On the Modelling of a Nonequilibrium Spherical Microwave Discharge at Atmospheric Pressure

I. R. Rafatov
Dept. Physics, Middle East Technical University, TR-06531 Ankara, Turkey

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1 Introduction

Microwave discharges at atmospheric pressure attract an increasing interest in recent years. Their study is stimulated by various scientific and technological applications in plasmochemistry, microelectronics, lighting, etc [1–3].

Adaptation of microwave discharges as sources of plasma for these applications needs a knowledge of the dependence of the properties of plasmas on external factors, namely, the electromagnetic field mode and frequency, nature, pressure and flow rate of the gas, geometric size of the discharge setup and the amount of the microwave power delivered to the plasma. Such a knowledge can be provided from the modelling of the processes occurring in the discharge plasma.

Microwave discharge models vary according to their geometry, scheme for microwave power feed, approach in the solution of the gas-dynamic, electrodynamic and thermal problems. The present work concerns with the modelling of a spherical microwave discharge at atmospheric pressure. Experiments show that a spatially localized plasma of spheroidal shape in a microwave discharge at atmospheric pressure can arise and stably exist as a stationary object sustained by a continuous power feed, in free space with microwave beam focusing [4–7], and without focusing of electromagnetic waves in metallic discharge chambers [8–10]. Numerical modelling of such a discharge has been done by many researchers on the assumption of a local thermal equilibrium (LTE) condition [1, 11, 13–15]. LTE approximation leads to significant simplification in the description of plasma. Under the LTE condition, plasma properties at any point within the plasma volume are uniquely defined by the two thermodynamic parameters, the temperature and the pressure, and by the chemical nature of the gas. Electromagnetic field serves as a supplier of the energy to the plasma as a whole. The complicated kinetics of the collisional elementary processes are taken into account in the transport coefficients (thermal conductivity, electric conductivity, diffusion, viscosity, etc.). Such an approach enables to keep the model simple, providing the main characteristics of the discharge plasma. In actual situations, as experiments show [1], the electron temperature in the discharge plasma, especially near the wall of the discharge chamber, is higher than the heavy particle (atom, ion, and molecule) temperature. This is not so noticeable in molecular gases, but in atomic gases the electron temperature can differ from the heavy particle temperature significantly ( [16], p 463). Therefore, a quantitative description needs, at least, adaptation of a two temperature plasma model, which suggests a more

∗Corresponding author: e-mail: rafatov@metu.edu.tr. Phone: +90 312 2103254, Fax: +90 312 2105099
realistic description of the mechanism of energy transfer from the electromagnetic field to heavy particles via electrons. In this approximation, plasma is considered to comprise two main species, the electrons and heavy particles, which are characterized by different temperatures [1, 11, 12, 17, 18].

Thermal non-equilibrium model of a spherical microwave discharge, based on a two-temperature plasma approximation, has been developed in [19]. The authors there took into account a first spherical mode of the applied electromagnetic field, and assumed an ionization equilibrium of the discharge plasma. In reality, recombination processes in plasma do not compensate the ionization processes, that result in ionization non-equilibrium. Model of a spherical discharge, which takes into account a deviation of the plasma from ionization equilibrium, has been considered in [20]. In [15, 21], authors investigated effect of higher modes of the electromagnetic field on the LTE [15] and on the thermal non-equilibrium [21] spherical microwave discharge plasma behavior. The present study is devoted to the further development of the works [20] and [21]. It aims to construct the numerical model of thermal and ionization non-equilibrium spherical discharge, which takes into account higher modes of the incident microwaves, and to investigate their effect on the discharge plasma properties.

The paper is organized as follows. In section 2 we describe the model, we list the assumptions adopted and the governing equations. As an example of the application of the model, section 3 presents results of calculation of the discharge plasma characteristics. Finally, section 4 contains the concluding remarks.

2 Modelling approach

Detailed description of the model and solution method in the case of the first spherical mode have been given previously in [20]. Present study will take into account higher modes of the electromagnetic waves.

It is supposed that the axial symmetric electric discharge is taking place inside a spherical chamber with a dielectric wall (figure 1) and is sustained by the microwave power, \( Q_{app} \), of convergent electromagnetic waves with the field components \( E = E(\theta, 0, 0) e^{i\omega t} \) and \( H = H(0, 0, \phi) e^{i\omega t} \). Other features of the model are as following. Plasma consists of two gas media of electrons and heavy particles (ions and atoms), which are characterized by Maxwell’s distribution for the speeds of particles with kinetic temperatures \( T_e \) and \( T_h \approx T_i \approx T_a \), and Boltzmann’s distribution for the exited levels. Recombination processes in plasma do not compensate the ionization processes. Processes are quasistationary. Plasma is quasineutral. Motion of gas can be neglected. Pressure in close to atmospheric. The energy of the applied electromagnetic field is absorbed basically by the electronic gas, while the heavy particles are heated up as a result of collisions with electrons. Radiation losses are negligible. Finally, dissipated energy is removed from the plasma by heat conductivity to the walls, which are held at a constant temperature. Validity of this approach has been discussed in [19].

