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Viscosity of Saturated Liquid Fluorocarbon Refrigerants from 273 to 353 K

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Viscosity measurements were carried out on saturated liquid fluorocarbon refrigerants using an improved capillary viscometer for 11 kinds of fluorocarbon refrigerants; CCl_3F (R11), CCl_2F_2 (R12), $CHClF_2$ (R22), $CBrF_3$ (R13B1), CH_3CHF_2 (R152a), CCl_2FCClF_2 (R113), $CHCl_2CF_3$ (R123), $CHClFCClF_2$ (R123a), CH_3CF_3 (R143a), $CClF_2CClF_2$ (R114), and CH_2FCF_3 (R134a), in the temperature range from 273 to 353 K. An equation is given to represent the viscosity as a function of temperature.

KEY WORDS: capillary viscometer; fluorocarbons; refrigerants; viscosity.

1. INTRODUCTION

There are significant differences among saturated liquid viscosity values for some liquefied fluorocarbon refrigerants previously reported. The present authors believe that these differences arise from large Reynolds numbers and inaccurate correction factors. In the present study, Reynolds numbers and correction factors were checked in the viscosity measurements of low-viscosity liquids, and viscometers were designed in order that Reynolds numbers were small and that the correction factor approached unity.

Using a designed viscometer, measurements of saturated liquid viscosities were performed in the temperature range from 273 to 353 K for the following fluorocarbon refrigerants: R11, R12, R22, R13B1, R152a, R113, R123, R123a, R143a, R114, and R134a.

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2. APPARATUS AND EXPERIMENTAL METHOD

Figure 1 shows four kinds of capillary viscometers designed for this study. The viscometers were made of borosilicate glass and can stand pressures up to 2 MPa. Table I shows dimensions and apparatus constants of these viscometers. Figure 2 shows a block diagram of the viscosity apparatus. The sample liquid was distilled through a valve (A) into the viscometer evacuated in advance, until a certain amount of liquid was collected in the lower bulb (H). Then, the viscometer was mounted in the transparent thermostat as shown in Fig. 2. When the thermostat was reversed by rotation, the liquid sample was transfered into the upper bulb (B) and the efflux bulb (D). After thermal equilibrium had been established, the thermostat was returned to the original position. Then liquid flowed down through the capillary (F) into the lower bulb (H), and the liquid surface in B fell. The efflux time was measured by an electric counter. The moment when the liquid surface passed through the mark (C) was determined by visual observation. The moment when the liquid surface passed through the mark (E) was determined by an optical detector connected with the electric counter.

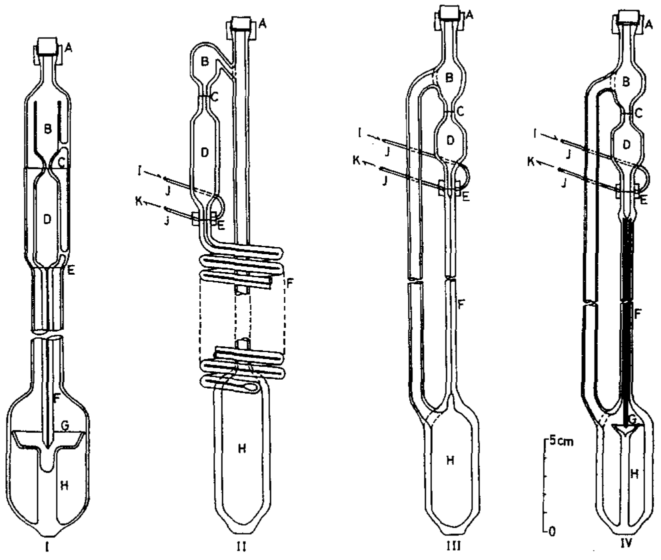

Fig. 1. Capillary viscometers. A, valve; B, upper bulb; C and E, fiducial marks; D, efflux bulb; F, capillary; G, pan; H, lower bulb; I, laser; J, optical fiber; K, photodiode.

Table I. Dimensions and Apparatus Constants of Viscometers

Viscometer	Volume of bulb, v (cm^3)	Inner diameter of capillary, d (cm)	Radius of upper meniscus, r_u (cm)	Radius of lower meniscus, r_l (cm)	Average driving head, h_{av} (cm)	Viscometer constant	
						$C_1 \times 10^7$	$C_2 \times 10^3$
I	5.0	0.050	0.6	1.7[b]	13	48.83	1.805
II[a]	4.7	0.050	0.5	0.2	12	3.700	1.432
III	2.5	0.025	0.7	0.05–0.1[c]	12	5.752–5.910	0
IV	3.0	0.025	0.7	0.7[b]	11	3.606	0

[a] The coil diameter of the viscometer, D, is 4.5 cm.

