Numerical Study of Microwave Plasma Characteristics in Moderate Pressure Used for Diamond Deposition

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Abstract

The present paper provides the results of numerical simulation obtained using a fluid model approach of pure hydrogen microwave plasma, at moderate pressure, in two-dimensional cylindrical geometry. The governing equations consist of continuity equations of electrons and ions in drift-diffusion approximation, as well as Poisson equation for self-consistent electric field. Dependencies of discharge characteristics, such as densities of charged particles and plasma potential, on the
main physical parameters namely the hydrogen pressure and the power density have been numerically studied. It is seen that the electron and ion densities were in the range of $5 \times 10^{16} - 3 \times 10^{17} \text{ m}^{-3}$ for the pressure range of 35-49 Torr. It is also shown that as the power density increases the charged particles densities increase significantly; and that the plasma potential varies slightly with pressure and power density.

**Keywords**: Plasma fluid model, Moderate pressure, Drift-diffusion, Numerical simulation.

1. Introduction

Recent years have seen intense studies of diamond films deposition by the chemical vapor deposition (CVD) method [7,11,15,30]. The interest in CVD diamond films is due to its various applications in the optical, mechanical, thermal and electronic domains [9]. Different types of CVD reactors are used, including hot filament, RF, DC and microwave discharges. The advantages of microwave discharge reactors are numerous; these include the absence of electrodes; high specific power contribution; high densities of excited and charged particles; and a relatively large area and high homogeneity of the films [11,18]. Moreover, microwave excited plasma have demonstrated good potential in moderate pressure plasma processing applications [28] including the chemical vapor deposition (CVD) of diamond films [14]. For this application, microwave plasma discharge is produced using feed gas consisting mainly of hydrogen and a few percents of hydrocarbon species such as methane. Hydrogen atoms constitute the main governing species for diamond deposition [1,3,5,7,19,20,29,31], this is due to the high reactivity of these atoms produced by electron impact dissociation of molecular hydrogen [5].

The improved understanding of the changes in diamond film characteristics and growth rate requires knowledge of processes taking place in the plasma volume, and parameters that govern discharge. Previous work has been performed to provide information on the key plasma parameters that control key species densities, such as hydrogen atoms, which are responsible for diamond growth mechanisms, growth rate and diamond film characteristics.

Indeed, Giquel et al [9] have reported the experimental results which show variations in diamond characteristics as plasma variables, like pressure and power, are systematically changed, and discussed the effects of these variables on the local parameters such as H-atom densities. Understanding of the atomic hydrogen molar fraction behavior versus pressure and average gas temperature has been studied by Grotjhon et al [15]. H-atom density in diamond deposition plasma reactors under conditions of high power density has been also estimated by Gicquel et al [7] using actinometry technique.
However, charged particles distribution in the plasma could be one of the most important factors which determine power efficiency of the diamond growth rate. Therefore, we have focused in this work on the distribution of the plasma density and electric potential, at moderate pressure, to provide information on the key plasma parameters that control the processes of diamond deposition.

In this paper, a numerical model analysis is presented for a two-dimensional cylindrical geometry. The model is based on the fluid approach with a drift-diffusion approximation for the particles charged fluxes. The main objective of this work is to study in particular the effect of the gas pressure and power density on the discharge characteristics such as electron and ion densities as well as plasma electric potential at moderate pressure.

The structure of this paper is as follows. In section 2, the physical model description is presented, while section 3 is devoted to numerical results and discussion. Finally, the conclusion is given in section 4.

2. Physical model description

Different numerical models were applied to describe the dynamics of plasma discharge in various applications. The modeling approaches can be classified as fluid methods, kinetic (particle) methods and their combinations, called hybrid methods [2, 24]. Among those methods, the fluid approach has been used widely because it helps to understand the physical structure of the discharge, and is less time consuming in numerical computation compared to the kinetic simulation [4].

