Numerical Simulation of Hydrogen Microwave Plasma Discharge Using a Fluid Model Approach

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Abstract

Microwave plasma processing technology plays a vitally important role in various fields such as electronic engineering and development of new materials. Further, it is one of the promising ways to synthesize large crystals of diamond in high growth rates. However, physical and chemical phenomena that occur in the plasma are very complex and strongly coupled. Understanding the correlation between the parameters of such phenomena, like electrons and ions density, electric field, plasma potential, microwave power and gas pressure, can significantly contribute to an efficient use of microwave plasma technology. Numerical modeling of plasma can be of great use to optimize the discharge parameters in order to improve the knowledge of plasma processing technology. This paper presents a numerical simulation of the impact of microwave power and gas pressure on the hydrogen microwave plasma discharge characteristics, using a fluid model approach which solves the electron and ion continuity equations, momentum transport equation
and the Poisson’s equation. The simulation results show a strong effect of power density and pressure on the species densities distribution in the plasma.

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1 Introduction

In recent years there has been a growing interest in microwave discharge plasma because of increasing number of its applications. It has been widely applied as a manufacturing method for etching or deposition [18] because it is clean and has high chemical reactivity [14]. Thus, microwave plasma created using a gas mixture consisting of primarily hydrogen with small additions of carbon-containing gases, has found an application in the chemical vapor deposition (CVD) of diamond films [10]. Indeed, it is one of the promising ways to synthesize large crystals of diamond in high growth rates [24]. Recently, it has been reported that approximately 10-mm-thick high-quality single-crystal diamonds have been synthesized by microwave plasma CVDs [23]. The hydrogen atoms play a key role in diamond CVD since they stabilize the growth of diamond and suppress the formation of graphite [6]. Different types of CVD reactors are used, in which hot filament, RF, DC and microwave discharges, and flames are used. The advantages of microwave discharge reactors are: absence of electrodes; high specific power contribution; high densities of excited and charged particles; and a relatively large area and high homogeneity of the film [9]. However, discharge plasma has physicochemical phenomena very complex and strongly coupled. Further, it is difficult to experimentally observe physical quantities of plasmas inside the reactors. Therefore, the numerical simulation of microwave plasma is a necessity to understand the plasma behavior inside the reactor, and to improve the knowledge for deposition or etching by means of plasma technology. The present paper offers a numerical simulation of a pure hydrogen discharge characteristics in a cylindrical reactor, using a fluid plasma approach. The hydrogen plasma example was chosen because diamond film deposition processes often consist of high percentages of hydrogen in the discharge. And, as shown by Koemtzopoulos et al, adding small percentages (e.g. 1%) of methane to a hydrogen discharge has only a minimal effect on the electron energy distribution function [22]. The results found concern mainly the effect of pressure and microwave power density on the discharge characteristics.

This paper is organized as follows: In section 2, the plasma discharge models
are presented, while section 3 describes the fluid approach. Section 4 presents the discharge parameters. Section 5 gives the numerical simulation. In section 6, the simulation results and discussion are presented. Finally, the conclusion is given in section 7.

2 Plasma discharge models

A number of different numerical models were published for the plasmas modeling used in a variety of applications. The most commonly used models are: fluid models [1, 13, 17], particle-in-cell/Monte Carlo (PIC/MC) models [16, 25] and hybrid models [3, 4]. All these modeling approaches have their specific advantages and limitations, and therefore, the choice of the model is often dictated by the gas discharge and conditions under study. Particle-in-cell/Monte Carlo (PIC/MC) models are generally known as the most accurate approach, because they consider the plasma species on their lowest microscopic level. The trajectory of each species is calculated using Newton’s laws, while the collisions between the plasma species depend on the cross sections, and are determined by random number [2]. However, these models are very time consuming because of a large number of particles in the plasma. Fluid models are relatively simple modeling way of plasmas compared with kinetic approaches. They are generally based on solving the continuity and transport equations for the various plasma species, in combination with Poisson equation, in order to obtain a self-consistent electric field distribution. This approach is particularly suitable for describing the detailed plasma chemistry. Indeed, a large number of different plasma species and chemical reactions can be included in the model, without too much computational effort. These models are widely used for the modeling of different kinds of plasmas [19], their main advantage is that their computational effort is still lower compared to the other numerical technique for plasma modeling. Hence, this makes a fluid model a useful tool for the modeling of discharge plasmas. Thus, many of the modeling effort performed to obtain a better understanding of the discharge mechanisms are based on a fluid approximation. Further, plasmas can also be described by the hybrid model which combines several models (e.g., fluid model and Monte Carlo model) into a modeling network. In this way, the advantages of the individual models can be combined, whereas the disadvantages can be avoided. In this study, a fluid plasma model is applied to describe the hydrogen discharge characteristics by solving the electron and ion continuity equations, momentum transport equations and the Poisson’s equation.
3 Fluid approach description

