

Gaseous slip models based on the Langmuir adsorption isotherm

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On the basis of Langmuir's theory of adsorption of gases on solids, a robust gaseous slip model is presented. The concept of accommodation coefficient and the difference of gas particles are explained within the new framework. It turned out that the Langmuir model recovers the Maxwell model in the first-order approximation in the case of the microchannel gas flow. In order to validate the new approach, the model is applied to problems of technical interests: pressure-driven microchannel gas flow and low Reynolds number gas flow past a sphere. With the help of previous theoretical and experimental results it is shown that with an adjustable parameter the model in low-speed creeping regime with moderate Knudsen numbers yields a prediction in qualitative agreement with the data. © 2004 American Institute of Physics. [DOI: 10.1063/1.1630799]

I. INTRODUCTION

The slip phenomenon has remained as an intriguing problem since it was observed in various experiments of low density gas flows. A typical example is the appearance of a minimum in the mass flow rate of gases in a tube flow.¹ Owing to the recent development of microscale devices in micro-electro-mechanical systems,^{2,3} there has been a renewed interest in understanding the basic nature of this problem. Since the mean free path in a rarefied condition or in flows involving the microscale geometry is not sufficiently small in comparison with the characteristic length, the theoretical investigation requires the molecular description of gases as embodied in the Boltzmann equation or the high-order fluid dynamic equations. Furthermore, the study of rarefied flows additionally requires information on the interaction of gas molecules with the solid surface, which plays a critical role in determining the drag exerted on the body and the heat transfer between the gas and the solid boundary.

Previous works on the formulations of governing equations and boundary conditions for the description of the slip flow have led to various models; the Burnett equations and the Grad moment equations for the governing equations and slip boundary conditions like the one derived by Maxwell.⁴ However, there remains the question of whether those models are robust enough to deal with the question of the high nonlinearity of the slip flow in various situations. The question in the case of the governing equations is related to whether the equations satisfy the basic physical laws such as the second law of thermodynamics, while in the case of boundary conditions it becomes whether the models reflect the true nature of gas-surface molecular interactions and whether their computational models are robust to be applicable to the multidimensional simulation of slip flow in complex flow configurations, which, for instance, model the separation in rarefied gas flows.⁵ There also exist the diffi-

culties of theoretical investigation using fully kinetic models such as the Boltzmann equation, since the theory has not been fully worked out for modifying the Boltzmann collision term that should correctly reflect the molecular collision with the surface and the state of knowledge of gas-surface molecular interaction is not fully satisfactory.

Recently some works⁶⁻⁸ have appeared, which answer some of the aforementioned questions, in particular, with regards to the derivation of the thermodynamically consistent macroscopic governing equations and the macroscopic slip boundary conditions that take into account the interaction of gas molecules and solid surfaces. An important finding obtained from these works was that, in addition to the nonlinear transport coefficients giving rise to, for example, the non-Newtonian effect on flow in the nonequilibrium flow regime, a detailed examination of the gas-surface interaction is necessary to describe the slip phenomena.^{9,10} Furthermore, it was shown that because the gas may interact only with the surface of the solid the gas-surface molecular interaction can be described with the help of the concept of adsorption,¹¹⁻¹³ which is a well-developed field of active research in physical chemistry of surfaces, in which it is observed that deposition of layers of molecules can occur on the solid surface. In this picture, gas molecules can get adsorbed on the surface and desorbed after being held by the intermolecular force field at the surface, and the fraction of adsorbed molecules can be determined by the Langmuir adsorption isotherm, for example. This idea resulted in a rather simple slip boundary condition for a monatomic gas readily available to the study of rarefied gas flows.^{9,10,14}

On the basis of this development, an attempt will be made in this work to reexamine the slip phenomena and to derive a robust slip model for rarefied and microscale gas flows. The approach taken here is similar to the one taken in the previous works^{9,10}—Langmuir's theory of adsorption isotherm—but the emphasis will be placed on the issues of practical implementation of the model under complex geometry configurations, extension to a diatomic gas or mixture,

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and validation of the model. For the validation process, which is a critical issue in the assessment of the accuracy and the demonstration of correctness of a model, a comparison with experimental data will be given with regards to the qualitative aspects. In order to avoid the uncertainty involving the numerics, the validation study is confined to analytically solvable problems—low-speed internal flow in a very long microchannel and low Reynolds number external flow—in which the interaction of gas molecules with the surface molecules remains significant but the nonlinearity arising from the non-Newtonian effect in bulk flow regions may be negligible.

The present paper is organized as follows. First, the origin of the slip phenomenon is reexamined with the help of recent results on the generalized hydrodynamic theory of nonequilibrium gas flows. In order to examine the differences behind the slip models, the popular model derived originally by Maxwell is summarized. Then in Sec. III the gaseous slip model based on Langmuir’s theory of adsorption is described in detail. The present derivation will follow the kinetic derivation based on a mechanical picture because it is easy to follow intuitively. For the statistical thermodynamic derivation of the Langmuir adsorption isotherm, the reader may be referred to the textbooks on physical chemistry of surfaces.¹³ In Sec. IV microchannel flows and low Reynolds number flow past a sphere are investigated to validate the new slip model and to show the feasibility of the approach for the study of slip phenomena taken in the present work. Finally concluding remarks are given in Sec. V.

II. SLIP IN NONEQUILIBRIUM GAS TRANSPORT

A. Origin of gaseous slip

The degree of nonequilibrium in phase space is best represented by the Knudsen number since the collision integral of the Boltzmann equation in nondimensionalized form is scaled by an inverse Knudsen number. Based on this observation the value of Knudsen number was used as the primary parameter to determine the degree of rarefaction and the degree of validity of macroscopic model. For example, the flow can be classified as in the continuum regime ($\text{Kn} \leq 10^{-3}$), the slip flow regime ($10^{-3} \leq \text{Kn} \leq 10^{-1}$), the transition regime ($10^{-1} \leq \text{Kn} \leq 10$), and the free molecular flow ($\text{Kn} > 10$). Even though this classification may serve as a guide to explain the various experiments and as a starting point for effective theoretical investigations there are many indications to make the picture of slip flow more complicated. For instance, the early study of the drag by a flat plate in the low-density regime¹⁵ showed that the parameter $\text{Kn} \cdot M$ is the dominant factor to determine its macroscopic property—the drag coefficient. A simple explanation of this result—trivial at first glance—can be obtained by examining how the non-equilibrium effects are described in the macroscopic level. It is straightforward to identify two primary nondimensional parameters Re and M by comparing inertial force, hydrostatic pressure, and viscous force terms in the conservation of law of momentum; the former being the ratio of inertial force to viscous force, and the latter the ratio of inertial force to the

