

Comparison of Methods to Compute High-Temperature Gas Viscosity

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A review of the basic equations for computing the viscosity of neutral and ionized species is presented. Four commonly used methods for determining viscosity of a gas mixture are discussed. The performance and accuracy of these methods are tested for 11-species air and hydrogen–helium gas mixtures for temperatures ranging from 200 to 20,000 K. The Gupta–Yos mixing rule gives acceptable results for weakly or nonionized flows and requires half the computer time of solving the full multicomponent equations. The Armaly–Sutton mixing rule is applicable to higher temperature, more strongly ionized flows as long as the tuning parameters for the method are appropriately set. The Wilke mixing rule is the least-accurate method, is actually slower than the Gupta–Yos mixing rules, and should be used only as a method of last resort.

Nomenclature

$b_{j0}(1)$	=	Sonine polynomial expansion coefficient, sec/cm ³
e	=	electron charge, 4.803e–10 esu
esu	=	gm ^{1/2} cm ^{3/2} /s
g	=	relative molecular velocity, cm/s
k_b	=	Boltzmann's constant, 1.38054e–16 erg/K
M	=	molar mass, g/mole
m	=	molar mass, g/particle
N_A	=	Avogadro's number, 6.0225e+23 mol ^{–1}
n	=	number density, cm ^{–3}
$Q^{(l)}$	=	total interaction cross section
r	=	intermolecular separation
T	=	temperature, K
x	=	mole fraction
Z	=	charge number
η	=	viscosity coefficient, g/(cm · s)
Λ	=	shielding parameter
λ	=	Debye shielding length
μ_{ij}	=	reduced mass
σ^2	=	rigid sphere collision diameter, Å ²
$\Omega_{ij}^{(1,1)}, \Omega_{ij}^{(2,2)}$	=	collision integrals, cm ³ /s
$\Omega_{ij}^{(1,1)*}, \Omega_{ij}^{(2,2)*}$	=	reduced collision integrals, dimensionless
Subscripts		
e	=	electron
f	=	field
i, j, k	=	species

Introduction

ACCURATE modeling of the transport of momentum, energy, and mass in high-temperature shock layers is required to obtain an accurate description of the aerothermal environments needed for

thermal protection system design. There are two aspects to modeling transport: 1) transport properties of each species and 2) transport of the gaseous mixture as a whole. In the present paper, the transfer of momentum is examined. Specifically, the present paper details the methods to compute the coefficient of viscosity of individual species and gas mixtures.

Literally hundreds of papers have been written on the subjects of species and mixture viscosity, for example, Refs. 1–27. These papers commonly use different, and often conflicting, nomenclatures and systems of units. Much of the substantive theoretical work on computing transport properties was performed in the 1950s and 1960s. These references can be difficult to find. The purpose of this paper is to provide in one source the details of computing the viscosity of both neutral and ionized species. This paper also presents and compares the mixture viscosity relations most commonly used for hypersonic flow simulations using a consistent nomenclature and system of units for each method.

The computation of the viscosity of a pure species depends on the type of species considered. Short-range intermolecular forces (valence and chemical forces) are the primary drivers of neutral species interactions, especially at higher temperatures. These types of forces decrease rapidly with distance, so that distant encounters do not contribute significantly to the viscosity coefficient. Neutral species viscosity is a function of temperature only.

Ionized species viscosity is different from neutral species viscosity in that both long-range (electrostatic) and short-range intermolecular forces influence interactions between ionized species. Electrostatic forces decrease much more slowly with distance than do short-range forces. Because the number of distant encounters is so great, electrostatic forces due to distant encounters between charged particles can have a significant, even dominant, effect on the transport properties of a molecule. In addition, Coulomb shielding by free electrons plays a key role in determining the effective interaction cross section. Therefore, ionized species viscosity is a function of both temperature and electron number density. The first part of this paper presents a brief review of the general expressions for computing neutral and ionized species viscosity.

The viscosity of a multicomponent gas mixture can be obtained by solving the Boltzmann equation. This is generally accomplished using a Sonine polynomial expansion. The determination of the Sonine expansion coefficients requires the solution of a system of equations equal to the number of species present in the gas mixture. A number of simplified methods have been developed over the years to approximate multicomponent gas viscosity.^{1–7} These approximate methods try to address the dual requirements of computational efficiency and physical accuracy. Generally speaking, the approximate methods were designed for low-to-moderate temperature, neutral gas mixtures. They may or may not be applicable to

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higher temperature gas mixtures where ionized species are present in significant amounts.

Three of the approximate mixture viscosity methods most commonly used in computational fluid dynamics (CFD) are evaluated in terms of efficiency and accuracy for an 11-species air mixture at temperatures ranging from 200 to 20,000 K. The approximate method results are compared against those obtained solving the multicomponent viscosity equations. Calculations are also performed using a hydrogen-helium gas mixture that approximates the Jovian atmosphere.

Viscosity of Neutral Species

The viscosity coefficient for a gas mixture can be obtained by solving the Boltzmann equation using a Sonine polynomial expansion (see Refs. 8 and 9). Because of the rapid convergence of the expansion, an accurate representation of the viscosity coefficient can be acquired by including only the first term. The equation for a single-species gas in grams per centimeter second reduces to

$$\eta_i = \frac{1}{2} k_b T b_{10}(1) = \frac{5}{8} (k_b T / \Omega_{ii}^{(2,2)}) \quad (1)$$

In Eq. (1), the $b_{10}(1)$ term is one of a series of Sonine expansion coefficients, which are discussed in more detail later. The viscosity collision integral $\Omega^{(2,2)}$ is one of a linear combination of integrals that are used to represent the Sonine polynomial expansion coefficients. As will be shown, an additional collision integral $\Omega^{(1,1)}$ will be required to compute the viscosity of a gas mixture. The collision integrals have units of cubic centimeter per second and take the following general form:

$$\Omega_{ij}^{(l,s)}(T) = \sqrt{\frac{k_b T}{2\pi\mu_{ij}}} \int_0^\infty \exp(-\gamma^2) \gamma^{2s+3} Q^{(l)}(g) d\gamma \quad (2)$$

$$\mu_{ij} = \frac{m_i m_j}{m_i + m_j} \quad (3)$$

$$\gamma^2 = \frac{1}{2} \frac{\mu_{ij} g^2}{k_b T} \quad (4)$$

The total interaction cross section $Q^{(l)}$ is computed by integrating the differential cross sections.⁸ Thus, the collision integral represents an orientationally and thermally averaged collision probability between two particles. The calculation of collision integrals from differential cross sections depends on the potential energy model. To simplify comparisons, Hirschfelder et al. presented the concept of a reduced collision integral⁸ that is the ratio of the collision integral evaluated using a given potential energy model divided by the collision integral evaluated using the rigid sphere model:

$$\Omega_{ij}^{(l,s)*} = \Omega_{ij}^{(l,s)} / [\Omega_{ij}^{(l,s)}]_{\text{rigid sphere}} \quad (5)$$