On the basis of these assumptions, discharge behavior is expressed by the system of Maxwell’s equations, energy balance equations for electrons and for heavy particles, equations for the structure of the plasma, which consist of continuity equation for electrons, condition of quasineutrality, equations of state and Dalton’s law,

\[
\nabla \times \mathbf{H} = -i\omega \varepsilon_0 c \mathbf{E}, \quad \nabla \times \mathbf{E} = i\omega \mu_0 \mathbf{H}, \quad (1)
\]

and

\[
\nabla \left( v_e n_e \left( \frac{5}{2} kT_e + U_i \right) \right) = \nabla \left( \lambda_e \nabla T_e \right) + Q_{E} - \dot{B}_{ch} \left( T_e - T_h \right), \quad (2)
\]
\n
\begin{equation}
\n\nabla \left( \lambda_e \nabla T_e \right) + B_{eh} \left( T_e - T_h \right) = 0,
\end{equation}

\begin{equation}
\n\nabla \left( \nu_e n_e \right) = n_e, \quad n_e = n_i, \quad p = n_e k T_e + \left( n_i + n_a \right) k T_h.
\end{equation}

Here \( \mu_0 \) and \( \epsilon_0 \) are the permeability and permittivity, respectively, of vacuum, \( p \) is the pressure, \( U_i \) is the ionization energy threshold, \( m \) and \( n \) are the mass and concentration of particles, \( k \) is the Boltzmann’s constant. The subscripts \( h, e, i \) and \( a \) refer to heavy particles, electrons, ions and atoms, respectively. \( Q_E = \frac{1}{2} \sigma \left| E \right|^2 \) expresses the density of the power dissipated within the plasma. \( \left| E \right| = \sqrt{E_r E_r^* + E_\theta E_\theta^*} \) (asterisk denotes a complex conjugate). Velocity of electrons, \( \nu_e \), is formed by the velocities of ambipolar and thermal diffusions, \( \nu_e = \nu_{am} + \nu_{th} \), with \( \nu_{am} = -D_{am} \nabla \ln n_e \) and \( \nu_{th} = -\frac{1}{2} D_{am} \nabla \ln T_e \). Net production of electrons is described by \( \dot{n}_e = K_i n_v n_a - K_r n_e^2 n_i \). Energy transfer from electrons to heavy particles is \( B_{eh} = \frac{3}{4} \delta_e \nu_e n_e k \), where \( \delta_e = 2 m_e / m_i \) and \( \nu_e \) denotes the elastic collision frequency. Complex relative permittivity of the plasma is \( \epsilon_e = 1 - \frac{\sigma_{am}}{\omega \epsilon_0} = \frac{1 - \sigma_{am}}{\omega \epsilon_0} \).

Plasma coefficients (electric conductivity, \( \sigma \), thermal conductivities, \( \lambda_i \) and \( \lambda_e \), ambipolar diffusion coefficient, \( D_{am} \), ionization and recombination coefficients, \( K_i \) and \( K_r \), etc.) are calculated as functions of the temperatures, wave frequency and the pressure by the same way as in [20].

Analysis of the spherical microwave discharge is made difficult by the fact that the vector nature of the electromagnetic field prevents the existence of spherically symmetric waves. In the modelling performed in [13, 14], the authors oriented to experiments [8–10], where a stationary spherical discharge several centimeters in size burnt in the metal discharge chamber \( \sim 10 \text{ cm in size}, \) under the applied power of \( \sim 5 \text{ kW at a wave frequency of 2.45 GHz} \). The electromagnetic field in [13, 14] was described in terms of the spherical harmonic \( E = (E_r(r) \cos \theta, E_\theta(r) \sin \theta, 0) \) and \( H = (0, 0, H_\varphi(r) \sin \theta) \), where \( r, \theta \) and \( \varphi \) are spherical coordinates. (Notice that this field configuration corresponds to our first mode.) In order to compromise between the spherical symmetry of temperature distribution and the angular asymmetry of heat release, the discharge was assumed to be irradiated uniformly from all directions with the ensemble of waves of the same amplitude, and source term in the energy balance equation was averaged over the polar angle \( \theta \). Moreover, the authors noticed that this assumption appears to be somewhat allied to the real situation in experiments [8–10]: as a result of the discharge chamber asymmetry, there must be a large number of scattered waves, propagating at different angles to each other. This approximation corresponds basically to the approach in our previous works [19, 20], where we took into account the first spherical mode of the applied electromagnetic field. Here, we attempt to extend this approach for the case of higher modes, taking into consideration the second and third modes, for thermal and ionization non-equilibrium plasma.

