Numerical Study of Flow and Plasma in an Inductive Chlorine Discharge

M. Shiozawa* and K. Nanbu*

*Institute of Fluid Science, Tohoku University, Sendai, Japan 980-8577

Abstract. Particle modeling of inductively coupled chlorine plasmas is performed. The flow field of background gas is simulated using the direct simulation Monte Carlo (DSMC) method and the structure of plasma is examined by use of the electron Monte Carlo simulation. Using the plasma current that is obtained from the electron Monte Carlo simulation, electromagnetic field and plasma structure are determined self-consistently, including effects of collisionless heating. Spatial distributions of electromagnetic field, electron density and temperature, and rates of dissociation and dissociative attachment are obtained together with flow fields of Cl₂ and Cl.

INTRODUCTION

Plasma processing has been used for fabricating semiconductors. The requirements such as high aspect-ratio etching of features and uniform etching of large diameter wafers are satisfied by use of high density plasmas in a low gas pressure. Inductively coupled plasma (ICP) is one of such high density plasmas and has been widely used in etching process. For a control of etching process it is important to understand the plasma structure in ICP reactors. Numerical simulations of ICPs have been performed for this purpose. There are mainly two approaches for the analysis of ICPs. One is the fluid model and the other is the particle model. ICP reactors are driven at a low gas pressure and hence the particle model based on the Boltzmann equation makes sense. There are several advantages in the particle model as compared with the fluid model. Electron energy distribution function (EEDF) and ion velocity distribution (IVD) can be obtained and also the effects of collisionless heating are included in the particle model as it stands. Economou et al. [1] used the direct simulation Monte Carlo (DSMC) method to examine reactive neutral and ion flow in an ICP reactor. Nanbu et al. [2] used the Monte Carlo method for obtaining the production rate of radicals and then used the DSMC method for a prediction of etch rate in an ICP reactor.

In the present study, the gas flow and plasma structure in inductive chlorine discharges are simulated simultaneously to examine the effect of the flow on the plasma structure. The flow fields are obtained using the DSMC method and the plasma structure is examined using the electron Monte Carlo simulation. The plasma structure and the induced electromagnetic field driven by the rf coil current are coupled. Collisionless heating is taken into consideration by direct sampling of the current density from the motions of electrons.

CALCULATION PROCEDURE

The geometry of the reactor is shown in Figure 1. The reactor consists of a quartz cylinder and a diffusion chamber. The substrate holder in the diffusion chamber is movable in z-direction. The flow, electromagnetic field, and plasma are assumed to be axisymmetric. The total height of the reactor is 684 mm. The radii of quartz cylinder and diffusion chamber are 100 mm and 174 mm, respectively. An 8-turn coil, which is driven by rf current, is wound around the quartz cylinder.

Particle modeling of ICP consists of three modules; DSMC calculation of flow, analysis of electromagnetic field, and electron Monte Carlo simulation. Flow fields of Cl₂ and Cl are calculated using the DSMC method and hence spatial distributions of density and temperature of Cl₂ and Cl are obtained. The electromagnetic field and the plasma structure are calculated using the DSMC results as follows. The gas density is used to determine the probabilities of collisions for e−Cl₂ and e−Cl. The gas temperature is used to sample the velocity of gas molecules colliding with electron. The
induced electric field is obtained from the Maxwell equations by assuming that the field is axisymmetric and hence it has only an azimuthal component. Electrostatic field is replaced by a simple sheath model. Collisionless motions of electrons in a small time step are traced in the electromagnetic field and hence the current density in Ampère’s law is sampled directly. We did not use Ohm’s law for the current density. Next collisions of $\text{e} - \text{Cl}_2$ and $\text{e} - \text{Cl}$ are considered.

The total number of collisional events is 20. The production rate of Cl, which is due to dissociation or dissociative attachment, is sampled. This rate is an input to the flow simulation.

**Flow field**

The flow fields of background gases are simulated using the DSMC method. In the DSMC method, the motions and collisions of neutral species are traced and hence time evolution of the flow fields is simulated. The simulated particles are neutral species $\text{Cl}_2$ and Cl; the etch product such as $\text{SiCl}_2$ and $\text{SiCl}_4$ are not considered because the etch rate of Si wafer is assumed infinitely small. Also, heavy ion $\text{Cl}_2^+$ and Cl$^+$ are neglected in the DSMC calculation because the densities of ions are one order smaller than those of $\text{Cl}_2$ and Cl.

