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Gas Viscosity of Difluoromethane from 298.15 to 423.15 K and up to 10 MPa

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The gas viscosity of difluoromethane was measured using an oscillating disk viscometer of the Maxwell type from 298.15 to 423.15 K at pressures up to 10 MPa. The results were fitted to an empirical equation as a function of temperature and density. Intermolecular force constants of the Lennard-Jones 12–6 potential were determined. In addition, the experimental density was compared with the calculated values from various equations of state previously reported.

Introduction

Chlorodifluoromethane (HCFC-22) has been used mostly as a refrigerant for air-conditioning systems, but it is scheduled to be phased out by 2030 due to its ozone depletion properties. Difluoromethane (HFC-32) has no ozone depletion potential (ODP) since there are no chlorine atoms in the molecule and has been considered as a substitute for HCFC-22. Various thermophysical properties of HFC-32 have recently been measured (Defibaugh et al., 1994; Holcomb et al., 1993; Kuwabara et al., 1992, 1995; Malbrunot et al., 1968; McLinden, 1990; Nishimura et al., 1992; Oliveira et al., 1993; Outcalt and McLinden, 1994; Qian et al., 1993; Ripple and Matar, 1993; Sato et al., 1994; Weber and Goodwin, 1993; Weber and Silva, 1994; Widiatmo et al., 1994), but its viscosity in the gas phase has been hardly measured (Dunlop, 1994). In this paper, the viscosity and density of HFC-32 gas were measured from 298.15 to 423.15 K at pressures up to 10 MPa. The results were fitted to an empirical equation for the viscosity as a function of temperature and density. Intermolecular force constants of the Lennard-Jones 12–6 potential were determined. In addition, the experimental density was compared with the calculated values from various equations of state previously reported (Defibaugh et al., 1994; Malbrunot et al., 1968; Nishimura et al., 1992; Outcalt and McLinden, 1994).

Experimental Section

Measurements were made using an oscillating disk viscometer of the Maxwell type, which could also be used for the measurement of the density. The experimental apparatus, the method of measurement, and the analysis of results were essentially the same as those described in the previous papers (Takahashi et al., 1988, 1989). The apparatus constant was determined using the viscosity of nitrogen recommended by Hanley and Ely (1973). The precisions in the measurements of temperature and pressure were ±0.01 K and ±0.5 KPa, respectively. The error in the measurements of density was ±0.12%, and the error in the viscosity was estimated to be ±0.3%.

The sample with 99.972 mass % purity was supplied by Daikin Industries, Ltd., Osaka, and was used without further purification. The physical properties of HFC-32 are given in Table 1 (Kuwabara et al., 1992; McLinden, 1990; Nishimura et al., 1992).

Results and Discussion

Figure 1 shows the temperature and pressure region (shaded parts) where the measurements of viscosity and density were made. The viscosity and density are given in Table 2 and Figures 2 and 3. In Figure 2, the intersects of the curve were observed around a temperature of 373.15 K, but there was no intersect in Figure 3. This behavior of the viscosity was known to be a characteristic of nonpolar gas.

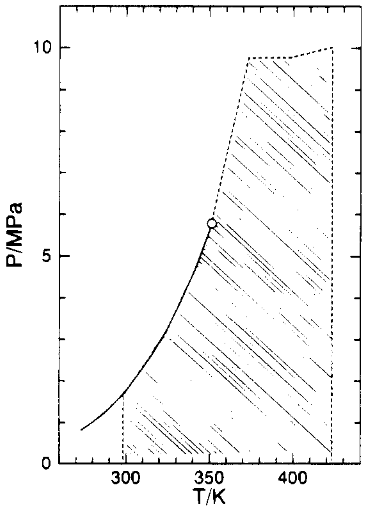

Figure 1. Temperature–pressure region of viscosity and density measurements, saturated vapor pressure, and critical point of difluoromethane: —, vapor pressure; ○, critical point.

