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Measurement of the Viscosity of HFC 134a in the Temperature Range 213–423 K and at Pressures up to 30 MPa

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The viscosity of HFC 134a was measured over the range of temperatures from 213 to 423 K and pressures up to 30 MPa. The experimental method was that of the capillary flow and a closed-circuit high-pressure viscometer was used. The sample fluid was circulated through a stainless-steel capillary from a high-pressure plunger system. The constant of the capillary was calibrated against the reference standard, pure water. The viscosity of the sample was calculated from the flow rate, the pressure drop at the capillary, and the capillary constant using the Hagen–Poiseuille equation. Measurements were made at a total of 39 points on eight isotherms. The measurement uncertainty of the viscosities was estimated as ±1.3%. Based on the present results, an empirical equation for the viscosity of HFC 134a has been correlated. The viscosity on the saturation line calculated by the equation compares with experimental viscosity data in other previous studies. There are rather considerable differences among these measurements. Comparisons of the data for HFC 134a with those for CFC 12 show that the viscosity of HFC 134a is similar in magnitude to that of CFC 12 at temperatures around 300 K but is higher at lower temperatures and lower at higher temperatures. The pressure gradients for these two corresponding substances are similar over the entire temperature range.

KEY WORDS: capillary viscometer; HFC 134a; 1, 1, 1, 2-tetrafluoroethane; viscosity.

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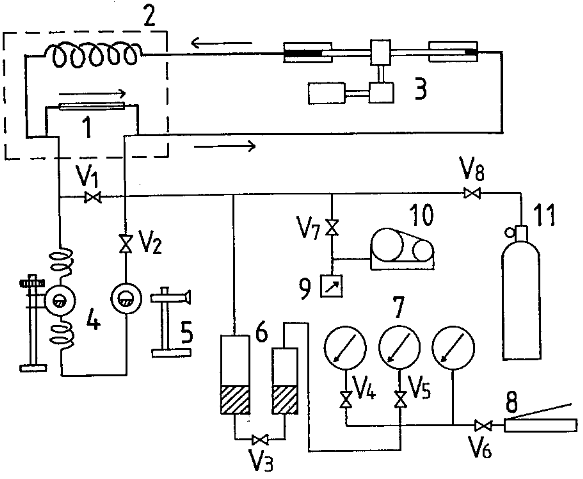
1. INTRODUCTION

Chlorofluorocarbons (CFCs) are widely used as refrigerator working fluids, blowing agents in polymer forms, and heat-pump working fluids. However, as CFCs are speculated to deplete the earth's stratospheric ozone layer, protocol calls for the regulation of their consumption. Some CFC alternatives whose general properties are similar to those of CFCs have been proposed. However, the thermophysical properties that are actually needed have not been determined yet; measurements of viscosity are especially scarce. In the present study, the viscosity of HFC 134a (1, 1, 1, 2-tetrafluoroethane; CH_2FCF_3), which is the alterative to CFC 12, was measured over the temperature range 213–423 K and at pressures up to 30 MPa in the compressed liquid and the supercritical states.

2. EXPERIMENTAL

The experimental method was that of the capillary method and a closed-circuit capillary viscometer was used. The experimental system as shown in Fig. 1 was similar to those used for other fluids under high pressure in our laboratory in the past. Details of the technique and the apparatus are given in our previous publications [1, 2]. The viscosity was calculated from a modified Hagen–Poiseuille equation,

$$\eta = \frac{\pi C \Delta P(1+\alpha \Delta t)^3}{8Ql} - \frac{m\rho Q}{8\pi l(1+\alpha \Delta t)} \tag{1}$$



1. Test section (capillary)
2. Thermostat
3. Circulating injector
4. Mercury manometer
5. Travelling microscope
6. Separator
7. Bourdon gage
8. Hand pump
9. Vacuum gage
10. Vacuum pump
11. Container of specimen fluid

V_1–V_8 Valves

Fig. 1. Schematic diagram of the experimental system.

where η (Pa · s) is the viscosity, ΔP (Pa) is the pressure drop between both ends of the capillary, C (m^4) is the capillary constant, Q (m$^3 \cdot$ s^{-1}) is the volumetric flow rate, l (m) is the length of the capillary, α (K^{-1}) is the thermal expansion coefficient of the capillary, ρ (kg · m^{-3}) is the density of the sample fluid, and m is the kinetic-energy correction factor ($m = 1.12$).

