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Measurements of the Viscosity of Liquid R22, R124, and R125 in the Temperature Range 273–333 K at Pressures up to 17 MPa

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Viscosity masurements of refrigerants R22, R124, and R125 in the liquid phase have been performed in the temperature range 273–333 K and at pressures up to about 17 MPa. A vibrating-wire instrument has been employed. The overall uncertainty of the experimental values is estimated to be ±0.5%. The experimental data have been represented by polynomial functions of temperature and pressure for the purposes of interpolation.

KEY WORDS: high pressure; liquid; refrigerants; R22; R124; R125; vibrating-wire technique; viscosity.

1. INTRODUCTION

As the time of the decision concerning the replacement of the old generation of refrigerants is approaching, the need for accurate measurements of the equilibrium as well as the transport properties becomes more pronounced. In the specific case of the viscosity in the liquid phase, few measurements exist and these are mostly at saturation conditions. Furthermore, in general, the dicrepancies between the investigators, up to 20% in extreme cases, far exceed the quoted uncertainties.

During the last 3 years we have undertaken a program of measurement of the transport properties of the old and new refrigerants in the liquid and vapor phases. We have, hence, already measured the viscosity of R11, R12, R141b, and R152a [1] and R134a and R32 [2], in the liquid

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phase up to about 20 MPa pressure. The data presented in this work conclude this series of measurements.

The viscosities of R22 (chlorodifluoromethane), R124 (1-chloro-1,2,2,2-tetrafluoroethane), and R125 (pentafluoroethane) have been measured with a vibrating-wire instrument. R124 and R125 are new refrigerants possible substitutes of R114 and R502 (an azeotropic mixture of R22 and R115), respectively.

As already discussed [1], the large discrepancies observed in the measurement of the viscosity of liquid refrigerants are attributed mostly to three main effects: impurities in the samples, electrolytic effects in the instruments, and incapability of some instruments to be calibrated with water. The measurements presented in this paper are believed, as it has been shown [1–3], to be free of all these effects. The vibrating-wire technique has already been proved to be capable of attaining high accuracy in the masurement of the viscosity of liquids [3]. The viscometer, calibrated against water, enables high-pressure measurements with an uncertainty of about ±0.5%.

2. EXPERIMENTS

The measurements were carried out in a vibrating-wire instrument that has been described in detail elsewhere [3]. It should however be pointed out that the entire instrument (including even electrical leads), is made out of stainless steel with the exception of the vibrating wire itself and the inner weight [3], which are made out of tungsten. This arrangement was found to be necessary to eliminate electrolytic effects [1, 3] due to the dissimilar metals present in the earlier version of the instrument. Sealing of the vessel was achieved by an especially fabricated PTFE-coated Viton O-ring to protect the O-ring from chemical attack [3]. Water, whose viscosity is very accurately known, is used for calibration of the viscometer. Taking all parameters into account, the uncertainty of the instrument is thus estimated to be ±0.5% while the precision and the reproducibility of the measurements are about ±0.1%. The use of water in the instrument served also as a check that no electrolytic effects are taking place. Samples were introduced after evacuation of the system and a stainless-steel 20- to 30-μm microfilter in the inlet of the instrument ensured that no solid particles from the bottles could enter. The viscosity of water was measured before and after each liquid to ensure the continuing good operation of the instrument. The temperature of the viscometer was measured by two platinum-resistance thermometers calibrated to ±10 mK, placed in the two ends of the pressure vessel. The pressure vessel was immersed in a 150-L water–glycol bath whose temperature stability and uniformity were better than ± 10 mK over hours. The pressure was registered by a Druck pressure gauge calibrated to ±0.2 MPa.

The R22 sample was supplied by SICNG Chemical Industries of Northern Greece S.A., whereas the R124 and R125 samples were supplied by Du Pont de Nemours International S.A., all at nominal purities better than 99.95%.

