Mathematical modelling of the supersonic induction plasma jet

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Abstract

In the paper we apply numerical simulation to study the supersonically accelerated argon plasma flow downstream of the induction plasma torch. We compare the jets exhausting from two different convergent-divergent nozzles by means of two-temperature model. The results show that the recombination resulting in electron gas heating is more essential in the jet flowing from the nozzle with a higher outlet Mach number. The composition of the jet exhausting from the nozzle with a lower outlet Mach number remains almost frozen until the end of the first expansion zone. These results confirm that the chamber pressure reduction and the nozzle design changing lead to the induction plasma jets with different chemical conditions. The conclusion is that depending on the industrial process, one can choose the proper torch nozzle geometry to have plasma with the required properties.

1. Introduction

Standard plasma sources for plasma processing and plasma chemistry applications such as radio-frequency inductively coupled plasma torches and direct current torches until recently have operated mostly in subsonic regime and the choice of the operating conditions was rather limited. By means of continues pumping, the pressure in the working chamber can be reduced enough to produce a supersonic plasma, which became of a great interest during the last decade. The examples of the successful supersonic plasma applications are plasma assisted chemical vapour deposition of diamonds and polymer-like films [1-4] and nanoscaled deposition. In most technological applications the plasma accelerates up to supersonic speed using a specific torch nozzle design [5]. Supersonic plasma jets are often out of local thermodynamic equilibrium (LTE). Nonequilibrium plasma, however, can be beneficial for industrial applications. In plasma deposition technique it is important to perform a fast transport of the reactive products obtained in the discharge region. Supersonic jet, where the chemistry is almost frozen, can be the advantageous transport tool comparing with the subsonic jet, where a set of chemical reactions takes place. This example shows that the investigation of the possible deviation from LTE in supersonic plasma is important to control the industrial processes. In the present paper, we simulate nonequilibrium effects in supersonic induction plasma jet of pure argon using the standard two-temperature model [6]. We do not investigate the discharge region; it is beyond the scope of the present paper. We examine instead the accelerated plasma flow downstream of the discharge. It is important to note that the results of the present study are applicable when the plasma is firstly formed and than accelerated. These conditions are typical for induction torches with a supersonic nozzle and low-pressure chamber. The purpose of the paper is to compare the plasma jets flowing from the supersonic nozzles of two different configurations. The schematic of induction torch with a supersonic nozzle is shown in Fig.1, the nozzle dimensions are given in Table 1. The working chamber pressure is the same for both cases, so that the effect of torch nozzle geometry on plasma properties is studied.

![Fig.1 Induction torch with a nozzle](image)

Table 1. Nozzle dimensions, mm

<table>
<thead>
<tr>
<th>M1.5</th>
<th>M3</th>
</tr>
</thead>
<tbody>
<tr>
<td>d_e</td>
<td>12.85</td>
</tr>
<tr>
<td>d_o</td>
<td>13.88</td>
</tr>
<tr>
<td>L</td>
<td>18.38</td>
</tr>
</tbody>
</table>

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2. Gas dynamic model

We assume that the pressure is high enough to describe plasma flow by continuum fluid dynamic equations. We incorporate a two-temperature model into the computational fluid dynamic code FLUENT (FLUENT is a registered trademark of FLUENT Inc. Ceterra Resource Park, 10 Cavendish Court, Lebanon, NH 03766 U.S.A.). The considered flow is mostly supersonic and the coupling between momentum and energy equations of Navier-Stokes system is very strong. That why is we choose a coupled technique [7] solving simultaneously the equations written in matrix form. The Navier-Stokes equations become numerically very stiff at low Mach number due to the disparity between the fluid velocity and the speed of sound. The numerical stiffness of the equations under these conditions results in poor convergence rates. This difficulty is overcome in FLUENT’s coupled solver by employing a technique called time-derivative preconditioning [8]. The jet is assumed to be turbulent; a realizable k-ε model [9] was used for turbulence simulation because this model predicts axisymmetric jet properties better than a usual k-ε model.

3. Physical model

Due to the comparatively high chamber pressure (about 14 Torr) in the considered problem, we can simplify the study making the following assumptions: the velocity distributions of electrons, atoms and heavy particles are assumed to be Maxwellian; the plasma is ideal. The temperature of ions is taken to be equal to the temperature of atoms and is called a heavy particle temperature; this temperature can deviate from the electrons temperature. The quasielectroneutrality assumption is valid, self-induced electric field is neglected. The mass fraction of double ions is negligible and only the first ionisation and three particle recombination reactions are taken into consideration.

