A Three Dimensional Two-Temperature Model of Loop-Type of Ar Inductively Coupled Thermal Plasmas for Large-Area Materials Processing

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Abstract:
A three dimensional two-temperature model was developed for a loop-type of inductively coupled thermal plasma (loop-ICTP). The loop-ICTP has uniquely been developed by ourselves for large-area materials processing, and a part of loop-ICTP is formed on a substrate directly with a linear shape. The developed model solves the mass, momentum and energy conservation equations for heavy particles, to derive the heavy-particle temperature and gas flow fields. In addition, energy conservation equation for electrons and Poussin's equations for vector potential were also solved to obtain the electron temperature and electromagnetic fields. This simulation can provide the spatial distributions of heavy temperature in the loop-tube and on the substrate.

Keywords: Thermal plasma, loop tube, Large area materials processing, Alternating quenching gas

1. Introduction

The inductively coupled thermal plasma (ICTP) has some advantages of high gas temperatures and high enthalpy, and then high radical density compared to the low-temperatures plasma. In addition, the ICTP has no contamination from the electrode impurity because it does not need electrodes. From these advantages, the ICTP has been widely used for various materials processing such as nanoparticle synthesis, waste destruction, etc. However, the conventional cylindrical type of ICTP torch is hardly suitable for large-area materials processing. This is due to the fact that the ICTP is essentially shrunk by the self-pinch effect from the Lorentz force and temperature pinch. To apply the ICTP to large-area materials processing, it is one candidate to develop another shape type of ICTP. Recently, we have developed a loop type of ICTP, which is established inside a loop quartz tube but a part of the ICTP is formed on the substrate directly with linear shape [1]−[3]. Then, this linear-ICTP from the loop-ICTP can be exposed to the whole surface by scanning the substrate in one direction perpendicular to the linear-ICTP. Previously, we actually formed a Ar-O_2 loop-ICTP and then linear-ICTP to make surface oxidation of a 2-inch Si(100) substrate [4]−[6]. As a result, the oxide layer was found to be fabricated on the 2-inch substrate only by 180 s exposure of the linear ICTP [6]. Furthermore, a 2-inch SiC substrate can be oxidized by exposure of Ar-O_2 loop-ICTP [7].

The key point of large-area surface modification is how to form an uniform linear ICTP on the substrate. In this report, we developed a 3D thermofluid model for Ar loop-ICTP to consider temperature distributions and gas flow pattern in the loop-tube and on the substrate. The developed model solves the mass, momentum and energy conservation equations of heavy particles and the energy conservation equation and Poisson's equation for vector potential, as well as Sasha's equation for electron density Through this simulation, we obtained the 3D gas flow pattern and the 3D spatial distributions of the heavy particle temperature, the electron temperature, electron density and electromagnetic fields in the Ar loop-ICTP.

2. Loop Type of Inductively Coupled Thermal Plasma Torch

Figure 1 depicts the configuration of the loop-ICTP torch that we developed for surface modification test. The torch consists of a loop quartz tube with a loop diameter of 100 mm, and an inner tube diameter of 8 mm. At the outlet of this tube, there installed a rectangular quartz vessel with a size of 52.5 mm high and 110 mm wide and 20 mm in depth. Inside the rectangular quartz vessel, a Si_3N_4 substrate holder with a size of $95 \times 12 \times 2$ mm^3 is placed. As a plasma forming gas, Ar is supplied from the top of the loop tube. The loop quartz tube is sandwiched by the coil where a rf current is supplied from an rf inverter power supply through an rf matching circuit. This rf current creates magnetic field perpendicular to the coil, and rotational electric field along the loop tube. This electric field sustains an Ar loop-ICTP in the tube and on the substrate holder. For this Ar loop-ICTP, a three dimensional (3D) modeling was made to obtain the temperature and gas flow fields.

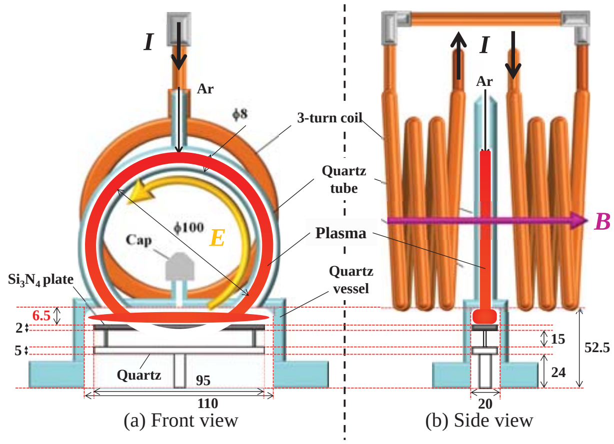

Fig. 1. Loop type of induction thermal plasma torch for the present calculation.