hydrostatic pressure. By a similar fashion, a composite parameter can be defined as the ratio of viscous force to the hydrostatic pressure p . Then this parameter,^{6,7} defined as

$$N_\delta = \frac{\eta u/L}{p} \approx \text{Kn} M \approx \frac{M^2}{\text{Re}}, \quad (1)$$

can best represent the degree of nonequilibrium in macroscopic thermodynamic space because the viscous force is a direct consequence of the nonequilibrium effect. The η is the Chapman–Enskog viscosity, u is the velocity, and L is the characteristic length. The importance of this parameter can be seen by examining the constitutive equations of various high order hydrodynamic models; the moment equation of Grad¹⁶ and Eu.^{6–8,17,18} Under the shear flow condition, which is in essence the same as the slip flow near a solid wall, the constitutive equations of the normal stresses $\hat{\Pi}_{yy}$ in the dimensionless form can be simplified into, in the order of approximation used by Grad¹⁶ and Eu,^{6,7}

$$\hat{\Pi}_{yy} = - \frac{2\hat{\Pi}_{xy\text{NS}}^2}{3 + 2\hat{\Pi}_{xy\text{NS}}^2}, \quad (2)$$

$$\hat{\Pi}_{yy} = - \frac{2\hat{\Pi}_{xy\text{NS}}^2}{3(\sinh \hat{R}/\hat{R})^2 + 2\hat{\Pi}_{xy\text{NS}}^2}, \quad (3)$$

where $\hat{R}^2 = 3\hat{\Pi}_{yy}(\hat{\Pi}_{yy} - 1)$ and the subscript NS represents the Navier–Stokes theory. The caret (^) over a symbol represents a quantity with the dimension of the ratio of the stress to the pressure, for example, $\hat{\Pi}_{yy} \equiv N_\delta(\Pi_{yy}/p)$ and $\hat{\Pi}_{xy\text{NS}} \equiv N_\delta(-\eta du/dy)/p$. The x and y denote the tangential and normal directions at the wall, respectively. Then the shear stress $\hat{\Pi}_{xy}$ is determined by the following stress constraint whose ultimate origin can be traced to the kinematic terms in the constitutive equations,

$$\hat{\Pi}_{xy} = \text{sign}(\hat{\Pi}_{xy\text{NS}}) \left[-\frac{3}{2}[\hat{\Pi}_{yy} + 1]\hat{\Pi}_{yy} \right]^{1/2}. \quad (4)$$

For simplicity, the isothermal flow of a monatomic gas is assumed in this derivation. It should be mentioned that the essence of these relations remains the same for other closures based on the moment method. Equations (2) and (3) of the normal stresses can be easily solved for a given value of the shear velocity gradient, equivalently, $\hat{\Pi}_{xy\text{NS}}$ and then the shear stresses follow from the stress constraint (4). The general properties obtained by this procedure can be found in Fig. 1 where the stresses by the Grad and Eu theories are illustrated against the shear velocity gradient. As expected, the normal stresses vanish near equilibrium (origin) or in the limit of small N_δ , and the shear stresses recover the value predicted by the Navier–Stokes theory. On the other hand, as the value of N_δ increases, the stresses deviate from those predicted by the linear relations and eventually all the components of the stress tensor, $p + \Pi_{yy}$ and Π_{xy} in the dimensional form, vanish. This unusual asymptotic behavior typical of free molecular flows indicates that the gas can slip near the solid wall, and consequently velocity slip can be

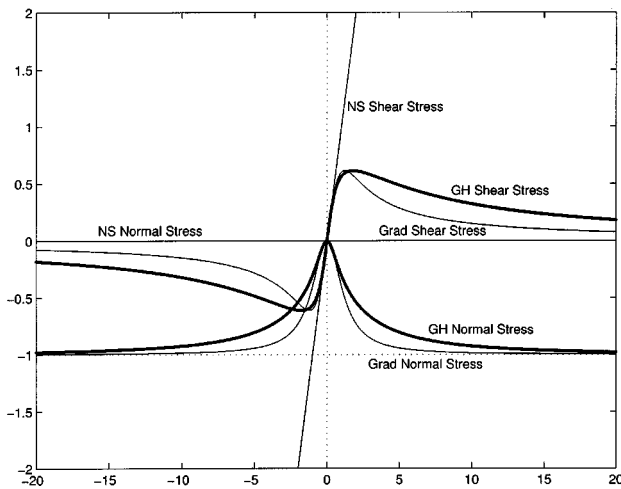


FIG. 1. Grad and Eu (generalized hydrodynamics; GH) constitutive relations to the Navier–Stokes theory in the shear flow of a monatomic gas. The horizontal axis represents the shear velocity gradient $\hat{\Pi}_{xy,NS}$.

observed in this limit because of a purely hydrodynamic reason, which has nothing to do with the gas–surface molecular interaction. The velocity slip by this mechanism comes from the nonlinear nature of the constitutive equations and thus it may be called the slip by the non-Newtonian effect of gas flow in highly nonequilibrium states. This finding can also be confirmed by theoretical investigations^{17,19} or numerical simulations of the high order hydrodynamic equations in a simple geometry.⁸ Therefore, it may be feasible to explain the slip phenomenon largely by two basic components; one the non-Newtonian effect in bulk flow region measured by the composite number N_δ and another the gas–surface molecular interaction measured by the Knudsen number Kn whose scale is the order of gas molecular size. If we classify the nonequilibrium parameter regimes based on Kn and N_δ , most of nonequilibrium gas flows of technological interests may come under one of the following:

- (1) Hypersonic rarefied regime: $Kn = O(10^{-3} \sim 10^{-1})$, $N_\delta = O(10^{-1} \sim 1)$.
- (2) Low-speed microscale (creeping) regime: $Kn = O(10^{-2} \sim 1)$, $N_\delta = O(10^{-5} \sim 10^{-2})$.
- (3) High-speed free-molecular regime: $Kn = O(1 \sim 10)$, $N_\delta = O(1 \sim 10)$.

