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 ± 1%, which deteriorates to ± 2% at the lowest temperatures and to ± 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-

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lyatomic gases. Consequently, the selection of the data to be employed in a correlation must be based entirely upon a critical evaluation of the measurement technique involved and the precision attained. Therefore following the procedures discussed elsewhere,[1,2] it is both convenient and necessary to define two categories of experimental data.

(i) Primary data. These are the results of measurements carried out with an instrument of high precision for which a complete working equation and a detailed knowledge of all corrections are available.

(ii) Secondary data. These are the results of measurements which are of inferior accuracy to primary data. The inferior accuracy may arise from operation at extreme conditions or from an incomplete characterization of the apparatus.

Only the primary data have been employed in the development of the correlation discussed here. The secondary data are used exclusively for comparison purposes.

2.1. Coefficient of Viscosity

The most accurate measurements of the viscosity of deuterium near atmospheric pressure have been performed with the oscillating disk viscometer at Brown University.[3,4] The measurements were made at room temperature only and have an accuracy that is estimated to be ± 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 *et al.*[5] and Menabde[6] obtained in oscillating disk viscometers, that are considered here as primary data. Both sets of authors estimated the uncertainty of their measurements to be ± 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 ± 2%. Notwithstanding this reservation, the measurements are among the small set 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 Michels *et al.*[7] have been proven to be accurate. The present estimate of the uncertainty in these results is ± 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 *et al.*[8] and Becker *et al.*,[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 Itterbeek and van Paemel[11] have also been excluded because they are superseded by later work of the same Laboratory.[5]

Table 1. Primary experimental data for the viscosity of normal deuterium

Reference	Technique	Temperature Range (K)	Estimated Uncertainty (%)
Kestin et al.[3,4]	Oscillating disc	293 - 303	± 0.2
Coremans et al.[5]	Oscillating disc	30 - 80	± 2.0
Menabde[6]	Oscillating disc	80 - 300	± 2.0
Michels et al.[7]	Capillary	290 - 350	± 0.2

3. Methodology

The viscosity η of a pure gas in the limit of zero density may always be written in the form[11]

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

where m is the molecular mass, T the absolute temperature, and k is Boltzmann's constant. The symbol σ represents a length scaling parameter and Ω^* 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 structureless particles, where the potential may be expressed in a universal reduced form

$$U^*(r^*) = U(r/\sigma)/\epsilon\,, \tag{2}$$

it can readily be shown[12] that the functional Ω^* 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 Ω^* 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.[1,2]

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.[12] 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

$$\frac{\eta_{D_2}}{\eta_{H_2}}\left(\frac{m_{H_2}}{m_{D_2}}\right)^{1/2} = 1\,. \tag{3}$$

On the basis of model calculations[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 correlation for the viscosity of hydrogen[2] 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

$$\ln[\Omega^*(T^*)] = 0.466\,49 - 0.570\,15(\ln T^*) + 0.191\,64(\ln T^*)^2 - 0.037\,08(\ln T^*)^3 + 0.002\,41(\ln T^*)^4, \quad (0.6 < T^* < 66), \tag{4}$$

in which

$$T^* = kT/\epsilon, \tag{5}$$

and where $\sigma = 0.2968$ nm and $\epsilon/k = 33.3$ 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

$$\frac{\eta_{D_2}}{\eta_{H_2}} \left(\frac{m_{H_2}}{m_{D_2}} \right)^{1/2} = f(T^*) . \tag{6}$$

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

$$f(T^*) = 1 - \exp(-2.8105 T^{*1/3}), \quad (0.9 < T^* < 10.5), \tag{7}$$

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.