3. Model

3.1. Assumptions

The developed model assumes that Ar loop-ICTP is in the following conditions: (i) The electron temperature T_e is treated separately to the heavy particle temperature T_h. Heavy particles gain the power transferred through elastic collisions with electrons. (ii) All the species have the same flow velocity, meaning that one-fluid model is adopted. (iii) The plasma is optically thin. (iv) Ionization equilibrium and excitation equilibrium are established.

3.2. Governing equation

A three dimensional (3D) thermofluid model with electromagnetic field for Ar loop-ICTP was developed using COMSOL Multiphysics Ver.5.3 [8]. Under the assumptions in the previous section, the following governing equations were solved:

Mass conservation equation:

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \boldsymbol{u}) = 0 \tag{1}$$

Equation of motion:

$$\rho \frac{\partial \boldsymbol{u}}{\partial t} + \rho \boldsymbol{u} \cdot \nabla \boldsymbol{u} = -\nabla p + \nabla \cdot \boldsymbol{\tau} \tag{2}$$

Stress tensor

$$\boldsymbol{\tau} = \eta_{\mathrm{WFF}}(\nabla \boldsymbol{u} + (\nabla \boldsymbol{u})^T) - \frac{2}{3}\eta_{\mathrm{eff}}(\nabla \cdot \boldsymbol{u}) \tag{3}$$

Energy conservation for heavy particles:

$$\rho C_{\mathrm{p}} \frac{\partial T_{\mathrm{h}}}{\partial t} + \rho C_{\mathrm{p}} \boldsymbol{u} \cdot \nabla T_{\mathrm{h}} = \nabla \cdot (\kappa_{\mathrm{eff}} \nabla T_{\mathrm{h}}) + E_{\mathrm{eh}} \tag{4}$$

Energy conservation for electrons:

$$\nabla \cdot (\kappa_{\mathrm{e}} \nabla T_{\mathrm{e}}) = e\mu_{\mathrm{e}} n_{\mathrm{e}} \boldsymbol{E} \cdot \boldsymbol{E} - E_{\mathrm{eh}} - P_{\mathrm{rad}}(T_{\mathrm{e}}) \tag{5}$$

Energy transfer from electrons to heavy particles through elastic collision:

$$E_{\mathrm{eh}} = \frac{2 m_{\mathrm{e}} m_{\mathrm{Ar}}}{(m_{\mathrm{e}} + m_{\mathrm{Ar}})^2} \frac{3}{2} k_{\mathrm{B}} (T_{\mathrm{e}} - T_{\mathrm{h}}) \times n_{\mathrm{e}} (n_{\mathrm{Ar}} \pi \bar{\Omega}_{\mathrm{eAr}}^{(1,1)} + n_{\mathrm{Ar^+}} \pi \bar{\Omega}_{\mathrm{eAr^+}}^{(1,1)}) \bar{v}_{\mathrm{eAr}} \tag{6}$$

Ionization equilibrium for Saha's equation:

$$\frac{n_{\mathrm{e}} n_{\mathrm{Ar^+}}}{n_{\mathrm{Ar}}} = \left(\frac{2\pi m_{\mathrm{e}} k_{\mathrm{B}} T_{\mathrm{e}}}{h^2} \right)^{\frac{3}{2}} \frac{2 Z_{\mathrm{Ar^+}}}{Z_{\mathrm{Ar}}} \exp\left(-\frac{E_{\mathrm{Ar}}}{k_{\mathrm{B}} T_{\mathrm{e}}} \right) \tag{7}$$