The typical case of hypersonic rarefied regime can be found in the prediction of the aerothermodynamic loads and heat transfer on hypersonic vehicles that operate in high altitude. For this range of Kn and N_δ it becomes apparent that the slip neither by the gas–surface molecular interaction nor by the non-Newtonian behavior of flow is large because the Knudsen number is small and the constitutive relation of shear flow remains to be linear. However, in this range of N_δ the constitutive relation of gas compression (shock wave) or expansion ceases to follow the linear relation,⁸ and hence it can be argued that the full kinetic theory with an accurate treatment of the collision term or fluid dynamic models that

carefully take into account the energy dissipation mechanism are necessary to properly treat this regime in which shock and expansion waves are abundant.

On the other hand, the opposite conclusion can be drawn for the low speed microscale regime whose typical example is a gas flow at atmospheric condition but in microscale geometry. First of all, owing to the extremely small value of N_δ the slip by the non-Newtonian effect is negligible in the bulk flow region and the linear theory is applicable to all flow regimes. According to the approach based on Kn and N_δ , the only noticeable effect by the high Knudsen number in this acoustic-dominated limit comes from the slip arising from the gas–surface molecular interaction. Therefore it may be argued that if the gas–surface molecular interaction is properly taken into account most of flows in this regime can be described without reckoning the complicated fully kinetic or high order fluid dynamic governing equations. It is from this observation that the present study focuses on the gas–surface molecular interaction and restricts its scope of validation to low speed microscale gas flows.

In the case of high speed free molecular regime, none of the simplifications available to the two aforementioned cases may be possible since all of nonequilibrium factors (high speed, low density, and small length scale) may be present. This regime may be found in mechanical devices operating near the vacuum condition or in high speed microscale gas flows. Since both Kn and N_δ are large well beyond the limit where no slip and the linear constitutive relation may hold, the slip effects from both the gas–surface molecular interaction and the non-Newtonian (nonlinear bulk transport) properties are equally important in this regime. As a consequence, the study of this regime may require a detailed description of nonlinear coupling effects between heat conduction and stresses in the gas, of which the prominent examples are thermal stress and thermal creep.^{20,21} Therefore the study of gas flows in this viscosity dominated limit may be considered the most challenging problem in both theoretical and computational aspects.

B. Maxwell slip model

A simple way to include the slip effect is to make a correction based on the degree of nonequilibrium near the wall surface which can be best represented by the shear stress. This idea can be traced to the work by Maxwell⁴ in which the following slip velocity boundary condition is proposed:

$$u = \varsigma \Pi_w, \quad (5)$$

where u , Π_w , and ς represent the slip velocity, the shear stress at the wall, and the slip coefficient, respectively. If the linear theory is applied to the constitutive equation of the shear stress, the slip velocity reduces to

$$u = \varsigma \left[-\eta \frac{du}{dy} \right]_w. \quad (6)$$

The slip coefficient ς can be further refined by using the kinetic derivation.²² If we introduce the concept of diffusive reflection of the gas molecule near the surface as the colli-

sion in which a molecule is temporarily absorbed at the surface and then re-emitted, the mean velocities q_1 and q_2 of molecules before striking and after leaving the stationary surface can be written as

$$q_2 = (1 - \theta)q_1,$$

where the accommodation coefficient θ represents the fraction of molecules undergoing diffusive reflection. Then the average tangential velocity of the gas at the surface u can be represented as the mean of the pre- and post-collision velocities,

$$u = \frac{1}{2}(q_1 + q_2) = \frac{2 - \theta}{2}q_1.$$

Nothing that the value of q_1 can be approximated by the average velocity at a distance of a mean free path from the surface where the last collision occurred, it may be written as

$$q_1 = u + l \left[\frac{du}{dy} \right]_w, \quad (7)$$

where l denotes the mean free path. On combining two equations, the slip velocity reduces to

$$u = \sigma l \left[\frac{du}{dy} \right]_w, \quad (8)$$

where the coefficient σ is defined as $(2 - \theta)/\theta$. The comparison between Eqs. (6) and (8) yields the following expression for the slip coefficient:

$$s = -\frac{\sigma}{\eta}l.$$

The slip model (8) by Maxwell becomes in the dimensionless form

$$u = \sigma \frac{\text{Kn}}{p} \left[\frac{du}{dy} \right]_w. \quad (9)$$

As expected, the only parameter appearing in this model is the Knudsen number, which measures the level of the slip by the gas-surface molecular interaction. Since the same method can be applied to derive the temperature slip boundary condition, the discussion of temperature slip is omitted in the present work.

From this model, it is also possible to derive more refined versions, for example, high order slip boundary conditions.^{23,24} All of these more elaborate models, however, yield the simple Maxwell slip model in the limit of small Knudsen number, so that it was argued that the Maxwell slip model should be sufficient for most of the slip phenomena. Nonetheless, as the need for treating the more challenging problem has grown rapidly in recent years, there exist some serious difficulties, especially in implementing the model for the numerical codes. The main difficulty comes from the accommodation coefficient σ whose values have to be found by other means. In practice, their values are chosen such that they fit best for the experimental data, and they are tabulated for various gases and surfaces. There remains, however, a need for developing an efficient procedure capable of ex-

plaining the subtle differences concerning the gas and surface temperature, and type of gas (for example, monatomic or diatomic gas) and surface molecules. Another difficulty arises also from the accommodation coefficient in the mathematical sense. The expression by the Maxwell slip model is not well-defined mathematically in the limit of vanishing diffusive reflection and, depending on situations, the value of slip is not bounded, which can cause severe problems in the numerical implementation of the model. It was reported in the study⁵ of modeling separation in rarefied gas flows that the Maxwell slip model can cause the reversal of slip velocity and an overshoot of slip velocity near the reattachment point. Furthermore, it can be shown by a simple analysis that in order to ensure the numerical stability for no change in the sign of vorticity at the wall the Knudsen number should be less than the size of the grid, restricting the range of the model significantly, especially if the grid size is refined near the wall. To deal with some of these difficulties, a slip model of the gas-surface molecular interaction based on Langmuir's theory of the adsorption of gases on solids is considered in the present work.

