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A Simple and Accurate Method for Calculating Viscosity of Gaseous Mixtures

By Thomas A. Davidson

UNITED STATES DEPARTMENT OF THE INTERIOR

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BUREAU OF MINES

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CONTENTS

ILLUSTRATIONS

TABLE

UNIT OF MEASURE ABBREVIATIONS USED IN THIS REPORT

AAD	average absolute deviation	RMS	root-mean-square deviation
°C	degree Celsius		

A SIMPLE AND ACCURATE METHOD FOR CALCULATING VISCOSITY OF GASEOUS MIXTURES

By Thomas A. Davidson[1]

ABSTRACT

The Bureau of Mines Helium Field Operations has developed a simple and accurate method for calculating the viscosity of gas mixtures. Only the composition of the mixture and the molecular weights and viscosities of the pure components in the mixture are required. The momentum fraction of a mixture is calculated from the composition. The fluidity is calculated as a quadratic function of the momentum fractions. The efficiency factor for transfer of momentum in collisions between bodies of different masses is derived and used with one empirical constant. The viscosity is the reciprocal of the fluidity. Results of the method are compared with 752 reported viscosities for 40 dilute binary systems at temperatures from -78 to 276.9° C. Using this method the average absolute deviation is 1.29%, the root-mean-square deviation is 1.99%, and the average deviation is -0.84%.

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INTRODUCTION

Accurate knowledge of the transport properties of fluids is essential for control of industrial processes. Many analytical and measurement devices are strongly dependent on the viscosity of a fluid or the thermal conductivity, which is proportional to the product of the viscosity and the heat capacity. Common examples include flowmeters, thermistors, and thermal conductivity detectors. In most applications, the direct measurement of viscosity is impractical, and a calculated value must be used. The accuracy of measurements made with such devices is limited by the accuracy of the determination of the viscosity of a gas mixture.

The Bureau of Mines has developed a method of calculating the viscosity of a mixture of gases that is both simple and accurate. Simplicity is measured by the amount of information needed to perform a calculation. Accuracy is measured by the root-mean-square (RMS) deviation of calculated results from experimental results.

VISCOSITY EQUATIONS

There are a variety of equations available for the estimation of the viscosity (μ_{mix}) of a mixture of gases. These are separated into three major groups: sums of partial viscosities of pure gases, sums of partial viscosities including viscosities for interacting components, and viscosities of generalized fluids based on the principle of corresponding states.

PARTIAL VISCOSITY SUMS

These equations express the viscosity of a mixture as a sum of partial viscosities. This approach was originally taken by Graham (*1*).[2] The assumption is made that the total momentum transferred between adjacent layers in a fluid is a sum of the momenta transferred by each component. On this basis, partial viscosities are added.

The equations in this group lack separate terms for the interaction of dissimilar molecules. The interaction is typically accommodated as an adjustment of the values for pure gas viscosities. The usual form of this adjustment is one or more added terms in the denominator.

The simplest such model is Graham's model, in which the viscosity of a mixture is approximated by summing the products of the viscosities (μ_i) of the individual components and their mole fractions (x_i):

$$\mu_{mix} = \Sigma_i(x_i \cdot \mu_i). \tag{1}$$

This model is adequate for many mixtures in which the components have nearly the same molecular weight. When the molecular weight ratio is different from unity, mixtures can deviate from this behavior greatly.

Wilke (*2*) chose to use an equation derived from simple kinetic theory that is easily extended to multicomponent systems. It requires the evaluation of a complicated coefficient (ϕ_{ij}) for each pair of components in a mixture. The evaluation requires only the viscosities and the molecular weights (M_i) of the individual components.

$$\phi_{ij} = \frac{\left[1 + \left(\frac{\mu_i}{\mu_j}\right)^{1/2} \cdot \left(\frac{M_j}{M_i}\right)^{1/4}\right]^2}{\frac{4}{\sqrt{2}} \cdot \left[1 + \frac{M_i}{M_j}\right]^{1/2}}. \tag{2}$$

Golubev (*3*) produced graphical estimates for Wilke's ϕ_{ij} coefficients. These coefficients are then used with the mole fraction in the calculation of the viscosity of the mixture:

$$\mu_{mix} = \Sigma_i \frac{x_i \cdot \mu_i}{x_i + \Sigma_{j \neq i} (x_j \cdot \phi_{ij})}. \tag{3}$$

Each term represents a partial viscosity for each component. The denominator of each term represents an effort to account for interactions between differing molecules by adjusting the partial viscosity for each species directly.

Wilke's equation is an extension of the equation derived by Schmick (*4*) for binary mixtures. Equation 2 and 3 can produce significant errors for real mixtures.

The equation of Herning and Zipperer (*5*) calculates partial viscosities without the evaluation of Wilke's coefficients:

$$\mu_{mix} = \frac{\Sigma\left(\mu_i \cdot x_i \cdot \sqrt{M_i}\right)}{\Sigma\left(x_i \cdot \sqrt{M_i}\right)}. \tag{4}$$

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[2] Italic numbers in parentheses refer to items in the list of references preceding the appendix at the end of this report.

