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A novel microwave plasma reactor with a unique structure for chemical vapor deposition of diamond films

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ABSTRACT

With the aid of numerical simulation, a novel microwave plasma reactor for diamond films deposition has been designed. The new reactor possesses a unique structure, neither purely cylindrical nor purely ellipsoidal, but a combination of the both. In this paper, the design strategy of the new reactor together with a simple but reliable phenomenological simulation method will be described. Preliminary experiments show that uniform diamond films of high quality could be deposited using the new reactor, and the deposition rate of diamond films is typically about 3 μm/h at 6 kW input power level on a 2 inch diameter silicon substrate.

1. Introduction

Diamond is a near-perfect material possessing many remarkable properties and thus having a broad range of high-tech application. During the last decades, research has been conducted on synthesizing diamond films by various methods such as hot filament, DC arc plasma jet and microwave plasma chemical vapor deposition (MPCVD). Among these techniques, MPCVD is the first choice for depositing high quality diamond films [1], though the technique has the shortcoming of low deposition rate of diamond films.

In order to improve the MPCVD technique, different types of MPCVD reactors have been developed, including the early tubular type [2], the bell jar type [3–8], the cylindrical cavity type [8,9], the multimode non-cylindrical cavity type [10] and the ellipsoidal cavity type [11]. Along with this development, the input power to the MPCVD reactors as well as the deposition rate of diamond films has been greatly increased. Because in every type of the MPCVD reactors, quartz has been selected as the material for fabricating microwave-transmitting and vacuum-sealing windows, etching of the windows by plasma has remained a major consideration in designing or choosing a MPCVD reactor for depositing diamond films. For example, when a bell jar type [3–8] or a cylindrical cavity type [8,9] MPCVD reactor is used, the input power of the reactor will usually be limited to about 3 kW, because too high an input power will result in the etching problem. The multimode non-cylindrical cavity type [10] and the ellipsoidal cavity type [11] MPCVD reactors could be operated at higher input power levels, because in the former type of reactors, the quartz windows are located beneath the deposition stage, so that it could be protected from the plasma etching. In the latter type of reactors, the diameter of the quartz bell jar is comparatively large. In this case, plasma etching is less a problem.

Recently, Li et al. [12] from our group reported on using a newly designed MPCVD reactor for depositing diamond films. The reactor adopted a similar design for the quartz microwave window as in the multimode non-cylindrical cavity type reactor [10], so as to ensure that the reactor could be operated at high power levels. More importantly, the cylindrical geometry of the new reactor was very simple, so that incorporation of multiple adjustment mechanisms to the reactor became possible. The later characteristic is advantageous as it could facilitate optimization of plasma distribution during diamond film deposition process. Unfortunately, later experiments showed that when this reactor was used, diamond films deposited could be contaminated because during the diamond deposition process, deposit of carbon impurities was likely to appear on top of the deposition chamber, especially when the deposition time was long and the input microwave power was high.

To circumvent the problem, we propose in this paper a new design for the MPCVD reactor. This reactor possesses a unique resonator cavity structure, which is neither purely cylindrical, as in the case of the cylindrical cavity type reactor [8,9], nor purely ellipsoidal, as in the case of the ellipsoidal cavity type MPCVD reactor [11], but it is composed of both a cylinder and half an ellipsoid. Furthermore, in optimizing the design of the reactor, a simple but effective phenomenological simulation method is used, which does not need to deal with a large number of chemical reactions/particle collisions and solution of many complex differential equations. Comparison between simulation and experimental results will show that the behavior of plasma in the MPCVD reactor could be modeled by using the simple phenomenological model. In addition, experimental results will show that the proposed reactor has

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circumvented the contamination problem, and that the reactor has the ability to prepare high quality diamond films at high input power level during long time operation.

2. Simulation methods

In the following, the simulation methods will be described which involve three interconnected modules.

2.1. Module for microwave electric field simulation

Microwave electric field in the MPCVD reactor can be modeled by solving the following Maxwell's equation

$$\nabla \times \times \mu_r^{-1}(\nabla \times \times E) - k_0^2\left(\varepsilon_r - \frac{j\sigma}{\omega\varepsilon_0}\right)E = 0 \quad (1)$$

where E is microwave electric field, $k_0 = 2\pi/\lambda_0$ the wave number of microwave in free space, λ_0 the wavelength of microwave with a frequency of 2.45GHz, ω the microwave angular frequency, ε_0 the electric permittivity, σ the plasma conductivity, ε_r and μ_r the relative permittivity and relative permeability, respectively. Among these parameters, the relative permittivity ε_r and plasma conductivity σ should be determined for two different kinds of regions:

(1). In the region where no plasma exists

$$\varepsilon_r = 1 \text{ and } \sigma = 0; \quad (2)$$

(2). In the region where a plasma exists [13,14]

$$\varepsilon_r = 1 - \frac{\omega_p^2}{\omega^2 + \nu_e^2} \quad (3)$$

$$\sigma = \nu_e \frac{\varepsilon_0\omega_p^2}{\omega^2 + \nu_e^2.} \quad (4)$$

Here, $\omega_p{}^2 = e^2 n_e / (m_e\varepsilon_0)$ is the plasma frequency [15], e and m_e the charge and mass of an electron, n_e the electron density, and ν_e the electron collision frequency which can be calculated for pure hydrogen plasma by the following relation

$$\nu_e \approx a\frac{p}{T_g} \quad (5)$$

where p and T_g are gas pressure and gas temperature of the hydrogen plasma, respectively, and the constant $a = 1 \times 10^{10}$ K·Pa^{-1}·s^{-1} [15].

2.2. Module for plasma simulation

Compared to the simulation of the microwave electric field, it is more complicated to simulate plasma. However, Ref. [16] has introduced a simple phenomenological model to describe plasma by supposing that the plasma density at each point could be seen as a result of balance between various electron generation and loss terms

$$\nabla \cdot \left(-D_e \nabla n_e\right) + R_{vr} \cdot n_e^2 + R_a \cdot n_e = R_i \cdot E^2 \cdot n_e \quad (6)$$

where n_e is the electron density, D_e the ambipolar diffusion coefficient of electrons, R_i the ionization coefficient of gas molecules due to collisions with electrons, R_{vr} the coefficient of electron recombination, and R_a the coefficient of attachment of electrons to neutral particles. Obviously, by using above phenomenological description there will be no need to use the assumption that plasma density is proportional to local electric field [13,15] which has been believed as too strong simplification. Meanwhile, the simulation deals with no complicated chemical reactions and particle collisions [7], so there will be no need to solve a large number of differential equations [17,18]. By simply solving Eq. (6), plasma density in the MPCVD reactor would be phenomenologically described.

Because in the process of diamond films deposition, the main component of reaction gas is hydrogen, we can simplify the plasma simulation by considering only the pure hydrogen discharge. In addition, in Eq. (4), D_e will be replaced by diffusion coefficient of hydrogen ions D_i [19] whose value could be taken from Ref. [20]. Moreover, the recombination coefficient R_{vr} could be inferred from Refs. [21] and [22] as 10^{-13} m^3/s, while the attachment term $R_a \cdot n_e$ could be neglected [15]. To determine the value for the ionization coefficient of hydrogen molecules, R_i, a series of simulation was made using different values of R_i, and then comparison was made between simulated plasma distribution and experimentally observed plasma configuration. By this way, the value of R_i could be roughly estimated.

2.3. Module of gas temperature distribution

In order to ensure that the plasma be modeled in a correct way, inclusion of a module to describe gas temperature is necessary, since some parameters in the plasma simulation are functions of gas temperature, such as the electron collision frequency ν_e.

Assuming that the gas convection in the vacuum chamber could be ignored, gas temperature distribution could be calculated by solving the following energy conservation equation

$$\nabla \cdot \left(-k\nabla T_g\right) = Q \quad (7)$$

where k is the thermal conductivity of the gas, and Q is the energy source. In using Eq. (7), we have assumed that as the pressure in the deposition chamber is relatively high and the mean free path of gas molecules is short, energy obtained by electrons from the microwave field will be immediately transferred to gas molecules. In the simulation, the absorbed microwave power density by plasma, $Q_h = \frac{e^2 \upsilon_e n_e |E|^2}{2\left(\omega^2 + \upsilon_e^2\right)}$, has been taken as the energy source.

3. Design of the new MPCVD reactor

As has been mentioned above, during operation of the original MPCVD reactor shown in Fig. 1(a) there will be a serious problem that diamond films would be contaminated by impurities which were likely to deposit on the plunger of the deposition chamber. The plunger has been designed as a part of the adjustment mechanism of the microwave resonator cavity. In the following, new designs to circumvent the problem will be described.

3.1. The first modification

A schematic of the initial modification made on the original MPCVD reactor is shown in Fig. 1(b). Compared with the original design (Fig. 1(a)), it could be seen that the structure of the resonant cavity has been greatly changed. Different from a purely cylindrical appearance, the modified reactor has half an ellipsoid as its upper resonant cavity. In addition, the movable plunger in the original design has been removed, but an adjustment mechanism has been retained in the new design allowing regulation of the chamber height and the plasma distribution during the diamond films deposition by moving the ellipsoidal ceiling up and down.

Distribution of microwave electric field in the MPCVD chamber was modeled, firstly without consideration of plasma. In Fig. 2(a) and (b) are shown the results of such a pure electric field simulation, for the original and the modified reactor, respectively. From Fig. 2(a), it can be seen that in the original design, microwave energy may partly dissipate on the bottom of the plunger after being coupled into the chamber, causing carbon impurities deposition there. On the contrary, in the modified