The Viscosity of Nonpolar Gases at Normal Pressures

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Experimental viscosity data available in the literature for fifty-two nonpolar gases have been utilized in conjunction with a dimensional analysis approach to relate the viscosity at atmospheric pressure to temperature. The substances investigated are both simple and complex and include the inert and diatomic gases, carbon dioxide, carbon disulfide, carbon tetrachloride, and the hydrocarbons up to *n*-nonane, including normal and isoparaffins, olefins, acetylenes, naphthenes, and aromatics. The dependence of the product $\mu^*\xi$ on reduced temperature was found to be the same for all of these substances, except helium and hydrogen.

Both theoretical considerations and dimensional analysis indicate that the viscosity product of a gas might depend on the compressibility factor at the critical point. However the results of this study show that for these nonpolar substances this viscosity product at normal pressure is independent of z_c and depends only on temperature.

The only information required for the calculation of viscosity with the relationships developed in this study is the molecular weight, critical temperature, and critical pressure of the substance. Values calculated with these relationships have been compared with 785 experimental points from all reliable sources of experimental data and produced an average deviation of 1.77%. Comparisons have also been made with the values calculated with the Licht-Stechert and Bromley-Wilke equations.

The development of accurate relationships for the transport properties of gases is currently receiving considerable interest as a result of demands brought forth by modern technological advances. Assuming that molecules are rigid spheres and employing the Lennard-Jones force constants ϵ/κ and σ, Hirschfelder, Curtiss, and Bird (*19*) have developed expressions for the transport properties of nonpolar gases at moderate pressures. For this case they propose that the first approximation of viscosity is

$$[\mu^*]_1 = 266.93 \times 10^{-5} \frac{\sqrt{MT}}{\sigma^2 \Omega^{(2,2)*}[T_N]} \qquad (1)$$

where the viscosity is in centipoises and the collision integral $\Omega^{(2,2)*}[T_N]$ is a tabulated function of the normalized temperature $T_N = T/\frac{\epsilon}{\kappa}$. Because the determination of the Lennard-Jones force constants requires reliable viscosity and/or PVT data, the values calculated with Equation (1) are of questionable accuracy when this equation is applied to gases for which such data are not available.

Thodos and co-workers (*9, 50*) have developed viscosity correlations for the monatomic and twelve diatomic gases in which they relate the viscosity ratio $\mu^*/\mu^*_{T_c}$ to T_R and obtained good agreement with experimental data. However this approach can be applied only to those gases for which reliable viscosity data are available. It would therefore be advantageous to relate the viscosity to the critical constants of the gas, since these constants are more readily available than experimental viscosity data and more dependable than Lennard-Jones force constants.

One of the earliest attempts to relate the viscosity of a gas to its critical constants was made by Kamerlingh Onnes (*30*) in 1881. Using kinetic theory he derived for the viscosity of a gas at its critical temperature and moderate pressures

$$\mu^*_{T_c}\left(\frac{T_c}{M^3 P^4_c}\right)^{1/6} = k \qquad (2)$$

In 1893 Sutherland (*55*), also using kinetic theory, related the viscosity of a gas to reduced temperature as follows:

$$\frac{\mu^*}{\mu^*_{T_c}} = \left[\frac{1+S}{T_R+S}\right] T_R^{3/2} \qquad (3)$$

Equation (3) has found considerable utility by a number of investigators.

From available viscosity data Licht and Stechert (*37*) determined k of Equation (2) to be 35 x 10^{-5} when the critical temperature is in degrees Kelvin, the critical pressure in atmospheres, and the viscosity in centipoises. They also determined S in Equation (3) to be approximately 0.8 for all gases. Combining Equations (2) and (3) Licht and Stechert proposed for the viscosity of a gas at moderate pressures

$$\mu^* = 63.0 \times 10^{-5}\left(\frac{M^3 P^4_c}{T_c}\right)^{1/6} \frac{T_R^{3/2}}{T_R + 0.8} \qquad (4)$$

Bromley and Wilke (*11*), using the approximate relationships that $\epsilon/\kappa = 0.75\ T_c$ and $\sigma = 0.833\ v_c^{1/3}$, simplified Equation (1) to produce the following equation for the viscosity of a gas at moderate pressures:

$$\mu^* = 333 \times 10^{-5} \frac{(M\,T_c)^{1/2}}{v_c^{2/3}} f\,(1.33\,T_R) \qquad (5)$$

where values of $f\ (1.33\,T_R)$ are tabulated by them.

