Measurements of the Viscosity of R11, R12, R141b, and R152a in the Temperature Range 270–340 K at Pressures up to 20 MPa

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This paper reports new measurements of the liquid viscosity of R11, R12, R141b, and R152a in the temperature range 270 to 340 K and pressures up to 20 MPa. The measurements have been carried out in a vibrating-wire instrument calibrated with respect to the standard reference value of the viscosity of water. It is estimated that the uncertainty of the present viscosity data is one of 0.5%. The experimental data have been represented by polynomial functions of temperature and pressure for the purposes of interpolation. A recently developed semiempirical scheme, based on considerations of hard-sphere theory, is employed to correlate successfully the viscosity and the thermal conductivity of these refrigerants as a function of their density.

KEY WORDS: high pressure; refrigerants; R11; R12; R141b; R152a; vibratingwire technique; viscosity.

1. INTRODUCTION

Among the fluids considered as substitutes for the environmentally harmful R11 (trichlorofluormethane) and R12 (dichlorodifluoromethane) refrigerants, R141b (1,1-dichloro-1-fluoroethane) and R152a (1,1-difluoroethane) are among the possible candidates. As the time of the final decision 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 liquid viscosity for the four aforementioned refrigerants, very few

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measurements exist, and these are mostly at saturation conditions. Furthermore, the discrepancies between the investigators, up to 20% in an extreme case, far exceed the quoted uncertainties.

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 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 measurement of the viscosity of liquids. The viscometer, calibrated against water over its whole range of measurements, enables high-pressure measurements with an uncertainty of about $\pm 0.5\%$. Measurements of the viscosity of water [1], alcohols [2], and refrigerants R134a and R32[3] have already been published.

2. EXPERIMENTS

The vibrating-wire viscometer employed for the present measurements has been described in detail elsewhere $\lceil 1-3 \rceil$ and is not repeated here. 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 [1], 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 with an especially fabricated PTFE-coated Viton O-ring to protect the O-ring from chemical attack [1]. 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 were taking place. Samples were introduced after evacuation of the system and a stainless-steel 20- to 30-µm filter 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 samples of R11 and R12 were supplied by SICNG Chemical Industries of Northern Greece S.A., both at stated purities of better than 99.95%. The sample of R141b was supplied by Elf Atochem S.A. and the sample of R152a by Du Pont de Nemours International S.A., both at a stated purity of better than 99.9%.

3. RESULTS

The measurements of the viscosity of the four refrigerants were carried out along four isotherms, 273.15, 293.15, 313.15, and 333.15 K, from above saturation pressure up to 20 MPa. The density values employed in the calculation of the viscosity of R11, R12, and R152a were obtained from empirical correlations published by Blanke and Weiß [4, 5], with a quoted uncertainty of $\pm 0.01\%$. These correlations were based on their measurements and cover all the present range. In the case of R141b, the density values were obtained from an equation published by Defibaugh et al. [6] with a quoted uncertainty of $\pm 0.05\%$. This equation, however, covered the pressure range up to 6 MPa only. Since to our knowledge no other measurement-based equation or measurements of the density of R141b are available, we have extrapolated this equation to 20 MPa. It should be noted that the extrapolated density values agree with the correspondingstates values produced by Diller et al. [7] within 0.1%.

Tables I to IV show the experimental measurements of the viscosity of R11, R12, R141b, and R152a. All measurements of each refrigerant have been correlated as a function of the reduced temperature, T_r (= T/T_c , where T_c is the critical temperature) and reduced pressure, P_r (= P/P_c , where P_c is the critical pressure), for the purpose of interpolation only, by an equation of the form

$$\eta = \sum_{i=0}^{2} \sum_{j=0}^{3} C_{ij} P_{r}^{i} T_{r}^{j}$$
(1)

The values of all constants are shown in Table V. Critical constants shown in Table V were obtained from the literature—R11 [8], R12 [4], R141b [6], and R152a [32]. In the same table the maximum deviation and the standard deviation of each fit are also shown. The largest maximum deviation of all viscosity measurements is 0.11%, while the largest standard deviation is 0.05%. It should also be point out that the above equation was employed to calculate the values at nominal temperatures shown in Tables I to IV, together with the experimental values.