The simulation of plasma processes can be based generally on two major approaches. One is the particle approach, which is carried out using a particle simulation technique that treats the plasma as a combination of particles (electron, ion, neutral). The other approach is the fluid method, which treats the plasma as a fluid and solves the equations obtained from the moments of the Boltzmann transport equation. The Boltzmann equation (BE) is a fundamental equation describing the transport of an ensemble of particles. It is given by the following form [5]:

\[
\left( \frac{\partial}{\partial t} + \vec{v} \cdot \vec{\nabla} + \frac{\vec{F}}{m} \cdot \vec{\nabla} \right) f(\vec{r}, \vec{v}, t) = \left( \frac{\partial f(\vec{r}, \vec{v}, t)}{\partial t} \right)_{\text{coll}}
\]

(1)

Here, \( f(\vec{r}, \vec{v}, t) \) is the distribution function, \( \vec{r} \) denotes the spatial position, \( \vec{v} \) denotes the velocity, and \( t \) denotes the time. \( m \) is the mass of the particle, \( \vec{F} \) denotes the external forces, and the term on the right side of the equation (1) represents the collision term of the Boltzmann equation, which accounts for changes of the electron velocity distribution function because of electrons collisions undergo mainly with neutrals but also with other electrons and ions. Equation (1) is a partial integro-differential equation in seven dimensions (three in space, three in velocity and time), and as such is extremely difficult to solve. The fluid model of plasma, reducing the complexities in the kinetic description, is based on partial differential equations which describe the macroscopic quantities such as density, flux, average velocity, pressure and temperature.

The equations for macroscopic quantities, called fluid equations, are obtained from the Boltzmann equation by taking velocity moments [12]. Thus, zero moment of the Boltzmann equation \( \int_{-\infty}^{+\infty} (BE) \, d^3\nu \) yields continuity equation for the particle density as:

\[
\frac{\partial n}{\partial t} + \vec{\nabla} \cdot (n \vec{u}) = \int_{-\infty}^{+\infty} \left( \frac{\partial f}{\partial t} \right)_{\text{coll}} \, d^3\nu
\]

(2)

Here, the particle density \( n \) and the average velocity \( \vec{u} \) are defined as \( n = \int_{-\infty}^{+\infty} f \, d^3\nu \) and \( \vec{u} = \frac{1}{n} \int_{-\infty}^{+\infty} \vec{v} f \, d^3\nu \). The source term on the right side of the continuity equation corresponds to the collision term of the Boltzmann equation.

The momentum transport equation (equation of motion) can be found as first moment of the Boltzmann equation \( m \int_{-\infty}^{+\infty} \vec{v} (BE) \, d^3\nu \) such as:

\[
m n \frac{\partial \vec{u}}{\partial t} + mn (\vec{u} \cdot \nabla) \vec{u} + \nabla \vec{P} - nq(\vec{E} + \vec{u} \times \vec{B}) = m \int_{-\infty}^{+\infty} (\vec{v} - \vec{u}) \left( \frac{\partial f}{\partial t} \right)_{\text{coll}} \, d^3\nu
\]

(3)
Where \( \overrightarrow{E} \) and \( \overrightarrow{B} \) are the electric and magnetic fields, respectively.