[b] Radius of pan.

[c] Tentative value.

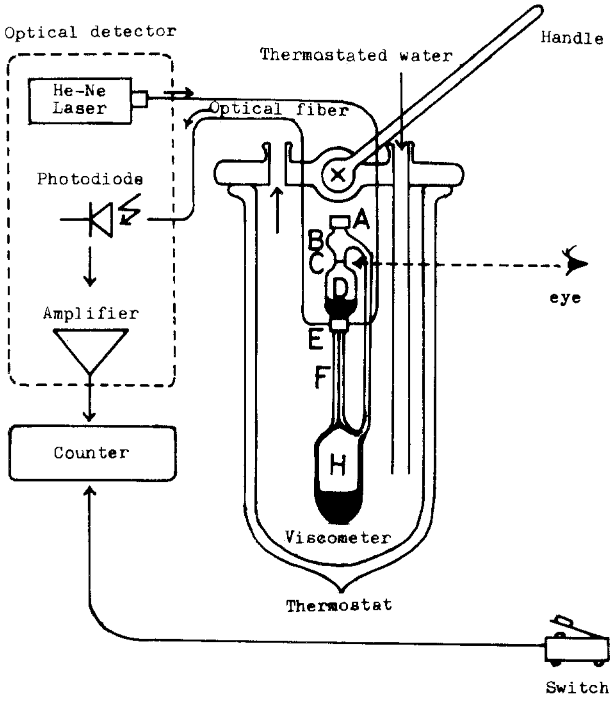

Fig. 2. Block diagram of the viscosity apparatus. Key is the same as in the legend to Fig. 1.

Table II. Reynolds Numbers and Correction Factors of Viscometers of Four Types for R12[a]

	Type of viscometer				
			III r_1 (cm)		
T (K)	I	II	0.05	0.10	IV
303.15					
Re	2100	200	340	340	250
k	0.9990	1.003	1.019	1.009	1.000
c	1.000	1.039	1.000	1.000	1.000
η	0.1941	0.1885	0.1906	0.1940	0.1910
323.15					
Re	2500	250	430	430	320
k	0.9992	1.002	1.015	1.007	1.000
c	1.000	1.076	1.000	1.000	1.000
η	0.1632	0.1550	0.1586	0.1617	0.1585
343.15					
Re	2800	290	530	530	390
k	0.9995	1.002	1.010	1.004	1.000
c	1.000	1.109	1.000	1.000	1.000
η	0.1393	0.1295	0.1318	0.1347	0.1320

[a] Re, Reynolds number; k, correction factor due to surface tension; c, correction factor due to coil curvature; η, viscosity (mPa · s).

Table III. Viscosity of Chloroform

		η (mPa · s)	
T (K)	ρ (kg · m^{-3}) [5]	Present work	Phillips *et al.* [6]
304.73	1467.0	0.5086	0.5093
319.42	1438.5	0.4434	0.4450
334.41	1408.7	0.3898	0.3922

Table IV. Viscosity of Fluorocarbon Refrigerants

T (K)	t (s)	ρ ($\mathrm{kg \cdot m^{-3}}$)	η ($\mathrm{mPa \cdot s}$)
	R11		
273.15	964.7	1534.3[a]	0.5337
283.15	873.8	1511.3[a]	0.4761
293.15	796.7	1487.8[a]	0.4274
303.15	731.7	1463.9[a]	0.3863
313.15	675.5	1439.5[a]	0.3506
323.15	626.5	1414.4[a]	0.3190
333.15	583.3	1388.6[a]	0.2921
343.15	545.1	1362.0[a]	0.2677
353.15	510.6	1334.5[a]	0.2457
	R12		
273.15	509.0	1396.8[a]	0.2564
283.15	471.1	1363.7[a]	0.2317
293.15	438.4	1328.9[a]	0.2100
303.15	409.9	1292.2[a]	0.1910
313.15	384.9	1253.1[a]	0.1739
323.15	362.9	1211.0[a]	0.1585
333.15	344.3	1165.2[a]	0.1446
343.15	328.5	1114.6[a]	0.1320
	R22		
273.15	459.0	1284.8[a]	0.2127
283.15	426.3	1249.9[a]	0.1921
293.15	397.8	1213.1[a]	0.1740
303.15	373.0	1173.8[a]	0.1579
313.15	351.6	1131.3[a]	0.1434
323.15	333.7	1084.6[a]	0.1305
	R13B1		
273.15	321.0	1703.0[a]	0.1971
283.15	300.9	1641.7[a]	0.1781
293.15	283.6	1574.4[a]	0.1610
303.15	269.8	1499.0[a]	0.1458
313.15	259.8	1411.7[a]	0.1323

[a] From Ref. 7.

