The Viscosity of Pure Substances in the Dense Gaseous and Liquid Phases

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Viscosities available in the literature for the gaseous and liquid states of eleven substances have been correlated with reduced density by the use of dimensional analysis and the Abas-zade expression for the residual viscosity to produce a single generalized relationship which is presented both graphically and analytically. The substances are argon, nitrogen, oxygen, carbon dioxide, sulfur dioxide, methane, ethane, propane, *i*-butane, *n*-butane, and *n*-pentane. The properties required for the calculation of viscosity with this relationship are the molecular weight, the critical constants, and the density of the substance at the temperature and pressure considered.

Separate relationships were developed for hydrogen, ammonia, and water which do not follow the consistent behavior of the other substances. Viscosity values for ethylene calculated with the generalized relationship compared favorably with the corresponding experimental values.

In 1944 Uyehara and Watson developed a generalized correlation for the prediction of the viscosity of a pure substance at any temperature and pressure (*79*). Although this correlation has proved to be of extreme utility for industrial calculations, current demands for highly accurate viscosity values in such areas as heat transfer and reactor design necessitate the development of a more exacting method for obtaining viscosities of both gases and liquids.

Recent studies on the prediction of the transport properties of pure substances have been primarily concerned with the viscosity and thermal conductivity of gases at normal pressures (*47, 74, 75*). Using a dimensional analysis approach and viscosity data reported in the literature for fifty-two nonpolar and fifty-three polar gases, Stiel and Thodos (*74, 75*) have developed relationships which can easily be applied for the prediction of the viscosity of any pure gas at moderate pressures (0.1 to 5 atm.). For nonpolar gases the following relationships resulted:

$$\mu^*\xi = 34.0 \times 10^{-5} T_R^{0.94} \quad \text{for} \quad T_R \leqq 1.50 \tag{1}$$

and

$$\mu^*\xi = 17.78 \times 10^{-5} [4.58 T_R - 1.67]^{5/8} \quad \text{for} \quad T_R > 1.50 \tag{2}$$

where $\xi = T_c^{1/6}/M^{1/2}P_c^{2/3}$. For polar gases the viscosity parameter $\mu^*\xi$ was found to depend on both T_R and z_c. This dependence can be expressed as follows for polar substances which exhibit hydrogen bonding:

$$(\mu^*\xi)z_c^{5/4} = [7.55 T_R - 0.55] \times 10^{-5} \tag{3}$$

Similarly for polar gases which do not exhibit hydrogen bonding

$$(\mu^*\xi)z_c^{2/3} = [1.90 T_R - 0.29]^{4/5} \times 10^{-4} \tag{4}$$

Values calculated from Equations (1), (2), (3), and (4) for all the substances investigated were found to compare favorably with the corresponding experimental values. Therefore it would be desirable to utilize a similar approach to develop relationships for the prediction of the viscosity of pure substances at high pressures in both the gaseous and liquid states.

In 1952 Abas-zade (*1*) proposed that the following relationship exists between the residual thermal conductivity of a liquid and its corresponding density:

$$k - k^* = \gamma \rho^s \tag{5}$$

Thodos and co-workers (*23, 33, 50, 66, 68*) have studied the transport properties of several individual substances over a complete range of temperatures and pressures and have found that Equation (5) and a similar expression for the residual viscosity $\mu - \mu^*$ give excellent agreement with experimental data for both gases and liquids. The information obtained from experimental data for the viscosity and thermal conductivity behavior at moderate pressures for these substances was utilized in conjunction with plots of the residual transport properties against density to develop correlations in which the reduced transport properties investigated for each substance were plotted against the reduced temperature for lines of constant reduced pressure. Although these reduced state correlations have proved to be of a high degree of accuracy, they are specific only to the substances investigated and can be extended to substances having similar critical compressibility factors and polarities only if reliable viscosity values are available for these substances for the establishment of their critical viscosities. Therefore in the present study the Abas-zade relationship and available atmospheric viscosities have been utilized in conjunction with a dimensional analysis approach to relate the viscosity of a substance at high pressures in both the gaseous and liquid states to its critical constants, since these constants are more readily available than experimental viscosity data.