The experimental apparatus is a closed-circuit high-pressure capillary viscometer, as shown in Fig. 1. Generally, the capillary for a viscometer is made of glass, but this is inapplicable to high-pressure viscometry, because of difficulty in connecting it with other parts of the apparatus made of steel. Therefore, a capillary made of stainless steel welded to the apparatus was used in the present measurement. Its length was 283 mm; outer diameter, about 2 mm; and inner diameter, about 0.3 mm.

The capillary constant, C, was determined by calibrating the capillary against the viscosity of water. And it was certain that the value of C determined against water viscosity was almost equal to that against nitrogen gas. It is considered that the critical Reynolds number (Reĉ) for a metallic

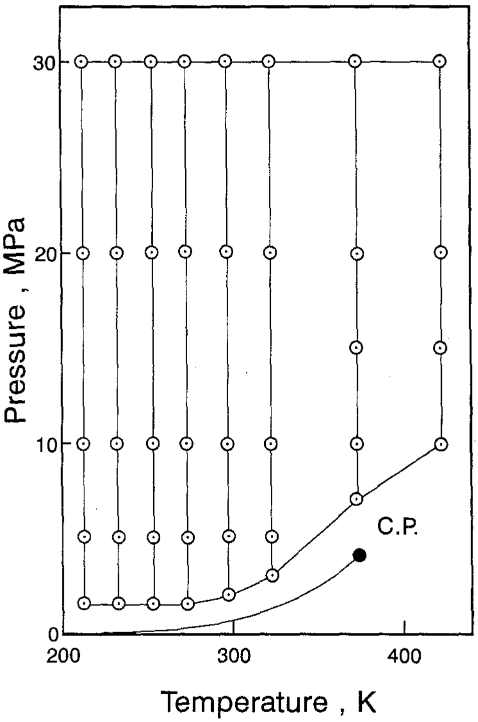


Fig. 2. Range and grid points of the present measurements.

capillary is lower than the generally accepted Reĉ because of roughness on the inside wall of a stainless-steel capillary. Therefore, the Reĉ of the present capillary was determined when calibrating *C*, and if the Reynolds number in a measurement was higher than Reĉ, the value of the viscosity was corrected according to the calibrated curve of *C*. The maximum amount of the correction when applied was about 3%.

The fluid was circulated from one end of a twin-headed injector (Fig. 1), through the capillary, to the other end of the injector. The pressure drop between the two ends of the capillary was measured with a high-pressure mercury manometer with a traveling microscope. The volumetric flow rate, *Q*, was determined by counting the movement of the plunger of the circulation injector. The pressure of the sample was measured with a Bourdon gauge and temperature at the capillary was measured with a Pt thermoresistor which was approved according to IPTS-68.

The sample fluid, HFC 134a, was manufactured by Showa Deako Co., The purity is more than 99.9 wt%. Impurities are residua, after evaporation, of less than 100 wt ppm, acids of less than 1 wt ppm, and H_2O of 20 ppm. The sample fluid was introduced into the viscometer circuit after evacuating the system.

In this study, the density data of the sample are needed for calculation of the viscosity. The density values were mostly calculated from the equation of state for HFC 134a of Piao et al. [3]. Since the equation is not effective at temperatures lower than 240 K, the density at lower temperatures was calculated by the Tait equation, which was correlated with the experimental density data [4] in the compressed liquid region. However, accurate density data were not available at pressures higher than 15 MPa in the supercritical states, and density in that range was calculated.