The expressions for the commonly used collision integrals $\Omega^{(1,1)}$ and $\Omega^{(2,2)}$ can be reexpressed in terms of the reduced collision integrals as

$$\begin{aligned} \Omega_{ij}^{(1,1)} &= \sigma^2 \sqrt{\pi k_b T / 2\mu_{ij}} (\Omega_{ij}^{(1,1)*}) \\ \Omega_{ij}^{(2,2)} &= 2\sigma^2 \sqrt{\pi k_b T / 2\mu_{ij}} (\Omega_{ij}^{(2,2)*}) \end{aligned} \quad (6)$$

Substituting Eq. (6) into Eq. (1) yields the commonly cited expression for the viscosity coefficient of a pure, neutral gas species:

$$\begin{aligned} \eta_i &= \frac{\frac{5}{8} k_b T \sqrt{m_i}}{2\sigma^2 \Omega_{ii}^{(2,2)*} \sqrt{\pi k_b T}} = \frac{\frac{5}{16} \sqrt{M_i / N_A} (k_b T)}{1.0e-16 \sqrt{\pi} \sigma^2 \Omega_{ii}^{(2,2)*}} \\ &= 2.6693e-5 \frac{\sqrt{M_i T}}{\sigma^2 \Omega_{ii}^{(2,2)*}} \end{aligned} \quad (7)$$

Note that Eq. (7) is a function of temperature only, which is consistent with kinetic theory. The factor of 10^{-16} is used to convert σ^2 from square angstrom to square centimeters.

Values of $\sigma^2 \Omega_{ii}^{(2,2)*}$ have been tabulated for many simplified intermolecular potentials and are available in the literature. Data for many neutral species interactions can be found in a variety of sources.^{1,10-15} A general discussion of the various interaction potential models can be found in Ref. 8. An evaluation of some of the sources of collision integral data is contained in Refs. 14 and 15.

Viscosity of Ionized Species

The nature of encounters between charged particles is different than that of neutral particles. Neutral particle encounters are governed primarily by short-range intermolecular forces. The effect of an encounter falls off rapidly with increasing distance such that distant encounters between neutral particles do not contribute significantly to the transport properties of a molecule. On the other hand, charged particle encounters are governed by both short-range and long-range (electrostatic) forces. Electrostatic forces decrease much more slowly with separation than do intermolecular forces. Because the number of distant encounters is so great, the electrostatic forces due to distant encounters between charged particles can have a significant, even dominant, effect on the transport properties of an ionized molecule. To a first approximation, the viscosity of an ionized gas can be obtained by solving the Boltzmann equation using the Debye potential (see Ref. 16)

$$V(r) = (1/r) \exp[-(r/\lambda)] \quad (8)$$

The Debye shielding length λ is discussed in the next paragraph. The Debye potential function represents the Coulomb interaction of ions and electrons. At very high temperatures, ion-ion interaction energies can be considerably different from a Coulomb potential energy. However, for most aerospace applications, the quantum mechanical deviation from a Coulomb potential will be small, and this effect can be neglected. For example, even at 10,000 K, the quantum mechanical correction for the $\sigma^2 \Omega_{ii}^{(2,2)*}$ value for atomic nitrogen ion is less than 0.05% of the screened Coulomb potential value (see Ref. 16).

As was already stated, electrostatic forces play a dominant role in determining the viscosity of an ionized species. However, because of the charge neutrality of most undriven ionized gases, the electrical charge of a particle beyond a certain distance is effectively neutralized by nearby particles of opposite charge. Another way of describing the situation is that, beyond a certain distance, charged particles are shielded from each other by intervening charges. In practice, this shielding effect has the effect of cutting off the integration of the collision cross section at a large distance known as the Debye shielding length. Interactions beyond this distance are ignored. The value of the Debye shielding length (in centimeters) can be obtained from¹⁷

$$\lambda = \sqrt{k_b T / 4\pi n_e e^2} = 6.905 \sqrt{T/n_e} \quad (9)$$

For the viscosity of an ionized gas species, in the absence of a magnetic field the coefficient of viscosity (in grams per centimeter second) for a fully ionized gas is given by¹⁸

$$\eta_i = 0.406 \frac{m_i^{\frac{1}{2}} (k_b T)^{\frac{5}{2}}}{Z^4 e^4 \ln \Lambda} = 2.207e-15 \frac{M_i^{\frac{1}{2}} T^{\frac{5}{2}}}{Z^4 \ln \Lambda} \quad (10)$$

The charge number Z is 1 for a mixture of singly ionized gas molecules. The shielding parameter Λ is given by¹⁸

$$\Lambda = (3/2 Z Z_f e^3) \sqrt{k_b T^3 / \pi n_e} = 12143 (T^{\frac{3}{2}} / Z Z_f \sqrt{n_e}) \quad (11)$$

The charge number of the field, Z_f , is 1 for a singly ionized gas. The viscosity of a fully ionized gas is due primarily to the positive ions. Viscous stresses due to electrons are generally negligible,¹⁸ but the electron number density must be obtained to calculate the shielding parameter.

