Investigation of the gas flow effect on an atmospheric pressure RF plasma torch

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Abstract. A cool atmospheric pressure non-thermal capacitively-coupled RF discharge is studied. It is created between two parallel electrodes – a powered one supplied by 13.56 MHz, and a grounded one. The feed gas argon flows via holes between the electrodes where it is ionized. The plasma torch is studied by means of a time dependent two-dimensional fluid model. A simplified kinetic scheme with four active species is considered, namely argon excited atoms (Ar*), atomic (Ar+) ions, molecular (Ar₂+) ions and electrons (e). The plasma dynamics in the space between the electrodes as well as in the extended region behind the grounded electrode is studied. The effect of the gas flow on the plasma is examined. Constriction of the plasma is induced by the field sustaining the discharge due to the sieve-like structure of the electrodes. As a result of the stationary gas flow the filaments extend beyond the electrodes ensuring a flow of active species in the afterglow.

1. Introduction

Atmospheric pressure non-thermal discharges continue to receive formidable interest during the last years. That is because they are easy to operate which consequently leads to numerous practical implementations. Particular interest attracts the radio-frequency atmospheric plasma jet. It is a device that features low gas temperature, that is versatile in selecting the gas chemistry, and adjustable in power input and gas flow rate. These advantages result in various applications such as surface treatment of polymers, surface modification, film coating and etching, developing of nanotechnologies, sterilization, and environmental applications [1–6]. Although there exists advanced experimental and modelling studies a complete picture of the physical and chemical processes determining the performance of this discharge is not obtained, yet. Most of the studies are on the gas breakdown [7–9], the transitions of the discharge from α to γ mode [10,11] and the concentration of radicals in the afterglow [12,13].

The present study focuses on a non-thermal, non-equilibrium, RF argon discharge at atmospheric pressure. It aims to give a better knowledge on the processes and the discharge characteristics inside
the plasma source and thus to help to get an insight into the phenomena within the region which is hardly accessible experimentally. It also provides information on the species densities and their spatial distributions in the effluent region. This is of crucial importance to improve laboratory and industrial applications. The model is used to study the dependence of the properties of the argon plasma on the geometry, the dimensions and the effect of the flow on the discharge composition and kinetics.

2. Model

A hybrid model is constructed that combines the chemical kinetics obtained with a time-dependent, two-dimensional fluid model with the flow provided by a Navier-Stokes solver.

The PLASIMO MD2D sub-model is used to carry out the time-dependent simulations [14]. It features a drift-diffusion description for the charged particles and accounts for the convective motion of the species. The calculation of the convection velocity is carried out by another PLASIMO sub-model. In such a way, a complete description of the constituting particles, the processes, the flow pattern and the discharge characteristics can be accomplished.

2.1. Plasma chemical kinetics

The electron energy distribution function (EEDF) is obtained by solving the Boltzmann equation using the Bolsig+ solver [15]. With the EEDF the electron transport coefficients: mobility \( \mu_e \) and diffusivity \( D_e \) and electron induced reaction rates \( k_i \) are calculated. A simplified kinetic scheme with four active species is considered consisting of argon excited atoms (Ar\(^*\)), atomic (Ar\(^+\)) ions, molecular (Ar\(_2^+\)) ions and electrons (e). The energy diagram of argon is presented in figure 1. The considered excited atoms are those of the 4s system treated as a lumped block of levels. This is justified by the high exchange probability of the population of the levels in this level block. The following elementary processes are included in the model: elastic scattering, excitation, direct, stepwise, and Penning ionization, dissociative recombination, molecular ion formation, diffusion and radiation losses with accounting for trapping. The elementary processes taken into account with the corresponding references for the cross sections and rate constants data are listed in Table I. For the ions, the local field approximation was used. The values for the mobility of atomic and molecular ions and excited atoms were adopted from references [16,17]. The diffusion coefficients of all the species were calculated from their mobilities using the Einstein relation.