Table 1. Physical Properties of Difluoromethane

physical property	
molecular formula	CH_2F_2
molecular weight	52.023
melting point	137 K (McLinden, 1990)
normal boiling point	221.41 K (Nishimura et al., 1992)
critical temperature	351.255 K (Kuwabara et al., 1992)
critical pressure	5.780 MPa (Nishimura et al., 1992)
critical density	424 kg·m^{-3} (Kuwabara et al., 1992)

The viscosity at atmospheric pressure, $\eta_1/(\mu\mathrm{Pa{\cdot}s})$, can be expressed as a function of temperature, T/K, in the following equation:

$$\eta_1 = T^{0.5}/(a_1 + a_2/T + a_3/T^2 + a_4/T^3) \qquad (1)$$

where a_i are constants ($i = 1-4$); $a_1 = -1.6434\ \mathrm{K}^{0.5}{\cdot}(\mu\mathrm{Pa{\cdot}s})^{-1}$, $a_2 = 2.5354 \times 10^3\ \mathrm{K}^{1.5}{\cdot}(\mu\mathrm{Pa{\cdot}s})^{-1}$, $a_3 = -7.7239 \times 10^5\ \mathrm{K}^{2.5}{\cdot}(\mu\mathrm{Pa{\cdot}s})^{-1}$, and $a_4 = 8.4516 \times 10^7\ \mathrm{K}^{3.5}{\cdot}(\mu\mathrm{Pa{\cdot}s})^{-1}$. The deviations of experimental viscosity at atmospheric pressure from eq 1 are shown in Figure 4. Two symbols, the open and filled versions, represent two sets of experimental data under the same conditions. The average and maxi-

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Table 2. Density ϱ and Viscosity η of Difluoromethane

P/MPa	ϱ/(kg·m^{-3})	η/(μPa·s)	P/MPa	ϱ/(kg·m^{-3})	η/(μPa·s)
T = 298.15 K					
0.1016	2.161	12.70	0.9664	23.33	12.50
0.1018	2.166	12.70	1.113	27.57	12.47
0.2362	5.114	12.65	1.260	32.08	12.48
0.3732	8.196	12.59	1.411	36.95	12.49
0.5178	11.66	12.56	1.558	42.21	12.48
0.6599	15.18	12.57	1.655	46.09	12.50
0.8207	19.35	12.54			
T = 323.15 K					
0.1019	1.825	13.67	1.644	38.01	13.72
0.1020	1.991	13.73	1.951	47.24	13.72
0.3972	7.977	13.65	2.302	59.20	13.85
0.7107	14.71	13.64	2.578	69.83	14.00
1.025	21.95	13.65	2.870	82.99	14.14
1.335	29.74	13.65	3.094	95.15	14.38
T = 348.15 K					
0.1008	1.825	14.70	3.824	101.5	15.78
0.1017	1.841	14.72	4.102	114.5	16.07
0.4814	8.962	14.68	4.345	127.5	16.40
0.8680	16.66	14.69	4.556	140.2	16.74
1.258	24.93	14.71	4.737	153.3	17.17
1.805	37.40	14.76	4.891	166.2	17.54
2.303	50.21	14.91	5.035	180.5	18.03
2.748	62.88	15.04	5.141	193.6	18.49
3.146	75.55	15.23	5.226	205.8	19.01
3.509	88.50	15.52	5.286	216.5	19.42
T = 373.15 K					
0.1015	1.711	15.67	5.966	165.9	18.74
0.1023	1.724	15.68	6.395	190.3	19.62
0.5001	8.619	15.66	6.875	223.6	20.94
0.9350	16.53	15.67	7.360	265.6	22.89
1.338	24.26	15.70	7.769	312.7	25.23
1.794	33.37	15.76	8.127	361.4	28.07
2.340	45.22	15.91	8.517	417.2	31.74
2.766	55.18	16.04	8.793	454.0	34.35
3.396	71.21	16.26	9.039	482.1	36.63
3.913	85.88	16.52	9.293	508.8	38.72
4.450	102.1	16.94	9.575	532.8	40.81
4.930	119.8	17.35	9.756	546.3	42.06
5.434	140.2	17.95			
T = 398.15 K					
0.1012	1.597	16.77	4.821	97.62	18.08
0.1014	1.637	16.77	5.300	110.6	18.39
0.5164	8.483	16.79	5.773	124.6	18.79
0.9253	15.47	16.82	6.283	141.1	19.29
1.340	22.31	16.87	6.828	160.0	19.87
1.782	30.31	16.93	7.331	179.1	20.53
2.333	40.86	17.09	7.862	201.5	21.39
2.734	49.08	17.16	8.392	226.2	22.33
3.333	61.65	17.36	8.895	251.5	23.46
3.834	73.06	17.56	9.278	272.6	24.43
4.353	85.67	17.81	9.771	300.6	25.86
T = 423.15 K					
0.1020	1.513	17.88	5.047	90.97	19.15
0.1022	1.516	17.88	5.495	101.2	19.41
0.5787	8.730	17.90	5.981	112.6	19.71
1.076	16.49	17.98	6.488	125.3	20.05
1.557	24.31	18.01	7.058	140.1	20.48
2.044	32.50	18.10	7.568	154.4	20.94
2.508	40.45	18.24	8.099	169.9	21.46
2.986	49.27	18.33	8.626	185.9	22.03
3.567	60.32	18.53	9.125	201.8	22.66
4.067	70.40	18.71	9.628	218.4	23.30
4.572	80.81	18.92	10.02	231.6	23.87

