Optimizing Nozzle Geometry for Controlling Properties of Molecular Beam with Heavy Organic Molecules

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Abstract. The flow in conical supersonic micronozzles and behind them by deposition of pentacene accelerated by helium was studied with the use of direct simulation Monte-Carlo method. The Knudsen number in critical cross-section of nozzles is 0.0043. A directedness of resulting accelerated pentacene flow was studied as a function of cone angle and geometrical Mach number of the nozzle. The intensity of heavy gas flow in the hypersonic region can be elevated by one order using supersonic nozzles.

Keywords: micronozzles, nozzle optimization, molecular beam, pentacene, supersonic molecular beam epitaxy.
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INTRODUCTION

In the history of rarefied gas dynamics the problem of gas flows in supersonic nozzles and in free jets has been solved as fundamental one as well as for practical application. Most important trends were concerned an elaboration of nozzles to get supersonic flat parallel flows, investigation of nonequilibrium (slow relaxing) flows, elaboration of gas dynamic sources for molecular beam systems, elaboration of gas sources for coating vacuum technologies. The aim of this work is the study of flows of gases with disparate masses in nozzles and jets behind them. The most specific task is the search of possibilities of gas dynamic compressing of heavy molecule stream lines by a flow formation in nozzles and behind them. In a row of cases the optimization of molecular beam systems used for a film deposition is necessary. One example is the deposition of semiconducting films from organic molecules such as pentacene [1].

The idea of using supersonic nozzles instead of sonic (orifice) ones to get intensive molecular beams belongs to Kantrovits [2]. The efficiency of this method occurred to be very high and there was no need to perform thorough exploring and systematic research. Later the more detailed knowledge were claimed by an elaboration of microthruster nozzles [3]. The well-grounded computational modeling of flows in microthruster nozzles was performed in [4, 5]. The comparison of computations on the base of Navier-Stokes equations and statistical modeling with experiments has shown the efficiency of DSMC.

The problem formulated in this work is distinguished by analysis of the gas mixture flow in a field of two-dimensional parameter gradients.

The main content of this work is the study of an influence of the conical nozzle geometry (opening angle and supersonic part length) on the pentacene flow intensity far away from the nozzle exit in a hypersonic part of the flow. For the evaluation of a principal possibility and tentative level of the pentacene flow intensity magnification the calculations were performed for one of typical regimes of experiments in the Institute of Photonics and Nanotechnology [1].

PROBLEM SETTING AND SIMULATION METHOD

The DSMC method [6] used in this work was based on prerequisites as follows: 1) particles displacement and collision phases can be separated; 2) the collision partners are chosen stochastically from the same collisional cell, regardless of their mutual disposition; 3) the aiming partners have been taken as occasional ones. The choice of collisional pairs was followed by majorant frequency scheme [7], when the number of collisions in a cell is subordinated to Poisson distribution with the average frequency being proportional to the number of all possible collisional pairs in a cell. The computational space is divided by quadrangle cells with sizes less than a molecular mean free path. To increase the number of particles for a calculation close to the axis the radial weight factors were applied; with these, particles moving off the axis are annihilated, and those moving to the axis are multiplied by collisions. The molecules are collided as Variable Soft Spheres. This model has 3 parameters: the average cross-section at some temperature, the exponent in the viscosity-temperature dependence, and the exponent for deflection angle cosine distribution to balance viscosity and diffusion. For rotational-translational and vibrational-translational energy exchange the Borgnakke-Larsen model was used. According to this model the part of collisions is assumed to be elastic, and other part is non-elastic with Boltzmann distribution of the total energy over translation and internal degrees of freedom. All model parameters are summarized in Table 1: d is collisional diameter at the stagnation temperature, ω is the exponent in the viscosity-temperature dependence, α is the scattering law exponent, ξ_{rot} and ξ_{vib} are quantities of rotational and vibrational degrees of freedom correspondingly. $Z_{rot(DSMC)}$ and $Z_{vib(DSMC)}$ are the average quantities of collisions per inelastic ones. On the boundary of computational domain the full absorption was prescribed. The diffuse scattering with full accommodation was prescribed for collisions of molecules with the nozzle wall. The molecular velocity distribution of the gas entering into the stagnation chamber was considered as maxwellian one.

TABLE 1. Simulation model parameters.

