Numerical investigation of flows with condensation in micronozzles

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Abstract. The paper considers the numerical simulation of the flow of argon with account for the condensation process in the micronozzle and behind it. To describe phase transitions, the initial mathematical model of viscous heat-conducting gas flow is supplemented with the equation of formation and growth of condensation nuclei in the flow. The developed mathematical model allows for simulating the process of gas condensation at low pressures and temperatures. It is shown that the condensate mass fraction in the flow is not less than 1% at the pressure and temperature of 5 bar and 200 K, respectively, when argon flows out of a micronozzle to the environment with the pressure of 0.01 Pa. At the nozzle exit, the size of condensed particles reaches 80 angstroms. The obtained results confirm the necessity to take into account the condensation phenomenon in micronozzle flows of inert gases.

1. Introduction

Modern trends in technological development focus on reducing the mass-size parameters, and lower power consumption has led to the emergence of a wide range of devices, in which micro- and nano-flows are implemented. An example of such devices is a supersonic micronozzle with a critical cross-section diameter much less than 1 mm. They can be used in micromotors for nanosatellites [1], as an “aerosol gun” for deposition of solid materials on a substrate in the production of solar cells and electronic microcircuits in the Collimated Aerosol Beam Direct-Write (CAB-DW) technology [2], to create a gas-jet target in a laser-plasma source of the shortwave radiation [3] or in studies of homogeneous condensation of various gases [4].

In [4] the results of experimental studies of homogeneous condensation of inert gases (argon, krypton, xenon) at different initial parameters of the outflow are given. They show that condensed phase clusters with the size of 6–10 nm are formed in the flow through a nozzle with a large expansion rate into a vacuum chamber. In [3], when modeling a xenon flow in micronozzles, it is concluded that the role of condensation in such flows is negligible.

The purpose of this study is to assess the contribution of condensation to the change of flow parameters and to identify the parameters of the condensation model [5] by comparing the simulation results with the known experimental data. Parametric studies based on numerical calculations will allow controlling flow parameters in condensed micro-nozzles in the future by changing the input parameters of the gas flow – the pressure and temperature of braking.
This work presents simulation of the argon microflow in a conical micronozzle with the radius of critical section of 170 μm, the cone apex angle of 8.6°, and the expansion degree of 36.6. The braking pressure varies from 2 to 10 bars, the braking temperature is 200 K, and the nozzle size and flow parameters are taken according to [4].

Due to the adiabatic expansion of the gas in the supersonic nozzle, a significant part of the initial enthalpy is converted into the kinetic energy. The peripheral flow near the nozzle walls is characterized by a high temperature due to viscous braking. In the central area of the supersonic part of the nozzle, the gas is cooled and a cold and dense gas core is formed in the flow. The formation of such a core is a necessary condition for the emergence of a condensed phase.

Simulation of the condensation process is based on thermodynamic properties of argon in the gaseous and condensed states. Due to the narrow range of argon in the liquid state (83.8 ÷ 87.3 K) and extremely low probability of condensation in this temperature range, argon in the solid state is taken because of the proper properties of substances [6], which is possible because of the properties of argon as an inert gas. The constancy of the heat capacity of inert gases at the temperatures above 24 K was shown in [7]. To determine the properties of the condensed (solid) phase and the heat of sublimation, data from [8, 9] were used. The viscosity and pressure dependences of saturated vapors on the temperature obtained as a result of this analysis were used in the model of argon motion in the micronozzle and in the model of gas condensation.

2. Mathematical models

2.1. Governing equations for viscous gas flow modeling

To simulate the flow of the viscous heat-conducting gas in a micronozzle, a system of equations of hydromechanics in the cylindrical coordinate system is used. Since the problem is solved in the axisymmetric statement, the initial system of equations is written in the form:

$$\frac{\partial Q}{\partial t} + \frac{\partial F_x}{\partial x} + \frac{\partial F_r}{\partial r} + F_{\theta 0} = \frac{\partial L_x}{\partial x} + \frac{\partial L_r}{\partial r} + L_{\theta 0},$$

where $Q$ is the vector of hydro-mechanical parameters (HMP); $F_x, F_r, F_{\theta 0}$ are the vectors of convective flows; $L_x, L_r, L_{\theta 0}$ are the vectors of diffusive flows; $x, r, \theta$ are the longitudinal and radial coordinates; and $t$ is the time. Vectors of HMP, convective and diffusive flows have the form:

$$Q = \begin{bmatrix} \rho \\ \rho u \\ \rho v \\ \rho e \end{bmatrix}, \quad F_x = \begin{bmatrix} \rho u \\ \rho u^2 + p \\ \rho u v \\ \rho u h \end{bmatrix}, \quad F_r = \begin{bmatrix} \rho v \\ \rho v u \\ \rho v^2 + p \\ \rho v h \end{bmatrix}, \quad F_{\theta 0} = \begin{bmatrix} \rho \frac{\partial v}{\partial r} \\ u \frac{\partial v}{\partial r} + \frac{v}{r} \frac{\partial u}{\partial r} \end{bmatrix},$$

$$L_x = \begin{bmatrix} 0 \\ \tau_{xx} \\ \tau_{xr} \\ u \tau_{xx} + v \tau_{rx} \end{bmatrix}, \quad L_r = \begin{bmatrix} 0 \\ \tau_{rx} \\ \tau_{rr} \\ u \tau_{rx} + v \tau_{rr} \end{bmatrix}, \quad L_{\theta 0} = \begin{bmatrix} 0 \\ 0 \\ \tau_{\theta r} - \tau_{\theta 0} \\ u \tau_{rx} + v \tau_{rr} \end{bmatrix},$$

here $u, v$ are the components of the velocity vector, $p$ is the pressure, $\rho$ is the density, $e$ is the specific energy, $h$ is the specific enthalpy, $\tau_{xx}, \tau_{rx}, \tau_{xr}, \tau_{rr}, \tau_{\theta 0}$ are components of the viscous stress tensor defined in the axisymmetric statement as follows:
\[
\tau_{xx} = 2\eta \frac{\partial u}{\partial x} - \frac{2}{3} \eta \text{div}\Omega,
\]
\[
\tau_{rr} = 2\eta \frac{\partial v}{\partial r} - \frac{2}{3} \eta \text{div}\Omega,
\]
\[
\tau_{\theta\theta} = -\frac{2}{3} \eta \text{div}\Omega,
\]
\[
\tau_{sr} = \tau_{rx} = \eta \left( \frac{\partial v}{\partial x} + \frac{\partial u}{\partial r} \right),
\]

where \( \eta \) is the coefficient of dynamic viscosity, \( \Omega = (u; v) \) is the velocity vector.

The divergence of the velocity vector is defined by the formula:
\[
\text{div}\Omega = \frac{\partial u}{\partial x} + \frac{\partial v}{\partial r} + \frac{\partial u}{\partial r} - \frac{v}{r},
\]

The total specific energy and total specific enthalpy are calculated by relations:
\[
e = \frac{u^2 + v^2}{2} + C_v T, \quad h = \frac{u^2 + v^2}{2} + C_p T.
\]

The state equation is used in the following form:
\[
p = \rho RT.
\]

Here \( C_v = 311 \text{J/(kg K)} \) is the specific heat capacity of gas at the constant volume, \( C_p = 520 \text{J/(kg K)} \) is the specific heat capacity of gas at the constant pressure, \( T \) is the temperature, \( R = 208 \text{J/(kg K)} \) is the specific gas constant, and \( \gamma = 1.67 \) is the adiabatic index.

2.2. The mathematical model of condensation

To describe the condensation effect, the initial model of equations (1)-(7) is supplemented with a mathematical model of formation and growth of condensation nuclei in the flow.

\[
\frac{\partial \rho G}{\partial t} + \frac{\partial \rho Gu}{\partial x} + \frac{\partial \rho Gv}{\partial r} + \rho Gv = g
\]

where \( G \) is the mass fraction of the condensate, and \( g \) is the mass speed of condensation calculated by the formula:
\[
g = 4\pi r_{cl}^2 \frac{p - p_S}{2\pi R_0 T} - N.
\]

In the equation (9), \( r_{cl} \) is the cluster radius, \( p_S \) is the saturated gas vapor pressure, \( R_0 = 8.31441 \text{J/(mol K)} \) is the universal gas constant, \( \mu = 39.948 \text{g/mol} \), and \( N = 10^{15} \text{m}^{-3} \) is the concentration of particles per unit volume.

The cluster size \( r_{cl} \) is determined by the ratio of the cluster mass \( m = \rho G / N \) to the mass of a single Argon molecule \( m_0 = \mu / N_A \) and is calculated by the formula:
\[
r_{cl} = r_0 \left( \frac{m}{m_0} \right)^{1/2},
\]
where $r_0 = 3.542\text{Å}$.