3. RESULTS

The measurements of the viscosity of R22 and R124 were performed along four isotherms, of 273.15, 293.15, 313.15, and 333.15 K. In the case of R125, a restriction was imposed as far as the higher temperature was concerned, due to its comparatively low critical temperature. Thus, the R125 viscosity was measured at 273.15, 293.15, and 313.15 K. All isotherms extended from above the saturation pressure up to about 17 MPa. The density values employed in the calculation of the viscosity of R22 were taken from the correlation proposed by Blanke and Weiss [4]. This correlation, based on their experimental data, has a quoted uncertainty of ±0.01%. For R124, the density values published by Kubota et al. [5], with a quoted uncertainty of ±0.09%, were used. It should, however, be pointed out that the lowest temperature at which density measurements were performed was 283.15 K. Since, to our knowledge, no other experimental measurements of the density exist, extrapolation to 273.15 K was necessary. The density values of R125 were taken from the equation given by Defibaugh and Morrison [6], which was based on their experimental measurements, with a quoted uncertainty of ±0.05%. The pressure range covered by their values extends to 6.3 MPa only, but since these were the only experimental measurements of the density available, this equation had also to be extrapolated to 15 MPa. To examine further the temperature extrapolation in the density of R124 and the pressure extrapolation in the density of R125, the corresponding-states values presented by Diller and Peterson [7] were compared with the aforementioned values. Thc agreement was within ±0.5%.

The measurements of the viscosity of R22, R124, and R125 are shown in Tables I, II, and III respectively. For the purpose of interpolation only, all measurements have been correlated as a function of the reduced temperature $T_r = T/T_c$ and the reduced pressure $P_r = P/P_c$ (where T_c and P_c are the critical temperature and pressure), by an equation of the form

$$\eta = \sum_{i=0}^{2} \sum_{j=0}^{3} C_{ij} P_r^i T_r^j \qquad (1)$$

Table I. Measurements of the Viscosity of R22

Pressure P (MPa)	Temperature T (K)	Viscosity $\eta(T, P)$ (μPa·s)	Density $\rho(T_{nom}, P)$ (kg·m^{-3})	Viscosity $\eta(T_{nom}, P)$ (μPa·s)
$T_{nom} = 273.15$ K				
1.30	273.194	212.6	1284	212.7
2.60	273.148	215.9	1289	216.0
3.90	273.178	219.3	1294	219.3
5.45	273.190	223.0	1299	223.1
6.90	273.176	226.6	1304	226.5
8.20	273.112	229.4	1309	229.5
9.60	273.123	232.8	1313	232.6
11.80	273.217	237.1	1320	237.3
$T_{nom} = 293.15$ K				
1.75	293.189	172.8	1214	172.8
3.60	293.189	177.3	1224	177.4
5.80	293.196	182.5	1235	182.7
7.90	293.221	187.6	1244	187.6
9.90	293.203	192.0	1252	192.2
11.65	293.189	196.0	1259	196.2
13.40	293.183	199.9	1266	200.0
15.00	293.210	203.4	1272	203.5
$T_{nom} = 313.15$ K				
2.05	313.269	137.2	1133	137.4
4.40	313.150	143.9	1151	143.9
5.80	313.207	147.7	1160	147.6
7.10	313.180	151.0	1169	151.0
8.85	313.166	155.2	1179	155.4
10.50	313.146	159.2	1188	159.3
12.35	313.210	163.4	1198	163.6
14.05	313.198	167.3	1206	167.3
$T_{nom} = 333.15$ K				
3.25	333.198	110.1	1042	110.3
4.50	333.150	114.7	1058	114.6
5.70	333.269	118.3	1072	118.4
7.00	333.214	122.3	1085	122.4
8.10	333.212	125.4	1095	125.5
9.30	333.225	128.5	1105	128.7
10.70	333.260	132.1	1116	132.2
12.20	333.173	135.6	1126	135.6

