Inertia effects in the compressed layer in front of a flat plate

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Abstract. The deceleration of heavy admixture in the compressed layer in front of the flat plate is studied numerically using the Direct simulation Monte Carlo (DSMC) method, on the example of helium+xenon and helium+argon gas mixtures. The energy spectrums of heavy molecules hitting the plate are analyzed. Two collision models with different scattering law but same diffusive and viscous effective cross-sections were used to test the sensitivity of results. The simple estimation method for mean energy of heavy component is derived, and its results are compared to those obtained in the simulation.

Keywords: flat plate, energy spectrum, shock layer, binary mixture, supersonic gas jet deposition.
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INTRODUCTION

The supersonic gas mixture jet deposition method, employing the acceleration of heavy molecules in a light carrier gas up to the speed of few km/s, which then make a way through the compressed layer in a rarefied condition and deposit on a surface, was proposed many years ago [1, 2]. This method is one of approaches to the creation of nanostructured thin films, particularly, films of organic semiconductor [3].

Let $\mathcal{M}$ designate a mass ratio of heavy and carrier molecules. During the phase of supersonic expansion into the vacuum, the heat energy is effectively converted to the energy of kinetic movement. For example, the expansion of pure helium with the stagnation temperature T_0 = 300K, yields the kinetic energy of up to 0.065 eV. This is valid for a gas mixture as well on condition that the concentration of heavy molecules is small enough not to distort the mass density. If the acceleration of the heavy admixture is effective, it will have the same velocity as the carrier gas, and the kinetic energy of heavy molecules will be $\mathcal{M}$ times higher compared to the energy of carrier molecules. For example, the xenon ($\mathcal{M}$ = 32.78) admixture may be accelerated up to 2.13 eV of kinetic energy. With such a great kinetic energy of heavy organic precursor molecules hitting the substrate, the properties of an obtained film may be unique from those deposited in near-equilibrium conditions [4].

However, a shock layer, consisting of a shock wave and compressed layer, is formed before the deposition target. The gas is almost stagnated in compressed layer, with its kinetic energy converted back to the heat energy, as long as the gas is dense enough. The rarefaction may be described by the Knudsen number Kn, that is a ratio of local mean free path and the size of the target. With $\mathrm{Kn} \gg 1$, no shock layer is formed. With $\mathrm{Kn} \ll 1$, the flow is close to continuum with the clear shock layer formed and the most of the kinetic energy lost. With $\mathrm{Kn} \sim 1$, only a part of the kinetic energy is lost. In the case of big mass ratio ($\mathcal{M} \gg 1$), a lot of collisions of heavy molecules with light carrier gas are required to drain their kinetic energy, so a considerable energy may remain even with quite low Knudsen number.

In theory, the kinetic energy of deposited particles may be controlled by changing the Knudsen number of the compressed layer. However, there is still no detailed theoretical description of inertia effects in a compressed layer that would allow precise predictions. Most investigators have to search experimentally conditions they need, basing on general provisions, or simulate the flow numerically.

In this work the movement of heavy particles through the compressed layer in front of a flat plate is studied on example of helium + xenon and helium + argon model mixtures. The study is performed numerically by using Direct Simulation Monte Carlo (DSMC) method based on Majorant Frequency Scheme [5]. Varied parameters include Knudsen number and xenon concentration. The energy spectrum of xenon particles colliding the plate is analyzed, together with total flow of xenon onto the plate.

The study is performed employing two different collision models (VSS [6] and VS [7]) that give similar transfer coefficients. Even the Navier-Stokes estimation of Boltzmann equation predicts the dependence of the velocity distribution function on collision model. The usage of two different scattering models gives the understanding of result universality.

This work offers the simple estimation method as well, which gives good prediction of mean kinetic energy of a heavy admixture, as long as its concentration is small enough not to disturb the flow. The estimation method is testified by both the helium+xenon and helium+argon simulations.