The measurements of the viscosity took place at 39 grid points in the temperature range 23–423 K and at pressures up to 30 MPa. The grid points where the viscosity of HFC 134a was measured in the experiments are shown in Fig. 2. The viscosity measurements were repeated twice or three times at each grid point in order to confirm the reproducibility of the results.

3. RESULTS AND DISCUSSION

The measured values of the viscosity of HFC 134a as well as the density and the kinematic viscosity are given in Table I. Error in the measurement of the pressure drop was ±0.2% and that in the flow rate was ±1.1% (including the error of the equation of state). Error in the capillary constant *C* was estimated as ±0.4%. Thus, the estimated accuracy of the

Table I. Experimental Results of the Viscosity of HFC 134a

Temperature T (K)	Pressure P (MPa)	Density ρ (kg·m^{-s})	Viscosity η (10^{-4} Pa·s)	Kinematic viscosity ν (10^{-7} m^2·s^{-1})
		Nominal temperature, 213 K		
213.11	1.73	1473.4	6.734	4.570
213.12	1.74	1473.4	6.727	4.566
213.08	4.75	1479.1	6.953	4.701
213.17	4.77	1478.9	6.944	4.695
213.11	10.07	1488.4	7.323	4.920
213.10	10.12	1488.5	7.325	4.921
213.20	19.65	1503.8	8.066	5.364
213.23	19.82	1504.0	8.041	5.347
213.29	29.62	1518.3	8.957	5.899
213.29	29.72	1518.4	8.934	5.884
213.27	29.81	1518.6	8.978	5.912
		Nominal temperature, 233 K		
233.01	1.76	1418.9	4.755	3.351
233.05	1.87	1419.1	4.752	3.348
233.00	5.03	1426.4	4.923	3.451
233.00	5.11	1426.6	4.944	3.466
233.02	9.93	1436.7	5.212	3.628
233.01	9.95	1436.8	5.205	3.622
232.99	19.81	1455.8	5.723	3.931
232.99	19.91	1456.0	5.728	3.934
233.01	29.60	1472.5	6.258	4.250
233.01	29.82	1472.9	6.251	4.244
		Nominal temperature, 253 K		
252.98	1.58	1360.9	3.546	2.606
252.94	1.59	1361.0	3.547	2.606
252.93	5.18	1371.5	3.699	2.697
252.94	5.20	1371.5	3.690	2.690
252.96	9.95	1383.9	3.888	2.809
253.00	10.01	1384.0	3.905	2.821
252.97	19.84	1406.5	4.316	3.068
252.97	20.01	1406.9	4.325	3.074
253.00	29.93	1426.1	4.742	3.325
252.91	30.19	1426.8	4.727	3.313

Table I. *(Continued)*

Temperature T (K)	Pressure P (MPa)	Density ρ (kg · m^{-s})	Viscosity η (10^{-4} Pa · s)	Kinematic viscosity ν (10^{-7} m^2 · s^{-1})
		Nominal temperature, 273 K		
273.08	1.65	1299.1	2.713	2.089
273.12	1.69	1299.2	2.729	2.100
273.13	5.07	1311.6	2.842	2.167
273.13	5.11	1311.7	2.842	2.167
273.04	10.05	1328.1	3.040	2.289
273.06	10.06	1328.0	3.028	2.280
273.04	20.26	1356.2	3.383	2.494
273.05	20.29	1356.3	3.388	2.498
273.00	30.01	1378.7	3.764	2.730
272.95	30.11	1379.1	3.729	2.704
272.99	30.14	1379.0	3.748	2.718
		Nominal temperature, 299 K		
298.54	2.17	1213.4	1.993	1.643
298.50	2.18	1213.6	1.988	1.638
298.53	5.07	1228.8	2.082	1.694
298.52	5.07	1228.9	2.081	1.693
298.53	10.15	1251.9	2.235	1.785
298.49	10.16	1252.1	2.245	1.793
298.54	19.82	1287.1	2.545	1.977
298.50	19.86	1287.3	2.551	1.982
298.57	29.70	1315.8	2.888	2.195
298.55	29.84	1316.2	2.876	2.185
		Nominal temperature, 324 K		
323.72	3.35	1118.9	1.484	1.326
324.04	3.35	1117.5	1.484	1.328
324.03	3.38	1117.8	1.488	1.331
324.00	5.46	1135.4	1.562	1.376
323.96	5.65	1137.0	1.571	1.382
324.00	10.32	1168.2	1.724	1.476
324.04	10.47	1168.9	1.724	1.475
323.98	19.87	1215.5	1.999	1.645
323.96	19.97	1215.9	2.009	1.652
324.00	29.64	1251.8	2.266	1.811
323.99	29.93	1252.8	2.260	1.804