This is a sum of partial viscosities, weighted by the square root of the molecular weight for each species. These weights are proportional to the momenta.

The reported accuracy of equation 4 for hydrocarbon mixtures is 1.5% average deviation and 5% maximum deviation, except for hydrogen-rich mixtures. Equation 4 is not recommended for use with mixtures containing significant hydrogen.

The problem of hydrogen is handled by Brokaw (*6*) in a method that requires effective collision diameters for all molecules and pairs, and dipole moments for polar constituents. Brokaw reports an average deviation of 0.7% and a maximum of 3.7% for 25 gas pairs and 280 mixtures. For binary mixtures, the average deviation is 0.6% and the maximum is 2.5%.

With the exception of Brokaw's equation, this group of equations is characterized by relative simplicity of use and limited accuracy.

VISCOSITY OF INTERACTION

This group of equations uses explicit terms for the interactions of dissimilar molecules. The form of these equations is that of a weighted average of the viscosities of the pure components, including an effective viscosity of interaction μ_{ij}.

Chapman and Cowling (*7*) give a complicated equation, involving the ratio of an eight-term polynomial to a six-term polynomial, that they call a first approximation for a binary gas mixture. Several terms of the equation are dependent on the evaluation of integrals involving the force law for molecular interactions. The application of this equation to a real mixture is not trivial, and the extension to multicomponent systems is not clear.

Chapman (*8*) presents a simplified form of the Chapman and Cowling equation which has four constants (μ_{12}, a_1, a_2, and b) for each pair of gases. These constants depend on the molecular masses, the law of force, and the temperature. For polyatomic gases, even an approximate force law may be unavailable. For this reason, these constants may be considered empirical.

$$\mu_{mix} = \frac{\mu_1 \cdot a_1 \cdot x_1^2 + \mu_{12} \cdot x_1 \cdot x_2 + \mu_2 \cdot a_2 \cdot x_2^2}{a_1 \cdot x_1^2 + b \cdot x_1 \cdot x_2 + a_2 \cdot x_2^2}. \qquad (5)$$

This form was found to agree with data by Schmitt (*9*) for argon-helium and hydrogen-oxygen mixtures to about 1%. The evaluation of the multiple constants makes use of this equation difficult, especially for multicomponent mixtures.

Kennard (*10*) simplifies equation 5 even further. For practical purposes he reduces (5) to a quadratic in mole fraction with one empirical constant μ_{12} for each pair of gases:

$$\mu_{mix} = \mu_1 \cdot x_1^2 + \mu_{12} \cdot x_1 \cdot x_2 + \mu_2 \cdot x_2^2. \qquad (6)$$

The equations in this group are generally the most accurate, but the application is hindered by requirements for determining multiple constants or the difficulty of extending the equations to multicomponent systems.

CORRESPONDING STATES

Methods based on the principle of corresponding states calculate properties for a hypothetical pure fluid based on estimated critical constants and other parameters for a mixture. The computer program of Ely and Hanley (*11*) applies this principle to relate a mixture to methane as a reference fluid. The viscosity correlation used for the reference fluid involves 20 coefficients. This program has an accuracy for methane of over 3% average absolute deviation (AAD). This can be considered a lower limit of the uncertainty expected for this program when applied to estimation of the viscosity of a mixture of gases. The authors report an AAD of 8.4% for 36 pure fluids of the 61 that they included in their data base. They also report an average absolute deviation of 6.95% for 455 mixtures representing 26 binary systems of hydrocarbons.

Addition of a pure fluid to the data base of the Ely and Hanley program requires 13 constants, including 3 critical constants, the molecular weight, the normal boiling point, the first 7 coefficients of a Taylor series in temperature for the heat capacity, and a parameter for correlating the PVT surface of the gas to that of methane. These latter constants may be difficult to obtain.

Methods based upon the principle of corresponding states have an inherent drawback. In any selected temperature and pressure range the accuracy suffers from the effort to fit data simultaneously in other ranges. The viscosities of the pure components of a mixture at the temperature and pressure of interest are usually known to the desired accuracy. An equation that describes viscosity of an intermediate range of mixtures at a fixed temperature and pressure can be expected to be more accurate than one that describes viscosity of a specified mixture over wide ranges of temperature and pressure.

Reid, Prausnitz, and Poling (*12*) compared several methods for calculating gas mixture viscosities, including those of Wilke (equation 3) and of Herning and Zipperer (equation 4). They used data for 10 gas pairs, represented by 34 binary mixtures. The pairs were selected to represent a broad range of gases, including hydrogen, ammonia and other polar molecules, light hydrocarbons, oxides, halogenated gases, nitrogen, and n-pentane. The RMS deviations for equations 3 and 4 respectively were 4.68% and 6.17%. For two methods based on corresponding-states calculations by Lucas, and by Chung and co-workers, Reid found RMS deviations of 5.29% and 6.45% respectively.

DEVELOPMENT OF THE EQUATION

To relate the viscosity (μ) of a gaseous mixture to its composition, a mixing equation was developed through logical analysis of the form that it should take, rather than from purely theoretical premises.