DIMENSIONAL ANALYSIS

From kinetic theory considerations Stiel and Thodos (*74*) assumed in the development of Equations (1), (2), (3), and (4) that the viscosity of a gas at moderate pressures was dependent only on the molecular weight, temperature, and critical constants of the substance. The Abas-zade relationship indicates that the residual viscosity $\mu - \mu^*$ should be a function of the density and dimensional constants which are specific to each substance. If by analogy to the previous work these constants are assumed to be the molecular weight and the critical constants of the substance, the application of the Rayleigh method of dimensional analysis to the variables leads to the following development:

$$\mu - \mu^* = \alpha T_c^a P_c^b v_c^c R^d M^e v^f \qquad (6)$$

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

$$a = d \qquad d = d$$

$$b = \frac{1}{2} - d \qquad e = \frac{1}{2}$$

$$c = -\frac{1}{6} - d - f \qquad f = f$$

By the use of these exponents Equation (6) becomes

$$(\mu - \mu^*)\xi = \beta z_c^m \rho_R^n \qquad (7)$$

where $\beta = \alpha/R^{1/6}$, $m = -d - \frac{1}{6}$, $n = -f$, and $\xi = T_c^{1/6}/M^{1/2}P_c^{2/3}$. Since ξ, z_c, and ρ_c are specific constants for each substance, Equation (7) is identical in form to the Abas-zade relationship for residual viscosity.

TREATMENT OF EXPERIMENTAL VISCOSITY DATA

Experimental viscosities for the gaseous and liquid phases available in the literature for fourteen substances were utilized in conjunction with experimental PVT data to establish the constant β and the exponents m and n of Equation (7). The substances considered ranged in complexity from hydrogen to water and included several hydrocarbons. The substances are listed in Table 1 along with their molecular weights, critical constants, and calculated values of ξ. The sources of the experimental viscosities and densities used in this work are also presented.

From the reported viscosities of each substance at high pressures and the corresponding experimental viscosities at atmospheric pressure the group $(\mu - \mu^*)\xi$ was calculated and plotted on log-log coordinates against the reduced density obtained from reported PVT data. Considerable scatter was noted in the experimental viscosity data of some of the investigators for the less dense region. Therefore for this region only those data were utilized which possessed internal consistency and which enabled the relationship between $(\mu - \mu^*)\xi$ and ρ_R to be extended properly into the dense gaseous and liquid regions. The resulting plots produced essentially identical curves for nearly all the substances. Since the critical compressibility factors of these normally behaving substances vary from 0.294 to 0.269, it can be concluded that for these substances the group $(\mu - \mu^*)\xi$ is independent of z_c and therefore $m = 0$ in Equation (7). In Figure 1 a composite curve relating $(\mu - \mu^*)\xi$ to ρ_R is presented for all the substances following this consistent behavior. Although not all the points in Figure 1 fall exactly on the selected curve, these deviations are not serious since the ordinate $(\mu - \mu^*)\xi$ represents a difference between viscosity values; consequently the values resulting from the use of Figure 1 should be closer to the experimental values than this figure indicates. It can be seen from Figure 1 that the exponent n in Equation (7) is not a constant and that the dependence of $(\mu - \mu^*)\xi$ on ρ_R is a com-

