Numerical and experimental study of a warming up effect of an underexpanded rarefied rf plasma jet outflowing into a flooded area

A Yu Shemakhin¹, V S Zheltukhin² and A A Khubatkhuzin²
¹ Kazan Federal University, Kremlyovskaya 18, Kazan, Tatarstan 420008, Russia
² Kazan National Research Technological University, Karl Marx Street 68, Kazan, Tatarstan 420015, Russia
E-mail: shemakhin@gmail.com

Abstract. A mathematical model of the rf plasma flow at 13.3–133 Pa in transition regime at Knudsen number values $8 \times 10^{-3} \leq Kn \leq 7 \times 10^{-2}$ and the nozzle pressure ratio $n = 10$ for the carrier gas is described. The model based on both the statistical approach to the neutral component of the rf plasma and the approach to the continuum model for electron and ion components. The results of plasma flow calculations performed both for an undisturbed flow and for the stream with a sample at a prescribed electric field are described. The effect of a warming up of a stream in a mixture zone confirmed by comparison of numerical results with experimental ones is found.

1. Introduction
Radio frequency (rf) plasma discharges at low pressures ($P = 13.3–133$ Pa) are successfully used for the modification of various materials: dielectrics, conductors, semiconductors [1–5]. The plasma shows the following properties: ionization degree is $10^{-7}–10^{-5}$, electron density $n_e$ is $10^{15}–10^{19}$ m$^{-3}$, the electron temperature $T_e$ is 1–4 eV, the temperature of atoms and ions $T_a$ ranges within $(3–4) \times 10^3$ K and $(3.2–10) \times 10^2$ K in the plasma bunch and in a plasma stream, respectively.

The feature of low pressure rf plasma stream is a transient flow mode between the free-molecule flow and static continuum. When the Knudsen’s parameter is in range of $8 \times 10^{-3} \leq Kn \leq 7 \times 10^{-2}$ as in the case under study, using the Navier–Stokes equation is not correct for flow description [6, 7]. At the same time, both electron and ion gases satisfy continuity, since their movement is determined not only by moving together with the gas flow, but also by the Coulomb force action that prevents the separation of charges [8].

The direct statistical Monte-Carlo (DSMC) modeling [9–12] is widely used for the numerical solution of problems of the rarefied gas dynamics. The method is based on the splitting of the Boltzmann’s equation on the movement and collision processes that allows us to describe the gas-dynamic processes in the transition mode for neutral environment. Heating atoms and ions in a low-temperature plasma occurs mainly due to elastic collisions of those with electrons, which is equivalent to the presence of the allocated heat source in the plasma stream. Therefore, both modification of the Bird’s method for calculation of rarefied rf plasma flows and bringing it in compliance with the continuous model of charged particles are required.
The hybrid mathematical model, combining a kinetic model for the carrier gas flow at Knudsen number values $8 \times 10^{-3} \leq Kn \leq 7 \times 10^{-2}$ with the continuous model for charged particles was developed earlier [13–15]. The aim of this work is to study the effect of low pressure rf plasma jet overheating and to compare the numerical results with experimental data.

2. Mathematical model of low pressure rf plasmas stream

Mathematical model of the low pressure rf plasma stream [13–15] is constructed by neglecting the following effects: the Hall’s effect, electron pressure gradient, the radiation energy loss, the electron attachment, the excitation of atoms, bulk recombination, formation of multiply charged ions and ions slipping. A direct electron impact as the basic mechanism of the formation of charged particles is assumed. We also assume that the ion density is equal to the electron one, the ion temperature coincides with the temperature of neutral atoms.

Let the radius of the cylindrical vacuum chamber be denoted by $R_{vk}$, its length—by $L_{vk}$, the radius of the plasma torch outlet—by $R_k$, the subscripts inlet, outlet, body, walls should be used for parameter values on inlet and outlet of the chamber, on the walls of the sample and the vacuum chamber, respectively.

The model includes:

1) the Boltzmann’s transport equation for neutral atoms:

$$\frac{\partial f}{\partial t} + c \cdot \frac{\partial f}{\partial r} + \tilde{F} \cdot \frac{\partial f}{\partial c} = S(f), \quad (1)$$

2) the equation of the electron continuity:

$$\frac{\partial n_e}{\partial t} - \text{div} (D_a \text{grad} n_e - v_a n_e) = \nu_i n_e \quad (2)$$

3) the equation of the electron heating:

$$c_p \rho_e \frac{\partial T_e}{\partial t} - \text{div} \left( \lambda_e \text{grad} T_e - \frac{5}{2} k_B n_e T_e v_e \right) + \frac{3}{2} k_B \delta \nu_e n_e (T_e - T_a) = \sigma E^2 - \nu_i n_e E_I. \quad (3)$$