VISCOSITY VERSUS FLUIDITY

Viscosity could be described as the resistance of a fluid to the transport of momentum. Consider a shear applied to a fluid by an apparatus such as a fan. A given shear will create less disturbance at a given distance from the point of application if the fluid is more viscous. The adding of partial viscosities pioneered by Graham is an approximation based on empirical results for very few systems. There is no physical basis for assuming that partial viscosities are additive.

Maxwell's equation (*13*) shows gas viscosity is the product of the mean free path (l) of the molecules, the density (d), and the average velocity (v) with a proportionality constant (C):

$$\mu = C \cdot l \cdot d \cdot v. \tag{7}$$

Results of different kinetic theory treatments for the numeric value of C range from 0.30967 to 0.5 (*7*, p. 218; *14*, *15*, *16*).

The mean free path of a molecule may be defined as the average velocity divided by the frequency (Z) of collisions with other molecules:

$$l = \frac{v}{Z}. \tag{8}$$

The mean free path is not a simple function of the state or composition of the fluid.

The overall collision frequency is a sum of terms for collisions (Z_{ij}) between all possible pairs of molecules:

$$Z = \Sigma_{i,j} Z_{ij}. \tag{9}$$

The collision frequency Z_{ij} for two molecules is proportional to the product of their concentrations. Z is therefore a sum of second degree terms in concentration. Z is thus more convenient than l for treatment of the properties of mixtures.

The reciprocal of the viscosity is called the fluidity (f). Fluidity may be described as the ability of a fluid to transport momentum. This quantity is directly related to the overall collision frequency. Combining equations 7, 8, and 9 to express fluidity produces the equation

$$f = \frac{\Sigma_{i,j} Z_{ij}}{C \cdot d \cdot v^2}. \tag{10}$$

In a mixture of gases at thermal equilibrium, the average kinetic energy (K) of each molecule is related to Boltzmann's constant (k) and absolute temperature (T) by

$$K = \frac{3 \cdot k \cdot T}{2}. \tag{11}$$

The kinetic energy of a molecule of mass (m) is given by

$$K = \frac{m \cdot v^2}{2}. \tag{12}$$

If V represents the molar volume of the gas, the density is m/V, and the denominator on the right of equation 10 becomes

$$C \cdot d \cdot v^2 = \frac{3 \cdot C \cdot k \cdot T}{V}, \tag{13}$$

which is proportional to the temperature and independent of the composition of the mixture.

The fluidity of a mixture of gases contains additive contributions based on molecular collision frequencies. These contributions can be conveniently expressed in terms of the concentrations and the properties of the components. A mixing equation for fluidity was developed. When the fluidity of a mixture is determined, the viscosity follows as the reciprocal of the fluidity.

MOMENTUM FRACTION

Mole fraction is often the variable of choice for expressing concentrative properties of mixtures. Viscosity is not a concentrative property. Viscosity and fluidity are associated with the transport of momentum from one point in the fluid to another. This suggests that the fluidity of a mixture of gases should depend upon the momenta of the separate components rather than just their concentrations. The momentum fraction is used instead of the mole fraction or concentration as the main compositional variable.

The momentum is related to the molecular weight and temperature. The momentum (p) of a molecule is given by

$$p = m \cdot v. \tag{14}$$

The momentum is then related to the temperature using equations 11 and 12:

$$p = \sqrt{(3 \cdot m \cdot k \cdot T)}\,. \tag{15}$$

At thermal equilibrium, the temperatures of all components of a mixture are the same. The average momentum of each component is thus proportional to the square root of its molecular weight. The Herning-Zipperer equation given above is equivalent to a momentum-weighted sum of partial viscosities.

The momentum fraction (y_i) of a component is defined as that fraction of the total momentum within a mixture that is associated with a particular component. If the mole fraction of a component is represented as x_i, the amount of momentum associated with that component is $x_i \cdot \sqrt{(3 \cdot M \cdot k \cdot T)}$, and the momentum fraction associated with a component of molecular weight M_i becomes

$$y_i = \frac{x_i \cdot \sqrt{M_i}}{\Sigma_i \left(x_i \cdot \sqrt{M_i} \right)}\,. \tag{16}$$

The following treatment of the fluidity and the viscosity of mixtures of gases uses this variable.

The frequency of collisions between two types of molecules Z_{ij} can be expressed in terms of the product of their momentum fractions

$$Z_{ij} \propto y_i \cdot y_j\,. \tag{17}$$

The fluidity of a pure gas therefore depends upon the square of the average momentum of the molecules. The fluidity mixing equation is a quadratic in momentum fraction.

The following equation, with scalar mixing coefficients B_{ij}, is used to express fluidity for a gas mixture

$$f = \Sigma_{i,j}(y_i \cdot y_j \cdot f_{ij} \cdot B_{ij})\,. \tag{18}$$

This equation is a mixing equation of second degree in the momentum fraction.

FLUIDITY OF INTERACTION

When $i \neq j$, f_{ij} represents an effective fluidity of interaction between the ith and jth component. When $i = j$, f_{ij} represents the fluidity of the pure gas i under the same conditions of temperature and pressure as those of the mixture. The fluidity of interaction is estimated with the quantity

$$f_{ij} = \frac{1}{\sqrt{\mu_i} \cdot \sqrt{\mu_j}}\,. \tag{19}$$

The scalar coefficients B_{ij} are related to the efficiency with which momentum is transferred between molecules in a collision. If equation 18 is to reduce to an identity for a pure gas, B_{ij} must be unity when $i = j$. In cases where $i \neq j$, B_{ij} will depend on the weights of the molecules in the collision.

