Viscosity of Saturated R152a Measured with a Vibrating Wire Viscometer

P. S. van der Gulik 1

Received September 22, 1994

Earlier reported values of the viscosity coefficient of the refrigerant R152a (l,l-difluoroethane) have been recalculated with an improved value for the mechanical damping of the vibrating wire viscometer. The measurements were taken along the saturation line both in the saturated liquid and in the saturated vapor every 10 K from 243 up to 393 K by means of a vibrating wire viscometer. The damping of the vibration of the wire is a measure for the viscosity provided that the mechanical damping is subtracted. The latter is usually measured in vacuum. It turns out that the damping value measured in this way depends on the vacuum pressure and on the way the wire has been handled before. It appeared that the damping applied previously, measured after 6 days of pumping, is too small, resulting in values of the viscosity coefficient which are too large. The effect on the data for the saturated-liquid viscosity is small, but the new saturated-vapor viscosity data agree much better with the unsaturated-vapor data reported by Takahashi et al.

KEY WORDS: 1,1-difluoroethane; mechanical damping; R152a; refrigerants; vibrating wire viscometer; viscosity.

1. INTRODUCTION

There exists current interest in the transport properties of alternative refrigerants which do not deplete the ozone layer and do not contribute to the greenhouse effect. One of these alternatives is the refrigerant R152a (1,1-difluoroethane). Since it is inflammable, it is preferably applied as one of the components of a refrigerant mixture or in isolated industrial applications. For the latter purpose an industrial need exists for the viscosity

¹ Van der Waals-Zeeman Laboratory, University of Amsterdam, Valckenierstraat 67, 1018 XE Amsterdam, The Netherlands.

coefficient along the saturation line, both in the saturated liquid and in the saturated vapor.

The vibrating wire viscometer is well suited for this type of measurement since it is simple and small, it does not disturb the system, and it does not use pressure differences. For the investigation of the viscosity of the boiling liquid it is mounted in the lower half of a relatively long pressure vessel, and for that of the saturated vapor upside down in the upper half, while in both cases the vapor-liquid interface is located in the other half of the vessel. This configuration was used in our investigation, and the results were reported in an earlier publication [1].

In the present paper, we analyze a refinement of the measuring method, which was found subsequently, and which leads to a recalculation of the earlier results. With our vibrating wire viscometer the damping of the vibration of the wire is a measure for the viscosity provided that the mechanical damping is subtracted. The value of the mechanical damping is measured separately in vacuum. We found that this value depends on the vacuum pressure and on the way the wire has been treated before. To remove as many impurities as possible, the system had been pumped thoroughly for 6 days. The mechanical damping was measured at the highest vacuum and this value was used for the calculations reported in $[1]$. Afterward, this value has turned out to be too small, resulting in values of the viscosity coefficient which are too large. Apparently, so many impurities were removed that the wire was no longer in the state corresponding to the mechanical damping effect when gas is present. The effect on the data for the saturated liquid is small, but the new saturated-vapor data are up to 17% lower than the previous data and agree much better with the unsaturated-vapor data of Takahashi et al. [2].

Details about the mechanical damping are given in the next section and the results of the recalculation are presented in Section 3.

2. THE MECHANICAL DAMPING

As mentioned above, the measurements were performed by means of a vibrating wire viscometer $\lceil 3-6 \rceil$. A thin tungsten wire, with a length of 15 mm and a radius of about 25 μ m, is set into vibration by the combined action of a magnetic field and a sinusoidal electric current with a frequency close to the resonance frequency of the wire. After the current is stopped, the voltage, induced by the decaying vibration, is amplified, sampled and stored. After Fourier transformation, the width *df* and frequency of the resonance line f are computed. The viscosity coefficient is now calculated from the density at the measured temperature and from *(df/f),* according to the working equations for the vibrating wire viscometer given in Refs. 5

and 6. In this procedure the mechanical damping of the wire $(df/f)_{\text{vac}}$ is subtracted from (df/f) . Further details of the apparatus and the method can be found in the previous article $\lceil 1 \rceil$.

