The effect of substrate holder size on the electric field and discharge plasma on diamond-film formation at high deposition rates during MPCVD

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Abstract
The effect of the substrate holder feature dimensions on plasma density \(n_e\), power density \(Q_{\text{mw}}\) and gas temperature \(T\) of a discharge marginal plasma (a plasma caused by marginal discharge) and homogeneous plasma were investigated for the microwave plasma chemical vapor deposition process. Our simulations show that decreasing the dimensions of the substrate holder in a radical direction and increasing its dimension in the direction of the axis helps to produce marginally inhomogeneous plasma. When the marginal discharge appears, the maximum plasma density and power density appear at the edge of the substrate. The gas temperature increases until a marginally inhomogeneous plasma develops. The marginally inhomogeneous plasma can be avoided using a movable substrate holder that can tune the plasma density, power density and gas temperature. It can also ensure that the power density and electron density are as high as possible with uniform distribution of plasma. Moreover, both inhomogeneous and homogeneous diamond films were prepared using a new substrate holder with a diameter of 30 mm. The observation of inhomogeneous diamond films indicates that the marginal discharge can limit the deposition rate in the central part of the diamond film. The successfully produced homogeneous diamond films show that by using a substrate holder it is possible to deposit diamond film at 7.2 \(\mu\)m h\(^{-1}\) at 2.5 kW microwave power.

Keywords: MPCVD, marginal discharge, diamond film, deposition rate

1. Introduction
Diamond combines many outstanding properties that make it an attractive material for many applications [1–4]. Microwave plasma chemical vapor deposition (MPCVD) is a well-known method to produce high-quality diamond [5–12]. However, the high deposition cost due to the low deposition rate represents a major obstacle for large-scale commercial use [13, 14]. A high deposition rate for diamond films has always been one of the primary goals in diamond research [15, 16]. Some studies have indicated that high power density can be used to increase the growth rate significantly during MPCVD diamond deposition [17–20]. Studies [17] show that increasing the input microwave power with a corresponding increase in pressure in the chamber can simultaneously improve the area of the diamond films as well as the power density. In addition, some studies indicate that for low microwave power, a significant increase in chamber pressure


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can compress the plasma to increase its power density. The plasma density contributes greatly to the diamond-film growth rate [18]. In order to use this information, Hemawan [18] redesigned a new substrate holder to increase the power density, and Yamada [20] added a cylindrical conductor made of Cu to replace the top wall of the chamber. Recently, we found the power density could be increased by decreasing the substrate size. However, the decreasing substrate size can easily affect the uniformity of the diamond films. In order to research the cause, the effect of substrate holder shape and size (dimensions) on the electric field and plasma characteristics during diamond-film deposition is investigated in this paper.

We pay particular attention to any correlation with the appearance of a marginally inhomogeneous plasma. We also investigate a method to modify and control the marginally inhomogeneous plasma. All these important characteristics of the reactor, including the electric field strength of microwaves, electron density, power density, as well as gas temperature are studied using a phenomenological model for hydrogen plasma [21–23]. Our experimental results show that marginally inhomogeneous plasma has a significant influence on the diamond deposition result. The homogeneous discharge plasma can reach a high deposition rate after optimizing the plasma.

2. The simulation model and simulated conditions

Figure 1 is a schematic diagram of the MPCVD equipment with a microwave frequency of 2.45 GHz. Silicon wafers with a thicknesses of 3 mm and diameter of 60 mm are commonly chosen as substrates for diamond deposition. Figure 1 shows that the reactor consists mainly of a cylindrical cavity that serves as a microwave cavity with a quartz window. A coaxial antenna is located at the top of the cavity. Substrate holder II (with the height b2) is adjustable and located at the bottom of the chamber. There is a groove in the surface of substrate holder II to receive substrate holder I. Substrate holder I is divided into upper and lower layers, and the heights of these two layers are b4 and b3, respectively. The other scales of the chamber are shown in figure 1. For this reactor, a change of the substrate size affects both the electric field and the plasma redistribution [17]. The dimensions b2, b3, b4 as well as the substrate diameter need to be adjusted to eliminate changes of plasma state.