2.1. Basic equations and assumptions

The physical model adopted in this work is based on a two-dimensional fluid description of charged particles (electrons and ions) partially ionized hydrogen gas at moderate pressure. The model consists of the particle and momentum balances equations, for ions and electrons, obtained from moments of the Boltzmann equation [23], which are combined with the Poisson equation, in order to obtain the electric potential. In such fluid simulation, the model can be based on the following equations:

\[
\frac{\partial n_e}{\partial t} + \nabla \cdot \vec{I}_e = n_e n_i k_{ion} - \alpha_r n_i n_e \tag{1}
\]

\[
\frac{\partial n_i}{\partial t} + \nabla \cdot \vec{I}_i = n_e n_i k_{ion} - \alpha_r n_i n_e \tag{2}
\]

\[
m_e \frac{\partial \vec{v}_e}{\partial t} + m_e \nabla \cdot (\vec{u}_e \cdot \vec{I}_e) + e n_e \vec{E} + \nabla \cdot (n_e k_B T_e) = -m_e \nu_{en} \vec{I}_e \tag{3}
\]

\[
m_i \frac{\partial \vec{v}_i}{\partial t} + m_i \nabla \cdot (\vec{u}_i \cdot \vec{I}_i) - e n_i \vec{E} + \nabla \cdot (n_i k_B T_i) = -m_i \nu_{in} \vec{I}_i \tag{4}
\]

\[
\nabla^2 \phi = \frac{e}{\varepsilon_0} (n_e - n_i) \tag{5}
\]
Equations (1) and (2) represent the electron and ion continuity equations respectively, while the equations (3) and (4) represent the momentum transport equations for electrons and ions respectively. Equation (5) is the Poisson’s equation. Where $n_e$ is the electron density; $n_i$ is the ion density; $n_n$ is the neutral density; $m_e$ is the electron mass; $m_i$ is the ion mass; $\vec{u}_e$ is the electron average velocity; $\vec{u}_i$ is the ion average velocity; $\vec{I}_e$ is the electron flux; $\vec{I}_i$ is the ion flux; $T_e$ is the electron temperature; $T_i$ is the ion temperature; $\vec{E}$ is the electric field; $\Phi$ is the electric potential; $\varepsilon_0$ is the permittivity of the vacuum; $k_{ion}$ is the inelastic rate constant for ionization; $\alpha_r$ is the recombination rate constant; $\nu_{en}$ is the electron-neutral collision frequency; $\nu_{in}$ is the ion-neutral collision frequency; and $K_B$ is the Boltzmann constant.

Nevertheless, the numerical resolution of the momentum equations requires much more computation time and a specific stabilization technique. Therefore, the drift-diffusion approximation is frequently used to reduce the momentum equations included in model, by the use of the algebraic expression for the charged particles fluxes instead of full equation of motion [10,12,13,22,25]. Indeed, the charged particles are assumed to be isothermal and the plasma to be weakly ionized. They lead to the drift-diffusion equations [4,21,23,27]. This approximation is then adopted in the present study to replace the momentum equation.

In summary, the fluid plasma model used in this paper is governed, in the steady state, by the following set of equations:

- Electron continuity equation:
  \[ \nabla \cdot \vec{I}_e = n_e n_n k_{ion} - \alpha_r n_i n_e \]  
  (6)

- Electron flux:
  \[ \vec{I}_e = -n_e \mu_e \vec{E} - D_e \nabla n_e \]  
  (7)

- Ion continuity equation:
  \[ \nabla \cdot \vec{I}_i = n_e n_n k_{ion} - \alpha_r n_i n_e \]  
  (8)

- Ion flux:
  \[ \vec{I}_i = n_i \mu_i \vec{E} - D_i \nabla n_i \]  
  (9)

The system is completed by the Poisson equation (5) which gives the electric potential.

The electric field is derived from the potential by:
\[ \vec{E} = -\nabla \Phi \]  
(10)

In the above equations, $\mu_{e,i}$ and $D_{e,i}$ are the electron and ion mobility and diffusion coefficients respectively.

The governing equations are discretized and solved in two-dimensional cylindrical coordinates (radial and axial directions as shown in Fig 1) using the finite difference method. Furthermore, the Newton-Raphson method is used to solve the nonlinear equations [6].

The boundary conditions adopted in the present model at the substrate and the edge of the plasma volume are:

- For the electron and ion densities:
- For the electric potential of the plasma:
  \( \phi = 0 \).