At the start of the investigation the complete fdling system and the vessel containing the viscometer were evacuated by means of a two-stage rotary oil pump to remove as many impurities as possible. The pumping was continued for 6 days until the damping *df* of the wire did not change any longer. At this vacuum, the mechanical damping of the wire was measured as 0.329 Hz at a frequency f of 3571.00 Hz. Consequently, the system was filled with vapor, pumped again to the same vacuum pressure, and then filled definitively. Delay of boiling, observed several times during the investigation, confirmed the success in removing impurities from the vessel. The value of $(df/f)_{\text{vac}}$ for the mechanical damping obtained in this way and used for the calculations in Ref. 1 was 0.9213×10^{-4} .

After the measurements on R 152a the mechanical damping of a new wire mounted in a different holder was determined in a high-vacuum vessel. The value obtained at a high-vacuum pressure of about 10^{-4} Pa $(10^{-6}$ mbar) was much too small, however, compared with the value which can be calculated from the damping in open air and the known viscosity of open air. Apparently, the sintered tungsten wire was in a different condition than before in the open air, presumably because it was degassed and adsorbed impurities were removed.

Consequently, a separate study was made of the behavior of tungsten wires in vacuum at room temperature. The results can be summarized as follows: down to 50 Pa the resonance width *df* decreases with the pressure. In the range from 10 to 50 Pa the behavior is unclear due to the transition to the Knudsen regime. Next, *dfis* constant in the range from 0.5 to 10 Pa. In the range between rough and high vacuum, 0.5 to 0.005 Pa, a stable state could not be established with the available equipment. Finally, at high vacuum, 0.005 Pa and lower, the value of *df* was about 15 % smaller. The correct value was again determined from the damping in open air and the known viscosity and density of air at room temperature and atmospheric pressure. We could conclude that the correct value of the mechanical damping is obtained at pressures of about 1 to 10 Pa after about 1 h of pumping with a two-stage rotary oil pump. Longer pumping with a good pump or pumping with a high-vacuum pump will degas the wire and remove adsorbed layers which normally form part of the wire. That procedure will produce too low values for the mechanical damping and, thus, too high values for the viscosity.

In the case of our measurements of the viscosity of R152a $\lceil 1 \rceil$, we pumped the system thoroughly for 6 days to remove impurities. So the value for the mechanical damping measured in these circumstances was too

low and therefore the values calculated for the viscosity coefficient too high. Looking back at our notes, we found a value of $df = 0.369$ Hz taken after 1 h of pumping, resulting in $(df/f)_{\text{vac}} = 0.000119$. This value agrees convincingly with the value 0.00012, which is calculated from the viscosity of open air as measured during the mounting process of the wire. Therefore, we have recalculated the viscosity coefficient of R152a with $\left(\frac{df}{f}\right)_{\text{max}} =$ 0.000119 as the value of the mechanical damping.

3. RESULTS

The measurement of the viscosity consists of measurement of the temperature of the sample fluid, calculation of the saturated-vapor pressure from the correlation given by Higashi et al. [7], and of the density at this state from the equation of state given by Tamatsu et al. $[8]$, and determination of the resonance width and frequency of the transversal vibration of the wire. This determination is repeated four or five times with different sampling times and subsequently averaged. The measurements were taken along the saturation line both in the saturated liquid and in the saturated vapor every 10 K from 243 up to 393 K.

The results of the recalculation of the viscosity coefficient of the refrigerant R152a are presented in Tables I and II and in Figs. 1-4. The tables present the measured temperatures T , the saturated-vapor pressures p calculated from the correlation given by Higashi et al. [7], the densities ρ calculated with the use of T and p from the equation of state given by Tamatsu et al. [8], and the viscosity coefficients η computed from ρ and the measured values of *df/f* The results are not presented in the order they were taken.

The new values of the viscosity of the saturated liquid are only slightly smaller than those reported earlier, the difference varying from 0.23% at 243 K to 0.6% at 373 K. One value at 273 K has been omitted.