In Table VI, the viscosity at saturation conditions is shown for the four refrigerants. These values have been obtained by the use of Eq. (1), while values for the saturation pressure, P_s , and saturation density, ρ_s , are obtained from the respective density references discussed previously.

In Figs. 1 and 2, the deviations of other investigators' experimental values of the viscosity of the four refrigerants at saturation, from the values calculated by Eq. (1), are shown. It is worth pointing out that except for the measurements of Phillips and Murphy [9] performed in 1970, all other measurements have been performed the last 3 years. The measurements of

| Pressure P (MPa) | Temperature T (K) | Viscosity $\eta(T, P)$ $(\mu Pa \cdot s)$ | Density $\rho(T_{nom}, P)$ $(kg \cdot m^{-3})$ | Viscosity η(T _{nom} , P) (μPa·s) |
|------------------------|-------------------------|---|--|---|
| | | $T_{\rm max} = 273.15 \rm K$ | | |
| | | - nom | | |
| 1.40 | 273.146 | 531.8 | 1536 | 531.6 |
| 3.15 | 273.164 | 540.4 | 1540 | 540.7 |
| 4.50 | 273.164 | 547.4 | 1542 | 547.3 |
| 5.90 | 273.144 | 554.3 | 1545 | 554.4 |
| 7.40 | 273.272 | 561.0 | 1548 | 561.9 |
| 9.00 | 273.325 | 569.0 | 1551 | 570.0 |
| 9.90 | 273.240 | 573.9 | 1553 | 574.6 |
| 16.00 | 273.199 | 605.2 | 1565 | 605.4 |
| | | $T_{\rm nom} = 293.15 {\rm K}$ | | |
| 2.30 | 293.173 | 430.1 | 1492 | 430.2 |
| 4.60 | 293,194 | 440.1 | 1498 | 440.2 |
| 6.90 | 293.189 | 449.9 | 1503 | 450.1 |
| 9.30 | 293.203 | 460.3 | 1509 | 460.5 |
| 11.70 | 293.173 | 470.8 | 1514 | 470.8 |
| 14.20 | 293.139 | 481.4 | 1520 | 481.7 |
| 16.60 | 293.201 | 491.8 | 1525 | 492.0 |
| 18.90 | 293.221 | 501.8 | 1529 | 502.0 |
| | | $T_{\rm nom} = 313.15 {\rm K}$ | | |
| 2.15 | 313,184 | 352.1 | 1444 | 352.2 |
| 4.40 | 313.214 | 360.5 | 1451 | 360.8 |
| 6.80 | 313.201 | 369.8 | 1458 | 369.9 |
| 9.20 | 313.162 | 378.9 | 1464 | 379.1 |
| 11.45 | 313.214 | 387.7 | 1470 | 387.6 |
| 13.70 | 313.198 | 396.0 | 1476 | 396.2 |
| 16.10 | 313.189 | 405.2 | 1482 | 405.4 |
| 18.50 | 313.207 | 414.2 | 1487 | 414.5 |
| | | $T_{\rm nom} = 333.15 {\rm K}$ | | |
| 1.60 | 333.141 | 291.1 | 1392 | 291.1 |
| 4.15 | 333.202 | 299.9 | 1402 | 300.1 |
| 6.60 | 333.145 | 308.6 | 1410 | 308.7 |
| 8.55 | 333.168 | 315.8 | 1416 | 315.5 |
| 10.50 | 333.205 | 322.2 | 1422 | 322.3 |
| 12.40 | 333.109 | 329.1 | 1428 | 329.0 |
| 14.40 | 333.246 | 335.8 | 1434 | 336.0 |
| 16.40 | 333.202 | 342.9 | 1440 | 343.1 |