Table I. *(Continued)*

Temperature T (K)	Pressure P (MPa)	Density ρ (kg · m^{-s})	Viscosity η (10^{-4} Pa · s)	Kinematic viscosity ν (10^{-7} m^2 · s^{-1})
		Nominal temperature, 373 K		
372.94	7.03	903.6	0.8483	0.9388
372.92	7.06	904.5	0.8532	0.9433
373.00	10.11	964.2	0.9997	1.037
372.99	10.17	965.3	0.9960	1.032
373.03	15.22	1025.2	1.162	1.133
373.03	15.27	1025.8	1.165	1.135
373.04	20.28	1066.9	1.301	1.219
373.04	20.33	1067.4	1.311	1.228
373.01	29.60	1123.0	1.511	1.345
373.01	29.89	1124.4	1.517	1.349
		Nominal temperature, 423 K		
423.10	10.10	645.4	0.4770	0.7391
423.05	10.11	646.3	0.4762	0.7368
423.15	15.07	813.6	0.7037	0.8649
423.17	15.07	813.5	0.7022	0.8632
423.24	19.95	893.4	0.8478	0.9489
423.24	19.99	893.9	0.8508	0.9517
423.29	29.69	987.9	1.080	1.093
423.29	29.81	988.9	1.081	1.093

measurement at the 95% confidence level is $\pm 1.3\%$. The pressure dependence of the viscosity of HFC 134a is shown in Fig. 3. At lower temperatures, the temperature dependence is more significant, and the pressure dependence is similar to that for the viscosity of other normal liquids.

Equation (2) for the viscosity of liquid HFC 134a was correlated from the values in Table I,

$$\begin{gathered} \eta = A + BP_r + C\ln(1 + P_r) \\ A = \sum_{i=0}^{3} a_i \exp\frac{i-1}{T_r}, \qquad B = \sum_{i=0}^{3} b_i \exp\frac{i-1}{T_r}, \qquad C = \sum_{i=0}^{3} c_i T_r^i \\ T_r = T/T_C, \qquad P_r = P/P_C \end{gathered} \tag{2}$$

where the values of a_i, b_i, and c_i are given in Table II, η (10^{-4} Pa · s) is the viscosity of HFC 134a, T_C is the critical temperature (374.3 K), and P_C is the critical pressure (4.0640 MPa). Figure 4 shows the deviation of the

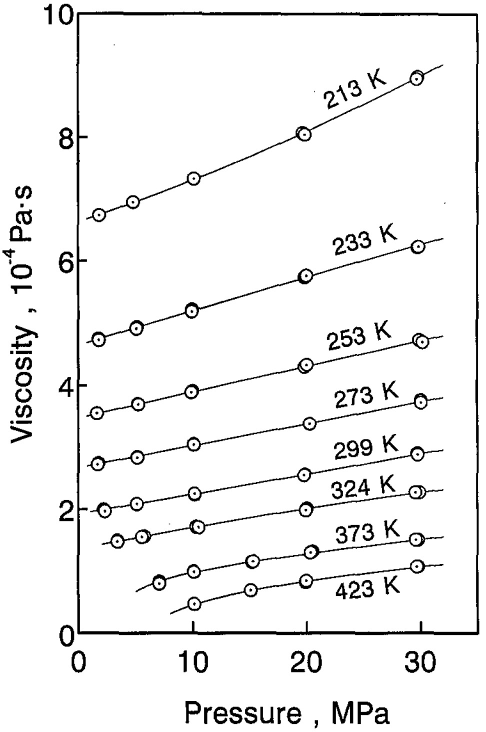


Fig. 3. Pressure dependence of the viscosity of HFC 134a.