Pair	d, Å	ω	α	ξ_{rot}	$Z_{rot(DSMC)}$	ξ_{vib}	$Z_{vib(DSMC)}$
He-He	2.16	0.652	1.318	–	–	–	–
Pentacene-He	7.87	0.5	1	3+0	4.6	10+0	1.75

Following data were used in calculations: the nozzle critical cross-section diameter $d_* = 65$ μm, the total stagnation pressure (helium+pentacene) $P_0 = 240$ kPa, the stagnation temperature $t_0 = 400$°C, the mole pentacene fraction was taken 0.1%; it was accepted that the gas expands into vacuum. This parameters correspond to Knudsen number in critical cross-section equal to 0.0043. The nozzle wall temperature $t_W = 125$°C. The pentacene molecular weight is 278 a. u. In real experiments[1] the gas mixture of pentacene vapor and helium is forming by sublimation of pentacene immediately before nozzle. It gives rise to uncertainty in concentration distribution in a mixture. But it is not reason for an obstruction in optimization calculations at a low pentacene concentration.

The solution of the problem on nozzle optimizations in the formulated set does not need in specification of nozzle skimmer disposition. Calculations were performed up to a distance, where the hypersonic flow with a practically radial expansion of both components was formed. It happens at Mach numbers M = 7–9 and higher.

The directedness of pentacene flow Q near the axis is determined as ratio of mass flux into the solid angle unit in the vicinity of axis at the right simulation boundary to the value for pure radial expansion into half-space from the hemispherical sonic surface. The injection rate through the hemispherical sources was prescribed equal to that through the nozzle. The value of Q was determined for conical nozzles with an opening angle 20°, 30°, 40° and geometrical Mach number M_G for monatomic gas at the nozzle exit equal 1, 4, 6, 8.

The nozzle geometrical Mach number M_G for a gas with given properties (a monatomic gas in our case) is conventionally uniquely connected with ratio of nozzle exit and critical cross-section surfaces. In gas dynamic terms, geometrical Mach number is the Mach number to be expected at a given flat, transversal to the axis, cross-section of a nozzle, supposing the flow is isentropic everywhere in the nozzle, and is plane-parallel in that given cross-section. One can use the Table 2 to compare the nozzles studied. Here, d_{ex} is the diameter of nozzle exit, L is the nozzle length.

[1] Deposition experiments were performed in Institute of Photonics and Nanotechnology, but the direct comparison of results is not yet available.

TABLE 2. Nozzles parameters for different geometrical Mach numbers.

$\mathbf{M}_G$	1	2	3	4	5	6	8	10	12
$\boldsymbol{d_{ex}/d_*}$	1	1.237	1.732	2.375	3.130	3.980	5.922	8.142	10.609
20°, $\boldsymbol{L/d_*}$	0	0.673	2.076	3.899	6.041	8.451	13.96	20.25	27.25
30°, $\boldsymbol{L/d_*}$	0	0.443	1.366	2.566	3.976	5.562	9.185	13.33	17.93
40°, $\boldsymbol{L/d_*}$	0	0.326	1.006	1.889	2.927	4.094	6.762	9.812	13.20

RESULTS

Now consider some results for the nozzle with the convergent part angle 157.4° and supersonic part opening 40°. Figure 1 shows: a) carrier gas streamlines with $\mathrm{M}_G = 6$; b) pentacene streamlines. The transversal separation of gases is visible behind a nozzle as different turn of streamlines.

a) b)

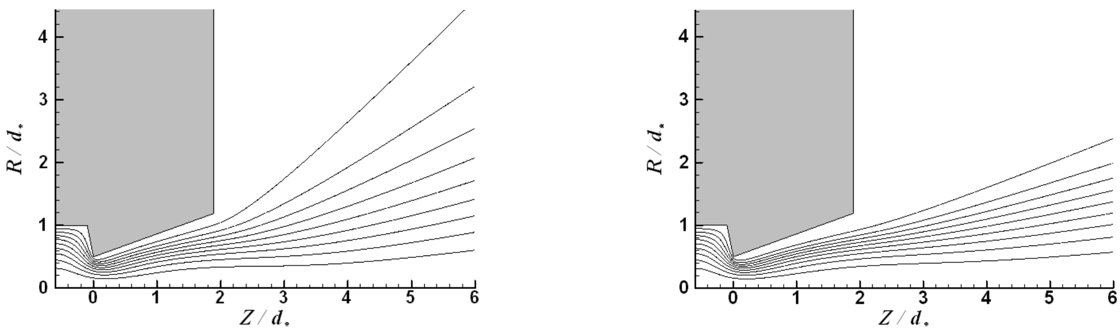

FIGURE 1. Helium (a) and pentacene (b) streamlines. The nozzle with angle 40°, $\mathrm{M}_G = 4$.

The change of the angle between a selected stream line and the axis indirectly characterize the focusing or defocusing of the flow, i.e. the expected Q value, while the final Q value may be estimated by this angle at the right boundary. As well, the component separation is indirectly characterized by the divergence between streamlines of both components, because the flow rate is constant between two streamlines in stationary case, and divergence of streamlines for both components means each component flows in different direction and the composition may change down the flow.

