GAS MOLECULE–MOLECULE INTERACTION AND THE GAS–SURFACE SCATTERING INFLUENCE ON THE RAREFIED GAS FLOW THROUGH A SLIT INTO A VACUUM

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The gas molecule–molecule interaction and the gas–surface scattering influence on the gas flow through a slit into a vacuum are investigated in a wide range of gas rarefaction using the direct simulation Monte Carlo method. To study the gas molecule–molecule interaction influence, we use the variable hard sphere and variable soft sphere models defined for an inverse-power-law potential and the generalized hard sphere model defined for the 12-6 Lennard-Jones potential. The Maxwell, Cercignani–Lampis, and Epstein models were used to simulate the gas–surface scattering. This study demonstrates that the gas molecule–molecule interaction significantly interferes with the gas flow through a slit, while the influence of the gas–surface scattering is negligibly small. The presented numerical results are in agreement with the corresponding experimental ones.

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1. INTRODUCTION

The study of gas flow through capillaries has both theoretical interest and practical applications. In particular, the latter are related to the gas–dynamic process simulation in micro- and nano-electromechanical systems (MEMS/NENS) [1]. To perform a simulation of the gas–dynamic process in MEMS/NENS, the following process parameters should be taken into account: the gas rarefaction, the gas flow velocity, the gas–surface scattering, the gas molecule–molecule interaction, the surface roughness, and the geometry and ratio of the surface area to the volume of the device. This study improves understanding of the gas–surface scattering and the gas molecule–molecule interaction effect on the gas flow through an infinitesimally thin capillary with the rectangular cross section (or slit) into a vacuum.

In our opinion, there is a shortage of studies available in the open literature providing estimates of the gas–surface scattering and the gas molecule–molecule interaction influence on the gas flow through a slit. The gas–surface scattering effect on the gas flow through a slit in the case where the pressure difference is small

(i.e., when the gas flow is rather slow) was investigated theoretically in [2]. The Maxwell specular–diffuse scheme was used there to simulate the gas–surface scattering, with the result that the gas–surface scattering does not significantly affect the mass flow rate through a slit for any gas rarefaction. As far as we know, there is no research in the open literature that describes the case of a large pressure difference, such as the gas flow into a vacuum, i.e., when the gas flow is rather rapid. In fact, the Mach number can be significantly greater than unity when gas expands into a vacuum [3].

Experimental research of the gas flow through a slit related to the role of the gas–surface scattering and gas molecule–molecule interaction in this process has been done a few decades ago [4]. The experimenters studied noble and some polyatomic gases flowing through glass slits in a wide range of the slit size and gas rarefaction. A noticeable difference was observed between the values of the dimensionless flow rate (normalized by its value in the free molecular flow regime) for various gases in the transition and free molecular flow regime. Strictly speaking, slits used in experimental research are short rectangular channels, and hence the difference can equally be attributed to the gas molecule–molecule interaction or the gas–surface scattering. In the case of

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the free molecular regime (i.e., when gas molecules collide with the surface much more often than with each other), this difference is due only to the different gas–surface scattering, but in the transition regime, it may be the result of both reasons.

Relatively recently, we have performed a series of experiments [5, 6], whose results demonstrate that the effect of gas–surface scattering on the mass flow rate through a tube (a capillary with a circular cross section) in the free molecular regime is sufficiently strong. The mass flow rate through a tube into a vacuum can vary significantly when the tube surface varies from contaminated to atomically clean conditions. For instance, the mass flow rate of helium (He) through a tube with an atomically clean silver (Ag) surface is 62 % higher than through a tube with contaminated surface. It is also 39 % higher for neon (Ne), 23 % for argon (Ar), and 15 % for krypton (Kr).

This study continues the investigation in [7], where we described the statement of the problem in detail and substantiated the chosen method of study, the direct simulation Monte Carlo method [8] based on the majorant frequency technique [9] with application of the weight-factor and subcell procedures; we also estimated the simulation parameters, in particular, the characteristics of a nonhomogenous grid and the time step length that guarantee the computation error not exceeding 0.2 %. The computation data on the mass flow rate through a two-dimensional slit into a vacuum in a wide range of rarefaction in the case of the hard sphere (HS) model for the gas molecule–molecule interaction and the diffuse law for the gas–surface scattering model is considered the key result of the previous study. The mass flow rate values presented in the previous work are in good agreement with the results published by other authors.

