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Viscosity of Gaseous HFC-134a (1,1,1,2-Tetrafluoroethane) Under High Pressures

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The viscosity of gaseous HFC-134a (1,1,1,2-tetrafluoroethane) was measured with an oscillating disk viscometer of the Maxwell type from 298.15 to 398.15 K at pressures up to 5.5 MPa. Intermolecular potential parameters for the Lennard–Jones 12-6 model were determined from the viscosity data at 0.1 MPa. The viscosity equation developed by Krauss et al. was applied to correlate the present viscosity data. In addition, the correlations proposed by Stiel and Thodos and by Lee and Thodos were tested for fitting the experimental viscosity data.

KEY WORDS: HFC-134a; Lennard–Jones potential; viscosity.

1. INTRODUCTION

There has recently been great interest in the thermophysical properties of HFC-134a (1,1,1,2-tetrafluoroethane), since it is considered to be an environmentally acceptable alternative to CFC-12. Furthermore, HFC-134a is one of the constituent components for ternary mixtures of HFC-134a + HFC-125 + HFC-32.

To establish a prediction method for the viscosity of the ternary mixture in the gas phase, we started to measure the viscosity of the three pure components and of the three binary gas mixtures of HFC-134a, HFC-125, and HFC-32. In 1990, we presented our first data for HFC-134a at a conference held in Sendai [1]. Subsequently, some researchers measured the viscosity of gaseous and liquid HFC-134a and also developed a viscosity equation from a theoretical point of view. Recently, Vogel pointed out to us that our previous data in Ref. 1 are about 5% higher than

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those of other researchers including the data obtained by his group; this discrepancy probably arises from impurities contained in the samples as suggested by the temperature dependence of the deviations between our previous data and the literature values [2]. Since the sample used in our previous study had already been discharged, we could not check the purity of the sample used in our previous study. Therefore, in this study we tried to measure the gas viscosity of HFC-134a again, with a new sample. The measurements were made at temperatures from 298.15 to 423.15 K and at pressures up to 5.5 MPa. The experimental results were correlated with the theoretical viscosity equation developed by Krauss et al. [10], and then compared with the correlations of Stiel and Thodos [3] and of Lee and Thodos [4].

The gas viscosity of HFC-134a at normal pressure has been measured by Nabizadeh and Mayinger [6], Dowdell and Matthews [7], and Wilhelm and Vogel [8]. Ruvinskii et al. [5] and Pasekov and Ustyushanin [9] measured the gas viscosity under high pressures. Krauss et al. [10] developed a generalized viscosity equation based on the experimental data published previously in the open literature before 1993.

2. EXPERIMENTS

The viscosity was measured with an oscillating disk viscometer of the Maxwell type. The gas density was measured with a high-pressure gas pipette. The experimental apparatus and procedure were essentially the same as those described in previous studies [11–13]. The apparatus constant at the experimental temperature and pressure conditions was determined by considering the viscosity data of nitrogen taken from Stephan et al. [14] and the nitrogen gas density data from Jacobsen and Stewart [15]. Temperature and pressure values have an uncertainty of ± 0.01 K and ± 0.5 kPa. Density values have an uncertainty of $\pm 0.03\ \mathrm{kg \cdot m^{-3}}$. The error in the measurements of viscosity was estimated to be $\pm 0.3\%$. The sample was supplied by Asahi Glass Co. Ltd. The purity of the sample, certified by the suppliers, was approximately 99.9%. The sample used was purified by distillation several times.

3. RESULTS

The experimental results for viscosity and density are presented in Table I and displayed in Figs. 1 and 2. As seen in Fig. 1, the curves as a function of pressure intersect around a temperature of 373.15 K, but the curves as a function of density do not, as seen in Fig. 2. This behavior of the viscosity is a well known characteristic of nonpolar and polar gases.

Table I. Viscosity of HFC-134a

P (MPa)	ρ $(\mathrm{kg \cdot m^{-3}})$	η $(\mu\mathrm{Pa \cdot s})$
	$T = 298.15$ K	
0.1001	4.230	11.815
0.2033	8.714	11.769
0.3064	13.458	11.748
0.4074	18.367	11.711
0.5182	24.044	11.689
0.6107	29.881	11.666
	$T = 323.15$ K	
0.1011	4.137	12.768
0.2029	7.921	12.738
0.2537	9.899	12.751
0.3090	12.125	12.745
0.3450	13.613	12.744
0.4277	17.146	12.714
0.5491	22.622	12.744
0.5521	22.759	12.724
0.6795	28.891	12.736
0.7268	31.263	12.751
0.8194	36.066	12.752
0.9246	41.766	12.763
1.0230	47.331	12.802
1.2111	58.607	12.852
	$T = 348.15$ K	
0.1014	3.607	13.771
0.1037	3.756	13.757
0.2069	7.426	13.740
0.2996	10.883	13.749
0.3021	10.977	13.714
0.4016	14.810	13.735
0.4875	18.208	13.726
0.5005	18.733	13.729
0.5981	22.708	13.729
0.6911	26.593	13.780
0.6995	26.592	13.766
0.8376	32.913	13.818
0.9925	39.886	13.837
1.1878	49.197	13.878
1.3426	57.100	13.944
1.4988	65.669	14.030
1.6506	74.708	14.156
1.8291	86.465	14.291
1.9730	97.016	14.459
2.1309	109.95	14.647
2.2361	119.47	14.838