Poisson's equation for vector potential:

$$\nabla \times (\nabla \times \boldsymbol{A}) = \mu_0 (\omega^2 \varepsilon - j e \mu_{\mathrm{e}} n_{\mathrm{e}} \omega) \boldsymbol{A} \tag{8}$$

$$\boldsymbol{B} = \nabla \times \boldsymbol{A} \tag{9}$$

$$\boldsymbol{E} = -j\omega \boldsymbol{A} \tag{10}$$

where ρ: mass density [kg/m^3], $\boldsymbol{u}$: gas flow vector [m/s], p: pressure [Pa], η_{eff}: effective viscosity [Pa·s], κ_{eff}: effective thermal conductivity for heavy particles [W/(m·K)], T_{h}: heavy particle temperature [K], T_{e}: electron temperature [K], C_{p}: specific heat [J/(kg·K)], P_{rad}: radiation power [W/m^3], μ_{e}: electron mobility [m^2/(V·s)], ω: angular frequency for coil current [rad/s], $\pi\bar{\Omega}_{\mathrm{eAr}}^{(1,1)}$, $\pi\bar{\Omega}_{\mathrm{eAr^+}}^{(1,1)}$: momentum transfer collision integral for e-Ar and e-Ar$^+$, respectively, $\bar{v}_{\mathrm{eAr}}$: relative thermal velocity between electron and Ar [m/s], m_{e}, m_{Ar}: masses of an electron and Ar [kg], n_{e}, n_{Ar}, $n_{\mathrm{Ar^+}}$: number densities of e, Ar and Ar$^+$ [m^{-3}], Z_{Ar}, $Z_{\mathrm{Ar^+}}$: internal partition function of Ar and Ar$^+$, E_{Ar}: ionization potential of Ar [J], ε: dielectric constant [F/m], e: electronic charge [C], k_{B}: Boltzmann constant [J/K], h: Planck constant [J·s], μ_0: permeability of vacuum [H/m].

As indicated here, energy transfer from electrons to heavy particles is considered due to mainly elastic collisions among them. The electron density n_{e} is calculated through Saha's equilibrium equation with T_{e}. Using this calculated n_{e}, the electrical conductivity is computed with collision integrals, T_{e} and T_{h}. Electromagnetic field is computed using Poisson equation for vector potential. It automatically determines the plasma column location from electromagnetic field and the electrical conductivity on the substrate. The k-ϵ model was used as turbulent flow model just for simplicity although its validity should be further discussed.

3.3. Calculation space and boundary condition

Fig. 2 shows the calculation space and boundary condition. The calculation space was taken to be a loop quartz tube and a rectangular quartz vessel. The plasma torch has one gas inlet from the top of the tube. In the quartz vessel, a substrate holder is placed. Table 1 summarizes calculation condition. From the top of the loop tube, Ar gas is supplied at a flow rate of Q_1=0.1 slpm at T_{h}=300 K. The inner walls of the loop-tube and vessel were set to be non-slip, while the outer wall of them was assumed to have T_{h}=300

Table 1. Calculation condition

Gas kind	Ar
Gas flow rate	Q_1=0.1 L/min
Pressure	10 torr
Coil current	140 A_{ems}
Current frequency	360 kHz

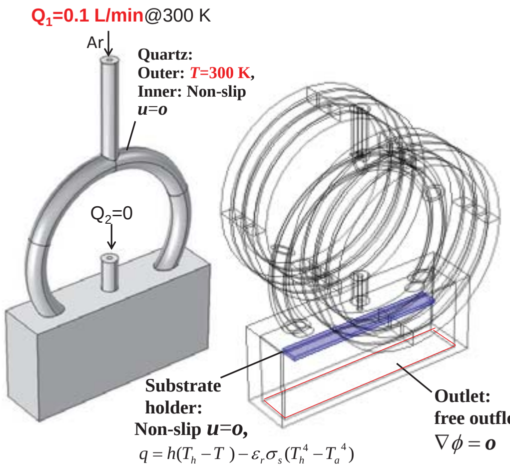

Fig. 2. Computational space and boundary condition for loop type induction thermal plasma torch.

K because these walls are all water-cooled in the actual experiments. The wall of the substrate holder was set to be adiabatic ($-\boldsymbol{n} \cdot \boldsymbol{q} = 0$) for simplicity. The rectangular quartz vessel has a free gas flow outlet at its bottom side. For calculation of electromagnetic field, a large volume sphere space was taken with infinite boundary. Pressure at a point on the bottom boundary was set to 10 Torr, and the coil current was assumed to have a root mean square value of 140 A_{ems} and a frequency of 360 kHz.

4. Results and discussions

4.1. Gas flow pattern

Fig. 3 shows the stream lines inside the loop-ICTP torch. In the loop tube, gas from the top of the tube flows along the tube, flowing out to the quartz vessel. This gas flow collides with the substrate holder, being divided into the right and left hand sides on the holder: One flow directs to the center of the torch, the other flows out finally to the bottom exit. On the center of the holder, the flow collides with the counter flow. Finally, all the flow goes out at the bottom exit.