Similarly, the energy equation can be found as second moment of the Boltzmann equation \( \left( \frac{1}{2} m \int_{-\infty}^{+\infty} \nu^2 (BE) d^3 \nu \right) \) as:

\[
\frac{1}{\gamma - 1} \left( \frac{\partial p}{\partial t} + \nabla \cdot (p \overrightarrow{u}) \right) + \nabla \cdot \overrightarrow{P} = \frac{1}{2} m \int_{-\infty}^{+\infty} (\overrightarrow{\nu} - \overrightarrow{u})^2 \left( \frac{\partial f}{\partial t} \right)_{\text{coll}} d^3 \nu
\]

\( \overrightarrow{Q} = \frac{1}{2} m \int_{-\infty}^{+\infty} (\overrightarrow{\nu} - \overrightarrow{u})(\overrightarrow{\nu} - \overrightarrow{u})^2 f d^3 \nu \) defines heat flux \( \overrightarrow{Q} \).

\( \overrightarrow{P} = m \int_{-\infty}^{+\infty} (\overrightarrow{\nu} - \overrightarrow{u})(\overrightarrow{\nu} - \overrightarrow{u}) f d^3 \nu \) defines pressure tensor \( \overrightarrow{P} \), and \( P_{ij} = p \delta_{ij} \) defines scalar pressure \( p \). \( \gamma \) is the ratio of specific heats.

This simulation consists of the particle and momentum equations for electrons and ions, which are combined with the Poisson’s equation.

In the steady state, the governing equations used in this study are given by:

\[
\nabla^2 \psi = \frac{e}{\varepsilon_0} (n_e - n_i) \quad (5)
\]

\[
\nabla \cdot \overrightarrow{J}_e = n_e n_n k_{ion} - \alpha_r n_i n_e \quad (6)
\]

\[
\nabla \cdot \overrightarrow{J}_i = n_e n_n k_{ion} - \alpha_r n_i n_e \quad (7)
\]

\[
\overrightarrow{J}_e = -n_e \mu_e \overrightarrow{E} - D_e \nabla n_e \quad (8)
\]

\[
\overrightarrow{J}_i = n_i \mu_i \overrightarrow{E} - D_i \nabla n_i \quad (9)
\]

Equation (5) represents the Poisson’s equation, which gives the electric interaction between electrons and ions [20], where \( \psi \) is the electric potential.

The electric field \( \overrightarrow{E} \) is derived from a scalar potential, \( \psi \), by: \( \overrightarrow{E} = -\nabla \psi \).

Equations (6) and (7) represent the electron and ion continuity equations, respectively. They are written by balances among convective diffusion, production/ vanishing due to ionization/ recombination.

Thus, the term on the left-hand side of equations (6) and (7) represents the particles flux variation in position, while the first and second terms on the right-hand side correspond to source terms due to ionization and recombination, respectively.

Equations (8) and (9) represent the momentum balances for electrons and ions, respectively, in which the species fluxes are expressed as the sum of drift and diffusion terms. Indeed, the first term on the right-hand side of these equations represents the migration of the charged particles under influence of an electric field, while the second term gives the diffusion due to particles concentration gradient.

The drift diffusion approximation reduces the number of partial differential equations included in the model by the use of the algebraic expression for particle flux (Equations (8) and (9)) instead of full equation of motion [12, 15].

In the above equations, \( n_e \) and \( n_i \) are the electron and ion densities, respec-
respectively; $\overrightarrow{J}_e$ and $\overrightarrow{J}_i$ are the electron and ion fluxes, respectively; $k_{\text{ion}}$ is the inelastic rate constant for ionization; and $\alpha_r$ is the recombination rate constant; $D_{e,i}$ and $\mu_{e,i}$ are the electron and ion diffusivities and mobilities, respectively.

4 Discharge parameters

The hydrogen plasma discharge is assumed to be partially ionized and partially dissociated. Then, the major particle interaction processes are the electron-$\text{H}_2$ molecule inelastic collision, electron-$\text{H}_2$ molecule elastic collision and electron-hydrogen ion recombination. The electron-$\text{H}_2$ inelastic collisions include the $\text{H}_2$ molecule ionization process. The rate coefficient in equations (6) and (7) for this collision process can be expressed using the Arrhenius relationship [21, 22] as:

$$k_{\text{ion}} = A_{\text{ion}} \exp \left( \frac{-\epsilon_{\text{ion}}}{K_B T_e} \right)$$

where $\epsilon_{\text{ion}}$ is the threshold energy for $\text{H}_2$ molecule ionization, $T_e$ is the electron temperature, $K_B$ is the Boltzmann constant and $A_{\text{ion}}$ is the pre-exponential factor, which is obtained by approximating the rate constant data at low electron temperatures to this relationship.

