Structure and LTE analysis of direct current supersonic plasma jets

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Abstract
Mathematical modeling and optical emission spectroscopy are applied to study the effect of the chamber pressure on the structure and nonequilibrium effects of supersonic plasma jets formed by direct current arc. The results show that when the chamber pressure is low, a strongly underexpanded jet is formed. For higher ambient pressure values, the core region of the jet changes to a moderately underexpanded structure. Both the modeling and the emission spectroscopy show that the axial electron number density in the jet is much closer to its frozen values than to the equilibrium values. Downstream of the jet core, the velocity drops but the LTE is not attained because of the correlation between the characteristic recombination and the hydrodynamic times.

1. Introduction
Direct current (DC) arc torches have found their applications as plasma sources in plasma chemistry and plasma processing [1], for rocket propulsion systems and for the simulation of the reentry conditions [2]. In the paper we consider the plasma flow downstream of the DC arc, formed in the convergent straight nozzle (Fig. 1). The torch operates with the central tungsten cathode and a water-cooled annular copper anode. The plasma gas is injected into the gap between the two electrodes. As the gas passes through the anode nozzle, it is heated, partially ionized and at forms a high velocity jet. Due to the fact that in this torch the plasma is formed and accelerated at the same time, it deviates from the local thermal-dynamic equilibrium (LTE). However most of the numerical studies of this plasma are made under the assumption of LTE everywhere in the flow except for the cathode shears [3]. In the paper by means of a two-temperature model we study the jet flow downstream of the nozzle using the results of the emission spectroscopy measurements to assume the boundary conditions at the chamber inlet. The considered jets are underexpanded, that is, if these jets the ambient chamber pressure $P_{ch}$ is lower than the inlet jet static pressure $P_{in}$. This equilibrium of the static pressure with the ambient pressure is performed in the jet core by means of an extremely complex nonuniform structure of expansion and compression zones. In fact, in the expansion regions the static pressure can be substantially lower than in the discharge region, which causes the increase of the deviation from LTE in the jet comparing with the discharge region. The nonequilibrium effects are high nonuniformity make the optical emission spectroscopy complicated requiring the implementation of Abel inversion procedure and a special care in Boltzmann plot data interpretation. As far as the modeling is concerned, the deviations from LTE prevents one from using the standard computational fluid dynamics codes. Even though only the pure argon jet is considered in the present study, the attention given to the nonequilibrium effects modeling makes the results be useful to predict the possible gas dynamics and chemical kinetics mechanisms in supersonic DC jets. The range of applications includes plasma assisted chemical vapor deposition of diamonds, silicon and polymer thin films.
2. Model

We assume that the chamber pressure is high enough to describe plasma flow by continuum fluid dynamic equations. We incorporated a two-temperature model [3] into an originally one-temperature computational fluid dynamic code FLUENT (FLUENT is a registered trademark of FLUENT Inc. Cererra Resource Park, 10 Cavendish Court, Lebanon, NH 03766 USA). We choose a coupled technique solving the Navier-Stokes equations written in matrix form. The jet is assumed to be turbulent, a RNG k-ε model was used for turbulence simulation. The electron mass fraction ε, transport equation, the electron temperature T_e transport equation as well as the two-temperature modification of the state equation, thermodynamic and transport properties are programmed and added to the FLUENT code. The details of the model can be found in Ref.[4]. We study the stationary axisymmetric jet of argon using the computational area shown in Fig.2. The boundary conditions are as follows: the electron temperature about 11000K is supposed to be out of equilibrium with the argon heavy particle temperature 7500K, the equilibrium value of the electron number density calculated at the electron temperature is assigned at the chamber inlet with the diameter 50mm. The computational area radius is R = 4 cm, its length is L_e = 0.4 m. Our analysis is limited to the jets exhausting from the convergent-straight nozzle (with Mach number M=1), having the static pressure at the inlet $P_{in}$=28kPa, while the chamber pressure is $P_{ch}$ = 635 kPa and the argon flow rate is 5slm. The supplied electrical power in the corresponding experiments is about 20kW. In the model we use a constant wall boundary condition for the heavy particle temperature $T_e = 300K$, a zero normal derivative condition for the electron temperature and non-slip velocity boundary condition. The computations start on a rather coarse nonstructural grid using first order upwind scheme. Then the grid is adapted and the calculations proceed using second order scheme until the residuals of all the equations become less than $10^{-8}$.