Table I. Measurements of the Viscosity of R11

| Pressure | Temperature | Viscosity | Density | Viscosity |
|---|-------------|---------------------------------|--------------------------|------------------------|
| P | T | $\eta(T, P)$ | $\rho(T_{\rm nom}, P)$ | $\eta(T_{\rm nom}, P)$ |
| (MPa) ———————————————————————————————————— | (K) | $(\mu Pa \cdot s)$ | (kg · m ^{- 3}) | (μPa·s) |
| | | $T_{\rm nom} = 273.15 {\rm K}$ | | |
| 1.15 | 273.242 | 252.7 | 1399 | 253.t |
| 2.65 | 273.215 | 258.2 | 1406 | 258.2 |
| 3.70 | 273.176 | 261.8 | 1410 | 261.8 |
| 5.55 | 273.142 | 267.9 | 1417 | 268.0 |
| 7.25 | 273.284 | 273.1 | 1423 | 273.5 |
| 9.25 | 273.176 | 279.8 | 1430 | 279.9 |
| 11.00 | 273.226 | 285.1 | 1435 | 285.3 |
| 12.70 | 273.199 | 290.3 | 1441 | 290.5 |
| | | $T_{\rm nom} = 293.15 {\rm K}$ | | |
| 1.80 | 293.169 | 206.3 | 1336 | 206.2 |
| 3.20 | 293.155 | 210.8 | 1343 | 210.9 |
| 5.30 | 293.153 | 217.8 | 1354 | 217.8 |
| 7.05 | 293.162 | 223.2 | 1362 | 223.3 |
| 9.10 | 293.173 | 229.7 | 1371 | 229.7 |
| 11.05 | 293.178 | 235.5 | 1379 | 235.6 |
| 13.20 | 293.173 | 241.8 | 1388 | 241.8 |
| 15.10 | 293.162 | 247.1 | 1395 | 247.2 |
| | | $T_{\rm nom} = 313.15 {\rm K}$ | | |
| 2.55 | 313.171 | 168.8 | 1267 | 168.9 |
| 4.40 | 313.189 | 175.2 | 1280 | 175.2 |
| 5.90 | 313.198 | 180.2 | 1290 | 180.2 |
| 7.30 | 313.219 | 184.6 | 1298 | 184.7 |
| 8.20 | 313.123 | 187.5 | 1304 | 187.5 |
| 9.30 | 313.187 | 190.8 | 1310 | 190.8 |
| 10.45 | 313.166 | 194.1 | 1316 | 194.3 |
| 11.65 | 313.219 | 197.6 | 1322 | 197.7 |
| | | $T_{\rm nom} = 333.15 {\rm K}$ | | |
| 3.00 | 333.221 | 137.4 | 1186 | 137.6 |
| 4.30 | 333.207 | 142.2 | 1200 | 142.4 |
| 5.40 | 333.202 | 146.3 | 1210 | 146.3 |
| 6.45 | 333.148 | 149.8 | 1220 | 149.9 |
| 7.90 | 333.276 | 154.5 | 1232 | 154.6 |
| 9.10 | 333.180 | 158.2 | 1241 | 158.4 |
| 10.70 | 333.301 | 162.9 | 1252 | 163.2 |
| 12.20 | 333.150 | 167.4 | 1262 | 167.4 |