It is known that discharges in microwave fields are characterized by fully nonlinear coupling existing between the electromagnetic field and the discharge plasma: the wave properties depend on the discharge plasma and conversely. Therefore, a self-consistent description of the discharge requires simultaneous solution of the electrodynamic problem and the equations describing the discharge plasma processes. We used basically the same solution method as in [20]. We solved independently the plasma maintenance equations and those of the electrodynamic problem and the equations describing the discharge plasma processes. We then determined the solution by equating the tangential components of the electromagnetic field at the wall of the discharge chamber, \( r = R \). Relation between the coefficients of the incident waves and the input power is

\[
Q_{\text{app}} = \frac{\mu_0 \omega}{k_0} \pi \sum_{n=1}^{\infty} \left( |C_n|^2 \frac{2}{2n+1} n(n+1) \right).
\]

\[
\frac{\nabla \left( \lambda_e \nabla T_e \right) + B_{eh} \left( T_e - T_h \right)}{\nabla \left( \nu_e n_e \right) = n_e, \quad n_e = n_i, \quad p = n_e k T_e + \left( n_i + n_a \right) k T_h.}
\]
Electric field components in terms of function $\chi$ are expressed by the equations

$$E_r = \frac{i}{\omega \epsilon_0 r^2 \sin \theta} \frac{\partial \chi}{\partial \theta}, \quad E_\theta = \frac{i}{\omega \epsilon_0 r^2 \sin \theta} \frac{\partial \chi}{\partial r}.$$ 

Reflection coefficient $\rho_n$, corresponding to the $n$th mode, is defined as a ratio of the respective reflected and input powers, $\rho_n = 1 - Q_{\text{dis}}^{(n)}/Q_{\text{app}}^{(n)} = |B_n|^2/|C_n|^2$.

Regarding the thermal and plasma composition problems, for simplification of the model and maintaining a spherical symmetry, the energy balance equations (2) and (3) and continuity equation for electrons (4) are averaged over the polar angle $\theta$, by multiplying the equations by $\sin \theta$ and integrating them over the interval $[0, \pi]$ on $\theta$. As a result, the source term in heat equation (2) is replaced with a $\langle Q_E \rangle_\theta = \frac{1}{2} \sigma \langle |E|^2 \rangle_\theta$, where $\langle |E|^2 \rangle_\theta = \langle E_r^* E_r \rangle_\theta + \langle E_\theta^* E_\theta \rangle_\theta$, and $\langle \ldots \rangle_\theta$ denotes averaging on angle $\theta$.

### 3 Results

The model has been applied to the microwave discharge sustained in argon at atmospheric pressure. Results presented in figures 2–9 are the discharge plasma and the wave characteristics, namely, $T_e$, $T_h$, $n_e$, $n_a$, and $E$, versus the radius, $R$, of discharge chamber, the mode and frequency, $\omega$, of the incident electromagnetic wave and the amount of microwave power, $Q_{\text{app}}$, delivered to the plasma. Here $E$ denotes the magnitude of the electric field, $|E|$, averaged over the angle $\theta$. The parameter range studied includes microwave power of $1 - 70$ kW, frequency, $\omega$, of $1 - 100 \times 10^9$ rad s$^{-1}$, and discharge chamber radius, $R$, of 0.5 – 20 cm.

![Fig. 2](Image 223x716 to 235x716) Typical result for the radial profiles of (a) heavy particle and electron temperatures, $T$ and $T_e$, (b) electric field magnitude, $E$, (c) electron and (d) atom number densities, $n_e$ and $n_a$. Discharge is sustained separately by the power of the first mode, second mode, or third mode. Dotted line corresponds to the ionization equilibrium model ($n_a = 0$) [21]. $Q_{\text{app}} = 10$ kW, $\omega = 40 \times 10^9$ rad s$^{-1}$, $R = 1$ cm. (Online colour: www.cpp-journal.org).

Figures 2 and 3 show the typical picture for the radial profiles of the electron and ion temperatures, the electric field magnitude and particle number densities, at wave frequency $\omega = 40 \times 10^9$ rad s$^{-1}$ and discharge chamber radius $R = 1$ cm. On the direction from the wall toward the discharge center, heating of electrons by microwave sharply decreases and electrons are rapidly thermalized with heavy particles to attain a single temperature, around 4000 K in the case of the first mode. (Notice that such a monotonicity of the solution profiles breaks down when incident microwave lengths are comparable with the size of the discharge chamber.) Results, presented in figure 2, have been obtained assuming that the discharge was sustained separately by the power of the first mode, second mode, or third mode, of the same power, $Q_{\text{app}} = 10$ kW. Apparently, at this parameter regime, effect of the different modes on the discharge characteristics differs significantly. Heating of the plasma sharply diminish in the case of higher modes. This is a direct consequence of strong increasing of incident wave reflection with increase in the order of mode. Precisely, reflection coefficients and dissipated power amounts, corresponding to
first three modes, are $\rho^{(1)} = 0.930$ and $Q^{(1)}_{\text{dis}} = 0.70$ kW, $\rho^{(2)} = 0.983$ and $Q^{(2)}_{\text{dis}} = 0.17$ kW, and $\rho^{(3)} = 0.998$ and $Q^{(3)}_{\text{dis}} = 0.02$ kW.