For this study, we have adapted the code used previously to investigate the effect of the gas molecule–molecule interaction and gas–surface scattering on the rarefied gas flow through the two-dimensional slit into a vacuum. As before, the key interest is in the dimensionless mass flow rate $Q^* = Q/Q_{fm}$, normalized by the free molecular value, as a function of the rarefaction parameter $\delta = hP/\mu v = \sqrt{\pi}/2\text{Kn}$, where h is the height of the slit and P, μ, and v are the gas pressure, viscosity, and the most probable molecular velocity in the upstream container far away from the slit. The rarefaction parameter δ is inversely proportional to the Knudsen number, defined for a slit as $\text{Kn} = \lambda/h$, where λ is the mean free path of gas molecules.

2. INFLUENCE OF THE GAS MOLECULE–MOLECULE INTERACTION

The procedure using an intermolecular potential based on the HS model is a simple gas molecule–molecule interaction (or collision) simulation procedure. The HS model is a special ($\eta = \infty$) case of the repulsive inverse-power-law (IPL) potential $U(r) \sim r^{-\eta}$, where r is the distance between molecules and η is the IPL exponent. According to this model, the molecule collision process is described by a constant total cross section σ_t and by isotropic scattering in the center-of-mass frame of reference. The HS model is suitable to simulate the elastic collision process of gas molecules. The theory based on the HS model postulates the relation $\mu \sim \sqrt{T}$ between the viscosity coefficient and the temperature.

A good fit to analytic calculations is the model of Maxwell's molecules, which is a special case of the IPL potential with $\eta = 5$. Indeed, according to this model, the probability of the collision of two molecules is a constant value; this fact significantly simplifies both theoretical analysis and the direct simulation Monte Carlo algorithm.

The simplicity and ease of the application are important virtues of the HS and Maxwell-molecule models. But these models are not applicable for simulating collisions in the case of real gases because they are "too hard" and "too soft" for the gas molecule–molecule interaction. A considerable disadvantage of these models is the absence of the cross-section dependence on the relative velocity of the colliding particles.

To eliminate this disadvantage, the variable hard sphere (VHS) model was developed in [10]. The VHS model is defined for the general IPL potential. According to the VHS model, the diameter d of colliding molecules is inversely proportional to the relative velocity of gas molecules v_r raised to the power $\omega - 1/2$, i.e., $\sigma_t \sim v_r^{-2\omega+1}$, where ω, being a parameter of the model, is the viscosity index derived from the temperature dependence of the viscosity coefficient. Both the IPL potential and the VHS model imply that the dependence of the viscosity coefficient μ on the temperature is proportional to T^ω. The viscosity index ω is related to the IPL exponent η as $\omega = 2/(\eta - 1) + 1/2$ and varies depending on the kind of the gas.

The primary drawback of the VHS model is that the theory based on it leads to formulas for the viscosity cross section σ_μ and the diffusion cross sections σ_D that do not correspond to the chosen IPL potential. Indeed, according to the VHS model, the ratio σ_μ/σ_D is a constant equal to $2/3$, as in the HS model. But ac-
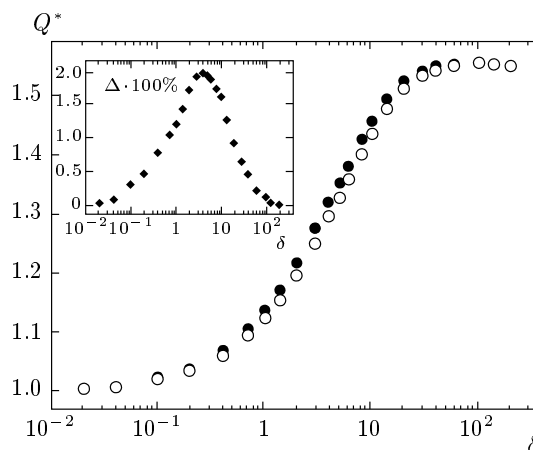
cording to the IPL potential, this ratio should depend on η.