Table I. *(Continued)*

P (MPa)	ρ $(\mathrm{kg \cdot m^{-3}})$	η $(\mu\mathrm{Pa \cdot s})$
	$T = 373.15$ K	
0.1013	3.317	14.735
0.1021	3.387	14.748
0.2223	7.627	14.726
0.3441	11.888	14.743
0.3913	14.684	14.762
0.4649	17.182	14.745
0.5924	21.338	14.767
0.6899	24.498	14.766
0.7347	25.969	14.797
0.8751	30.748	14.819
0.9287	32.664	14.842
1.0057	35.516	14.866
1.1806	42.497	14.912
1.4236	53.388	15.012
1.6742	65.897	15.150
1.9260	79.436	15.347
2.1604	91.288	15.509
2.3992	105.23	15.751
2.6156	119.81	16.065
2.8425	136.83	16.414
3.0051	150.75	16.733
3.1914	168.36	17.202
3.3609	187.57	17.744
3.4745	202.61	18.204
3.5849	219.60	18.716
3.6850	238.36	19.396
3.7457	252.68	19.938
3.7974	264.90	20.410
3.8509	283.66	21.167
3.8802	293.86	21.633
3.9057	310.41	22.413
3.9284	324.12	23.084
	$T = 398.15$ K	
0.1017	3.136	15.685
0.1023	3.182	15.708
0.2212	6.994	15.661
0.3550	11.329	15.680
0.3978	12.729	15.729
0.4756	15.297	15.711
0.6098	19.784	15.740
0.6923	22.587	15.756
0.7548	24.734	15.760
0.8896	29.435	15.801

Table I. *(Continued)*

P (MPa)	ρ ($kg \cdot m^{-3}$)	η ($\mu Pa \cdot s$)
0.9872	32.919	15.851
1.0212	34.129	15.847
1.2867	43.933	15.938
1.5843	55.505	16.057
1.8812	67.728	16.234
2.1520	79.551	16.401
2.3792	90.027	16.590
2.6394	102.74	16.830
2.8571	114.07	17.064
3.1544	130.75	17.422
3.3953	145.50	17.783
3.6688	163.87	18.264
3.9110	181.90	18.810
4.1809	204.36	19.485
4.4571	230.48	20.346
4.6870	255.14	21.116
4.9117	282.27	22.380
5.1739	318.38	24.113
5.3574	346.94	25.564
5.5060	372.32	27.118
5.6336	395.88	28.693
	$T = 423.15$ K	
0.1012	2.952	16.600
0.2329	6.837	16.604
0.3592	10.623	16.640
0.5065	15.104	16.670
0.6525	19.620	16.691
0.8011	24.294	16.722
0.9477	28.988	16.740
1.0926	33.707	16.804
1.2412	38.638	16.855
1.4017	44.105	17.026
1.7122	54.965	17.153
2.0545	67.361	17.290
2.5294	85.901	17.566
2.9683	104.95	17.954
3.3723	122.69	18.338
3.5408	131.05	18.546
3.9296	150.88	19.106
4.3629	174.96	19.673
4.6852	194.39	20.290
5.1011	221.58	21.170
5.3497	239.09	21.691
5.5125	251.09	22.300

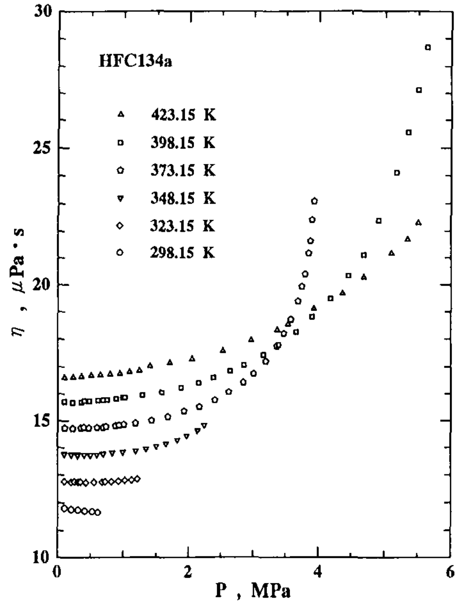

Fig. 1. Viscosity of HFC-134a as a function of pressure.

