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Density of 1,1-Dichloro-1-Fluoroethane (HCFC 141b) as a Function of Temperature and Pressure[1]

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The density of HCFC 141b has been measured at several temperatures between 260 and 320 K, and pressures up to 20 MPa, with a mechanical oscillator densimeter. The densimeter was calibrated with 2,2,4-trimethylpentane, whose density was obtained from a correlating equation with 0.3% uncertainty. The density data obtained for HCFC 141b has a reproducibility of 0.05%, and an uncertainty of 0.3%. The data obtained were fitted to a Tait-type equation, which reproduced the experimental densities within 0.11% and were compared with the data obtained in other works.

KEY WORDS: density; HCFC 141b; pressure; temperature; Tait equation.

1. INTRODUCTION

The thermophysical properties of environmentally acceptable refrigerants are very important for obtaining a correct design of the new equipment for the refrigeration, air conditioning and polymer foam industries. HCFC 141b is a possible replacement for CFC 11 as a blowing agent for expanded thermal insulation foams. It has an ozone depletion potential = 0.1 and a global warming potential = 150, compared with the corresponding values for CFC 11 of 1 and 1500. Data for the density of this fluid in the liquid region are scarce, and restricted mostly to the low-pressure side of the phase diagram.

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In our current program of research, we have measured the density of 1,1-dichloro-1-fluoroethane (HCFC 141b) as a function of temperature and pressure, with a mechanical oscillator densimeter.

2. EXPERIMENTS

In these measurements a mechanical oscillator densimeter Anton Paar-type DMA60, equipped with a high-pressure cell DMA 512, was used. Details of the instruments and the system coupled to measure the temperature and pressure and to fill the cell are described in previous publications [1, 2].

This type of densimeter operates in a relative mode. In this work the densimeter was calibrated with 2,2,4-trimethylpentane (purity, 99.8%) and vacuum, between 261 and 323 K and pressures up to 20 MPa. The linearity of the instrument has been verified before with several calibrants, with densities ranging from 170 to 1600 $\mathrm{kg \cdot m^{-3}}$ [1]. The working equation in this case is [2, 3]

$$\rho = \frac{\rho_1(\tau^2 - \tau_0^2)}{\tau_1^2 - \tau_0^2} \tag{1}$$

where ρ_1 and τ_1 are the density and the vibration period of the calibrating substance—2,2,4-trimethylpentane. τ_0 is the vibration period of the cell in vacuum and τ is the vibration period of the sample—HCFC 141b.

The densities of 2,2,4-trimethylpentane were obtained from the work of Malhotra and Woolf [4], who recommended, after IUPAC, this fluid as a secondary calibration fluid for temperatures in the range 273.15 to 373.15 K and pressures up to 300 MPa, with a maximum uncertainty of 0.1%. Those authors also proposed a Tait equation to correlate the variation of density with pressure and temperature that could reproduce all the available experimental data in the literature within 0.1%, and capable of predicting the density of 2,2,4-trimethylpentane to within 0.3%, between 170 and 473 K and pressures up to 980 MPa. The vibration period of 2,2,4-trimethylpentane was correlated as a function of temperature (K) and pressure (MPa) by a Tait-type equation, as described earlier [3]:

$$\tau_1^2 = \tau_R^2(T) \Big/ 1 - E \ln\left(\frac{F(T) + P}{F(T) + P_R}\right) \tag{2}$$

$$\tau_R^2(T) = 13.802 + 4.7694 \times 10^{-3} T \tag{3}$$

$$F(T) = 0.97861 + 1.4671 T - 3.4288 \times 10^{-3} T^2 \tag{4}$$

The parameter E was found to be equal to 2.1672×10^{-2}. τ_R is the vibration period at a reference pressure P_R, taken as 0.1 MPa, E and F are empirical constants. Equation (2) reproduced the experimental values of τ_1^2 within the experimental error.

The vibration period of the cell in vacuum was fitted as a function of temperature through Eq. (5):

$$\tau_0 = 3.479284 + 8.18 \times 10^{-4} T \tag{5}$$

This equation reproduces the experimental values of τ_0 with an uncertainty of 0.01 %.