Although Eq. (10) is useful in computing the viscosity of a fully ionized gas, it is necessary to develop an expression for the collision integrals of an ion-ion interaction to compute the mixture viscosity

of a gas containing both neutral and ionized species. When Eqs. (10) and (7) are compared, it is possible to derive an expression for $\sigma^2\Omega_{ij}^{(2,2)*}$ (in square angstrom) for an ionized species interaction:

$$\sigma^2\Omega_{ij}^{(2,2)*} = 1.209e + 10(\ln \Lambda / T^2) \quad (12)$$

Gupta et al.¹ used a slightly modified form of Eq. (12) to compute the ion-ion viscosity collision integral. His results are in generally good agreement with Spitzer.¹⁸ In addition, Mason et al.¹⁷ have presented tables of ionized species collision integral data using both attractive and repulsive screened potentials as a function of a normalized temperature. More recently, Stallcop et al.¹⁶ presented an expression to compute the repulsive shielded Coulomb collision integral in terms of a reduced temperature. Their expression, which closely reproduces the data of Ref. 17, was a curve fit to their detailed computational chemistry calculations of the potential energies and transport cross sections of ions:

$$\sigma^2\Omega_{ij}^{(N,N)*} = 5.0e + 15(\lambda / T^*)^2 \ln\{D_N T^* [1 - C_N \exp(-c_N T^*)] + 1\} \quad (13)$$

The reduced temperature T^* is defined by the following expression:

$$T^* = 4132.5(T^{\frac{3}{2}} / \sqrt{n_e}) \quad (14)$$

The values for the constants in Eq. (13) for the repulsive potential are reproduced from Ref. 16 in Table 1. In addition, new curve fits for the attractive (ion-electron) potential have been generated from the data in Ref. 17 and are also given in Table 1. Strictly speaking, the values in Table 1 are valid for $T^* > 4$; however, this condition will be met except at very high temperatures and/or densities. For example, the T^* value for 11 species equilibrium air at 25,000 K and 100 kPa is 43.

Table 2 shows computed values of $\sigma^2\Omega_{ij}^{(2,2)*}$ using both Eq. (13) (Spitzer¹⁸ and Gupta et al.¹) and Eq. (14) (Mason et al.¹⁷ and Stallcop et al.¹⁶). The results are based on an equilibrium electron number density computed using an 11-species air chemistry model at a reference pressure of 100 kPa. The results show that Eq. (13) gives somewhat higher values for $\sigma^2\Omega_{ij}^{(2,2)*}$ than Eq. (14). However, the values are within 10% of each other, which is a smaller uncertainty than exists for most binary interactions; therefore, in practice both methods of evaluating $\sigma^2\Omega_{ij}^{(2,2)*}$ are acceptable.

Of greater concern is the approximation made by Gupta et al.¹ in the computation of $\sigma^2\Omega_{ij}^{(2,2)*}$ for ionized species pairs. They assume

that the ratio of viscosity and momentum transfer collision integrals for ionized species pairs is given by¹

$$A_{ij}^* = \frac{\Omega_{ij}^{(2,2)*}}{\Omega_{ij}^{(1,1)*}} = \frac{1.36Z^4 Q_c}{0.795Z^4 Q_c} = 1.71 \quad (15)$$

Thus, they assume the collision integral ratio for ionized species pairs has a constant value of 1.71 for all temperatures and electron number densities. However, the screened Coulomb potential results of Mason et al.¹⁷ and Stallcop et al.¹⁶ clearly show a temperature dependence of this ratio, for both attractive and repulsive potentials. Furthermore, the value of 1.71 given by Gupta et al.¹ appears unrealistically high at all temperatures and electron number densities. Both Mason et al.¹⁷ and Stallcop et al.¹⁶ indicate values of this ratio for repulsive potentials ranging from 1.02 to 1.26.

Collisions Between Ions and Neutrals

Collisions between ions and neutrals can be a significant contributor to the mixture viscosity of a partially ionized mixture. Although it is beyond the scope of this paper to discuss the details of such interactions, in practice they are governed by similar types of short-range forces that govern neutral interactions, coupled with several long-range forces, including polarizability and dispersion effects. Once an acceptable model for the intermolecular potential is selected, calculation of the collision integrals proceeds as for neutral interactions, and the resulting binary viscosity coefficient is a function of temperature only. Mason and McDaniel¹⁹ provide a detailed discussion of the relevant collision theory.

Viscosity of Multicomponent Gas Mixtures

The viscosity coefficient of a multicomponent gas mixture can be obtained by solving the Boltzmann equation using a Sonine polynomial expansion. Because of the rapid convergence of the Sonine polynomials, an accurate representation of the mixture viscosity coefficient can be acquired by including only the first term in the expansion⁸

$$\eta = \frac{1}{2} k_b T \sum_{j=1}^{NS} n_j b_{j0}(1) \quad (16)$$

In Eq. (16), NS is the number of species in the mixture and $b_{j0}(1)$ are Sonine expansion coefficients. The definition of the expansion coefficients consists of a complicated combination of bracket integrals, details of which are provided in Ref. 8. Chapman and Cowling²⁸ demonstrated that the bracket integrals could be replaced by a linear combination of collision integrals. When this simplification is used, the expansion coefficients can be determined from the following system of equations:

$$\sum_{j=1}^{NS} \frac{Q_{ij}^{00}}{R_{i0}} b_{j0}(1) = -1, \quad i = 1, 2, \dots, NS \quad (17)$$

where

$$\frac{Q_{ij}^{00}}{R_{i0}} = \frac{16 m_i}{15 m_j} \sum_{k=1}^{NS} \frac{n_k m_k}{(m_i + m_k)^2} \times \left[5m_j(\delta_{ij} - \delta_{jk})\Omega_{ik}^{(1,1)} + \frac{3}{2}m_k(\delta_{ij} + \delta_{jk})\Omega_{ik}^{(2,2)} \right] \quad (18)$$

The Kronecker delta δ_{mn} is 1 if $m = n$ and 0 otherwise. Thus, the determination of multicomponent viscosity requires the solution of a system of equations whose size is equal to the number of species in the gas mixture. The cost of computing multicomponent viscosity increases rapidly as the number of species in the gas mixture increases.