4.2. Electron temperature and heavy particle temperature

Figs. 4(a) and (b) show the spatial distributions of T_e and T_h, respectively. Inside the loop-tube, T_e reaches to 9700 K as inedited in panel (a). This is attributed to the fact that strong induced electric field is present there, re-

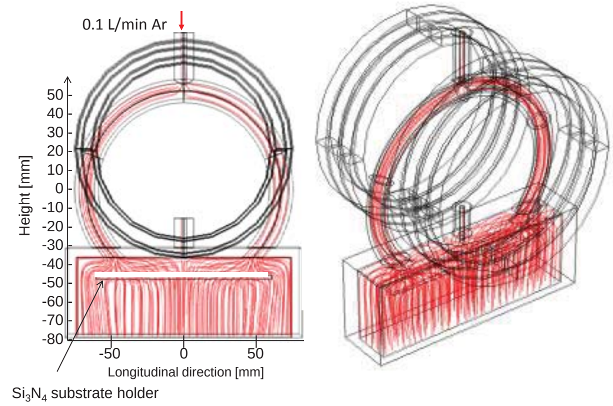

Fig. 3. Streamlines in Ar loop-type of induction thermal plasma torch.

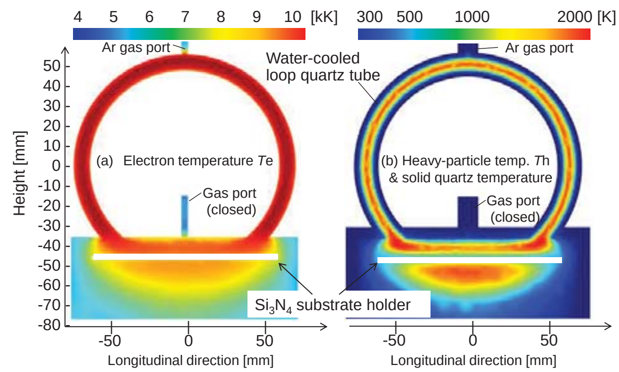

Fig. 4. Streamlines in Ar loop-type of induction thermal plasma torch (I_{coil}=140 A_{Mrs}).

sulting in high joule heating to electrons. As seen in this figure, T_e of 9700 K is almost uniformly distributed in the loop-tube, while T_e is 9000 K on the substrate holder. This high T_e elevates T_h in 10 torr plasma. From panel (b), T_h is about 1700 K almost uniformly along the center of the loop-tube, whereas T_h is around 400 K at the wall because of water-cooled. This value of T_h is mainly determined by energy balance between the energy transfer from electrons and thermal conduction to the tube wall. On the other hand, T_h is 1900 K at two stagnation positions around the outlet of the tube where the convection loss is low.

As depicted in Figs.4(a) and (b), T_e is largely different from T_h. To indicate a difference between T_e and T_h clearly, a relative deviation between T_e and T_h, defined by $(T_e - T_h)/T_e$, is presented in the left panel of Fig. 5. At the tube wall,$(T_e - T_h)/T_e \sim 0.96$ because T_e =9000 K and T_h =400 K. On the other hand, along the center of the loop-tube, $(T_e - T_h)/T_e \sim 0.84$ is found. This deviation between T_e and T_h originates from high electric field strength inside the tube. The right panel of Fig. 5 indicates the electric field strength. Inside the loop-tube, the electric field strength reaches 250–300 V/m in root mean square value. Thin electric field elevates T_e, and then T_h through the elec-

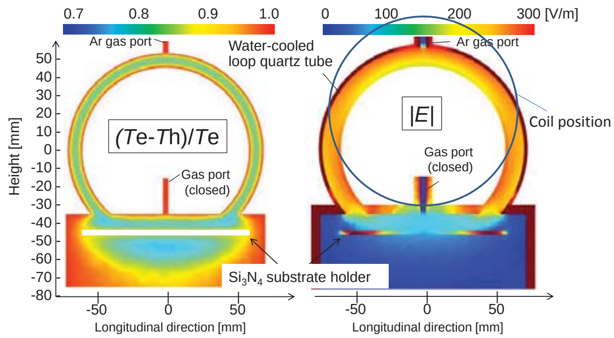

Fig. 5. Deviation between electron temperature and heavy particle temperature (left), and the distribution of electric field strength (right) in Ar loop-type of induction thermal plasma torch (I_{coil}=140 A_{RMS}).