3. Optical emission spectroscopy measurements

The spectroscopic measurements are performed using 1m Jobin Yvon monochromator with a 1200 grooves/mm grating. Plasma jet is projected into the monochromator entrance slit. The radial profiles of argon excitation temperature and electron number density are obtained by interpreting the measurements performed in the several sections with different distance from the nozzle exit. The average excitation temperature is obtained from Boltzmann plot of several isolated argon atom spectral lines that represent three distinctive groups. In this method the straightforward interpretation of spectroscopy data is possible only if the populations of the excited states obey the Boltzmann distribution. In that case the expression for the emission coefficient $a_{nm}$ corresponding to the transition between the atomic levels m and n can be written in the form that represents a straight line with a slope proportional to the excitation temperature.
In the absence of equilibrium the linear regression of experimental points in the Boltzmann plot evaluates only the average excitation temperature that can deviate substantially both from the electron and heavy particle temperatures. To determine the electron number density the continuum emissivity is used.

4. Effect of the chamber pressure on the gas dynamic structure of the jet

The structure of the DC jets for chamber pressure values $P_{ch}=6.5$, 13, 26 and 39kPa resulted from the simulations is shown in Fig.2 representing the temperature and the Mach number profiles and Fig.3a,b) showing the axial static pressure, temperature, velocity, Mach number and electron number density. We can see that when the chamber pressure is low (6.5kPa and 13kPa) strongly underexpanded jets are formed. The first expansion zone in these jets ends

![Image](image.png)

with a Mach disk followed by small subsonic region after which the jet again becomes supersonic. For the higher pressure values, the initial core region of the jet becomes shorter and it changes to a moderately underexpanded structure with a sequence of alternating oblique expansion and compression zones. The expansions correspond to the regions where thermal energy transfers into kinetic energy. They are followed by the compressions where kinetic energy transfers into thermal energy. Physically, these oscillations of static pressure along the jet axis are similar to the damped oscillations of the pendulum. Therefore, the curve showing the axial static pressure in the moderately underexpanded jet can be well reproduced by the solution of the damped oscillation equation

$$ P(x) = P_{ch} + (P_{in} - P_{ch}) \exp (-\sigma x) \cos (\omega x) $$

Analyzing Fig.3 showing the axial static pressure for chamber pressure values $P_{ch}=26$ and 39kPa, one can find the approximate values for the damping coefficient $\sigma$ and the frequency $\omega$ of the oscillations

$$ \sigma = 4 \lambda_{\text{jet}}, \quad \omega = 8 \pi \lambda_{\text{jet}} \left(1 - \frac{1}{\text{Re}_{\text{jet}}}ight) $$

where the following observation are taken into account: there are four full periods in the core region in both jets; the length of the jet core $\lambda_{\text{jet}}$ is proportional to the pressure ratio $\alpha$. Fig.3 show that the higher the chamber pressure the more oblique the waves are. In addition, the higher the chamber pressure the closer to the inlet the first shock zone position is. Due to the supersonic nature of the first expansion zone, the plasma starts to expand without

![Image](image.png)