To account for the effect of condensation in the energy equation of system (1), the right side of the equation contains an additional term \( \Delta H_S \) that takes into account the heat of the phase transition [9]:

\[
\Delta H_S = 83.8058 \left[ 11.126 \left( 1 - \frac{T}{83.8058} \right)^5 + 11.188 \left( \frac{T}{83.8058} \right)^{0.8755} \right] R_0. \tag{11}
\]

The state equation changes as follows:

\[
p = \rho RT (1 - G). \tag{12}
\]

The dependence of the saturated argon vapor pressure on the flow temperature is given by the formula:

\[
\ln p_s = \frac{S_0 - S_z^0}{R_0} - \frac{\Delta H_S}{R_0 T}, \tag{13}
\]

where $S_0$ is the entropy of the gas phase [6], $S_z^0$ is the entropy of the solid phase [8], and $\Delta H_S$ is the heat of sublimation [9].

In modeling, the effect of temperature on the argon viscosity coefficient is taken into account in accordance with recommendations for pure gases given in [10].

3. Problem statement

The problem is solved by the method of control volumes based on the integral form of conservation laws (1–3) in laminar approximation. Discretization of the computational domain is performed using quadrangular cells, the total number of which is 307200 at the detailed resolution of the boundary layer.

Control volumes are chosen to coincide with the grid cells. The main variables are the mean values of HMP (density, pressure, energy, and flow velocity components) across the grid cells at the central points of cells.

Determining both convective and diffusive fluxes at the control cell boundary is done by linear approximation of the solution in each cell. The monotonous character of approximation is ensured by a limiting multiplier. An explicit Euler scheme is used to integrate the equations over time.

Calculations are performed using the time step determined from the estimate of viscous and non-viscous flows in cells of the computational grid. The minimum grid cell edge is 0.5 μm. This cell size significantly reduces the time step. When the condensation model is involved, the time step is limited to a value of $10^{-10}$s. As a result, the Courant number in calculations does not exceed the value of 0.05.

The solution procedure implies a multistep calculation algorithm that includes stepwise switching between the non-viscous solver, viscous algorithm, finite-volume grid crushing procedure, and addition of the condensation model. The described numerical schemes and algorithms are implemented in the author's code.

When modeling the outflow from a conical nozzle, it is necessary to consider the configuration of the gas jet that is formed behind the nozzle exit. Therefore, the computational domain includes both the nozzle itself and a part of the space behind it. The integration domain has the following boundaries: inlet, impermeable walls, and outlet.

At solid boundaries, the no-slip and impermeability conditions are set. At the inlet of the domain the stagnant flow conditions are $T^* = 200$ K, $p^* = 5$ bar. The gas flows out to the region of deep vacuum with the pressure of 0.01 Pa. At the outlet, depending on the flow-out mode, either a parameter drift (supersonic flow) or a fixed ambient pressure is set.
After the gas flow is established, the condensation model is involved. The origin of condensation is determined by the temperature taken as 45 K on the basis of solving a one-dimensional problem [11]. The problem is solved by the relaxation method. Calculation results are shown in Figures 1–3.

Figure 1 shows the distribution of the gas temperature and Mach number of the flow in the expanding part of the nozzle and behind it. The temperature distribution corresponds to the viscous gas flow. A braking zone with the increased gas temperature is formed near the walls. In the flow core, the gas temperature changes from a braking temperature of 200 K at the nozzle inlet to 10 K at the outlet (Figure 2). Due to a significant temperature decrease, the gas flow accelerates up to 6 Mach behind the nozzle.

![Figure 1](image1.png)

**Figure 1.** Distribution of the temperature (a) and Mach number (b).

The change in the axial temperature is shown in Figure 2. The appearance of the condensed phase in the flow leads to an increase in the gas temperature in the region of condensation. In Figure 2, this region corresponds to the outlet cross section of the nozzle (x~11 mm).

![Figure 2](image2.png)

**Figure 2.** Temperature diagram on the axis.

The percentage of the condensate in the flow and the radius of the particles are shown in Figure 3. A significant portion of the condensed gas corresponds to the area behind the nozzle exit. At the nozzle exit the particle radius is 80 angstroms and corresponds to the experimental data [4].
Conclusions
The numerical simulation of the argon outflow in vacuum has been performed. Our studies confirm that the developed mathematical model allows for calculating the viscous heat-conductive flow of the inert gas and modeling the process of gas condensation at low temperatures and pressures. Numerical simulation of the flow at a braking temperature of 200 K and a braking pressure of 5 bars has been performed as well. The beginning of the condensation process is shown to correspond to the nozzle exit. Behind the nozzle, there is the forming of a flow region filled with the condensate with particle sizes up to 100 angstroms. Thus, even for argon with the low phase transition temperature (only 45 K), the mass fraction of the condensate in the flow reaches 1%. It means that for heavier inert gases, for example, xenon or krypton, the condensate fraction will be significantly higher and one cannot neglect the condensation phenomenon when calculating such flows.

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