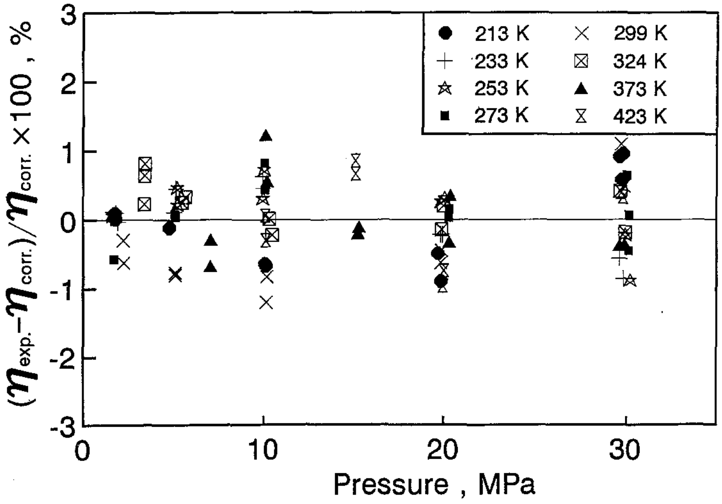


Fig. 4. Deviations of the measured viscosity data for HFC 134a from Eq. (2).

Table II. Coefficients of Eq. (2)

$a_0 = -28.3390$	$b_0 = -2.03324$	$c_0 = 3.48185$
$a_1 = 18.1691$	$b_1 = 1.29841$	$c_1 = -12.4685$
$a_2 = -4.13120$	$b_2 = -0.269815$	$c_2 = 13.2131$
$a_3 = 0.514252$	$b_3 = 0.0280058$	$c_3 = -3.74703$

experimental data from Eq. (2). The standard deviation is 0.54% and the maximum deviation is 1.22%. It is certain that the saturated liquid viscosity calculated by Eq. (2) agrees with the production of the experimental viscosities on each isotherms. The viscosities in the present work were compared with the experimental viscosities in other studies. The deviations of the data of Ruvinskii [5] in the compressed liquid region from Eq. (2) are shown in Fig. 5. The data of Ruvinskii agree with the equation except a few data around the critical point. Small disagreement around the critical point might show that Eq. (2) might not be applicable in the region close to the critical point. The viscosity on the saturation line calculated by Eq. (2) compares with experimental viscosity data of Shankland et al. [6] and those of Kumagai and Takahashi [7]. Their data and values by Eq. (2) are shown in Fig. 6. There are significant differences among these three measurements. The values of Kumagai and Takahashi are about 4%

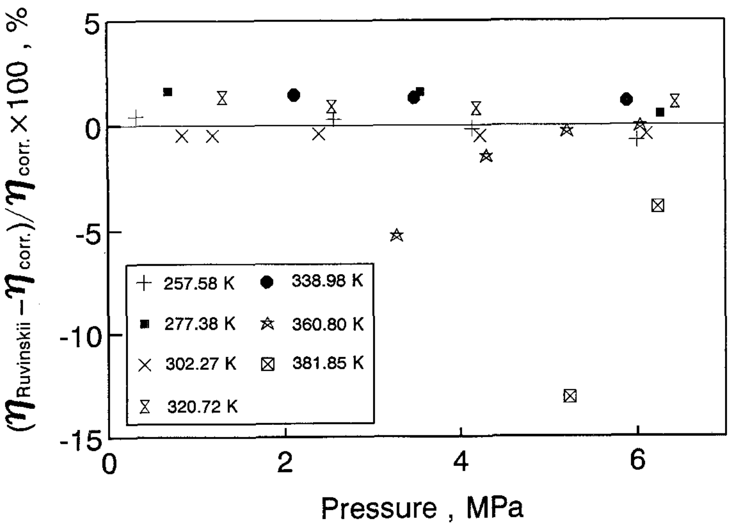


Fig. 5. Deviations of the viscosity data for HFC 134a of Ruvinskii [5] from Eq. (2).

