The main properties of microwave argon plasma at atmospheric pressure

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Abstract. Plasma torch sustained by surface wave at atmospheric pressure is theoretically studied by means of 1D model. A steady-state Boltzmann equation in an effective field approximation coupled with a collisional-radiative model for high-pressure argon discharge is numerically solved together with Maxwell’s equations for an azimuthally symmetric TM surface wave. The axial dependences of the electrons, excited atoms, atomic and molecular ions densities as well as the electron temperature, the mean power per electron and the effective electron-neutral collision frequency are determined. A strong dependence of the plasma properties on the discharge conditions and the gas temperature is obtained.

1. Introduction
Plasma production at atmospheric pressure is a field in plasma physics which develops headlong because of the easy way for operating with these type of plasmas and thus for numerous practical applications. Among the atmospheric pressure plasmas the ones produced and sustained by means of microwaves draw a particular interest. Because the electrodes or excitation structures, serving to impose the electric field that maintains the plasma, do not need to be located within the plasma vessel, this eliminates electrode corrosion problems, reduces gas contamination, and substantially lowers the production costs. The specified advantages results in wide opportunities for applications in industry [1], materials treatment [2,3], environmental protection [4–6], sterilization [7], biomedical treatment [8] and etc.

This paper deals with the numerical modeling of surface wave sustained plasmas at atmospheric pressure which are a particular class of microwave discharges. They are distinguished for independence of the plasma properties on the type of the wave launcher which allows studying of various configurations. The devices generating the plasma are simple, efficient and easy to operate with. The surface wave technique allows for an exceptionally flexible control of the plasma parameters and of its dimensions, in a wide range of wave frequencies and gas pressures. The created plasma is over-dense plasma \( n > n_{\text{cutoff}} = m_0 \omega_c^2 / 4 \pi e^2 \), which characterizes with both axial and radial inhomogeneity.

The organization of the paper is as follows: section 2 depicts the physical pattern accounted in the model. In section 3 a description of the self-consistent model is given. Section 3.1 presents the main electrodynamic equations as well as the boundary conditions used for obtaining the wave propagation characteristics. In section 3.2 the processes occurring in the plasma, the atomic and molecular energy levels, and the equations considered in order to build a collisional-radiative model of high pressure...
argon discharge are described. Section 3.3 looks into the self-consistent connection between the former two parts of the modelling. Finally, section 4 shows the results obtained by means of applying the complete model.

2. Physical pattern
The discharge in a surface wave plasma source is sustained by electromagnetic wave. Propagating along the interface between the plasma and the dielectric tube the field of the wave penetrates in the plasma, therefore the name of the wave comes up. The propagating surface electromagnetic wave creates the discharge and at the same time the discharge is a medium for the wave propagation. The wave gradually transfers its power to the plasma, more precise to the electrons. The energy the electrons receive from the wave is expended on different elementary processes which ensure the creation and sustaining the plasma. When the absorbed power is no longer enough for compensation the losses of radiation, diffusion and recombination toward the walls of the tube the discharge quenches.

3. Self-consistent model
The model, as it was mentioned above, consists of two parts: electrodynamic and kinetic ones, and a link which allows deriving self-consistently all discharge and wave characteristics.

3.1. Electrodynamic part
The model is created under the following assumptions:

▫ the plasma is in a steady state
▫ the electromagnetic wave maintaining the plasma is an azimuthally symmetric (m = 0) HF TM surface wave with components \( \mathbf{E}_{hf} = (E_r, 0, E_z) \) and \( \mathbf{B}_{hf} = (0, B_{\phi}, 0) \)
▫ the plasma density \( n \), the wave number \( k \) and the wave amplitude are slowly varying functions of the axial coordinate \( z \)
▫ this is numerical one-dimensional axial model where the radially averaged plasma density is used, \( n = \frac{2}{R^2} \int_0^R r \, dr \, n(r) \)

The electrodynamic part of the model is based on Maxwell’s equation for an azimuthally symmetric TM surface wave with angular frequency \( \omega \) propagating along a plasma cylinder with radius \( R \) surrounded by vacuum. Maxwell’s equations give the wave equations for the radial, azimuthal and axial components of the wave. The one for the \( E_z \) component of the wave electric field in cylindrical coordinates is:

\[
\frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial E_z}{\partial r} \right) + \frac{\partial^2 E_z}{\partial z^2} + \left( \frac{\partial}{\partial z} \ln \varepsilon \right) \frac{\partial}{\partial z} E_z + \frac{\omega_p^2}{c^2} \varepsilon E_z = 0
\]  

(1)

Here \( \varepsilon \) is the plasma permittivity of the medium which for vacuum is \( \varepsilon_v = 1 \) and for plasma:

\[
\varepsilon_p = 1 - \frac{\omega_p^2}{\omega(\omega + i \nu_{eff})} = 1 - \frac{\omega_p^2}{\omega^2} \left( 1 + \frac{\nu_{eff}^2}{\omega^2} \right)^{-1} + i \frac{\nu_{eff} \omega_p^2}{\omega \omega^2} \left( 1 + \frac{\nu_{eff}^2}{\omega^2} \right)^{-1}
\]  

(2)

The wave equations are solved for the case \( \nu_{eff} \neq 0 \) without any assumptions although in our case \( \nu_{eff} \geq \omega \), \( \omega_p \) is the usual electron plasma angular frequency and \( \nu_{eff} \) is the effective electron–neutral collision frequency for momentum transfer.