Figure 3 shows the radial profiles of the same functions as in figure 2, under the same parameter regime, but obtained under the assumption that the discharge was sustained by the power of the first mode, or combination of first two modes, or combination of first three modes. The power $Q_{\text{app}} = 10$ kW, delivered to the discharge, was divided equally between modes in the case of their combinations. As figure 3 shows, heating of plasma decreases with the increase in the order of mode taken into account.

Such an effect of higher modes on the discharge plasma behavior remains the same, when we vary the amount of power, $Q_{\text{app}}$, delivered to the discharge, keeping the wave frequency, $\omega = 40 \times 10^9$ rad s$^{-1}$, and discharge chamber size, $R = 1$ cm, unchanged. Results are presented in figure 4, for the modes taken into account separately, and in figure 5, for combinations of modes. Higher modes are mostly reflected from the discharge plasma, $\rho^{(3)} \gg \rho^{(2)} \gg \rho^{(1)}$.

Figure 4 shows the radial profiles of the same functions as in figure 2, but now the discharge is sustained by the power of the first mode, by combination of first two modes, or by combination of first three modes. (Online colour: www.cpp-journal.org).

Effect of higher modes on discharge characteristics may change significantly when taking different wave frequency or discharge chamber radius. More precisely, effect of higher modes strongly depends on the ratio of
incident wave length and size of the discharge chamber. For testing, we calculated discharge characteristics, as functions of wave frequency, \( \omega \), (at fixed discharge chamber radius, \( R \)) and as functions of discharge chamber radius, \( R \) (at fixed wave frequency, \( \omega \)), at certain input powers, \( Q_{\text{app}} \), for first three modes, taken them into account separately, and for their combinations. When testing for the combinations of modes, the input microwave power was divided equally on modes. Results for the reflection coefficient, \( \rho \), and dissipated power, \( Q_{\text{dis}} \), are given in figures 6–9.

As results show, at rather small wave frequencies, higher modes almost totally reflected from the discharge (figures 6 and 7). With increasing in the frequency, the effect of the higher modes became comparable with the effect of the first mode. At further increase in the wave frequency, difference in the effect of different modes monotonically decreases.

Effect of the higher modes depending on the size of the discharge chamber is qualitatively similar. Notice that the profiles of reflection coefficients versus the radius of the discharge chamber resemble respective profiles for the wave frequency (compare figures 6 and 8).
Fig. 8 (a) Reflection coefficient, \( \rho \), and (b) dissipated power, \( Q_{\text{dis}} \), as functions of the radius of the discharge chamber, \( R \). Discharge is sustained separately by the power of the first mode, second mode, or third mode. Dotted lines correspond to the ionization equilibrium model (\( \dot{n}_e = 0 \)) [21]. \( Q_{\text{app}} = 1 \text{ kW}, \omega = 15 \times 10^9 \text{ rad s}^{-1}. \) (Online colour: www.cpp-journal.org).

Notice that figures presented include also results obtained in [21] within the framework of the ionization equilibrium model. In this case, plasma composition was derived from the Saha equation, to which continuity equation for electrons (4) reduces subject to the condition \( \dot{n}_e = 0 \). As comparison shows, at the beginning parts of the interval for \( \omega \) (figure 6) and of the interval for \( R \) (figure 8), respective plots for \( \rho \) and \( Q_{\text{dis}} \), corresponding to different model solutions, are almost coincide.

4 Concluding remarks

We have presented results of calculations of discharge plasma characteristics in argon at atmospheric pressure versus the external control parameters (the mode, frequency and power of the applied electric field and the radius of the discharge chamber).

Discharge model assumed a thermal and ionization non-equilibrium of plasma and took into account first three modes of incident electromagnetic waves.

Numerical results have been compared with the results of [21] for the ionization equilibrium discharge plasma model.

Calculations showed that effect of higher modes on the discharge characteristics is determined by the ratio of the incident microwaves length and the discharge chamber size. With increase in the wave frequency, \( \omega \), effect of higher modes becomes comparable with the effect of the first mode. At further increase in \( \omega \), difference between different mode effect monotonically diminishes. Effect of the higher modes dependent on the size of the discharge chamber is qualitatively similar.

References