EFFICIENCY OF MOMENTUM TRANSFER

A dilute gas may be defined as a gas in which potential energy due to intermolecular forces becomes insignificant compared to translational kinetic energy at some point in the path of a molecule between collisions, Thus the mean free path in a dilute gas is no less than twice the effective range of the intermolecular forces. Under this constraint, simultaneous collisions between three or more bodies are sufficiently rare that they do not contribute to the overall properties of the system.

The efficiency of the transfer of momentum in a collision between two bodies depends upon their masses. The exact form of this dependence can be found with elementary mechanics (*17*).

Consider the elastic collision of two spherical bodies of mass m_1 and m_2 respectively, with initial velocities v_{1i} and v_{2i}. While the bodies are within the effective range of the forces of interaction, accelerations induced by these forces alter the details of their trajectories. Beyond the range of these forces the trajectories are asymptotic to the trajectories required to satisfy conservation of momentum and energy.

Components of velocity perpendicular to the line between the centers of the bodies can be resolved into contributions to the angular and translational momenta of the two-body system. These momenta are unaltered by collisions. Only the radial momenta of the two bodies are altered by a collision. The components of the velocities parallel to the line between the centers are sufficient to describe the transfer of momentum.

The equations for conservation of momentum and energy for this system relate the masses, initial velocities, and final velocities (v_{1f}) and (v_{2f}):

$$m_1 \cdot v_{1i} + m_2 \cdot v_{2i} = m_1 \cdot v_{1f} + m_2 \cdot v_{2f}\,. \tag{20a}$$

$$\frac{m_1 \cdot v_{1i}^2}{2} + \frac{m_2 \cdot v_{2i}^2}{2} = \frac{m_1 \cdot v_{1f}^2}{2} + \frac{m_2 \cdot v_{2f}^2}{2}\,. \tag{20b}$$

v_{1f} and v_{2f} can be found by solving these equations algebraically:

$$v_{1f} = \frac{(m_1 - m_2)}{(m_1 + m_2)} \cdot v_{1i} + \frac{2 \cdot m_2}{(m_1 + m_2)} \cdot v_{2i}. \quad (21a)$$

$$v_{2f} = \frac{(m_2 - m_1)}{(m_1 + m_2)} \cdot v_{2i} + \frac{2 \cdot m_1}{(m_1 + m_2)} \cdot v_{1i}. \quad (21b)$$

Multiplying equation 21a by m_1 yields an expression for the final momentum of body 1:

$$m_1 \cdot v_{1f} = \frac{(m_1 - m_2)}{(m_1 + m_2)} \cdot m_1 \cdot v_{1i} + \frac{2 \cdot m_1 \cdot m_2}{(m_1 + m_2)} \cdot v_{2i}. \quad (22)$$

The final momentum of body 1 is the sum of two terms. The first term represents what remains of its initial momentum after transferring some momentum to body 2. The second term represents the portion of the momentum of body 2 that body 1 received in the collision.

The efficiency (E_{21}) with which body 2 transfers its momentum to body 1 is found by dividing the second term by the original momentum of body 2:

$$E_{21} = \frac{2 \cdot m_1}{(m_1 + m_2)}. \quad (23)$$

Repeating this with (21b) and m_2 produces

$$E_{12} = \frac{2 \cdot m_2}{(m_1 + m_2)} \quad (24)$$

for the efficiency with which body 1 transfers momentum to body 2.

The mean efficiency for this interaction, weighted for the momenta of the bodies, is

$$E_{1,2} = \frac{y_1 \cdot E_{12} + y_2 \cdot E_{21}}{y_1 + y_2}. \quad (25)$$

This can be simplified using equation 16 to the following expression:

$$E_{1,2} = \frac{2 \cdot \sqrt{m_1} \cdot \sqrt{m_2}}{(m_1 + m_2)}, \quad (26)$$

which has a maximum value of unity for bodies of equal mass. It is also nonnegative and is symmetrical with respect to interchange of indices. We should expect this for our mixing coefficients.

The coefficient B_{ij} is assumed to be dependent upon $E_{i,j}$ by a simple power law, with the exponent (A) determined empirically:

$$B_{i,j} = E_{i,j}^{A}. \quad (27)$$

This exponent is assumed to be the same for all pairs and is the only empirical constant in the mixing equation.

FLUIDITY OF A MIXTURE

Including equation 27 for the effect of dissimilar masses on the efficiency of momentum transfer refines the expression in equation 18 for the fluidity of a gas mixture.

$$f = \Sigma_{i,j} \left[\frac{y_i \cdot y_j}{\sqrt{\mu_i} \cdot \sqrt{\mu_j}} \cdot E_{i,j}^{A} \right]. \quad (28)$$

The symmetry of the expression is such that the ij terms are equal to the ji terms.