Equation (16) can be recast in terms of mole fractions x_j , and the leading term can be pulled inside the Sonine polynomial expression yielding the expression

$$\eta(1) = \sum_{j=1}^{NS} \frac{1}{2} x_j n k_b T b_{j0}(1) = \sum_{j=1}^{NS} x_j \bar{b}_{j0}(1) \quad (19)$$

Table 1 Shielded Coulomb collision integral curve-fit coefficients

n	Attractive potential			Repulsive potential		
	C_n	c_n	D_n	C_n	c_n	D_n
1	-0.476	0.0313	0.784	0.138	0.0106	0.765
2	-0.146	0.0377	1.262	0.157	0.0274	1.235

Table 2 Shielded Coulomb collision integral $\sigma^2\Omega_{ij}^{(2,2)*}$ for ion-ion interactions, assuming an equilibrium electron number density at a pressure of 100 kPa

Temperature, K	n_e, cm^{-3}	Stallcop et al., ¹⁶	Spitzer, ¹⁸
		Eq. (15)	Eq. (13)
5,000	6.115×10^{13}	3040	3051
6,000	2.556×10^{14}	1939	1970
7,000	6.923×10^{14}	1347	1382
8,000	2.198×10^{15}	947	986
9,000	6.805×10^{15}	678	721
10,000	1.711×10^{16}	506	548
11,000	3.572×10^{16}	391	430
12,000	6.378×10^{16}	313	348
13,000	9.905×10^{16}	258	289
14,000	1.353×10^{17}	220	247
15,000	1.646×10^{17}	192	215

In Eq. 19, $\eta(1)$ indicates that this is a first-order mixture viscosity and that $\bar{b}_{j0}(1) = 1/2n k_b T b_{j0}(1)$. The system of equations shown in Eq. (17) can be recast in terms of $\bar{b}_{j0}(1)$ as

$$\sum_{j=1}^{NS} H_{ij} \bar{b}_{j0}(1) = x_i, \quad i = 1, 2, \dots, NS \quad (20)$$

where

$$H_{ij} = \frac{32}{15} x_i \frac{m_i}{m_j} \sum_{k=1}^{NS} \frac{x_k m_k}{(m_i + m_k)^2} \times \left[5m_j (\delta_{ij} - \delta_{jk}) \frac{\Omega_{ik}^{(1,1)}}{k_b T} + \frac{3}{2} m_k (\delta_{ij} + \delta_{jk}) \frac{\Omega_{ik}^{(2,2)}}{k_b T} \right] \quad (21)$$

The collision integrals in Eq. (21) are general collision integrals. As already noted, general collision integrals are often expressed as the collision integral for the rigid-spheremodel multiplied by a reduced collision integral, $\Omega^{(l,s)*}$. When this convention is used, the equation for the H_{ij} terms becomes

$$H_{ij} = B \frac{x_i}{\sqrt{T}} \sum_{k=1}^{NS} \frac{x_k \sqrt{\mu_{ik}}}{(m_i + m_k)} \times \left[\frac{5}{3} (\delta_{ij} - \delta_{jk}) \sigma^2 \Omega_{ik}^{(1,1)*} + \frac{m_k}{m_j} (\delta_{ij} + \delta_{jk}) \sigma^2 \Omega_{ik}^{(2,2)*} \right] \quad (22)$$

All terms in Eq. (22) are expressed in cgs units. The constant B is given by the following relation:

$$B = (32/15)(3/2)2.0e-16\sqrt{\pi N_A/2k_b} = 52979 \quad (23)$$

See Ref. 20 for a discussion of the physical significance of the diagonal and off-diagonal elements of H . For a gas containing a single species, the equation for H_{ij} reduces to the following:

$$H_{ii} = 37462(\sigma^2 \Omega_{ii}^{(2,2)*} / \sqrt{M_i T}) \quad (24)$$

When Eq. (24) is incorporated into Eqs. (19) and (20), the standard expression for the viscosity of a pure species [Eq. (7)] is recovered. Equation (22) is sometimes expressed in terms of a reduced collision integral ratio,

$$H_{ij} = B \frac{x_i}{\sqrt{T}} \sum_{k=1}^{NS} \frac{x_k \sqrt{\mu_{ik}}}{(m_i + m_k)} \sigma^2 \Omega_{ik}^{(2,2)*} \times \left[\frac{5}{3} \frac{1}{A_{ik}^*} (\delta_{ij} - \delta_{jk}) + \frac{m_k}{m_j} (\delta_{ij} + \delta_{jk}) \right] \quad (25)$$

The reduced collision integral ratio A^* is the ratio of $\Omega^{(2,2)*}$ to $\Omega^{(1,1)*}$ for a given species pair. Equations (22) and (25) contain the general form for the H_{ij} coefficients. The general expression can be simplified for both diagonal and off-diagonal terms. For off-diagonal terms, $i \neq j$, the Kronecker delta terms $\delta_{ij} - \delta_{jk}$ and $\delta_{ij} + \delta_{jk}$ will only have nonzero values when $k = j$. This condition removes the summation from the equation leaving the terms

$$H_{ij} = B \frac{x_i x_j}{(m_i + m_j)} \sqrt{\frac{\mu_{ij}}{T}} \sigma^2 \Omega_{ij}^{(2,2)*} \left[-\frac{5}{3} \frac{1}{A_{ij}^*} + 1 \right] \quad (26)$$

Some approximate mixture viscosity formulas assume that $A^* = 5/3$, in which case the off-diagonal terms reduce to zero. The expression for the diagonal terms can be slightly simplified to

$$H_{ij} = B \frac{x_i}{\sqrt{T}} \sum_{k=1}^{NS} \frac{x_k \sqrt{\mu_{ik}}}{(m_i + m_k)} \sigma^2 \Omega_{ik}^{(2,2)*} \times \left[\frac{5}{3} \frac{1}{A_{ik}^*} (1 - \delta_{ik}) + \frac{m_k}{m_i} (1 + \delta_{ik}) \right] \quad (27)$$

The system of equations represented by Eq. (20) can be solved for the $\bar{b}_{j0}(1)$ terms using a standard pivoted Gaussian elimination routine. Another commonly used technique to solve the system of equations is by using Cramer's rule (see Ref. 8). The mixture viscosity is then found by the ratio of the determinant of an augmented H matrix over the determinant of the H matrix, where the augmented matrix includes an extra row and column containing the species mole fractions. Cramer's rule is the most commonly cited technique to solve for multicomponent viscosity. However, Cramer's rule is less stable and more prone to numerical error than pivoted Gaussian elimination (see Ref. 21). As will be shown, in the "results," Cramer's rule also requires more CPU time.