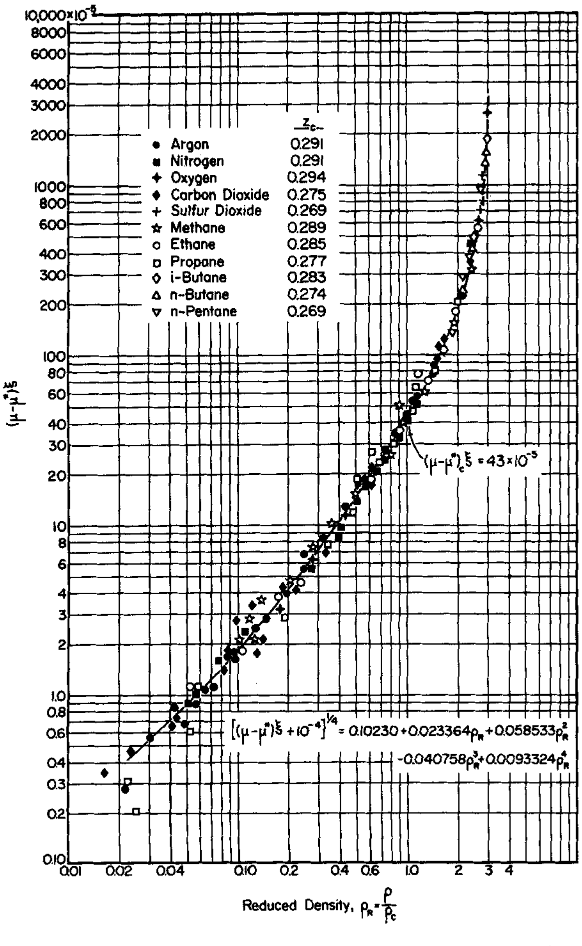

Fig. 1. Relationship between the residual viscosity modulus $(\mu\text{-}\mu^*)\xi$ and reduced density ρ_R for normally behaving substances.

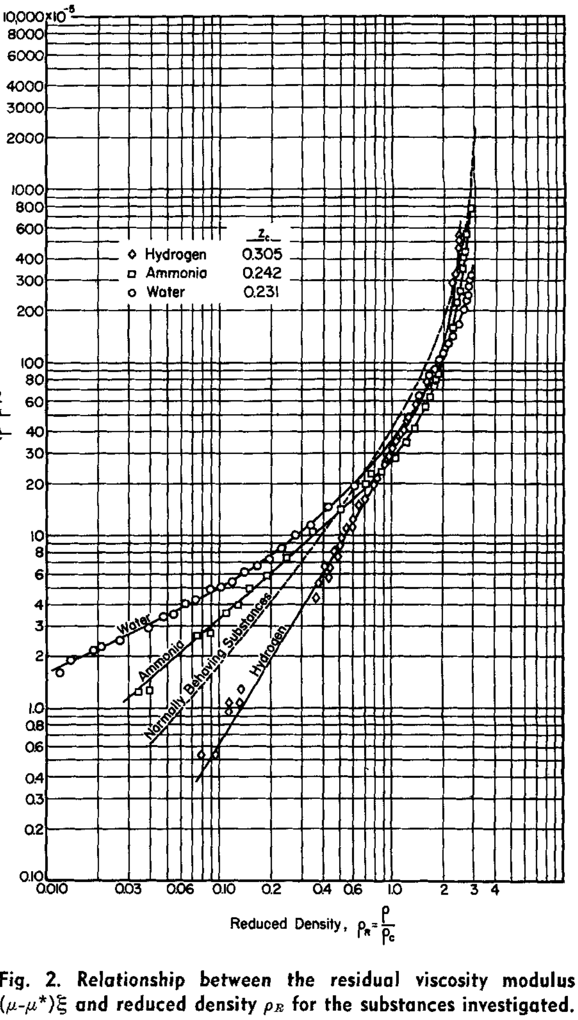

Fig. 2. Relationship between the residual viscosity modulus $(\mu\text{-}\mu^*)\xi$ and reduced density ρ_R for the substances investigated.

plex but continuous function. An IBM-650 computer was used to calculate the coefficients of first, second, third, and fourth degree polynomials with the method of least squares. It was found that the following fourth degree polynomial was necessary to define accurately the analytical relationship over the entire range of reduced densities:

$$[(\mu-\mu^*)\xi+10^{-4}]^{1/4}=0.10230+0.023364\,\rho_R+0.058533\,\rho_R^{2}-0.040758\,\rho_R^{3}+0.0093324\,\rho_R^{4} \quad (8)$$

Alternate analytical expressions for limited regions of reduced densities were developed which are less complex in application than Equation (8). For densities in the range $0.3 \leq \rho_R \leq 2.0$ the following relationship accurately represents the data:

$$(\mu-\mu^*)\xi=[23.12\,e^{1.079\rho_R}-25]\times 10^{-5} \quad (9)$$

Similarly for $\rho_R < 0.3$ the following expression can be employed:

$$(\mu-\mu^*)\xi=11.0\times 10^{-5}\,[e^{1.584\rho_R}-1] \quad (10)$$

The relationship presented in Figure 1 was found to be applicable to all the substances investigated with the exception of hydrogen, ammonia, and water. The relationships between $(\mu-\mu^*)\xi$ and ρ_R for these three substances are presented in Figure 2 along with the curve for the normally behaving substances. The curves of Figure 2 diverge at low densities and increase very rapidly in a narrow band at the high densities.