For the saturated vapor the difference varies from 2.3 % at 373 K up to 17 % at 243 K. For the calculation of these data, a reselection of the raw data has been performed whereby only the results of normal Fourier transformations have been taken into account, and for the sake of consistency, for 243 K another set of data has been chosen.

Since the differences for the saturated liquid are small, only the vapor data are shown in the figures. Figure 1 shows both the newly calculated values of the viscosity coefficient (filled squares) and the old ones (open squares), plotted as a function of the temperature. The critical temperature is also indicated. Values of η measured in the unsaturated vapor by Takahashi et al. [2] are indicated by small crosses, whereas the open circles represent extrapolated saturated-vapor data calculated by means of a

Viscosity of Saturated 152a 871

| Temperature (K) | Pressure (MPa) | Density $(kg \cdot m^{-3})$ | Viscosity $(\mu \, Pa \cdot s)$ |
|--------------------|-------------------|--------------------------------|------------------------------------|
| | | | |
| 243.15 | 0.0778 | 1021.5 | 337.5 |
| 243.16 | 0.0778 | 1021.5 | 338.7 |
| 243.17 | 0.0778 | 1021.5 | 338.3 |
| 253.25 | 0.1216 | 1001.0 | 282.7 |
| 253.25 | 0.1216 | 1001.0 | 283.2 |
| 262.98 | 0.1805 | 980.6 | 246.7 |
| 263.05 | 0.181 | 980.5 | 246.2 |
| 263.05 | 0.181 | 980.5 | 246.4 |
| 273.21 | 0.265 | 958.4 | 218.9 |
| 273.24 | 0.265 | 958.3 | 219.2 |
| 283.21 | 0.373 | 935.6 | 194.5 |
| 283.25 | 0.374 | 935.5 | 194.5 |
| 283.25 | 0.374 | 935.5 | 194.3 |
| 293.18 | 0.513 | 911.7 | 172.6 |
| 293.19 | 0.513 | 911.7 | 172.7 |
| 293.20 | 0.513 | 911.7 | 173.4 |
| 303.23 | 0.691 | 886.3 | 154.6 |
| 303.23 | 0.691 | 886.3 | 153.9 |
| 313.16 | 0.909 | 859.5 | 138.0 |
| 313.16 | 0.909 | 859.5 | 137.8 |
| 323.19 | 1.178 | 830.4 | 123.5 |
| 323.19 | 1.178 | 830.4 | 123.4 |
| 333.23 | 1.503 | 798.8 | 109.0 |
| 333.24 | 1.504 | 798.7 | 108.6 |
| 343.17 | 1.887 | 764.3 | 96.15 |
| 343.18 | 1.887 | 764.2 | 96.09 |
| 353.15 | 2.342 | 725.1 | 84.25 |
| 353.15 | 2.342 | 725.1 | 84.18 |
| 353.16 | 2.342 | 725.0 | 84.22 |
| 363.17 | 2.879 | 678.5 | 73.02 |
| 363.18 | 2.880 | 678.5 | 72.93 |
| 363.19 | 2.880 | 678.4 | 72.83 |
| 372.92 | 3.491 | 620.2 | 61.41 |
| 372.92 | 3.491 | 620.2 | 61.33 |
| 372.95 | 3.492 | 620.0 | 61.00 |
| | | | |

Table I. Viscosity of R152a in the Saturated-Liquid Phase

Fig. 1. The viscosity coefficient of the saturated vapor as a function of the temperature: (\blacksquare) new recalculated values; (\Box) previous values for comparison; $(+)$ unsaturated-vapor values and (O) saturated-vapor values, both of Takahashi etal. [2], the latter extrapolated according to their correlation. The solid line is a polynomial fitting curve as a guide for the eye; the critical temperature is also indicated.

