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Revised Viscosities for HFC-134a + Glycol Mixtures from 273 to 333 K

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The liquid viscosity of six binary mixtures of HFC-134a with glycols [ethylene glycol, diethylene glycol, triethylene glycol, tetraethylene glycol, polyethylene glycol(400), and polypropylene glycol(2000)] was measured in the temperature range from 273 to 333 K. The viscosity was measured with an improved capillary viscometer instead of the rolling-ball viscometer reported in our previous work. The measurement uncertainty was estimated to be 1.6%. Most of the present data are higher than previous data, and the gap between the two sets of data increases further in low-viscosity regions. The present results obtained with laminar flow are more reliable than previous results, which are indefinite because of the limit of usefulness of the rolling-ball viscometer in low-viscosity regions.

KEY WORDS: ethylene glycol; diethylene glycol; HFC-134a; polyethylene glycol(400); polypropylene glycol(2000); tetraethylene glycol; triethylene glycol; viscosity.

1. INTRODUCTION

In our study of the viscosity of mixtures of new refrigerants with refrigerant oils, we previously obtained measurements for mixtures of HFC-134a with glycols in the temperature range from 273 to 333 K [1]. The viscosity of the mixtures decreases by a factor of about 1000 with temperature and composition of HFC-134a. The low viscosity near the critical Reynolds numbers cannot be measured accurately with the rolling-ball viscometer. In the present work, the viscosity of the mixtures was measured with an improved capillary viscometer, which can be used over a wide range of viscosity including a low-viscosity range.

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2. EXPERIMENTS

The purities of the samples of ethylene glycol (EG), diethylene glycol (DEG), triethylene (TEG), tetraethylene glycol (TTG), and HFC-134a were 99, 99, 99, 95, and 99.9%, respectively. The average molecular weights of polyethylene glycol (PEG) and polypropylene glycol (PPG) were 400 and 2000, respectively.

The viscosity was measured using the capillary viscometer shown in Fig. 1. This viscometer has the following characteristics.

1. Viscosity can be measured over a wide range by using several capillary tubes with different inner diameters in a pressure cell.
2. Small amounts of the sample (about 10 cm^3) can be weighed on a balance in order to calculate the exact composition.
3. The simple construction makes the viscometer easy to clean.
4. The correction for the composition is small because of the small vapor phase occupied by the sample in the viscometer.

The viscometer was located in a water bath whose temperature was regulated to within 30 mK. The temperature was measured with standard

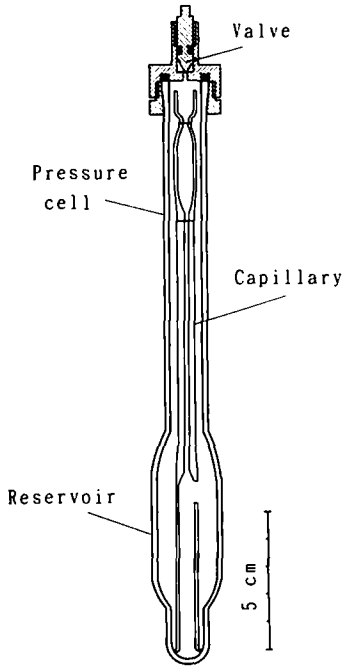

Fig. 1. Capillary viscometer.

Table I. Dimensions of Capillary Tubes and Apparatus Constants Used in the Viscometer

Tube No.	Inner diameter (mm)	Apparatus constant		Standard liquid
		C_1	C_2	
1	0.8	0.1182	10.725	JS5, JS10
2	1.0	0.3595	0	JS10
3	1.5	1.359	0	JS20

mercury thermometers calibrated at the National Research Laboratory of Metorology (Japan) within an accuracy of 30 mK. The maximum uncertainty in the viscosity, resulting from an absolute accuracy of temperature measurement, is estimated to be less than 0.7% for polypropylene glycol(2000) at 273.15 K. The procedure was described in detail previously [2]. The viscosity η (mPa·s) is calculated from the measured efflux time t, ranging from 24 to 1530 s, with a reproducibility of 0.5%, using the following equation:

$$\eta = C_1 \rho t - C_2 \rho / t \tag{1}$$

where C_1 and C_2 are the apparatus constants, and ρ (kg·m^{-3}) is the density of the sample liquid obtained from literature data [1]. The apparatus constants for the viscometer equipped with the three kinds of capillary tubes listed in Table I were calibrated with standard liquids (JS5,