For a pure gas, equation 28 reduces to the usual definition of fluidity:

$$f = \frac{1}{\mu}. \quad (29)$$

Equation 28 is easily applied to mixtures of any number of components. It contains one empirical constant which remains the same for all mixtures of gases.

In the case of a simple binary mixture, equation 28 assumes the form

$$f_{12} = \frac{y_1^2}{\mu_1} + 2 \cdot \frac{y_1 \cdot y_2}{\sqrt{\mu_1} \cdot \sqrt{\mu_2}} \cdot E_{1,2}^{A} + \frac{y_2^2}{\mu_2}. \quad (30)$$

Using equation 16, equation 28 can be recast in mole fractions as a ratio of quadratics. For a binary mixture it is comparable to equation 5, the simplified Chapman equation. However the four empirical constants per binary mixture required for equation 5 are replaced by functions of the pure component masses and viscosities. Equation 28 equals the simplified Chapman equation in accuracy, as will be shown below.

The only information required to apply equation 28 to a mixture is the composition, along with the molecular weight and viscosity of each component at the temperature and pressure of interest.

RESULTS

The value of the empirical constant A was estimated using reported viscosities for 35 gas pairs. The quantity $B_{i,j}$ was calculated from data for 164 mixtures using equation 18. The logarithm of $B_{i,j}$ was then plotted against the logarithm of $E_{i,j}$ (fig. 1). The RMS deviations were found to be relatively insensitive to small changes in A. The miminum RMS deviation for the predicted viscosities is 1.28%, occurring at a value of 0.375 for A. The value 1/3 yields an RMS deviation of 1.45%. The plotted slope of 1/3 was selected as simple and consistent with the data.

The dependence of $B_{i,j}$ on $E_{i,j}$ suggests that the interaction of molecules of different masses does not conform to the conditions necessary for application of the principle of corresponding states. Transfer of momentum during collisions between molecules depends specifically on the masses of the individual molecules. Viscosity is inherently composition dependent and is not well modeled by a hypothetical pure fluid.

Equation 28 was tested against 752 experimental values reported by various workers for 40 gas pairs. Reported temperatures for the viscosities ranged from -78 to 276.9° C. Pressures were not generally reported but are assumed to be low enough for the gases to be treated as dilute. Table 1 summarizes the comparison of calculated and experimental values for each mixture. Data for three gas pairs were obtained from two different sources each. The average deviation (bias) is -0.84%. The AAD is 1.29%. The overall RMS error is 1.99%. The maximum RMS error found for a given system is 5.82%, for the neon-carbon dioxide pair.

The calculated values were then correlated with experimental values for the 752 data points, as shown in figure 2. The measured viscosities ranged from 7.31 to 43.10 micropascal seconds (73.1 to 431.0 micropoises).

Three of the five gas pairs of table 1 with the highest RMS error include ammonia as one component. Calculations for the ammonia-ethylene system with equation 28 provide excellent agreement with 1.58% RMS error (1.45% AAD).

Wilke reported difficulty with fitting his equation to the hydrogen-argon system. Equation 3 fits the hydrogen-argon data of Trautz and Binkele (*23*) with 4.73% RMS error (4.33% AAD). Equation 28 fits the same data with 1.58% RMS error (-1.43% AAD).

The Herning-Zipperer method (equation 4) has a reported 1.5% average deviation for hydrocarbons. Equation 28 fits four pairs of hydrocarbon gases with 0.33% RMS error (0.16% AAD). Data were examined for 15 pairs that included hydrogen as 1 component. Equation 28 fits 12 of these pairs within 2% RMS error (1.30% AAD). The Herning-Zipperer equation is not recommended for use with mixtures containing significant hydrogen.

Table 1.—Accuracy of equation 28 compared with reported viscosities of various binary gas mixtures

Binary mixture	Reference	Points used	RMS error, %	Binary mixture	Reference	Points used	RMS error, %
CH_4/C_2H_6	20	20	0.11	H_2/O_2	21	24	1.08
N_2/CO	21	24	.12	N_2/CO_2	19	9	1.16
C_3H_6/C_3H_8	3	12	.13	H_2/C_3H_8	22	24	1.23
CO/O_2	21	18	.20	Ne/Ar	18	5	1.30
He/Ne	23	11	.22	C_2H_4/CO	21	24	1.31
N_2/O_2	21	19	.22	H_2/C_3H_6	3	36	1.39
N_2O/CO_2	22	19	.25	H_2/CO_2	22	24	1.47
C_2H_6/C_3H_8	20	12	.29	H_2/Ar	23	16	1.58
CH_4/C_3H_8	20	12	.33	NH_3/C_2H_4	3	24	1.58
He/Ne	18	10	.38	H_2/2-C_4H_8	3	32	1.82
He/Ar	24	12	.39	C_2H_4/O_2	21	15	2.10
N_2/NO	3	6	.40	H_2/N_2O	22	16	2.22
H_2/N_2	25	24	.55	N_2O/C_3H_8	22	16	2.28
He/Ar	23	8	.56	Ar/CO_2	19	12	2.64
H_2/C_2H_4	26	33	.59	CO_2/C_3H_8	22	16	2.80
H_2/He	23	12	.65	He/Kr	19	16	3.74
H_2/CH_4	20	16	.67	NH_3/N_2	3	20	3.80
H_2/CO	25	24	.82	H_2/NH_3	3	23	4.39
Ne/Ar	23	12	.90	H_2/Ne	23	16	4.49
He/N_2	19	15	.92	NH_3/O_2	3	15	4.91
N_2/C_2H_4	21	24	.99	Ne/CO_2	27	10	5.82
H_2/C_2H_6	20	16	1.00				

Total points = 752, RMS error = 1.99%, AAD = 1.29%, bias = -0.84%.