Solution of the system of NS equations given by Eq. (20) yields the exact first-order approximation of the multicomponent mixture viscosity. However, due to the complexity of this system of equations, they are seldom used in practice for the computation of viscosity in a CFD code. Instead, an approximate mixing rule is typically employed to produce reasonable results at a much lower computational cost. In the next section, several of the mixing rules most commonly applied to hypersonic airflows are introduced, and the relative merits of each scheme are discussed.

Wilke Mixing Rule³

Wilke's mixing rule³ was developed in 1950 through application of kinetic theory to the full first-order Chapman-Enskog relation. In addition to the already defined A^* ratio, the assumptions of Wilke can be explained succinctly with the following collision integral ratio definitions:

$$B_{ik} = \Omega_{ik}^{(2,2)*} / \Omega_{kk}^{(2,2)*}, \quad F_{ik} = \Omega_{ik}^{(2,2)*} / \Omega_{ii}^{(2,2)*} \quad (28)$$

First, Wilke assumed that the collision integral ratio $A^* = 5/3$ for all interactions, which makes all off-diagonal terms in the H matrix equal to zero. In addition, Wilke assumed that B_{ik} and F_{ik} are equal to 1 for all interactions, which is analogous to assuming that all binary interactions have the same (hard sphere) cross section. With these assumptions, the mixture viscosity can be expressed as an appropriately weighted sum of the individual species viscosities and is given by

$$\eta = \sum_{j=1}^{NS} \frac{x_j \eta_j}{\phi_j} \quad (29)$$

The scale factor in Eq. (29) is given by the following expression:

$$\phi_i = \sum_{k=1}^{NS} \left[1 + \sqrt{\frac{n_i}{\eta_k}} \left(\frac{M_k}{M_i} \right)^{\frac{1}{2}} \right]^2 / \sqrt{8 \left(1 + \frac{M_i}{M_k} \right)} \quad (30)$$

Wilke's mixing rule can be expected to yield reasonable results for nonpolar and nonionized gas mixtures, for which the off-diagonal terms of H can be shown to be much smaller than the diagonal elements and for which all binary interactions have the same general form. However, for polar or ionized gases, Wilke's mixing rule has been shown to give poor results (see Ref. 22). It is commonly assumed that Wilke's mixing rule³ is the fastest of the mixture viscosity formulations and as such is the most frequently used. However, this assumption will be shown to be incorrect in the "Results."

Armaly-Sutton Mixing Rule²

Armaly and Sutton developed an approximate mixture viscosity formulation² that significantly improves the accuracy of the computed mixture viscosity for ionized gases with only a small increase in complexity. They started with the multicomponent expression (25) and neglected the off-diagonal terms without assuming $A^* = 5/3$. Armaly and Sutton used an approximate formula for the binary collision diameter:

$$\sigma_{ij}^2 = (\sigma_i + \sigma_j)^2 / 4 = (2.6693e-5\sqrt{T}) / 4 \times \left[m_i^{\frac{1}{2}} / \sqrt{\eta_i \Omega_{ii}^{(2,2)*}} + m_j^{\frac{1}{2}} / \sqrt{\eta_j \Omega_{jj}^{(2,2)*}} \right]^2 \quad (31)$$

Inserting Eq. (31) into Eq. (25) for a diagonal term leads to

$$\eta = \sum_{i=1}^{NS} \frac{x_i^2}{H_{ii}} \quad (32)$$

The H_{ii} term in Eq. (32) is given by

$$H_{ii} = x_i \sum_{k=1}^{NS} \frac{2x_k \mu_{ik}}{\eta_{ik}(m_i + m_k)} \left[\frac{5}{3} \frac{(1 - \delta_{ik})}{A_{ik}^*} + \frac{m_k}{m_i} (1 + \delta_{ik}) \right] \quad (33)$$

The binary viscosity η_{ik} is defined by

$$\eta_{ik} = \frac{\sqrt{32\mu_{ik}}}{\left[(m_i^{\frac{1}{2}} / \sqrt{\eta_i}) F_{ik} + (m_k^{\frac{1}{2}} / \sqrt{\eta_k}) B_{ik} \right]^2} \quad (34)$$

The B_{ik} and F_{ik} parameters are defined by Eq. (28).

Rather than assuming constant values for the A^* , B , and F parameters as was done by Wilke,³ Armaly and Sutton² evaluated them using available cross-section data for air and hydrogen-helium mixtures. The numbers assigned to these parameters by Armaly and Sutton are as follows. The quantity A^* has the value 1.25 for all interactions except those of an atom with its own ion. In that situation, the recommended value was 1.1. However, for air species this value does not properly account for the resonant charge exchange process, which dominates the momentum transfer cross section. A more reasonable number for air species, based on quantum mechanical calculations for the N-N⁺ and O-O⁺ interactions,²³ is 0.21.

The F_{ik} parameter was assumed equal to 1 for all interactions, and B_{ik} was determined by solution of Eq. (32). The resulting values for B_{ik} were then grouped according to interaction type and a best-fit value was selected. According to Armaly and Sutton,² B_{ik} is given a value of 0.78 for neutral-neutral interactions, 0.15 for neutral-ion interactions, 0.2 for neutral-electron interactions, and 1.0 for ion-ion, ion-electron, and electron-electron interactions. Finally, it is assumed that $B_{ik} = F_{ki}$. This situation presents a rather curious inconsistency in the Armaly-Sutton formulation. Even though mathematically $B_{ik} = F_{ki}$, the value given to F_{ki} is always 1 where the value given to B_{ik} varies by interaction type. However, in practice this inconsistency is not important because the values of B_{ik} and F_{ik} are determined simply as best-fit parameters.

The Armaly-Sutton mixing rule² is typically expressed in a form similar to that used by Wilke's mixing rule,³ where the mixture viscosity is given by Eq. (29). In this form, the scaling parameter ϕ_i is given by

$$\phi_i = x_i + \sum_{k \neq i} x_k \left[\frac{5}{3} \frac{1}{A_{ik}^*} + \frac{M_k}{M_i} \right] / \left[1 + \frac{M_k}{M_i} \right] \times \left[F_{ik} + B_{ik} \sqrt{\frac{n_i}{\eta_k}} \left(\frac{M_k}{M_i} \right)^{\frac{1}{2}} \right]^2 / \sqrt{8 \left(1 + \frac{M_i}{M_k} \right)} \quad (35)$$

The Armaly-Sutton mixing rule has shown² to yield significantly better results than Wilke's mixing rule³ for ionized gases, at essentially no additional computational cost.