The abnormal behavior of hydrogen results from its significant quantum deviations (*17*). The relationship for hydrogen can be expressed analytically as follows:

$$[(\mu-\mu^*)\xi+10^{-4}]^{1/4}=0.10616-0.042426\,\rho_R+0.17553\,\rho_R^{2}-0.12295\,\rho_R^{3}+0.028149\,\rho_R^{4} \quad (11)$$

The inconsistent behavior of ammonia and water can undoubtedly be attributed to the significant hydrogen bonding effects exhibited by these substances. For ammonia the resulting analytical relationship is

$$[(\mu-\mu^*)\xi+10^{-4}]^{1/4}=0.10670+0.022655\,\rho_R+0.035749\,\rho_R^{2}-0.032153\,\rho_R^{3}+0.0089998\,\rho_R^{4} \quad (12)$$

Similarly for water the relationship is

$$[(\mu-\mu^*)\xi+10^{-4}]^{1/4}=0.10721+0.040646\,\rho_R+0.0026282\,\rho_R^{2}-0.0054430\,\rho_R^{3}+0.0017979\,\rho_R^{4} \quad (13)$$

Although sulfur dioxide is moderately polar, it can be seen from Figure 1 that in the liquid region its viscosity behavior is identical to that of the nonpolar substances investigated. An additional study is necessary to establish the exact viscosity behavior of polar substances in the dense gaseous and liquid phases. It can be inferred from Equations (3) and (4) resulting from the previous moderate pressure studies that $(\mu-\mu^*)\xi$ might be dependent on z_c for these substances. However it can be concluded that the relationship of Figure 1 accurately represents the viscosity behavior of nonpolar gases for the gaseous and liquid regions and is at least a fairly accurate representation of the viscosity behavior in these regions for polar gases which do not exhibit hydrogen bonding. For nonpolar gases Equations (1) and (2) should be used to calculate values of $\mu^*\xi$, while Equation (4) should be used to calculate this quantity for polar gases which do not exhibit hydrogen bonding.

RESULTS AND CONCLUSIONS

Although Equations (8), (9), and (10) are the best analytical representations of the curve of Figure 1, viscosity values for the dense gaseous and liquid phases are more easily and accurately determined directly from Figure 1 than from these equations. Comparisons between values of $(\mu-\mu^*)\xi$ calculated from Equations (8), (9), and (10) and those obtained from an enlarged plot of Figure 1 are presented below:

	$(\mu-\mu^*)\,\xi\times 10^5$		
ρ_R	Figure 1	Equation (8)	Equations (9) and (10)
0.10	1.78	2.24	1.89
0.20	4.20	4.12	4.10
0.35	8.70	8.13	8.73
0.50	14.5	13.7	14.6
0.70	24.5	23.8	24.2
1.00	43.0	44.5	43.0
1.25	64.0	66.2	64.0
1.50	91.0	92.0	91.8
1.75	126	125	128
2.00	175	172	175
2.25	260	252	
2.50	405	408	
2.75	740	750	
3.00	1,600	1,570	

Since Equations (9) and (10) are easier to apply than Equation (8), these comparisons indicate that their use is preferable for reduced densities $\rho_R < 2.0$. The use of these equations for the calculation of the viscosity of a nonpolar gas is illustrated in the following example.

Example

Calculate the viscosity of ethylene at 50°C. and 89.3 atm. At these conditions the density of ethylene is 0.184 g./cc.