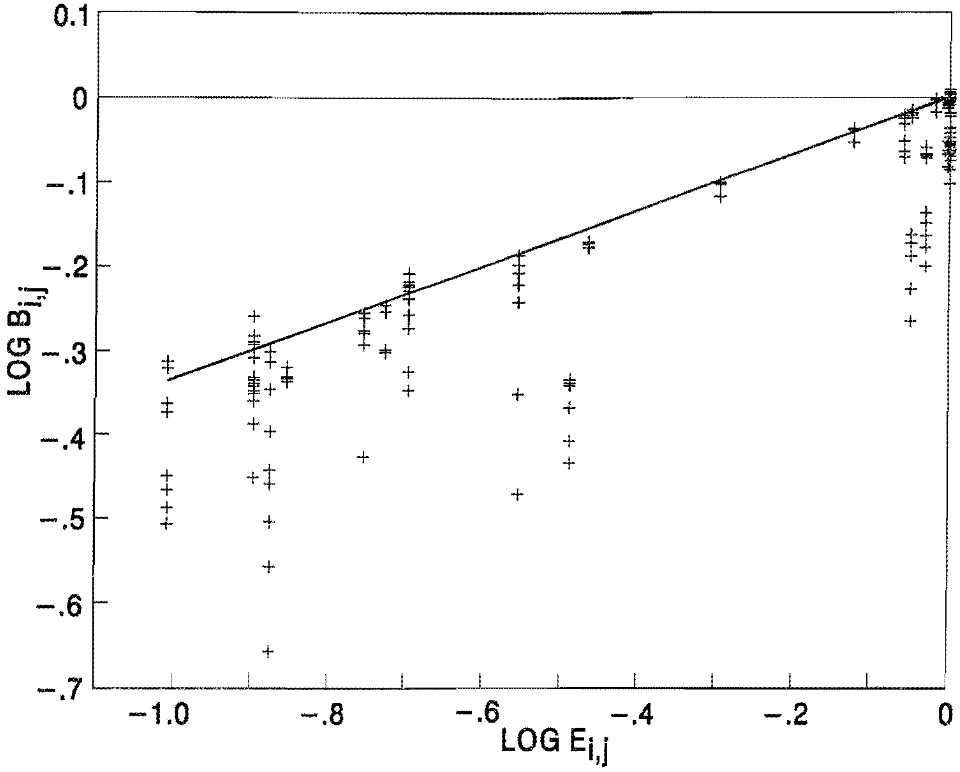

Figure 1.—Evaluation of the empirical constant A using 164 mixtures.

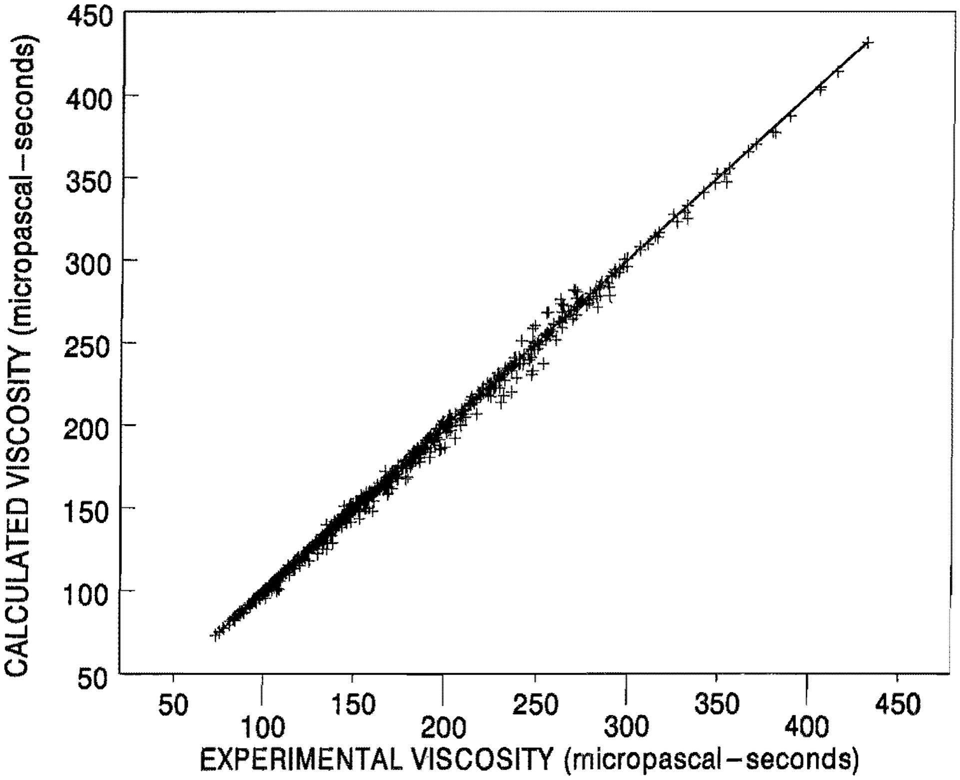

Figure 2.—Comparison of calculated and experimental viscosities for 752 mixtures - line has slope of unity.