![Figure 1. Simplified energy level diagram of the argon atom](image)

**Table I.** Elementary processes included into the model and corresponding references for the cross sections and rate constants data.

| Process                      | Notation                                      | Reference |
|------------------------------|-----------------------------------------------|-----------|
| Elastic scattering           | e + Ar \(\rightarrow\) e + Ar                 | [18]      |
| Excitation                   | e + Ar \(\rightarrow\) e + Ar\(^*\)          | [19]      |
| Direct ionization            | e + Ar \(\rightarrow\) e + e + Ar\(^+\)       | [20]      |
| Stepwise ionization          | e + Ar\(^*\) \(\rightarrow\) e + e + Ar\(^+\) | [21]      |
| Radiation                    | Ar\(^*\) \(\rightarrow\) Ar + h\(\nu\)        | [21]      |
| Penning ionization           | Ar\(^*\) + Ar\(^*\) \(\rightarrow\) Ar\(^+\) + e + Ar | [22] |
| Molecular ion formation      | Ar + Ar\(^+\) \(\rightarrow\) Ar + Ar\(_2^+\)  | [21,22]  |
| Dissociative recombination   | Ar\(_2^+\) + e \(\rightarrow\) Ar + Ar\(^*\)  | [23]      |
2.2 Plasma dynamics
The plasma dynamics part couples the transport of the relevant species (electrons, ions, excited atoms), to the production and destruction balances of these species, the electric field and the electron energy. The governing fluid equations are:
- the continuity equations for the excited atoms, ions and electrons;
- the transport equations of the considered particles solved in a drift–diffusion approximation taking a convective velocity field \( v_{\text{conv}} \) into account;
- the energy equation for the electrons;
- the Poisson equation for the electric field.

The coupled differential equations are solved by the so-called “modified strongly implicit method” [24] using an extra stabilization method [25].

2.3 Fluid dynamics
To calculate the convection velocity field \( v_{\text{conv}} \), a fluid dynamics module is used to calculate the mass-averaged velocity field. For a given mass density field \( \rho \) it calculates the velocity \( v \) and the pressure field \( p \). These quantities are determined from the combined solution of the:
- mass continuity equation and the
- momentum balance equation

3. The experimental set-up
Figure 2 gives a photo and a scheme of the atmospheric pressure plasma jet that was considered in this study. It consists of two parallel perforated electrodes separated by a variable gap of 1 to 2 mm. One of the electrodes is driven by a RF between 30 and 100 W at 13.56 MHz, while the other electrode is grounded. Feed gas made of argon (or helium) and up to 0.1 vol% of other gases, flows between the electrodes at a rate of 20 up to 60 L/min. The gas emerges from the holes and impinges on a substrate placed 2 to 15 mm downstream, where cleaning or etching of surface material takes place [12].

4 Results
4.1 Steady discharge
We model the atmospheric pressure plasma torch using a periodic 2D Cartesian geometry in which the hole-pattern shown in figure 2 is seen as a repetition of identical cells. Such a unit cell is depicted in figure 3. It includes half of a hole and half of an electrode between the two holes. The powered electrode is on the left, the plasma bulk in the middle and a similar electrode pattern on the right.

**Figure 2.** Photo and schematic representation of the atmospheric pressure plasma jet

**Figure 3.** Schematic of a unit cell from the perforated electrodes geometry.
grounded electrode is displaced in vertical direction. On the top and the bottom of the unit cell mir rror boundary conditions are applied. In figure 3, \( l_p \) denotes the distance between the electrodes, \( w_h/2 \) the half-width of the hole, \( w_e/2 \) the half-width of the electrode and \( d \) the thickness of the electrode. The calculations are performed for a unit cell with \( d = 1 \) mm, \( w_h = 1 \) mm, \( w_e = 2 \) mm and \( l_p = 2 \) mm. For the gas temperature 350 K is taken [13]. The applied voltage is – 300 V.

Searching for an optimal configuration in which we have maximum uniformity and homogeneity of the plasma, we examined first the effect of the set-up dimensions on the discharge characteristics for the non-convective case; i.e. \( v_{\text{conv}} = 0 \). The spatial distributions of plasma potential, volume charge and electron density are shown in figure 4. These are obtained by averaging over one period of the electric cycle. As shown in figure 4(a), the electric potential has a large gradient close to the electrodes. The sheath regions formed around the electrodes are observable in figure 4(b) as domains with high volume charge. This charge extends also inside the holes. As a result a zigzag particle density pattern and higher density structures perpendicular to the plane of the electrodes and stretched inside the holes are formed (figure 4(c)).