For the VHS model, as well as for the HS model, the isotropic scattering law holds for molecular collisions, but this law is not realistic in general. In [11, 12], the variable soft sphere (VSS) model was suggested, which can be used for any kind of the intermolecular potential with definite viscosity and diffusion cross sections. According to this model, the diameter d of colliding particles depends on the relative velocity of molecules as in the VHS model, but the angle of the change of the relative velocity direction after the molecule collision (the deflection angle χ) is defined by the scattering law $b = d \cos^{\alpha}(\chi/2)$, which is more realistic than the isotropic law (here, b is the impact parameter). The value of α should be chosen so as to make the values of the viscosity cross section σ_{μ} and the diffusion cross section σ_D agree with the chosen interaction potential. In the case of an IPL potential, the parameters involved in the VSS model can be determined explicitly, and therefore the VSS model is typically used in practical calculations only for IPL potentials.

However, the IPL potential, which describes only the repulsion between molecules, becomes incorrect in the low-temperature range, where attraction dominates. It is known that under the condition $kT/\varepsilon \gg 1$, the interaction potential can be considered to be fully repulsive, and in the case $kT/\varepsilon \ll 1$, it is attractive; ε denotes the well depth of the repulsive–attractive interaction potential of colliding particles and k is the Boltzmann constant.

To take attractive intermolecular forces into account, the authors of [13] suggested the so-called generalized hard sphere (GHS) model of the intermolecular potential that contains both repulsive and attractive components. In the GHS model, the post-collision direction of the gas molecule relative velocity is the same as in the HS model, but the total cross section σ_t is a function of the impact energy of the collision.

The GHS model was further developed in [14] in the case of heavy gas molecules interacting through an attractive–repulsive potential when a strong attraction is assumed. According to the authors, if the intermolecular potential is represented as a sum of a repulsive and an attractive potential, $U = U^{att} + U^{rep}$, then, specifically for the 12-6 Lennard-Jones potential with strong attraction, $U^{att}/U^{rep} \sim (\varepsilon/E)^{1/6}$, where E is the impact collision energy. For instance, for Xe, the ratio $(\varepsilon/E)^{1/6}$ equals 1.15 at the temperature $T = 77.4$ K and 0.75 at $T = 1000$ K. Therefore, attractive intermolecular forces can make a significant contribution to the gas molecule–molecule interaction of heavy gas

The dimensionless mass flow rate Q^* through a two-dimensional slit into a vacuum as a function of the rarefaction parameter δ for the VSS model, for He (solid) and Xe (open) gases. The inset shows the dimensionless difference Δ in the Q^* values for these gases

molecules even in the case of rather high temperatures.

A generalized soft sphere (GSS) model was suggested in [15], where the total cross section σ_t was assumed to be the same as in the GHS model, but the deflection angle χ was as in the VSS model. A simplifying assumption used in [15] − a fitting formula for the collision integrals in the case of the Lennard-Jones potential − allows explicitly determining the parameters involved in the GSS model.

The variable sphere (VS) model developed in [16] provides the viscosity cross section σ_{μ} and the diffusion cross sections σ_D consistent with those of any realistic intermolecular potential and obeys a simple scattering law that is even simpler than the isotropic law. However, even the author himself noted that the model should be tested with many rarefied gas flow problems.

For this study, we used the VHS and VSS models defined for the IPL potential to analyze the influence of the gas molecule–molecule interaction on the mass flow rate through a slit. The computations were performed for He and Xe gases; the ω and α parameters for those gases were taken from tables A1 and A3 in [8]. The obtained results were compared with similar computations in [7] for the HS model, which does not require knowing the kind of the gas. To study how the gas molecule attraction affects the mass flow rate, the GHS model for heavy gas molecules was used for Xe molecules at various gas temperatures. The GHS model parameters are taken from [14] in the case of strong attraction. The gas–surface scattering was modeled as being completely diffuse in all cases.

