

Viscosity of Saturated Liquid for 1,1,1,2-Tetrafluoroethane

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In this paper, a capillary viscometer is described for measurement of the viscosity of 1,1,1,2-tetrafluoroethane (HFC-134a) under high pressure. The viscosity of saturated liquid HFC-134a has been measured over the temperature range of 233.15–333.15 K. The purity of the HFC-134a sample used in the experiment is 99.95 mass %. A saturated liquid viscosity correlation has also been proposed. The experimental data for various researchers are compared with the correlation.

Introduction

The replacement of chlorofluorocarbons (CFCs) has greatly accelerated since the establishment of the Montreal Protocol, especially the Copenhagen Congress in November 1992. 1,1,1,2-Tetrafluoroethane (HFC-134a) is the most promising alternative to CFC-12. Various thermophysical properties of HFC-134a have been measured. The saturated liquid viscosity data of HFC-134a from various researchers, however, are not consistent with each other. The temperature ranges listed in Table 1 are the ranges at which experiments were carried out by the individual researchers. In this paper a capillary viscometer is presented with which the viscosity can be measured under pressure. The viscosity of saturated liquid HFC-134a has been measured over the temperature range of 233.15–333.15 K, and the results are compared with the literature. We also present a saturated liquid viscosity correlation for HFC-134a.

Apparatus and Experimental Method

The Ostwald viscometer and its modification, the Ubblohe viscometer, which are both capillary viscometers, have a tendency for the glass to shatter when there is a high pressure difference between the inside and outside of the viscometer. In addition, they are inconvenient for filling and handling. For the reasons above, we designed a special capillary viscometer which can be operated at pressures up to 6 MPa and handled conveniently. With this viscometer, the viscosity of the saturated or subcooled liquid can be measured at various temperatures.

The new viscometer is shown in Figure 1. A modified Ostwald capillary viscometer was used as the body and fitted in a stainless steel pressure (6 MPa) vessel. Three pressure valves were fitted from outside of the vessel for filling, raising the liquid, and pressure connection.

Since the pressures both inside and outside of the capillary are always equal except for small pressure differentials, a glass capillary can be used. The high pressure of the sample liquid is exerted on the stainless steel vessel only. The sight glasses were made of fused silica of thickness 15 mm. Tetrafluoroethylene gaskets were used to prevent leakage at high pressure.

Prior to the experiment, the capillary was cleaned with nitric acid and rinsed with distilled deionized water. After assembly, the vessel was evacuated and then charged with HFC-134a through valve 3. The amount of HFC-134a charged was controlled by mass.

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Table 1. Comparison of Experimental Saturated Liquid Viscosity Data of HFC-134a^a

ref	author	physical state	T_{min}/K	T_{max}/K	points compared	method
1	Shankland	sl	251	343	7	cv
2	Kumagai	sl	273	343	6	cv
3	Oliveria	l, sl	238	343	49	vw
4	Diller	l, sl	175	320	27	tc
5	Ripple	sl	250	306	5	cv

^a Abbreviations: sl, saturated liquid; l, liquid; cv, capillary viscometer; vw, vibrating wire; tc, torsional crystal.