For ethylene $\xi=\dfrac{T_c^{1/6}}{M^{1/2}P_c^{2/3}}$

$$=\frac{(282.4)^{1/6}}{(28.05)^{1/2}\,(50.0)^{2/3}}=0.0356$$

At $t = 50$°C. the reduced temperature is $T_R = (273.2 + 50)/282.4 = 1.144$, and

TABLE 1. BASIC CONSTANTS AND SOURCES OF VISCOSITY DATA

	Critical constants					Sources of viscosity data (dense gaseous and liquid phases)	Sources of PVT data (dense gaseous and liquid phases)
	T_c, °K.	P_c, atm.	ρ_c, g./cc.	z_c	ξ		
Hydrogen	33.3	12.80	0.031	0.305	0.230	*21, 22, 29, 45, 80, 65*	*65*
Argon	151.2	48.00	0.531	0.291	0.0276	*28, 34, 43, 59*	*25*
Nitrogen	126.2	33.5	0.311	0.291	0.0407	*28, 44, 58, 59*	*11*
Oxygen	154.8	50.1	0.430	0.294	0.0301	*20, 21, 59, 80*	*11*
Carbon dioxide	304.2	72.85	0.467	0.275	0.0224	*15, 34, 43, 73*	*32*
Sulfur dioxide	430.7	77.8	0.524	0.269	0.0189	*2, 73*	*42*
Ammonia	405.5	111.3	0.235	0.242	0.0284	*12, 34, 69, 73*	*24*
Water	647.4	218.3	0.32	0.231	0.0192	*26, 56, 67, 70, 71, 78*	*77*
Methane	191.1	45.8	0.162	0.289	0.0464	*3, 15, 36, 76*	*41*
Ethane	305.5	48.2	0.203	0.285	0.0357	*3, 72, 76*	*4, 8, 9, 30, 35, 40, 46, 52, 54, 62, 81*
Propane	370.0	42.0	0.220	0.277	0.0334	*3, 15, 38, 60, 72, 76*	*5, 6, 10, 13, 16, 18, 19, 48, 55, 61*
i-Butane	408.1	36.0	0.221	0.283	0.0327	*38, 64*	[*]
n-Butane	425.2	37.5	0.228	0.274	0.0321	*38, 64, 76*	*7, 14, 16, 18, 31, 49, 51, 57, 63*
n-Pentane	469.8	33.3	0.232	0.269	0.0317	*39, 53*	[*]

[*] Density data for *n*-butane used for these substances.

the reduced density is $\rho_R = 0.184/0.227 = 0.811$. Therefore from Equation (1)

$$\mu^*\xi = 34.0 \times 10^{-5}\,(1.144)^{0.94} = 38.6 \times 10^{-5}$$

From Equation (9)

$$(\mu - \mu^*)\,\xi = [23.12\, e^{1.079\,(0.811)} - 25] \times 10^{-5} = 3.05 \times 10^{-5}$$

Consequently,

$$\mu\,\xi = (30.5 + 38.6) \times 10^{-5} = 69.1 \times 10^{-5}$$

$$\text{and } \mu = \frac{69.1 \times 10^{-5}}{0.0356} = 1{,}941 \text{ centipoises}$$

For *this* temperature and pressure Comings, Mayland, and Egly (*15*) report a viscosity value of $\mu = 1{,}975$ centipoises. Thus the calculated value compares well with the corresponding experimental value. Similarly viscosities were calculated for ethylene at five other conditions of temperature and pressure for which Comings et al. report experimental values. The six calculated viscosities produced an average deviation of 4.0% from the corresponding experimental values. In these calculations density values were obtained from a reduced density plot for *n*-butane. Thus the use of experimental density data for ethylene and values of $(\mu - \mu^*)\,\xi$ obtained directly from Figure 1 should result in even lower deviations between calculated and reported viscosities.

For the viscosity at the critical point Hougen and Watson (*27*) present the following equation developed from the work of Licht and Stechert (*37*):

$$\mu_c\,\xi = 77.0 \times 10^{-5} \qquad (14)$$

From Figure 1 at $\rho_R = 1.0$, $(\mu - \mu^*)_c\,\xi = 43.0 \times 10^{-5}$. Also from Equation (1) $\mu^*\,\xi = 34.0 \times 10^{-5}$ at $T_R = 1$. Thus the value of $\mu_c\,\xi$ resulting from this study is identical to that presented in Equation (14).