Fig.4. Axial static pressure predicted by the model and formula (1)
having the information about the downstream conditions. That is why the very beginning of the expansion is similar for all the considered chamber pressure values. The higher the chamber pressure is the earlier the plasma shocks and the expansion turns to the compression. The results of the modeling on the first shock positions are in good agreement with the emission spectroscopy measurements data (Fig. 3). Note that the extrapolation of these dependencies to much lower chamber pressure cases leads to the erroneous conclusions. In fact, if one allows the chamber pressure to drop substantially, than starting from its particular value the shocks begin to thicken, they can move closer to the inlet. This regime can be called a transitional regime. The continuation of the chamber pressure reduction results in the transfer to the scattering regime, where any shocks disappear. To categorize these regimes a rarefaction parameter $\xi = \frac{d_0(P_e, P_i)}{P_\infty}$ can be used [3], where $d_0$ is the chamber inlet diameter, $P_e$ is the stagnation pressure at chamber inlet, $P_\infty$ is the heavy particle temperature at the chamber inlet. In our study $\xi < 5$ dynes-sec/m$^2$ and the results are applicable for the study of the continuous regime only.

5. Effect of the chamber pressure on nonequilibrium effects in the jet

Fig. 5 a and b) compare the heavy particle temperature and electron temperature along the jet axis for the two chamber pressure values. In the expansion zones, the temperature of the electrons heated by the three particle recombination drops less than the heavy particle temperature does (Fig. 5). In the shock region, however, the kinetic energy transfers to the thermal energy of the heavy particles, so that their temperature rises sharply. The electrons also experience shock due to the forces establishing electroneutrality and due to the energy transfer from the heavy particles. We can see 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. The mentioned facts result in the more pronounced maxima and minima of axial heavy particle temperature than the corresponding electron temperature, so that even zones with higher heavy particle temperature than electron temperature can occur. Figs 6 show the comparison of the axial electron number density predicted by the model with the results of the emission spectroscopy measurements and with its equilibrium and frozen values. Along the jet axis the values of the electron number density become higher than the equilibrium values $n_e$, calculated by the Saha equation at the electron temperature.
Furthermore, in the jet core, these values are very close to the frozen values \( n_e \) obtained when the recombination ionization reaction is excluded from the consideration. The modeling results on the electron number density along the jet agree well with the emission spectroscopy data. The further from the axis the better this agreement is. Some disagreement between modeling and experimental data can be explained by the fact that the flat profile for the electron number density is assumed in the model while in reality it has an axial maximum. Downstream of the core due to the recombination, the electron number density is lower than the frozen one. In this region, both the degrees of the ionization and thermal nonequilibrium increase. This observation can be explained by the following phenomena: First, the three-particle recombination heats the electrons. Due to the electron heating by the recombination, the deviation from thermal equilibrium does not decrease downstream of the jet core. Moreover, this recombination is not sufficient to reach the Saha equilibrium. It can seem unrealistic, because of the fact that the velocity drops essentially downstream of the jet core. Nevertheless, as it is shown in Ref. [6], even a low-velocity flow can be far from equilibrium. Not only velocity drops downstream of the core, but also the temperatures. The temperature changes cause the changes of the equilibrium constant for the recombination-ionization reaction. In other words, there is a correlation between the gas dynamic characteristic time and the characteristic time of the chemical reaction. Because of this correlation, the equilibrium condition \( \tau_\text{gas} \approx \tau_\text{chem} \) (where \( \tau_\text{gas} \) is the gas dynamic characteristic time and \( \tau_\text{chem} \) is the recombination characteristic time) is not satisfied and the plasma deviates from the ionization equilibrium. Moreover, in the core region along the axis, the following condition of freezing is satisfied: \( D_\text{ex} < D_\text{ion} \approx 1 \), where \( D \) is the Damköhler number, \( D_\text{ex} \) is the electron recombination potential, \( \tau_\text{ex} \) is the ionization degree. Fig. 11 a) and b) show predicted by the model heavy particle and electron temperature radial profiles for the chamber pressures considered. 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 wave. The electron number density radial profiles (Fig. 7) obtained from the modeling also reflect the position of the compression, expansion zones and barrel shock waves. The emission spectroscopy data on excitation temperature and electron number density, however, do not have off-axis maximum in the expansion regions. The excitation temperature stays high along the jet, it does not drop in the expansion regions. These facts can be explained by the averaging of the excitation temperature over all the levels. The excitation temperature defined in this way in recombining plasma does not correspond to the
The absence of the axial excitation temperature minimums in the expansion regions can also be explained by the application of Abel transformation. The weak point of this procedure is that the error of the calculated local quantity increases towards the centerline of the plasma column. This effect is negligible if the emission intensity profile has axial peak, however it becomes essential in case of well pronounced intensive axial minimum as can be observed in the expansion zones. In general, the results confirm that in the jet core the deviation from LTE is maximal in low pressure jets, the higher the pressure the closer to equilibrium the plasma is there.