Table II. Measurements of the Viscosity of R12

| Pressure | Temperature | Viscosity | Density | Viscosity |
|----------|-------------|----------------------------------|--------------------------|------------------------|
| Р | Т | $\eta(T, P)$ | $\rho(T_{nom}, P)$ | $\eta(T_{\rm nom}, P)$ |
| (MPa) | (K) | (µPa s) | (kg · m ^{- 3}) | (μPa·s) |
| | | $T_{\rm nom} = 273.15 {\rm K}$ | | |
| 0.85 | 273.233 | 544.8 | 1281 | 545.5 |
| 2.70 | 273.208 | 555.4 | 1284 | 555.4 |
| 4.70 | 273.192 | 565.8 | 1288 | 566.2 |
| 6.90 | 273.242 | 577.6 | 1291 | 578.0 |
| 8.60 | 273.183 | 586.5 | 1294 | 587.1 |
| 10.30 | 273.245 | 595.2 | 1297 | 596.3 |
| 12.10 | 273.251 | 605.4 | 1300 | 606.0 |
| 14.35 | 273.164 | 618.1 | 1303 | 618.1 |
| | | $T_{\rm nom} = 293.15 {\rm K}$ | | |
| 215 | 293 201 | 435.4 | 1247 | 435.9 |
| 4.30 | 293.201 | 445.1 | 1251 | 445.4 |
| 6.75 | 293.201 | 456.1 | 1256 | 456.3 |
| 9.00 | 293.194 | 466.1 | 1260 | 466.3 |
| 11.20 | 293.153 | 476.0 | 1264 | 476.1 |
| 14.10 | 293.187 | 488.9 | 1270 | 489.0 |
| 16.60 | 293.183 | 499.9 | 1274 | 500.1 |
| 18.60 | 293.267 | 508.6 | 1278 | 509.0 |
| | | $T_{\rm nom} = 313.15 \text{ K}$ | | |
| 1.80 | 313.143 | 352.3 | 1209 | 352.1 |
| 4.30 | 313.136 | 361.7 | 1214 | 361.7 |
| 7.00 | 313.189 | 372.0 | 1220 | 372.0 |
| 9.35 | 313.178 | 381.0 | 1225 | 380.9 |
| 11.40 | 313.166 | 388.6 | 1229 | 388.7 |
| 13.70 | 313.159 | 397.5 | 1234 | 397.5 |
| 16.40 | 313.168 | 407.5 | 1239 | 407.8 |
| 18.90 | 313.130 | 417.3 | 1243 | 417.3 |
| | | $T_{\rm nom} = 333.15 \text{ K}$ | | |
| 1.20 | 333.198 | 288.0 | 1167 | 288.1 |
| 3.05 | 333.159 | 294.6 | 1172 | 294.5 |
| 5.20 | 333.198 | 301.7 | 1178 | 302.0 |
| 6.90 | 333.102 | 308.1 | 1182 | 307.9 |
| 8.80 | 333.230 | 314.3 | 1186 | 314.6 |
| 11.65 | 333.166 | 324.5 | 1193 | 324.5 |
| 14.10 | 333.102 | 333.1 | 1198 | 333.0 |
| 16.45 | 333.244 | 341.2 | 1202 | 341.2 |

Table III. Measurements of the Viscosity of R141b

| Pressure | Temperature | Viscosity | Density | Viscosity |
|----------|-------------|----------------------------------|---------------------------|------------------------|
| P | T | $\eta(T, P)$ | $\rho(T_{\text{nom}}, P)$ | $\eta(T_{\rm nom}, P)$ |
| (MPa) | (K) | $(\mu Pa \cdot s)$ | (kg m) | $(\mu Pa \cdot s)$ |
| | | $T_{\rm nom} = 273.15 {\rm K}$ | | |
| 1.45 | 273.226 | 218.6 | 962 | 218.9 |
| 3.25 | 273.206 | 223.2 | 966 | 223.4 |
| 4.85 | 273.178 | 227.3 | 970 | 227.3 |
| 6.60 | 273.135 | 231.6 | 974 | 231.6 |
| 8.30 | 273.176 | 235.7 | 978 | 235.7 |
| 10.00 | 273.203 | 239.6 | 982 | 239.8 |
| 11.75 | 273.229 | 243.6 | 985 | 243.9 |
| 13.30 | 273.178 | 247.6 | 988 | 247.5 |
| | | $T_{\rm nom} = 293.15 \text{ K}$ | | |
| 1.65 | 293.231 | 174.9 | 916 | 175.1 |
| 4.15 | 293.267 | 181.0 | 924 | 181.0 |
| 6.40 | 293.217 | 186.1 | 930 | 186.2 |
| 8.45 | 293.208 | 190.7 | 936 | 190.9 |
| 10.85 | 293.180 | 196.1 | 943 | 196.2 |
| 13.10 | 293.208 | 200.9 | 948 | 201.0 |
| 15.40 | 293.153 | 205.9 | 954 | 205.9 |
| 17.70 | 293.189 | 210.5 | 959 | 210.6 |
| | | $T_{\rm nom} = 313.15 {\rm K}$ | | |
| 1.95 | 313.239 | 140.8 | 865 | 141.0 |
| 3.80 | 313.230 | 145.4 | 873 | 145.5 |
| 5.55 | 313.162 | 149.7 | 880 | 149.6 |
| 7.20 | 313.262 | 153.0 | 886 | 153.4 |
| 9.50 | 313.205 | 158.3 | 895 | 158.4 |
| 11.30 | 313.239 | 162.1 | 901 | 162.1 |
| 13.15 | 313.143 | 165.9 | 906 | 165.9 |
| 15.00 | 313.171 | 169.6 | 912 | 169.4 |
| | | $T_{\rm nom} = 333.15 \text{ K}$ | | |
| 2.40 | 333.260 | 114.9 | 806 | 114.9 |
| 3.50 | 333.198 | 117.8 | 814 | 117.9 |
| 5.00 | 333.230 | 121.6 | 823 | 121.7 |
| 6.50 | 333.207 | 125.4 | 831 | 125.4 |
| 8.00 | 333.157 | 128.8 | 839 | 128.8 |
| 9.65 | 333.257 | 132.4 | 847 | 132.4 |
| 11.40 | 333.168 | 135.9 | 854 | 136.0 |
| 13.20 | 333.189 | 139.2 | 862 | 139.3 |

