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# Self-enhanced plasma discharge effect in the deposition of diamond-like carbon films on the inner surface of slender tube

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#### **1. Introduction**

Recently, internal plasma processing of narrow halls and tubes are strongly desired in industry and a lot of efforts have been made in this filed  $[1-7]$ . As an emerging technique on inner surface films deposition, enhanced glow discharge plasma immersion ion implantation and deposition (EGD-PIII&D) has been intensively investigated both experimentally and theoretically  $[8-11]$ . EGD-PIII&D is conducted using a special hardware configuration composed of a small pointed hollow anode and large tabular cathode. In contrast to other techniques [\[4–7\],](#page--1-0) the EGD-PIII&D is cost-effective and easy to be installed. Meanwhile, it is high efficient to conduct the deposition of diamond-like carbon (DLC) films on the inner surface of the quartz tube with an inner diameter as small as 0.9 mm and length-to-diameter ratio of over 100 [\[10\].](#page--1-0) The DLC film deposition rate under this process can reach as high as 1.3  $\mu$ m/min with minor destruction on the substrate [\[10\].](#page--1-0) These unique characteristics introduce the EGD-PIII&D technique 10 times faster than traditional methods to deposit DLC films [\[12–15\].](#page--1-0) As noted previously [\[8\],](#page--1-0) such high deposition rate was explained to be attributed to the high ionization degree of neutral particles resulting from the short electron mean free path. However, as noticed in this study,

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#### A B S T R A C T

Enhanced glow discharge plasma immersion ion implantation and deposition (EGD-PIII&D) have been proved to be highly effective for depositing diamond-like carbon (DLC) films on the inner surface of the slender quartz tube with a deposition rate of 1.3  $\mu$ m/min. Such a high-efficiency DLC films deposition was explained previously as the short electrons mean free path to cause large collision frequency between electrons and neutral particles. However, in this paper, we found that the inner surface material of the tube itself play a vital role on the films deposition. To disclose the mechanism of this phenomenon, the effect of different inner surface materials on plasma discharge was experimentally and theoretically investigated. Then a self-enhancing plasma discharge is discovered. It is found that secondary electrons emitted from the inner surface material, whatever it is the tube inner surface or deposited DLC films, can dramatically enhance the plasma discharge to improve the DLC films deposition rate.

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the implantation current was kept increasing continuously during the whole process of EGD-PIII&D to deposit DLC films, especially in the first 30 s. This makes us realize that the previously-determined mechanism may be deficient. By considering that during the deposition of DLC films on the inner surface of a slender tube, the electron mean free path inside the tube is much less than the length of the tube but still much larger than the inner diameter of the tube. This would not only produce a large amount of electron-neutral particle collisions but also lead to high frequency of electrons-inner surface impact. Given the progress being made, however, there is still a lack of systematic research on studying the effect of the electroninner surface impact on deposition of DLC films during the process of EGD-PIII&D.

Following this line of thought, the present work aims to elucidate the effect of inner surface materials of the slender insulating tube on plasma discharge. First, the implantation currents in the EGD-PIII&D with tubes made of different materials were measured. Based on the experimental results, the numerical simulations was then conducted to obtain the pressure gradient distribution and the ion density in the experimental region. Subsequently, the electron mean free path was introduced to explore how the inner surface materials affect the plasma discharge. Based on the combination of experimental, simulative and theoretical study, a self-enhanced plasma discharge effect was discovered. We found that the inner surface material, whatever it is the tube inner surface or deposited









**Fig. 1.** Schematic of the EDG-PIII &D system for inner surface treatment of tubes.

films, themselves can enhance the plasma discharge to improve the DLC films deposition rate.

### **2. Experimental setup and simulation details**

## 2.1. Experimental details

The schematic diagram of the EGD-PIII&D system was depicted as Fig. 1. The whole EGD-PIII&D system is inside the vacuum chamber. The base pressure is  $2 \times 10^{-3}$  Pa. As described in [\[10\],](#page--1-0) in brief, a glass chamber with a height of 222 mm and an inner diameter of 174 mm was placed on an aluminum substrate. The insulating tube was pre-cleaned ultrasonically in a deionization water bath, then rinsed in ethanol, and finally inserted into the central hole at the top of the glass chamber after dried up. The inside of the glass chamber is pumped through the interstices between the aluminum plate and the glass chamber. To investigate the effect of the inner surface material of the tube on plasma discharge, two experiments were designed and conducted: In Experiment I, acetylene  $(C<sub>2</sub>H<sub>2</sub>)$ gas was bled into the glass chamber through the tube at a flow rate of 10 sccm. The length of the tube was 100 mm, and the outer and inner diameters were 7 mm and 0.9 mm respectively. The material of the tube was quartz glass with main component of  $SiO<sub>2</sub>$ ; In Experiment II, two tubes with different materials were used. One of the tubes was made of quartz glass, while the other was made of lucalox with main component of  $Al_2O_3$ . Both of the tubes had 6 mm inner diameter and 8 mm outer diameter with the length of 100 mm. Instead of  $C_2H_2$ , Argon (Ar) gas was used with varied flow rates of 20 sccm, 30 sccm, 40 sccm and 50 sccm. In both experiment I and II, a negative voltage of 10 kV with a pulse duration of 100  $\mu$ s and repetition rate of 50 Hz was applied to the aluminum substrate after the gas pressure inside the vacuum chamber reached a stable state. As shown in Fig. 1, the positive electrode of the high voltage pulse power supply was connected to ground via a resistor (R=12  $\Omega$ ) in order to measure the implantation current. The resulting traces were recorded by a digital oscilloscope (Agilent Technologies DSO-X 2024A). In order to compare the implantation currents, the values of implantation currents at 99  $\mu$ s were taken to calculate the difference in currents.

#### 2.2. Description of the simulation model

Numerical simulation based on the Particle-In-Cell/Monte Carlo (PIC/MC) method was performed. Following the simulation pro-



**Fig. 2.** Schematic diagram of the EGD-PIII&D simulation model.

cedure described elsewhere [\[16–18\],](#page--1-0) a 3D Cartesian coordinate system in the x-y-z space was adopted and the schematic diagram of the PIC/MC simulated region was depicted. As shown in Fig. 2, the simulated region covers the x-y-z space with a volume of 228  $\times$  228  $\times$  360 mm<sup>3</sup>. In accordance with the experimental conditions, an initial argon plasma density of  $10^{15}$  m<sup>-3</sup> is adopted and represented by 175444 PIC particles equally distributed in the glass chamber and the tube in the initial stage. The collisions between electrons and neutral particles were handled by a Monte Carlo algorithm based on the efficient null-collision method [\[19–21\].T](#page--1-0)hree types of the collisions were considered, including elastic scattering  $(e + Ar \rightarrow e + Ar)$ , excitation  $(e + Ar \rightarrow e + Ar^*)$  and ionization  $(e + Ar)$  $Ar \rightarrow e + e + Ar^{+}$ ) [\[22–25\].](#page--1-0) The incident electron energy dependence of secondary electron emission coefficients of  $SiO<sub>2</sub>$  [\[26\]](#page--1-0) and  $Al_2O_3$  [\[27\]](#page--1-0) could be found. In the numerical simulation based on PIC/MC, the negative volatge was set as 10 kV including a rise time of 1  $\mu$ s, the same as the starting period of the pulse in our experiment. However, to shorten the computation period, we only calculated the first 10  $\mu$ s which we think it was enough to schematically show the development of the plasma. As the mass of the ion was much larger than that of the electron, the time step was set

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