The simplified Chapman equation (equation 5) is reported to fit the helium-argon pair and the hydrogen-oxygen pair to 1%. Equation 28 fits the helium-argon pair to 0.56% RMS error (Trautz' data) and to 0.39% RMS error (Kestin's data). The respective AADs are 0.48% and 0.32%. This is an improvement over the simplified Chapman equation. The hydrogen-oxygen pair is fitted to 1.08% RMS error (0.84% AAD). This is comparable to the simplified Chapman equation.

Ten data points were found to contribute over 20% of the sum of the squared deviations. With the exception of these 10 data points, absolute errors were less than 6.5%. The remaining set was fitted with an RMS error of 1.81% (1.19% AAD) and an average deviation (bias) of -0.74%. Five data points fitted with absolute errors over 6.5% were from the neon-carbon dioxide pair. The other five points were from gas pairs that include ammonia as one component.

The 25 data points that showed the largest absolute errors in the fit were from only 4 sets of data. Three of these sets were for gas pairs that included ammonia, and one set was for the hydrogen-neon pair. Neglecting these 4 gas pairs, the other 706 data points are fitted with less than 4.75% maximum absolute error. The RMS error for the reduced set is 1.47% (0.99% AAD); the bias is -0.51%.

Of all the models described, only Brokaw's appears able to fit data more closely. Brokaw's model requires effective collision diameters for all components, and dipole moments if applicable, in addition to pure component viscosities and molecular weights.

The development of our model assumes elastic collisions of spheres. Nonspherical molecules have internal energy levels for rotational, vibrational, and electronic energies. If these internal energy levels are comparable to thermal energies, interaction becomes possible between the translational energy and internal energy levels. The collisions are then no longer elastic. This may account for the difficulty in describing data for ammonia mixtures.

Several models were compared that require only the viscosity and molecular weight for each component against data for the binary systems of helium-neon (*18*) and helium-nitrogen (*19*) at 20° C. These systems were selected because the high molecular weight ratio and the difference between the pure gas viscosities would accentuate differences between the models. The models used were the linear model (equation 1), Wilke's equation (equation 3), and the Herning-Zipperer model (equation 4), as well as equation 28. The calculated results are plotted against the data in figures 3 and 4.

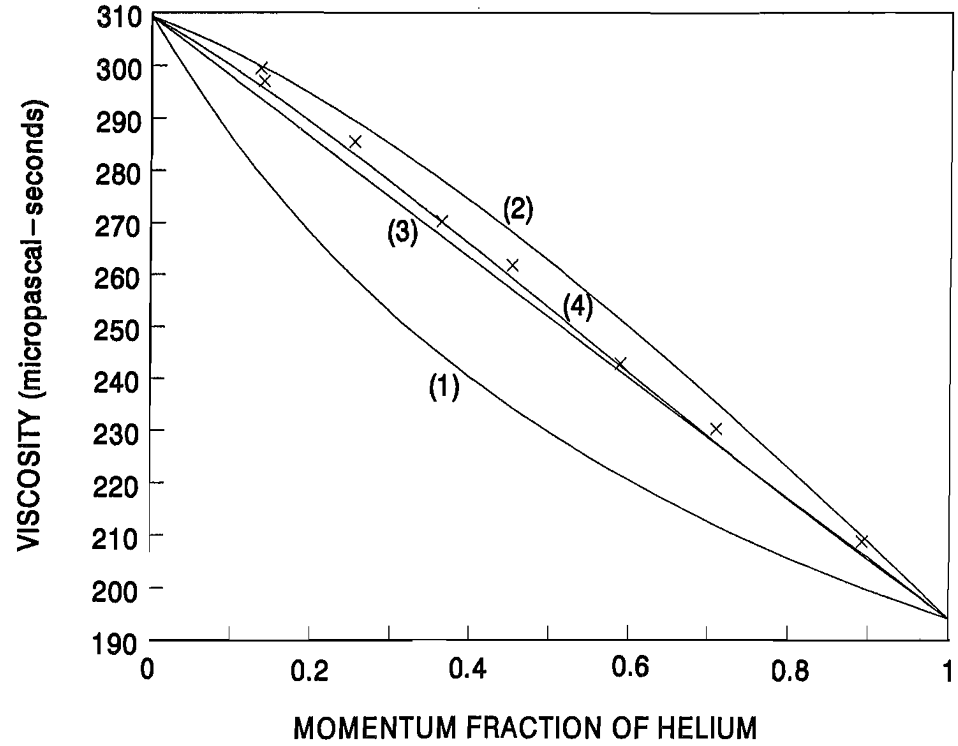

Figure 3.—Comparison of viscosity equations for helium-neon system at 20° C. (1) Linear equation (*1*); (2) Wilke equation (*2*); (3) Herning-Zipperer equation (*5*); (4) equation 28 of this work; (+) data of Kestin and Nagashima (*18*).