Table IV. Measurements of the Viscosity of R152a

| Coefficients C _" (µPa·s) | R11 | R12 | R141b | R152a |
|--|------------|-----------|------------|---------------|
| C_{00} | 10,148.6 | 3,162.34 | 12,673.8 | 2,817.96 |
| $C_{\rm ot}$ | - 37,890.8 | -8,729.07 | - 49,402.6 | - 7.662.37 |
| C_{02} | 49,761.7 | 8,717.06 | 67,183.0 | 7,401.25 |
| C_{03} | - 22,489.2 | -3,102.53 | - 31,283.9 | - 2,504.58 |
| C_{10} | 160.22 | 119.18 | 205.64 | 125.52 |
| C_{11} | - 389.49 | - 280.89 | - 529.35 | - 302.45 |
| C_{12} | 261.29 | 188.02 | 366.56 | 199.69 |
| C_{13} | 0 | 0 | 0 | 0 |
| C_{20} | 0 | - 16.25 | 0 | -16.82 |
| C_{21} | 0 | 44.01 | 0 | 46.49 |
| C 22 | 0 | - 30.44 | 0 | - 32.46 |
| C_{23} | 0 | 0 | 0 | 0 |
| $P_{\rm c}$ (MPa) | 4.4026 | 4.129 | 4.230 | 4.517 |
| $T_{\rm c}$ (K) | 471.15 | 384.981 | 477.26 | 386.41 |
| Max. dev. (%) | 0.11 | 0.11 | 0.08 | 0.11 |
| σ (%) | ± 0.03 | ± 0.03 | ± 0.04 | <u>±</u> 0.05 |

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



Fig. 1. Percentage deviations of the viscosity measurements of R11 and R12 along the saturation line, from Eq. (1). (\ominus) Ref. 9; (\Box) Ref. 11.

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Phillips and Murphy [9] were performed in an Ubbelohde suspended level-type viscometer calibrated with diethyl ether. The uncertainty of the measurements is not quoted. The deviations from the present values rise to 20%. The viscometer employed by Phillips and Murphy contained a coiled capillary. Their neglect of the effects of the curvature of the capillary upon the efflux times measured in the viscometer could have contributed to systematic errors in the reported viscosities, especially at low viscosities. The viscosity measurements of R152a of van der Gulik [10] were carried