Gupta et al.-Yos Mixing Rule

Yos²⁴ and later Gupta et al.¹ introduced the following mixture viscosity correlation:

$$\eta = \sum_{i=1}^{NS} \left(x_i / \sum_{k=1}^{NS} \frac{x_k}{M_i} \Delta_{ik}^{(2)} \right) = \sum_{i=1}^{NS} \left(x_i / \sum_{k=1}^{NS} \frac{x_k}{M_i} \frac{16}{5} \pi \sigma^2 \Omega_{ik}^{(2,2)*} \sqrt{\frac{2\mu_{ik}}{\pi k_b T}} \right) \quad (36)$$

This mixture viscosity relation ignores all off-diagonal elements of H and makes further simplifications to the diagonal elements.

When expressed using the H_{ii} terms, the Gupta et al.-Yos mixture viscosity relation becomes

$$\eta = \sum_{i=1}^{NS} \frac{x_i^2}{H_{ii}} \quad (37)$$

The H terms are defined according to the following expression:

$$H_{ii} = \sum_{k=1}^{NS} \frac{x_i x_k m_k}{m_i + m_k} B \frac{\sigma^2 \Omega_{ik}^{(2,2)*}}{\sqrt{\mu_{ik} T}} = B \frac{x_i}{m_i \sqrt{T}} \sum_{k=1}^{NS} x_k \sigma^2 \Omega_{ik}^{(2,2)*} \sqrt{\mu_{ik}} \quad (38)$$

The B constant is defined by Eq. (23). The Gupta et al.-Yos formulation is, thus, more exact than Wilke's mixing rule³ because the true nature of the viscosity collision integrals is taken into account. However, the simplifications employed will cause poor agreement between Gupta et al.-Yos and the multicomponent formulation for significantly ionized flows. This was noted by Gupta et al.,¹ and this mixing rule was only designed for weakly or nonionized flows. Despite this, the Gupta et al.-Yos formulation as presented here remains the most commonly used method for computing mixture viscosity in hypersonic CFD codes.

Note that Yos²⁵ later proposed a modified formulation that approximately accounts for the effects of the off-diagonal terms by summing and adding them to the diagonal (also see Ref. 1). This formulation was shown²⁵ to give results in good agreement with the multicomponent formulation. However, because the cost of implementation is comparable to that of the full multicomponent formulation,¹ the method was not examined in this paper.

Results

Two standard benchmarks for evaluating mixing rules are accuracy and cost. The first aspect investigated was the computational cost (efficiency) of the mixture relations. A viscosity test bed code (written in C) was created containing functions for each of the earlier described mixture viscosity methods. For the Wilke³ and Armaly-Sutton² formulations, the species viscosities were calculated directly from Eq. (7) for neutrals and Eq. (10) for ions. However, similar results would be obtained (both in terms of efficiency and accuracy), if the neutral species viscosities were computed using a Blottner et al. style expression.²⁶

To create an apples-to-apples comparison of the results generated by each mixture viscosity method, the same set of collision integral data was used in each method. The data presented by Murphy and Arundell¹⁴ were chosen both because the collision integral data are of high quality and are readily available in the literature. These data have been used by other authors^{5,27} to validate transport property computations. The only binary interaction for which Murphy's data were not used was the N₂-N₂ interaction, where the coefficients suggested by Murphy did not match low-temperature experimental viscosity data.⁸ Therefore, data from Stallcop et al.,²⁹ which agree well with the experimental data, were used instead. Equation (13) was used to compute the collision integrals for the ion-ion, ion-electron, and electron-electron interactions.

To evaluate the computational efficiency of each mixture viscosity method, the clock() routine was invoked before and after each viscosity method call. The difference in the return value between the two clock() method calls is the time required to execute the viscosity method. Inside each viscosity method, the viscosity calculations were repeated 200,000 times to isolate the viscosity calculations themselves from any one-time operations such as temporary variable definitions and array memory allocations.

Two sets of runs were completed. One set used a 5-species air model and the other an 11-species air model. The results for these computations are shown in Table 3. The results were normalized with respect to the time required for Wilke's mixing rule³ to compute the viscosity of that mixture. There is a consistency in the results (as one would expect) between the 5- and 11-species air models. The cost of each method is roughly proportional to the square of the

[‡] Data available online at <http://WebBook.nist.gov> [cited July 2001].

Table 3 Relative computational cost of the methods used to compute mixture viscosity

Method	5-species air	11-species air	11 vs 5 species
Wilke ³	1.0	1.0	4.554
Armaly-Sutton ²	1.054	1.065	4.603
Gupta et al. ¹ -Yos ²⁴	0.891	0.919	4.697
Multicomponent (Gaussian)	1.927	2.098	4.958
Multicomponent (Cramer's rule)	1.979	2.250	5.177

number of species (25 vs 121). The 11-species air model requires about five times the computer time of the 5-species model, as one would anticipate.

Note from Table 3 that Wilke's mixing rule³ is not the most efficient method. The Gupta et al.¹-Yos²⁴ method uses less CPU time than Wilke³ for both the 5- and 11-species air models. This result is obvious if one looks at the higher-order mathematic function calls associated with each method. The Gupta et al.-Yos mixture expression requires fewer calls to the power (exponentiation) and square root functions and, thus, is slightly faster.