PROBLEM SETTING AND SIMULATION METHOD

Two-dimensional setting of the problem is considered. The flat plate, having zero thickness, the wideness H and infinite length, is placed transversal to the oncoming equilibrium plane-parallel supersonic gas mixture flow. The flow parameters are: the temperature T_∞ = 29.27 K, and the velocity V_∞ = 672.5 m/s. For pure helium, these correspond to the Mach number M = 5 and the stagnation temperature T_0 = 273.15 K. The plate temperature was fixed at T_0. Varied parameters are: the Knudsen number $\mathrm{Kn}_\infty = \frac{1}{\sqrt{2} n_\infty \sigma_{11} H}$ and the molar fraction of heavy admixture C_{Xe}. Here n_∞ is a density of undisturbed flow, σ_{11} is the He-He collision cross-section at temperature T_0, H is the width of the plate.

The rectangular simulation area was used. The problem has a plane of symmetry that allows computing only a half of the problem by placing a mirror plane. The diffusive reflection on the plate was set for a carrier gas. Heavy molecules either reflect as a carrier gas in one case or are fully adsorbed in other. A combination of full adsorption and the source of undisturbed flow molecules was set on other boundaries. For collisions, two molecule scattering models were used: Variable Soft Spheres (VSS) [6] and Variable Spheres (VS) [7]. In VSS model, deflection cosine is random, sampled from power-distribution with exponent α. In VS, the deflection angle χ cosine is fixed. Both models allow balancing diffusion and viscosity effects. As VS model gives smaller total cross-section, the VSS model was used to calculate Knudsen numbers. Model parameters at T_0 are shown in Table 1. The ratio of xenon and helium molecular masses is 32.78, the argon/helium ratio is 10.

TABLE 1. Collision model parameters.

Pair	ω	VSS		VS	
		d_{ref}, Å	α	d_{ref}, Å	χ
He-He	0.660	2.30	1.32	1.65	101.8º
He-Xe	0.755	3.97	2.20	2.61	87.3º
Xe-Xe	0.850	5.65	1.33	4.04	101.6º
He-Ar	0.725	3.20	1.64	2.21	95.7º
Ar-Ar	0.810	4.11	1.40	2.91	100.2º

The result of a simulation, along with the flow macroparameters, is the kinetic energy spectrum of heavy molecules hitting the plate.

ESTIMATING THE MEAN ENERGY

When the oncoming supersonic flow stagnates in front of the plate, the compressed layer is formed with thin shock wave as a boundary between the compressed layer and the undisturbed flow. The gas looses the velocity in the shock wave and is almost stagnated in the compressed layer. In the practical case, when the molecular mass of heavy admixture is much higher than the molecular mass of carrier gas, heavy molecules need a lot of collisions with stagnated carrier molecules to loose the kinetic energy. The thicker is the shock layer, the more energy is lost. Let consider the thickness of shock layer to be N mean free paths, the mean temperature in the shock layer is T_S, and the mean energy of accelerated molecules is E_0. The following considerations are to be used: a) for unambiguity, the shock layer starts when the 50% of the carrier gas velocity is lost; b) being fully stagnated, heavy molecules would reach the plate with mean kinetic energy $2kT_S$; c) collisions are described with the hard sphere model, so that $\frac{2\mathcal{M}}{(\mathcal{M}+1)^2}$ is the fraction of the heavy molecule energy lost in a single collision, as well as the fraction of carrier gas molecule energy transferred to the heavy one; d) the distance between collisions of heavy molecule with carrier gas molecules is $\lambda_{12} \approx \frac{v_2}{n_0 \sigma_{12} c_{r12}} \approx \lambda_0 \sqrt{2} \frac{\sigma_{11} v_2}{\sigma_{12} c_{r12}}$; e) the relative velocity is almost equal to the heavy molecules velocity: $\frac{v_2}{c_{r12}} \sim 1$, so