We will use the subscripts \( a \), \( ion \), and \( e \) to denote atom, ion and electron values correspondingly. The electron mass fraction \( C_e \) transport equation in axisymmetric cylindrical coordinates \( x, r \) is written in the form

\[
\frac{\partial}{\partial x} \left( \rho U C_e \right) + \frac{1}{r} \frac{\partial}{\partial r} \left( \rho V C_e \right) = \frac{\partial}{\partial x} \left( \rho D_{amb} \mu_a \frac{\partial C_e}{\partial x} \right) + \frac{1}{r} \frac{\partial}{\partial r} \left( \rho D_{amb} \mu_a \frac{\partial C_e}{\partial r} \right) + S_e,
\]

where \( U \) is the gas velocity axial component, \( V \) is the gas velocity radial component, \( \rho \) is the gas density, \( \mu_a \) is the turbulent viscosity, \( Sc_e = 0.9 \) is the turbulent Schmidt number, and \( D_{amb} \) is the coefficient of ambipolar diffusion [10]. \( S_e \) is the source term, which includes the changes of electron concentration due to the thermal ionization and three particle recombination reactions \( Ar^+ + e^- \leftrightarrow Ar^0 + e^- + e^- \). The expression used for this term is the following

\[
S_e = K_{ion} \rho \rho C_a C_{ion} - K_{rec} \rho^3 C_e^2 C_{ion},
\]

where the \( m \) and \( C \) are the species mass and mass concentration respectively, \( K_{ion} \) is the ionization rate, \( K_{rec} \) is the rate of recombination [6]. The electron temperature transport equation is

\[
\frac{\partial}{\partial x} \left( \rho n_e T_e \right) + \frac{1}{r} \frac{\partial}{\partial r} \left( \rho r V n_e T_e \right) = \frac{\partial}{\partial x} \left( \frac{2 \lambda_e}{k} \frac{\partial T_e}{\partial x} \right) + \frac{1}{r} \frac{\partial}{\partial r} \left( \frac{2 \lambda_e}{k} \frac{\partial T_e}{\partial r} \right) - \frac{2}{5k} E_{eh} - \frac{2}{5k} W - \frac{2}{5k} R^0,
\]

where \( T_e \), \( n_e \) and \( \lambda_e \) are the electron temperature, number density and thermal conductivity correspondingly, \( k \) is the Boltzmann constant. The right hand side of the equation contains the term \( E_{eh} \) representing the net volumetric energy exchange between the electrons and the heavy
particles, the ionization losses term \( W \), and the net volumetric radiation losses term \( R^0 \). The energy exchange term is calculated as follows

\[
F_{ch} = \frac{m_e}{m_a} n_e \bar{v}_{ch} (T_e - T). 
\]

where the elastic collision frequency \( \bar{v}_{ch} \) is a sum of the electron-atom \( v_{ea} \) and electron-ion \( v_{ei} \) collision frequencies according to the formulas

\[
\bar{v}_{ch} = v_{ea} + v_{ei}, \quad v_{ea} = n_e \sigma_{ea} (8kT_e / \pi m_a)^{1/2}, \quad v_{ei} = n_e \sigma_{ei} (8kT_e / \pi m_i)^{1/2}. 
\]

The electron-atom cross-section \( \sigma_{ea} \) is calculated according to Ref. [6], the electron-electron collision cross-section \( \sigma_{ee} \) is assumed to be the same as that of ion-electron [11]. The ionisation loss term contains \( l = 15.45 \text{eV} \) - the argon ionisation potential, and electron production term \( W \), written in the following form

\[
W = \frac{\partial}{\partial x} \left( (l + \frac{1}{2} \frac{\partial}{\partial r} (rV_n e) \right) 
\]

The volumetric radiation loss is represented as in Ref. [12]. The radiation to the ground state is mathematically taken into account in the \( S_e \) source term. The state equation is written in a two-temperature form. The computational domain used for modeling an axisymmetric jet flowing from the convergent-divergent nozzles is shown in Fig.2. At the nozzle inlet the total mass flow rate \( 60 \text{slm} \) is given. The constant heavy particle temperature \( 10000 \text{K} \) is assumed to be equal to the electron temperature and equilibrium value of the electron number density is assigned at the inlet. At the wall we use a constant condition for the heavy particle temperature, a zero normal derivative condition for the electron temperature and non-slip velocity boundary condition. At the chamber outlet the pressure \( P_e = 1800 \text{Pa} \) is specified. The computations start on a rather coarse grid than the grid is adapted and the calculations proceed until the residuals of all the equations become less than \( 10^{-6} \).

4. Nozzle design effect on the gas dynamic structure of the jet

Fig.2 represents the calculated contours of the Mach number and the heavy particle temperature in M3 (top) and M1.5 (bottom) jets. It can be noticed that because of the
deviation from isentropic flow conditions and because of the effect of viscosity, the transition from the subsonic flow to supersonic one occurs a little downstream from the nozzle critical section. Besides, the Mach numbers at the nozzle outlets deviate from the predicted ones by the isentropic assumption. Both jets are underexpanded, because the static pressure at the chamber inlet is higher than the ambient static pressure, as can be seen from Fig.5. The model predicts well all the main features of a moderately underexpanded supersonic jet. The flow along the jet axis within the computational area is wholly supersonic. By means of the alternating expansion and compression zones, the pressure tends to equilibrate with the ambient chamber pressure. One can see from Fig.2-5 that the first expansion wave is the strongest one, the further from the nozzle the weaker the oblique waves are until the static pressure becomes equal to the ambient chamber pressure. The axial velocity rises about one hundred times in the supersonic nozzle. It means that the supersonic jet performs very fast transport of the reactive species from the torch to the substrate, providing small residence time.