III. GAS-SURFACE MOLECULAR INTERACTION: LANGMUIR SLIP MODEL

Another way to include the slip effect near the surface is to take into account the interfacial interaction between the gas molecules and the surface, which itself consists of molecules. It is well known through numerous studies¹¹⁻¹³ on surfaces that the gas molecules do not in general rebound elastically but inelastically interact with the surface of the solid owing to a long range attractive force, and consequently the gas molecules can get adsorbed on the surface (condensation) and then desorbed after some time lag (evaporation). This picture results in the deposition of a layer with a thickness of one or more molecules on the surface. This is known as adsorption in the literature. On the basis of this concept of adsorption it is possible to derive a slip model of the gas-surface molecular interaction.

A. Langmuir's theory of adsorption

The critical part in the theory of adsorption rests on the amount of gas adsorbed, which is in general a function of pressure and temperature. If we consider the amount of gas at constant temperature, the so-called adsorption isotherm can be defined. There exist various adsorption isotherms, but among them the isotherm developed by Langmuir¹¹ stands out most because it has proven to be extremely useful both in explaining experimental data and for theoretical treatments. The Langmuir adsorption isotherm can be derived from the molecular viewpoint by employing statistical thermodynamics. In the present study we will follow the derivation^{9,10} based on a mechanical picture which is essentially that given by Langmuir. A more elaborate derivation may be found in the textbook on physical chemistry of surfaces.¹³

Let us assume that there are N sites per unit area of the surface interacting with the gas molecules. Also, let α be the fraction of the surface covered by adsorbed atoms at thermal equilibrium. Then the number of sites which are not covered

is $N(1-\alpha)$. If we model this interaction as a chemical reaction in which the gas molecule m and the site s form the complex c , the equilibrium constant K can be expressed in the concentration of m , s , and c ,

$$K = \frac{C_c}{C_m C_s}. \quad (10)$$

Since $C_m = p/k_B T_w$, $C_s = N(1-\alpha)$, $C_c = N\alpha$ where k_B is the Boltzmann constant, we can derive the following form known as the Langmuir hyperbolic adsorption isotherm:

$$\alpha = (1-\alpha)\beta p$$

or

$$\alpha = \frac{\beta p}{1 + \beta p}, \quad (11)$$

where $\beta = K/k_B T_w$ and K is a function of the interfacial interaction parameters. The fraction α is a function of the pressure p , the equilibrium constant K , and the surface temperature T_w . As the pressure increases, its value approaches unity, implying that most of molecules are at thermal equilibrium.

This derivation can also be extended to treat the adsorption of a diatomic gas, such as nitrogen, on the solid surface.¹² In order that a gas molecule approaching the surface may be adsorbed on the surface, two particular elementary sites must be vacant. The chance of one of these sites being vacant is $(1-\alpha)$; that both sites will be vacant is $(1-\alpha)^2$. Thus the rate of condensation is proportional to $(1-\alpha)^2$. Evaporation only occurs when adsorbed atoms are in adjacent sites. The chance that an atom shall be in a given site is α and therefore the chance that atoms shall be in adjacent sites is equal to α^2 . Thus the rate of evaporation of molecules from the surface is proportional to α^2 . From this reasoning, the following adsorption isotherm for a diatomic gas can be found:

$$\alpha^2 = (1-\alpha)^2 \beta p$$

or

$$\alpha = \frac{\sqrt{\beta p}}{1 + \sqrt{\beta p}}. \quad (12)$$

It is also straightforward to treat the adsorption of a gas mixture within the present formulation. Suppose that a mixture consists of two gas components (nitrogen and oxygen) and V denotes the fraction of components in volume, then the fraction of the surface at thermal equilibrium may be expressed as ($V_{N_2} + V_{O_2} = 1$)

$$\alpha = V_{N_2} \frac{\sqrt{\beta_{N_2} p}}{1 + \sqrt{\beta_{N_2} p}} + V_{O_2} \frac{\sqrt{\beta_{O_2} p}}{1 + \sqrt{\beta_{O_2} p}}. \quad (13)$$

B. Langmuir slip models

With information of the fraction of surface covered at equilibrium α determined by the Langmuir adsorption isotherm, it is possible to develop a slip model for the gas–

surface molecular interaction. The velocity and temperature slip can be expressed, in the dimensional form, as^{9,10}

$$u = \alpha u_w + (1-\alpha)u_g, \quad (14)$$

$$T = \alpha T_w + (1-\alpha)T_g, \quad (15)$$

where the subscript g denotes the local value adjacent to the wall—for example, a mean free path away from the wall. The only parameter requiring a further investigation is β or specifically the equilibrium constant K . The equilibrium constant may be calculated by using the partition function as follows:

$$K = \frac{Q_c}{Q_m Q_s}. \quad (16)$$

The partition function of the sites Q_s can be assumed unity because the sites are fixed in the surface. The partition function Q_m of the gas molecules per unit volume can be expressed as a combination of translational and internal energy and thus it becomes

$$Q_m = Q_m^{\text{int}} Q_m^{\text{trans}} = Q_m^{\text{int}} (2\pi m k_B T/h^2)^{3/2}, \quad (17)$$

where m and h are the molecular mass and the Planck constant, respectively. The partition function Q_c depends on the details of the gas–surface molecular interaction and thus it takes different forms for different models of the motion on the surface. In the case of physical adsorption, the complex would have a loose structure and thus it may be described as an almost translational motion of the gas molecule in the field of an attractive potential energy of magnitude D_e . In this model, the partition function of the complex Q_c may be expressed as

$$Q_c = Q_c^{\text{int}} Q_c^{\text{trans}} = Q_c^{\text{int}} (2\pi m k_B T/h^2)^{3/2} A \ell \exp\left(\frac{D_e}{k_B T_w}\right), \quad (18)$$

where A is the mean area of a site and ℓ is a mean collision distance between the wall surface and the gas molecules. The inclusion of the term $\exp(D_e/k_B T_w)$ means that Q_c is referred to the gaseous state. The D_e denotes the potential parameter (heat of adsorption) which measures how strong the surface–molecular interaction is and it will have different values for the different combination of the gas molecules and the surface atoms. Then the equilibrium constant reduces to

$$K = \frac{Q_c^{\text{int}}}{Q_m^{\text{int}}} A \ell \exp\left(\frac{D_e}{k_B T_w}\right). \quad (19)$$