mum deviations were ±0.08% and ±0.18%, respectively. On the other hand, the viscosity value at atmospheric pressure and 298.15 K reported by Dunlop (1994) was 12.36 μPa·s, and its deviation from eq 1 was −2.71%.

Intermolecular force parameters of the Lennard-Jones 12−6 potential, ϵ/k and σ, were calculated by using the theoretical equation given by Hirschfelder et al. (1964). The parameters which fit the viscosity at atmospheric pressure were ϵ/k = 204.0 K and σ = 0.491 nm. The average deviation of the experimental values from the above-mentioned theoretical equation with these parameters was ±0.35%, and the maximum was ±0.70%.

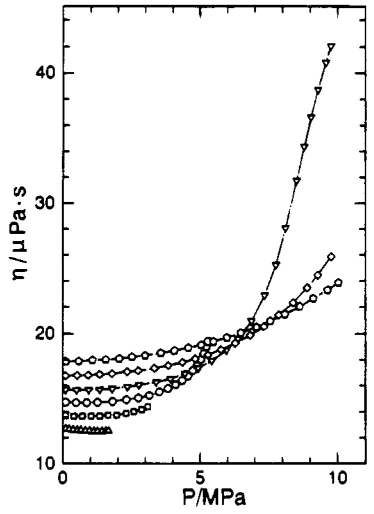

Figure 2. Viscosity η as a function of pressure P for Difluoromethane: △, 298.15 K; □, 323.15 K; ○, 348.15 K; ▽, 373.15 K; ◇, 398.15; ⬠, 423.15 K.

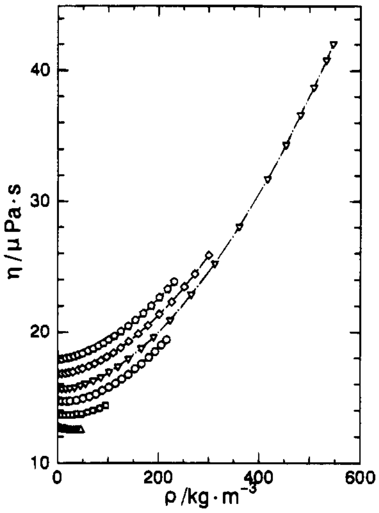

Figure 3. Viscosity η as a function of density ϱ for Difluoromethane: △, 298.15 K; □, 323.15 K; ○, 348.15 K; ▽, 373.15 K; ◇, 398.15; ⬠, 423.15 K.

Table 3. Constants in Eqs 2−7

constant	value
b_{11}/(μPa·s·K)	-3.2425×10
b_{12}/(μPa·s)	4.6097×10^{-1}
b_{13}/(μPa·s·K^{-1})	4.1097×10^{-2}
b_{21}/(μPa·s·(kg·m^{-3})$^{-1}$)	-1.2714×10^{-2}
b_{22}/(μPa·s·K^{-3}·(kg·m^{-3})$^{-1}$)	2.7075×10^{-10}
b_{31}/(μPa·s·(kg·m^{-3})$^{-2}$)	1.4978×10^{-4}
b_{32}/(μPa·s·K^{-3}·(kg·m^{-3})$^{-2}$)	-8.5144×10^{-13}
b_{41}/(μPa·s·K^{-1}·(kg·m^{-3})$^{-4}$)	-5.1452×10^{-8}
b_{51}/(μPa·s·(kg·m^{-3})$^{-6}$)	2.4270×10^{-16}

