ignition energy, and a small burning velocity. It is also thermally stable with no significant corrosion to metals. It is a nonozone-depleting substance, having an atmospheric lifetime of 11 days. Its GWP is approximately 12 for a 20 year time horizon and 4 for a 100 year time horizon,[4] while R-134a is 3590 for a 20 year time horizon and 1420 for a 100 year time horizon.

Regarding toxicity, the data demonstrate a low potential, similar to R-134a, by tests on male rats and mice.[5] As far as the ATEL (Acute Toxicity Exposure Limit) is concerned, HFO-1234yf also has a favorable value (101 000 ppm).

HFO-1234yf thermodynamic properties are very similar to R-134a: boiling point, critical point, and liquid and vapor density are comparable to R-134a.[6] The rise of interest on this specific fluid is also witnessed by several recent papers.[7–14]

In a recent paper from the same research group,[15] the saturated vapor pressure region in a wide temperature range was studied.

In spite of all these favorable considerations, to our best knowledge no experimental data on the superheated vapor region data of this fluid have been published so far in the open literature.

In this paper, the superheated vapor region pressures of this fluid were measured by means of an isochoric apparatus. Data were collected over a wide temperature range, from (243 to 373) K. Experimental results were compared with REFPROP 8.0 prediction,[16] obtained with a preliminary equation developed also with present data,[17] and were fitted with the Martin−Hou equation of state.[18]

Experimental Section

Materials. The sample was produced by the French group Arkema and donated by Centro Ricerche FIAT, Italy. It was then degassed to remove air and other noncondensable gases by immersing it in liquid nitrogen and evacuating. It was then brought to room temperature and was again subjected to the freezing, evacuating, and thawing process. This procedure was repeated several times. Its purity was checked by gas chromatography using a thermal conductivity detector and was found to be better than 99.95 % on a molar basis by analysis of peak area.

Apparatus. The basic experimental setup has already been described elsewhere,[19] so it is only briefly outlined here. Two twin thermostatic baths were filled with different silicone oils (Baysilone M10 and Baysilone M100, Bayer). After charging with the sample, the setup could be operated over two temperature ranges, approximately from (210 to 290) K and from (290 to 360) K, depending on which bath was used. The two silicone oils have different kinematic viscosity values [(10 and 100) cSt at room temperature, respectively]. The one with lower kinematic viscosity, due to its higher volatility, was applied only for the low-temperature range, while that with a greater viscosity was applied only at high temperatures. The spherical cells and pressure transducer are immersed in one of the two thermostatic baths. An auxiliary thermostat was used to reach below-ambient temperatures. The cell volume was

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Table 1. Experimental $P-V-T$ Data for R134a and Deviations with REFPROP 8.0[10]

T / K	P / kPa	V / $dm^3 \cdot mol^{-1}$	P_{REF} / kPa	dP / kPa	dP / %
		m/g = 1.486			
253.09	107.6	18.7	107.6	0.0	0.01
282.80	121.4	18.8	121.5	−0.1	0.06
292.89	126.2	18.8	126.1	0.1	0.06
313.10	135.4	18.8	135.3	0.1	0.06
333.03	144.3	18.8	144.3	0.0	0.02
352.93	153.2	18.8	153.2	0.0	0.01
372.84	162.1	18.9	162.1	0.0	0.00
		m/g = 2.053			
272.92	158.9	13.6	159.4	−0.4	0.28
292.87	171.9	13.6	172.3	−0.4	0.25
313.10	184.9	13.6	185.3	−0.4	0.20
333.05	197.5	13.6	197.9	−0.3	0.18
353.04	210.0	13.6	210.4	−0.4	0.17
372.99	222.4	13.6	222.8	−0.3	0.15
		m/g = 4.282			
282.69	329.1	6.5	329.4	−0.3	0.10
302.58	357.7	6.5	358.0	−0.3	0.09
323.07	386.5	6.5	386.7	−0.2	0.06
342.96	413.7	6.5	414.0	−0.3	0.07
362.89	440.7	6.5	441.0	−0.3	0.07
372.82	454.0	6.5	454.3	−0.3	0.07
		m/g = 22.412			
333.03	1610.8	1.25	1610.9	−0.1	0.00
343.00	1706.9	1.25	1707.7	−0.8	0.05
352.94	1798.6	1.25	1800.0	−1.4	0.08
362.90	1888.6	1.25	1891.1	−2.5	0.13
372.87	1976.6	1.25	1979.2	−2.6	0.13