| | Temperature | Pressure | Density | Viscosity |
|-------------|-------------|----------|-----------------------------------|---------------------------------|
| Fluid | (K) | (MPa) | $(\text{kg} \cdot \text{m}^{-3})$ | $\eta(I_{s}, P_{s})$ (µPa·s) |
| | | | | |
| RII | 273.15 | 0.04 | 1533 | 524.8 |
| | 283.15 | 0.06 | 1510 | 468.5 |
| | 293.15 | 0.09 | 1487 | 420.7 |
| | 303.15 | 0.13 | 1463 | 379.8 |
| | 313.15 | 0.18 | 1438 | 344.7 |
| | 323.15 | 0.24 | 1413 | 314.1 |
| | 333.15 | 0.31 | 1387 | 286.6 |
| D1 2 | 777 16 | 0.21 | 1206 | 250.1 |
| R12 | 273.15 | 0.31 | 1390 | 230.1 |
| | 283.15 | 0.43 | 1303 | 224.0 |
| | 293.15 | 0.37 | 1329 | 202.0 |
| | 303.15 | 0.73 | 1293 | 161.0 |
| | 313.15 | 0.90 | 1234 | 103.3 146 P |
| | 323.15 | 1.22 | 1213 | 140.8 |
| | 333.15 | 1.53 | 1108 | 131.9 |
| R141b | 273.15 | 0.03 | 1280 | 541.0 |
| | 283.15 | 0.04 | 1262 | 478.8 |
| | 293.15 | 0.07 | 1243 | 426.6 |
| | 303.15 | 0.09 | 1224 | 382.9 |
| | 313.15 | 0.13 | 1205 | 345.8 |
| | 323.15 | 0.18 | 1185 | 313.6 |
| | 333.15 | 0.25 | 1164 | 284.7 |
| P152a | 273.15 | 0.26 | 959 | 215.9 |
| RTJ2a | 273.15 | 0.20 | 936 | 197.8 |
| | 203.15 | 0.57 | 912 | 172.3 |
| | 293.15 | 0.51 | 887 | 154.3 |
| | 312 15 | 0.09 | 860 | 138.4 |
| | 212.12 | 1 1 2 | 821 | 174.6 |
| | 222.15 | 1.10 | 800 | 1125 |
| | 535.15 | 1.50 | 000 | 112.3 |

Table VI. Viscosity of R11, R12, R141b, and R152a at Saturation



Fig. 2. Percentage deviations of the viscosity measurements of R141b and R152a along the saturation line, from Eq. (1). (\oplus) Ref. 9; (\blacktriangle) Ref. 10; ((1)) Ref. 11; (\blacksquare) Ref. 12; (\bigcirc) Ref. 13; (\bigtriangledown) Ref. 14.

out in a calibrated vibrating-wire instrument and at a quoted uncertainty of about +1%. The maximum deviation of these values from those calculated by Eq. (1) is -2.9%. The measurements of Kumagai and Takahashi [11, 12] were carried out in a capillary viscometer calibrated with water and chloroform with a quoted uncertainty of about 1%. Except in the case of the viscosity of R12, in all other three refrigerants these values agree with the present set within the mutual uncertainties. In the case of the viscosity of R12 a different slope was observed, with deviations rising to 10% at the highest temperature. Since no other investigator has measured the viscosity of R12 recently, no other comment can be made except that this difference between the two sets is alarming. Diller et al. [13] measured the viscosity of R141b employing a torsional-crystal viscometer. Its reproducibility was checked by ethane to 2%. The uncertainty of the measurements is not quoted. This set of measurements deviate from the present values up to 5%. Finally, Arnemann and Kruse [14] employed a falling-body viscometer calibrated with various oils and water, to measure the viscosity of R152a. The uncertainty of the measurements is not quoted. This set shows deviations of up to 2.5% from the present values.

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In the case of high-pressure measurements of the viscosity, to our knowledge the only available set of measurements is that of Diller et al. [13] at 300 K. This set seems to be shifted in pressure from the present values by about 3%.

4. DISCUSSION

Whereas Eq. (1) is suitable for interpolation, it has little or no value for extrapolation. For such purposes it has been shown [15–17] that a correlation in terms of the molar volume, V, is much more suitable. In the case of the viscosity and the thermal conductivity, this scheme, described in detail elsewhere [15], suggests that the dimensionless viscosity, η^* , and thermal conductivity, λ^* , defined by the equations

$$\eta^* = 6.035 \times 10^8 \left[\frac{1}{MRT} \right]^{1/2} \eta V^{2/3}$$
 (2)

$$\lambda^* = 1.936 \times 10^7 \left[\frac{M}{RT}\right]^{1/2} \lambda V^{2/3} \tag{3}$$

are functions of the reduced molar volume $V_r = (V/V_o)$, where V_o is a characteristic molar volume of the liquid, weakly dependent on temperature. In the above two equations (all quantities in SI units), M represents the molar mass, R the universal gas constant, and T the absolute temperature. According to this scheme [15] the aformentioned functions were calculated to be