The viscosity, η/(μPa·s), can be expressed as a function of the temperature, T/K, and density, ϱ/(kg·m^{-3}), over the range of measurements as follows:

$$\eta = b_1 + b_2\varrho + b_3\varrho^2 + b_4\varrho^4 + b_5\varrho^6 \qquad (2)$$

$$b_1 = b_{11}/T + b_{12} + b_{13}T \qquad (3)$$

$$b_2 = b_{21} + b_{22}T^3 \qquad (4)$$

$$b_3 = b_{31} + b_{32}T^3 \qquad (5)$$

Table 4. Deviation of Experimental Density from Various Equations of State[a]

temp/K (data point)	eq of Malbrunot		eq of Nishimura		eq of Defibaugh		eq of Outcalt	
	max dev	av dev	max dev	av dev	max dev	av dev	max dev	av dev
298.15 (13)	+8.92	±3.67 (13)	−0.31	±0.16 (13)	+0.40	±0.22 (13)	+2.01	±0.56 (13)
323.15 (12)	+8.42	±3.38 (9)	−0.51	±0.24(11)	+0.28	±0.14 (11)	+3.72	±0.87 (9)
348.15 (20)	+5.29	±2.07 (9)	+2.14	±0.40 (20)	−0.27	±0.12 (13)	+3.18	±0.76 (9)
373.15 (25)	+2.98	±1.09(9)	−0.91	±0.26 (14)	−1.11	±0.25 (12)	+1.99	±0.39 (9)
398.15 (22)	+2.24	±1.09 (7)	−0.46	±0.28 (11)	−1.15	±0.31 (11)	+1.95	±0.67 (7)
423.15 (22)	+1.72	±0.54 (10)	−0.94	±0.50 (14)	−0.78	±0.23 (15)	+1.68	±0.37 (10)
total (114)	+8.92	±2.10 (57)	+2.14	±0.32 (83)	−1.15	±0.21 (75)	+3.72	±0.59 (57)

[a] Dev = $100(\varrho_{\text{exptl}} - \varrho_{\text{calcd}})/\varrho_{\text{exptl}}$.

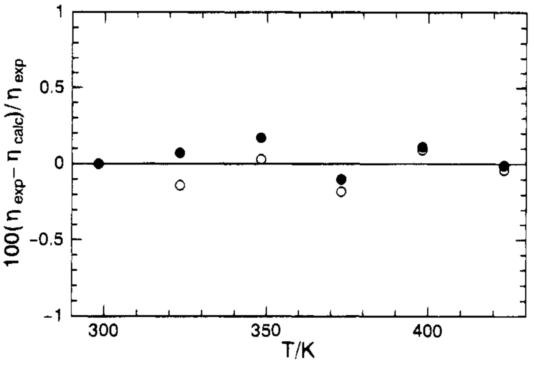

Figure 4. Deviation of experimental viscosity at atmospheric pressure from eq 1.

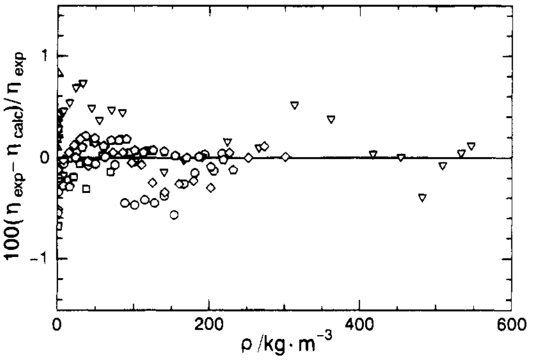

Figure 5. Deviation of experimental viscosity from eq 2. Symbols are the same as in Figures 2 and 3.

$$b_4 = b_{41}/T \qquad (6)$$

$$b_5 = b_{51} \qquad (7)$$

where b_{ij} are constants ($i = 1-5, j = 1-3$) and the values and units of each constant are listed in Table 3. The deviations of experimental viscosity from eq 2 are shown in Figure 5, and the average and maximum deviations were ±0.23% and ±0.93%, respectively.

Further, the experimental density was compared with the calculated values from various equations of state previously reported (Defibaugh et al., 1994; Malbrunot et al., 1968; Nishimura et al., 1992; Outcalt and McLinden, 1994). The maximum and average deviations of experimental density from each equation are shown in Table 4. Nishimura's equation has been developed only in the pressure range up to 6 MPa, and Defibaugh's equation has been used in the density range less than 2.56 mol·dm^{-3}. Malbrunot's and Outcalt's equations have been applied in the density range less than 1.54 mol·dm^{-3}. The calculated values from Nishimura's and Defibaugh's equations were in good agreement with the experimental values in the wide range of measurements.

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