The reactions considered in this study are:

- Ionization:

  $$e + \text{H}_2 \rightarrow e + \text{H}_2^+ + e$$

  $$\text{H}_2^+ + \text{H}_2 \rightarrow \text{H}_3^+ + \text{H}$$

- Recombination:

  $$e + \text{ion} \rightarrow \text{neutral}$$

It should be noted that the only neutral species considered in the ion and electron simulations was the $\text{H}_2$ species and that the dominant ionic species in the plasma is $\text{H}_3^+$ [21, 22].

The rate and transport parameters that are used in the model are summarized in the table 1 [21]:
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\[ \mu_{en} = 1.0 \times 10^{24} m^{-1} V^{-1} s^{-1} \]
\[ D_{en} = 5.0 \times 10^{23} m^{-1} s^{-1} \]
\[ \mu_{i} n_{i} = 3.5 \times 10^{22} m^{-1} V^{-1} s^{-1} \]
\[ D_{i} n_{i} = 3.5 \times 10^{21} m^{-1} s^{-1} \]
\[ A_{\text{ion}} = 1.0 \times 10^{-14} m^3 s^{-1} \]
\[ \alpha_{r} = 1.0 \times 10^{-13} m^3 s^{-1} \]
\[ \varepsilon_{\text{ion}} = 15.4 eV \]

Table 1: Rate and transport parameters.

The electron diffusivity and mobility are written, respectively, as:

\[ D_{e} = \frac{K_B T_e}{m_e \nu_{en}} \]
\[ \mu_{e} = \frac{e}{m_e \nu_{en}} \]

where \( \nu_{en} \) is the electron-neutral (in this case, electron-H\(_2\)) momentum transfer frequency, \( e \) is the elementary charge and \( m_e \) is the electron mass. The collision frequency for electron-H\(_2\) molecule momentum transfer is relatively independent of the electron temperature, so it can be written as [22]:

\[ \nu_{en}(H_2) = 1.44 \times 10^{12} \times \frac{\text{Pressure}(\text{Torr})}{T_n(K)} \]

where \( T_n \) is the neutral temperature, which can be represented by the translational temperature of H\(_2\) gas.

When working with pure hydrogen discharges, there are two key parameters that govern the process. These are the input microwave power and the pressure in the deposition reactor [11].

One of the best ways to increase hydrogen atom density and then growth rates is to increase the power density of the plasma. This can be done either by increasing simultaneously pressure and power keeping constant the plasma volume [8] or by increasing only pressure while keeping constant the power, in this latter case the plasma volume decreases [7].

The empirical equations used for the translational temperature of H\(_2\) gas, and
the discharge volume, are [22]:

\[
\text{Translational temperature}(K) = 228.6 + 374.3 \times \text{Incident power}(KW) + 16.5 \times \text{Pressure}(\text{Torr}) \pm 94.2
\]

\[
\text{Plasma Volume}(cm^3) = 449.7 + 116.2 \times \text{Incident power}(KW) - 18.1 \times \text{Pressure}(\text{Torr})
\]

\[
+ 57.1 \times [\text{Incident power}(KW)]^2
\]

\[
+ 0.25 \times [\text{Pressure}(\text{Torr})]^2
\]

\[
- 5.4 \times \text{Pressure}(\text{Torr}) \times \text{Incident power}(KW)
\]

\[
\pm 15.4
\]

In this study, it is assumed that 100% of the microwave power coupled into the reactor is absorbed by the plasma.

5 Numerical simulation

The simulation region in this work has a cylindrical form; therefore, the discharge behavior can be assumed to be \(\phi\) symmetric and the discretization of the equations reduced to a two-dimensional problem. Thus, the simulation region remains in the r-z plane only.

