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# Viscosity of the Saturated Liquid Phase of Three Fluorinated Ethanes: R152a, R143a, and R125

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Data are reported for the viscosity of three saturated liquids over a temperature range from 255 K to 323 K. The liquids studied are the fluorinated compounds 1,1-difluoroethane (R152a), 1,1,1−trifluoroethane (R143a), and pentafluoroethane (R125). A capillary viscometer constructed of stainless steel and sapphire was used to obtain the data. The viscosity measurements have an expanded uncertainty of 2.4%. A free volume model of viscosity was used to correlate the data.

## Introduction

This paper reports the viscosity of the saturated liquid phase of three fluorinated ethanes proposed as refrigerants. The chemical names, and the ASHRAE standard designations in parentheses, of the measured compounds are 1,1-difluoroethane (R152a), 1,1,1-trifluoroethane (R143a), and pentafluoroethane (R125).

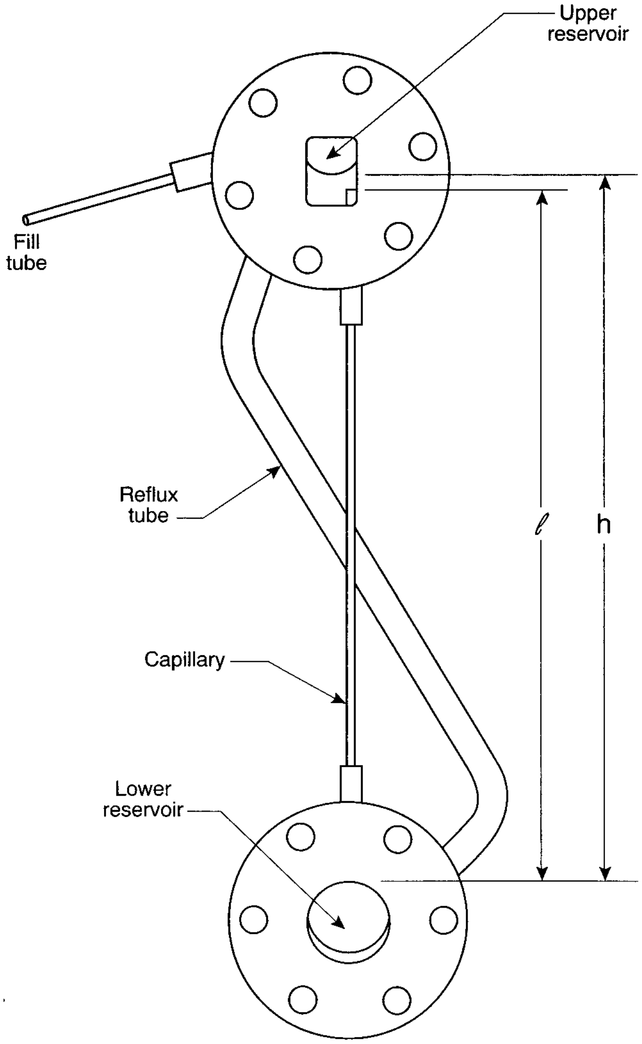

**Figure 1.** Schematic of the viscometer. The upper and lower fluid reservoirs are shown without the sapphire windows and the flanges that hold the windows in place. Holes bored in the upper reservoir body connect the reflux tube to the top of the upper reservoir and to the fill line.

## Experimental Methods and Uncertainties

The viscometer used to acquire the data, shown schematically in Figure 1, was modeled on a previous viscometer (Ripple, 1992). It is similar to an Ubbelohde capillary viscometer in design and was constructed out of stainless steel with sapphire windows to withstand the high vapor pressures of the refrigerants. To eliminate a systematic effect that depended on the curvature of the capillary in the previous viscometer (White, 1929), the present viscometer was built with a straight capillary of length $l = 14.8$ cm and bore diameter $d = 0.236$ mm. The measured data consist of data pairs of temperature, $T$, and the rate of fall, $\dot{h}$, of the liquid−vapor meniscus in the upper reservoir of the viscometer. The viscosity, $\eta$, is related to $\dot{h}$ through the working equation (Hardy, 1962)