The temperature inside the system during the measurements was determinated with a Thermocoax-type E thermocouple, within 0.07 K, on IPTS 68. The results were converted to ITS 90. The pressure was measured with a pressure transducer, Sedeme-TS 205, connected to a pressure sensor, Sedeme-CMR, within 8 kPa. The e.m.f. of the thermocouple and the signal of the pressure sensor were determined with a multimeter Keithley 195.

The vibration period of a sample of HCFC 141b, with an estimated purity of 99.8 %, was measured between 260 and 320 K and pressures up to 20 MPa with an experimental uncertainty less than 1×10^{-5}.

The reproducibility of the density data obtained with this instrument has been proved to be of the order of 0.01 % [3]. Some of the present

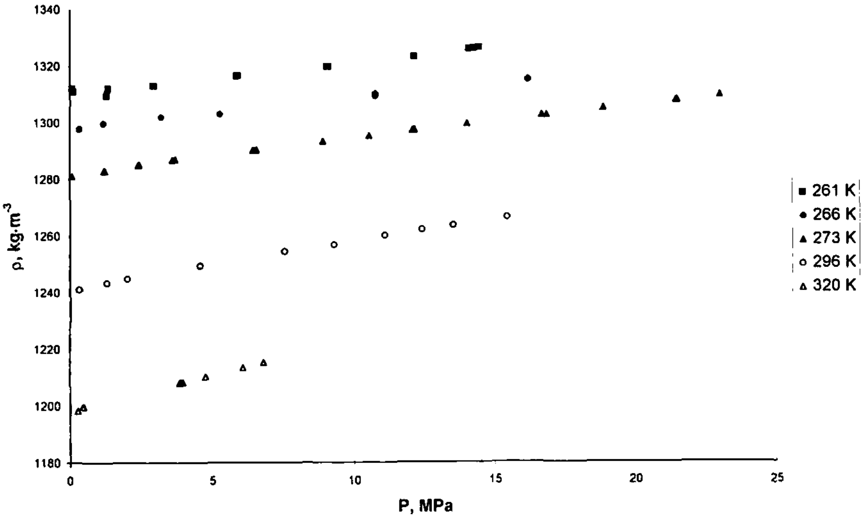

Fig. 1. Experimental $P\rho T$ data for compressed liquid HCFC 141b.

measurements, however, are reproducible only to 0.05%. Because the density data of the calibrating fluid was obtained from a correlation equation obtained from Ref. [4], with an uncertainty of 0.3%, the uncertainty of the data reported in this paper is 0.3%.

3. RESULTS

A set of 304 experimental values of the vibration period of HCFC 141b and the densities calculated by application of Eq. (1) was obtained. Table I presents a sampling of these data averaged for close values of pressure, for each pseudoisotherm. The error introduced by the average processes is much smaller than the uncertainty of the experimental data. Figure 1 shows all experimental data obtained for compressed liquid HCFC 141b.

Table I. Density of 1,1-Dichloro-1-fluoroethane (HCFC 141b)

T (K)	P (MPa)	ρ ($\mathrm{kg \cdot m^{-3}}$)
260.37	0.145	1311.0
260.01	0.100	1312.0
259.98	0.100	1312.2
261.24	1.313	1309.7
261.24	1.316	1309.7
260.54	1.358	1312.2
260.49	1.354	1312.3
260.49	1.354	1312.3
260.85	2.949	1313.1
260.85	2.964	1313.1
260.86	5.875	1316.6
260.86	5.902	1316.7
260.85	5.925	1316.7
260.85	5.917	1316.8
260.97	9.053	1320.0
260.97	9.099	1320.0
260.96	9.100	1320.1
260.91	12.137	1323.6
260.67	14.064	1326.5
260.67	14.232	1326.7
260.67	14.422	1327.0