$\lambda_{12} \approx \lambda_0\sqrt{2}\frac{\sigma_{11}}{\sigma_{12}}$. Here v_2 is the mean velocity of heavy component, n_0 is the carrier gas density, σ_{12} is the carrier-admixture collision cross-section, σ_{11} is the carrier-carrier collision cross-section, c_{r12} is the mean relative velocity, λ_0 is the mean free path of carrier molecules. The number of collisions of heavy molecule with carrier molecules is then $\frac{N\lambda_0}{\lambda_{12}}$. These yield the following expression:

$$E \approx 2kT_S + (E_0 - 2kT_S) \cdot \exp\left(-\frac{\mathcal{M}}{(1+\mathcal{M})^2}\sqrt{2}N\frac{\sigma_{12}}{\sigma_{11}}\right) \quad (1)$$

The only parameter unknown here is N. Obviously, it should depend on the Knudsen number and Mach number.

The problem of flow over a flat plate, infinite in one direction, placed into the transversal supersonic flow, is studied in [8] for the $\mathrm{M} = 5$, and the N is determined as a function of Knudsen number, by the DSMC simulation of the hard sphere gas. Results for $\mathrm{M} = 5$ are still valid for $\mathrm{M} > 5$, as the velocity of the gas is close to its limit and further increase of the Mach number does not change the thickness of shock layer considerably.

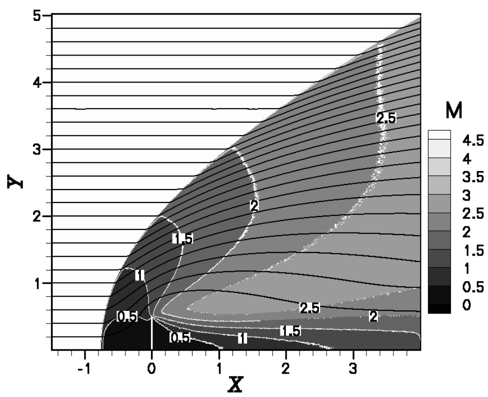

FIGURE 1. The typical flow around a flat plate.

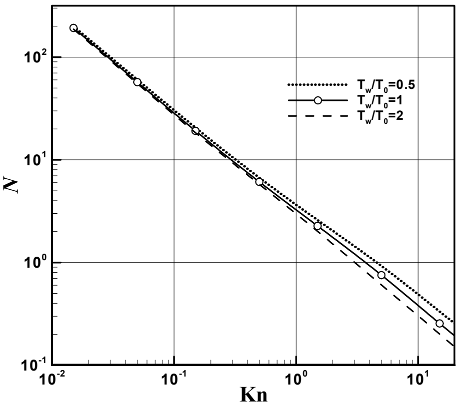

FIGURE 2. The dependence of the shock layer thickness in mean free paths on the Knudsen number.

Figure 1 shows the typical flow around the flat plate, $\mathrm{M} = 5$, $\mathrm{Kn}_\infty = 0.015$ and plate temperatue T_W equal to the stagnation temperature T_0. Figure 2 shows the thickness of the shock layer in mean free paths for three temperatures of the plate. One can see the thickness weakly depends on plate temperature at $\mathrm{Kn}_\infty < 1$.

In the case of the model more complex than hard spheres, the hard sphere cross-section σ_{11} should be determined using the carrier gas viscosity in the compressed layer, while σ_{12} should be determined using the admixture-carrier diffusion coefficient, in the compressed layer.

THE CASE OF FULL ADSORPTION OF HEAVY COMPONENT ON THE PLATE

The Figure 3 integrates energy spectrums of xenon obtained by DSMC simulations. The xenon concentration was considered low (0.1%), and no xenon was reflected from the plate. Comparing them, one can see, that, with the original (free-molecular) spectrum being almost symmetrical, it further widens to the left ($\mathrm{Kn}_\infty = 10$), then becomes more symmetrical again, and then widens to the right ($\mathrm{Kn}_\infty = 0.1$), approaching the equilibrium exponent-tailed one. As expected, lower Knudsen numbers yield lower energies. With $\mathrm{Kn}_\infty = 0.3$, 50% of energy is still remaining. One can see, the energy spectrum is the same for both VSS and VS model, the reason is the high mass ratio, so that a lot of helium-xenon collisions are required for considerable change in energy, and more collisions better smooth out scattering details.