If we introduce an approximation that the internal partition functions are the same for the two states m and c it can be further simplified, and then the parameter β takes the following form:

$$\beta = \frac{A \ell}{k_B T_w} \exp\left(\frac{D_e}{k_B T_w}\right). \quad (20)$$

A more detailed derivation of the equilibrium constant K may be found in Refs. 10 and 13. When the characteristic length L is taken equal to ℓ then the parameter can be, after some manipulation, simplified into the form

$$\beta = \sqrt{\frac{\pi}{2}} \frac{A}{4c^2 d_r^2} \frac{T_r}{T_w} \exp\left(\frac{D_e}{k_B T_w}\right) \frac{1}{p_r \text{Kn}}, \quad (21)$$

where the subscript r stands for the reference state and

$$c = \left[\frac{2\sqrt{\pi}}{5} A_2(\nu) \Gamma[4 - 2/(\nu - 1)] \right]^{1/2}.$$

Here Γ denotes the gamma function, d is the diameter of the molecule, and ν is the exponent of the inverse power law for the gas particle interaction potential. The $A_2(\nu)$ is a pure number; its tabulated values are available in the monographs on kinetic theory.²² In deriving this equation, in addition to the equation of state and the definition of c , the following relations were used:

$$l = \sqrt{\frac{\pi}{2}} \frac{\eta}{\rho \sqrt{RT}},$$

$$\eta = \frac{5}{8A_2(\nu) \Gamma[4 - 2/(\nu - 1)] d^2} \sqrt{\frac{mk_B T}{\pi}},$$

$$d = \left(\frac{\kappa}{2k_B T} \right)^{1/(\nu - 1)},$$

where R is a gas constant and κ is the coefficient of the inverse power laws. Finally, if the area covered by a molecule A is approximated as $\pi d^2/4$, the parameter β reduces to

$$\beta = \frac{1}{4\omega \text{Kn}} \frac{1}{p_r}, \quad (22)$$

where

$$\omega = \sqrt{\frac{2}{\pi}} \frac{4c^2}{\pi} \left(\frac{T_w}{T_r} \right)^{s+1/2} \exp\left(\frac{-D_e}{k_B T_w}\right)$$

and $s = \frac{1}{2} + 2/(\nu - 1)$. The coefficient ω will be shown to be very similar to the slip coefficient σ of the Maxwell model in the next section. For most of molecular interaction models, the value of the heat of adsorption D_e falls under the range

$$D_e = O(10^{-1} \sim 10) \text{ kcal/mol.}$$

Its value may be inferred from experimental data or theoretical prediction of intermolecular forces. It was known from previous studies^{12,13} that the lighter gas molecules such as helium have smaller values of the heat of adsorption in comparison with the heavier molecules such as argon. The easier way to determine its value, however, would be the use of the experimental data of some benchmark problems, which will be given in the next section.

This completes the derivation of a gaseous slip model based on the Langmuir adsorption isotherm. In summary, the idea of the Langmuir slip model can be explained by the statement that all the molecules adsorbed are desorbed after some time lag with a totally random velocity and thus the fraction of molecules undergoing diffusive reflection is proportional to the fraction of molecules adsorbed out of all the molecules approaching the surface. And this fraction of adsorbed molecules can be given by the Langmuir hyperbolic adsorption isotherm. In order to discuss its characteristics let

us consider the simplest case; velocity slip of a monatomic gas at the stationary surface. The resulting slip model can be expressed as

$$\frac{u}{u_g} = (1 - \alpha) = \frac{1}{1 + \beta p} \quad (23)$$

or in the dimensionless form

$$u = \frac{1}{1 + p/4\omega \text{Kn}} = \frac{1}{1 + \bar{\beta} p}, \quad (24)$$

where

$$\bar{\beta} = \frac{1}{4\omega \text{Kn}} \quad (25)$$

and ω is a function of ν , T_w , and D_e . Thus the slip in the Langmuir model depends largely on the parameters Kn , D_e , ν , and T_w . As the density or pressure increases, the slip velocity vanishes. On the other hand, as the Knudsen number increases, the dimensionless slip velocity approaches unity, meaning the full slip to the reference velocity. Notice that due to its special form the slip value in the present model is always bounded; $0 \leq u \leq u_g$. This ensures the preservation of the sign of vorticity near the wall. Therefore, the present model circumvents the difficulty of the Maxwell model related to the reversal of slip velocity and overshoot of slip velocity near the reattachment point in a separated gas flow. In connection with this, it is instructive to note that the following second-order Maxwell model considered in the work of Beskok and Karniadakis (Ref. 23) can be explained in the context of the Langmuir model,

$$u = \sigma \left(\frac{\text{Kn}}{p} \left[\frac{du}{dy} \right]_w + \frac{(\text{Kn}/p)^2}{2!} \left[\frac{d^2 u}{dy^2} \right]_w + \dots \right)$$

$$\simeq \frac{1}{-b/\sigma + p/\sigma \text{Kn}} \left[\frac{du}{dy} \right]_w, \quad (26)$$

where b is a parameter to be determined. The first equality in this equation can be derived by keeping the higher-order terms in the Taylor series of the mean velocity of molecules q_1 in Eq. (7). In that work the following parameter was proposed:

$$b = \frac{1}{2} \left[\frac{d^2 u / dy^2}{du / dy} \right]_w$$

and set as $b = -1$. The reasoning for this modification becomes obvious when we compare models (24) and (26) and also note that the two consecutive derivatives in the Taylor series expansion of the function (24) or a similar function always have a different sign. The ultimate origin of this property is associated with the boundedness of the slip velocity in the limit of large Knudsen number, which is automatically satisfied in the case of the Langmuir slip model.

The implementation of the present model to the multi-dimensional numerical codes is straightforward. Since its mathematical form is of Dirichlet type rather than of Neumann type, there is no need for changing the basic structure from the stick boundary condition. Furthermore, if we inter-

pret the value of the slip velocity as the magnitude of the velocity vector and use the local information adjacent to the surface for the direction of the velocity, the present model can be applied to gas flows under any flow configurations since the gas molecule–surface interaction is local with its range being a few nanometers.