The streamline bending inside nozzle demonstrates the boundary layer effect, pronounced as boundary layer displacement. The qualitative consequence of this is the focusing of pentacene toward the axis (estimated Q enlargement), as well as a carrier gas, in comparison with a conical flow without boundary layer (in the case of non-viscid gas or a gas with specular reflection at the wall). The noticeable scattering of a gas behind the nozzle with $\mathrm{M}_G = 4$ testifies to defocusing of a flow (estimated Q decreasing) and points on the extension of the nozzle to increase the final Q value.

The role of boundary layer and separation processes are clearer from the representation of Mach number (Figure 2) and concentration (Figure 3) isolines.

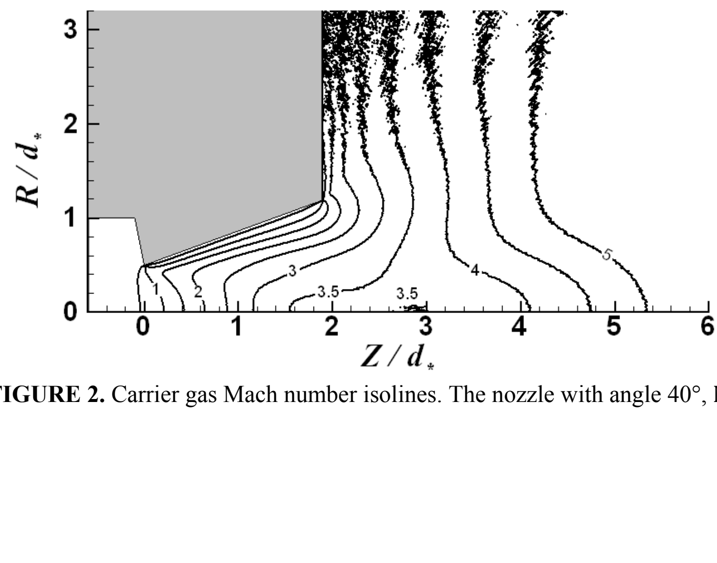

FIGURE 2. Carrier gas Mach number isolines. The nozzle with angle 40°, $\mathrm{M}_G = 4$.

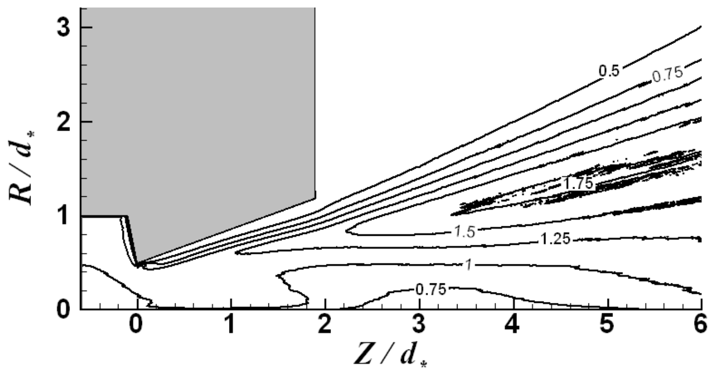

FIGURE 3. Pentacene relative concentration isolines. The nozzle with angle 40°, $\mathrm{M}_G = 4$.

The computation provides to determine all gas parameters in a flow field. The important information one can get from the comparison of transversal axial component velocity distributions in cross-section at geometrical Mach number $\mathrm{M}_G = 5$ for nozzles with $\mathrm{M}_G = 6$ at the exit and different angle openings (Figure 4). The velocity is normalized by the value of critical velocity. The velocity profiles for three considered nozzles at first sight are not very different, but one can notice that the isentropic (non-disturbed by boundary layer) flow core is clearly pronounced only in the case of the nozzle with the angle 40°. For this nozzle the flow slip by the wall (at the $\frac{R}{d_*} = 1.56$) is maximal, that correlates with the larger velocity gradient by the wall.

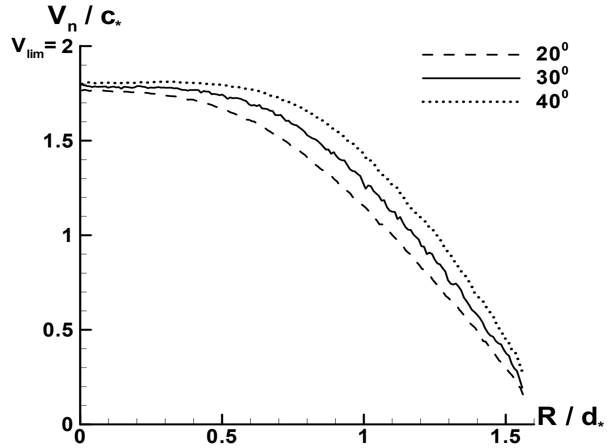

FIGURE 4. Helium axial velocity component in cross-section $\mathrm{M}_G = 5$ for different nozzles of $\mathrm{M}_G = 6$.