Fig. 2. The viscosity coefficient of the saturated vapor as a function of the density: (\blacksquare) new recalculated values; (\square) previous values for comparison; $(+)$ unsaturated-vapor values and (O) saturated-vapor values, both of Takahashi et al. [2], the latter extrapolated according to their correlation. The solid line is a polynomial fitting curve through the open circles,

Fig. 3. The viscosity coefficient of the saturated vapor as a function of the pressure: (\blacksquare) new recalculated values; (\Box) previous values for comparison; $(+)$ unsaturated-vapor values and (O) saturated-vapor values, both of Takahashi et al. [2], the latter extrapolated according to their correlation. The solid line is a polynomial fitting curve through the open circles.

DENSITY, kg·m³

Fig. 4. The kinematic viscosity coefficient $v = \eta/\rho$ of the saturated vapor as a function of the density. The solid line is a polynomial fitting curve as a guide for the eye.

| Temperature (K) | Pressure (MPa) | Density $(kg \cdot m^{-3})$ | Viscosity $(\mu \text{Pa} \cdot \text{s})$ |
|--------------------|-------------------|--------------------------------|---|
| 242.98 | 0.0772 | 2.610 | 10.39 |
| 242.98 | 0.0772 | 2.610 | 10.32 |
| 253.12 | 0.1209 | 3.974 | 10.45 |
| 253.12 | 0.1209 | 3.974 | 10.27 |
| 263.16 | 0.1818 | 5.833 | 10.57 |
| 263.20 | 0.1820 | 5.841 | 10.57 |
| 273.22 | 0.2645 | 8.329 | 10.80 |
| 273.22 | 0.2645 | 8.330 | 10.79 |
| 273.22 | 0.2645 | 8.330 | 10.82 |
| 283.18 | 0.3729 | 11.583 | 10.61 |
| 283.18 | 0.3729 | 11.583 | 10.62 |
| 283.19 | 0.3729 | 11.584 | 10.56 |
| 293.21 | 0.5135 | 15.824 | 10.63 |
| 293.21 | 0.5135 | 15.824 | 10.53 |
| 303.22 | 0.6908 | 21.25 | 10.84 |
| 303.22 | 0.6909 | 21.25 | 9.90 |
| 303.23 | 0.6909 | 21.26 | 10.56 |
| 303.23 | 0.6910 | 21.26 | 9.84 |
| 313.19 | 0.9097 | 28.14 | 10.64 |
| 313.19 | 0.9098 | 28.14 | 10.48 |
| 313.19 | 0.9098 | 28.14 | 10.54 |
| 323.19 | 1.178 | 36.94 | 11.05 |
| 323.19 | 1.178 | 36.94 | 10.94 |
| 333.16 | 1.501 | 48.15 | 11.59 |
| 333.18 | 1.501 | 48.17 | 11.56 |
| 343.20 | 1.888 | 62.75 | 12.33 |
| 343.20 | 1.888 | 62.75 | 12.33 |
| 343.20 | 1.888 | 62.76 | 12.22 |
| 353.19 | 2.344 | 82.00 | 12.69 |
| 353.19 | 2.344 | 82.00 | 12.77 |
| 353.19 | 2.344 | 82.00 | 12.70 |
| 363.16 | 2.879 | 108.57 | 13.86 |
| 363.16 | 2.879 | 108.58 | 13.85 |
| 373.07 | 3.500 | 148.67 | 14.96 |
| 373.07 | 3.501 | 148.69 | 14.90 |
| 373.08 | 3.501 | 148.72 | 14.88 |

Table 1I. Viscosity of R152a in the Saturated-Vapor Phase

correlation formula provided by these authors. The solid line is a polynomial fitting curve meant only as a guide for the eye.

The same data are shown as a function of the density in Fig. 2 and as a function of the pressure in Fig. 3. Here the solid lines are fitted to the four extrapolated saturated-vapor values of Takahashi et al. [2]. The figures show a good agreement of our new data above room temperature with this fit. The deviation of the values at 373 K is presumably due to the use of different density values near the critical point. For the low temperatures, however, deviations from this fit and a large spread in the data are observed. The transition is marked by the spread in the data taken at 303 K. An explanation can perhaps be found in the plot of the kinematic viscosity as a function of the density (Fig. 4). The kinematic viscosity is the quantity, which in principle is measured with a vibrating wire viscometer. In Fig. 4, the 303 K point marks the transition from the nearly horizontal to the nearly vertical part of the plot. It shows that at low temperatures fluctuations in temperature and pressure have a large influence on the measurements. This is an extra source of inaccuracy, as was also experienced during the measurements.