Table IV. (*Continued*)

	R152a		
273.15	618.6	958.4[a]	0.2138
283.15	567.5	935.4[a]	0.1914
293.15	523.4	911.5[a]	0.1720
303.15	485.0	886.2[a]	0.1550
313.15	451.5	859.1[a]	0.1399
323.15	422.9	829.6[a]	0.1265
333.15	398.2	797.2[a]	0.1145
343.15	377.3	761.4[a]	0.1036
	R113		
273.15	1602.7	1621.2[a]	0.9369
283.15	1399.7	1596.2[a]	0.8057
293.15	1236.9	1576.5[a]	0.7032
303.15	1102.4	1553.1[a]	0.6175
313.15	991.3	1529.1[a]	0.5467
323.15	897.3	1504.4[a]	0.4868
333.15	816.9	1478.9[a]	0.4356
343.15	747.7	1453.0[a]	0.3918
353.15	687.1	1424.6[a]	0.3530
	R123		
273.15	1029.6	1524.2[b]	0.5659
283.15	923.6	1499.1[b]	0.4993
293.15	834.4	1474.0[b]	0.4435
303.15	759.5	1448.6[b]	0.3967
313.15	695.7	1422.8[b]	0.3569
323.15	640.3	1396.4[b]	0.3224
333.15	591.1	1369.2[b]	0.2918
343.15	548.0	1340.9[b]	0.2650
353.15	510.0	1311.4[b]	0.2412
	R123a		
273.15	1083.9	1528.4[b]	0.5974
283.15	969.9	1504.3[b]	0.5261
293.15	875.5	1479.5[b]	0.4671
303.15	795.3	1453.9[b]	0.4170
313.15	726.3	1427.4[b]	0.3738
323.15	667.5	1400.1[b]	0.3370
333.15	616.6	1371.8[b]	0.3050
343.15	571.3	1342.5[b]	0.2766
353.15	530.6	1312.2[b]	0.2511

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[b] From Ref. 8.

Table IV. (*Continued*)

R143a			
273.15	439.9	1022.2[b]	0.1621
283.15	406.0	985.6[b]	0.1443
293.15	376.9	946.0[b]	0.1286
303.15	353.0	904.1[b]	0.1151
313.15	334.1	856.0[b]	0.1031
323.15	320.5	800.5[b]	0.0925
R114			
273.15	863.2	1529.4[a]	0.4760
283.15	774.8	1500.9[a]	0.4193
293.15	700.0	1471.4[a]	0.3714
303.15	636.6	1440.8[a]	0.3307
313.15	581.4	1408.8[a]	0.2954
323.15	534.9	1375.4[a]	0.2652
333.15	493.9	1340.1[a]	0.2387
343.15	457.5	1302.7[a]	0.2149
353.15	425.9	1262.6[a]	0.1939
R134a			
273.15	584.5	1294.5[b]	0.2728
283.15	530.5	1260.4[b]	0.2411
293.15	484.0	1225.4[b]	0.2139
303.15	444.6	1187.9[b]	0.1904
313.15	410.3	1147.2[b]	0.1697
323.15	381.0	1102.6[b]	0.1515
333.15	356.7	1052.7[b]	0.1354
343.15	336.7	996.7[b]	0.1210