The boundary layer in the nozzle with small opening angle (20°) reaches the axis and the flow in the nozzle center becomes to be non-isentropic. The attainment of high exit Mach numbers with a low flow angle scattering is not possible in such nozzles. Such limitations are peculiar to nozzles with larger angle opening as well, but only for much larger geometrical Mach numbers.

Figure 5 presents real Mach numbers at the flow axis in the exit section of different nozzles. This data expose the tendency of real Mach number growth retarded by increasing of geometrical Mach number of nozzle exit. These results reflect quantitatively the known displacement effect of developing boundary layer that diminishes an expansion rate in the nozzle and retards the creation of high Mach number flow at a nozzle exit. The point for a nozzle with angle 40° and $\mathrm{M}_G = 4$ (Figure 5) is distinguished from a general regularity. More thorough analysis has shown, that for a given transonic configuration of the nozzle the shock wave structure close to a sonic orifice has generated mentioned above peculiarities.

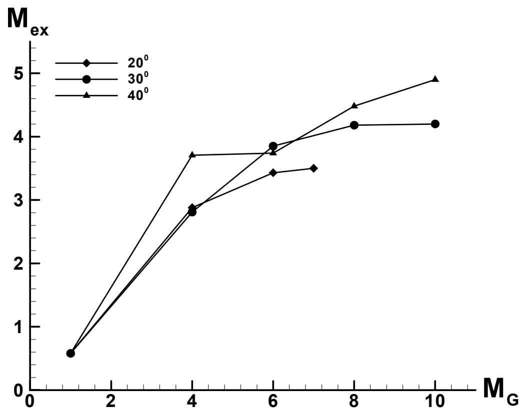

FIGURE 5. Mach numbers on the axis at the nozzle exit.

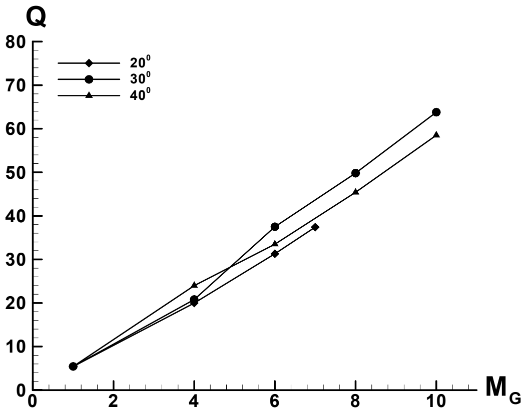

FIGURE 6. The directedness Q of pentacene flow for different nozzles.

The variation of relative intensity of heavy gas flux Q in the remote hypersonic region, where the angle displacement of streamlines is not essential, is shown in Figure 6 as dependence of geometrical Mach number in a nozzle exit M_G. The point $\mathrm{M}_G = 1$ belongs to sonic nozzle.

When addressing to Figures 1(a, b) one can notice that in the nozzle there are 3 different zones with the streamline bending causing different inertial gas separation. In the transonic region the convergent-divergent flow is favored by high heavy gas concentration to the axis; then due to influence of a boundary layer the tendency to the formation of a flat parallel flow is founded. At last, just behind the nozzle there is a streamline bending the more essential the less Mach number at the nozzle exit. All these effects can be different for nozzles with different angle and length. In the light of above outlined the dependence $Q(\mathrm{M}_G)$ in Figure 6 is very close to linear and practically coinciding for all nozzles (one can accept in error limits), seems to be unexpected result. It is very important to stress this main result of the performed investigation. The intensity of heavy component in a hypersonic region can be elevated even one order or more using nozzles with the geometrical Mach number for monatomic gas up to 10 and higher. It is evident now that this is just for nozzles with opening angle more 30°. A-priori one can note that the limit value of Q can be obtained by achieving flat parallel flow in which the intensity will be decreased by flow expansion in the process of parallel flow formation.

Data in Figure 6 testify to possible further magnification of Q.

CONCLUSION

The performed investigation shows wide possibilities of molecular beam system optimization of uniform and non-uniform system on the way of the choice of nozzles by numerical flow modeling.

The benefit of this work is much wider. In past time one of the problems of experimental research in rarefied gas dynamic was the formation of flat parallel isentropic flow by expansion in nozzles. The shaping of a supersonic nozzle part generatrix is hampered by strong influence of boundary layer changing with variation of flow parameters. It means that a specific nozzle with fixed geometry can be useful only for one regime characterized by Knudsen and Reynolds numbers. Modern computational possibilities take off the problem of the optimal nozzle design at given parameters of pumping for a given vacuum system.

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