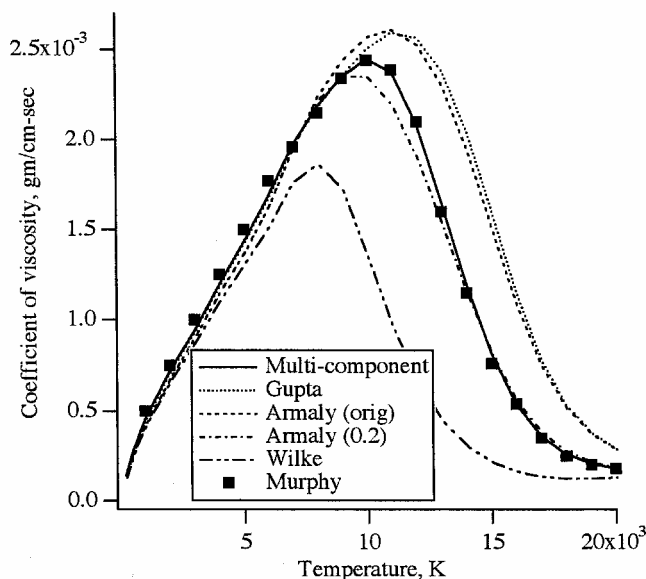
Because of the need to solve a system of equations, the multicomponent viscosity methods require about twice the computer time as the other methods. In addition, it appears that the multicomponent viscosity methods become comparatively more expensive as the number of species increases, although the increased cost ratio from 5- to 11-species is not large (about 10%). Using Gaussian elimination to solve the system of equations is slightly faster than using Cramer's rule. Because Gaussian elimination is also more stable and reliable method to solve a system of equations than Cramer's rule (see Ref. 21), it should be the method of choice when used in conjunction with multicomponent viscosity.

Computational efficiency is only one consideration when choosing a mixture viscosity methodology. A more important consideration is the accuracy of a given method. The multicomponent, the Gupta et al.¹-Yos,²⁴ Armaly-Sutton,² and Wilke³ methods were tested under equilibrium conditions for an 11-species air model over temperatures ranging from 200 to 20,000 K at reference pressures of 100 and 10 kPa.

To ensure that the methods were properly implemented, the results were compared against those of Murphy and Arundell¹⁴ and Murphy,¹⁵ who computed the viscosity of air using a detailed Chapman-Enskog method. The results of Murphy and Arundell¹⁴ and Murphy¹⁵ were, thus, chosen both because the collision integral data that they collected are of high quality and because they are readily available in the literature. These data have been used by other authors^{5,27} to validate transport property computations. As was already stated, to make a meaningful comparison with their results, it was necessary to use the Murphy and Arundell¹⁴ and Murphy¹⁵ collision integral data in the present study for all binary interactions. Note that as long as a consistent set of collision integral data are used for all computations, it is not necessary to evaluate the accuracy of the collision integral data to judge the relative merits of the mixing rules (although the choice of A^* for resonant charge exchange interactions in the Armaly-Sutton² rule complicates things somewhat). In all cases the multicomponent Chapman-Enskog method is the benchmark against which the various mixing rules are compared.

The results for the various methodologies are shown in Fig. 1. The multicomponent results and Murphy and Arundell¹⁴ and Murphy¹⁵ data match very closely along the entire temperature range indicating that the multicomponent mixture viscosity function was properly implemented in the viscosity test bed code. The curve shows a rising tendency until about 10,000 K, where the mixture viscosity value begins to decrease. This is because, although each individual binary viscosity coefficient increases with increasing temperature, ionized species viscosity is an order of magnitude or more less than neutral species viscosity. Above 10,000 K, the ionized species interactions begin to dominate the gas mixture, and the mixture viscosity begins to decrease.

The Gupta et al.¹-Yos²⁴ mixing rule matches the multicomponent value quite closely at low temperatures (weakly ionized gas

**Fig. 1** Mixture viscosity for equilibrium 11-species air at 100 kPa.

mixture). Above 9000 K when ionized species become significant, the Gupta et al.-Yos method overpredicts the mixture viscosity. This overprediction is in part because the off-diagonal terms of H , which account for collisional transfer of momentum between species, have been neglected in this formulation. At temperatures above 14,000 K, the Gupta et al.-Yos value is about twice the multicomponent value.

The value obtained by the Armaly-Sutton² mixing rule depends on the values assigned to the A^* , B , and F parameters. When the A^* value recommended in the original paper ($A^* = 1.1$) for atom-ion interactions with their own ion is used, the Armaly-Sutton results are very close to those obtained using the Gupta et al.¹-Yos²⁴ rule. Using a more representative value of 0.21 for the resonant atom-ion interactions produces a curve much closer to the multicomponent value. The different Armaly-Sutton² curves shown in Fig. 1 suggest that the Armaly-Sutton method can be tuned for different gas mixtures, assuming collision integral data are available. This was suggested by Armaly and Sutton in their original paper.² The Armaly-Sutton results are somewhat less accurate than those of Gupta et al.-Yos at low temperatures (about 4% lower at 500 K), but in general provide quite good results over the entire temperature range, with an accuracy comparable to the full multicomponent method and a cost comparable to results using the Gupta et al.-Yos mixing rule.

Not surprisingly, the least accurate mixture viscosity result is obtained using Wilke's mixing rule.³ At 500 K, the Wilke result is 7% lower than the multicomponent value. Wilke's mixing rule is very inaccurate at temperatures above 7500 K (when the flow becomes ionized) predicting a mixture viscosity value that is up to four times less than the multicomponent value.

Another set of mixture viscosity calculations were performed at a reference pressure of 10 kPa. This corresponds roughly to the postshock pressure of a Mach 20 normal shock wave at 60-km altitude. The results are shown in Fig. 2. The main physical difference between the two sets of computations is that dissociation and ionization occur at a lower temperature at a pressure of 10 kPa than they would at 100 kPa. Also because the pressure is lower, the electron number density at a given temperature will be lower at 10 kPa than it will be at 100 kPa.

The general nature of the curves is the same at 10 kPa as it was at 100 kPa with one exception. The Armaly-Sutton² method result with $A^* = 0.21$ for atom-ion interactions does not compare as closely to the multicomponent result. The separation between the two curves above 9000 K is larger for $p = 10$ kPa than it was for $p = 100$ kPa. This implies that a single set of Armaly-Sutton coefficients may not be generally applicable to any thermodynamic condition.