estimated to be (273.5 ± 0.3) cm^3 at room temperature,[19] and the cell volume change with temperature was taken into account.[20,21] The pressure and temperature data acquisition systems were identical to those of the previous apparatus.[20,21] A PID device was used to control the temperature, which was measured using a calibrated resistance thermometer; the total uncertainty of the temperature measurements was ± 0.02 K. The charging procedure has been described elsewhere.[22] The uncertainty in the measurement of the mass inside the cell was estimated to amount to ± 5 mg. The volume of the cell, piping, and pressure transducer cavity is measured with an uncertainty of ± 0.0003 dm^3. From the uncertainties in the mass and volume measurements, the uncertainty in calculated molar volume was estimated to be about 1 %.

The uncertainty in the pressure measurements stems from the uncertainty of the transducer and null indicator system and the pressure gauges. The uncertainty of the digital pressure indicator (Ruska, mod. 7000) is ± 0.003 % of its full scale. The total uncertainty in the pressure measurement, considering also the temperature fluctuations due to bath instability, was found to be always lower than ± 0.8 %.

Results and Discussion

To check the reliability of the experimental setup, 24 superheated vapor region points along four isochores were taken for R-134a approximately in the same temperature range (from (253 to 373) K) of the measurements taken for HFO-1234yf. This fluid was chosen as a reference because of its very well-known thermophysical properties, and the Tillner−Roth equation of state[23] implemented in REFPROP 8.0 is able to calculate densities for R-134a with typical uncertainties of 0.05 %. In Table 1, the experimental data for R-134a are reported together with deviations with data calculated by REFPROP 8.0 calculations. Absolute and relative deviations were reported in Figures 1 and 2, respectively. Defining AAD (P) as

$$\mathrm{AAD}\,(P) = \frac{1}{n}\sum_{i=1}^{n}[|(P_{\mathrm{exp}} - P_{\mathrm{calc}})/P_{\mathrm{exp}}|]\cdot 100 \qquad (1)$$

the measured data were well represented by the equation of state, and deviations showed an AAD (P) = 0.1 %.

In total, 136 experimental points for HFO-1234yf were collected along 12 isochores in the superheated vapor region. The data are reported in Table 2. The $P-V-T$ measurements were taken in a temperature range from (243 to 373) K, at pressures from (84 to 3716) kPa, and for molar volumes from (0.25 to 22.91) $dm^3 \cdot mol^{-1}$. For the calculations, the molar masses of 102.03 $g \cdot mol^{-1}$ and 114.04 $g \cdot mol^{-1}$ were adopted for R134a and R1234yf, respectively.

Since no experimental data were available in the open literature, our experimental results were compared with the preliminary equation of state[16,17] developed at NIST and reported in Figures 3 and 4. Deviations showed an AAD (P) = 0.34 %. Results compared with REFPROP 8.0 prediction were well within the experimental uncertainty (calculated to be always lower than 0.8 %), shown with the dashed line in Figure 4.