Figure 4 shows the results for helium-argon mixture, for VSS model only. The tendency is similar, but spectrums are more asymmetrical and the energy is lost faster.

On Figures 5 and 6, the mean energy, estimated by formula (1), taking the N from Figure 2 and using recalculated Knudsen number, is compared with actual computed mean and most probable energy values, for xenon and argon correspondingly. Additionally, the penetrability G of heavy molecules through the compressed layer is shown, $G = 1$ corresponds to the heavy molecule flow of $n_\infty V_\infty$. One can see, the penetrability of the shock layer is high in the whole range of Knudsen numbers where the energy can be effectively controlled.

Figure 7 shows the energy spectrum for $\mathrm{Kn}_\infty = 3$ and different xenon concentrations (0.1%, 4.8% and 20%), with the fixed density of helium component. One can see no considerable difference. This testifies the independence of energy spectrum of heavy admixture concentration, as soon as all heavy molecules are adsorbed on the plate. Note that even 20% concentration of xenon is hard to achieve experimentally reaching the same gas mixture velocity as in pure helium, so a consideration of higher xenon concentrations is not required.

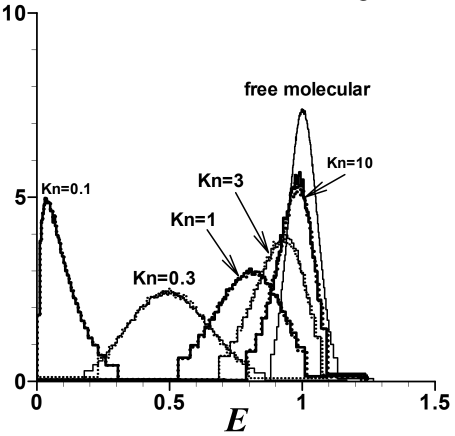

FIGURE 3. Energy spectrums of xenon (concentration is 0.1%) on different Knudsen numbers. Dotted lines are for VS model, solid are for VSS.

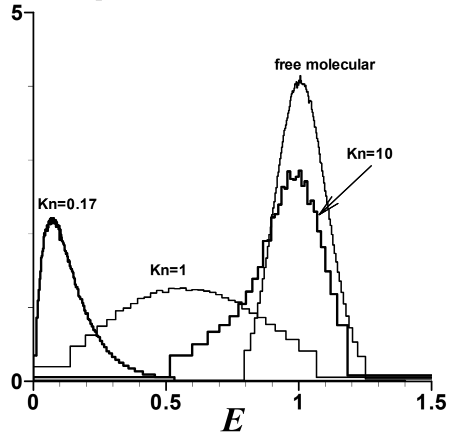

FIGURE 4. Energy spectrums of argon on different Knudsen numbers.

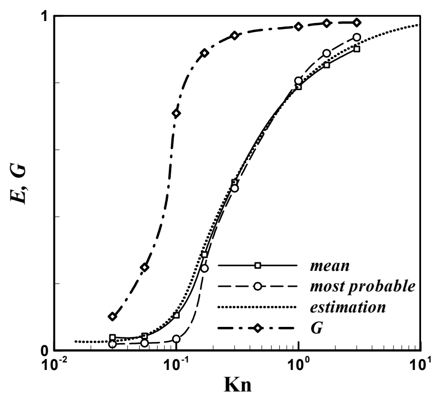

FIGURE 5. The comparision of actual and predicted xenon energy, and the penetrability of the shock layer for xenon.

