Density and Viscosity Measurements of 1,1,1,2-Tetrafluoroethane (HFC-134a) from 199 K to 298 K and up to 100 MPa

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New density results for liquid 1,1,1,2-tetrafluoroethane (HFC-134a) along five isotherms from 199 K to 298 K and at pressures up to 100 MPa are presented. The measurements were performed with a new vibrating-wire instrument operated in the forced mode of oscillation and were carried out on a round-robin sample of refrigerant HFC-134a. The viscosity of the fluid was measured simultaneously with the density, using the same vibrating-wire sensor, at temperatures above 248 K over the entire pressure range. The results are compared with recent literature data. The precision of the present values is ±0.03% for the density and ±0.6% for the viscosity. The corresponding estimated accuracies are ±0.05% and ±2.5%, respectively.

Introduction

The hydrofluorocarbon 1,1,1,2-tetrafluoroethane (HFC-134a) is a possible substitute for dichlorodifluoromethane (CFC-12) in refrigeration applications. The absence of chlorine in HFC-134a makes it less harmful to the ozone layer than CFC-12. As the use of alternative refrigerants spreads, a better knowledge of their properties is required. The amount of thermodynamic data available for HFC-134a has increased greatly in the last years, and an international standard equation of state has been formulated (Tillner-Roth and Baehr, 1994). In the field of transport properties, divergences among reported values for HFC-134a has led to the organization of a round-robin set of measurements of viscosity and thermal conductivity (Assael *et al*., 1995) to investigate the causes of discrepancies, such as impurities.

The present measurements were made on one of the round-robin samples but are additional to the tests of transport properties. Density and viscosity data for HFC-134a are still scarce at low temperatures in the compressed liquid, and the results reported here extend the pressure range of the sets found in the literature. Our results were obtained using a vibrating-wire technique in the forced mode. The instrument has been presented and validated elsewhere (Pádua *et al*., 1994, 1995), where its ability to perform accurate measurements up to high pressures and in a wide temperature range was assessed. Although this instrument has been biased toward a high sensitivity to the fluid density, the working equations allow the viscosity of the fluid to be obtained simultaneously, as a secondary result of the density measurements.

Method

The instrument here employed was designed primarily as a hydrostatic weighing densimeter. In this class of methods, the density of a fluid is determined by measuring the buoyancy force acting on a sinker of known volume. It is advantageous to have a simple-shaped sinker, so that its volume can be known accurately at different conditions of temperature and pressure. This makes hydrostatic techniques more accurate in principle than volumetric methods. In the latter, the accuracy is limited by a deficient knowledge of the volume of the measuring cell, requiring calibration over the entire range of experimental conditions, and by the absence of reference fluids at conditions away from ambient. Despite their advantage in terms of accuracy, the use of hydrostatic densimeters at high pressures has been conditioned by difficulties in the measurement of the apparent weight of the sinker inside a thick-walled pressure vessel. The main difference between the present method and other hydrostatic techniques (Wagner *et al*., 1995; Haynes *et al*., 1976; Hales and Gundry, 1983) is that no magnetic suspension mechanism is used to transmit the buoyancy force to an exterior balance. Instead, a vibrating-wire sensor replaces the balance and is located inside the cell.

In the vibrating-wire densimeter, the sinker is suspended vertically from a thin tungsten wire whose tension depends upon the buoyancy force. To measure this tension, the wire is forced to undergo transverse vibrations and its resonance curve is plotted. The system for driving the wire and detecting its motion is based on an electromagnetic coupling: the wire is placed inside a permanent, uniform magnetic field and a sinusoidal electrical current of specified frequency is fed through it to provide a driving force; the velocity amplitude of the wire is then deduced from the electromotive force arising from the motion of the wire inside the magnetic field (Pádua *et al*., 1994, 1995). The difference between the resonant frequencies observed in the fluid and in vacuum is sensitive mainly to the fluid density, whereas the change in the width of the resonance peaks is predominantly an effect of the fluid viscosity, which provides damping to the wire motion. The present authors have recently shown (Pádua, 1994; Pádua *et al*., 1995) that a practical instrument can be made to conform to a rigorous theoretical model which includes the hydrodynamic effects of the wire motion in the fluid (Retsina *et al*., 1986). The parameters that enter this model are

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clearly identifiable as real physical quantities characteristic of the measuring cell, such as the density, length and radius of the wire, and the volume and mass of the sinker.