The experimental $P-V-T$ measurements were also fitted with the Martin−Hou equation of state in its original expression[18]

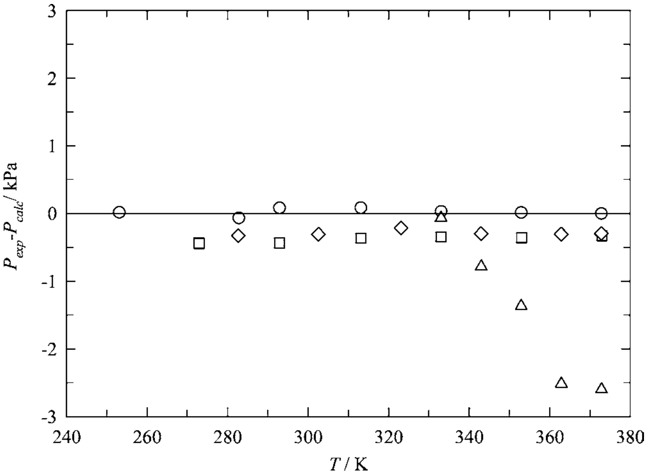

Figure 1. Scatter diagram of absolute pressure deviations produced from REFPROP 8.0 for R134a. ○, m = 1.486 g; □, m = 2.053 g; ◇, m = 4.282 g; △, m = 22.414 g.

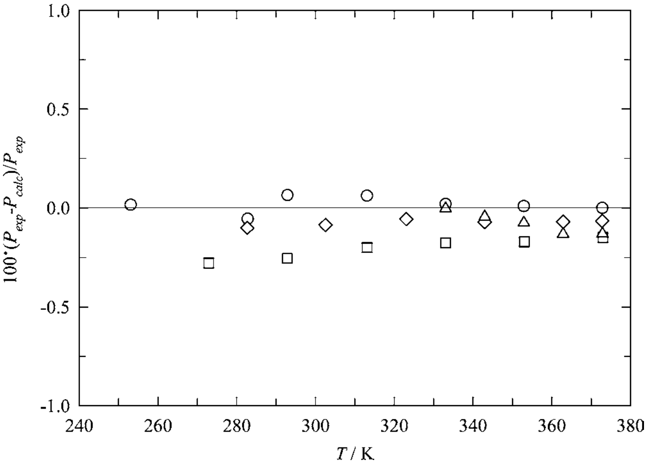

Figure 2. Scatter diagram of relative pressure deviations produced from REFPROP 8.0 for R134a. ○, m = 1.486 g; □, m = 2.053 g; ◇, m = 4.282 g; △, m = 22.414 g.