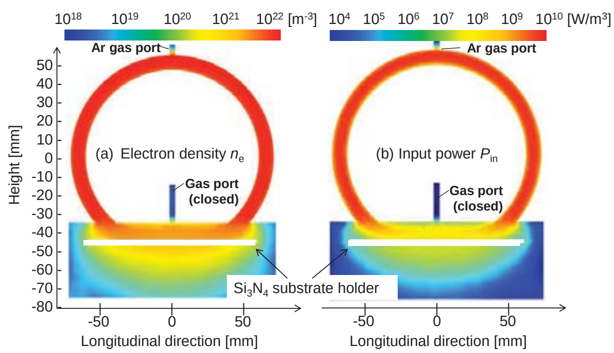

Fig. 6. Distribution of electron density and input power density in Ar loop-type of induction thermal plasma torch (I_{coil}=140 A_{Roms}).

tron energy transfer by collision with heavy particles.

4.3. Electron density and input power

Figs. 6(a) and (b) illustrates the distribution of n_e and the input power density P_{in} by joule heating, respectively. The quantity n_e is about 2.5×10^{22} m^{-3} in the loop tube. This value is almost similar to the value under thermal equilibrium condition. On the other hand, n_e reaches to 7.1×10^{20} m^{-3} on the substrate holder. The above result suggests that a loop-plasma in the tube and on the holder is formed with a circular electron current along the loop. As a result, the loop plasma absorbs the input power density P_{in} of 1.1×10^9 W/m^3 in the loop tube, and 7.1×10^7 W/m^3 on the substrate holder. The reason why P_{in} is lower in the tube than on the substrate is due to larger cross section of the plasma on the substrate compared to in the tube.

4.4. Heavy particle temperature distribution on the substrate holder

It is important to consider the uniformity in T_h on the substrate in case that this loop-ICTP is used for surface modification process. Fig. 7 indicates the T_h distribution on the holder. Here, the substrate holder has a length of 95 mm ($-47.5 < x < +47.5$ mm) and a depth of 15 mm

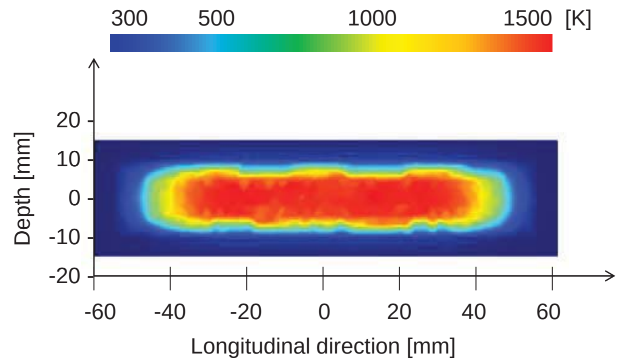

Fig. 7. Distribution of surface temperature of Si_3N_4 substrate holder irradiated by Ar loop-type of induction thermal plasma (I_{coil}=140 A_{RMS}).

($-7.5 < y < 7.5$ mm). As shown in this figure, T_h has 1350 K almost uniformly in 50 mm long on the holder. This suggests that the loop-ICTP could be useful to uniform surface modification for 50 mm long substrate.

5. Summary

In this report, a 3D two-temperature thermofluid model was developed for Ar loop-ICTP. As a result, it was found that T_e and T_h reach 9700 K and 1700 K, respectively, in the tube for a rf coil current of 140 A_{rums}. The loop-ICTP can have high electron density over 10^{21} m^{-3}. The input power is injected into the loop ICTP along the circular loop.

References

[1] Y. Tanaka, et al., 12th Asia Pacific Conf. Plasma Technol. & 27th Symp.Plasma Sci. for Materials (APCPST12/SPSM27), 2014.9, Adelaide, Australia

[2] H.Irie, et al., ISPlasma/IC-PLANTS 2015, A1-P-03, 2015.3, Nagoya, Japan

[3] Y. Maruyama, et al., IEEE Trans. Plasma Science, 44, 3164-3171, 2016

[4] Y. Tanaka, et al. 14th High-Tech Plasma Processes, S6-13, 2016.7, Munich, Germany

[5] T. Tsuchiya, et al. Int. Symp. Plasma Chem. (ISPC-23), P2-16-7, 2017.7, Montreal, Canada

[6] T. Tsuchiya, et al. , Plasma Chem. & Plasma Process., 28, 599-620, 2018

[7] Y. Tanaka, et al. ISPlasma2018/IC-PLANTS2018, 05pB11O, 2018.3, Nagoya, Japan

[8] COMSOL Multiphysics; `http://www.comsol.com/`