Microwave energy is transmitted into the chamber through the entrance. A microwave resonance occurs if the size of the MPCVD chamber is properly selected. When microwave energy is introduced into the chamber, it will be concentrated just above the substrate, where a hemispherically-shaped plasma will be ignited by the energy from the microwave in the presence of negative pressure. Diamond films are then deposited onto the substrate.

To study the influence of substrate holder size on the plasma characteristic, we use the phenomenological model of hydrogen plasma, which was reported by our group previously [21, 22]. It enables us to simulate important characteristics of the plasma, including the electric field strength of the microwave, electron density, power density, and gas temperature. Furthermore, the microwave power in all simulations is set to 2.5 kW, gas pressure to 15 kPa, temperature on the substrate surface to 1150 K, temperature of the substrate holder I surface to 850 K, temperature at the other parts to 320 K, the initial electron density to $7 \times 10^{17} \text{ m}^{-3}$, microwave frequency to 2.45 GHz, and the substrate
thickness to 3 mm. The substrate diameter is the same as the diameter of the top part of substrate holder I. All the scal conditions of simulation are shown in table 1. A more detailed operating principle, parameters, and boundary conditions can be found in earlier studies [21–23].

3. Results and discussion

3.1. The effect of substrate holder dimension in the radial direction on the electric field and plasma characteristics

Before studying the effect of substrate holder characteristics on the plasma, the effect of substrate holder characteristics on the electric field distribution needs to be better understood. Figure 2(a) shows the electric field strength of the reactor obtained for a vacuum without plasma when diamond (within a diameter of 60 mm) is deposited in the MPCVD cavity. In this case, b2, b3, and b4 are 10, 19, and 9 mm, respectively. Figure 2(a) shows that the electric field strength is comparatively low and the distribution of the electric field is relatively large. As a result, the effect of the electric field igniting plasma is not strong. The plasma is affected by the substrate in the MPCVD chamber. More precisely, above a certain size of the substrate, there must be a large plasma area to keep the power density relatively low. This is the reason for the typical low deposition rate of diamond films for large-area substrates with low microwave power [18, 24, 25]. Figure 2(a) shows that there is a 30 mm diameter area in the central region, which has a higher electric field strength than the fringe region. The distribution of the electric field is considered to be homogeneous. The electric field strength increases, and the maximum electric field strength appears in the fringe region of the substrate when changing the diameter of the top part of substrate holder I to 30 mm—see figure 2(b). The electric field adheres to the substrate and the substrate holder. The electric field strength increases further if the top part of substrate holder I has a cylindrical shape. In other words, the decrease of the substrate holder dimension in the radial direction increases the electric field strength. However, the maximum electric field strength gradually shifts to the peripheral (i.e. marginal) region of the substrate. We refer to this phenomenon as ‘marginal discharge’—see figure 2(c). For the same reason, the volume increase reduces the electric field strength and also decreases the maximum electric field strength in the fringe region of the substrate. Figure 2 shows that the substrate attracts the electric field. When the relatively large electric field (corresponding to the 60 mm diameter substrate) generated above a small-sized substrate (corresponding to 30 mm diameter substrate) increases the electric field strength, the increasing electric field can easily produce the marginal discharge. In addition, the plasma distribution correlates with the electric field [17], and the marginal discharge can affect the uniformity of the plasma. Peripherally inhomogeneous plasma is also referred to as marginally inhomogeneous plasma in this paper.

