The Viscosity of Normal Deuterium in the Limit of Zero Density

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This paper contains a new representation of the viscosity of normal deuterium in the limit of zero density as a function of temperature. The correlation is based upon the semiclassical kinetic theory of polyatomic gases and a body of critically evaluated experimental data. The similarity of the intermolecular pair potentials of normal hydrogen and normal deuterium is employed to extrapolate the correlation for deuterium beyond the range of the experimental data. In the temperature range 250-350 K the accuracy of the representation of the viscosity is estimated to be $\pm 1\%$, which deteriorates to $\pm 2\%$ at the lowest temperatures and to $\pm 4\%$ at the highest temperatures.

Key words: correlation; deuterium; viscosity.

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1. Introduction

The recent advances in measurement techniques, with concurrent progress in automation and data reduction techniques, are a motivation for a reassessment of the transport property data for fluids. The viscosity of normal deuterium gas has not been measured as frequently as has the same property for other gases. For this reason it has not been possible to base a representation of the viscosity of deuterium on experimental data alone. Instead it has been necessary to combine a small body of experimental data with an estimate of the viscosity based on the similarities between the pair potentials of deuterium and hydrogen. In principle, it would be possible to make an estimate of the thermal conductivity in a similar fashion. However, the theory on which such an estimate would have to be based is less reliable than that for the viscosity so that it is not included in the present treatment.

2. Experimental Data

There are no independent criteria that can be used to assess the accuracy of measurements of the viscosity of po-
The most accurate measurements of the viscosity of deuterium near atmospheric pressure have been performed with the oscillating disk viscometer at Brown University.\textsuperscript{3,4} The measurements were made at room temperature only and have an accuracy that is estimated to be \( \pm 0.2\% \). Because the original evaluation of the data made use of a complete working equation for the instrument and because the measurements have been confirmed by independent studies over a number of years, the results are classified as primary data.

There are two further sets of data arising from the work of Coremans \textit{et al.}\textsuperscript{5} and Menabe\textsuperscript{6} obtained in oscillating disk viscometers, that are considered here as primary data. Data sets of authors estimated the uncertainty of their measurements to be \( \pm 1\% \). However, the measurements were taken without the benefit of the full theory of the instrument and we must therefore recognize that the accuracy may be worse than that claimed and is closer to \( \pm 2\% \). Notwithstanding this reservation, the measurements are among the smallest that extend to low temperatures and the results are consistent with the high accuracy data at room temperature. Therefore, the inclusion of these data in the category of primary data is justified provided that they are assigned an appropriate weight. The remaining primary data for the viscosity of deuterium have been obtained with a capillary viscometer. Although in general, the capillary viscometer measurements are of inferior precision and accuracy to those from the oscillating disk instrument, the results of Michals \textit{et al.}\textsuperscript{7} have been proven to be accurate. The present estimate of the uncertainty in these results is \( \pm 0.2\% \), which is in accord with that claimed by the authors. Table 1 summarizes the primary data used for the viscosity correlation.

The remaining viscosity data for normal deuterium have been classified as secondary. Included among them are the results of Barua \textit{et al.}\textsuperscript{8} and Becker \textit{et al.}\textsuperscript{9} obtained with an oscillating disk viscometer. The results of these two sets of measurements deviate systematically from the high-accuracy data at room temperature—an indication of undetected systematic errors. The early results of van Itterbeeck and van Paemel\textsuperscript{10} have also been excluded because they are superseded by later work of the same laboratory.\textsuperscript{5}

### 3. Methodology

The viscosity \( \eta \) of a pure gas in the limit of zero density may always be written in the form\textsuperscript{11}

\[
\eta = \frac{5}{16} \left( \frac{mkT}{\pi} \right)^{\frac{1}{2}} \frac{1}{\sigma^2 \Omega^* (T^*)},
\]

where \( m \) is the molecular mass, \( T \) the absolute temperature, and \( k \) is Boltzmann's constant. The symbol \( \sigma \) represents a length scaling parameter and \( \Omega^* \) is a functional of the pair potential for the interaction between the molecules of the gas. In the case of spherically symmetric pair potentials for monatomic particles, where the potential may be expressed in a universal reduced form

\[
U^*(r^*) = U(r/a)/\epsilon,
\]

it can readily be shown\textsuperscript{12} that the functional \( \Omega^* \) is a function only of the reduced temperature \( T^* = kT/\epsilon \), and that it is universal among the same interactions. For polyatomic gases interacting through nonspherically symmetric pair potentials, the functional \( \Omega^* \) depends not only upon the intermolecular pair potential but also upon the internal energy states of the molecules. Nevertheless, Eq. (1) provides a convenient basis for the representation of experimental data.\textsuperscript{12}