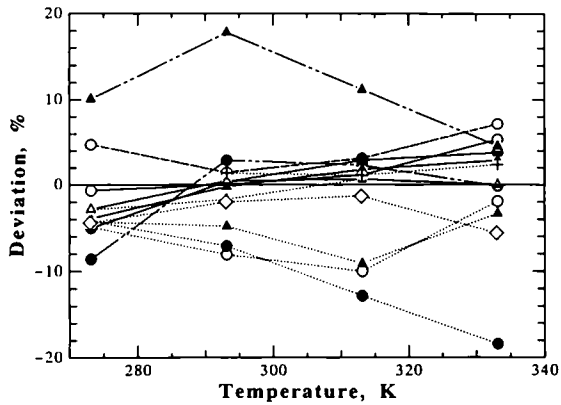

Fig. 2. Deviations of the experimental viscosity of pure glycols from the literature data. Deviation = $100(\eta_{\text{lit.}} - \eta_{\text{present work}})/\eta_{\text{present work}}$. (○) EG; (●) DEG; (▲) TEG; (△) TTG; (+) PEG; (◇) PPG; (·····) Ref. 1; (— · ·—) Ref. 3; (——) Ref. 4; (– – –) Ref. 5.

Table II. Viscosity (η) of HFC-134a + Glycol Mixtures as a Function of the Weight Fraction of Glycol (x)

T (K)							
273.15		293.15		313.15		333.15	
x	η (mPa·s)	x	η (mPa·s)	x	η (mPa·s)	x	η (mPa·s)
			HFC-134a + EG				
1.0	58.8	1.0	20.9	1.0	9.39	1.0	4.84
0.986	57.2	0.986	20.6	0.987	9.31	0.987	4.85
0.968	55.1	0.969	20.2	0.972	9.15	0.972	4.77
		0.941	19.2	0.944	8.81	0.955	4.69
0	0.2728[a]	0	0.2139[a]	0	0.1697[a]	0	0.1354[a]
			HFC-134a + DEG				
1.0	123.0	1.0	36.2	1.0	14.7	1.0	7.38
0.921	84.3	0.924	27.7	0.922	11.8	0.927	6.26
0.852	59.2	0.856	20.9	0.864	9.89	0.871	5.46
			HFC-134a + TEG				
1.0	171.4	1.0	48.4	1.0	19.2	1.0	9.40
0.935	127.7	0.937	37.5	0.916	14.4	0.922	7.54
0.814	63.5	0.821	22.9	0.835	11.0	0.849	6.33
0.718	34.6	0.723	14.2	0.737	7.44	0.750	4.67
			HFC-134a + TTG				
1.0	215.0	1.0	58.4	1.0	22.7	1.0	11.0
0.932	146.8	0.934	42.5	0.938	17.6	0.941	8.97
0.807	66.3	0.811	22.7	0.823	10.8	0.832	6.13
			HFC-134a + PEG(400)				
1.0	—	1.0	118.5	1.0	43.9	1.0	21.1
		0.942	87.1	0.946	33.9	0.950	17.3
0.842	172.8	0.848	53.1	0.864	23.6	0.877	13.1
0.766	105.0	0.775	35.6	0.795	17.6	0.814	10.7
0.682	57.9	0.686	20.3	0.694	10.2	0.706	6.34
0.597	30.0	0.600	12.4	0.614	6.84	0.626	4.68
			HFC-134a + PPG(2000)				
1	2118	1	438.4	1	147.5	1	66.6
0.834	360.2	0.840	116.6	0.854	53.8	0.866	30.6
0.720	113.5	0.725	46.3	0.737	25.2	0.749	16.3
0.641	54.2	0.646	26.4	0.662	15.9	0.677	11.7
0.566	27.0	0.572	14.8	0.585	9.81	0.598	7.66
0.409	7.71	0.496	8.93	0.506	6.22	0.517	5.20
0.356	4.91	0.414	4.74				

[a] From Ref. 2.

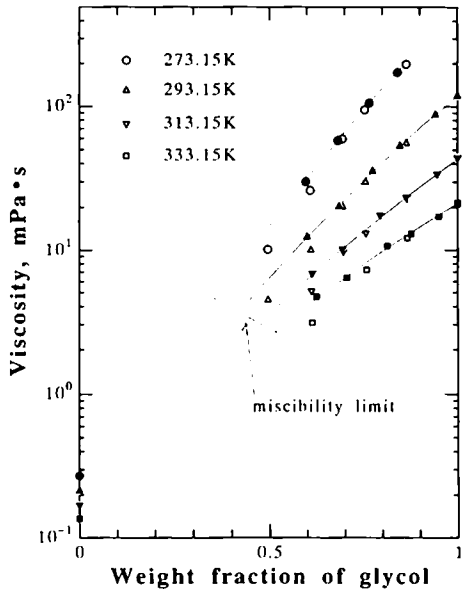

Fig. 3. Comparison of the viscosities obtained from the literature and in the present work for a HFC-134a + PEG mixture. Filled symbols (capillary): present work and Ref. 2 (pure HFC-134a). Open symbols (rolling ball): Ref. 1.