The viscosity, η (mPa · s), was calculated with the following equation,

$$\eta = C_1(k/c)\,\rho t - C_2\rho/t \qquad (1)$$

where C_1 and C_2 are the viscometer constants, ρ ($\mathrm{kg \cdot m^{-3}}$) is the density of the sample liquid, t(s) is the efflux time. k is the surface-tension correction factor for the driving head given by

$$k = 1 - 2(\gamma/gh_{\mathrm{av}}\rho)(1/r_u - 1/r_1) \qquad (2)$$

Here c is a numerical coefficient, representing the increase in resistance due to the curvature of the coil in the viscometer of type II, and it is given by

$$c^{-1}=1-[1-(11.6/\kappa)^{0.45}]^{2.22} \tag{3}$$

$$c^{-1}=1 \text{ (no correction)} \qquad \text{for } \kappa \leqslant 11.6 \tag{4}$$

$$\kappa=\mathrm{Re}(d/D)^{0.5} \tag{5}$$

In Eqs. (2–5), $\gamma(\mathrm{N \cdot m^{-1}})$ is the surface tension of the liquid, $g(\mathrm{m \cdot s^{-2}})$ is the acceleration due to gravity, $h_{\mathrm{av}}(\mathrm{m})$ is the average driving head, $r_{\mathrm{u}}(\mathrm{m})$ is the radius of the upper meniscus, $r_{\mathrm{l}}(\mathrm{m})$ is the radius of the lower meniscus, Re is Reynolds number, $d(\mathrm{m})$ is the inner diameter of the capillary, and $D(\mathrm{m})$ is the diameter of the coil [1]. The surface tension values were taken from the handbooks for H_2O [2], $CHCl_3$ [2], R11 [3], R12 [3], R22 [3], R13B1 [3], and R114 [3] and were estimated by the MacLeod–Sugden correlation [4] for other refrigerants.

Table II shows Reynolds numbers and the correction factors of the viscometers of four types in the case of R12. The viscometer of type I gave high Reynolds numbers. The viscometer of type II, which was miniaturized by coiling the long capillary tube (1.5 m), yielded low Reynolds numbers but gave a much larger correction factor, c, than unity. The correction factor, c, increased to about 1.1 at 343 K. The viscometer of type III of a suspended-level type yielded low Reynolds numbers, but it was difficult to measure the radius of meniscus r_{l} at the capillary end accurately and to estimate the correction factor, k. For example, $\eta=0.1318\ \mathrm{mPa \cdot s}$

Table V. Constants in the Viscosity Equation (6), with η in $\mathrm{mPa \cdot s}$ and T in K[a]

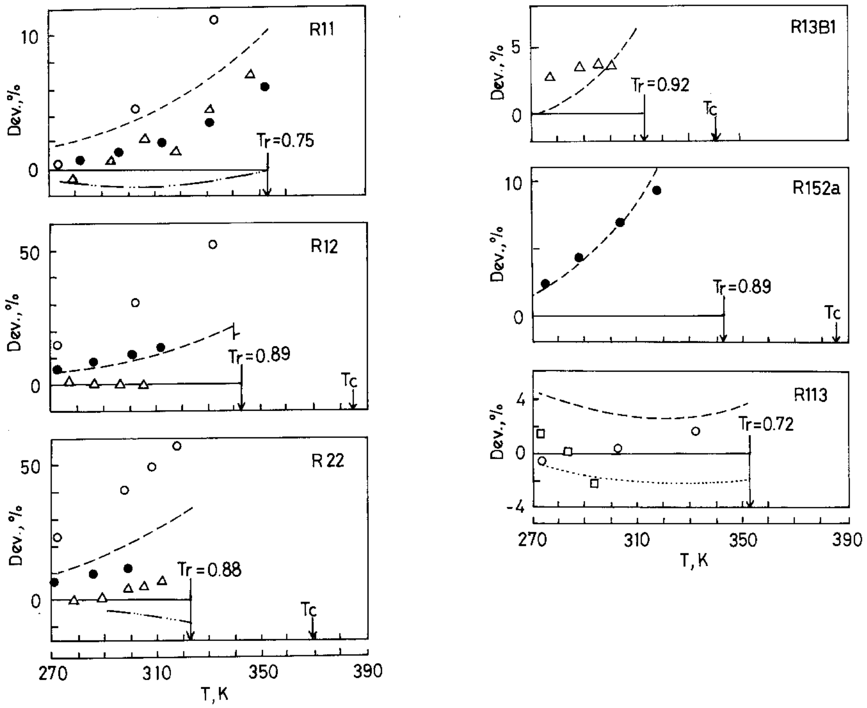
		$\ln \eta = A + B/T + CT + DT^2$					
Refrigerant	Temp. range (K)	A	$B \times 10^{-3}$	$C \times 10^{2}$	$D \times 10^{5}$	Av. dev.	Max. dev.
R11	273–353	−8.446	1.2848	1.7420	−2.2029	0.04	0.12
R12	273–343	−6.928	0.9066	1.4634	−2.3442	0.02	0.03
R22	273–323	−0.4470	0.2319	0.7139	0.0	0.02	0.04
R13B1	273–313	−0.1116	0.1769	−0.7907	0.0	0.03	0.04
R152a	273–343	−7.807	1.0102	1.6530	−2.6127	0.01	0.02
R113	273–353	−13.694	2.0657	3.2253	−3.6781	0.05	0.13
R123	273–353	−11.093	1.6032	2.5615	−3.1391	0.03	0.05
R123a	273–353	−9.172	1.4167	1.9553	−2.5066	0.04	0.08
R143a	273–323	−0.7046	0.2884	−0.7947	0.0	0.03	0.06
R114	273–353	−8.597	1.2980	1.8424	−2.5860	0.03	0.08
R134a	273–343	−7.639	1.0543	1.6602	−2.7532	0.01	0.03