In all these investigations the viscosity was determined to be a function of the molecular weight and the critical constants of the gas. In order to determine the proper dependence of the viscosity of nonpolar gases on these and other variables a dimensional analysis approach has been employed in this study.

DIMENSIONAL ANALYSIS

Kinetic theory indicates that the viscosity of a gas at moderate pressures does not vary with pressure but is dependent on temperature and molecular weight. It is to be expected that the viscosity of the gas also depends on the critical constants T_c, P_c, and v_c. Applying the Rayleigh method of dimensional analysis to these variables one gets

$$\mu^* = \alpha T^a\, T^b_c\, M^c\, P^d_c\, v^e_c\, R^f \qquad (6)$$

The dimensions involved are mass, length, time, and temperature. The dimensional analysis produces the following values for the exponents:

$$\begin{array}{ll} a = f - b & e = -1/6 - f \\ b = b & d = 1/2 - f \\ c = 1/2 & f = f \end{array}$$

TABLE 1. BASIC CONSTANTS, SOURCES OF VISCOSITY DATA, AND AVERAGE DEVIATIONS

	M	T_c, °K.	P_c, atm.	z_c	ξ	Average deviation, %	References
Monatomic							
Helium	4.003	5.206	2.26	0.306	0.381	1.88	*17, 18, 24, 26, 27, 43, 70, 71, 76*
Neon	20.183	44.5	26.86	0.307	0.0466	1.06	*14, 26, 27, 43, 47, 69, 70, 76*
Argon	39.944	151.2	48.00	0.291	0.0276	1.50	*6, 25, 27, 43, 47, 49, 70, 73, 76*
Krypton	83.80	209.4	54.18	0.292	0.0184	2.04	*41, 43,*
Xenon	131.3	289.8	57.64	0.290	0.0151	0.79	*41, 43, 60*
Diatomic							
Hydrogen	2.016	33.3	12.8	0.305	0.230	1.26	*2, 18, 22, 23, 26, 29, 31, 33, 54, 58, 59, 62, 63, 64, 66, 67, 68, 70, 75, 76*
Nitrogen	28.02	126.2	33.5	0.291	0.0407	1.62	*23, 28, 29, 58, 59, 64, 70, 74, 75, 76, 77*
Oxygen	32.00	154.8	50.1	0.293	0.0301	1.66	*21, 23, 29, 59, 64, 70, 75, 76*
Carbon monoxide	28.01	133.0	34.5	0.294	0.0402	3.61	*27, 58, 64, 75*
Nitric oxide	30.01	180	64	0.251	0.0271	2.35	*29, 75, 76*
Fluorine	38.00	144	55		0.0257*	2.23	*32*
Chlorine	70.91	417.2	76.1	0.276	0.0180	1.90	*8, 65, 76*
Bromine	159.8	584	102	0.306	0.0104*	2.49	*7, 44*
Iodine	253.8	785	116		0.0080*	1.32	*8*
Hydrocarbons							
Methane	16.04	191.1	45.8	0.289	0.0464	1.05	*1, 3, 12, 20, 29, 36, 45, 66, 70, 75, 76*
Ethane	30.07	305.5	48.2	0.285	0.0357	1.15	*1, 13, 20, 36, 51, 56, 66*
Propane	44.09	370.0	42.0	0.277	0.0334	1.68	*1, 3, 12, 20, 36, 51, 56, 62, 66, 76*
n-Butane	58.12	425.2	37.5	0.274	0.0321	1.31	*35, 36, 48, 56, 76*
n-Pentane	72.15	469.8	33.3	0.269	0.0317	1.41	*4, 36, 57*
n-Hexane	86.17	507.9	29.9	0.264	0.0315	2.29	*13, 36, 38, 57*
n-Heptane	100.2	540.2	27.0	0.260	0.0316	2.68	*36, 39*