Table 2. Experimental $P-V-T$ Data for HFO-1234yf

T/K	P/kPa	V/$dm^3 \cdot mol^{-1}$	T/K	P/kPa	V/$dm^3 \cdot mol^{-1}$
	m/g = 1.367		362.86	534.0	5.31
243.08	84.5	22.8	367.83	542.5	5.31
248.02	86.5	22.8	372.81	550.9	5.31
249.15	87.0	22.8		m/g = 6.669	
253.07	88.5	22.8	282.75	430.2	4.68
258.03	90.4	22.8	287.93	447.1	4.68
263.08	92.4	22.8	292.81	457.2	4.68
267.98	94.2	22.8	297.89	467.6	4.68
273.04	96.2	22.8	302.91	477.9	4.68
277.99	98.0	22.8	308.11	488.4	4.68
282.95	99.9	22.8	313.11	498.5	4.68
287.79	101.7	22.8	318.09	508.5	4.68
292.76	103.6	22.8	323.07	518.3	4.68
297.80	105.5	22.8	328.06	528.3	4.69
303.12	107.4	22.8	333.04	538.1	4.69
308.11	109.3	22.8	333.05	537.8	4.69
313.11	111.2	22.8	338.03	547.6	4.69
318.10	113.0	22.8	343.01	557.3	4.69
323.08	114.9	22.9	347.99	567.0	4.69
328.07	116.8	22.9	352.96	576.6	4.69
333.04	118.8	22.9	357.93	586.2	4.69
338.03	120.6	22.9	362.92	595.8	4.69
343.00	122.5	22.9	367.89	605.3	4.69
347.97	124.3	22.9	372.87	614.8	4.69
352.96	126.1	22.9		m/g = 7.481	
357.92	128.0	22.9	293.10	508.2	4.17
362.89	129.8	22.9	303.12	531.3	4.17
367.87	131.9	22.9	313.11	554.0	4.17
372.82	133.8	22.9	323.09	576.5	4.18
	m/g = 1.629		333.05	598.7	4.18
243.07	98.0	19.1	343.01	620.6	4.18
253.01	105.1	19.1	352.96	642.4	4.18
262.97	109.8	19.1	362.92	664.0	4.18
272.92	114.4	19.1	372.88	685.4	4.18
282.62	118.8	19.1		m/g = 10.518	
292.99	123.5	19.2	303.11	703.6	2.97
303.12	128.1	19.2	313.11	738.1	2.97
313.12	132.7	19.2	323.09	771.8	2.97
323.10	137.1	19.2	333.05	804.7	2.97
333.06	141.6	19.2	343.02	837.4	2.97
342.99	146.1	19.2	352.96	869.4	2.97
352.98	150.6	19.2	362.93	901.2	2.98
362.93	155.0	19.2	372.84	932.4	2.98
372.88	159.4	19.2		m/g = 12.327	
	m/g = 2.621		318.11	861.5	2.53
262.93	171.7	11.9	328.06	901.7	2.53
272.84	179.3	11.9	338.02	941.5	2.54
282.68	186.6	11.9	347.98	980.2	2.54
292.58	193.9	11.9	357.93	1018.7	2.54
302.70	201.4	11.9	367.88	1057.0	2.54
313.12	209.0	11.9	372.85	1075.9	2.54
323.09	216.2	11.9		m/g = 23.602	
333.06	223.4	11.9	338.00	1538.8	1.32
343.00	230.6	11.9	347.94	1624.6	1.32
352.98	237.7	11.9	357.95	1709.0	1.33
362.96	244.9	11.9	367.92	1791.4	1.33
372.88	252.0	11.9		m/g = 37.550	
	m/g = 5.893		347.98	2157.7	0.833
283.00	394.6	5.29	352.99	2237.2	0.833
287.99	403.8	5.29	357.95	2314.1	0.833
292.95	412.8	5.29	362.89	2389.2	0.833
298.08	422.1	5.30	367.88	2463.9	0.834
303.09	431.1	5.30	372.85	2537.2	0.834
308.10	439.9	5.30		m/g = 39.663	
313.09	448.7	5.30	347.94	2217.8	0.789
318.09	457.5	5.30	352.91	2303.2	0.789
323.08	466.1	5.30	357.86	2385.6	0.789
328.06	474.7	5.30	362.83	2467.7	0.789
333.04	483.3	5.30	367.80	2548.9	0.789
338.01	491.8	5.30	372.80	2629.5	0.789
342.99	500.3	5.31		m/g = 124.890	
347.96	508.8	5.31	362.92	3067.5	0.251
352.91	517.2	5.31	367.92	3386.9	0.251
357.88	525.6	5.31	372.88	3715.9	0.251

$$P = \frac{RT}{(v-b)} + \frac{A_2 + B_2T + C_2e^{-5.475T/T_c}}{(v-b)^2} + \frac{A_3 + B_3T + C_3e^{-5.475T/T_c}}{(v-b)^3} + \frac{A_4}{(v-b)^4} + \frac{B_5T}{(v-b)^5} \qquad (2)$$

where

$$b = V_C - \frac{\beta V_C}{15Z_C} \qquad (3)$$

$$Z_C = \frac{P_CV_C}{RT_C} \qquad (4)$$

$$\beta = 20.533Z_C - 31.883Z_C^2 \qquad (5)$$

$$T' = T_C(0.9869 - 0.675T_C) \qquad (6)$$

The Martin–Hou EoS was regressed minimizing the AAD (P) and acting on two parameters: $M = (P - P_c)/(T - T_c)$ and T_B, the Boyle Temperature. According to the original paper,[18] T_B

