Numerical Simulation on $Ar/CH_4/H_2$ Induction Thermal Plasma Field with Triangular Power Modulation at Reduced Pressures for Diamond Film Growth

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Abstract: The influence of triangular modulated input power waveforms was numerically on temperature field and feedstock CH_4/H_2 gas transport in modulated induction thermal plasma (MITP) to consider the suitable modulated input power waveforms for diamond film growth. Results indicated that the radical particle fluxes contributing diamond film growth on the substrate holder increased by highly modulating input power waveforms.

Keywords: Thermal plasma, Diamond, Modulation.

1. Introduction

It is well known that the diamond has attractive excellent features of highest hardness, high thermal conductivity of 209 W/(m K), and also a wide band gap of 5.47 eV, which is useful for the next generation high-voltage semiconductor material[1]. Therefore, it is still desired to develop an effective method for diamond growth. Microwave plasma CVD is widely used for diamond growth. However, microwave plasma offers a relatively small area of diamond growth and then a larger-area growth method is necessary to be developed with high growth rate[2]. The authors have been investigating the application of RF modulated induction thermal plasma (MITP) to diamond film growth. This MITP offers time-controlled heat flux and time-controlled radical particle flux on the substrate holder[3]. Our previous research showed that irradiation of $Ar/CH_4/H_2$ plasma at reduced pressures increased the growth rate of diamond film in the experiments[4]. In present study, we numerically calculated the timevarying temperature and gas flow fields, and CH_4/H_2 mass fraction distribution with changing the modulation waveform. In addition, radical particle fluxes above the substrate holder were estimated to consider the relation between the radicals contributing diamond film growth and input power waveforms at a reduced pressure.

2. Numerical model

2.1 Calculation space and calculation condition

Figure 1 depicts MITP torch configuration. The MITP torch consists of a quartz tube, a 8-turn RF coil and a water-cooled metallic tube for feedstock gas feeding and a Mo substrate holder. The RF coil current generates a induction plasma inside the torch. The coil current can be modulated following a given waveform. The substrate holder is located at 40 mm below the coil end. For diamond growth, a monocrystalline diamond substrate is placed on the substrate holder. For this MITP torch, the calculation was made to obtain the temperature and gas flow field.

Figure 2 shows a schematic cross-sectional view of the plasma torch (z < 330 mm) and the chamber (z > 330 mm). The wall temperature was fixed at 300 K. Argon was supplied from the end of the torch (z = 0 mm) at 20 slpm each in the axial and swirl directions. The water-cooled tube was inserted from the top of the torch to the axial position z = 160 mm. Through this tube, the feedstock CH_4/H_2 gas was supplied with a flow rate of 0.02/2 slpm as a feedstock gas. The pressure in the plasma torch and chamber was set to 20 torr. Figure 3 shows the definition of modulation waveform in input power. The triangular modulation waveform in input power was defined by the ratio of rising time to cycle (RR) and the ratio of maximum peak power to minimum peak power (SPCL). Higher RR leads to saw-tooth waveform, and lower SPCL corresponds to higher modulation degree in input power. In the present work, input coil power was modulated in 25 triangular waveforms.

2.2 Calculation equations

For different RRs and SPCLs, the temperature and gas flow fields, and CH_4/H_2 mass fraction distribution in MITP were numerically obtained. For the calculation space, the coupled equations of conservation of mass, momentum, and energy were solved as well as Maxwell's equation, and the transport equation of CH_4/H_2 gas on the assumption of

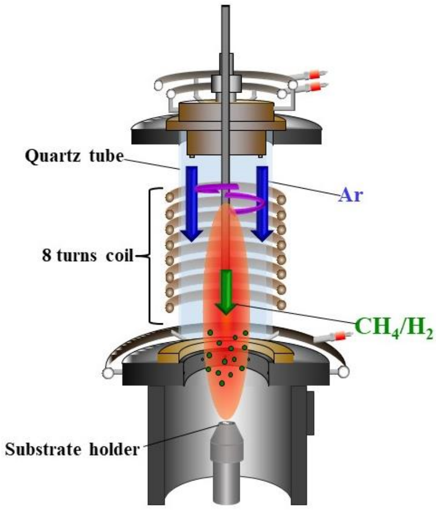


Fig. 1. MITP torch for diamond growth

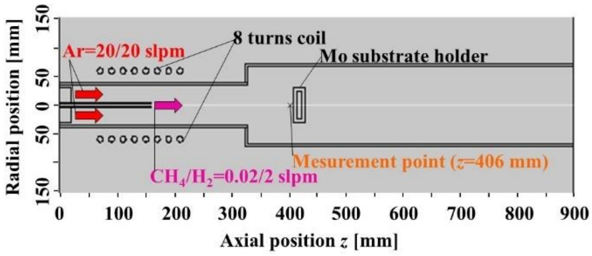
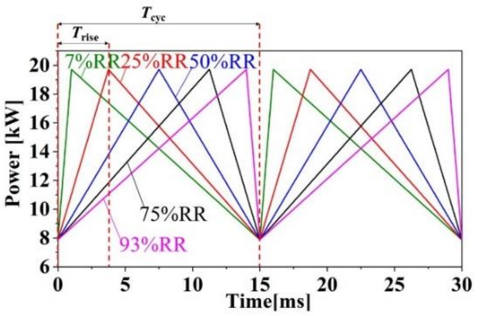
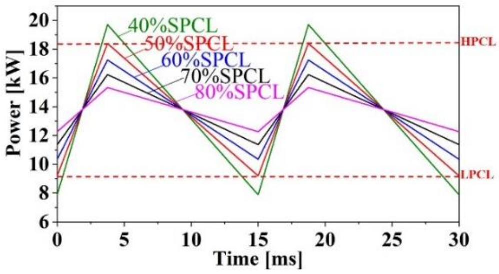