n-Octane	114.2	569.4	24.6	0.258	0.0318	3.11	*36, 38, 39*
n-Nonane	128.2	595.4	22.6	0.250	0.0320	a	*39*
i-Butane	58.12	408.5	36.0	0.282	0.0327	1.15	*20, 36, 48, 56*
2-Methylbutane	72.15	461.0	32.9	0.268	0.0318	2.23	*4, 36, 38, 46*
2, 2-Dimethylpropane	72.15	433.8	31.6	0.269	0.0290	a	*36*
2, 2-Dimethylbutane	86.17	489.4	30.7	0.274	0.0308	a	*36*
2, 4-Dimethylpentane	100.2	520.3	27.4	0.269	0.0312	1.49	*36*
2, 2, 3-Trimethylbutane	100.2	531.5	29.75	0.269	0.0296	3.63	*15*
2, 2, 4-Trimethylpentane	114.2	544.1	25.4	0.267	0.0321	4.26	*36, 38*
Ethylene	28.05	282.4	50.0	0.267	0.0356	1.81	*12, 36, 56, 58, 64, 72*
Propylene	42.08	365.1	45.4	0.280	0.0324	2.06	*1, 56, 61, 72*
i-Butylene	56.10	417.9	39.5	0.275	0.0315	2.72	*56*
Butene-1	56.10	419.6	39.7	0.276	0.0314	0.93	*56*
Butene-2	56.10	430	41.0	0.274	0.0308	1.77	*36, 56, 61*
Pentene-2	70.13	475.0	36.0	0.268	0.0306	2.27	*4*
i-Amylene	70.13	464.8	33.9	0.260	0.0317	3.48	*4, 56*
Hexene-1	84.16	503.7	31.2	0.262	0.0310	1.00	*38*
1, 3-Butadiene	54.09	425.2	42.7	0.270	0.0304	a	*36*
Ethyne	26.04	309.2	61.6	0.274	0.0326	2.08	*1, 34, 56, 75, 76*
Propyne	40.06	401.2	52.8	0.266	0.0305	1.89	*56*
Cyclopropane	42.08	406.7	57.2	0.287	0.0283	0.70	*36*
Cyclopentane	70.13	512.1	44.6	0.276	0.0269	0.88	*36*
Methylcyclopentane	84.16	532.8	37.4	0.272	0.0278	3.41	*38*
Cyclohexane	84.16	553.2	40.0	0.272	0.0267	1.38	*13, 40, 57*
Cyclooctane	112.2	620.2	32.9	0.261	0.0269	a	*38*
Benzene	78.11	562.2	48.6	0.274	0.0244	1.41	*13, 38, 40, 46, 53, 57*
Toluene	92.13	594.0	41.6	0.271	0.0251	1.58	*40*
1, 3, 5-Trimethylbenzene	120.19	641.2	33	0.282	0.0260	a	*15*
Miscellaneous carbon compounds							
Carbon dioxide	44.01	304.2	72.85	0.275	0.0224	1.23	*10, 12, 29, 34, 42, 62, 70, 74*
Carbon disulfide	76.13	552	78.0	0.298	0.0180	3.04	*53, 56*
Carbon tetrachloride	153.8	556.4	45.0	0.272	0.0183	3.40	*8, 52, 57*

* Use $\xi = 0.0396$ for fluorine, $\xi = 0.0115$ for bromine, and $\xi = 0.00905$ for iodine.
a Average deviation greater than 5%; critical constants and reliability of viscosity data uncertain.

With these exponents Equation (6) becomes

$$\mu^* \frac{T_c^{1/6}}{M^{1/2} P_c^{2/3}} = \beta z_c^m T_R^n \qquad (7)$$

where $z_c = P_c v_c / R T_c$. For convenience the group $T_c^{1/6}/M^{1/2} P_c^{2/3}$ will be referred to in this study as ξ. This group is a characteristic constant for each substance and is the same group suggested by Kamerlingh Onnes (*30*) and later utilized by Licht and Stechert (*37*). However, contrary to their conclusions, dimensional analysis indicates that μ^* is a function not only of T_R