Table II. Measurements of the Viscosity of R124

Pressure P (MPa)	Temperature T (K)	Viscosity $\eta(T, P)$ (μPa·s)	Density $\rho(T_{nom}, P)$ (kg·m^{-3})	Viscosity $\eta(T_{nom}, P)$ (μPa·s)
$T_{nom} = 273.15$ K				
1.50	273.155	353.2	1424	353.1
3.35	273.142	361.8	1432	361.9
5.25	273.164	371.0	1439	370.9
7.00	273.190	378.7	1445	379.0
8.40	273.206	385.4	1449	385.6
10.60	273.183	395.7	1456	395.8
12.10	273.183	402.5	1460	402.7
13.90	273.199	410.8	1465	410.9
$T_{nom} = 293.15$ K				
2.00	293.185	278.0	1384	278.0
4.70	293.178	289.6	1396	289.7
6.70	293.226	298.1	1404	298.2
8.85	293.281	306.8	1412	307.2
10.40	293.201	313.3	1418	313.6
12.25	293.102	321.2	1425	321.1
14.50	293.153	330.0	1432	330.1
16.70	293.226	338.3	1440	338.7
$T_{nom} = 313.15$ K				
2.00	313.173	220.5	1322	220.6
3.40	313.148	226.5	1331	226.5
5.10	313.159	233.4	1340	233.6
7.40	313.191	242.6	1353	242.8
9.40	313.157	250.7	1363	250.6
11.30	313.198	257.6	1372	257.8
13.00	313.155	264.1	1380	264.1
14.85	313.148	271.0	1387	270.7
$T_{nom} = 333.15$ K				
1.60	333.191	173.9	1237	174.0
2.50	333.191	177.9	1246	178.0
3.80	333.200	183.7	1257	183.7
5.20	333.207	189.5	1269	189.5
6.60	333.166	195.3	1279	195.3
8.10	333.202	201.1	1290	201.2
9.50	333.180	206.5	1299	206.5
12.70	333.154	218.1	1318	218.1

Table III. Measurements of the Viscosity of R125

Pressure P (MPa)	Temperature T (K)	Viscosity $\eta(T, P)$ (μPa·s)	Density $\rho(T_{nom}, P)$ (kg·m^{-3})	Viscosity $\eta(T_{nom}, P)$ (μPa·s)
		$T_{nom} = 273.15$ K		
2.45	273.219	212.1	1333	212.2
4.05	273.139	219.5	1344	219.5
5.80	273.126	227.2	1355	227.1
7.30	273.158	233.2	1364	233.4
9.00	273.215	240.1	1373	240.3
10.40	273.249	245.6	1380	245.7
12.40	273.183	253.1	1391	253.2
14.45	273.155	260.4	1402	260.5
		$T_{nom} = 293.15$ K		
2.90	293.194	161.9	1239	162.1
4.40	293.194	168.9	1254	169.0
5.85	293.201	175.5	1268	175.4
7.50	293.244	182.3	1282	182.4
9.20	293.201	189.0	1295	189.2
10.90	293.196	195.3	1308	195.7
12.45	293.203	201.3	1320	201.3
14.10	293.201	207.0	1331	207.0
		$T_{nom} = 313.15$ K		
5.40	313.235	131.7	1158	132.0
6.55	313.267	137.3	1174	137.3
7.80	313.189	142.8	1191	142.9
9.20	313.178	148.6	1207	148.7
10.65	313.175	154.1	1224	154.3
12.10	313.228	159.5	1239	159.5

This equation was also used to calculate the viscosity values at nominal temperatures, shown in Tables I, II, and III. The values of all constants, as well as the maximum and the standard deviations of each fit, are listed in Table IV. The maximum deviation of all fits is 0.16%, whereas the maximum standard deviation is ±0.07%. The critical constants for R22 and R124 were taken from the respective density references discussed earlier, whereas in the case of R125 the critical temperature and pressure were taken from the work of McLinden [8].