To study the effect of the substrate holder dimension in the radial direction on the plasma, we used the phenomenological model for hydrogen plasma to simulate the plasma state—see figure 3. Figure 3 shows the electron density \(n_e\), power density \(Q_{\text{pow}}\), and the gas temperature \(T\) when the top diameters of substrate holder I are 60 and 30 mm, respectively. Figure 3 shows that the maximum electron density as well as the power density shift into the peripheral

### Table 1. Scale conditions of simulation used in figures 2–6.

<table>
<thead>
<tr>
<th>Figure</th>
<th>b2 (mm)</th>
<th>b3 (mm)</th>
<th>b4 (mm)</th>
<th>Substrate diameter (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>2</td>
<td>10</td>
<td>19</td>
<td>9</td>
<td>—</td>
</tr>
<tr>
<td>3</td>
<td>10</td>
<td>19</td>
<td>9</td>
<td>60 ((a1–a3)/30 ((b1–b3))</td>
</tr>
<tr>
<td>4</td>
<td>10</td>
<td>19</td>
<td>1–10</td>
<td>30</td>
</tr>
<tr>
<td>5</td>
<td>10</td>
<td>16–23</td>
<td>5</td>
<td>30</td>
</tr>
<tr>
<td>6</td>
<td>8–16</td>
<td>14</td>
<td>5</td>
<td>30</td>
</tr>
</tbody>
</table>

Figure 2. Electric field intensity at 2.5 kW for various substrate holder structures are obtained for a vacuum without plasma. (a) Cylinder with 60 mm diameter, (b) circular truncated cone with 30 mm (top) and 60 mm (bottom) diameter, respectively, and (c) cylinder with 30 mm diameter.
area, and marginally inhomogeneous plasma develops, which correlates with the distribution of the electric field—see figure 3. Figures 3(a1) and (b1) show that the maximum electron density increases from $3.2 \times 10^{17}$ m$^{-3}$ to $4.6 \times 10^{17}$ m$^{-3}$ after decreasing the substrate holder dimension in the radial direction. Figures 3(a2) and (b2) show that the maximum power density increases from 28.2 to 66.1 W cm$^{-3}$. It is clear that the increase of the electron density and the power density increases the diamond deposition rate [26, 27]. Figures 3(a3) and (b3) show that the plasma gas temperature increases when the substrate holder dimension in the radial direction decreases. Although the numerical simulation aims to maximize electron density, and power density in the MPCVD equipment, the peripheral discharge affects the uniformity of the diamond films [27]. Therefore, it is necessary to study systematically the effect of the substrate holder structure on the plasma.

3.2. The effect of substrate holder dimension in the axis direction on the plasma characteristics

Based on the analysis of our results in section 3.1, the effect of the substrate holder dimension in the axis direction on the plasma characteristics was studied using the simulation described here. The values of plasma density, power density, and gas temperature in the following figures are the maxima within the plasma area.

Figure 4 shows the effect of $b_4$ (substrate holder I) on the plasma. Clearly, when $b_4$ increases from 1 to 9 mm, the electron density continues to rise. However, the marginally inhomogeneous discharge begins to appear in the fringe region of the substrate when $b_4$ is about 6 mm. The intensity of the marginally inhomogeneous plasma begins to decrease in the fringe region of the substrate when $b_4$ is about 6 mm. The power density shows a similar behavior. The results of the physical simulation are shown in figure 4. The gas temperature reaches a maximum around $b_4 = 5$ mm, and
decreases when marginal discharge appears. There is also no marginally inhomogeneous plasma when $b_4$ is between 1 and 5 mm. Considering these dependencies, we can try to reduce $b_4$ to eliminate the inhomogeneous plasma. Knowing the strong inhibiting effects of marginally inhomogeneous plasma, the following values could not be simulated if marginal discharge occurred.