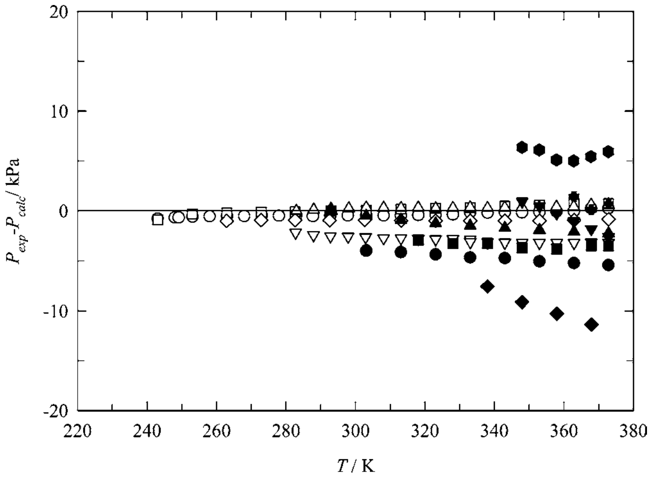

Figure 3. Scatter diagram of absolute pressure deviations produced from REFPROP 8.0 for HFO-1234yf. ○, m = 1.367 g; □, m = 1.629 g; ◇, m = 2.621 g; △, m = 5.893 g; ▽, m = 6.669 g; ▲, m = 7.481 g; ●, m = 10.518 g; ■, m = 12.327 g; ◆, m = 23.602 g; ▼, m = 37.550 g; ⬢, m = 39.663 g; +, m = 124.890 g.

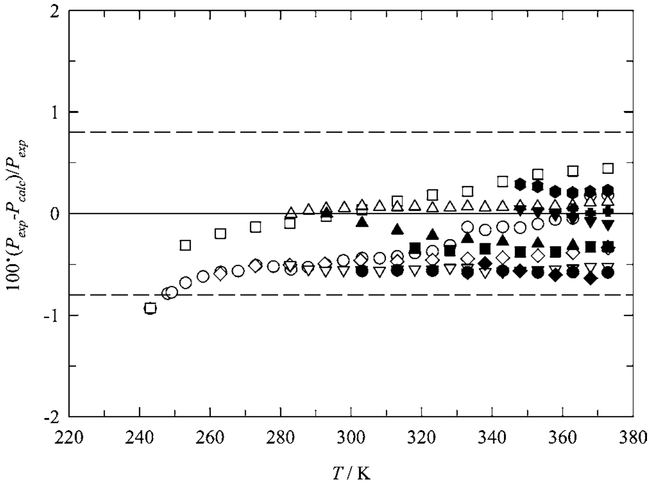

Figure 4. Scatter diagram of relative pressure deviations produced from REFPROP 8.0 for HFO-1234yf. The dashed line represents the experimental uncertainty. ○, m = 1.367 g; □, m = 1.629 g; ◇, m = 2.621 g; △, m = 5.893 g; ▽, m = 6.669 g; ▲, m = 7.481 g; ●, m = 10.518 g; ■, m = 12.327 g; ◆, m = 23.602 g; ▼, m = 37.550 g; ⬢, m = 39.663 g; +, m = 124.890 g.

Table 3. Parameters for the Martin−Hou EoS

critical parameters			fitted parameters		equation constants							
T_C	P_C	V_C	T_{Boyle}									
K	kPa	$dm^3 \cdot mol^{-1}$	K	M	A_2	B_2	C_2	A_3	B_3	C_3	A_4	B_5
367.95	3382.0	0.2386	551.66	0.752	−28999357.58	48775.01	29307214.31	4787634452.68	−7552243.73	−5642815560.72	−181061886091.12	18047800638.03

spanned from (500 to 1000) K, while the M value spanned from 0.1 to 0.9, taking as a reference the values obtained from the critical isometric estimated by REFPROP 8.0.[16] All the obtained parameters were summarized in Table 3.

To minimize the AAD (P), the 1 plus 1 Evolution Strategy technique[24] based on the evolutionary fast algorithm was developed.