$$\frac{\eta}{\rho_l - \rho_v} = \frac{C}{\dot{h}} - m\frac{\rho_l}{(\rho_l - \rho_v)}\frac{A}{8\pi l}\dot{h} \qquad (1)$$

where $C$ is a calibration constant, $\rho_l$ and $\rho_v$ are densities of the liquid and vapor phases, and $A = 1.20$ cm$^2$ is the cross section of the upper reservoir. The term proportional to $m$ is a correction for inertial or kinetic energy effects. Simple theories give $m = 1$, but for glass viscometers, an empirical expression of $m = 0.037\mathrm{Re}^{1/2}$ has been used (Cannon *et al.*, 1963), where $\mathrm{Re} = (4A\dot{h}\rho_l)/(\pi\eta d)$ is the Reynolds number. We have chosen to use the empirical expression for the data in this paper. The maximum difference between these two models is equivalent to a 0.6% difference in $\eta$ for the data reported here. Because the inertial energy term is less than 1% of the term $C/\dot{h}$ for all data in this paper, $A$ and $l$ need not be known to high accuracy. The constant $C = (6.415 \pm 0.033) \times 10^{-12}$ m$^3$/s$^2$ in eq 1 was determined by calibrating the viscometer with toluene of 99.9% purity at temperatures between 24 °C and 40 °C (Gonçalves *et al.*, 1987). Toluene was chosen because its viscosity is well characterized in the literature; water could not be used as a calibration fluid because of its large capillary length (Rowlinson and Widom, 1982). Calibrations performed before and after the data measurements in this paper were obtained gave values of $C$ that differed by less than 0.3%, consistent with the short-term statistical

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**Table 1. Expanded Uncertainties (2*u*) of the Viscosity Measurements**

| | |
|---|---|
| type A uncertainty | 1.0% |
| type B uncertainties: | |
| pressure head variations | 0.9% |
| drainage films | 1.5% |
| calibration with toluene | 0.5% |
| inertial energy correction | 1.2% |
| total type B | 2.2% |
| total expanded uncertainty, *U* | 2.4% |

uncertainty of the measurements. The purity of the samples was determined by gas chromatography to be 99.7 mol % for R152a, 99.9% for R125, and 99.8% for R143a. All samples were degassed by twice freezing the sample in liquid nitrogen and pumping off any volatile gases remaining. The viscometer was filled by first evacuating the viscometer and the transfer lines and then distilling the fluorinated ethanes from a transfer vessel at room temperature into the viscometer. The fluorinated ethanes condensed in the lower reservoir of the viscometer, which was cooled to a temperature in the range 0 °C to −40 °C.

The uncertainty of the measurements is expressed in the terminology recommended by the ISO (ISO, 1993). Contributions to the overall uncertainty are termed either type A or type B uncertainties. Type A uncertainties are derived from statistical analysis of the data; type B uncertainties are evaluated through other experiments, calculations, or any other means independent of the data. In many, but certainly not all, cases, type A uncertainties are equivalent to “random” uncertainties and type B uncertainties are equivalent to “systematic” uncertainties. The expanded uncertainty of the viscosity measurements is $U = 2u_c = 2.4\%$, where $u_c$ is the combined standard uncertainty including both type A and type B uncertainties. The various contributions to $U$ are listed in Table 1.

As a check on the reproducibility of the data, two separate samples were used to obtain data for both R143a and R152a. The average statistical deviation between runs is $u = 0.5\%$, which we take as the type A standard uncertainty. As an additional check, a set of measurements for each fluid was obtained covering the full range of temperatures and with the first and last data points differing by less than 10 °C. In the fitting of the data, no shifts between the first and last data points were seen for any of the fluids.