Figure 4 shows that when $b_4$ increases from 1 to 5 mm, the electron density and power density can reach a local maximum with a homogeneous state. In order to systematically study the effect of the substrate holder dimension (in the direction of the axis) on the plasma characteristics, we investigated what effect $b_3$ has on the electron density, power density, and gas temperature—see figure 5. When $b_2$ is 10 mm and $b_4$ is 5 mm, both the electron density and power density increase when $b_3$ increases from 16 to 23 mm. When $b_3$ is 22 mm, both the electron density and power density reach their highest values, $4.11 \times 10^{16} \text{ m}^{-3}$ and $61.9 \text{ W cm}^{-2}$, when the plasma is uniform. When $b_3$ continues to increase to 23 mm, the marginally inhomogeneous plasma arises again and the electron density and power density increase. The temperature decreases when marginal discharge appears.

In conclusion, the increase of dimensions in the axis direction can easily produce marginally inhomogeneous plasma. Assuming there is no marginally inhomogeneous plasma, the variation of the electron density, power density, and temperature after decreasing $b_4$ can be tuned by adjusting $b_3$ (the height of substrate holder I). All these can be summarized into one hypothesis: while the changes of electron density, power density, and temperature, are randomly triggered after decreasing $b_3$ and $b_4$, the plasma state can be tuned to produce the state shown in figure 5 by increasing $b_2$.

### 3.3. Tunability of the movable substrate holder

In order to validate the hypothesis in section 3.2, we set $b_3$ and $b_4$ to 14 and 5 mm, respectively. We then simulated the effect that the variation of $b_2$ has on electron density, power density, and temperature—see figure 6. Figure 6 reveals that, when $b_3$ and $b_4$ are 14 and 5 mm, respectively, the marginally inhomogeneous plasma is avoided if $b_2$ is within 8 and 15 mm. This plasma state is desirable for the deposition of homogeneous diamond films. However, the electron density and power density show a tendency to increase if $b_2$ increases. To assure the uniformity of the plasma, the electron density and the power density reach maximum at $40.7 \times 10^{16} \text{ m}^{-3}$ and $61.5 \text{ W cm}^{-2}$, respectively, when $b_2$ is 15 mm. After that, when $b_2$ is 16 mm, the marginally inhomogeneous plasma appears again. Figure 6 shows that the temperature increases first and then decreases—similar to figures 4 and 5. However, although the plasma is comparatively uniform, the average deposition may not be the desired rate, because the power density is not the highest when $b_2$ is 10 mm. To achieve the maximum deposition rate, while considering the uniformity of diamond films as well, substrate holder II should be adjusted to 15 mm where marginal discharge plasma could be avoided and the power density is high. This method to avoid the marginal discharge is different from the method in [27].

In summary, we conclude that the substrate holder dimension in the axis direction, $b_3$ and $b_4$, can be combined for different values. The change in the plasma state resulted from the different combinations of substrate holder heights, $b_3$ and $b_4$, and can be improved by adjusting movable substrate holder II, i.e. by adjusting the height of substrate holder II $b_2$. In addition, by altering $b_2$ we could modify the uniformity of the plasma and electron density distribution, as
well as the power density, which optimizes the power density and improves the deposition rate to be as high as possible. Furthermore, substrate holder II could be adjusted when marginally inhomogeneous plasma happened to occur and its height was reduced by 1 mm. As a result, homogeneous diamond films can be produced with a higher deposition rate.

### 3.4. Validating the simulation with experiments

In order to validate the reliability of the simulation, a substrate holder with \( b_3 = 14 \text{ mm} \) and \( b_4 = 5 \text{ mm} \) was designed and manufactured. The experiments involving marginally inhomogeneous plasma for diamond deposition were conducted with this substrate holder. The value of \( b_2 \) used in the experiment with a marginally inhomogeneous plasma was 16 mm. The diamond film deposited with this substrate holder is marked as sample I. Marginally inhomogeneous plasma appears already at the beginning of the experiment. By contrast, the value of \( b_2 \) used in the experiment with homogeneous plasma is 15 mm. The diamond film deposited by this substrate holder is marked as sample II. In order to ensure no marginally inhomogeneous plasma throughout the entire deposition, the deposition time of sample II is 45 h. Otherwise, the thicker diamond film might influence the distribution of plasma and the simulation results cannot be validated, because the thickness of the diamond film increases with time. The deposition conditions for these two experiments are listed in Table 2.