[a] Ave. dev. (%) $= \{[\sum (\eta_{\mathrm{exp}} - \eta_{\mathrm{eq}})/\eta_{\mathrm{eq}}]/n\} \times 100$. Max. dev. (%) = maximum of $\{(\eta_{\mathrm{exp}} - \eta_{\mathrm{eq}})/\eta_{\mathrm{eq}}\} \times 100$; η_{eq}, viscosity calculated from Eq. (6); η_{exp}, viscosity in Table IV.

Table VI. Experimental Investigations of Viscosity of Liquid Fluorocarbon Refrigerants

Investigator	Year	Refrigerant	Temp. range (K)	Press. range (MPa)	Method	Purity (%)	No. of data	Accuracy (%)	Ref. No.
Benning et al.	1938	R11	243–333	Sat'n[a]	Roll. ball	—	4	—	9
		R12	243–332	Sat'n	Roll. ball	—	5	—	
		R22	240–318	Sat'n	Roll. ball	—	5	—	
		R113	243–333	Sat'n	Roll. ball	—	4	—	
		R114	242–332	Sat'n	Roll. ball	—	5	—	
Lilios	1957	R12	213–243	Sat'n	Capillary (coil)	—	4	2.20	10
			213–243	Sat'n	Roll. ball	—	4	—	
		R13B1	203–218	Sat'n	Roll. ball	—	4	—	
		R113	243–293	Sat'n	Roll. ball	—	6	—	
Gordon et al.	1969	R11	245–347	Sat'n	Capillary	—	9	—	11
		R12	245–305	Sat'n	Capillary	—	7	—	
		R22	246–312	Sat'n	Capillary	—	8	—	
		R13B1	246–301	Sat'n	Capillary	—	7	—	
Phillips et al.	1970	R11	209–352	Sat'n	Capillary (coil)	99.90	9	—	12
		R12	202–312	Sat'n	Capillary (coil)	99.90	9	—	
		R22	201–299	Sat'n	Capillary (coil)	99.90	8	—	
		R152a	200–318	Sat'n	Capillary (coil)	99.90	9	—	
Nagashima et al.	1975	R11	273–453	0.5–11	Capillary	99.998	51	1.6	13
Fujioka et al.	1977	R22	293–353	3–20	Capillary	99.985	16[b]	0.87	15
Nagashima et al.	1985	R113	273–473	0.5–30	Capillary	99.991	32[b]	0.83	16
Shankland et al.	1988	R134a	251–343	Sat'n	Capillary (coil)	99.9	12	—	17

[a] Saturation pressure.
[b] Graphically presented.



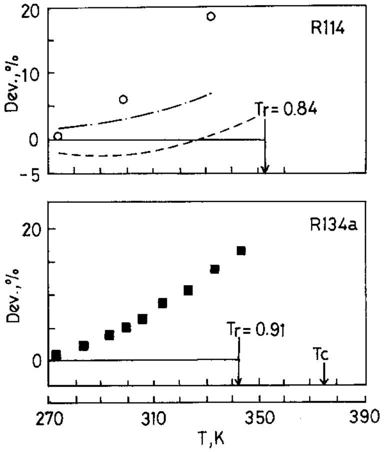


Fig. 3. Deviations of literature values (η_{lit}) from those calculated by Eq. (6) (η_{eq}). Dev. $= 100\,(\eta_{\mathrm{lit}} - \eta_{\mathrm{eq}})/\eta_{\mathrm{eq}}$. (○) Ref. 9; (□) Ref. 10; (△) Ref. 11; (● and ---) Ref. 12; (-–-–) Ref. 13; (———) Ref. 14; (-----) Ref. 15; (--------) Ref. 16; (■) Ref. 17.

assuming that $r_1 = 5 \times 10^{-4}$ m, and $\eta = 0.1347$ mPa·s assuming that $r_1 = 1.0 \times 10^{-3}$ m. In the case of low-viscosity liquids, the sample liquid sometimes flowed out in droplets from the capillary end. The viscometer of type IV was equipped with a pan at the capillary end, and the accurate r_1 value could be determined. The correction factor of this viscometer due to surface tension, k, is unity as $r_u \to r_1$.