Comparing eq 2 with the experimental temperature and volume data, the deviations from the experimental pressure were calculated for each data point, and the results are shown in Figures 5 and 6. Deviations showed an AAD (P) = 0.44 %, and few points of the results in Figure 6 were out of the experimental uncertainty limit, again shown with the dashed line.

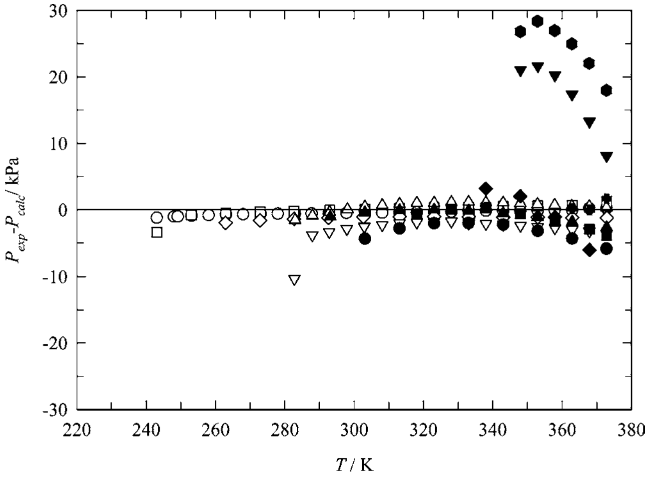

Figure 5. Scatter diagram of absolute pressure deviations produced from from eq 2 for HFO-1234yf. ○, m = 1.367 g; □, m = 1.629 g; ◇, m = 2.621 g; △, m = 5.893 g; ▽, m = 6.669 g; ▲, m = 7.481 g; ●, m = 10.518 g; ■, m = 12.327 g; ◆, m = 23.602 g; ▼, m = 37.550 g; ⬢, m = 39.663 g; +, m = 124.890 g.

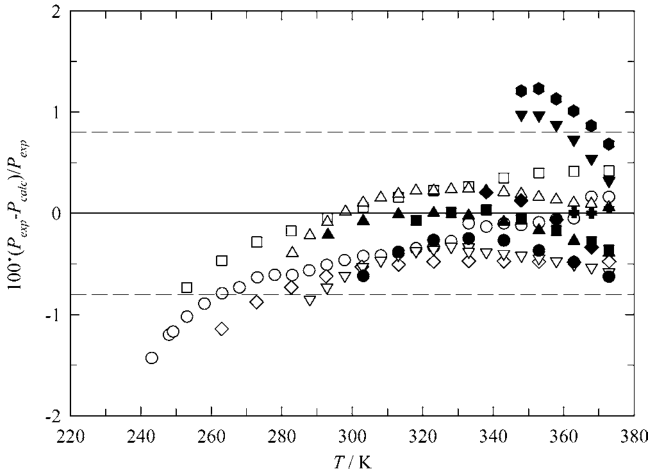

Figure 6. Scatter diagram of relative pressure deviations produced from eq 2 for HFO-1234yf. The dashed line represents the experimental uncertainty. ○, m = 1.367 g; □, m = 1.629 g; ◇, m = 2.621 g; △, m = 5.893 g; ▽, m = 6.669 g; ▲, m = 7.481 g; ●, m = 10.518 g; ■, m = 12.327 g; ◆, m = 23.602 g; ▼, m = 37.550 g; ⬢, m = 39.663 g; +, m = 124.890 g.

However, both predictions, excluding two series, generally showed the same trend of deviations for the different isochores. In addition, the isochores with lower charge showed a slight increase of deviation with the lowering of temperatures for both models.

Conclusions

The measurements of 136 experimental points along 12 isochores for the superheated vapor region were obtained using a constant-volume apparatus for HFO-1234yf. To check the reliability of the experimental setup, 24 experimental superheated vapor points along four isochores were taken for R-134a in the same temperature range of the present paper measurements, and good consistency with REFPROP 8.0 was found. The experimental data were compared with a preliminary equation of state developed also with present data. The experimental points were also regressed with the Martin−Hou equation of state. Results were generally within the experimental uncertainty limit.

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