Here $t$ is time, $c$ and $r$ are vectors of velocity and the coordinates of the atoms, respectively, $f(c, r, t)$ is the velocity distribution function of neutral atoms, $S(f)$ is the collision integral, $\tilde{F}$ is the reduced force which effects neutral atoms under elastic collisions with electrons, $D_a$ is ambipolar diffusion coefficient, $\nu_i$ is the ionization frequency, $v_a$ is the gas stream velocity, $\lambda_e$ is thermal conductivity coefficient of electrons, $c_p$ is the heat capacity of the electron gas, $v_e$ is the electron gas velocity, $\nu_e$ is elastic collision frequency of electrons and atoms, $\sigma$ is plasma conductivity, $E$ is electric field strength, $E = |E|$, $E_I$ is ionization potential, $k_B$ is the Boltzmann’s constant, $\delta = m_e/2m_a$ is the fraction of energy which electrons loss in elastic collisions, $m_e, m_a$ are the weights of the electron and the atom, respectively, $\rho_e = m_e n_e$ is the electron gas density. The expression $\nu_i n_e$ in the right hand of the equation (2) describes the rate of burning of charged particles. The expression $\nu_i n_e E_I$ in the right hand of the equation (3) describes the energy lost under the ionisation process.

Coefficients $D_a, \nu_i, \lambda_e$ are functions of the electron temperature [14–17],

$$\tilde{F} = -\frac{1}{m_a} \text{grad} W_T, \quad W_T = \int E_e \text{dV} dt, \quad E_e = \frac{3}{2} k_B \delta \nu_e n_e (T_e - T_a).$$

The system (1)–(3) is considered under the following initial conditions:

$$f(c, r, 0) = f_0(c, r), \quad n_e|_{t=0} = n_{e0}, \quad T_e|_{t=0} = T_{e0}, \quad (4)$$
and the following boundary conditions:

\[ n_e|_{\text{inlet}} = n_{e,\text{inlet}}, \quad n_e|_{\text{outlet}} = n_e|_{\text{walls}} = 0, \quad n_e|_{\text{body}} = 0, \]
\[ T_e|_{\text{inlet}} = T_{e,\text{inlet}}, \quad T_e|_{\text{outlet}} = T_e|_{\text{walls}} = 300 \text{ K}, \quad \frac{\partial T_e}{\partial n}|_{\text{body}} = 0, \]

where: \( f_0 \) is Maxwell velocity distribution function, \( \mathbf{n} \) is the normal vector to the surface, \( n_{e,0}, T_{e,0}, n_{e,\text{inlet}}, T_{e,\text{inlet}} \) are the preset values of the corresponding functions in an initial timepoint. The impermeability conditions on the \( \text{body} \) and \( \text{walls} \) boundary surfaces as well as soft boundary conditions on the \( \text{inlet} \) and \( \text{outlet} \) borders are defined for \( f(c, \mathbf{r}, t) \).

The equations (1)–(3) are closed by following relations:

\[
\mathbf{v}_a(r, t) = \int_{-\infty}^{\infty} c f(c, \mathbf{r}, t) \, dc, \quad P = n_a k_B T_a,
\]

\[
\mathbf{v}_e = \mathbf{v}_a - (D_a/n_e) \, \text{grad} \, n_e, \quad \sigma = \frac{n_e c^2 \nu_e}{m_e \left( \nu_e^2 + \omega^2 \right)},
\]

\[
T_a = \frac{m_a}{3 k_B} \left( \overline{c^2} - \overline{v_a^2} \right), \quad \overline{c^2} = \int_{-\infty}^{\infty} c^2 f(c, \mathbf{r}, t) \, dc,
\]

Here \( n_a \) is the neutral atom density, \( e \) is the electron charge, \( \omega = 2\pi f \) is the cyclic frequency of the electric field, \( f \) is the generator frequency.

The Bird’s method was modified to take into consideration the distributed heat source density \( W_T \). A two-step iterative process is developed to solve the problem (1)–(7). At the first step, a solution of (1) was found by the Bird’s DSMC method [9] to determine \( \mathbf{v}_a \) and \( T_a \). Then, these values are used to solve problems (2), (4), (5) and (3), (4), (6). Further, the solutions of these problems \( n_e \) and \( T_e \) are used to solve the equation (1) taking into account the distributed heat source power density \( W_T \). The process is repeated until the maximum of successive approximation ratios becomes less than the specified tolerance. The software package for calculating low pressure rf plasma flow was developed by using OpenFOAM [18] environment and C++ program language for Linux OS.