IV. VALIDATION STUDY

As the slip phenomena have remained a knotty problem, the validation study of the slip models has also been considered a very delicate and, often in many cases, vexatious task. The cause of this situation can be explained by recalling that not only the slip effect from the gas–surface molecular interaction but also the non-Newtonian effect is required to validate the slip model. The latter, especially, means that complicated nonlinear constitutive equations beyond the linear theory must be taken into account. Therefore, if one is interested in undertaking the validation study for some problems with non-negligible non-Newtonian effect, one is immediately confronted with a daunting task to consider the nonlinear constitutive equations, which cannot be performed without reckoning on a numerical method or an approximate technique. In principle, high order equations such as the Burnett equation or particle simulation methods^{25,26} may be employed to calculate the non-Newtonian effect in the bulk region, but it has not been very effective owing to numerical uncertainty, in particular, for the low speed gas flows.

In the present study we will circumvent this difficulty by considering a unique situation in which only the slip effect from the gas–surface molecular interaction is important but the non-Newtonian effect in the bulk region is small. Fortunately, this can be found in the *low speed microscale* gas flows,^{27–29} which have been investigated by recent experiments. Since the Mach number is extremely small in this case, for relatively large values of the Knudsen number the non-Newtonian effect can be assumed to be very small. As a result, the Navier–Stokes equations can be used as the governing equations. Furthermore, the low Mach number assumption can lead to a perturbation solution of the Navier–Stokes equations in the case of the very long channel flow.

A. Pressure-driven microchannel gas flows

As a first test case, pressure-driven internal isothermal gas flows in a very long microchannel of high aspect ratio will be considered. With a correction factor accounting for the different geometry, the flow in a very long microtube can also be treated by the present method. Using the method described by Arkilic *et al.*,²⁷ the compressible Navier–Stokes equations, at the zeroth order of ϵ , may be written as

$$\epsilon \frac{\partial(pu)}{\partial x} + \frac{\partial(pv)}{\partial y} = 0, \quad (27)$$

$$\frac{24}{\delta} \frac{dp}{dx} = \frac{\partial^2 u}{\partial y^2}, \quad (28)$$

where

$$\delta = 24 \frac{N_\delta}{\epsilon}.$$

In this expression ϵ represents the ratio of the channel height (H) to its length (L). It should be noted that the term associated with the bulk viscosity of a diatomic gas does not enter the present perturbation analysis to the lowest order and thus the present equations also hold for a diatomic gas. The streamwise coordinate x and the coordinate y normal to the wall are nondimensionalized by L and H . The u and v denote the streamwise and normal velocity components. The geometry of the channel is represented by $0 \leq x \leq 1$ and $-1/2 \leq y \leq 1/2$. The reference state is chosen as the exit conditions ($x=1$). The reference velocity u_r is the area-averaged streamwise velocity at the channel exit, or the maximum velocity condition in this particular flow problem. Critical constraints on which the present analysis is based are

$$\epsilon \ll 1, \quad M \ll 1, \quad O(\epsilon/N_\delta) \approx 1. \quad (29)$$

At this stage a few comments must be added with regard to the applicability range of the present analysis and the implication of the aforementioned constraints. First, in these microchannel gas flows the pressure may strongly change owing to the viscous effects even though the speeds may not be sufficiently high for the Mach number to go beyond the traditional threshold of 0.3.³⁰ For this reason the flow is treated as compressible rather than incompressible. Second, the value of ϵ and N_δ should be very small and at the same time should be considered finite. Once infinitely long channel or incompressibility is assumed, the resulting pressure distribution along the channel is always linear, which can exclude all the subtle effects relating to rarefaction and compressibility from the beginning. It can be said from these points that a careful check on the underlying assumptions must be made if one tries to compare the present analysis with other studies.

1. Velocity slip in a monatomic gas

With the Langmuir slip model of a monatomic gas (23) or (24), the dimensionless slip velocity at the surface can be expressed as

$$u = \frac{u(x, y=0)}{1 + \bar{\beta}p}.$$

First, by applying flow symmetry conditions and the aforementioned slip boundary condition to Eq. (28), and then by inserting the so determined streamwise velocity profile into Eq. (27), we can derive an equation for the velocity normal to the wall. By further applying the vanishing normal velocity at the wall, a solvability condition of the pressure distribution can be derived,

$$(p^2)'' = -\frac{3}{\bar{\beta}} p'', \quad (30)$$

where the prime denotes the differentiation in the streamwise coordinate x . Finally, by utilizing the pressure conditions at the inlet and exit, the following exact solutions—

dimensionless streamwise velocity $u(x,y)$, the pressure distribution along channel $p(x)$, and the dimensional mass flow rate \dot{m} —can be obtained:

$$u(x,y) = -\frac{3}{\delta} p' \left[1 - 4y^2 + \frac{1}{\bar{\beta} p} \right], \quad (31)$$

$$1 - x = \frac{1 - p^2 + 3[1 - p]/\bar{\beta}}{1 - p_{in}^2 + 3[1 - p_{in}]/\bar{\beta}}, \quad (32)$$

$$\dot{m} = \frac{H^3 W p_{out}^2}{24 \eta L R T} \delta, \quad (33)$$

where

$$\delta = -p'_{x=1} \left(2 + \frac{3}{\bar{\beta}} \right) = p_{in}^2 - 1 + 3(p_{in} - 1)/\bar{\beta}.$$

Here the $p'_{x=1}$ represents the x derivative of the pressure profile at the exit condition. The W denotes the channel width. As expected, in the pressure-driven internal flow the value of δ is determined in terms of the pressure difference and plays a dominant role in the mass flow rate. In the limit of large $\bar{\beta}$ or small Kn, the continuum (no-slip) solutions can be obtained: for example, the pressure distribution along the channel takes the following nonlinear form:

$$p(x) = [x(1 - p_{in}^2) + p_{in}^2]^{1/2}.$$

A very instructive result can be obtained from this simple analysis of the low speed microchannel flow. By applying the same technique, if the following equivalence relation is introduced,

$$\sigma = \omega, \quad (34)$$

it is straightforward to show that exactly the same solutions can be derived by using the first-order Maxwell slip model (9) in place of the Langmuir slip model. This is a very surprising result since the two slip models are developed from the totally independent consideration of the gas–surface molecular interaction. The implications of this finding are twofold: with the introduction of relation (34) a new physical meaning may be assigned to the accommodation coefficient of the Maxwell slip model; and the Langmuir slip model recovers the Maxwell slip model in the first-order approximation in the case of the microchannel flow. The latter means that previous validation results of the microchannel flow using the Maxwell model—for example, the works by Arkilic *et al.*^{27,31}—can carry over to the validation study of the present model without any change.