The type B uncertainties are substantially larger than the type A uncertainties. The combined type B uncertainty can be thought of as the uncertainty in our modeling of the viscometer by the working equation, eq 1. There is a small uncertainty resulting from the limited accuracy of the known viscosity of the calibration liquid, toluene. More substantial uncertainties are introduced in using the viscometer for liquids with surface tensions and viscosities that differ substantially from the values of toluene in the temperature range from 24 °C to 40 °C. Unfortunately, there are no reliable calibration liquids with properties similar to those of the measured compounds. Uncertainty contributions resulting from the use of the viscometer for liquids other than toluene are discussed in the next three paragraphs.

As mentioned above (Cannon *et al.*, 1963), there is a discrepancy between two common models for the inertial energy term in eq 1. The maximum magnitude of this discrepancy for the measurements reported in this paper was taken as the standard uncertainty for this effect.

When the viscometer is inverted and then turned upright, a film of liquid is left on the surface of the upper reservoir adjacent to the vapor phase (Landau and Levich, 1942). The thickness of this film and the rate at which it drains into the reservoir will depend on the surface tension, the density, and the viscosity of the liquid. The drainage from this film will affect the measured flow times of the liquid from the upper reservoir. There is an uncertainty in the flow rate through the capillary for the measured refrigerant compounds because the measured liquids have substantially different properties than the toluene used as a calibration liquid. Calculations of expected film thicknesses for toluene and for typical refrigerant compounds were performed. Experimental measurements also were made of the dependence of the measured viscosity value on the value of $h$ at which eq 1 was applied. These results were used to determine the uncertainty due to drainage films.

Differences in surface tension between the calibration liquid and the measured liquids also will introduce an uncertainty in the model for liquid flow through the viscometer. The liquid−vapor interface in the upper reservoir and the interface at the capillary exit in the lower reservoir are both slightly curved. The curvature of the interface is proportional to a pressure head across the interface (Rowlinson and Widom, 1982). Because eq 1 does not include this pressure term, there is a corresponding uncertainty in the viscosity determination. A study of the effects of different shapes of the capillary exit (Gonçalves *et al.*, 1991) demonstrated that the resultant error in the determination of viscosity is minimized with a capillary opening abruptly into the lower reservoir, as in the present viscometer. Optical measurements of the curvature of the liquid−vapor interface at the capillary exit and theoretical models of the curvature of the interface in the upper reservoir were used to determine possible variations in the pressure head caused by variations in the liquid surface tension.

To find $\eta$ from eq 1, the densities of the liquid and vapor phases must be measured or estimated. As reported in two separate papers, liquid densities were measured independently (Defibaugh and Morrison, 1992; Defibaugh and Moldover, 1997). Vapor densities were calculated by Weber (1994) from measured saturation vapor pressures and estimated virial coefficients. The standard uncertainty for $\rho_l - \rho_v$ is $u = 0.1\%$, which is negligible.

The viscometer was maintained at a constant temperature by immersion in a stirred liquid bath. The standard uncertainty of the temperature values is $u = 0.01$ °C, which includes the uncertainty of the thermometer used to determine the temperature of the stirred bath and uncertainties for the gradients and fluctuations of the bath.

Evaporation and condensation of the test fluid within the viscometer caused by temperature gradients can affect the observed rate of fall of the meniscus in the upper reservoir of the viscometer. To verify that gradients did not affect the data, tests were performed with wide variations in the bath heat load at a fixed temperature, with different intensities of illumination on the viscometer, and with variations in the position of the viscometer within the bath. No changes in the results were observed within the statistical uncertainty of the viscosity values. This negative result is consistent with our model of thermal and mass flows within the viscometer and our measurements of the bath gradients.