Table I. *(Continued)*

T (K)	P (MPa)	ρ ($kg \cdot m^{-3}$)
266.54	0.366	1297.8
266.54	0.366	1297.8
266.49	0.366	1298.0
266.29	1.201	1299.9
266.29	1.204	1300.0
266.43	3.224	1302.1
266.43	3.225	1302.1
266.97	5.303	1303.2
266.92	10.760	1310.1
266.92	10.760	1310.1
267.06	10.775	1309.6
266.84	10.775	1310.4
266.87	10.790	1310.3
267.12	16.150	1315.6
267.08	16.161	1315.7
267.08	16.162	1315.8
273.66	0.100	1281.2
273.66	1.230	1283.1
273.66	1.256	1283.1
273.68	2.424	1285.1
273.68	2.461	1285.2
273.68	3.619	1286.9
273.68	3.715	1287.1
273.68	6.454	1290.3
273.68	6.487	1290.4
273.68	6.511	1290.4
273.68	6.575	1290.5
273.68	6.584	1290.5
273.68	8.903	1293.6
273.68	10.550	1295.7
273.68	12.080	1297.7
273.68	12.110	1297.7
273.68	12.150	1297.7
273.68	14.000	1300.0
273.68	14.006	1300.1
273.68	16.649	1303.2
273.68	16.815	1303.3
273.68	18.840	1305.7
273.68	18.837	1305.7
273.68	21.436	1308.7
273.68	21.460	1308.7
273.68	21.485	1308.7
273.68	23.020	1310.4
273.68	23.017	1310.4

Table I. *(Continued)*

T (K)	P (MPa)	ρ ($kg \cdot m^{-3}$)
296.20	0.340	1241.0
296.20	0.367	1241.1
296.30	1.319	1243.4
296.30	1.324	1243.5
296.28	2.041	1244.9
296.37	4.589	1249.4
296.31	4.598	1249.6
296.40	7.557	1254.5
296.40	7.570	1254.6
296.56	9.295	1257.0
296.44	11.102	1260.3
296.41	12.404	1262.5
296.48	13.502	1263.9
296.48	13.513	1264.0
296.47	15.408	1266.9
320.12	0.286	1198.3
320.12	0.286	1198.3
320.08	0.463	1199.8
320.08	0.488	1199.9
320.10	3.858	1208.2
320.10	3.887	1208.2
320.10	3.972	1208.5
320.10	4.782	1210.4
320.07	6.098	1213.5
319.99	6.818	1215.3

4. DISCUSSION

The values of the densities of HCFC 141b, presented in Table I, have an estimated reproducibility of 0.05%. A fit as a function of temperature and pressure, through a Tait-type equation, similar to Eqs. (2) to (4), was made:

$$\rho = \rho_R(T) \Big/ 1 - C \ln\left(\frac{D(T) + P}{D(T) + P_R}\right) \tag{6}$$

$$\rho_R(T) = 1799.5 - 1.8826T \tag{7}$$

$$D(T) = -177.4 + 2.323T - 5.291 \times 10^{-3} T^2 \tag{8}$$

In Eqs. (6), (7), and (8) the density is expressed in $kg \cdot m^{-3}$, T in K, and P in MPa. The parameter C was found to be equal to 0.06490. ρ_R is the density at a reference pressure P_R, taken as 0.1 MPa, and C and $D(T)$ are an empirical constant and a temperature-dependent function, respectively. Equation (7) reproduces the extrapolated densities to within 0.17%. The complete set of equations can reproduce the densities of HCFC 141b with an error less than 0.11%, and it is valid between 260 and 320 K and pressures up to 19.8 MPa. Figure 2 shows the deviations of the data from Eq. (6). It can be seen that the deviations never exceed 0.25%.

We can compare our results with the data of Maezawa et al. [5], Kumagai and Takahashi [6], Defibaugh et al. [7], Matsuo et al. [8], and Malhotra and Woolf [9] and with data obtained by us previously with the same method with a densimeter at atmospheric pressure [10]. Figure 3 shows the deviations from Eq. (6). The deviation in relation to the saturation line obtained by Maezawa et al. is about −0.7%, increasing to −1.1% for the end of the temperature range; for the compressed liquid, the maximum deviation is −1.1% at the higher pressure. The deviations of the values presented in the work of Kumagai and Takahashi are about −0.3%, except for 323.15 K, where it is −0.7%. The deviation in relation to the saturation line proposed by Defibaugh et al. varies from −0.3 to −0.5%; for the compressed liquid the deviations range from −0.2% in the middle of the temperature range to −0.6% in the end of the range. The deviations of the data of Defibaugh et al. are smaller than the deviations of