Table 1. The dimensionless mass flow rate Q^* through a two-dimensional slit into a vacuum for the HS, VHS, VSS, and GHS models for different gas rarefaction parameters δ

δ	HS	VHS He/Xe	VSS He/Xe	GHS for Xe $T = 77.4/300/1000$ K
0.1	1.025	1.022/1.020	1.024/1.021	1.107/1.085/1.070
1	1.147	1.136/1.122	1.139/1.125	1.398/1.352/1.315
10	1.473	1.456/1.435	1.460/1.437	1.559/1.554/1.551
100	1.561	1.559/1.558	1.559/1.558	1.543/1.546/1.548

First, to test the code procedures that simulate molecule collisions, we performed simulation of a spatially homogeneous collision relaxation. For each procedure, we obtained the expected relaxation to the Maxwellian distribution. After that, these procedures were incorporated into the main code that simulates the gas flow through a slit into a vacuum.

The Figure shows the results of computation of the dimensionless mass flow rate Q^* through the two-dimensional slit into a vacuum as a function of the rarefaction parameter δ when the VSS model is used, for He and Xe gases. According to the figure, the mass flow rate in the transition flow regime is noticeably higher for helium than for xenon. Because the gas–surface scattering was modeled as being completely diffuse for both gases, the difference in the mass flow rate can only be explained by different molecule–molecule interactions. Higher values of the dimensionless mass flow rate for light inert gases such as He and Ne in comparison with the mass flow rate for heavier inert gases Ar, Kr, and Xe in the transition regime were also confirmed experimentally [4].

The inset to the figure shows the dimensionless difference in the values of the mass flow rate for those gases defined as

$$\Delta = (Q^*_{\mathrm{He}} - Q^*_{\mathrm{Xe}})/Q^*_{\mathrm{Xe}}$$

as a function of the gas rarefaction parameter δ. According to the inset, the maximum effect of the gas molecule–molecule interaction on the mass flow rate is observed when the δ value is about 4; the effect then reaches 2 %. It is interesting to note that the well-known Knudsen minimum of the dimensionless mass flow rate through a long rectangular channel has been observed both in theory [17] and in experiment [4] when the gas rarefaction value δ is close to 1.

It is also shown in the inset that the dimensionless mass flow rates are the same in the free molecular (small δ) and hydrodynamic (large δ) regimes. That is,

when one of these gas flow regimes is reached, the gas molecule–molecule interaction stops influencing the gas flow through a slit into a vacuum. In the case of the free molecular regime, this result is obvious.

Table 1 presents the results of computations of the dimensionless mass flow rate Q^* through a two-dimensional slit into a vacuum for the HS, VHS, VSS, and GHS models for various rarefaction parameters δ. As the table demonstrates, in the case of models based on the IPL potential (HS, VHS, and VSS), the results for the HS model are maximal because it represents the model of the gas molecule–molecule interaction that is "too hard". Minimal results should be observed in the case of the "too soft" model of Maxwell molecules.

By comparing the results for the VHS and VSS models (which differ by the scattering law in molecular collisions) for a given gas, for instance, VHS-He and VSS-He, we can conclude that variations in the scattering law do not entail any changes in the mass flow rate exceeding the computation error. But comparing the simulation results for He and Xe, for instance, VSS-He and VSS-Xe, which differ in the total cross section σ_t, convinces us that a variation in σ_t does lead to a measurable change in the mass flow rate.

Table 1 contains the values of the mass flow rate Q^* for the GHS model for Xe at the temperatures 77.4, 300, and 1000 K. As the table shows, the presence of the attractive component in the model of the intermolecular potential leads to a noticeable increase in the Q^* value in the transition flow regime even in the case of a rather high temperature 1000 K. As the temperature decreases, the effect of the attractive component increases, and hence the Q^* value also increases, as is also demonstrated by this table.

An increase in the mass flow rate values in the transition flow regime accompanied by a decreasing temperature is in agreement with the reliable experiment in [18], where the dependence of the mass flow rate through a rather long capillary systems in the temper-

ature range 77.4–293 K was studied. However, the observed systematic increase in the mass flow rate as the temperature decreases was attributed by the authors only to a change in the gas–surface scattering.

3. INFLUENCE OF THE GAS–SURFACE SCATTERING

The best known model of the gas–surface scattering is the Maxwell model, the so-called specular–diffuse scheme. According to this scheme, the fraction α_M of molecules falling on the surface reflects diffusely, i.e., comes into equilibrium with the surface and is then emitted in accordance with the cosine law, and the remaining fraction $1 - \alpha_M$ is reflected speculary.