The same kind of vibrating-wire sensor has been used for viscosity measurements in a different fashion and several viscometers have been built (Assael *et al.*, 1992b; Oliveira, 1991), both for liquid- and gas-phase measurements. In these viscometers the transient decay of the damped oscillations is measured with respect to time and related to the fluid viscosity. The theoretical model of the viscometer (Retsina *et al.*, 1987) is essentially the same as the model of the densimeter, but the buoyancy effect is irrelevant since the damping acting on the wire depends on the hydrodynamic contribution only. In the case of the densimeter, the buoyancy effect must be maximized relative to the hydrodynamic effect. In the present instrument, the fact that the wire is subject to the hydrodynamic effect provides a means to measure the fluid viscosity simultaneously with its density, but because density is the prime quantity of interest, the uncertainties in the viscosity will probably not be as small as could be attained with a purpose-built viscometer. Simultaneous measurements of both properties have been attempted in the past using a vibrating-disk viscometer (Krall *et al.*, 1987).

An independent determination of some of the cell parameters with the necessary accuracy poses particular difficulties. Two of them (radius of the wire and volume of the sinker) were therefore determined by measurement of the resonant characteristics of the wire in a fluid whose density and viscosity at a certain thermodynamic state are known independently. To guarantee the highest accuracy, this was done at 298 K and 0.1 MPa (Pádua *et al.*, 1995). Hence, using one datum for the density and one for the viscosity of a reference fluid, a set of cell parameters was obtained and then used to produce measurements in different fluids over wide ranges of temperature and pressure. The effects of temperature and pressure on the physical dimensions of the tungsten wire and aluminum sinker are accounted for using the thermal expansion and bulk modulus coefficients of these materials.

The vibrating-wire instrument, possessing a complete set of theoretically-based working equations (Retsina *et al.*, 1986; Pádua *et al.*, 1995), requires no calibration procedures as a function of temperature, pressure, or measurement range. The reference experiment provides the most accurate, practical means devised to obtain the values of two cell constants, which are real physical parameters, not empirical calibration constants. If the two parameters could be measured by independent, absolute methods with sufficient accuracy, that would render the present technique absolute. In our previous publication (Pádua *et al.*, 1995) an account of the tests performed on the apparatus and its theoretical model was given, where a careful comparison of results was made with literature data of the highest accuracy on three fluids. From this and other previous work (Pádua *et al.*, 1994), it was concluded that the measured densities have a precision of ±0.03% and an estimated accuracy of ±0.05%. The viscosity is obtained with a larger uncertainty—a precision of ±0.6% and an estimated accuracy of ±2.5%.

Experimental Section

The apparatus and the measurement technique are described elsewhere (Pádua *et al.*, 1994, 1995), and the equipment was used without significant changes for the present measurements. Temperature stability during the measurements is better than ±0.002 K, and the accuracy of the readings is ±0.01 K (ITS-90). The pressure readings are accurate to ±0.03 MPa up to 35.0 MPa, and ±0.07 MPa up to 138 MPa. Thermometer calibrations were done against a triple point of water cell and a 25 Ω platinum thermometer (Tinsley model 5187L) recently calibrated at the National Physical Laboratory, Teddington, U.K. Pressure calibrations were done against a dead-weight gauge (Ruska model 2450). The cell parameters used for the present measurements and their determination procedure were presented in detail by Pádua *et al.* (1995).