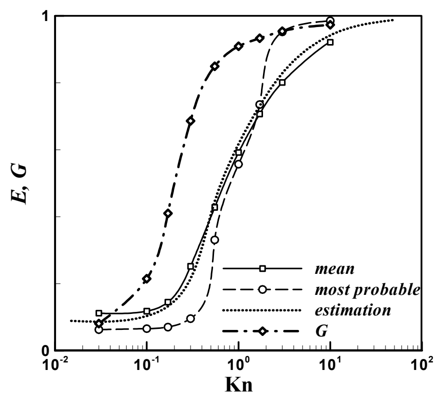

FIGURE 6. The comparision of actual and predicted argon energy, and the penetrability of the shock layer for argon.

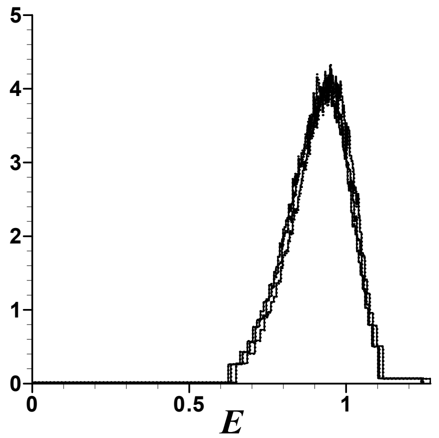

FIGURE 7. Energy spectrums of xenon, for different xenon concentrations, $Kn_\infty = 3$.

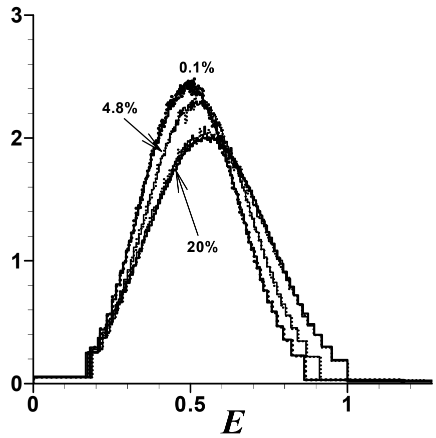

FIGURE 8. Energy spectrums of xenon, for different xenon concentrations, $Kn_\infty = 0.3$.

Figure 8 shows the xenon energy spectrum on different concentrations for $Kn_\infty = 0.3$. This time the difference is noticeable, because with the higher xenon concentration, its lost energy heats the compressed layer, rising the T_S. In the continuum limit, it corresponds to the rise of stagnation temperature. The difference is not because of Xe-Xe collisions, the simulation of 20% case with blocked Xe-Xe collisions gives the same result as with Xe-Xe collisions active.

Figure 9 shows density and velocity profiles in the plane of symmetry, for 0.1% and 20% cases. One can see, the shock layer profile is notably different in 20% case, as the xenon presses the shock layer towards the plate making it thinner geometrically. Interestingly, it almost does not change the velocity of the xenon in front of the plate.

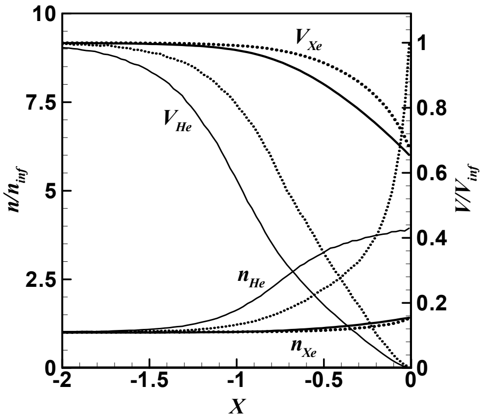

FIGURE 9. Density and velocity profiles in the shock layer for $Kn_\infty = 0.3$, xenon concentrations of 0.1% (solid lines) and 20% (dotted lines).