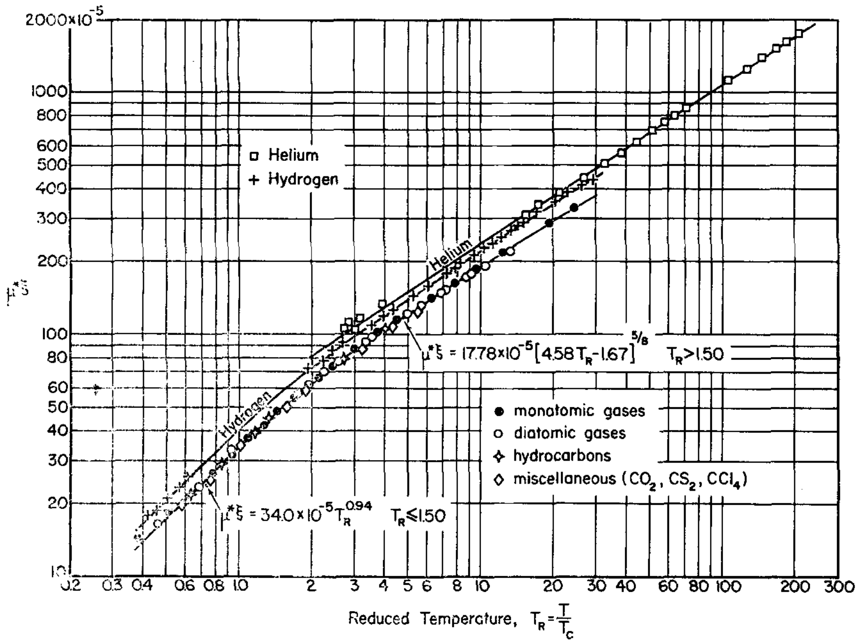

Fig. 1. Relationships of $\mu^*\xi$ and T_R for nonpolar gases at moderate pressures.

but also of z_c. Therefore the product $\mu^*\xi$ becomes a unique function of both z_c and T_R when experimental evidence is utilized to support this conclusion.

TREATMENT OF VISCOSITY DATA

The experimental viscosities at atmospheric pressure for fifty-two nonpolar gases were used to establish the constant β and the exponents m and n of Equation (7). The only experimental data used were those that were found to be internally consistent for each gas. For those gases for which a large number of data was available only the best values of each investigator were included. The gases considered included the monatomic and diatomic gases, carbon dioxide, carbon disulfide, carbon tetrachloride, and hydrocarbons, including normal paraffins, isoparaffins, olefins, acetylenes, naphthenes, and aromatics. These substances are listed in Table 1 along with their molecular weights, critical constants, and calculated values of ξ. The sources of the experimental viscosities used in this work are also listed.

For the reported viscosities of each substance the group $\mu^*\xi$ was calculated and plotted vs. T_R on log-log coordinates. These plots produced essentially identical curves for nearly all the gases. Since many of these substances have widely varying z_c values, it can be concluded that the group $\mu^*\xi$ is independent of z_c and that $m = 0$ in Equation (7). Thus for nonpolar gases the group $\mu^*\xi$ depends only on temperature:

$$\mu^*\xi = \beta T_R^{\,n} \qquad (8)$$

In Figure 1 a composite curve relating $\mu^*\xi$ to T_R is presented for all the substances following this consistent behavior. This figure indicates that the exponent n is constant for $T_R < 1.5$ but varies with reduced temperature above $T_R = 1.5$. The relationship for $T_R < 1.5$ has been determined to be

$$\mu^*\xi = 34.0 \times 10^{-5}\, T_R^{0.94},\ (T_R \leq 1.5) \qquad (9)$$

Above $T_R = 1.5$ the group $\mu^*\xi$ can be related to temperature as follows:

$$\mu^*\xi = 17.78 \times 10^{-5}\, [4.58\, T_R - 1.67]^{5/8},\ (T_R > 1.5) \qquad (10)$$