Table 1. Experimental Density and Viscosity of 1,1,1,2-Tetrafluoroethane

p/MPa	ρ/kg·m^{-3}	η/mPa·s	p/MPa	ρ/kg·m^{-3}	η/mPa·s
		$T = 199.11$ K			
0.80	1512.7		45.10	1572.7	
7.10	1522.7		60.10	1589.9	
15.10	1534.2		80.10	1612.7	
25.10	1548.0		94.60	1629.0	
35.10	1560.5				
		$T = 223.05$ K			
0.80	1447.5		35.10	1505.3	
5.10	1456.7		45.10	1519.3	
10.10	1464.8		60.10	1538.7	
15.10	1473.8		80.10	1561.8	
20.10	1482.9		100.10	1581.7	
		$T = 248.20$ K			
0.80	1374.4	0.401	27.10	1433.2	0.513
3.10	1380.6	0.411	35.10	1447.3	0.547
6.10	1388.2	0.424	45.10	1464.1	0.588
10.10	1397.8	0.442	60.10	1486.2	0.649
15.10	1409.1	0.462	80.10	1512.0	0.735
20.10	1419.7	0.483	100.10	1534.6	0.824
		$T = 273.15$ K			
0.73	1296.0	0.279	27.10	1372.4	0.375
3.08	1304.7	0.288	35.10	1389.9	0.404
6.10	1315.0	0.300	45.12	1409.5	0.438
10.10	1327.8	0.315	60.10	1435.1	0.490
15.12	1342.3	0.333	80.10	1464.8	0.562
20.10	1355.6	0.350	100.10	1490.4	0.636
		$T = 298.11$ K			
0.82	1207.6	0.205	24.97	1304.4	0.283
2.61	1217.1	0.212	32.10	1324.0	0.303
5.10	1230.0	0.221	45.10	1355.1	0.341
7.60	1241.9	0.229	60.10	1385.0	0.383
10.10	1252.7	0.238	80.10	1418.5	0.441
14.10	1268.4	0.250	100.10	1447.2	0.497
19.10	1286.0	0.265			

The round-robin sample of 1,1,1,2-tetrafluoroethane has been supplied by ICI with a stated purity of 99.9% (Assael *et al.*, 1995), which has been confirmed after the measurements had been carried out. The major impurity found was HFC-134, in a concentration below 0.1%. The water content was on the order of a few tens of ppm. The test fluid has been introduced in the measuring cell without further purification through a 65 μm filter.

Results and Discussion

The results for the density of 1,1,1,2-tetrafluoroethane along five isotherms from 199.11 K to 298.11 K are listed in Table 1, together with the viscosity measurements. For each temperature, the density, ρ, and viscosity, η, are shown as a function of pressure, p. About half of the values in each isotherm were obtained while increasing the pressure, and the remaining values, along decreasing pressures; no sign of hysteresis could be found. At the lowest temperature, signs of solidification appeared around 94.7 MPa. At the lower two isotherms we give only the density because we are not convinced we were able to maintain the same level of accuracy for the viscosity measurements that may be expected from the instrument and is characteristic of the remaining measurements. This

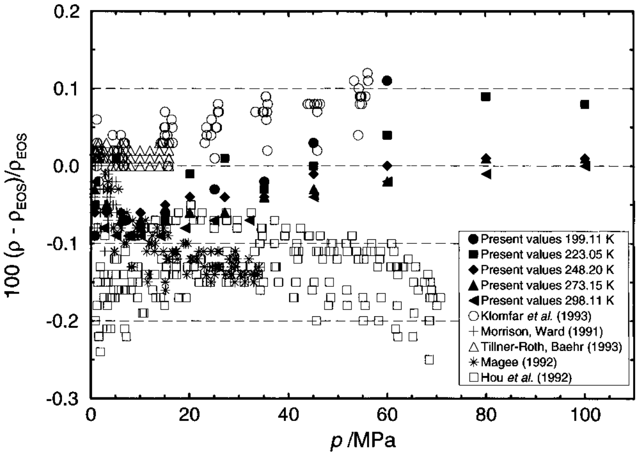

Figure 1. Deviations of the present density results for HFC-134a and of literature data from the EOS of Tillner-Roth and Baehr (1994).

disagreement at low temperatures will be the subject of further investigation.

In Figure 1 we present a deviation plot comparing our density results with the equation of state of Tillner-Roth and Baehr (1994), which is the standard recommended by Annex 18 of the International Energy Agency. The typical accuracy claimed for this EOS is ±0.05% in the density. Its pressure limit is 70 MPa, so we extrapolated it up to 100 MPa to compare with our values. All of our data except two points show deviations within ±0.10%, the agreement being generally very good even above 70 MPa. The points which have larger deviations belong to the isotherm at 199.11 K and correspond to the highest pressures of 80.10 MPa and 94.60 MPa. Their deviations are +0.3% and 0.5%, respectively, and they are not shown in the plot. We note that in the compressed liquid below room temperature only one set of direct density data (Tillner-Roth and Baehr, 1993) was considered in the formulation of the EOS. This set has a minimum temperature of 243 K and a maximum pressure of 16 MPa. At higher pressure and/or lower temperatures only heat-capacity and speed-of-sound data were considered (Tillner-Roth and Baehr, 1994).