The primary experimental data for normal deuterium cover only the temperature range from 30 to 350 K and are inadequate to form the basis of a wide-ranging correlation. We have therefore begun the correlation with a different approach. We first recognize that the intermolecular pair potentials of hydrogen and deuterium are essentially identical.\textsuperscript{1} Thus, at sufficiently high temperatures such that the differences in the nuclear spins of the two molecules have no effect on the viscosity, we may use Eq. (1) to write

\[
\frac{\eta_{D_2}}{\eta_{H_2}} \left( \frac{m_{H_2}}{m_{D_2}} \right)^{1/2} = 1.
\]

On the basis of model calculations\textsuperscript{12} this result is expected to be accurate to better than 0.1% for temperatures above 300 K. Thus we may employ Eq. (1) together with our previous correction for the viscosity of hydrogen\textsuperscript{10} to represent this viscosity of normal deuterium above 350 K, without incurring any significant error beyond that inherent in the hydrogen viscosity.
The viscosity of hydrogen is represented by Eq. (1) with
\[
\ln[\Omega^*(T^*)] = 0.46649 - 0.57015(\ln T^*) + 0.19164(\ln T^*)^2 - 0.03708(\ln T^*)^3 + 0.00241(\ln T^*)^4,
\]
(0.6 < T* < 66), \hspace{0.5cm} (4)

in which
\[T^* = kT/e,\]
and where \(\sigma = 0.2968\) nm and \(e/k = 33.3\) K. Thus, Eqs. (1)–(5) yield a representation of the viscosity of normal deuterium over the temperature range 350–2200 K (10.5 < T* < 66).

Below 350 K the nuclear spin effects become increasingly important so that in the temperature range down to 30 K we have based the correlation on the primary experimental data. For the sake of consistency with the high-temperature correlation, we have employed a representation that makes use of the viscosity of hydrogen by defining
\[
\eta_{H_2} = \left( \frac{m_{H_2}}{m_{D_2}} \right)^{1/2} \eta_{D_2},\]
(6)

The experimental values of \(f(T^*)\) are shown in Fig. 1 and rise, as expected, asymptotically to unity near ambient conditions. By means of a weighted least-squares fit we have then represented \(f(T^*)\) by the equation
\[
f(T^*) = 1 - \exp(-2.8105T^{*1/3}),\]
(0.9 < T* < 10.5),

which departs insignificantly from unity at \(T = 350\) K. The resulting function is also plotted in Fig. 1. Together with Eqs. (1), (4), and (5), the correlation of Eq. (6) describes the viscosity of normal deuterium over the temperature range 30 < T < 350 K.

4. The Viscosity Correlation

The viscosity of normal deuterium is represented by Eqs. (1) and (2)–(7), over a temperature range 30 < T* < 2200 K (0.90 < T* < 66). Table 2 presents values of the viscosity at selected temperature intervals in this range. Figure 2 contains a plot of the deviations of the primary experimental data from this correlation. The experimental data depart from the correlation by no more than \(\pm 0.7\%\) over the entire temperature range. Our estimate of the uncertainty in the viscosity correlation within the temperature range 250–350 K is based upon the errors in the experimental data themselves and is \(\pm 1\%\). At lower temperatures the uncertainty must be increased to \(\pm 2\%\) to allow for the rather greater errors in the experimental data. At temperatures above 350 K where the correlation is based on a correspondence principle it is more difficult to assess the uncertainty, but accounting for the uncertainty in the viscosity of hydrogen, the overall error should be no more than \(\pm 4\%\) at the highest temperatures.

![Fig. 1. The values of the function \(f(T^*)\) deduced from the primary viscosity data as a function of temperature (Refs. 3–7).](image)

![Fig. 2. Deviations of the primary experimental data for the viscosity of normal deuterium from the correlation of Eqs. (1) and (3)–(5) (Refs. 3–7).](image)
Fig. 3. Deviations of the secondary experimental data for the viscosity of normal deuterium from the correlation of Eqs. (1) and (3) (5) (Refs. 8–11).

Deviations of the secondary data from the correlation are displayed in Fig. 3.

5. Conclusions

A concise representation of the viscosity of normal deuterium over the temperature range 30–2200 K has been developed. The correlation has an accuracy of ±1% between 250 and 350 K, which deteriorates to ±2% at the lowest temperature and ±4% at the highest temperature.

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7. References