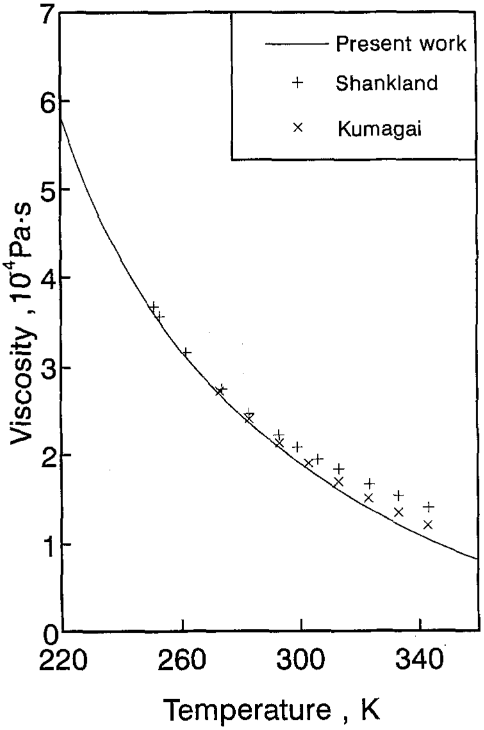


Fig. 6. Comparison of the viscosity data for HFC 134a of various authors on the saturation line (liquid).

larger and those of Shankland et al. are over 8% larger than those of the present study in the temperature range up to 300 K, where Eq. (2) is considered to be applicable on the saturation line. As the temperature increases, the difference increases further.

HFC 134a is proposed as an alternative to CFC 12. The experimental value [actually, values from Eq. (2)] of the present study are compared with the viscosities of CFC 12 [8] as shown in Fig. 7. The pressure dependence on the viscosities of HFC 134a agrees with that of CFC 12. The viscosities of CFC 12 and HFC 134a agree at around 300 K, but at temperatures below 300 K the viscosity of HFC 134a is higher than that of CFC 12. The opposite situation occurs when the temperature is higher than 300 K.

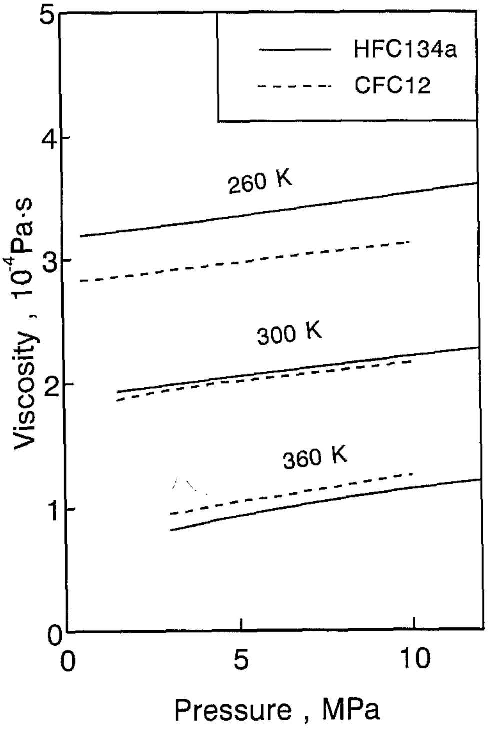


Fig. 7. Comparison of the viscosity of HFC 134a with that of CFC 12.

4. CONCLUSIONS

In the present study, the viscosity of HFC 134a in the compressed liquid and the supercritical states was determined with a closed-circuit capillary viscometer in the temperature range 213–423 K and at pressures up to 30 MPa. The results show the behavior of the viscosity of liquid HFC134a over a wide range of temperature and pressure. In the lower pressure range, reasonably good agreement was obtained with previous data of Ruvinskii. Comparison with the viscosity of CFC 12 is also described.

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