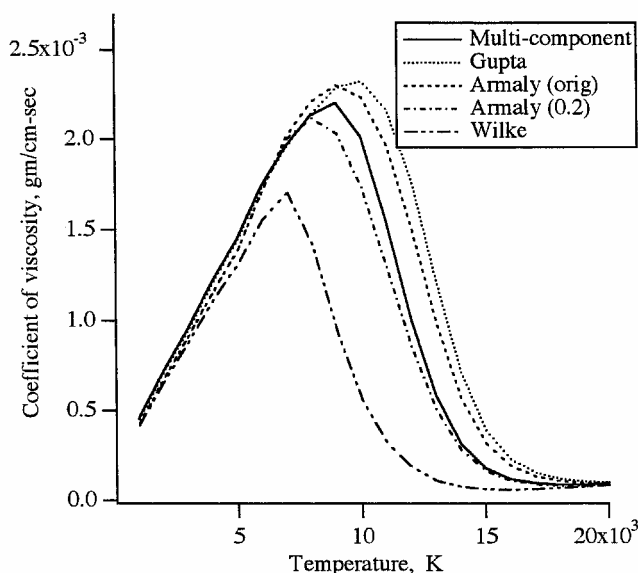


Fig. 2 Mixture viscosity for equilibrium 11-species air at 10 kPa.

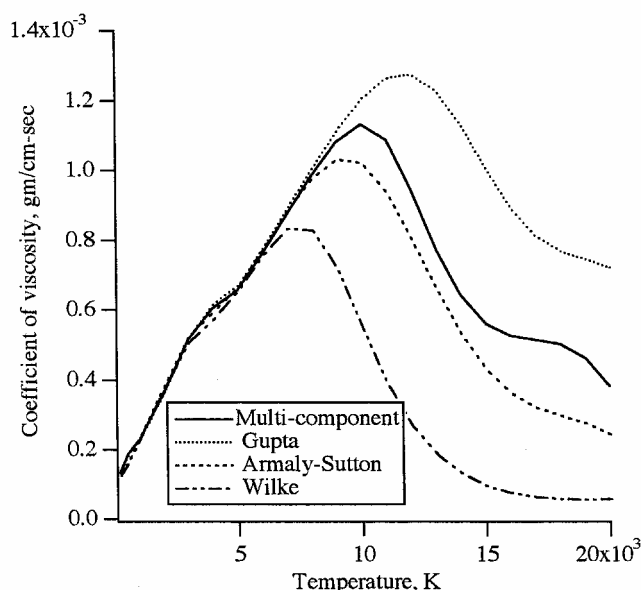


Fig. 3 Mixture viscosity for an equilibrium hydrogen-helium gas mixture at 100 kPa.

To examine the behavior of the mixture viscosity methodologies when applied to a different gas mixture, viscosity coefficients were computed for a six species, H_2 , H , He , H^+ , He^+ , and e , hydrogen-helium gas mixture at a reference pressure of 100 kPa for temperatures ranging from 200 to 20,000 K. Collision integrals for the relevant binary interactions were taken from Biolsi.³⁰ The initial, that is, low-temperature, mole fractions of H_2 and He were 0.864 and 0.136, respectively. This approximates the composition of the Jovian atmosphere. The Armaly-Sutton² coefficients used were the same as with air with the exceptions that the value of A^* for the $H-H^+$ interaction was set to be 0.18 and the value for the $He-He^+$ interaction was 0.025. These numbers follow the recommendations given in Ref. 2, which were also based on Biolsi's collision integral data.³⁰

The results of these calculations are shown in Fig. 3. At lower temperatures where the gas mixture consists mostly of neutral species, the four methods all compute approximately the same viscosity value. Above 7000 K, the results from using Wilke's mixing rule³ begin to deviate significantly from the other methods. Above 10,000 K when significant ionization begins to occur, the Gupta et al.¹-Yos²⁴ method tends to overpredict the mixture viscos-

ity. The Armaly-Sutton² results are closer to the multicomponent values than the Gupta et al.-Yos data; however, the correlation between the Armaly-Sutton² and multicomponent results is not as good for the hydrogen-helium gas mixture as it was for 11-species air. This again suggests that the Armaly-Sutton coefficients may have to be tailored to the specific gas mixture under consideration.

Conclusions

A review of the basic equations for computing the viscosity of neutral and ionized species was presented. The viscosity of neutral species is a function of temperature only, and that of ionized species viscosity is a function of both temperature and electron number density. Four commonly used methods for determining mixture viscosity were discussed. The performance and accuracy of these methods were tested for 11-species air and hydrogen-helium gas mixtures at temperatures ranging from 200 to 20,000 K, and recommendations for use in various situations are summarized next.

Solving the full multicomponent viscosity equations yields the most accurate result, but also requires at least twice the computer time of the other methods. If the multicomponent method is used, the system of equations should be solved using Gaussian elimination rather than with Cramer's rule. If computer time is not an issue and if adequate collision integral data exist for the gas mixture, the multicomponent viscosity method should be used for high-temperature applications.

Use of the Gupta et al.¹-Yos²⁴ mixing rule gives accurate results until significant ionization occurs in the gas mixture. At low temperatures, it is the approximate method that most closely matches the multicomponent results. This method requires the least computer time, even less than that used by Wilke's mixing rule.³ One disadvantage of the Gupta et al.-Yos method is that it requires reasonably accurate collision integral data for each species pair in the gas mixture. The Gupta et al.-Yos method appears applicable for low-to-moderate temperature applications where CPU time is an issue and where adequate collision integral data exists.

If the A^* , B , and F parameters are properly set, the Armaly-Sutton² mixing rule produces fairly accurate results over a wide temperature range and requires direct knowledge of only pure species collision integrals. The correlation with the multicomponent results continues even at temperatures where ionized species dominate the gas mixture. The Armaly-Sutton² method requires about half the CPU time of solving the full multicomponent equations and about 15% more time than the Gupta et al.¹-Yos²⁴ method. The Armaly-Sutton method appears applicable for high-temperature ionized gas flows where CPU time is an issue or for gas mixtures where adequate collision integral data does not exist for all binary interactions. The Armaly-Sutton method can be tuned through the A^* , B , and F parameters. A single set of parameters may not be applicable to every possible thermodynamic state.

Wilke's mixing rule³ is the least accurate method and requires about 10% more CPU time than does the Gupta et al.¹-Yos²⁴ method. The Wilke mixing rule should only be used for low-to-moderate temperature applications where either adequate collision integral data do not exist or where the Armaly-Sutton² parameters are unknown. Wilke's mixing rule³ should be the method of last resort.

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