## Results

Tables 2−4 list all of the results. The simple functional form

$$\frac{1}{\eta} = B\left(\frac{1}{\rho_l} - \frac{1}{\rho_0}\right) \quad (2)$$

**Table 2. Saturated Liquid Viscosity and Density of the Liquid and Vapor Phases for R125**

| $T$/K | $\rho_l$/kg·m$^{-3}$ [a] | $\rho_v$/kg·m$^{-3}$ [b] | $\eta$/mPa·s |
|---|---|---|---|
| 255.95 | 1395 | 24 | 0.2592 |
| 263.08 | 1365 | 31 | 0.2339 |
| 265.20 | 1356 | 33 | 0.2267 |
| 273.19 | 1320 | 43 | 0.2021 |
| 283.12 | 1272 | 58 | 0.1751 |
| 293.79 | 1215 | 81 | 0.1493 |
| 298.17 | 1190 | 92 | 0.1398 |
| 303.07 | 1160 | 107 | 0.1287 |

[a] From Defibaugh and Morrison, 1992. [b] From Weber, 1994.

**Table 3. Saturated Liquid Viscosity and Density of the Liquid and Vapor Phases for R143a**

| $T$/K | $\rho_l$/kg·m$^{-3}$ [a] | $\rho_v$/kg·m$^{-3}$ [b] | $\eta$/mPa·s |
|---|---|---|---|
| 255.62 | 1083 | 16 | 0.1966 |
| 260.67 | 1067 | 18 | 0.1845 |
| 267.38 | 1046 | 23 | 0.1697 |
| 273.74 | 1024 | 28 | 0.1566 |
| 285.58 | 982 | 41 | 0.1349 |
| 290.14 | 964 | 46 | 0.1274 |
| 295.07 | 944 | 54 | 0.1190 |
| 302.64 | 912 | 67 | 0.1080 |
| 306.87 | 892 | 75 | 0.1017 |
| 308.43 | 885 | 78 | 0.0992 |

[a] From Defibaugh and Moldover, 1996. [b] From Weber, 1994.

**Table 4. Saturated Liquid Viscosity and Density of the Liquid and Vapor Phases for R152a**

| $T$/K | $\rho_l$/kg·m$^{-3}$ [a] | $\rho_v$/kg·m$^{-3}$ [b] | $\eta$/mPa·s |
|---|---|---|---|
| 254.71 | 1000 | 4 | 0.2630 |
| 258.04 | 993 | 5 | 0.2530 |
| 263.27 | 982 | 6 | 0.2366 |
| 272.24 | 962 | 8 | 0.2124 |
| 282.94 | 937 | 12 | 0.1893 |
| 289.46 | 921 | 14 | 0.1749 |
| 300.98 | 892 | 20 | 0.1541 |
| 313.13 | 860 | 28 | 0.1347 |
| 313.20 | 859 | 29 | 0.1358 |
| 322.27 | 833 | 35 | 0.1214 |
| 323.00 | 831 | 35 | 0.1211 |

[a] From Defibaugh and Moldover, 1996. [b] From Weber, 1994.

**Table 5. Values of the Fitting Parameters for the Best Fit of Eq 2 to the Data[a]**

| liquid | $B$/10$^7$ s·m$^{-2}$ | $\rho_0$/kg·m$^{-3}$ | % deviation |
|---|---|---|---|
| R125 | 2.6816 | 1745.4 | 0.11 |
| R143a | 2.4101 | 1404.8 | 0.23 |
| R152a | 2.2028 | 1208.9 | 0.38 |

[a] The column labeled deviation gives the sum of root-mean-square deviations of the data from the fit divided by the degrees of freedom of the fit, as a percentage of viscosity.

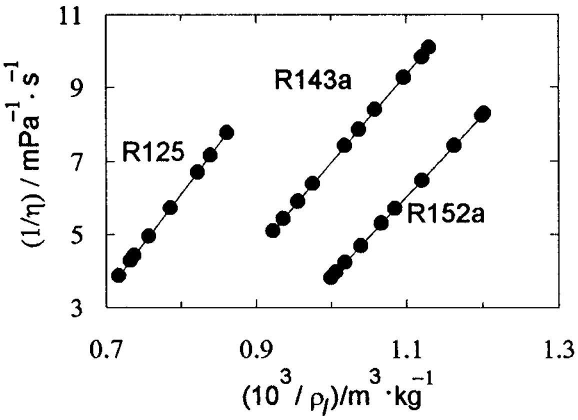

**Figure 2.** Reciprocal of the viscosity as a function of the reciprocal of the liquid density for three liquid fluorinated compounds at the saturated vapor pressure. The lines show the parameterization of eq 2, with parameter values as listed in Table 5.