Another interesting observation can be made if the reference velocity in the Langmuir slip model is changed into the overall maximum velocity, rather than the velocity adjacent to the surface or the *local* centerline velocity in the channel. In this case, the slip boundary condition becomes

$$u = \frac{u(x=1,y=0)}{1 + \bar{\beta} p},$$

and then a solvability condition can be written as

$$(p^2)'' = -\frac{3(1 + \bar{\beta})}{\bar{\beta}(1 + \bar{\beta} p)^2} p'_{x=1} p'. \quad (35)$$

Even though this ordinary differential equation of $p(x)$ seems at first glance very complicated, it can be analytically solved by utilizing the following integral formula:

$$\int \frac{p^2 + C_1 p}{p + C_2} dp = \frac{p^2}{2} + (C_1 - C_2)p + C_2(C_2 - C_1) \ln(p + C_2),$$

where C_1 and C_2 are integration constants. Applying the pressure conditions at the inlet and exit results in the following solutions:

$$u(x,y) = -\frac{3}{\delta} \left[p'(1 - 4y^2) + \frac{1 + \bar{\beta}}{\bar{\beta}(1 + \bar{\beta} p)} p'_{x=1} \right], \quad (36)$$

$$1 - x = \frac{f(1) - f(p)}{f(1) - f(p_{in})}, \quad (37)$$

where

$$\delta = -p'_{x=1} \left(2 + \frac{3}{\bar{\beta}} \right) = \frac{2\bar{\beta}(2\bar{\beta} + 3)[f(p_{in}) - f(1)]}{(2\bar{\beta}^2 - 3)}.$$

In this expression the function $f(p)$ is defined as

$$f(p) \equiv \frac{ap}{\bar{\beta}^2} + \frac{p^2}{2} - \frac{a}{\bar{\beta}^2} \left(\frac{1}{\bar{\beta}} - \frac{a}{\bar{\beta}^2} \right) \ln(\bar{\beta}^2 p + \bar{\beta} - a),$$

where

$$a = \frac{3\bar{\beta}(\bar{\beta} + 1)}{3 - 2\bar{\beta}^2}.$$

In the later discussion these solutions will be referred to as the *global* Langmuir solution, as opposed to the local Langmuir solution obtained in the previous case. In order that the solutions shall remain meaningful, the following condition from the well-posedness of the parameter δ that includes a natural logarithm term should be satisfied:

$$\frac{\bar{\beta}^2 p_{in} + \bar{\beta} - a}{\bar{\beta}^2 + \bar{\beta} - a} > 0$$

or

$$\bar{\beta} > \frac{[1 + 6p_{in}(p_{in} - 1)]^{1/2} - 1}{2p_{in}}. \quad (38)$$

This condition, mainly of the pressure difference p_{in} and $\bar{\beta}$ or the Knudsen number, simply tells us that for a given Knudsen number there exists an upper limit of the pressure difference. Since the value of N_δ or δ for a given value of ϵ is proportional to the pressure difference, this in turn means that the validity of the present analysis represented by condition (29) is ensured only when the aforementioned condition is met. The presence of such constraint whose mathematical origin can be traced to the appearance of the term

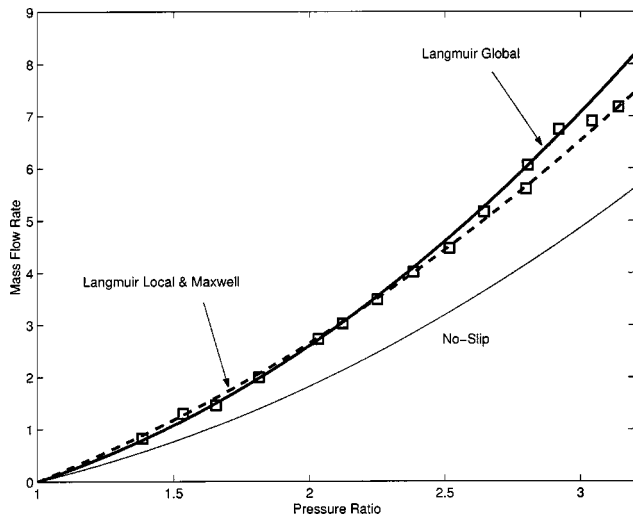


FIG. 2. Mass flow rate (kg/s) of helium gas in the scale of 10^{-12} depicted as a function of pressure ratio ($Kn=0.158$). The square represents the experimental data (Ref. 29).

$p'_{x=1}$ in the differential equation (35) may be explained with the help of the argument of Ref. 32 that whereas the calculations in the first-order approximation in general do not show in themselves the range of their validity, higher order calculations may show the range of their validity within the formulation. Thus it is possible to infer that the higher order effects have been incorporated in the present global model.

With these analytical results in hand, comparisons with experimental data are performed for helium gas in Figs. 2–4. A recent experimental work on gas flows in uniform microchannels by Shih *et al.*²⁹ is utilized. The dimensions of the microchannel are as follows:

$$H = 1.2 \mu\text{m}, \quad L = 4000 \mu\text{m}, \quad W = 40.0 \mu\text{m}.$$

The channel is manufactured by the method originally designed to fabricate integrated circuit chips and is made of silicon with a very smooth surface. The mass flow rate and

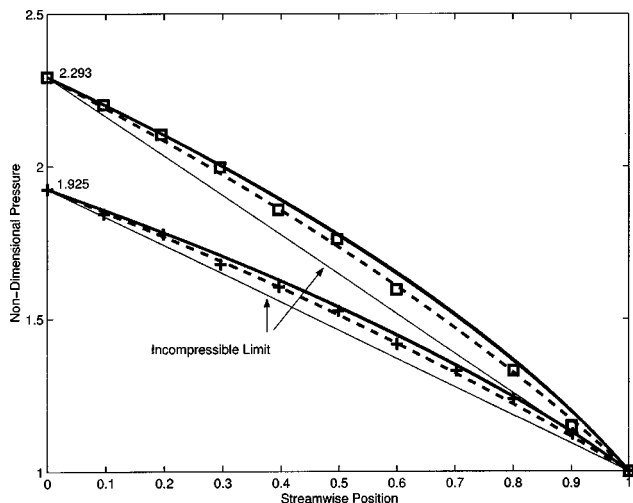


FIG. 3. Pressure distribution of helium gas along the microchannel. The Maxwell (or Langmuir local) model by the dashed lines; the Langmuir global model by the thick solid lines; the experimental data (Ref. 29) by the symbols.