5. Summary and conclusions

Numerical modeling and optical emission spectroscopy measurements are performed to understand the effect of chamber pressure on the structure and chemical kinetics mechanisms in the supersonic DC plasma. Argon plasmas exhausting into a chamber with different pressure values are compared. The results show that when the chamber pressure is low a strongly underexpanded jet with a Mach disk is formed. For the higher pressure values, the initial core region of the jet changes to a moderately underexpanded structure with alternating oblique expansion and compression zones. The structure of these zones in moderately underexpanded jet can be described by the damped oscillation equation. The shock zone position predicted by the modeling are in good agreement with the emission spectroscopy data. Along the jet core, the deviation from local thermodynamic equilibrium is more pronounced in the expansion regions and less pronounced in the compression zones. The axial electron number density is higher than its equilibrium values and is close to the frozen values in the jet core. Downstream of the core plasma recombines and the recombination heats the electrons. This recombination, however, is not sufficient to reach predicted by Saha equation electron number density there because of the correlation between the gas dynamic and recombination time scales.

Acknowledgements: Financial support by the National Sciences and Engineering Research Council of Canada and the Ministry of Education of the Province of Quebec is gratefully acknowledged.

References
4. S. E. Sobczak, M. I. Boudlos Supersonic induction jet modeling

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Numerical Simulation of the Functionalization for the Radio Frequency Inductively Coupled Plasma

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Abstract

The functions of the plasma flow such as electrical conduction, thermal conduction and chemical reaction can be enhanced by seeding vaporized alkali metal with low ionization potential. In the present study, numerical simulation is conducted for the radio frequency inductively coupled plasma functionalized by seeding a small amount of alkali metal vapor and the effects of seeding gas injection flow rate and applied frequency on the plasma characteristics are clarified.

1. Introduction

Plasma can be regarded as one of multifunctional fluids [1] since it has high energy density, chemical reactivity and variable transport properties such as electrical conductivity. Furthermore, plasma can be easily controlled by applying an electromagnetic field. In the various kinds of plasma, a radio frequency inductively coupled plasma (RF-ICP) has advantages of large volume, clean high energy and chemical reactivity since it is produced without electrodes. Besides the chemical reaction time is comparatively long due to the low plasma velocity. It has, therefore, been extensively used in reactive plasma spraying, synthesis of ultrafine powders and decomposition of hazardous substances such as freon and dioxin. In these industrial applications, it is very important to enhance the functions of plasma and control them precisely. It is well known that seeding a small amount of vaporized alkali metal with low ionization potential into plasma is one of effective methods for the functionalization of plasma [2][3]. Plasma controllability will also be enhanced with increase in the electrical and thermal conductivities by the functionalization. There have, however, been a few detailed papers as to functionalization by seeding [4].

In the present study, it is numerically investigated how its functions and thermostream characteristics of the RF inductively coupled argon plasma are influenced and enhanced by seeding vapor potassium or cesium at atmospheric pressure taking into account the ionization, recombination and diffusion of plasma species. The effects of gas injection flow rate and applied frequency on the RF argon plasma with seeding are also clarified.

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