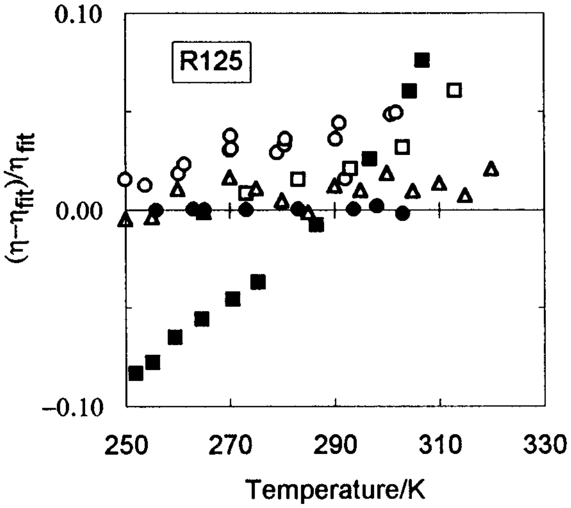

**Figure 3.** Deviations from the fit in Table 5 for the liquid viscosity of R125. The key for the data is as follows: ● for the present work, □ for the data of Assael and Polimatidou (1994), △ for the data of Diller and Peterson (1993), ○ for the data of Ripple and Matar (1993), and ■ for the data of Oliveira and Wakeham (1993).

has been fitted to the data values for $\eta$ and $T$, where $B$ and $\rho_0$ are fitted constants. The density of the liquid phase is a function of temperature along the saturation boundary, and this gives the temperature dependence of $\eta$. Diller *et al.* (1993) have demonstrated that for pressures below a few megapascals, eq 2 describes compressed liquids as well as saturated liquids. Table 5 lists the parameters of the best-fit curves shown in the figures as well as the standard deviation of the data from the curves. The reciprocal of the viscosity is plotted versus the reciprocal of the density in Figure 2. Deviations of the data and selected data from the literature from the best-fit curves are shown in Figures 3−5.

Over the range of temperature values for the present work, the results for R125 are consistent within the mutual uncertainty with results obtained by Diller and Peterson (1993), Assael and Polimatidou (1994), and Ripple and Matar (1993). All four of these sets of data are inconsistent with the results of Oliveira and Wakeham (1993). The paper by Assael and Polimatidou (1994) attributes this discrepancy to the use of an impure sample by Oliveira and Wakeham.

For R143a, the viscosity values of Kumagai and Takahashi (1991) are 3% to 8% higher than the values from the present work, with the largest deviations corresponding to the highest measured temperatures. Kumagai and Takahashi used a working equation for their capillary viscometer that assumed that the flow rate was approximately proportional to $\rho_l/\eta$. However, the gravitational force driving the flow is in fact proportional to $\rho_l - \rho_v$. To correct for this effect, we have corrected the viscosity values given by Kumagai and Takahashi by multiplying the viscosity by a factor of $(\rho_l - \rho_v)/\rho_l$. There is a 0.5% uncertainty in this correction because the vapor phase density of the fluid used for calibrating the viscometer was not reported. Both the corrected and uncorrected values are plotted in Figure 4. The corrected values agree with the present values to better than 1%, well within the mutual uncertainty of 3%.

