Numerical optimization of hydrogen microwave plasma reactor for diamond film deposition

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Abstract. We present the results of optimization of microwave plasma generation in a prototypical diamond film deposition device by means of numerical simulation. The modification of the device was done by changing the shape of microwave resonant chamber, where the plasma generation occurs, in a way to focus the discharge near the outlet nozzle. The best results were obtained for a configuration with an addition of a conducting rod located on the axis of the chamber. This modified configuration provided a two orders of magnitude increase in electron number density near the nozzle exit and about 10 time increase (up to 5%) in dissociation degree of hydrogen molecules.

1. Introduction

During the past decades, there has been much progress in the technology of surface coatings with various properties. One of the widely used methods in this industry is chemical vapor deposition (CVD). The diamond films which are obtained using this technology have a variety of unique properties such as high values of surface hardness, thermal conductivity and breakdown voltage. One important element to improve the efficiency of film deposition is high mole fraction of atomic hydrogen in the mixture, which plays a key role in a surface chemical reaction of transition from different molecular structures of carbon to diamond [1]. The usual way to increase the hydrogen dissociation degree is the use of electric discharge (ark, glow, or microwave) in the reaction chamber.

A microwave discharge has a certain advantage over the other discharge types as it allows focusing the discharge energy far from the electrode surface, which minimize the electrode erosion and prohibits the contamination of the substrate by the electrode material.

A new deposition method has recently emerged, in which an activated mixture formed in a microwave plasma chamber flows into a vacuum in a form of supersonic jet [2]. This method allows working with mixtures at near-atmospheric pressures in the discharge chamber. The current paper is aimed to develop an efficiency optimization strategy for such novel type of devices that are not yet deeply studied.

The microwave energy distribution in the chamber might be very inhomogeneous and depends on many parameters. The two main parameters which influence the local intensity of microwave heating are the chamber geometry and the distribution of the electrical conductivity inside the chamber. The electrical conductivity in the microwave gas discharge originates mainly from the free electrons distribution which rapidly changes during the ionization process. Optimization of the chamber geometry may lead to a higher stability of the discharge and focusing of the microwave energy in the region where the highest temperatures and electron concentrations are needed.
2. Computational details
In this paper, we present the results of optimization of the geometrical parameters of the microwave resonant chamber. A baseline simulation case was chosen to mimic an experimental setup from [2]. The setup consists of the upper chamber figure 1a, where the microwave port is located, and a lower discharge chamber separated by a 10 mm quartz plate. In the discharge chamber gas (H\textsubscript{2}) enters through the side wall inlets and after being ionized passes through the small outlet nozzle at the center of the bottom wall of the chamber. The diameter and height of the chamber were 100 mm and 105 mm, respectively. Such sizes were taken for the entire chamber to have a microwave resonance frequency of 2.45 GHz.

Figure 1. The case geometry (a). Numerical grids for the baseline (b), and modified (c) cases.

The simulations were conducted with the finite element modeling software COMSOL Multiphysics in axisymmetric formulation. This software package is well-suited for microwave plasma simulations [3,4]. The numerical grid is shown in figure. 1b,c. The grid was selected after the resolution study on a sequence of refined grids.

The problem under consideration consists of many interacting complex physical phenomena. The solution of all of them in a coupled formulation seems hardly possible. For that matter a simplified physical formulation of the problem was proposed, consisting only in plasma drift diffusion equations coupled with the microwave radiation equation for a hydrogen gas. The used electron impact reactions are listed in Table 1.

| Reaction                  | Type             | Cross-section data source |
|---------------------------|------------------|----------------------------|
| e + H\textsubscript{2} = e + H\textsubscript{2} | Elastic collision | [5]                        |
| e + H\textsubscript{2} = 2e + H\textsuperscript{+} | Ionization       | [6]                        |
| e + H\textsubscript{2} = e + 2H | Dissociation     | [6]                        |
| e + H = e + H\textsuperscript{+} | Elastic collision | [6]                        |
| e + H = 2e + H\textsuperscript{+} | Ionization       | [6]                        |
Such simplification assumes that all of the energy is transferred through the electron temperature, and only the electron impact reactions are the source of hydrogen dissociation. This might be true only for the initial stage of the discharge when the gas temperatures are still too low for the thermal dissociation. The gas flow is also not considered because the flow timescale is much larger than the timescale of electron number density dynamics. Since the flow inside the chamber is subsonic (i.e. slow compared to the charged species drift velocities), its effect on electron and ion dynamics was considered negligible.