Fig. 2. Calculation space



(a) RR = 7, 25, 50, 75, 93%, SPCL = 40%



(b) RR = 25%, SPCL = 40, 50, 60, 70, 80%

Fig. 3. Triangular modulated input power waveforms

local thermodynamic equilibrium for simplicity.

This calculation requires the thermodynamic and transport properties of the plasma. The thermodynamic and transport properties were calculated as a function of temperature and CH_4/H_2 mass fraction using the calculated equilibrium composition and the collision integrals between species by the first-order approximation of Chapman-Enskog method.

Then, the particle fluxes of each of species on the substrate (z = 406 mm) were also calculated by using the numerically calculated temperature, gas flow velocity, and mass fraction of CH_4/H_2, and the calculated equilibrium particle composition.

3. Calculation results

3.1 Temperature and CH_4/H_2 mass fraction

Figures 4 and 5 depict the calculated results for temperature and CH_4/H_2 mass fraction fields in the MITP at RR=93% and SPCL=40%, respectively. The condition RR=93% corresponds to almost a saw-tooth power modulation waveform condition. From the time t=276 ms to t=282 ms, the instantaneous input power increases linearly in this case. As seen in this figure, the high-temperature plasma is present between the coil and the substrate holder. As the instantaneous input power increases, the temperature rises especially near the substrate holder around axial position of 300 mm < z < 400 mm. This indicates that modulation can change heat flux onto the substrate. Following the temperature variation, CH_4/H_2 mass fraction distribution also changes as shown in Fig.5. Feedstock gas CH_4/H_2 is supplied from the tip of the water-cooled feeding tube on the axis from the top of the plasma torch. At t=276 ms, CH_4/H_2 mass fraction is high above 0.85 between the tip of the water-cooled feeding tube at z=160 mm and z=370 mm. At t=279 ms, the region of high CH_4/H_2 mass fraction reaches to the substrate holder at z=410 mm. Then at t=282 ms, the high mass fraction region of CH_4/H_2 is present only near the tip of the feeding tube around 160 mm < z < 240 mm.

Temperature and CH_4/H_2 mass fraction at the speficied position changed cyclically versus time. Figure 6(a) shows the time evolutions in temperature above the substrate holder (z = 406 mm). As seen, the temperature changes with time following the power modulation. The modulation amplitude in temperature depends on the SPCL. At SPCL=40%, the temperature varies markedly from 3600 K to 8700 K. This indicates that SPCL can control instantaneous heat flux onto the substrate. The time evolutions in CH_4/H_2 mass fraction above the substrate holder (z = 406 mm) can be seen in Fig.6(b). The CH_4/H_2 mass fraction also changes with time following the power modulation depending on SPCL.This implies that particle flux of each of species can be controlled by the power modulation.

3.2 Particle fluxes onto the substrate

The particle fluxes onto the substrate were evalvuated from the calculated temperature, gas flow velocity and CH_4/H_2 mass fraction at the substrate surface, and the particle composition data. Figure 7 shows the time change in C_2 and C_2H particle fluxes above the substrate holder (z = 406 mm) as examples. The neutral particles C_2 and C_2H

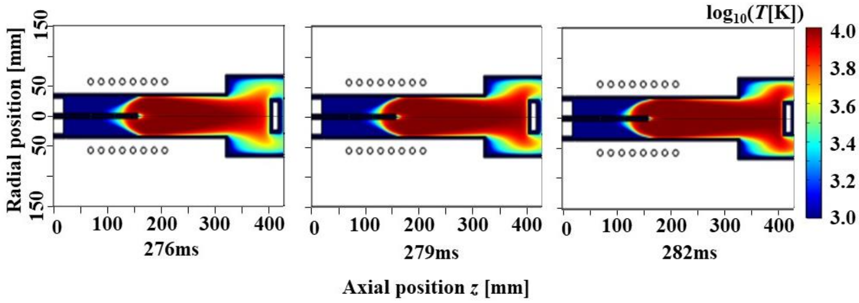


Fig. 4 Time variation in temperature distribution in MITP at RR=93% and SPCL=40%.