Obviously, the specular–diffuse scheme is based on the relatively simple physical approach, and hence this model has a number of inaccuracies in describing the gas scattering by a surface. In particular, a single parameter α_M characterizes both the energy and momentum exchange of gas molecules with the surface. However, it is known that the processes of the momentum and energy exchange differ and that momentum is acquired or lost much faster than energy. This is one of the shortcomings of the Maxwell model.

More sophisticated physical models of gas–surface scattering were developed in [19] and [20]. In particular, in the case of isotropic scattering tangential to the surface, the accommodation coefficients of the kinetic energy corresponding to the normal molecular velocity and of the tangential momentum are two parameters in the Cercignani–Lampis (CL) model [20].

In addition, the Epstein [19] and CL [20] models allow deriving the dependence of the gas–surface scattering process on the ratio between the velocity of gas molecules incident on the surface and the surface temperature. This is the fundamental distinction from the Maxwell model. Indeed, because α_M is constant, the outcome of the gas–surface scattering (diffuse or specular) is independent of this ratio. This fact is important in studying nonisothermal gas flows. Recently, we were able to show that computations based on the CL and Epstein models provide a more satisfactory description of the gas–surface scattering for nonisothermal gas flows than the commonly used Maxwell model [21].

Nonetheless, the Maxwell model is successfully used in many practical calculations, for instance, in isothermal gas flow computations. We used this model to interpret our experiment on studying the dependence of the isothermal gas flow through a capillary on the chemical composition of the capillary surface [5]. In

Table 2. The dimensionless mass flow rate Q^* through a two-dimensional slit into a vacuum in the case of the diffuse ($\alpha_M = 1$) and specular–diffuse ($\alpha_M = 0.71$) scattering for different gas rarefaction parameters δ

δ	Diffuse	Specular–diffuse
0.1	1.025	1.024
1	1.147	1.143
10	1.473	1.469
100	1.561	1.557

particular, in studying the flow of He through a capillary with an atomically clean surface of Ag, we obtained the smallest value of α_M in the open literature, equal to 0.71. At the same time, on so-called contaminated surfaces (surfaces that did not undergo a special treatment), the gas–surface scattering is close to the diffuse law. Hence, we can assume that in the case of the gas flow through a capillary, the parameter α_M ranges from 1 (diffuse scattering) to 0.71 (specular–diffuse one). It then follows that in order to estimate the influence of the gas–surface scattering on the mass flow rate through a capillary, it suffices to consider these two extreme cases.

The results of computation of the mass flow rate Q^* through a two-dimensional slit into a vacuum are presented in Table 2 in the cases of the diffuse and specular–diffuse scattering of He on an atomically clean surface of Ag. The gas molecule–molecule interactions were simulated using the HS model in both cases.

As Table 2 demonstrates, for any rarefaction δ, the result of the mass flow rate computation obtained using the Maxwell specular–diffuse scheme competes within the computation error with the result of using the diffuse law. Similar results were obtained when using the CL and Epstein models with significantly nondiffuse scattering parameters.

The central conclusion from the aforesaid is that the gas–surface scattering does not noticeably influence the mass flow rate through a slit into a vacuum. This fact is related to the absence of the lengthy border of the gas–surface scattering along the gas moving path in the case of a slit. Considerable influence of the gas–surface scattering should be observable in capillaries with a long enough border, i.e., in sufficiently long capillaries.

4. CONCLUSION

Using the direct simulation Monte Carlo method, we have computed the gas mass flow rate Q^* through a two-dimensional slit into a vacuum in the range of the rarefaction parameter δ from 0.02 to 200 for the different models of the gas molecule–molecule interaction and gas–surface scattering. The maximum gas molecule–molecule interaction effect on the mass flow rate is observed in the transition flow regime when the δ value is about 4. It is revealed that variations in the scattering law in molecular collisions do not lead to considerable changes in the mass flow rate, but variations in the total cross section affect the value of Q^* significantly. Incorporating attraction into the gas molecule–molecule interaction model leads to a significant increase in the mass flow rate in the transition flow regime. The gas–surface scattering does not noticeably affect the value of the mass flow rate. The obtained results agree with the corresponding experimental ones [4, 18].

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