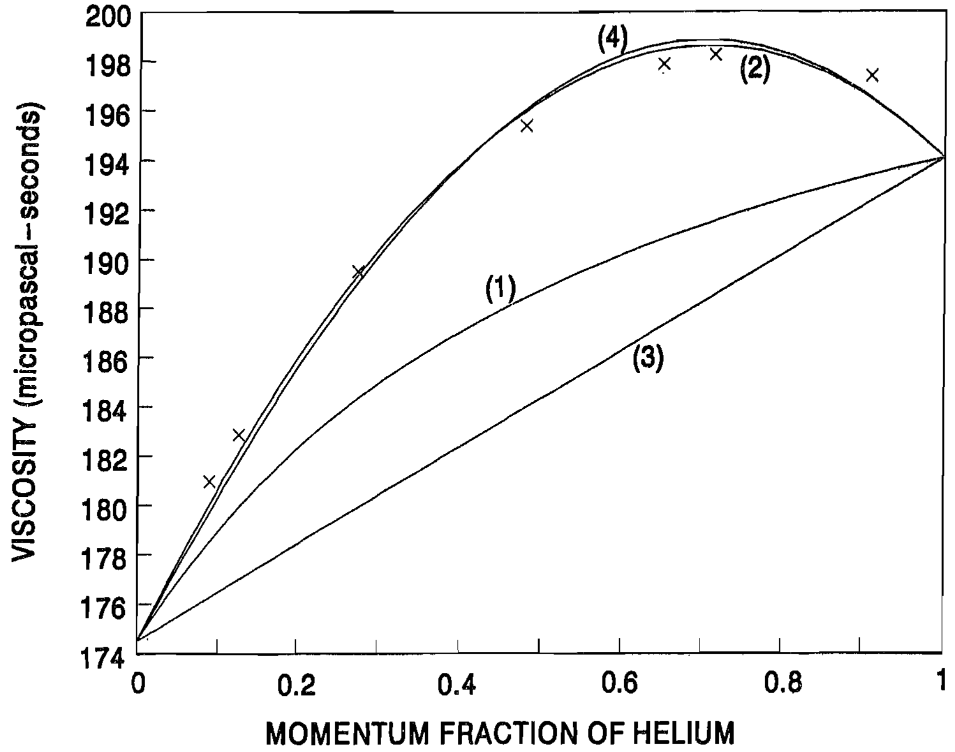

Figure 4.—Comparison of viscosity equations for helium-nitrogen system at 20° C. (1) Linear equation (*1*); (2) Wilke equation (*2*); (3) Herning-Zipperer equation (*5*); (4) equation 28 of this work; (+) data of Kestin, Kobayashi, and Wood (*19*).

Figure 3 compares the models to the data for the helium-neon system. In this system the more massive molecule has greater viscosity. The linear model seriously underestimates the actual viscosity. Wilke's equation significantly overestimates, but the errors are smaller than with the linear model. The simple equation of Herning and Zipperer becomes a straight line when plotted against momentum fraction. In this case, the Herning-Zipperer equation fits the data better than all other models except equation 28.

Figure 4 compares the models to the data for the helium-nitrogen system. In this system the less massive molecule has greater viscosity. The interaction of the differences in viscosity and the differences in molecular weight produces a dramatically changed plot. The Herning-Zipperer equation is now worse than a linear model. The model of Wilke is now able to fit the data almost within experimental error. Equation 28 performs as well as Wilke's in this case.

CONCLUSIONS

A simple method for calculating the viscosity of a mixture of gases has been developed. The development is based on additive contributions to the fluidity and use of the momentum fraction as the main compositional variable. The efficiency of transfer of momentum between bodies of different masses in an elastic collision is considered. One global empirical constant is used. The model is easily applied to multicomponent mixtures.

The only data required by this equation to calculate viscosity for a mixture of known composition are the molecular weights and the viscosities of the pure components at the temperature and pressure of interest. For most binary mixtures of gases, the equation performs at least as well as the best available method using comparable data. The deviations found are 1.29% AAD and 1.99% RMS. The bias is -0.84% and the maximum RMS error found for one system is 5.82%.

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APPENDIX.—LIST OF SYMBOLS

A	exponent of momentum transfer efficiency
a	constant in simplifed Chapman equation
B	mixing coefficient
b	constant in simplifed Chapman equation
C	Maxwell's viscosity proportionality constant
d	density of gas
E	efficiency of momentum transfer
f	fluidity
i	designator for one component of a mixture
j	designator for another component of a mixture
K	average kinetic energy
k	Boltzmann's constant
l	mean free path
M	molecular weight
m	mass
μ	viscosity
p	momentum
ϕ	Wilke's coefficient
T	absolute temperature
V	molar volume
v	average velocity
v_f	final velocity
v_i	initial velocity
x	mole fraction
y	momentum fraction
Z	overall collision frequency
Z_{ij}	binary collision frequency

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