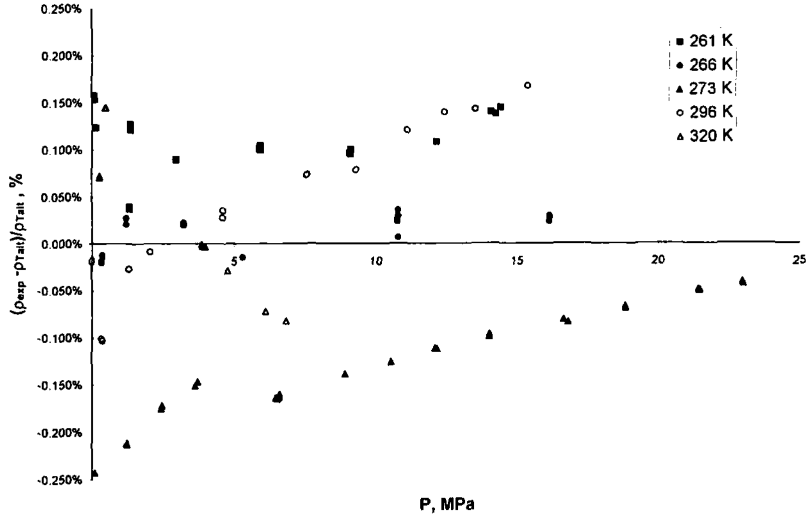

Fig. 2. Deviations of experimental data from values calculated with Eq. (6).

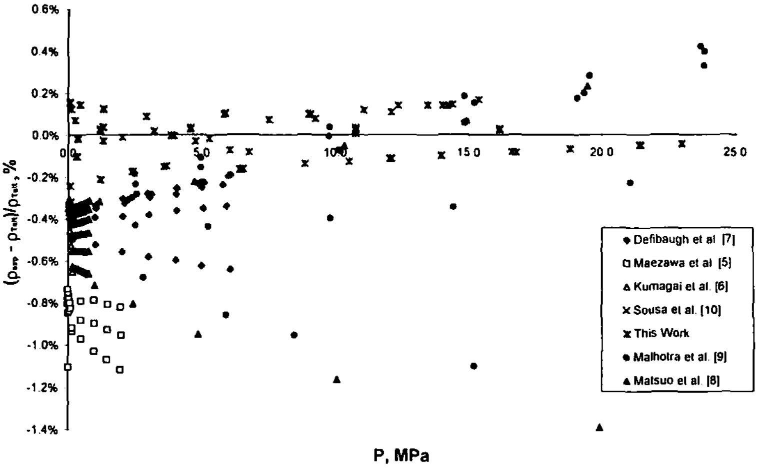

Fig. 3. Deviations of the densities of HCFC 141b, obtained by Eq. (6), and the values reported in other works.

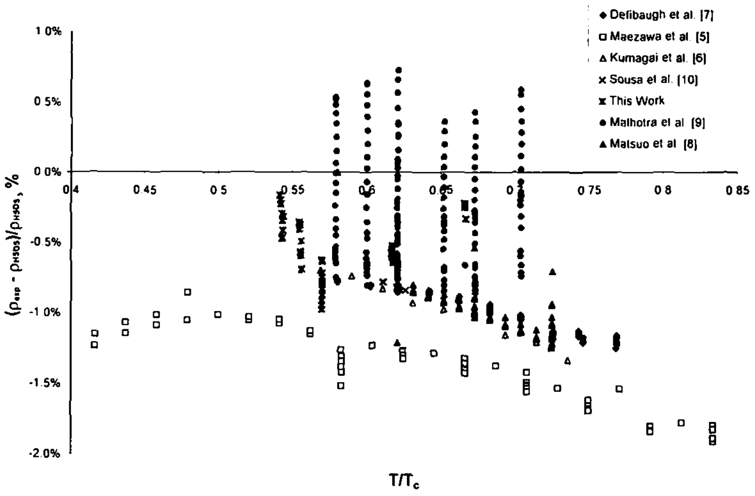

Fig. 4. Comparison between the densities of HCFC 141b obtained by different authors. The baseline is the prediction from the hard-sphere DeSantis reduced equation of state developed by Fialho and Nieto de Castro (11).