In the free motion stage of molecules, we consider injection of source gas $\text{Cl}_2$ from the top of the reactor, pumping of gases from the annular exit between the wall of the diffusion chamber and the substrate holder, production of Cl by reactive collisions of $\text{Cl}_2$, reflection of $\text{Cl}_2$ and Cl on the chamber wall, and recombination of Cl on the wall. The flux of injected $\text{Cl}_2$ is assumed to be uniform in the radial direction. The pumping of gases is treated by introducing the pumping probability $P_{\text{pump}}$; when a simulated particle is incident on the annular exit at $z = 0$, the particle is removed with the probability $P_{\text{pump}}$ and diffusely reflected with a probability $(1 - P_{\text{pump}})$. The probability $P_{\text{pump}}$ is adjusted in such a way that the total gas pressure agrees with a computational condition. The production rate of Cl, which is due to dissociation ($\text{e} + \text{Cl}_2 \rightarrow \text{e} + 2\text{Cl}$) or dissociative attachment ($\text{e} + \text{Cl}_2 \rightarrow \text{e} + \text{Cl}^- + \text{Cl}$), is obtained in advance in the electron Monte Carlo simulation. The production of Cl is one of the couplings of the plasma and flow field. The Cl atoms produced by dissociation or dissociative attachment are hot. Their kinetic energy is 1.5 eV and 1.1 eV for dissociation and dissociative attachment [3]. The velocities of the hot atoms are sampled from the Maxwell distribution. The recombination ($\text{Cl} \rightarrow 1/2\text{Cl}_2$) on the chamber wall is the loss of Cl and the production of $\text{Cl}_2$. One simulated Cl particle incident on the wall is removed with the probability $P_{\text{rcmb}}$. If a recombination is realized, one $\text{Cl}_2$ is produced with a probability $(1/2)W_{\text{Cl}}/W_{\text{Cl}_2}$, where $W_{\text{Cl}}$ and $W_{\text{Cl}_2}$ are the weights of Cl and $\text{Cl}_2$, respectively. The weight $W_s$ for a species $s$ is defined by $W_s = n_s V_s/N_s$, where $n_s$ is the number density, $V_s$ is the volume of a cell, and $N_s$ is the number of simulated particles in $V_s$.

For collisions between neutral species, we consider $\text{Cl}_2 - \text{Cl}_2$, Cl–Cl and $\text{Cl}_2$–Cl collisions in time step $\Delta t$. In a chlorine discharge, the source gas is $\text{Cl}_2$ and the radicals Cl are produced by the dissociation or dissociative attachment
of Cl$_2$. Therefore, the density of Cl is largely different from that of Cl$_2$; if we use a common weight for all species, the number of simulated particles with a lower number density is small and hence the fluctuation of sampled data will become large. In order to suppress the fluctuations, the collisions between molecules with different weights are treated by the weight algorism [4]. All molecules are regarded as hard sphere. The collisions between the same species is treated by the maximal collision number method [4]: the maximal collision number in $\Delta t$ for $s - s$ ($s = \text{Cl}_2$ or Cl) collisions is

$$N_{ss} = \frac{1}{2} W_s N_s (N_s - 1) (g_{ss})_{\text{max}} \sigma_{ss} V_e^{-1} \Delta t,$$

where $N_s$ is the number of simulated particles in a cell, $(g_{ss})_{\text{max}}$ is the maximal relative speed and $\sigma_{ss}$ is the cross section for $s - s$ collision. For Cl$_2$--Cl collisions, the weight $W_s$ for Cl$_2$ is not equal to $W_r$ for Cl. The maximal collision number for $s - r$ (Cl$_2$--Cl) collisions, $N_{sr}^*$, is chosen as

$$N_{sr}^* = \max(W_s, W_r) N_s N_r (g_{sr})_{\text{max}} \sigma_{sr} V_e^{-1} \Delta t,$$

where the $\max(W_s, W_r)$ is to return the largest value and $\sigma_{sr}$ is the cross section for $s - r$ collision. The number $N_{sr}^*$ is an overestimate. To compensate it a collision is regarded to occur with a probability of $(g_{sr})/(g_{sr})_{\text{max}}$ and furthermore, if a collision is realized, the simulated particle of species $s$ changes the velocity with a probability $W_r/\max(W_s, W_r)$ and the particle of species $r$ does with a probability $W_s/\max(W_s, W_r)$. Post-collision velocities are determined by Riechelmann and Nanbu’s method [5] because the flow is axisymmetric.