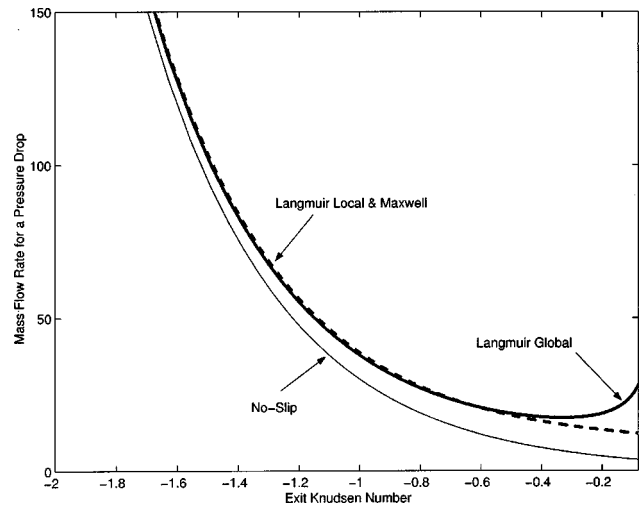


FIG. 4. Variation of mass flow rate of helium gas per pressure drop ($p_{in} = 2$) as a function of the exit Knudsen number in logarithmic scale. The dimensional value can be recovered by multiplying a factor $\sqrt{(\pi/2)} \times (H^2 W / 24 L \sqrt{RT})$.

the pressure distribution along the channel are measured. Other important conditions concerning the experimental setup and the physical properties of helium gas are

$$T = 293 \text{ K}, \quad p_{out} = 1 \text{ atm}, \quad R = 2077 \text{ J/kg K},$$

$$\eta = 1.97 \times 10^{-5} \text{ N s/m}^2, \quad \nu = 14, \quad c = 1.046.$$

The Knudsen number based on the density at the exit and the channel height is 0.158. According to the conventional classification based on the Knudsen number, the flow belongs to the transition regime. Even though the pressure difference is raised by a maximum value of 2.14 atm, the range of the nonequilibrium parameter N_δ remains an order of $O(\delta\epsilon/24) \approx 10^{-4}$, implying that the flow falls safely under the low speed microscale (creeping) regime. By following the same spirit as found in other studies, the values for the adjustable parameter are taken so as to best fit the experimental data. The accommodation coefficient σ of the Maxwell slip model and the adsorption coefficient of the local Langmuir slip model chosen in the present work are

$$\sigma = 0.721, \quad \omega = 0.721 \text{ or } D_e = 0.252 \text{ kcal/mol}.$$

In the case of the global Langmuir slip model the following value turns out to fit best the experimental data:

$$\omega = 0.408 \text{ or } D_e = 0.584 \text{ kcal/mol}.$$

In Fig. 2, dimensional mass flow rates of helium gas flows in the microchannel are depicted as a function of pressure ratio. Using the values assigned to the accommodation and adsorption coefficients, all of the slip models—either Maxwell or Langmuir—seem to predict the experimental values qualitatively correctly. Only a minor difference was found for the flow driven by the high pressure difference in which the global Langmuir slip model tends to predict slightly larger mass flow rate. This may reflect its characteristics associated with the higher order effects. On the other hand, the continuum model without slip correction underestimates the mass flow rate, especially in the case of high

of the inverse Knudsen number. To be consistent with the diffusive reflection assumption, the following values are assigned to the accommodation coefficients:

$$\sigma = 1, \quad \omega = \frac{3}{4}.$$

As can be seen in Fig. 7, the Stokes model without slip correction overestimates the drag coefficient, especially in the high Knudsen number regime, while the Maxwell and Langmuir slip models yield values closer to the experimental or accurate theoretical data. In particular, qualitative agreement with the data can be found near the low Knudsen number regime. In addition, a closer look indicates that in comparison with the Maxwell model the Langmuir model yields a prediction in better agreement with the data. On the other hand, both models deviate from the data in the high Knudsen number regime and fail to converge at the free molecular limit. This is, however, not an unexpected result, since the slip by the non-Newtonian effect of gas flow is excluded in the present slip model. Thus the discrepancy at the free molecular limit may imply that the inclusion of a proper nonlinear constitutive equation becomes essential in describing the flow past a sphere with high Knudsen numbers.

V. CONCLUDING REMARKS

As a step toward developing a robust slip model for rarefied and microscale gas flows, the theory based on Langmuir's adsorption isotherm has been studied. The emphasis has been placed on the extension to a diatomic gas and issues of practical implementation of the model under complex flow configurations. In order to circumvent the difficulty involving the nonlinearity of the constitutive equation the validation study was restricted to an analytically solvable situation where only the interaction of gas molecules with the surface molecules remains significant. With the help of recent experimental results on the microchannel gas flow and various theoretical and experimental results on the low speed gas flow of a sphere it is shown that with an adjustable parameter the model in low-speed creeping regime with moderate Knudsen numbers yields a prediction in qualitative agreement with the data. In the course of this study it has also been found that the new model can provide a more detailed explanation of the accommodation coefficient and the role played by the difference in gas molecules.

Owing to the unavailability of experimental data, the present study was limited to the velocity slip phenomena, but it is readily extendable to the temperature slip case. The preliminary study of the temperature slip associated with the convective heat transfer in the microtube or microchannel indicates that the influence of the temperature slip can be analyzed with the help of the solutions expressed in terms of hypergeometric functions. However, the extension of the present work to other complicated problems, for example, liquid slip, may present nontrivial challenges. The study of these problems will be reported in the future work.

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