The viscosity values at saturation conditions, presented in Table V, were obtained from Eq. (1). The saturation pressure, P_s, as well as the saturation density, ρ_s, for R22 were taken from Platzer et al. [9]; those for R124 and R125, from McLinden [8].

Table IV. Coefficients and Constants of Eq. (1)

	R22	R124	R125
C_{00} (μPa · s)	1136.26	7339.90	1730.45
C_{01} (μPa · s)	−1720.76	−23181.80	−2838.64
C_{02} (μPa · s)	631.85	25690.60	1165.34
C_{03} (μPa · s)	0	−9829.60	0
C_{10} (μPa · s)	327.56	140.01	213.31
C_{11} (μPa · s)	−805.02	−319.52	−482.26
C_{12} (μPa · s)	514.28	205.83	296.92
C_{13} (μPa · s)	0	0	0
C_{20} (μPa · s)	−81.57	−3.79	−29.92
C_{21} (μPa · s)	204.99	12.38	73.01
C_{22} (μPa · s)	−129.28	−10.21	−45.49
C_{23} (μPa · s)	0	0	0
P_c (MPa)	4.990	3.660	3.631
T_c (K)	369.30	395.65	339.40
Max. dev. (%)	0.12	0.11	0.16
σ (%)	±0.05	±0.04	±0.07

Table V. Viscosity of R22, R124, and R125 at Saturation

Fluid	Temperature T_s (K)	Pressure P_s (MPa)	Density ρ_s (kg · m^{-3})	Viscosity $\eta(T_s, P_s)$ (μPa · s)
R22	273.15	0.50	1281	210.5
	283.15	0.68	1246	190.1
	293.15	0.91	1210	170.8
	303.15	1.19	1171	152.6
	313.15	1.53	1128	135.9
	323.15	1.94	1082	120.7
	333.15	2.43	1030	107.4
R124	273.15	0.16	1436	346.7
	283.15	0.23	1404	305.7
	293.15	0.33	1371	270.6
	303.15	0.45	1337	240.5
	313.15	0.59	1301	214.5
	323.15	0.78	1263	191.8
	333.15	1.00	1222	171.3
R125	273.15	0.67	1320	203.9
	283.15	0.91	1272	177.7
	293.15	1.21	1219	154.0
	303.15	1.57	1160	132.9
	313.15	2.01	1090	114.7

4. DISCUSSION

The deviations of various investigators' experimental values of the viscosity along the saturation line from those correlated by Eq. (1) are plotted in Fig. 1, as a function of the temperature. In the case of R22 five other sets of measurements have been reported. Although four of five sets have been performed the last 5 years, the general picture is not a good one, with deviations rising up to 8%. More specifically, Diller et al. [10] measured the viscosity of R22 using a torsional-crystal viscometer with a