NOTATION

a,b,c,d,e,f = exponents in Equation (6)
k = thermal conductivity, cal./sec. cm. °K.
k^* = thermal conductivity at normal pressures, cal./sec. cm. °K.
m = exponent in Equation (7)
M = molecular weight
n = exponent in Equation (7)
P = pressure, atm.
P_c = critical pressure, atm.
P_R = reduced pressure, P/P_c
R = gas constant
s = exponent in Equation (5)
T = temperature, °K.
T_c = critical temperature, °K.
T_R = reduced temperature, T/T_c
v = molar volume, cc./g.-mole
v_c = critical molar volume, cc./g.-mole
z_c = compressibility factor at the critical point, P_cv_c/RT_c

Greek Letters

α = constant in Equation (6)
β = constant in Equation (7)
γ = constant in Equation (5)
μ = viscosity, centipoises
μ^* = viscosity at normal pressures (0.1 to 5 atm.), centipoises
μ_c = viscosity at the critical point, centipoises
ξ = viscosity parameter, $T_c^{1/6}/M^{1/2}P_c^{2/3}$
ρ = density, g./cc.
ρ_c = density at the critical point, g./cc.
ρ_R = reduced density, ρ/ρ_c

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Manuscript received August 15, 1961; revision received November 13, 1961; paper accepted November 16, 1961. Paper presented at A.I.Ch.E. New York meeting.

A Note on Latent Heat Transport in Nucleate Boiling

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A simple one-dimensional transport calculation is made which indicates that latent heat transport, by simultaneous evaporation and condensation at different portions of the bubble surfaces, can account for the major portion of the total heat flux in the neighborhood of the departure from subcooled nucleate boiling (burnout). This differs from the previously held view that the stirring action of the bubbles accounted for most of the heat flow in subcooled nucleate boiling. Other evidence is discussed which favors the latent heat transport theory.

Until recently it seemed quite probable that latent heat transport played a minor role in nucleate boiling heat transfer. This belief was based on observations of the rate of vapor production in subcooled nucleate boiling of water at atmospheric pressure (*1* to *3*). These measurements indicated that only 1 to 2% of the total heat flux could be accounted for by the latent heat content of the vapor evolved at the surface. Tentatively it was concluded that the bubbles acted principally as stirring devices which increased the turbulent intensity within the boundary layer next to the heating surface. This would imply that it would make no difference whether the bubbles were filled with vapor or with inert gas. Indeed it has been shown (*4*) that nonboiling heat transfer coefficients are increased by a factor of 2 or 3 by generating inert gas bubbles at the heating surface, although the resulting heat fluxes, even at the highest generation rate of inert gas bubbles, were in the range of the lower limit of nucleate boiling.

This leads to the suspicion that some other mechanism, such as transport by net flow of vapor away from the heating surface, may dominate near the upper limit of nucleate boiling. For this to be true there would have to be significant transport by simultaneous vaporization near the face of the bubbles and condensation at the top. This further implies that the heat transfer coefficients at the surface of the steam bubbles must be considerably greater than those calculated by Gunther and Kreith (*2*), who assumed that the bubbles were surrounded by a stationary film of liquid. The requirements for latent heat transport to be significant were discussed by Snyder and by Bankoff (*20*).

Direct experimental evidence concerning these points has been quite difficult to obtain because of the statistical nature of the boiling transfer and because of the small time and length scales for the temperature variations in the liquid. Recently however some direct measurements of the heat transfer coefficient at the surface of a rapidly growing and collapsing steam bubble in a turbulent subcooled liquid stream (*5*) have been presented. These coefficients ranged from 13,000 to the phenomenally high figure of over 300,000 B.t.u./hr./sq. ft./°F. and indicate that at least several maximum bubble volumes of steam flow through the bubble during its lifetime. From these measurements it was shown that latent heat transport becomes the dominant mechanism near the upper limit of nucleate boiling in the subcooled nucleate boiling experiments of water at atmospheric pressure performed by Gunther (*1*).

The purpose of this note is to point out, apart from this direct evidence, several other pieces of indirect evidence and to present some simple calculations which indicate that latent heat transport dominates at high fluxes. This means that as the bubbles become closely packed, the principal heat transfer into the bulk stream occurs from the bubble surfaces, rather than from the portions of the solid surface between the bubbles. This seems quite reasonable on the basis of physical intuition.

Some significant results can be obtained simply by examining the shape of the curves of a maximum heat flux in nucleate boiling vs. reduced pressure as reported for water (*6*) and several organic liquids (*7*). These