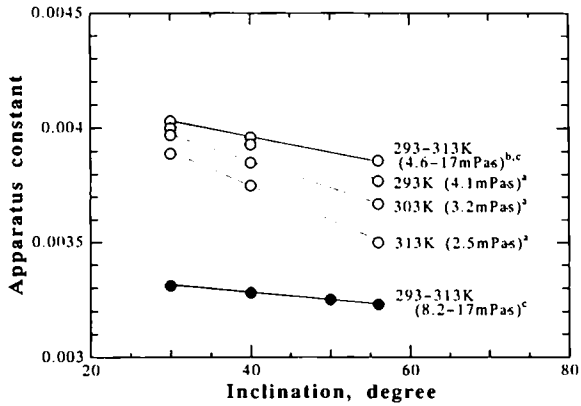

Fig. 4. Dependence of the apparatus constant on the temperature and the inclination of the tube in the rolling-ball viscometer. (○) Glass ball; (●) steel ball. Superscripts a, b, and c determined using standard liquids JS5, JS10, and JS20, respectively.

Table III. Parameters in Eq. (2) and Deviation of Experimental Viscosity Data from Eq. (2)

Parameter	HFC-134a + EG	HFC-134a + DEG	HFC-134a + TEG	HFC-134a + TTG	HFC-134a + PEG(400)	HFC-134a + PEG(2000)
f_1	2.18670	−2.09492	−3.78126	−2.11605	−4.17241	−1.09544
$10f_2$	−0.88175	0.10478	0.34651	1.03317	0.61702	−0.45750
f_3	1.37368	8.32498	14.0727	9.42909	18.2700	7.35264
$10f_4$	0.050824	−1.67508	−1.75868	−3.78333	−3.06886	1.06536
10^3f_5	−1.15061	2.76317	0.76825	0.79872	0.34060	1.09596
f_6	0.68665	−0.44242	−6.02655	−2.14874	−11.3173	0.08263
$10f_7$	1.39509	1.82103	0.94799	2.73456	2.27340	−1.85295
10^3f_8	2.75160	−5.32451	−0.84996	−0.67290	0.74218	−1.98557
f_9	−0.17497	−0.98077	0.88072	0.20431	3.26980	1.31706
$10f_{10}$	−1.11830	−0.91967	−0.23277	−0.69461	−0.52666	0.37372
10^3f_{11}	−1.36905	2.89870	0.43789	0.23608	−0.74340	1.38691
Av. dev. (%)[a]	0.8	1.0	1.1	1.3	1.3	2.0
Max. dev. (%)[b]	1.7	2.2	2.8	3.0	4.2	5.4

[a] $100[(\sum |\eta_{exp} - \eta_{calc}|/\eta_{calc})/n]$.

[b] Maximum of $100(|\eta_{exp} - \eta_{calc}|/\eta_{calc})$.

JS10, and JS20), the viscosity of which was known, between 298 and 313 K, with an accuracy of 0.4%. The accuracy of the viscosities was estimated to be 1.6%. The measurements were performed at Reynolds numbers of less than 22, which is well below the critical value of 2300.

3. RESULTS

Viscosity values for pure glycols in the present work agree with the literature data [1, 3–5] within 10% except for those DEG obtained for DEG by Kumagai et al. [1] and, for TEG by Obermeier et al. [3] as shown in Fig. 2. In Table II, the experimental data are listed for the viscosity of six binary mixtures of HFC-134a with glycol. Most of the present data are higher than previous data [1] obtained with a rolling-ball viscometer, and the gap between the two sets of data increases in the low-viscosity regions. A representative result for the viscosity of HFC-134a with PEG mixture is shown in Fig. 3. The present results obtained in laminar flow are more reliable than previous data [1], which are indefinite because of the significant dependence of the apparatus constant on both the temperature and the inclination of the tube in low-viscosity regions, as shown in Fig. 4. The viscometer should essentially have a fixed apparatus constant calculated from the dimensions of the viscometer according to the method of Hubbard and Brown [6].

Experimental viscosity values η (mPa·s) were fitted to the following equation in the range of miscibility.

$$\ln \eta = (f_1 + f_2 T) + (f_3 + f_4 T + f_5 T^2)x + (f_6 + f_7 T + f_8 T^2)x^2 + (f_9 + f_{10} T + f_{11} T^2)x^3 \quad (2)$$

where x is the weight fraction of glycol, T is the temperature (K), and $f_{1\ 11}$ are adjustable parameters. The parameters in Eq. (2) and the deviations of the experimental data from Eq. (2) are listed in Table III.

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