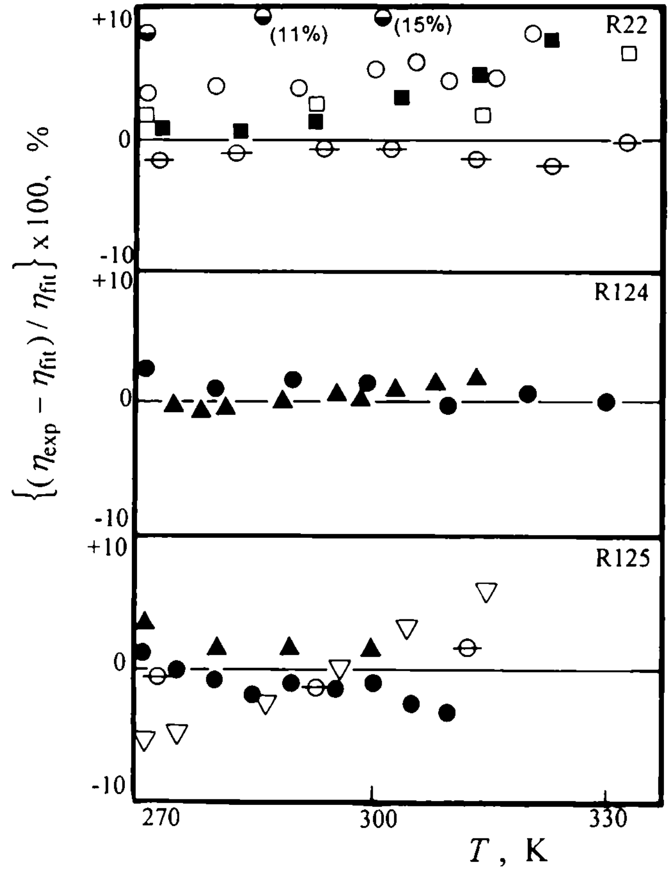

Fig. 1. Percentage deviations of the measurements of the viscosity of R22, R124 and R125 along the saturation line, from Eq. (1), as a function of the temperature: (●) Diller and Peterson [7]; (○) Diller et al. [10]; (■) Kumagai and Takahashi [11]; (□) Arnemann and Kruse [12]; (⊖) Bivens et al. [13]; (◒) Phillips and Murphy [14]; (▲) Ripple and Matar [15]; (▽) Oliveira and Wakeham [16].

±2% reproducibility checked by ethane. These values are higher than the present representation by as much as 8%. Similarly, the measurements of Kumagai and Takahashi [11], performed in a series of capillary viscometers calibrated with water and chloroform with an uncertainty of 0.5%, show deviations that rise up to 8%. Arnemann and Kruse [12] measured the viscosity of R22 with a falling-ball instrument calibrated with liquids of known viscosity. No uncertainty is quoted. Their maximum deviation from the present values rises up to 6%. The measurements, however, performed by Bivens et al. [13] in a capillary-tube viscometer, with an uncertainty of 1.2%, lie below the present representation by up to 2%, which is within the mutual uncertainty of both instruments. Finally, the R22 measurements of Phillips and Murphy [14], performed in an Ubbelohde suspended level-type viscometer, calibrated with diethyl ether, and no uncertainty mentioned, show large deviations amounting to 15%. This viscometer contained a coiled capillary, and their neglect of the effects of the curvature of the capillary upon the efflux times measured could have contributed to systematic errors in the reported viscosities, especially at the low refrigerant values.

In the case of the viscosity measurements of R124 along the saturation line, only two other sets of measurements have been reported. Here the general picture is much better. Diller and Peterson [7] measured the viscosity of R124 with a torsional crystal viscometer and, in this case, with a quoted ±3% precision. These values show deviations of up to 2.4%. The measurements of Ripple and Matar [15], performed in a calibrated-capillary viscometer with an uncertainty of ±4%, show deviations of up to 2%. The deviations of both these sets lie within the mutual uncertainty of the instruments.

The viscosity of R125 along the saturation line was reported by four investigators. A vibrating-wire instrument was employed by Oliveira and Wakeham [16] to measure the viscosity of R125 with an accuracy of ±0.5%. Their values deviate by as much as 5.7% from the ones calculated by Eq. (1), which far exceeds the limits of mutual uncertainty. This deviation, according to discussions held between the two groups, is probably attributed to the fact that the sample employed was one of the first samples produced and hence of a debatable purity. Diller and Peterson [7] measured also the viscosity of R125 with the same torsional crystal viscometer reported above, with a ±3% precision. These values are lower than the present set by up to 3.4%, which is within mutual uncertainty. It is interesting to note that although Diller's measurements of R22 disagree with the present set, in the cases of R124 and R125 the measurements are within the mutual uncertainty. Ripple and Matar [15], already mentioned in the discussion of R124, also reported viscosity measurements of R125.