Table 2. The viscosity of normal deuterium in the limit of zero density

T/K	η/μPa s	T/K	η/μPa s	T/K	η/μPa s	T/K	η/μPa s
30	2.084	520	18.41	1080	30.63	1640	41.14
35	2.430	530	18.65	1090	30.83	1650	41.32
40	2.759	540	18.89	1100	31.03	1660	41.49
45	3.072	550	19.13	1110	31.23	1670	41.67
50	3.371	560	19.37	1120	31.42	1680	41.84
55	3.658	570	19.61	1130	31.62	1690	42.02
60	3.934	580	19.85	1140	31.82	1700	42.20
65	4.200	590	20.08	1150	32.02	1710	42.37
70	4.458	600	20.32	1160	32.21	1720	42.55
75	4.707	610	20.55	1170	32.41	1730	42.72
80	4.950	620	20.78	1180	32.60	1740	42.89
85	5.186	630	21.02	1190	32.80	1750	43.07
90	5.410	640	21.25	1200	32.99	1760	43.24
95	5.640	650	21.48	1210	33.19	1770	43.41
100	5.860	660	21.70	1220	33.38	1780	43.59
110	6.285	670	21.93	1230	33.57	1790	43.76
120	6.695	680	22.16	1240	33.76	1800	43.93
130	7.091	690	22.39	1250	33.96	1810	44.10
140	7.474	700	22.61	1260	34.15	1820	44.28
150	7.847	710	22.83	1270	34.34	1830	44.45
160	8.210	720	23.06	1280	34.53	1840	44.62
170	8.564	730	23.28	1290	34.72	1850	44.79
180	8.910	740	23.50	1300	34.90	1860	44.96
190	9.249	750	23.72	1310	35.10	1870	45.13
200	9.582	760	23.94	1320	35.29	1880	45.30
210	9.909	770	24.16	1330	35.48	1890	45.47
220	10.23	780	24.38	1340	35.67	1900	45.64
230	10.55	790	24.60	1350	35.85	1910	45.81
240	10.86	800	24.81	1360	36.04	1920	45.98
250	11.16	810	25.03	1370	36.23	1930	46.14
260	11.47	820	25.25	1380	36.41	1940	46.31
270	11.77	830	25.46	1390	36.60	1950	46.48
280	12.06	840	25.68	1400	36.79	1960	46.65
290	12.35	850	25.89	1410	36.97	1970	46.81
300	12.64	860	26.10	1420	37.16	1980	46.98
310	12.93	870	26.31	1430	37.34	1990	47.15
320	13.21	880	26.53	1440	37.53	2000	47.32
330	13.49	890	26.74	1450	37.71	2010	47.48
340	13.77	900	26.95	1460	37.89	2020	47.65
350	14.04	910	27.16	1470	38.08	2030	47.81
360	14.31	920	27.36	1480	38.26	2040	47.98
370	14.58	930	27.57	1490	38.44	2050	48.14
380	14.85	940	27.78	1500	38.63	2060	48.31
390	15.12	950	27.99	1510	38.81	2070	48.47
400	15.38	960	28.19	1520	38.99	2080	48.64
410	15.64	970	28.40	1530	39.17	2090	48.80
420	15.90	980	28.61	1540	39.35	2100	48.96
430	16.16	990	28.81	1550	39.53	2110	49.13
440	16.42	1000	29.02	1560	39.71	2120	49.29
450	16.67	1010	29.22	1570	39.89	2130	49.45
460	16.92	1020	29.42	1580	40.07	2140	49.62
470	17.17	1030	29.62	1590	40.25	2150	49.78
480	17.42	1040	29.83	1600	40.43	2160	49.94
490	17.67	1050	30.03	1610	40.61	2170	50.10
500	17.92	1060	30.23	1620	40.78	2180	50.27
510	18.17	1070	30.43	1630	40.96	2190	50.43
						2200	50.59

4. The Viscosity Correlation

The viscosity of normal deuterium is represented by Eqs. (1) and (3)–(7), over a temperature range $30 \leqslant T \leqslant 2200$ K ($0.90 \leqslant T^* \leqslant 66$). 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 ±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 ±1%. At lower temperatures the uncertainty must be increased to ±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 ±4% at the highest temperatures.

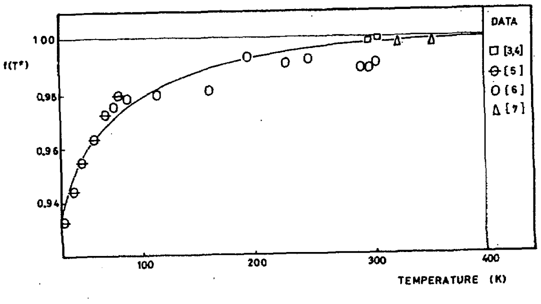

FIG. 1. The values of the function $f(T^*)$ deduced from the primary viscosity data as a function of temperature (Refs. 3–7).

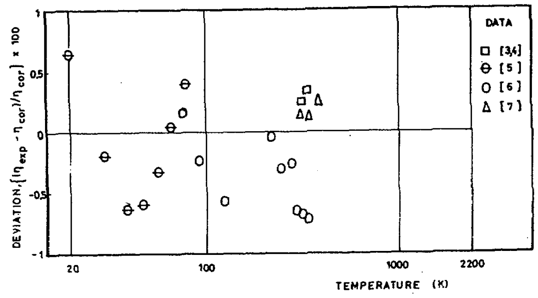

FIG. 2. Deviations of the primary experimental data for the viscosity of normal deuterium from the correlation of Eqs. (1) and (3)–(5) (Refs. 3–7).

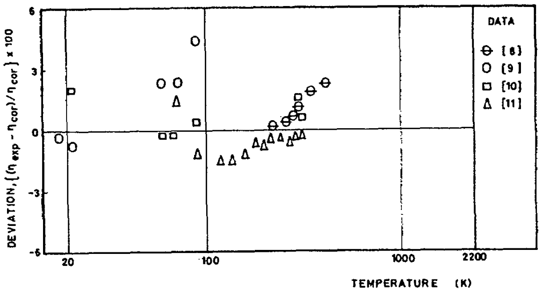

FIG. 3. Deviations of the secondary experimental data for the viscosity of normal deuterium from the correlation of Eqs. (1) and (3)–(5) (Refs. 8–11).

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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