The governing equations of this problem can be rewritten in cylindrical coordinates. Thus, the Poisson’s equation (5) can be given as:

\[
\frac{\partial^2 \psi}{\partial r^2} + \frac{1}{r} \frac{\partial \psi}{\partial r} + \frac{\partial^2 \psi}{\partial z^2} = \frac{e}{\varepsilon_0} (n_e - n_i) \quad (10)
\]

The equations (6) and (8) give the electron continuity equation as:

\[
- D_e \left[ \frac{\partial^2 n_e}{\partial r^2} + \frac{1}{r} \frac{\partial n_e}{\partial r} + \frac{\partial^2 n_e}{\partial z^2} \right] + \mu_e \left[ \frac{\partial \psi}{\partial r} \frac{\partial n_e}{\partial r} + \frac{\partial \psi}{\partial z} \frac{\partial n_e}{\partial z} \right]
\]

\[
+ \mu_e \left[ \frac{\partial^2 \psi}{\partial r^2} + \frac{1}{r} \frac{\partial \psi}{\partial r} + \frac{\partial^2 \psi}{\partial z^2} \right] n_e + \alpha_r n_i n_e - k_{ion} n_n n_e = 0 \quad (11)
\]

Similarly, the equations (7) and (9) give the ion continuity equation as following form:

\[
- D_i \left[ \frac{\partial^2 n_i}{\partial r^2} + \frac{1}{r} \frac{\partial n_i}{\partial r} + \frac{\partial^2 n_i}{\partial z^2} \right] - \mu_i \left[ \frac{\partial \psi}{\partial r} \frac{\partial n_i}{\partial r} + \frac{\partial \psi}{\partial z} \frac{\partial n_i}{\partial z} \right]
\]

\[
- \mu_i \left[ \frac{\partial^2 \psi}{\partial r^2} + \frac{1}{r} \frac{\partial \psi}{\partial r} + \frac{\partial^2 \psi}{\partial z^2} \right] n_i + \alpha_r n_i n_e - k_{ion} n_n n_e = 0 \quad (12)
\]

We discretize these equations in two-dimensional cylindrical coordinates (r and z directions as shown in figure 1), by using the finite difference method with
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centred scheme. Hence, the discretization of the Poisson’s equations (10) is given by:

\[
\begin{align*}
\frac{\psi(i + 1, j) - 2\psi(i, j) + \psi(i - 1, j)}{(\Delta r)^2} + \frac{\psi(i + 1, j) - \psi(i - 1, j)}{2i(\Delta r)^2} \\
+ \frac{\psi(i, j + 1) - 2\psi(i, j) + \psi(i, j - 1)}{(\Delta z)^2} - \frac{e}{\varepsilon_0} [n_e(i, j) - n_e(i, j)] = 0 \quad (13)
\end{align*}
\]

The discretization of the electrons continuity equation (11) is given as:

\[
\begin{align*}
&- D_e \left[ \frac{n_e(i + 1, j) - 2n_e(i, j) + n_e(i - 1, j)}{(\Delta r)^2} + \frac{n_e(i + 1, j) - n_e(i - 1, j)}{2i(\Delta r)^2} \right] \\
&- D_e \left[ \frac{n_e(i, j + 1) - 2n_e(i, j) + n_e(i, j - 1)}{(\Delta z)^2} \right] + \mu_e \left[ \frac{\psi(i + 1, j) - \psi(i - 1, j)}{2(\Delta r)} \right] n_e(i, j) \\
&+ \mu_e \left[ \frac{\psi(i, j + 1) - \psi(i, j - 1)}{2(\Delta z)} \right] n_e(i, j) - n_n k_{ion} n_e(i, j) + \alpha e n_e(i, j) n_e(i, j) = 0 \quad (14)
\end{align*}
\]

Similarly, the discretization of the ions continuity equation (12) is given as:

\[
\begin{align*}
&- D_i \left[ \frac{n_i(i + 1, j) - 2n_i(i, j) + n_i(i - 1, j)}{(\Delta r)^2} + \frac{n_i(i + 1, j) - n_i(i - 1, j)}{2i(\Delta r)^2} \right] \\
&- D_i \left[ \frac{n_i(i, j + 1) - 2n_i(i, j) + n_i(i, j - 1)}{(\Delta z)^2} \right] - \mu_i \left[ \frac{\psi(i + 1, j) - \psi(i - 1, j)}{2(\Delta r)} \right] n_i(i, j) \\
&- \mu_i \left[ \frac{\psi(i, j + 1) - \psi(i, j - 1)}{2(\Delta z)} \right] n_i(i, j) - n_n k_{ion} n_i(i, j) + \alpha e n_i(i, j) n_e(i, j) = 0 \quad (15)
\end{align*}
\]