5. Nozzle design effect on the deviations from LTE in the jet

Fig.3 a), b) compare the heavy particle temperature and electron temperature along the jet axis for two nozzle configurations. Both the heavy particles and the electron temperatures drop inside the nozzle. However, as can be seen from the figures, the electron temperature does not drop as much as the heavy particle temperature does. This is especially true for the nozzle with the higher Mach number. The higher Mach number, the more the temperature drop along the nozzle axis, the more essential the recombination becomes. In the three-particle recombination, the electrons as third particles gain the energy. At the outlet of the M=3 nozzle the electrons are heated more by the recombination than at the outlet of the M=1.5 nozzle. The axial degree of thermal nonequilibrium $\theta = T_e / T$ correlates well with the local Mach number (Fig.4). We can conclude that the deviation from the local thermal equilibrium is higher in the expansion regions. In these regions the pressure drops and the velocity rises. In the compression regions, where the pressure rises and the velocity drops, the electron temperature is closer to the temperature of the heavy particles. In the supersonic induction jet, the electron number density is rather convective flux controlled than ionization-recombination reaction controlled. That is why, its values are several orders of magnitude higher than the equilibrium values $n_{e\, eq}$ calculated by the Saha equation at the electron temperature,
and closer to the frozen values $n_{\phi}$. One can find these deviations from ionization equilibrium in the both jets. Fig. 5 shows the axial profile of $n_e/n_{eq}$, the degree of ionization nonequilibrium and Fig. 6 shows $n_e/n_{\phi}$, the deviation from the frozen chemistry assumption. The axial degree of ionization nonequilibrium is diversely proportional to the static pressure (Fig. 5). Using Fig. 6 and making comparison of Fig. 3 a) and b) we can conclude that in the jet flowing from $M=1.5$ nozzle, the chemistry is close to frozen until the end of the first expansion zone, while in the jet formed by $M=3$ nozzle the recombination is more significant. These facts explain why the deviation from the thermal equilibrium is in general more pronounced in the jet exhausting from $M=3$ nozzle, than from $M=1.5$ nozzle.

The thermal energy gained in the compression regions or lost in the expansion zones from the kinetic energy transfer is proportional to the mass of the particle. This fact explains why the gas compression and expansion influence more the gas of the heavy particles, than the electron gas. The electrons also experience compression and expansion following the ions due to the electrostatic forces establishing the electroneutrality. The mentioned facts result in the more pronounced maximums and minimums of axial heavy particle temperature than the corresponding electron temperature, so that even regions with higher heavy particle temperature than electron temperature can occur. There is not enough time for full relaxation after each of the first several oblique waves so that the next wave forms in a nonequilibrium gas. Fig. 7 shows the temperatures radial profiles for the $M=3$ nozzle configurations. These profiles have the form of the curve with one axial maximum in the compression zones and with off axial maximum in the expansion zone, where the maximum corresponds to the barrel shock. The electron temperature and electron number density radial profiles (Fig. 7) also reflect the position of the compression, expansion zones and barrel shocks. One can see that the deviation from thermal equilibrium is very pronounced in the fringes of the jet where the gradients of all macroscopic parameters are very big.

6. Summary and conclusions
To understand the chemical kinetics mechanisms in the plasma quenched by the nozzle expansion detailed study of the different deviations from LTE in that flow is performed. Argon plasmas exhausting from two configurations of the convergent-divergent nozzles are compared by means of the numerical modeling. The considered jet core is characterized by rather long and strongly nonuniform supersonic region with alternating zones of oblique expansion and compression waves by means of which the static pressure tends to equilibrate with the chamber pressure. Along the jet axis, the deviation from thermal equilibrium is more essential in the jet flowing from the nozzle with the higher Mach number. This fact is due to the increased role of the recombination resulting in the electron gas heating. Therefore, a
conclusion can be made that the nozzle design and the chamber pressure changing leads to the plasma jets with different properties. The chemical conditions in these jets can be in the following range:

- equilibrium (when one uses the atmospheric pressure chamber and a straight cylindrical nozzle);
- frozen (when one uses low Mach number nozzle and moderately subatmospheric chamber pressure);
- recombining (when one uses high Mach number nozzle and moderately subatmospheric chamber pressure or for any nozzle when very low chamber pressure is used).

In general, the results prove that the supersonic configuration of induction plasma allows avoiding the ionisation (and possibly dissociation) processes downstream of the torch. This configuration can separate spatially the different functions of the plasma source such as the dissociation and ionisation of the original material, transport of the produced reactive particles towards the substrate and surface processing. We can conclude, that in this configuration one can easily control the properties of the plasma jet in the transport region by choosing the appropriate nozzle design.

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References