Similar expressions are obtained for the radial electric field component \( E_r \) and for the azimuthal magnetic field component \( B_{\phi} \). With assumption for weak axial density inhomogeneity we seek the solutions of the wave equations in the form:
where and are cylindrical functions. The solutions we are looking for are combinations of the modified Bessel’s functions zeroth and first order.

Using the boundary condition at \( r = R \) we obtain the local wave dispersion relation:

\[
\frac{e_p}{a_p} I_1(a_p) + \frac{1}{a_v} K_1(a_v) = 0
\]

with \( a_p^2 = \tilde{k}^2 - \sigma^2 e_p \) and \( a_v^2 = \tilde{k}^2 - \sigma^2 \), where \( \tilde{k} = k_R + i k_i R \) is the dimensionless complex wave number with \( k_R \) the propagation coefficient and \( k_i \) the attenuation coefficient, \( \sigma = \omega R/c \) and \( I_0, I_1, K_0, K_1 \) are the modified Bessel functions of zeroth and first order.

The second main equation of the electrodynamic part of the model is the wave energy balance equation given in the form:

\[
\frac{d}{dz} S = -Q
\]

where \( S \) is the energy flux along the plasma column and \( Q \) is the power absorbed by the electrons at unit column length. The energy flux is the sum of the axial components of Poynting’s vectors averaged over the wave period and integrated over the plane normal to the plasma column:

\[
S(z) = 2\pi \int_0^r d\rho \rho S^\rho_z + 2\pi \int_1^\infty d\rho \rho S^\rho_v
\]

which in our case reads:

\[
S = \frac{1}{4} \omega R^4 E^2 \text{Re} \left[ \frac{\tilde{k} e_p}{a_p^2 - a_p^2} \left( \frac{1}{a_p} I_1(a_p) - \frac{1}{a_p} I_1(a_p^*) \right) \right] + \frac{1}{4} \omega R^4 E^2 \text{Re} \left[ \frac{\tilde{k}}{a_v^2 - a_v^2} \left( \frac{1}{a_v} K_1(a_v^*) - \frac{1}{a_v} K_1(a_v) \right) \right]
\]

The wave power dissipated per unit length is given by the relation:

\[
Q(z) = 2\pi \int_0^R drr \langle j, E \rangle = \frac{\omega}{4} \text{Im}(e_p) R^2 E^2 \int_0^1 d\rho \left( |F^\rho_r|^2 + |F^\rho_z|^2 \right)
\]

and is equal to:

\[
Q = \frac{\omega}{4} \text{Im}(e_p) R^2 E^2 \left( \frac{1}{a_p^2 - a_p^2} \left( \frac{1}{a_p} I_1(a_p) - \frac{1}{a_p} I_1(a_p^*) \right) \right) + \frac{1}{a_v^2 - a_v^2} \left( \frac{1}{a_v^*} I_1(a_v^*) - \frac{1}{a_v^*} I_1(a_v) \right)
\]

3.2. Kinetic part

The detailed modelling of any kind of plasma requires the knowledge of the discharge processes. In this study we consider argon plasma. In general the kinetic description consists of:

- electron kinetics
The electron kinetics includes calculation of the electron energy distribution function (EEDF), which determines the electron transport parameters, the rates of elementary processes, and several other important quantities as $\nu_{\text{eff}}$ – the effective electron–neutral collision frequency for momentum transfer, a key parameter for the modeling of MW discharge maintained of surface wave. The EEDF is derived from the electron Boltzmann equation, solved in our case using the two-term Legendre polynomials expansion. The electron energy balance equation giving the energy for sustaining an electron-ion pair in the discharge $\theta$, which is another key parameter:

$$\theta \equiv \theta_{\text{elast}} + \theta_{\text{exc}} + \theta_{\text{ion}} + \theta_{\text{diff}} + \theta_{\text{rec}} - \theta_{\text{Pen}}$$

as well as the electron particle balance equation are considered here. The superscripts denote the energy expanded on different processes, respectively elastic, inelastic collisions, direct and stepwise ionization, diffusion, recombination and Penning ionization. In additions of these processes the associative ionization, dissociative and three-body recombination, conversion to molecular ions and radiative transitions are taken into account.