Accordingly, the accuracy of the measurements is estimated to vary from 0.5 % for the saturated-liquid data to 2% for the high-temperature saturated-vapor data, while for the low-temperature vapor data the inaccuracy is much larger, perhaps up to 13 % when the fits are correct.

For the sake of completeness, the correlations for the dynamic viscosity of the saturated liquid given in Ref. 1 have also been recalculated. The results are nearly the same as before and so are the conclusions. The coefficients are given in Table III. A , B , C , and D are related to the correlation formula

$$
\ln \eta = A + (B/T) + CT + DT^2 \tag{1}
$$

based on the idea of Andrade [9] that the viscosity of a liquid is an exponential function of the temperature. This formula represents the

> Table lIl. Coefficients in Eqs. (1) and (2)

 $A = -47.08926$ $B=4172.9041$ $C = 0.12753767$ $D=-0.0001535643$ $a_0 = -24.530479$ $a_1 = 0.49206785$ $a_2 = -0.0010181847$ recalculated data within 1%. a_0 , a_1 , and a_2 are related to the reciprocal of the viscosity, i.e., the fluidity, as a function of the molar volume. For isotherms this function is mostly linear $\lceil 4, 10, 11 \rceil$, but for the present case this cannot be expected, and therefore it is extended to

$$
1/\eta = a_0 + a_1 V + a_2 V^2 \tag{2}
$$

With the exception of the three lowest temperatures, it gives the viscosity data within 0.5% of the experimental values. At 243 K the deviation is -2.5% ; at 253 K, $+0.9\%$; and at 363 K, $+1.5\%$. In this case, the contribution of the third term is an order of magnitude smaller than those of the first two terms.

ACKNOWLEDGMENTS

This research was performed under contract with the Dutch Agency for Energy and the Environment BV (NOVEM) in the framework of the program New Energy Conversion Technologies (NECT) that is financed by the Netherlands Ministry of Economic Affairs. The author gratefully acknowledges the help of B. E1 Boubkari in performing the measurements in vacuum.

REFERENCES

- I. P. S. van der Gulik, *hit. J. Thermophys.* 14:851 (1993).
- 2. M. Takahashi, C. Yokoyama, and S. Takahashi, J. *Chem. Eng. Data* 32:98 (1987).
- 3. N. J. Trappeniers, P. S. van der Gulik, and H. van den Hooff, *Chem. Phys. Lett.* 70:438 (1980).
- 4. P. S. van der Gulik and N. J. Trappeniers, *Physica A* 135:1 (1986).
- 5. R. Mostert, P. S. van der Gulik, and H. R. van den Berg, *Physica A* 156:909 (1989).
- 6. P. S. van der Gulik, in *Experimental Thermodynamics, Iioi. IIL Measurement of the Transport Properties of Fluids,* W. A. Wakeham, A. Nagashima, and J. V. Sengers, eds. (IUPAC, Blackwell Scientific, Oxford, 1991), p. 79.
- 7. Y. Higashi, M. Ashizawa, Y. Kabata, T. Majima, M. Uematsu, and K. Watanabe, *JSME Int.* J. 30:1106 (1987).
- 8. T. Tamatsu, H. Sato, and K. Watanabe, *Int. J. Refrig.* 16:347 (1993).
- 9. E. N. da C. Andrade, *Phil. Mag.* 17:698 (1934).
- 10. P. S. van der Gulik, R. Mostert, and H. R. van den Berg, *Physica A* 151:153 (1988).
- 11. P. S. van der Gulik, R. Mostert, and H. R. van den Berg, *High Temp.-High Press* 23:87 (1991).