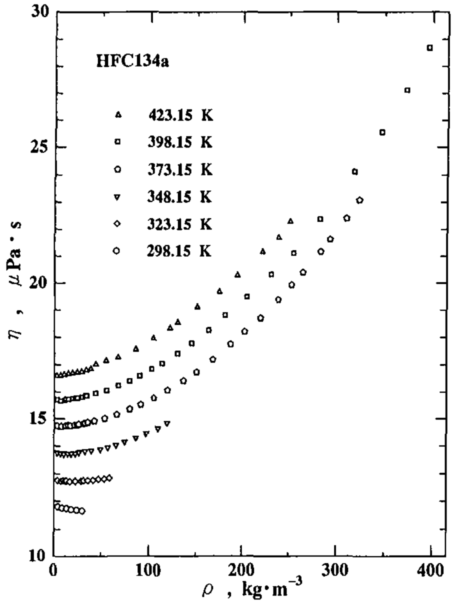

Fig. 2. Viscosity of HFC-134a as a function of density.

From Fig. 2, it can be seen that a negative initial density slope was observed for the viscosity isotherm at 273.15 K. This behavior has also been observed in our previous studies of polar gases [16–21].

For the gas viscosity at 0.1 MPa, the present results agree quite well with the viscosity data of Wilhelm and Vogel [8]. The data of Nabizadeh and Mayinger [6] are about 0.5% higher than the present results, and the data of Paskov and Ustyushanin [9] and Ruvinskii et al. [5] are about 0.5–2.0% lower. While the present data agree with those of Dowdell and Matthews [7] within ±0.5%, the data of Dowdell and Matthews show a weaker temperature dependency than the present results and those of Wilhelm and Vogel [8]. The viscosity data at 0.1 MPa of Paskov and Ustyushanin [9] were obtained by extrapolation based on using the initial density dependence data of the viscosity as measured by Wilhelm and Vogel [8]. The viscosity data of Ruvinskii et al. [5] were determined by extrapolating their data at high pressures to 0.1 MPa.

Krauss et al. [10] have expressed the viscosity at atmospheric pressure, $\eta_0/(\mu\mathrm{Pa\cdot s})$, using the following equation:

$$\eta_0 = \frac{0.2696566 T^{0.5}}{\sigma^2 \Omega(T^*)} \tag{1}$$

where T is the temperature in K, σ is a length scaling factor in nm, and Ω is a non-dimensional collision integral. The dimensionless temperature T^* is defined as:

$$T^* = kT/\varepsilon \tag{2}$$

where k is Boltzmann's constant in $\mathrm{J\cdot K^{-1}}$ and ε/k is the energy scaling factor. For the collision integral Ω, a formula presented by Bich et al. [22], shown by Eqs. (3) and (4), is used.

$$\ln \Omega(T^*) = \sum_{i=0}^{4} a_i (\ln T^*)^i \tag{3}$$

with coefficient a_i:

$$\begin{gathered} a_0 = 0.4425728, \qquad a_1 = -0.5138403, \qquad a_2 = 0.1547566 \\ a_3 = -0.02821844, \qquad a_4 = 0.001578286 \end{gathered} \tag{4}$$

Krauss et al. [10] determined the values of the energy and length scaling factors, ε/k and σ, from the data of Takahashi et al. [1], Nabizadeh and Mayinger [6], and Dowdell and Matthews [7]. Since the data of

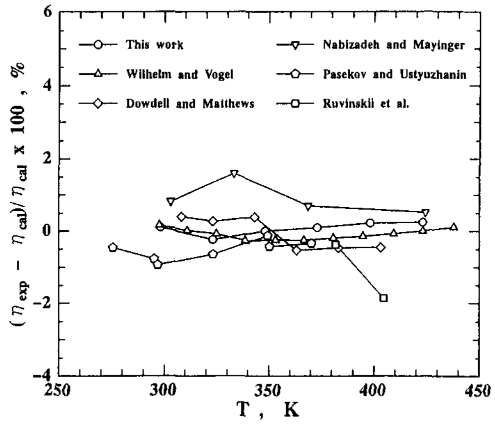

Fig. 3. Deviations of experimental viscosity values of HFC-134a at 0.1 MPa from those calculated with Eq. (1).

Takahashi et al. [1] have been found to be erroneous, we redetermined the values of these two factors with the use of the present results and the data of Wilhelm and Vogel [8]. The following values were obtained from a least-squares fit; $\varepsilon/k = 301.06$ K and $\sigma = 0.5016$ nm. Figure 3 shows the deviation of our data from Eq. (1). It can be seen that fairly good agreement is obtained with Eq. (1).