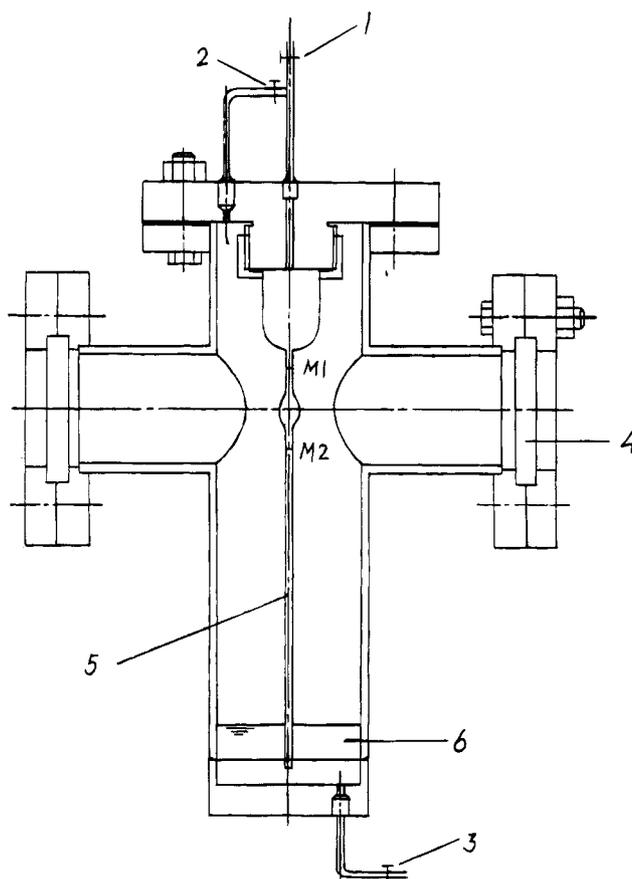


Figure 1. Viscosity experimental apparatus: (1, 2, 3) valves; (4) fused silica glass; (5) viscometer cell; (6) measured liquid.

The viscometer was mounted on a stainless steel seat and put in a bath with temperature controlled to ± 5 mK for all measurements. The bath temperatures were determined on ITS-90 with a certified platinum resistance

Table 2. Verification of the Computations for the Apparatus Constants with Water and Acetone

fluid	T/K	t/s	$\nu_{\text{calcd}}/(\text{mm}^2\text{s}^{-1})$	$\nu_{\text{sta}}/(\text{mm}^2\text{s}^{-1})$	$100(\nu_{\text{calcd}} - \nu_{\text{sta}})/\nu_{\text{sta}}$
water	313.15	358.20	0.6576	0.658	0.06
acetone	303.15	204.25	0.3736	0.370	0.97

thermometer. At least half an hour was required for temperature equilibration.

After temperature equilibrium had been established, with valve 2 turned off, valve 1 was turned on slightly, so that the capillary was connected with ambient air. The liquid sample was transferred into the two bulbs as a result of the high pressure of HFC-134a vapor in the vessel (the saturation pressure of HFC-134a is higher than ambient pressure at temperatures above -16.1°C ; at temperatures below -16.1°C , a suction from the outlet of valve 1 was needed). Valve 1 was then turned off and valve 2 turned on so as to connect the capillary with the vessel to equilibrate the pressure. The liquid flowed down through the capillary due to gravitation. The moments when the liquid surface passed through marks M1 and M2 were determined by visual observation. The efflux time was measured by a mechanical stopwatch graduated in 0.1 s and operated manually. From the efflux time, the viscosity of the sample liquid was calculated by the following equations:

$$\nu = AT - B/t \quad (1)$$

$$\eta = \nu/\rho \quad (2)$$

where ν is the kinematic viscosity of the fluid, t is the efflux time, η is the viscosity, ρ is the density of the fluid, and A and B are the apparatus constants.

Apparatus Calibration

The apparatus constants A and B were calibrated with water and acetone. In the calibrating experiment, the efflux times for water (25°C) and acetone (20°C) were 481.15 and 228.20 s, respectively. The viscosities for water (25°C) and acetone (20°C) used were 0.884 and 0.418 mm^2/s , respectively, to give the following apparatus constants:

$$A = 0.001839, B = 0.4017$$

So

$$\eta/(\text{mm}^2\text{s}^{-1}) = 0.001839(t/\text{s}) - 0.4017/(t/\text{s}) \quad (3)$$

Table 2 shows the viscosity of water and acetone at various temperatures calculated from eq 3. The calculated values were found to agree with the literature values within $\pm 1.0\%$.

Results and Analysis

The purity of the HFC-134a sample used in the experiment is 99.95 mass % with less than 10 ppm of water. The major impurity is HFC-134. The sample was provided by Shanghai Institute of Organic Chemistry.

The saturated liquid viscosities for HFC-134a over the temperature range of 233.15–333.15 K were measured. Twenty-one measuring points were chosen with equal temperature steps of 5 K.