$$\log\left[\frac{\eta^{*}}{R_{\eta}}\right] = 1.0945 - 9.26324V_{r}^{-1} + 71.0385V_{r}^{-2} - 301.9012V_{r}^{-3} + 797.69V_{r}^{-4} - 1221.977V_{r}^{-5} + 987.5574V_{r}^{-6} - 319.4636V_{r}^{-7}$$
(4)

$$\log\left[\frac{\lambda^*}{R_{\lambda}}\right] = 1.0655 - 3.538 V_r^{-1} + 12.120 V_r^{-2} - 12.469 V_r^{-3} + 4.562 V_r^{-4}$$
(5)

where R_n and R_{λ} are constants accounting for deviations from the behavior of smooth hard spheres [15]. In the case of pure *n*-alkanes [15], aromatic hydrocarbons [16], alcohols [17], and some refrigerants [18], experimental measurements were used to calculate the temperature dependence of the characteristic molar volumes and the values of the above constants. It was thus shown [15–18] that this scheme can correlate and predict the viscosity, the thermal conductivity, and the diffusion coefficient with a 5% uncertainty over the temperature range 100–400 K and up to 600 MPa pressure.

| | R11 | R12 | R141b | R152a |
|--|-----------|-----------|------------|------------|
| R, | 1.200 | 0.980 | 1.400 | 1.014 |
| R, | 1.640 | 1.510 | 1.585 | 1.470 |
| $a_{\rm n}$ (10 ⁵ m ³ · mol ⁻¹) | 6.751667 | 10.04425 | 16.56554 | 15.55339 |
| $a_1 (10^7 \text{m}^3 \cdot \text{mol}^{-1} \cdot \text{K}^{-1})$ | -0.507283 | - 3.47825 | - 10.19797 | - 11.48044 |
| a_{2} (10 ¹⁰ m ³ mol ⁻¹ K ⁻²) | 0.272575 | 5.72568 | 32.81475 | 37.49422 |
| $a_3 (10^{12} \text{m}^3 \cdot \text{mol}^{-1} \cdot \text{K}^{-3})$ | 0 | 0 | - 3.69437 | - 4.09760 |
| · · · · · · · · · · · · · · · · · · · | | | | |

Table VII. Constants R_n and R_k and Coefficients a_i of Eq. (6)

All the aforementioned viscosity measurements together with other existing thermal conductivity measurements were consequently used to calculate the temperature dependence of the characteristic molar volumes, V_{o} (m³ mol⁻¹) and the values of the constants R_{η} and R_{λ} for each liquid. The characteristic molar volume, V_{o} , was subsequently fitted to a cubic expansion of the absolute temperature as

$$V_{0} = \sum_{i=0}^{3} a_{i} T^{i}$$
 (6)

| | | D (| Temp. | Press. | No. | Devia | ition |
|--------|---------------|-------------|---------|---------------|------------|-------|-------|
| Liquid | author | Rel. No. | (K) | max. (MPa) | oi data | 5-10% | >10% |
| R11 | Present work | | 273-334 | 20 | 39 | _ | |
| | Kumagai | 11 | 273-354 | Sat. | 9 | _ | _ |
| | Phillips | 9 | 254-352 | Sat. | 7 | 1 | — |
| R12 | Present work | | 273-334 | 16 | 39 | _ | _ |
| | Kumagai | 11 | 273-344 | Sat. | 8 | 8 | _ |
| R141b | Present work | | 273-334 | 19 | 39 | _ | _ |
| | Diller | 13 | 250-320 | 30 | 27 | _ | |
| | Kumagai | 12 | 273-354 | Sat. | 9 | — | — |
| R152a | Present work | | 273-334 | 18 | 39 | _ | _ |
| | Kumagai | 11 | 273-344 | Sat. | 8 | _ | |
| | Arnemann | 14 | 253-333 | Sat. | 8 | | |
| | van der Gulik | 10 | 253-373 | Sat. | 33 | | _ |
| | Phillips | 9 | 259-319 | Sat. | 5 | 1 | |
| Total | | | | | 270 | 10 | 0 |

 Table VIII.
 Comparison of Experimental Viscosity Values with those

 Calculated from Eqs. (2)-(6)

The coefficients a_i and the constants R_η and R_λ for each liquid are shown in Table VII. The above Eqs. (2)-(6) form a consistent set of equations for the correlation and prediction of the viscosity and thermal conductivity of R11, R12, R141b, and R152a.