3. EXPERIMENTAL RESULTS

The apparatus constants of the viscometer of type I were calibrated using water [2] through the temperature range of 278–348 K. Chloroform measured with the viscometer of type I was used as the standard for the calibration of the apparatus constants of the viscometers of types II–IV, since it was difficult to pass water through the capillary in these viscometers. Table III shows the viscosity of chloroform measured by the viscometer of type I together with the literature data [6]. The present values were found to agree with the literature values within 0.6%.

The viscosity of the fluorocarbon refrigerants was measured with the viscometer of type IV over the temperature range from 273 to 353 K or to the temperatures at which the saturated vapor pressures reached 2 MPa. The temperature was controlled to within ±0.03 K. The experimental viscosities together with the density data [7, 8] are presented in Table IV. The efflux time values in Table IV represent the averages of five readings at the same temperature, and the reproducibility is 0.1%. The time ranged from 260 to 1600 s and the Reynolds numbers ranged from 17 to 630 depending on the substance and temperature. The estimated error of the viscosity values was less than 0.5%. The purities of samples analyzed by gas chromatography were better than 99.8% except for R123 (99.5%).

The present viscosity values were fitted to the equation,

$$\ln \eta = A + B/T + CT + DT^2 \qquad (6)$$

where T(K) is the absolute temperature, and A, B, C, and D are constants. Table V gives the constants and shows that Eq. (6) reproduces the present data within the estimated error.

4. COMPARISON WITH VALUES FROM THE LITERATURE

Table VI shows the previous experimental investigations of the viscosity of fluorocarbon refrigerants. Figure 3 shows deviations of the

Table VII. Deviations of Correlations (η_{cor}) Given by Gordon et al. [11], Li and Poole [18], and Srinivasan and Murthy [19] from the Present Experimental Viscosities (η_{eq})^a

Refrigerant	T_c (K)	T_r range	No. of present data, n	Gordon et al.		Li and Poole		Srinivasan and Murthy	
				Av. dev.	Bias	Av. dev.	Bias	Av. dev.	Bias
R11	471.2	0.58–0.75	9	3.7	−2.6	1.8	−1.4	6.4	6.4
R12	385.0	0.71–0.89	8	3.5	−3.5	14.9	−14.9	5.6	−4.9
R22	369.2	0.74–0.88	6	9.1	−9.1	19.4	−19.4	12.2	−12.2
R13B1	340.2	0.80–0.92	5	11.6	11.6	19.0	−19.0	10.3	−10.3
R152a	386.6	0.71–0.89	8	50.3	−50.3	19.9	−19.9	32.5	−32.5
R113	487.2	0.56–0.72	9	6.9	−3.3	6.2	4.1	17.7	−17.7
R123	459.5	0.59–0.77	9	6.6	−5.2	3.9	1.7	3.8	−0.1
R123a	465	0.59–0.76	9	—	—	6.0	5.5	—	—
R143a	346.3	0.79–0.93	6	13.0	−13.0	20.3	−20.3	31.0	−31.0
R114	418.9	0.65–0.84	9	4.4	−4.2	6.5	0.3	2.6	1.4
R134a	375	0.73–0.91	8	10.9	−10.9	9.4	−7.0	13.1	−13.1

^{a} Av. dev. (%) = $\{[\sum (\eta_{cor} - \eta_{eq})/\eta_{eq}]/n\} \times 100$. Bias (%) = $\{[\sum (\eta_{cor} - \eta_{eq})/\eta_{eq}]/n\} \times 100$.

literature values from those calculated by Eq. (6). The ASHRAE equation [14] based on the lietrature data shows poor agreement with the present results.

Table VII shows the deviations of the viscosities given by previous correlations [11, 18, 19] from those presented in this publication. There is little to choose among the correlations, but the reproducibilities are invariably poor for the hydrogen-containing refrigerants.

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