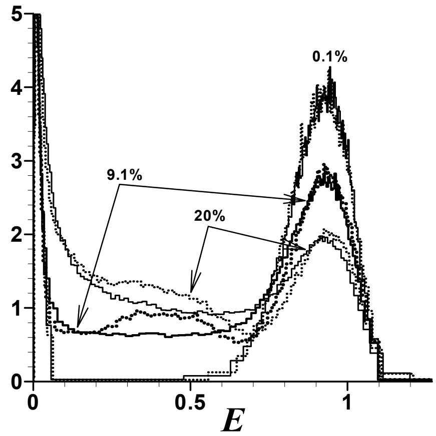

FIGURE 10. Xenon energy spectrums on 100% diffuse reflection of xenon from the plate. $Kn_\infty = 3$. Solid lines are for VSS model, dotted lines are for VS model.

THE EFFECT OF REFLECTED HEAVY MOLECULES

With no slow xenon molecules scattered from the plate, Xe-Xe collisions do not affect the energy much. However, with a presence of considerable amount of slow heavy molecules scattered from the plate, this cannot be true. The energy is lost in heavy-heavy collisions much faster, with 50% of energy on average transferred in a single collision.

Figure 10 shows energy spectrums of xenon for the case $Kn_\infty = 3$ and diffuse reflection of xenon from the plate. For low xenon concentration (0.1%), the spectrum consists of the same high energy constituent as in full adsorption case, with an additional low-energy constituent originated from the slow xenon molecules scattered from the plate. For higher xenon concentrations, the high energy constituent keeps almost the same profile but becomes weaker, whereas a new middle-energy constituent appears. One can see that VSS and VS spectrums of middle-energy constituent are noticeably different. VS model gives the visible knap at 40% of energy, while VSS model gives a smoother spectrum. The collision model affects the energy spectrum because of too few Xe-Xe collisions, with not enough He-Xe collisions to smooth this difference. At least 3 collisions are usually considered for translational relaxation, so at least 3 Xe-Xe collisions should be expected to smooth the energy spectrum enough for it to be the same for different collision model.

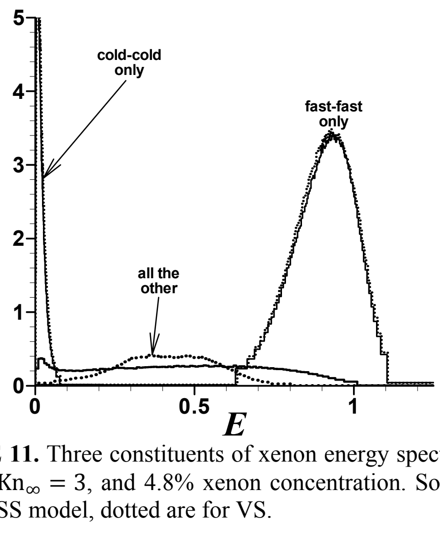

FIGURE 11. Three constituents of xenon energy spectrum for the case $Kn_\infty = 3$, and 4.8% xenon concentration. Solid lines are for VSS model, dotted are for VS.

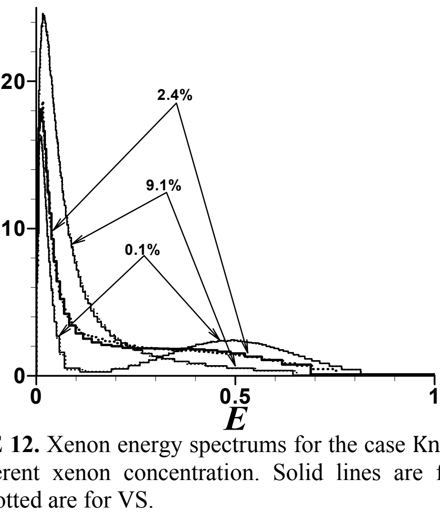

FIGURE 12. Xenon energy spectrums for the case $Kn_\infty = 0.3$ and different xenon concentration. Solid lines are for VSS model, dotted are for VS.