These equations are applicable to all nonpolar gases at atmospheric pressure with the exceptions of helium and hydrogen, whose relationships are also presented in Figure 1. The abnormal behavior of these gases results from their significant quantum deviations (5). Figure 1 shows that the hydrogen curve is parallel to the curve for the normally behaving gases. Below $T_R = 1.50$ (50°K.) the viscosity of hydrogen in centipoises can be related directly to the absolute temperature in degrees Kelvin as follows:

$$\mu^* = 6.43 \times 10^{-5}\, T^{0.94},\ T \leq 50°\text{K.} \qquad (11)$$

Above this temperature

$$\mu^* = 90.71 \times 10^{-5}\, [0.1375\, T - 1.67]^{5/8} \qquad (12)$$

For helium the viscosity relationship is linear in the region 15°K. < T < 1,100°K. This relationship can be expressed as follows:

$$\mu^* = 46.78 \times 10^{-5}\, T^{0.656},\ T > 15°\text{K.} \qquad (13)$$

The critical constants of fluorine, bromine, and iodine are not well established. The limited viscosity data for these three gases produce curves that are parallel to the curve for the normally behaving gases, indicating that one or more of the critical constants used in the determination of ξ for these substances may be incorrect. Chlorine, for which the critical constants are known to be fairly reliable, conforms to the normal behavior presented in Figure 1. The critical temperatures of these halogens have been examined (*16*) and were found to be consistent with the critical temperatures of other gases. Therefore it can be assumed that if one of the critical constants is in error, this constant is more likely the critical pressure than the critical temperature. If this is the case, the corrected critical pressures of these gases can be determined by establishing the proper ξ values which make the viscosity curves of these gases coincide with the curve for the normally behaving gases. These values of ξ and the corresponding critical pressures for these halogens were found to be

	ξ	P_c, atm.
Fluorine	0.0396	29
Bromine	0.0115	88
Iodine	0.00905	97

Thus this approach offers a useful method for establishing either the critical temperature or the critical pressure of a gas for which reliable viscosity values are available and for which one of these constants is known.

The critical pressures for bromine and iodine calculated by this approach are reasonably close to the reported values for these substances. However the critical pressure of fluorine appears to be significantly different from the reported value, P_c = 55 atm. Therefore the inconsistent viscosity behavior of fluorine is probably due to some other factor, which could be the unreliability of the viscosity data reported by the only investigator (32) or the polymerization of the molecules of the gas.

RESULTS AND CONCLUSIONS

The validity of Equations (9) and (10) has been checked for the fifty-two nonpolar substances included in this study. The corrected ξ values of fluorine, bromine, and iodine were used in the calculation of viscosities for these gases from Equations (9) and (10). The viscosity values calculated with these equations for all the gases considered in this study, except hydrogen and helium, were compared with the corresponding experimental values. Hydrocarbons for which the critical constants had to be estimated and for which the reliability of their viscosity data was uncertain were excluded in the comparison if they produced values which deviated by more than 5%. Equation (9) reproduced 560 viscosity values with an average deviation of 1.83%, while Equation (10) reproduced 225 viscosities with an average deviation of 1.62%. Viscosities calculated for hydrogen with Equations (11) and (12) produced an average deviation of 1.26% for ninety-seven experimental points. For helium an average deviation of 1.88% resulted for thirty-one experimental points with Equation (13).

The method developed in this study compares favorably with those of Bromley-Wilke and Licht-Stechert. For 211 randomly selected experimental points the viscosities were compared with the values calculated with the Bromley-Wilke equation, the Licht-Stechert equation, and Equations (9) and (10). The results of these comparisons are

	Average deviation, %
Bromley-Wilke	2.62
Licht-Stechert	2.17
Equations (9) and (10)	1.64

The Bromley-Wilke viscosity relationship given in Equation (5) can be modified by substituting $v_c = z_c RT_c/P_c$ to produce

$$\mu^*\xi = 17.63 \text{ x } 10^{-5} \frac{f(1.33\, T_R)}{z_c^{2/3}} \quad (14)$$

Since the results of this investigation show that $\mu^*\xi$ is independent of z_c, the dependence of $\mu^*\xi$ on z_c inherent in the Bromley-Wilke equation may be responsible for the higher deviations resulting from the use of their equation. Because of the contradictory influence of z_c there is a fundamental difference between Equations (9) and (10) and the Bromley-Wilke relationship given by Equation (5).