The present values of the viscosity at high pressures were compared with those calculated form the viscosity equation proposed by Krauss et al. [10]. Based on the excess viscosity concept, Krauss et al. [10] developed the following viscosity equation for HFC-134a applicable for gas and liquid states except those very close to the critical region:

$$\eta(\rho, T) = \eta_0(T) + \Delta_R\eta(\rho) \tag{5}$$

where the first term of the r.h.s. is the dilute gas viscosity given by Eq. (1), and the second term is the excess term. The excess term is represented by

$$\Delta_R\eta/H_c = \sum_{i=1}^{3} e_i(\rho/\rho_c)^i + \frac{e_4}{(\rho/\rho_c) - e_5} + \frac{e_4}{e_5} \tag{6}$$

where $\rho_c = 515.25\ \mathrm{kg \cdot m^{-3}}$ is the critical density and $H_c = 25.21\ \mu\mathrm{Pa \cdot s}$ is a so-called pseudo-critical viscosity. The coefficients in Eq. (6) are:

$$e_1 = -1.89758, \quad e_2 = 0.256449, \quad e_3 = -0.301641$$

$$e_4 = -23.1648, \quad e_5 = 3.44752$$

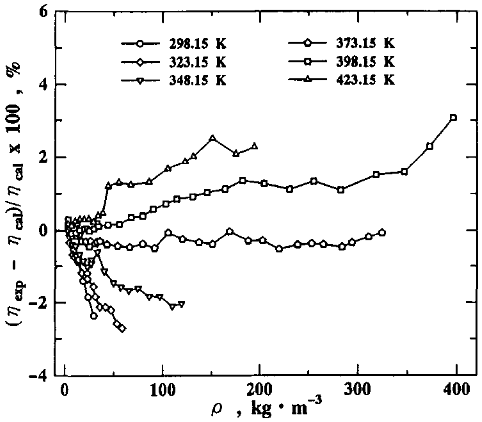

Fig. 4. Deviations of experimental viscosity values of HFC-134a from those calculated with Eq. (5).

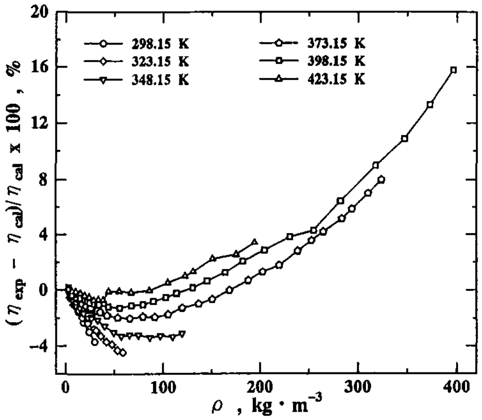

Fig. 5. Deviations of experimental viscosity values of HFC-134a under high pressures from those calculated with Stiel–Thodos correlation.

Table II. Physical Properties of HFC-134a

Molar weight mass	102.031 g · mol^{-1}
T_c	374.27 K
P_c	4.065 MPa
V_c	199 cm^3 · mol^{-1}
Dipole moment	1 D

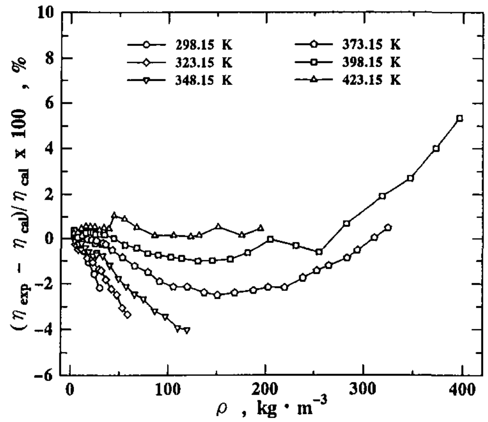

Fig. 6. Deviations of experimental viscosity values of HFC-134a under high pressures from those calculated with Lee–Thodos correlation.

Figure 4 shows the deviations of the present results from Eq. (5). For comparison, the deviation of the literature values of Nabizadeh and Mayinger [6] and Pasekov and Ustyushanin [9] are also shown. The data of Nabizadeh and Mayinger [6] show a larger density dependence than those of the present data. For engineering applications, we tried to correlate the present results with the viscosity equations proposed by Stiel and Thodos [3] and Lee and Thodos [4]. Table II lists the physical properties of HFC-134a taken from the JAR Thermodynamic Tables [23]. Figures 5 and 6 show the deviations of the present results from the correlations of Stiel and Thodos [3] and Lee and Thodos [4], respectively. Although Stiel and Thodos, and Lee and Thodos recommended their own equations for the viscosity at 0.1 MPa, we applied Eq. (1) for calculations of the viscosity at normal pressure in both correlations. It can be seen that the Lee–Thodos correlation represents the present results within 5%, which is satisfactory for practical purposes. However, the Stiel–Thodos correlation gives large deviations at high densities.

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