In Table VIII a comparison of the experimental viscosity values with those calculated from Eqs. (2)-(6) is presented. Only the R12 viscosity measurements of Phillips and Murphy [9] have not been included in the analysis for the reasons discussed in the previous section. For consistency purposes, for R141b and R152a the density values used in the analysis were the ones discussed in this work. In the case of R11 and R12 and to include a wider range of temperatures and pressure, density values were obtained from Platzer et al. [8]. The ranges of temperature and pressure shown in the table are restricted to the available density and saturation pressure

| | . | | Temp. | Press. | No. | Devia | ition |
|--------|-----------|-------------|---------|--------|------|-------|-------|
| Liquid | author | Rel. No. | (K) | (MPa) | data | 5-10% | >10% |
| R11 | Assael | | 253-334 | 21 | 35 | | |
| | Kitazawa | 20 | 232-323 | 21 | 11 | _ | _ |
| | Yata | 21 | 233-397 | Sat. | 9 | 2 | 1 |
| | Takizawa | 25 | 233-274 | Sat. | 5 | _ | |
| | Tauscher | 26 | 233-349 | Sat. | 12 | 1 | — |
| R12 | Assael | 19 | 253-334 | 23 | 40 | _ | _ |
| | Yata | 21 | 264-361 | Sat. | 9 | 6 | 1 |
| | Venart | 22 | 300-380 | 21 | 48 | 5 | _ |
| | Geller | 23 | 250-374 | 60 | 39 | 4 | _ |
| | Tauscher | 26 | 253-294 | Sat. | 5 | 1 | — |
| R141b | Gurova | 29 | 270-300 | Sat. | 7 | _ | |
| | Papadaki | 31 | 273-290 | Sat. | 3 | _ | |
| | Yata | 28 | 273-354 | 21 | 15 | 7 | _ |
| R152a | Assael | 18 | 253-334 | 23 | 37 | 1 | _ |
| | Kesselman | 24 | 253-334 | Sat. | 9 | _ | _ |
| | Tauscher | 26 | 253-299 | Sat. | 5 | _ | |
| | Gross | 27 | 253-314 | 7 | 24 | 1 | |
| | Yata | 28 | 265-343 | 31 | 20 | _ | |
| | Kim | 30 | 273-324 | 21 | 20 | 6 | |
| Total | | | | | 353 | 34 | 2 |

 Table IX. Comparison of Experimental Thermal Conductivity Values with Those

 Calculated from Eqs. (2)-(6)

data. A total of 270 experimental measurements was considered. Only 10 (3.7%) of them were found to deviate more than 5% from the scheme, and none more than 10%.

Finally, in Table IX a comparison of the experimental thermal conductivity values with those calculated from Eqs. (2)-(6) is presented. A total of 353 experimental measurements was considered. Of these measurements, 34 (9.6%) were found to deviate more than 5% from the scheme, and only 2 were found to deviate more than 10%.

Considering the fact that the experimental measurements shown cover a temperature range 250–400 K and a pressure range from saturation to 30 MPa, the deviations presented are regarded as quite satisfactory.

5. CONCLUSION

New measurements of the viscosity of R11, R12, R141b, and R152a have been presented. The measurements were performed in a recently modified vibrating-wire instrument and cover a temperature range from 270 to 340 K at pressures up to 20 MPa. The overall uncertainty in the reported data is $\pm 0.5\%$, an estimate confirmed by the measurement of the viscosity of water. From the discussion of the results, it seems that still more accurate measurements are required.

Based on the present measurements and other thermal conductivity measurements, a recently developed semiempirical scheme is successfully employed to correlate and predict the viscosity and thermal conductivity of these four refrigerants over the temperature range 250–400 K and up to 50 MPa pressure, with an uncertainty better than 5%.

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