Here, \(i\) and \(j\) denote the grid indices in the \(r\) and \(z\) directions respectively, while \(\Delta r\) and \(\Delta z\) denote the space steps.
The boundary conditions for the fluid plasma model at the substrate and the edge of the plasma volume; figure 1; are:

\[ n_e = n_i = 0 \]
\[ \psi = 0 \]

and at the centerline \((r = 0)\):

\[ \frac{\partial n_e}{\partial r} = \frac{\partial n_i}{\partial r} = \frac{\partial \psi}{\partial r} = 0. \]

Figure 1: Active Plasma Zone

In order to solve the system of nonlinear discretized equations, we applied in this simulation the Newton-Raphson iteration method.

6 Simulation results and discussions

A numerical simulation of a hydrogen plasma discharge using a fluid model approach in cylindrical geometry has been performed. The main input parameters for this model include the pressure and microwave power. The spatial distribution of plasma density at a given microwave power density is determined as shown in Figure 2 and Figure 3. The results show that the plasma density is maximal in the plasma volume near the center of the discharge \((r = 0\,cm)\), and decreases in the edges and near the substrate region.
By increasing simultaneously power and gas pressure keeping constant the plasma volume, the evolution of axial and radial profiles of electron density, for different power densities, is also calculated and presented in Figures 4-7. Thus, Figures 4 and 5 present the evolution of electron density along the axial direction in the hydrogen discharge at a fixed radial position (r=0 and r=3.5 cm respectively), for different power densities, where the substrate is situated at the position z = 0.
It is shown that the electron density increases significantly above the substrate with the axial position until it reaches its maximum value at the center of the discharge where $z = 1.75\,cm$, and then it diminishes to vanishing at the edge of the plasma.
The radial profiles of electron density in the discharge for different power densities are shown in Figures 6 and 7. It can be seen, from these figures, that the electron density decreases from its maximum value at the center \( r=0 \) to a minimum value at the edge of the plasma.

Figure 6: Radial profile of electron density for different power densities at \( z =1 \) cm

Figure 7: Radial profile of electron density for different power densities at \( z =3 \) cm

It is also seen from the figures 4 - 7, that the increasing in microwave power
density from 18.88 to 26.20 W/cm$^3$ under constant plasma volume leads to an important increasing of electron density in the center of the discharge. This result evidences the strong effect of microwave power density which is due to a coupled action of pressure and power.

The variation of the maximum density of electrons in the pure hydrogen discharge can be represented versus power density as shown in Figure 8. Its profile is seen to vary linearly in the plasma volume from $5.5 \times 10^{17} m^{-3}$ to $8.1 \times 10^{17} m^{-3}$ as the power density is increased from around 16W/cm$^3$ to 26W/cm$^3$.

Figure 8: Maximum density of electrons versus power density.

Figure 9 shows the dependence of the maximum density of electrons on pressure for hydrogen discharge at an absorbed power of 1900W. the most
prevailent feature is that the electron density increases as the pressure increases. Indeed, the maximum density of electrons rises from $5 \times 10^{16} \text{m}^{-3}$ to around $3.3 \times 10^{17} \text{m}^{-3}$ when the pressure is increased from 35 Torr to 49 Torr. This result shows clearly the important influence of pressure on the electron density. The enhancement in the production of electron density can be related to the increase in the gas temperature which correspond to the increase in pressure.

7 Conclusion

In this paper, a fluid plasma model is presented to describe the hydrogen microwave plasma discharge characteristics by solving the electron and ion continuity equation, momentum transport equation and the Poisson’s equation. The governing equations are discretized and solved in two-dimensional cylindrical coordinates using the finite difference method. Moreover, Newton-Raphson iteration is applied in order to solve the nonlinear equations. The characteristics of discharge, including in particular the plasma density were investigated. Because plasma distribution could be one of the most important factors which determine power efficiency of the diamond growth rate, we have focused on distributions of electrons number density to provide information on the key plasma parameters that control the processes of diamond deposition. As a result of the calculations using this model, distribution of electrons density is obtained for various conditions of power and pressure. The simulation results show a strong effect of gas pressure and power density, on the discharge characteristics, such as plasma density.

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