the data of Maezawa et al. for the same pressure (about one-third). Matsuo et al. [8] and Malhotra and Woolf [9] extended the measurements to very high pressure. Within the overlap in pressure, the deviations of Matsuo et al. [8] vary from 0.3 to −1.3%, while those of Malhotra and Woolf [9] deviate from −1.1 to 0.4%. Our previous data at atmospheric pressure [10] do not deviate by more than 0.3%.

Figure 4 shows the comparison of all sets of data with the predictions obtained with a general scheme for the estimation of halocarbon liquid-state densities based on a modified hard-sphere DeSantis equation of state developed by Fialho and Nieto de Castro [11]. This scheme estimates the density of pure liquids solely from the critical constants and molecular weight to better than 1.5% for $T^* < 0.9$. The deviations of all sets of data do not amount to more than 2.0% (Maezawa et al.), a general trend obtained previously for several halocarbons. It is remarkable that the correlation predicts the densities of this fluid to very high pressures (400 MPa) to within 1%, when in its development no data taken at pressures greater than 30 MPa were used for any fluid.

5. CONCLUSIONS

The density of HCFC 141b as a function of pressure for different temperatures has been measured with a vibrating tube densimeter with an estimated reproducibility of 0.05% and an uncertainty of 0.3%. The comparison with existing data shows a very good agreement at low pressures and an agreement within 1.3% over the whole pressure range measured. We conclude that the data reported agree well with the data reported previously, although some of the authors deviate between themselves and with our data by more than the mutual uncertainties. This is possibly due to the presence of impurities in the sample measured. This has proven to be one of the major obstacles in comparing the results of different authors, especially for the measurements obtained earlier, as HCFC 141b is very difficult to obtain with a high purity. The presence of impurities, especially water, can mask the results and modify the dependence on pressure encountered in the different sets of measurements. The quoted impurity of our samples is 99.8%, while that of Maezawa et al. [5] is 98.8%, that of Defibaugh et al. [7] is 99.9%, and that of Matsuo et al. [8] is 99.9%. The data of Malhotra and Woolf [9] were obtained with a sample of unspecified purity.

The present results seem to recommend further measurements of the density of HCFC 141b in the compressed liquid region to ascertain the reason for these discrepancies, possibly establishing a round-robin exercise with the same samples.

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REFERENCES

1. A. Tavares Sousa, C. Nieto de Castro, R. Tufeu, and B. Le Neindre, *High Temp.-High Press.* **24**:185 (1992).
2. A. Tavares Sousa and C. Nieto de Castro, submitted for publication.
3. A. Tavares Sousa, P. Fialho, C. Nieto de Castro, R. Tufeu, and B. Le Neindre, *Fluid Phase Equil.* **80**:213 (1992).
4. R. Malhotra and L. A. Woolf, *Int. J. Thermophys.* **11**:1059 (1990).
5. Y. Maezawa, H. Sato, and K. Watanabe, *J. Chem. Eng. Data* **36**:151 (1991).
6. A. Kumagai and S. Takahashi, *Int. J. Thermophys.* **14**:339 (1993).
7. D. R. Defibaugh, A. R. H. Goodwin, G. Morrison, and L. A. Weber, *Fluid Phase Equil.* **85**:271 (1993).
8. S. Matsuo, Y. Tanaka, H. Kubota, and T. Makita, *J. Chem. Eng. Data* **39**:903 (1994).
9. R. Malhotra and L. A. Woolf, *Fluid Phase Equil.* **92**:195 (1994).
10. A. T. Sousa, P. Fialho, and C. Nieto de Castro, *Int. J. Thermophys.* **15**:375 (1994).
11. P. S. Fialho and C. A. Nieto de Castro, *Fluid Phase Eq.* (1995), in press.