Table 3 and Figure 2 give the results together with the relative deviations of the data from our correlation:

$$\ln(\eta/\text{mPa}\cdot\text{s}) = 8.559810 - 508.5909/(T/\text{K}) - 0.03899179(T/\text{K}) + 0.00003557719(T/\text{K})^2 \quad (4)$$

Table 3. Experimental Results of Saturated Liquid Viscosity for HFC-134a

T/K	$\rho/(\text{kgm}^{-3})$	$\eta/(\text{mPa}\cdot\text{s})$	$100(\eta - \eta_{\text{calcd}})/\eta_{\text{calcd}}$
233.15	1.416 049	0.4666	1.6
238.15	1.401 581	0.4236	-1.5
243.15	1.386 866	0.3912	-2.8
248.15	1.371 893	0.3804	0.8
253.15	1.356 631	0.3628	2.6
258.15	1.340 986	0.3312	0.0
263.15	1.325 153	0.3097	-0.2
268.15	1.309 106	0.2915	0.1
273.15	1.292 574	0.2726	-0.1
278.15	1.275 771	0.2562	0.0
283.15	1.258 606	0.2413	0.2
288.15	1.241 049	0.2274	0.5
293.15	1.223 062	0.2111	-0.7
298.15	1.204 573	0.1980	-1.0
303.15	1.185 579	0.1885	0.0
308.15	1.165 936	0.1778	0.0
313.15	1.145 685	0.1676	0.0
318.15	1.124 619	0.1595	0.6
323.15	1.102 609	0.1490	-0.5
328.15	1.079 517	0.1437	1.4
333.15	1.055 131	0.1329	-0.9

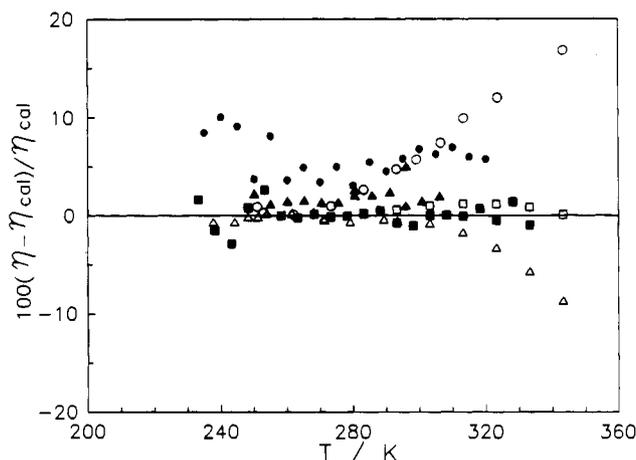


Figure 2. Relative deviation of the experimental data from eq 4: (■) this work; (△) ref 3; (□) ref 2; (○) ref 1; (▲) ref 5; (●) ref 4.

The relative deviations of the data reported in Table 1 from eq 4 are also shown in Figure 2. The following can be seen from Figure 2:

(1) At the lower and intermediate temperature range from 233.15 to 300.15 K, the relative deviations of the present data and those of Kumagai et al. (2) and Oliveria et al. (3) from eq 4 are less than 1%. The deviations of the data reported by Ripple et al. (5) depart from eq 4 by no more than 3%.

(2) At the higher temperature range from 300.15 to 330.15 K, the relative deviations of the present experimental data and Kumagai's data (2) from eq 4 do not exceed 1.3%. The results of Oliveria et al. (3) show the larger negative deviations, but those of Shankland et al. (1) show large positive deviations. At higher temperatures, the deviations are larger, and the maximum deviations are -8% and $+16.8\%$, respectively.

(3) The relative deviations of data reported by Diller et al. (4) from eq 4 are about 3–5%. The lower the temperature, the larger the deviation; the maximum deviation is 10%.

Conclusion

The viscosity of saturated liquid for HFC-134a has been measured in the temperature range from 233.15 to 333.15

K. We also present a saturated liquid viscosity correlation. The comparison of the experimental results with this correlation has also been discussed.

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