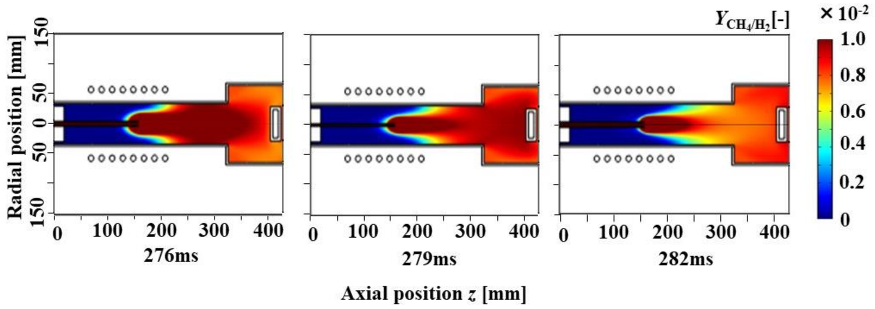


Fig. 5 Time variation in CH_4/H_2 mass fraction distribution in MITP at RR=93% and SPCL=40%.

are known as key species for diamond growth. As seen in this figure, both instantaneous C_2 and C_2H fluxes change significantly with time following the power modulation. Furthermore, their value change can be varied by SPCL. Lower SPCL increases the amplitude in modulation of C_2 and C_2H fluxes. This indicates that irradiation radical fluxes can be controlled by the power modulation.

In order to study the fluxes onto the substrate versus SPCL, the time-averaged flux for each of species were calculated per cycle for comparison. Figure 8 presents the time-averaged C_2 and C_2H fluxes above the substrate holder (z = 406 mm) as functions of SPCL and RR. From the point of view of SPCL, the time-averaged fluxes of C_2 and C_2H have a tendency to increase as SPCL of modulated input power decreased. Lower SPCL corresponds to higher modulation degree. On the other hand, from the point of view of RR, time-averaged flux of C_2 and C_2H had only a slight change by changing RR. This result shows that the fluxes contributing diamond film growth such C_2, C_2H can increase as SPCL of modulated input power waveforms decreases. In other words, highly modulated input power waveforms contributes the flux contributing diamond film growth. The reason for SPCL dependent fluxes was considered that highly modulated input power waveforms causes dissociation of CH_4/H_2 feedstock gas and effective transport of dissociated radicals onto the substrate.

4. Conclusion

This paper numerically studied the influence of triangular modulated input power waveforms on temperature field and feedstock CH_4/H_2 gas transport in MITP for diamond film growth. Results indicated that the radical particle fluxes contributing diamond film growth on the substrate holder increased by highly modulating input power waveforms.

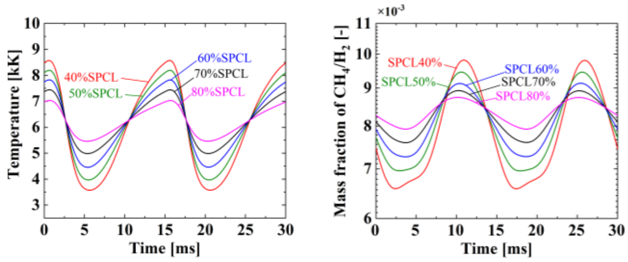
5. Reference

[1] N.Donato, *et al.*, *J.Phys.D:Appl.Phys.*, **53**(9), 093001(2020)

[2] H. Yamada, *et al.*, *Appl. Phys. Exp.*, **3**, 051301(2010)

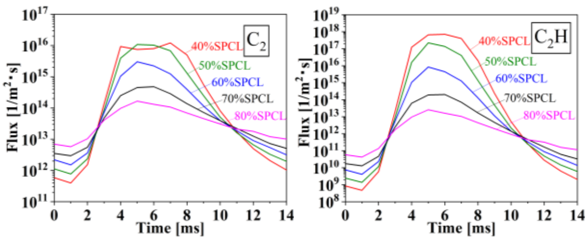
[3] Y. Tanaka, *et al.*: *Appl.Phys. Let.* **90**, 071502 (2007)

[4] K. Hata, *et al.*: *J.Appl.Phys.*, **126**, 223302 (2019)



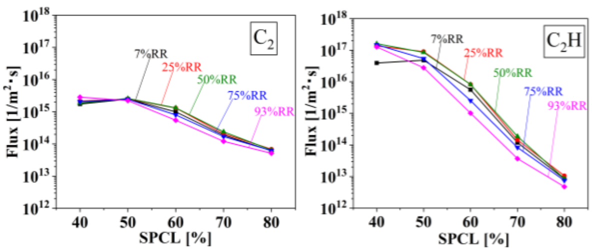
(a)Temperature (b)Mass fraction of CH_4/H_2

Fig. 6. Temperature and mass fraction of CH_4/H_2 above the substrate holder (z = 406 mm) versus time (RR = 93%, SPCL = 40, 50, 60, 70, 80%)



(a)C_2 (b)C_2H

Fig. 7. Time change in C_2 and C_2H neutral particle fluxes above the substrate holder (z = 406 mm) (RR = 93%, SPCL = 40, 50, 60, 70, 80%)



(a)C_2 (b)C_2H

Fig. 8. Time-averaged flux of neutral particle above the substrate holder (z = 406 mm)