Figure 11 shows separately three constituents of the xenon energy spectrum, for case of 4.8% xenon concentration. To split the spectrum, the incident xenon molecules (fast) and the reflected xenon molecules (slow) were tagged. The collision of differently tagged xenon molecules or a tagged xenon molecule with an untagged one makes them both untagged. One can make certain that only the middle-energy (untagged) molecules have different energy spectrum for VSS and VS model.

It is important to note, that collision of slow and fast heavy molecules are rather high-energetic, meaning that it can change the properties of the deposited precursor molecules.

Figure 12 shows xenon energy spectrums for the case $Kn_{\infty} = 0.3$. Although the position of high-energy knap is not changed, its amplitude decays much faster, because the smaller Knudsen number makes both the Xe-He and Xe-Xe collisions more frequent with the same xenon percentage. On a lower Knudsen number, the elevated xenon concentration makes the energy spectrum more complex and less predictable. This means, for deposition purpose, the concentration of heavy molecules in shock layer at lower Knudsen numbers should be carefully controlled. The amplitude of low-energy peak is considerably higher than in the case of higher Knudsen numbers, because it is harder for xenon molecules to leave the shock layer. One can see that, again, difference in VSS and VS spectrums is less considerable; the higher number of Xe-Xe collisions together with elevated amount of Xe-He collisions smooth the difference between two collision models.

CONCLUSION

The energy spectrum of heavy molecules accelerated by the carrier gas and penetrated the shock layer is numerically studied on the example of helium+xenon model mixture. Additionally, the simple and generalized method for estimating the mean value of energy is proposed for the case of negligible effect of heavy molecules reflected from the plate; the method is successfully testified numerically on helium+xenon and helium+argon model mixtures. As long as the amount of heavy molecules reflected from the plate is negligible, the concentration of heavy admixture does not affect the energy spectrum much and the penetrability of the shock layer is high in the whole practical range of Knudsen numbers.

For higher Knudsen numbers, the reasonably low concentration of heavy molecules reflected from the plate into the shock layer, just decrease the amount of fast heavy molecules approaching the plate. With less than 2-3 heavy-heavy collisions along the path, with the reflected molecules involved, the energy spectrum depends on the details of heavy-heavy collision model. Besides, the high energy of those collisions may change the properties of the precursor molecules. For lower Knudsen numbers, the dependence of energy spectrum on the concentration of heavy molecules reflected from the plate may be complex and less predictable. In this case the concentration of heavy admixture should be controlled somehow in the deposition process.

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REFERENCES

1. A. K. Rebrov. *Vacuum technologies and equipment: VI Intern. Conf. Proc.*, 2003, pp. 113-122 (in Russian).
2. R. V. Maltsev, A. K. Rebrov. *Proc. of 24th Intern. Symp. on Rarefied Gas Dynamics*, AIP Conference Proceedings, Vol. 762, 2005, pp. 1229-1234.
3. F. De Angelis, T. Toccoli, A. Pallaoro, N. Coppede, L. Mariucci, G. Fortunato, S. Iannotta. *Syntetic metals* **146**, 291 (2004)
4. S. Iannotta, T. Toccoli, A. Boschetti, P. Scardi. *Synthetic Materials* **122**, pp. 221-223 (2001).
5. M. S. Ivanov and S. V. Rogazinski, *Soviet J. Numer. Anal. Math. Modelling* **3** (6), 453 (1988).
6. G. A. Bird, *Molecular Gas Dynamics and the Direct Simulation of Gas Flows*, Clarendon press, Oxford, 1994.
7. H. Matsumoto. *Proc. of 23rd Int. Symp. on Rarefied Gas Dynamics*, AIP Conference proceedings, Vol. 663. Melville, New York, 2003, pp. 358-365.
8. R. V. Maltsev and A. K. Rebrov. *Fluid Dynamics* **40** (1), MAIK Nauka/Interperiodica, pp. 140-147 (2005).