The effect of the distance between the electrodes on the discharge properties is shown in figure 5 where the plasma densities for three \( l_p \)-values are plotted. The snapshots are taken at the moment in the RF cycle when a maximum of the negative potential (at the left electrode) is reached. It can be seen in figure 5(a) that for smaller gap-spacing the plasma density is higher but also more unevenly distributed. With the increase of the gap (see also figure 4(c)) the electron density within the so called filaments decreases the plasma becomes more homogeneous. The optimal gap spacing is found to be \( l_p = 2.5 \) mm. Higher distances result in a drop of the density of the electrons and thus of all the other species in the discharge. This is shown in figure 5(c).

Another topic of interest is the influence of the inequality between the aspect ratio AR of the left and right electrode. This AR is the ratio of the width of the hole and the electrode, \( w_h/w_e \). Figure 6 shows what happens with the electron density and the volume charge distribution if we keep for the left electrode \( w_h/w_e \) fixed to 1/2 while for the right electrode for the ratios \( w_h/w_e \) 0.75/2.25 and

**Figure 4.** Spatial distribution of the electric field in V (a), volume charge in nC (b) and electron density in m\(^{-3}\) (c) between perforated electrodes. The set-up dimensions are: \( l_p = 2 \) mm, \( d = 1 \) mm, \( w_h/2 = 0.5 \) mm, \( w_e/2 = 1 \) mm.

**Figure 5.** Electron density distribution in m\(^{-3}\) for a distance between the electrodes: \( 1 \) mm – (a), \( 2.5 \) mm – (b) and \( 3 \) mm – (c), \( d = 1 \) mm, \( w_h/2 = 0.5 \) mm, \( w_e/2 = 1 \) mm.
1.25/1.75 are applied. The variation of the AR results in changes of discharge species density and charge distribution within the sheath regions. The plasma density (mainly within the structures with higher density) increases as the width of the holes in the right electrode decreases. The positive charge is concentrated within these holes and in the sheath region of the electrode against them (the left one). Wider holes result in lower concentration of positive charge within the holes and within the sheath of the corresponding electrode.

4.2. The effect of gas flow

In order to study the role of the gas flow in the atmospheric pressure RF discharge, calculations of the velocity field distribution have been done. The calculations are performed for unit cell with the same dimensions as mentioned in Sec. 4.1. The fluid viscosity of argon at about 350 K is computed to be $\mu = 3.5 \times 10^{-5}$ Pa. For calculating the initial velocities corresponding to different gas flow vessel radius $r = 1.75$ cm is used.

The profile obtained for inlet gas flows of 30 l/min is shown as representative of the velocity field distribution in figure 7. The velocity field has its highest absolute values at the entrances of the holes especially at the edges, within the holes and in the plasma bulk close to the grounded electrode. A backward flow toward the powered electrode is also formed in the bulk.

The effect of the flow on particle distributions is shown in figure 8. The electron density profiles shown there correspond to gas supply 30 and 60 l/min. The gas flow modifies the distributions. It leads to a reduction of the electron density at the inlet holes and an increase of the density at the outlet. For comparison see also figure 4(c). It also increases the concentration of reactive species at some distance behind the discharge.

Figure 6. Distribution of electron density in m$^{-3}$ (left) and volume charge in nC (right) for a gap spacing of 2 mm and different ratios $w_h/w_e$ of the hole/electrode width on the right side: top 0.75/2.25 and bottom 1.25/1.75.

Figure 7. Spatial profiles of velocity field distribution at gas flow supply: 30 l/min.
5. Conclusion
A model of an RF capacitively coupled discharge has been developed and used to obtain key plasma parameters and to gain insight into the physical processes that determine its behavior. The spatial distributions of plasma density, potential and volume charge are obtained and the effect of the geometry on these characteristics is investigated. The role of the gas flow is studied and it is found that it changes the distributions of the plasma particles between the electrodes and thus affects the plasma chemistry kinetics. The flow is found to be an effective transport mechanism to bring radicals downstream to the region where the plasma-surface interaction takes place.

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