### 3. Calculations of low pressure rf plasma flow overheating

The gas-dynamic characteristics of both low pressure rf plasma undisturbed flow and the stream with overflowing sample in the vacuum chamber of \( R_{vk} = 0.2 \text{ m}, L_{vk} = 0.5 \text{ m} \) and \( R_{lk} = 0.012 \text{ m} \) at the center of the base plate were calculated. It is assumed that a cylindrical sample with the radius \( R_b = 0.03 \text{ m} \) and a height \( L_b = 0.02 \text{ m} \) is located in the plasma jet at a distance \( L_{ib} = 0.2 \text{ m} \) from the inlet.

Flow input parameters are the following: the plasma forming gas is argon, gas flow rate \( G \) is 0.12–0.24 g/s, pressure \( P_{\text{inlet}} \) is 35–85 Pa, the temperature \( T_{\text{inlet}} \) is 400–600 K, the degree of ionization \( \delta_i = 10^{-4} \). The initial pressure in the vacuum chamber \( P_0 \) is 3.5–8.5 Pa. As shown by calculations, steady state flow is established for \( t \sim 10^{-2} \text{ s} \). The plots of velocity, temperature and pressure distribution of the carrier gas, the concentration of electrons, and the electron temperature were mostly given in earlier works [13–15]. However, those results do not describe the effects of gas overheating in the mixing zone where plasma flow and stationary gas are mixed in the vacuum chamber. Figures 1 and 2 show the results of calculations of the plasma jet characteristics at \( G = 0.24 \text{ g/s}, T_{\text{inlet}} = 500 \text{ K}, \) gas velocity \( v_{\text{inlet}} = 1000 \text{ m/s}, \) \( P = 60 \text{ Pa}, P_0 = 6 \text{ Pa}, \) with the two latter corresponding to the nozzle pressure ratio \( n = 10 \). The curves in figure 1 show that the pressure decreases at a distance of 0.05–0.15 m from the
The radial distribution of the gas pressure in the cross-section of stream flowing around
the sample.

Figure 1. Radial distribution of the gas pressure in the cross-section of stream flowing around
the sample.

inlet (curves 2, 3) and increases near the sample (curve 4), which results from deceleration
of the flow. The lower pressure zone is created right downstream of the sample, and the pressure
stabilizes further downstream of the sample, as in the case with the model without sample. As
a result, numerical experiments revealed the heating effect of the jet on the periphery of the
stream in the mixing zone of the plasma flow and the stationary gas in vacuum chamber. The
curves on figure 2 show that at a distance of 0.01 m from the inlet, the maximum temperature
(\(\sim 650\) K) is created on the periphery of the jet, and the minimum temperature (\(\sim 520\) K) is
observed at the center (curve 1).

The radial temperature profile is completely aligned across the stream at a distance from
the sample. Thus, at a distance of 0.05 m from the inlet, the temperature in the mixing zone
\(T \sim 460\) K and that at the flow axis \(T \sim 350\) K (curve 2). The gas overheating is most likely
to be due to a sharp deceleration of the gas atoms at \(z = 0.15\) m by reason of collision with a
stationary gas. Radial temperature profile is leveled at a distance of \(z = 0.15\) m from the inlet
closer to a bell-shaped profile; the maximum temperature \(T_a = 650\) °C reaches at the center of
flow (curve 3). The gas is cooled to 570 °C at the distance of 0.2 m when the flow is collided with
the sample, and at a distance of \(\sim 0.03\) m from the axis a drastic temperature fall by \(\sim 30\) °C
is observed (curve 4). This effect is connected with the interaction of several factors. Firstly,
the gas is cooled due to both flow expansion and interaction with the colder surface having the
constant temperature (300 K), which corresponds to the sample being cooled. Secondly, braking
flow leads to gas heating, whereby the radial profile of the peak in the center of the jet becomes
narrower as compared with curve 3, and a gap at the distance of 0.3 m from the axis appears.