As before, these values show deviations within mutual uncertainty. Finally, the measurements performed by Bivens et al. [13] in a capillary-tube viscometer, with an uncertainty of 1.2%, show deviations of ±2% from the present set, which is, as in the case of R22, within the mutual uncertainty of both instruments.

Figure 2 shows the deviations of high-pressure viscosity measurements of other investigators from the values correlated by Eq. (1), as a function of the pressure. The deviations of the high-pressure measurements of Diller et al. [10] and Diller and Peterson [7] follow the discussion of the

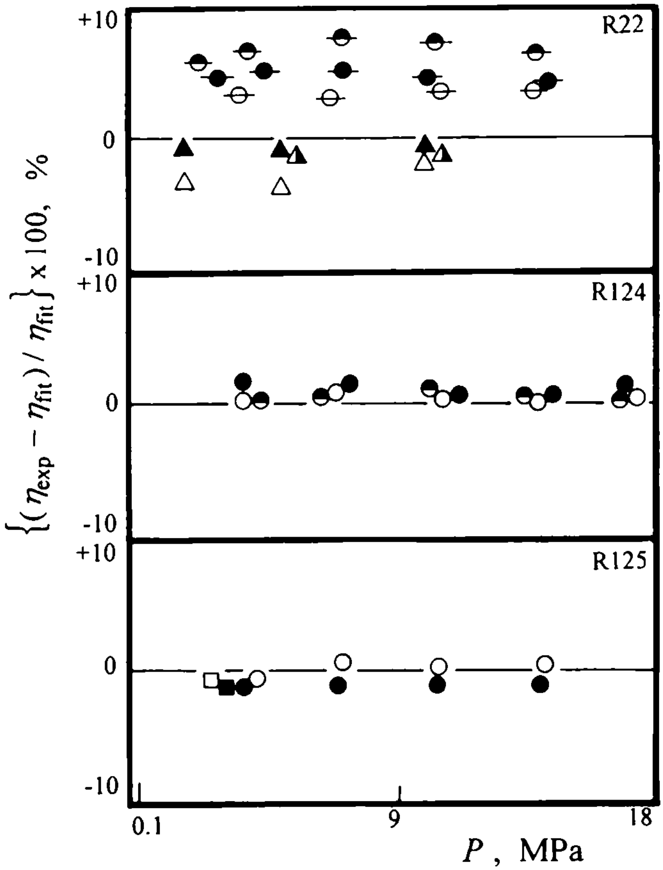

Fig. 2. Percentage deviations of the high-pressure measurements of the viscosity of R22, R124 and R125, from Eq. (1), as a function of pressure: (○) 270 K, (●) 300 K, and (◓) 320 K, Diller and Peterson [7] (⊖) 270 K, (-●-) 300 K, and (-◓-) 320 K, Diller et al. [10]; (△) 273 K, (▲) 293 K, and (◭) 313 K, Geller [17]; (□) 273 K and (■) 298 K, Wilson et al. [18].

saturation values, i.e., large deviations in the viscosity of R22 but good agreement in R124 and R125. In the case of the viscosity of R22, high-pressure measurements, performed in a capillary-flow viscometer with an uncertainty of ±3%, have also been reported by Geller [17]. These values lie below the present set by up to 3%, which is within the mutual uncertainty of the instruments. Finally, Wilson et al. [18] reported viscosity measurements for R125, performed in a water-calibrated capillary instrument, with a ±2% uncertainty. These data agree with the present set within mutual uncertainty.

5. CONCLUSION

Viscosity measurements for refrigerants R22, R124, and R125, performed in a recently modified vibrating-wire instrument, are reported. The measurements cover a temperature range of 273–333 K and a pressure range from above saturation pressure to about 17 MPa. The experimental values have an uncertainty of ±0.5%, confirmed by the measurement of the viscosity of water. Comparison of the present results with those reported by other investigators reveals, in some cases, discrepancies which cannot be accounted for. Although the general picture could be regarded as satisfactory, surely more accurate measurements of the viscosity are necessary.

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