The heavy particles kinetics involves calculation of the particle balance equations for the excited atoms as well as for the ions. The species considered in our model are Ar, Ar(4s), Ar(4p), Ar(3d), Ar(5s), Ar(5p), Ar(4d), Ar(6s), Ar$^+$, Ar$^{2+}$, Ar$^{3+}$ and Ar$^{3+}$. The calculations are done assuming all excited states as blocks of levels with effective energy corresponding to the energy of the center. The heavy particle kinetics coupled with the electron kinetics allows us to determine the atomic and molecular ion number densities, the populations of the excited states as well as $\theta$ and $\nu_{\text{eff}}$ as a function of the electron number density.

### 3.3. Self-consistent link

The self-consistent link between the electrodynamic and kinetic parts of the model is the balance of the wave and electron energy. On one hand $Q$ is the wave power absorbed per unit length in the discharge. On the other hand, $Q$ is the power expended by electrons in elastic and inelastic collisions:

$$Q = \pi R^2 n_e \theta$$

So the values of $Q$ derived from the both parts of the model must be equal. The relation between $S$ and $Q$ is given by the wave energy balance equation

$$\frac{d}{dz} S = -Q$$

The local dispersion relation yields the electron density for a given wave number, and the solution of the wave energy balance equation provides the dependence of the electron density on the axial position $n_e(z)$. Having obtained the axial dependence of the electron density, the kinetic part is recalculated again for appropriate axial position. This provides the spatial distribution of the discharge characteristics.

### 4. Results

Numerical calculations have been done for an argon plasma column sustained by electromagnetic wave with frequency $\alpha 2\pi = 2.45$ GHz, wave power: 500 W, and gas...
pressure: 1 atm, varying the gas temperature for fixed plasma radius $R = 3.7$ mm and varying the tube radius for fixed gas temperature $T_g = 3500$K.

In Figure 1 the phase (a) and the attenuation (b) diagrams for the four plasma radii are shown. The parts of the diagrams displayed with solid lines satisfied the requirement the phase coefficient to be higher or equal to the attenuation coefficient $k_r \geq k_i$ and the parts of them presented with dashed lines correspond to $k_r < k_i$. With the decrease of the radius the value of the wave number, for which $k^* \equiv k_r = k_i$ is satisfied, also decreases but the plasma frequency $\omega_p$ (and the plasma density $n_e$) corresponding to this point gets higher.

The numerical calculations in the kinetic part of the model allow us to obtain the dependence of the electron energy distribution function on the plasma radius. In Figure 2 the EEDFs calculated for four different discharge radii ($R = 15$, 7.5, 3.7, and 1.8 mm) are plotted. The solid lines in colours denote the distribution functions for the electron densities close to the launcher corresponding to the fourth radii ($n_e = 4.5 \times 10^{18}$, 1.4 x $10^{19}$, 4.5 x $10^{19}$, 1.9 x $10^{20}$ m$^{-3}$ respectively) and the black dotted line denotes the electron density close to the end of the discharge, which is of the same order $1 \times 10^{19}$ m$^{-3}$ for the fourth cases. It can be seen that with the reduction of the radius the electron density increases and the EEDF gets closer to Maxwellian one.

Using the complete self-consistent model the axial distributions of the wave and discharge parameters are obtained. Figure 3 presents the axial distributions of the plasma density (a) and the wave power flux (b) as functions of the discharge dimensions. One can see that the increase of the plasma radius results in a shorter plasma column and lower electron number density (figure 1(a)). For smaller plasma radii the electron density which corresponds to $k^* \equiv k_r = k_i$ is close to the value where the wave power flux $S = 0$ W. With the increase of the radius the attenuation coefficient becomes higher than the propagation one before this point, e.g. before the power flux to be dissipated completely (figure 1(b)). Bearing in mind that the regions of the phase and attenuation diagrams corresponding to $k_r < k_i$ (figure 1, dotted lines) cannot be associated with wave propagation so we can conclude that in this case the end of the discharge should be set by the requirement $k_r = k_i$. 

Figure 2. Electron energy distribution function at different plasma densities corresponding to discharges with different radii.

Figure 3. Axial distribution of the electron density (a) and wave energy flux (b) for different radii of the discharge.
Figure 4 shows the axial dependences of the argon excited states population as well as for the argon dimmer and atomic and molecular ions densities for discharges with radii 7.5 (figure 4 (a) and (b)) and 3.7 mm (figure 4 (c) and (d)).

5. Conclusions
The wave and discharge characteristics are obtained for different discharge conditions. The calculations show that: (i) the stationary discharge in pure Argon at atmospheric pressure can be sustained only in small radius tubes; (ii) the EEDF is non-Maxwellian one in most of the discharge conditions and gets closer to Maxwellian with increasing the plasma density (which is also a result of plasma radius reduction); the electron temperature is always higher than the gas temperature; the plasma density (even at the column end) is much higher than the cut-off limit, \( n_e >> n_{\text{cut-off}} \).

6. Acknowledgements
This work was supported by the Bulgarian National Fund for Scientific Research under Grant F 1401/04 and by the Fund for Scientific Research of the University of Sofia under Grant No 157/2008.

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Figure 4. Axial distributions of the excited atoms (a, c) and the ions (b, d) densities for discharge radii 7.5 and 3.7 mm.