Four sets of densities from the literature are compared below with the present measurements. Their deviations from the EOS of Tillner-Roth and Baehr (1994), in the temperature region where there is overlap with our results, are shown in Figure 1.

Of the data of Klomfar *et al.* (1993), 76 points overlap with our densities, between 205 K and 298 K and up to 56 MPa. These measurements were obtained using a constant-volume piezometer, with an uncertainty of ±0.15%. It is seen that the data of Klomfar *et al.* (1993) show positive deviations but agree with the EOS and with the present measurements within roughly +0.12%. They exhibit a slight but noticeable trend with pressure which is similar to the trend of our results. The data of Morrison and Ward (1991) were obtained with a vibrating-tube densimeter and comprise 31 points along four isotherms which fall within our temperature range. The upper pressure of this set is 6 MPa, and the claimed uncertainty is ±0.2%. This set shows negative deviations from the EOS, reaching −0.11%, and superimpose well with our measurements in this lower pressure range. The densities of Tillner-Roth and Baehr (1993) were also measured with a vibrating-tube instrument, and from this set 107 points were selected. They overlap with out results from 243 K to 293 K and extend up to 16 MPa. The reported accuracy for the densities is from within (0.14 to 0.4) $kg \cdot m^{-3}$, which corresponds to approximately ±0.05%. They coincide perfectly with the EOS and do not deviate from our values by more than +0.10%. Additional compressed liquid densities have been reported by Magee (1992), which were obtained during isochoric heat capacity measurements. These results cover the entire temperature range of the present work and extend up to 35 MPa. A total of 94 points were suitable for comparison. They show negative deviations from the EOS, with a maximum of −0.15% at the highest pressures. The agreement with the present results is within ±0.1%. Finally, Hou *et al* (1992) published densities of liquid HFC-134a up to 70 MPa measured with a continuously weighed pycnometer, whose claimed accuracy is ±0.1%. A total of 192 points along 6 isotherms from 200 K to 300 K were considered. These points show systematic negative deviations from the EOS that reach −0.25% at the lower and higher pressure limits, but no more than −0.15% in between. The maximum deviations from our densities are about −0.30%, but at pressures from 10 MPa to 40 MPa are of the order of −0.1% at most.

The overall comparison of our compressed liquid densities with the Tillner-Roth and Baehr (1994) EOS and with four of the five literature data sets does not contradict our previous assessment of an accuracy of ±0.05% for the vibrating-wire technique (Pádua *et al.*, 1995). Except for the two previously mentioned points, all the deviations agree with any of the sets of data and with the EOS within their mutual uncertainty. From an experimental point of view, nothing occurred during the measurements of the last two points at 199.11 K which could justify a loss of accuracy. It is true that, as the viscosity of the fluid increases, the larger damping to which the wire is subjected causes the resonance curves to broaden, and consequently, there is a loss of resolution in the detection of the maximum. This imposes an upper limit to the viscosity of the fluid which can be measured with a given vibrating-wire instrument without compromising the precision of the densities. From the measurements done so far using the present apparatus (Pádua, 1994; Pádua *et al.*, 1994, 1995), we evaluate this maximum viscosity to be about 6 mPa·s, whereas in the present study the highest viscosity attained was certainly below half of this limit.

With the vibrating-wire instrument measurements could not take place exactly at saturation, owing to the possibility of formation of bubbles caused by the motion of the wire, or even to the very slight heating resulting from the driving current and viscous dissipation. By design (Retsina *et al.*, 1986; Pádua, 1994), the temperature uncertainty arising from this heating is below 1 mK. In our previous publication (Pádua *et al.*, 1995) we presented orthobaric densities of HFC-134a that were calculated by performing a small extrapolation using the Tait equation fitted to each of the isotherms of Table 1. The vapor pressure for each temperature was taken from the equation of state for 1,1,1,2-tetrafluoroethane (Tillner-Roth and Baehr, 1994). A level of agreement comparable to the one found in the present work has been observed both with the EOS and with several literature data sets.