The temperature difference between the axis and the edges of the stream is 100 K, probably
resulted from gas flow around the sample under cooling. The temperature distribution over the
cross section is completely aligned with the jet at a distance of 0.3 m from the inlet, the profile
becomes flattened. The maximum gas temperature \(T_a = 420\) K is achieved at the flow axis. Gas
flow cooling is caused by the expansion and uniformity of the profile of the jet cross section due
to elastic collisions of electrons with atoms.
4. Comparison of numerical results with experimental data
Experimental measurements of the plasma temperature at the exit of the plasma torch with thermocouples were performed to confirm the revealed laws. The maximum allowable temperature was 1000 °C. The experiments were conducted at the equipment which was described in [1–3] for the rf inductive discharge jet. The equipment allows the rf plasma flows to be generated with the parameters specified in the introduction to this paper. Thermocouples
Figure 4. Radial temperature distribution of the carrier gas at the outlet of the plasmatron (experiment).

were placed on the flange with a certain pitch along the radius (Figure 3). The flange is placed at the inlet to the vacuum chamber. The temperature was measured along the jet radius at a small distance from the inlet. The experiment revealed that the temperature at the second thermocouple is higher than that in the jet center at the inductor current of 1.8 A, and at the pressure in the vacuum chamber (for pumping) of 60 Pa (figure 4), which indicates the presence of hot spots. Thus, experiments confirmed the calculation results.

5. Conclusion
The peculiarities of the gas temperature distribution in the low pressure rf plasma jet in the vacuum chamber at sample overflowing are found as a result of the numerical simulation at flow parameter nozzle pressure ratio of 10. Gas overheating in the mixing zone where the plasma stream and stationary gas are mixed in the vacuum chamber is observed at the vicinity of the inlet. The difference between the maximum temperature and the temperature on the flow axis is 110 K. The radial temperature distribution is aligned at a distance from the inlet. In the zone of stream collision with the sample being processed the temperature jump caused by the braking of a stream is observed. Temperature profile becomes uneven behind a sample that is caused by the expansion of a stream, and due to interaction with colder sample. Temperature profile in the cross section of a stream is leveled at the distance of 0.3 m from the inlet.

The effect of plasma stream overheating on the periphery of the stream is consistent with experimental results.

Acknowledgments
The work was funded by the Russian Foundation for Basic Research (according to the research project No. 16-31-60081, the theoretical part) and the Ministry of Education and Science of the Russian Federation (project No. 2196, the experimental part).
References

[1] Abdullin I S, Zheltukhin V S, Sagiiev I R and Shayekhov M F 2007 *Modifikaciya Nanosloev v Vysokochastotnoj Plazme Ponizhennogo Davleniya* (Kazan: Izdatel’stvo Kazanskogo Tekhnologicheskogo Universiteta)

[2] Khubatkhuzin A A, Abdullin I S, Gatina E B, Zheltukhin V S and Shemakhin A Y 2012 *Vestn. Kazan. Tekhnol. Univ.* **15** 72

[3] Khubatkhuzin A A, Abdullin I S, Bashkirtsev A A and Gatina E B 2012 *Vestn. Kazan. Tekhnol. Univ.* **15** 71

[4] Shatayeva D R, Kulevtsov G N and Abdullin I S 2014 *Vestn. Kazan. Tekhnol. Univ.* **17** 73

[5] Miftakhov I S, Fadeyev A O and Shayekhov M F 2013 *Vestn. Kazan. Tekhnol. Univ.* **16** 42

[6] Belotserkovsky O M 1984 *Chislennoe Modelirovanie v Mekhanike Sploshnykh Sred* (Moscow: Nauka)

[7] Dulov V G and Lukyanov G A 1984 *Gazodinamika Processov Istecheniya* (Moscow: Nauka)

[8] Mitchell M and Kruger C H 1973 *Partially Ionized Gases* (New York: John Wiley and Sons)

[9] Bird G A 1976 *Molecular Gas Dynamics and the Direct Simulation of Gas Flows* (Clarendon Press: Oxford)

[10] Pechatnikov Y M 2002 *Vakuumnaya tehnika i tehnologiya* **12** 227

[11] Pechatnikov Y M 2004 *Prikl. Fiz.* **2** 19

[12] Bird G A 1998 *Computers Math. Appl.* **35** 1

[13] Zheltukhin V S and Shemakhin A Y 2011 *Uch. Zap. Kazan. Univ., Ser. Fiz.-Mat. Nauki* **4** 135

[14] Zheltukhin V S and Shemakhin A Y 2014 *Math. Model. Comput. Simul.* **6** 101

[15] Abdullin I S, Zheltukhin V S, Khubatkhuzin A A and Shemakhin A Y 2014 *Matematicheskoe Modelirovanie Gazodinamiki Strujnykh Techenij Vysokochastotnoj Plazmy Ponizhennogo Davleniya* (Kazan: Izdatel’stvo Kazanskogo Tekhnologicheskogo Universiteta)

[16] Raizer Y P 1991 *Gas Discharge Physics* (Berlin Heidelberg: Springer-Verlag)

[17] Lyumberopoulos D P and Economou D J 1993 *J. Appl. Phys.* **73** 3668

[18] OpenFOAM OpenFOAM Foundation. *Free Open Source CFD 2011–2016* http://www.openfoam.org