The considered simplified formulation however is useful in the process of optimization of the discharge chamber design as it allows estimating the focusing efficiency of the microwave plasma heat source. The obtained data on the heating power deposition may be then used as an estimate for the heat source for the solution of thermal dissociation/heat transfer problem which is in plan for further research.

| Parameter | Description                  | Value  |
|-----------|------------------------------|--------|
| $p_0$     | Chamber Pressure (torr)      | 50     |
| $P_{dep}$ | Deposited microwave power (W)| 500    |
| $n_e^0$   | Initial electron density ($m^{-3}$) | $10^{15}$ |
| $T_e^0$   | Initial electron temperature (eV) | 1      |
| $n_{H^+}$ | Initial $H_2^+$ concentration | $10^{15}$ |
| $x_{H^+}$ | Initial H mole fraction      | $10^{-8}$ |
| $f$       | Microwave frequency (GHz)    | 2.45   |

Because of nonlinearity of the equations the simulations were conducted in a frequency transient formulation, i.e. in a time domain for drift-diffusion equations and in frequency domain for the microwave power deposition. The simulations were run until the equilibrium was reached between the microwave and drift-diffusion parts. The simulation parameters are presented in Table 2.

Figure 2. The microwave electric field distribution for the baseline (a) and modified (b) cases.

3. Results and discussion
The main idea of improving the efficiency of the microwave plasma jet device is to increase the microwave energy deposition locally near the nozzle, where the jet should be formed.

The baseline case discharge chamber configuration [2] is a simple cylindrical domain with conducting walls. In the beginning of simulation, before the ionization starts the chamber has a
uniform distribution of electrons. At that time the microwave electric field is concentrated near the bottom of the domain, where the outlet nozzle is located. However, when the equilibrium solution is reached, and the electron distribution become inhomogeneous (figure 2a), the microwave electric field acquires two maxima near the top and bottom walls of the chamber due to the gradients in electric conductivity created by free electron distribution (figure 3a). This reduces the magnitude of the field near the nozzle and hence the local heating intensity. The maximum of deposited power density distribution becomes shifted away from the bottom wall.

In experimental results such behaviour should be reflected in the lower stability of the discharge and lower operating pressures.

To improve the field focusing effect, a modification to the chamber is introduced by addition of a conducting rod at the axis of the chamber which is attached to the quartz layer and has a diameter of 6 mm (figure 1c). The rod has a hemispherical tip near the bottom wall which is lifted above the nozzle by 2 mm. Such a configuration was selected among several others as it shows the most effective field concentration near the outlet nozzle at the bottom wall.

For the modified case the microwave electric field is concentrated in a much smaller volume near the nozzle exit. The field value is about 5 times higher than in the baseline case. That leads to a significantly higher electron number concentration near the outlet nozzle. Figure 4 shows the increase in electron number density up to 100 times in that region. At the same time, in the middle of the chamber the electron concentration is lower than in the baseline case which is another effect of the field focusing. The electron temperature (figure 3c, d) is also higher in the modified case (up to 2 times 2.4 eV vs. 1.2 eV in unmodified case).

![Figure 3. The electron number density (a, b) and electron temperature (c, d) for the baseline (a, c), and modified (b, d) cases.](image-url)
The most important property for optimization is power deposition (i.e. heating of the mixture) by electron collisions. The power deposition is shown in figure 5c,d. With the proposed modification the deposited power density grows about one order of magnitude, form $10^6$ to $10^7$ W/m$^3$ in the near-nozzle region. This promises a significant effect on the heating of gas and consequent thermal dissociation.

The hydrogen dissociation degree from the electron impact reactions is low for the considered conditions, however, for the modified case it grows near the nozzle from 0.25% to 5.5% (figure 4b, figure 5b) that may become a sufficient addition to the thermal dissociation and increase the efficiency of the device.
Conclusion
The modification of a microwave activated CVD plasma jet device that increases the concentration of the microwave energy by means of plasma effects near the outlet nozzle of the jet is proposed. The modification consists of including a thin conducting rod that concentrates the microwave field near its tip. The effect of such modification on the ionization rate near the nozzle is quite significant and should improve the resulting plasma jet performance. While the simulation uses a very simplified approach with only electron impact reactions considered in the hydrogen gas, it still can provide useful information for the planned optimization of the experimental setup. The further experiments will provide more information on the modified device efficiency.

Acknowledgements
The research was carried out within the state assignment of FASO of Russia for Kutateladze Thermophysics Institute SB RAS, and with financial support from RFBR (project No. 18-29-19069/18).

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