Moreover, at the centerline of the discharge where \( r=0 \text{cm} \), we have the following conditions:
\[
\frac{\partial n_e}{\partial r} = \frac{\partial n_i}{\partial r} = \frac{\partial \phi}{\partial r} = 0.
\]

### 2.2. Rate and transport parameters

We concentrated on pure hydrogen microwave plasma throughout the model developed in this work, because the diamond film deposition processes often consist of high percentages of hydrogen gas; and the addition of small percentages of methane to a hydrogen discharge has only a minimal effect on the electron energy distribution function [8,28]. Thus, the hydrogen molecules \( \text{H}_2 \) are the most abundant neutral species in the plasma studied here, and the dominant ionic species are the \( \text{H}_3^+ \) [26,28]. Therefore, the major processes of chemical interactions involving the dominant plasma species that are considered in the model include the ionization reactions between electrons and hydrogen molecules and the recombination reactions between electrons and hydrogen ions. The ionization of hydrogen molecules is the process responsible for creation of charged particles in the plasma:
\[
e + \text{H}_2 \rightarrow e + \text{H}_2^+ + e
\]
\[
\text{H}_2^+ + \text{H}_2 \rightarrow \text{H}_3^+ + \text{H}
\]

The rate of this ionization reaction can be expressed using the Arrhenius relationship [26,28]:
\[
k_{\text{ion}} = A_{\text{ion}} \exp \left( \frac{-\varepsilon_{\text{ion}}}{K_B T_e} \right)
\]
Where $E_{\text{ion}}$ is the threshold energy for $\text{H}_2$ molecule ionization, $A_{\text{ion}}$ is the pre-exponential factor, $K_B$ is the Boltzmann constant and $T_e$ is the electron temperature.

The loss of charged particles is due to the recombination reaction between the electrons and ions in the plasma volume:

$$e + H_3^+ \rightarrow H_2 + H$$

The recombination reaction is characterized by the recombination rate constant $\alpha_r$ which appears in equations of the model.

The rate and transport parameters used in the present work are summarized in the following table [26]:

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$D_e n_n$</td>
<td>$5.0 \times 10^{23} \text{ m}^{-1}\text{s}^{-1}$</td>
</tr>
<tr>
<td>$D_i n_n$</td>
<td>$3.5 \times 10^{21} \text{ m}^{-1}\text{s}^{-1}$</td>
</tr>
<tr>
<td>$\mu_e n_n$</td>
<td>$1.0 \times 10^{24} \text{ m}^{-1}\text{V}^{-1}\text{s}^{-1}$</td>
</tr>
<tr>
<td>$\mu_i n_n$</td>
<td>$3.5 \times 10^{22} \text{ m}^{-1}\text{V}^{-1}\text{s}^{-1}$</td>
</tr>
<tr>
<td>$A_{\text{ion}}$</td>
<td>$1.0 \times 10^{-14} \text{ m}^3\text{s}^{-1}$</td>
</tr>
<tr>
<td>$E_{\text{ion}}$</td>
<td>15.4 eV</td>
</tr>
<tr>
<td>$\alpha_r$</td>
<td>$1.0 \times 10^{-14} \text{ m}^3\text{s}^{-1}$</td>
</tr>
</tbody>
</table>

**Table: Rate and transport parameters [26].**

### 3. Simulation results and discussion

We present the results of the numerical modeling constructed and developed for pure hydrogen discharge in cylindrical geometry at moderate pressure. The results obtained are designed to identify the discharge behavior by studying the effect of the main physical parameters such as power density and pressure, on the plasma characteristics like electron and ion densities as well as plasma electric potential.

#### 3.1. Electron and ion densities

Figs 2 and 3 compare the axial variations of electron and ion densities in the plasma at a fixed radial position ($r=0\text{cm}$), for power density of 18.88W/cm$^3$ and 26.20W/cm$^3$ respectively. While Figs 4 and 5 show the comparison between radial profiles of electron and ion densities at a fixed axial position ($z=2\text{cm}$), for power density of 18.88W/cm$^3$ and 26.2W/cm$^3$ respectively.
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We clearly see that the electron and ion densities are very close; and that the evolution of the ion density follows exactly that of the electron, with a very slight difference especially at 26.2W/cm$^3$. Thus, the densities of electron and ion are, as expected, almost equal in the discharge; which shows the quasi-neutrality of the plasma.