For R152a, deviations of values from the literature for the saturated liquid viscosity from the values of the present

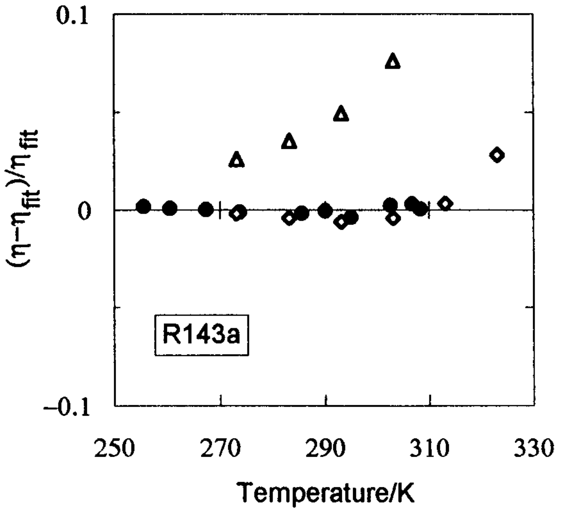

**Figure 4.** Deviations from the fit in Table 5 for the liquid viscosity of R143a. The key for the data is as follows: ● for the present work, △ for the data of Kumagai and Takahashi (1991), and ◇ for the data of Kumagai and Takahashi with a correction applied for the vapor phase density, as described in the text.

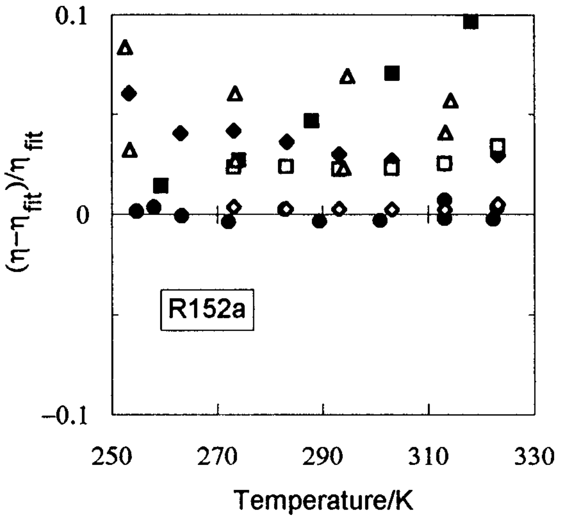

**Figure 5.** Deviations from the fit in Table 5 for the liquid viscosity of R152a. The key for the data is as follows: ● for the present work, ◆ for the revised data of van der Gulik (1995), ◇ for the data of Kumagai and Takahashi (1991), □ for the data of Assael *et al.* (1994), ■ for the data of Phillips and Murphy (1979), and △ for the data of Arnemann and Kruse (1991). The data of Phillips and Murphy and the data of Kumagai and Takahashi have been corrected for the vapor phase density, as discussed in the text.

work range from 0% to +8% (Arnemann and Kruse, 1991; Kumagai and Takahashi, 1991; van der Gulik, 1993, 1995; Assael *et al.*, 1994). The viscosity values of Kumagai and Takahashi and the values of Phillips and Murphy have been corrected for the vapor phase density, as described in the paragraph above. The corrected values of Kumagai and Takahashi agree within 1% with the present values, well within the mutual uncertainty. Two older sets of data for R152a (Phillips and Murphy, 1979; Mears *et al.*, 1955) give larger deviations, but both sets were obtained using a coiled capillary viscometer without correction for the capillary curvature (White, 1929). Substantial errors at low viscosity values may result from this practice. The data of Mears *et al.* is as much as 60% higher than all other data sets and is not shown in Figure 5.

One round-robin study (Assael *et al.*, 1996) found that for R134a, measurements by three groups on the same sample were consistent to approximately 3%. The authors concluded that impurities in the liquids may be the source of larger discrepancies. However, our results and the corrected results of Kumagai and Takahashi for R143a and R152a, both obtained with capillary viscometers, are remarkably consistent even though different fluid samples were used. Similarly for R152a, the two sets of data obtained with vibrating wire viscometers (Assael *et al.*, 1994; van der Gulik, 1995) agree well in spite of the use of different samples. The difference between viscosity values obtained with the two types of viscometers ranges from 2% near ambient temperatures to 6% at the lowest temperatures. We suggest that systematic effects common to each type of viscometer, such as surface tension effects in the capillary viscometers or adsorption effects in the vibrating wire viscometers, may be as important as impurities in accounting for these deviations.

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