When the Licht-Stechert relationship given in Equation (4) was superimposed on the curve of Figure 1, it was seen that their relationship is capable of reproducing values of $\mu^*\xi$ with small deviations up to $T_R = 10$. For $T_R > 10$ the $\mu^*\xi$ values calculated with the Licht-Stechert equation begin to show significant deviations from the corresponding experimental values. These deviations continue to increase at higher temperatures. However no significant deviations above $T_R = 10$ are encountered with Equation (10). A comparison of $\mu^*\xi$ values above $T_R = 10$ calculated with both the Licht-Stechert equation and Equation (10) with experimental values produced the following results:

$\mu^*\xi \times 10^5$

T_R	Experimental	Licht-Stechert	Equation (10)
10	190	186	190
15	248	231	246
20	295	273	296
25	338	305	341

Thus it is seen that at $T_R = 25$ a deviation of 9.8% results from the use of the Licht-Stechert equation. An attempt was made to adjust the constants k and S of the Licht-Stechert equation in order to improve its consistency with all of the available data. However no values for these constants were found which would produce a relationship which did not show significant viscosity deviations at high temperatures.

NOTATION

a,b,c,d,e,f = constants, Equation (6)
k = universal constant, Equation (2)
m = constant, Equation (7)
M = molecular weight
n = constant, Equation (7)
P_c = critical pressure, atm.
R = gas constant, 82.05 cc. atm./g.-mole °K.
S = Sutherland constant
T = absolute temperature, °K.
T_c = critical temperature, °K.
T_N = normalized temperature, $T/\frac{\epsilon}{\kappa}$
T_R = reduced temperature, T/T_c
v_c = critical volume, cc./g.-mole
z_c = compressibility factor at critical point, $P_c v_c/R T_c$

Greek Letters

α = constant, Equation (6)
β = constant, Equation (7)
ϵ = maximum energy of attraction for Lennard-Jones potential, erg.
κ = Boltzmann constant, 1.3805 x 10^{-16} erg./°K.
μ^* = viscosity at moderate pressure (0.2 to 5 atm.), centipoises
$\mu^*_{T_c}$ = viscosity at moderate pressure and critical temperature, centipoises
ξ = viscosity parameter, $T_c^{1/6}/M^{1/2} P_c^{2/3}$
σ = collision diameter for Lennard-Jones potential, Å.
$\Omega^{(2,2)*}[T_N]$ = collision integral for Lennard-Jones potential

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The Drag Coefficients of Single Spheres Moving in Steady and Accelerated Motion in a Turbulent Fluid

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Drag coefficients of aerodynamically smooth spheres having a density variation of from 0.252 to 1.91 g./cc. and a diameter variation from 1.56 to 3.21 mm. were obtained for acceleration rates varying from 103.5 ft./sec.^{2} to -30 ft./sec.^{2} and for relative intensities of up to 45%. The particle-to-Eulerian macroscale ratios varied from 0.50 to 0.16, and the diameter-to-Eulerian microscale ratios varied from 10 to 2.

The drag coefficients were found to be a function of the particle Reynolds number and of the relative intensity but not of the acceleration and relative macro-and-microscale variations.

A transition theory for the system investigated is presented, which predicts that the product of the critical Reynolds number and the square of the relative intensity should be a constant; it is supported by the experimental results obtained.

The theoretical analysis of the momentum transfer occurring in multiparticle dilute phase solids-gas flow systems has been hindered by an inability to estimate the fluid drag forces which act on the individual particles owing to their movement relative to the fluid. The fluid drag force is related to the relative motion by the use of a coefficient of drag defined by

$$R = (1/2) C_D A_p \rho_o U_R^2 \qquad (1)$$

Reliable drag coefficients have been evaluated only for bodies in steady motion with respect to turbulent-free flows, and these conditions are not encountered in most solids-gas systems. The drag coefficients indirectly obtained from pressure-drop data (*1, 2, 3*) are invalidated by the assumptions made in their calculation. More direct methods in which the velocities of individual particles have been measured (*4, 5, 6, 7, 8, 9*) suffer in that the fluid velocity and turbulence parameters along the particle trajectory were not determined.

To completely characterize a momentum transfer situation in a turbulent fluid the turbulent parameters

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