**Fig.2** : Axial distributions of electron density $n_e$, and ion density $n_i$, for power density of 18.88W/cm$^3$ at $r=0$cm.

**Fig.3** : Axial distributions of electron density $n_e$, and ion density $n_i$, for power density of 26.20W/cm$^3$ at $r=0$cm.
From these results, one can also deduce a significant increase in the density of ions and electrons with increasing the power density. Indeed, an increase in the charged particles densities from $6 \times 10^{17} \text{ m}^{-3}$ to around $8 \times 10^{17} \text{ m}^{-3}$ at the position ($r=0\text{ cm}$ and $z=2\text{ cm}$) is observed as the power density is increased from $18.88 \text{ W/cm}^3$ to $26.20 \text{ W/cm}^3$. This effect of power density on the charged particles densities can be related to the elevation of gas temperature as the power density increases.
increases. Indeed, increasing the micro-wave power density results in a higher gas temperature [7,16,17].

In Fig. 6, we have presented the variation of the maximum density of ions and electrons versus gas pressure, at a given power density. As can be seen, the charged particles densities increase strongly from around $5 \times 10^{16} \text{m}^{-3}$ to $3.3 \times 10^{17} \text{m}^{-3}$ when the gas pressure is increased from 35Torr to 49Torr, evidencing the important effect of pressure on the electron and ion densities.

![Graph showing variations of the maximum density of ions and electrons versus pressure for power of 1900W.](image)

*Fig. 6: Variations of the maximum density of ions and electrons versus pressure for power of 1900W.*

The radial and axial distributions of the plasma density for several pressure conditions have been also obtained. Thus, Figs. 7 and 8 present the evolution of the radial and axial profiles of electron density, for different pressures (35Torr, 40Torr, 45Torr and 48Torr) at a constant power (1900W). The results show clearly the strong impact of pressure on the plasma density. It is also shown that the maximum density is reached near the center of the discharge.
**Fig. 7:** Electron density evolution along the z axis \((r=0\, cm)\) for different pressures, at absorbed power of 1.9 kW.

**Fig. 8:** Radial evolution of the electron density \((z=2\, cm)\) for different pressures, at absorbed power of 1.9 kW.
The strong increase in the densities of charged particles as gas pressure is increased can be attributed to the increase in the collision frequency with pressure.

3.2. Plasma electric potential

The effect of pressure and power density on the plasma electric potential is also determined and studied in this work. Thus, Figs. 9 and 10 present the axial and radial distributions of the plasma potential profiles for different conditions of pressure (35Torr, 45Torr and 55Torr) at a fixed power (1900W). While the variation of the maximum potential as a function of power density is shown in Fig. 11.

**Fig.9:** Axial distribution of plasma potential \((r=0\text{cm})\) for different pressures, at a power of 1,9 kW.

**Fig.10:** Radial distribution of plasma potential \((z=3\text{cm})\) for different pressures, at a power of 1,9 kW.
It is clearly seen in Figs. 9 and 10 that the plasma potential slightly increases with pressure; and that the maximum value is reached near the center of the discharge (r=0cm and z=2.4cm). It is also seen in Fig. 11 that when power density increases the maximum value of plasma electric potential changes very slightly.

This slight change in the electrical potential of the plasma as a function of pressure and power density can be explained by the fact that the increase of these parameters leads, as we have seen above, to an increase in the density of electrons and ions simultaneously.

4. Conclusion

The purpose of this paper was to describe the effect of some of the main variables (pressure and power density) on the hydrogen microwave plasma characteristics at moderate pressure. A numerical model based on the fluid approach with a drift-diffusion approximation for the charged particles fluxes in two-dimensional cylindrical geometry has been developed. It was seen from the results obtained, that an increase in power density coupled to the plasma induces a significant increase in the electron and ion densities, which justifies clearly the important action of power density. It was also shown that the charged particles densities increase strongly as a function of the gas pressure, evidencing the strong effect of pressure on the production of electrons and ions in the plasma. It was also seen that the electric potential of the plasma varies slightly with the power density and the pressure; and that the maximum value of the potential is reached near the center of the plasma.
References


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