A viscosity correlation scheme for HFC-134a, encompassing single phase liquid and gas, a saturation line, and a critical region, has been published by Krauss *et al.* (1993). However, the application of this scheme is limited to temperatures above 290 K and densities below 1400 $kg \cdot m^{-3}$. Therefore, we chose to correlate the present values with an equation based on the hard-sphere theory of transport in dense fluids, which has been applied successfully to numerous alkanes (Assael *et al.*, 1992a) and to HFC-134a as well (Oliveira and Wakeham, 1993). A

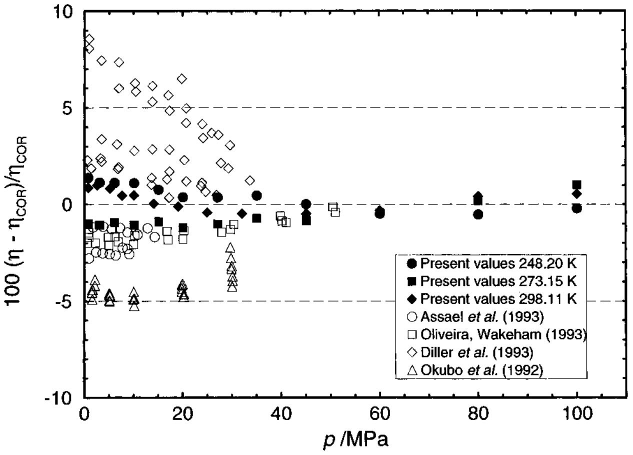

Figure 2. Deviations of the present viscosity results for HFC-134a and of literature data from the correlation using eqs 1 and 2.

Table 2. Coefficients of Eq 2

i	c_i
0	0.8780
1	1.8430
2	−0.1567

Table 3. Values for the Characteristic Volumes V_0

T/K	$10^6 V_0$/m^3·mol^{-1}
248.20	45.86
273.15	45.06
298.11	44.36

reduced, dimensionless viscosity, η^*, is defined by

$$\eta^* = 6.035 \times 10^8 (MRT)^{-1/2} V^{2/3} \eta \qquad (1)$$

where M is the molecular mass, R is the gas constant, T is the temperature, and V is the molar volume. The reduced viscosity should be a function of the ratio of the molar volume to a close-packed volume, V_0, only. This characteristic hard-sphere volume generally decreases linearly with increasing temperature, which can be interpreted as an account for the finite steepness of the real intermolecular potential.

We obtained a reference value for V_0 at 298.11 K by interpolation of the values reported by Oliveira and Wakeham (1993). Then, a characteristic volume for each of our isotherms was found by superimposing the isotherms on a single curve with the form

$$\ln \eta^* = \sum_{i=0}^{2} c_i [V_0/(V - V_0)]^i \qquad (2)$$

The coefficients c_i were determined by least-squares fitting to the entire set of points and are listed in Table 2. This correlation represents our measurements with typical deviations of ±1%, as can be seen from the deviation plot in Figure 2. The characteristic volumes are collected in Table 3. They exhibit an almost perfect linear dependence with temperature which guarantees a safe interpolation for the purpose of comparison with literature values at different temperatures.

For comparison, we chose from the literature four sets of viscosity measurements obtained using three different experimental techniques. Those of Assael *et al.* (1994) were measured using a vibrating-wire instrument in the transient mode with an uncertainty of ±0.5%. Two isotherms at 273.15 K and 293.15 K comprising 16 points, at pressures up to 9.1 MPa and 14.3 MPa, respectively, are suitable for comparison. These measurements show deviations from our correlation of about −2% with no visible trend, which is commensurate with the mutual uncertainties. Oliveira and Wakeham (1993) reported another set of measurements made using a vibrating-wire instrument in the transient mode, from which 22 points along two isotherms at 293.35 K and 303.14 K can be compared with our values. The estimated uncertainty of these measurements is ±0.6%, and they extend up to 51 MPa. Maximum deviations from the present correlation reach −3% at the lower pressures, but a total coincidence is observed at high pressures. Further comparisons were made with the results of Okubo *et al.* (1992), who used a capillary viscometer up to 30 MPa and claim an accuracy of ±1.3%. The 31 points along three isotherms at 253 K, 273 K, and 299 K show deviations from the present correlation in general between −5% at low pressures and −2% at 30 MPa. Again, this is roughly within the mutual uncertainties. Finally, we considered the torsional-crystal measurements of Diller *et al.* (1993) which extend to 30 MPa. The uncertainties associated with these data are of the order of 3%. The 44 points compared show positive deviations from our values, up to 2% at 250 K, up to 3.5% at 270 K, but up to 8% at low pressures at 300 K.

The overall agreement is good especially in view of the fact that four different techniques have been used to measure the viscosity of this polar fluid.

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