To evaluate the structure and quality of the diamond film, the sample was characterized using a scanning electron microscope (Quanta 450, FEI) and a Raman spectroscope (Raman-11, Nanophoton Corporation). For the Raman spectroscopy measurement, an Ar-ion laser, operated at 514.5 nm, was used.

Figure 7 shows the diamond films deposited using the technological parameters listed in Table 2 and their corresponding fracture morphology. Figure 7(a) shows the diamond film prepared using a marginally inhomogeneous plasma. The fringe region of the diamond film is thicker than in the central area. The fracture morphology of the films shown in Figure 7(a) suggests that the maximum thickness of the fringe region is 1.16 mm (corresponding to a diamond deposition rate of about 11.6 \( \mu \text{m h}^{-1} \) ) and the minimum thickness of the central area is 611.7 \( \mu \text{m} \) (corresponding to a diamond deposition rate of about 6.1 \( \mu \text{m h}^{-1} \) ). The difference in the deposition rate between the fringe region and central region can be explained as follows. The marginally inhomogeneous plasma increases the power density locally along the

<table>
<thead>
<tr>
<th>Sample</th>
<th>( b_2 ) (mm)</th>
<th>Power (kW)</th>
<th>Pressure (kPa)</th>
<th>( \text{CH}_4/\text{H}_2 ) flow rate (sccm)</th>
<th>Temperature (°C)</th>
<th>Time (h)</th>
</tr>
</thead>
<tbody>
<tr>
<td>I</td>
<td>16</td>
<td>2.5</td>
<td>15</td>
<td>18/300</td>
<td>920</td>
<td>100</td>
</tr>
<tr>
<td>II</td>
<td>15</td>
<td>2.5</td>
<td>15</td>
<td>18/300</td>
<td>920</td>
<td>45</td>
</tr>
</tbody>
</table>

Figure 7. Photos of freestanding diamond film (a1) and (b1) and the cross-sectional morphology (a2) and (b2) of sample I and II, respectively.
Figure 8. Raman spectra of sample I and II.

edge of the substrate, but the microwave power fed into the cavity is constant. Therefore, the higher power density limits the power density in the central region. Figure 7(b1) shows the diamond film prepared using the homogeneous plasma. The uniform thickness is clearly visible in figure 7(b2). The thickness of the diamond film is 325 μm (which corresponds to a diamond deposition rate of approximately 7.2 μm h⁻¹, which is fast for 2.5 kW microwave power). The deposition rate is higher than in sample I. The non-uniformity in the sample’s thickness is less than 3%. This indicates that homogeneous plasma can ensure the uniformity of diamond film, and a suitable substrate holder dimension can improve the deposition rate.

To characterize the quality of samples I and II for the high deposition rate, Raman scattering spectra were recorded—see figure 8. We find high-intensity diamond Raman peaks around 1331.8 and 1332.8 cm⁻¹. The corresponding full widths at half maximum of the diamond peaks are approximately 4.5 and 5.5 cm⁻¹. Our results suggest that the diamond films are of high quality despite the high deposition rate.

4. Conclusions

In this paper, the effect of substrate holder parameters on the produced plasma was simulated using a phenomenological model for hydrogen plasma. The results show that the size parameters of the substrate holder affect the plasma characteristics significantly. The decrease of dimensions in the radial direction and increase of dimensions in the axis direction can produce marginally inhomogeneous plasma. The marginally inhomogeneous plasma can be controlled i.e. tuned. The formation of a homogeneous plasma and optimized power density could be accomplished by changing the height of the movable substrate holder. During diamond-film deposition, the marginally inhomogeneous plasma could reduce the deposition rate in the central region, while diamond-film growth with a deposition rate of up to 7.2 μm h⁻¹ (at 2.5 kW) could be achieved using homogeneous plasma.

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