**Electromagnetic field**

Electric field consists of the induced electric field driven by the rf coil current and the electrostatic field governed by the Poisson equation. We use the cylindrical coordinate system ($r$, $\theta$, $z$). The induced electric field has only the azimuthal component $E_\theta$. The electrostatic field forms sheaths near all walls and has radial and axial components. We assume that the effect of electrostatic field is restricted to a narrow region near the wall and introduce a sheath model instead of solving the Poisson equation. The sheath model employed is mentioned later.

The induced electric field is obtained from Maxwell equations. The electric field and current density are assumed to vary as $\exp(j \omega t)$, where $\omega$ is the angular frequency of the coil current and $t$ is the time. The electric field $E_\theta$ can be written as

$$E_\theta = \hat{E}_\theta \exp(j \omega t),$$

where $\hat{E}_\theta$ is the complex amplitude. The amplitude $\hat{E}_\theta$ is governed by the equation

$$\left( \frac{\partial^2}{\partial r^2} + \frac{1}{r} \frac{\partial}{\partial r} + \epsilon_0 \mu_0 \omega^2 - \frac{1}{r^2} + \frac{\partial^2}{\partial z^2} \right) \hat{E}_\theta = j \omega \mu_0 \hat{J}_\theta,$$

where $\epsilon_0$ is the dielectric constant of vacuum, $\mu_0$ is the permeability of vacuum, and $\hat{J}_\theta$ is the complex amplitude of the current density. The current density $\hat{J}_\theta$ was given by the Ohm’s law in our previous work [6] by assuming cold plasma. The assumption of cold plasma may not be valid in a low gas pressure. Here we used a different approach. In the particle modeling, since the motions for the charged particles are traced, the current density can be directly sampled from the velocities of simulated particles. The current density depends mostly on the motions of electrons. It is given by

$$J_\theta = \mathbb{R}\{\hat{J}_\theta \exp(j \omega t)\} = \frac{1}{V} \sum (q W_e v_\theta),$$

where $V$ is the volume of a cell, the summation is for all simulated electrons in the cell, $q(< 0)$ is the electronic charge, $W_e$ is the weight of the simulated electron, and $v_\theta$ is the azimuthal component of the electron velocity.

The time-averaged power deposition into a unit volume of plasma is given by

$$P_{\text{dep}} = \frac{1}{2} \mathbb{R}\left( \hat{J}_\theta \hat{E}_\theta \right) = \frac{1}{2} |\hat{J}_\theta| |\hat{E}_\theta| \cos(\Delta \phi),$$

where $\Delta \phi = \phi_{E_\theta} - \phi_{J_\theta}$ is the phase difference between $\mathbb{R}(E_\theta)$ and $\mathbb{R}(J_\theta)$. 

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When the electric field is obtained by solving equation (4), the induced magnetic field is given by Faraday’s law. The radial and axial components of the magnetic field are

\[
\hat{B}_r = -\frac{j}{\omega} \frac{\partial \hat{E}_\theta}{\partial z}, \tag{7}
\]
\[
\hat{B}_z = \frac{j}{\omega} \frac{\partial}{\partial r} (r \hat{E}_\theta). \tag{8}
\]

Electron Monte Carlo simulation

If the time step is small, motion and collision of electrons can be separated by the principle of decoupling [4]. Ions are not considered because we replaced the electrostatic field by a sheath model. The motion of an electron is traced by solving

\[
m \frac{dv}{dt} = q(E + v \times B), \tag{9}
\]
where \(m\) is the mass of an electron, \(v\) is the velocity, \(E\) is the electric field, and \(B\) is the magnetic flux density. The electric field \(E\) has the azimuthal component \(E_\theta\) produced by the coil current and plasma current. The components \(E_r\) and \(E_z\) resulting from the Poisson equation are replaced by the simple sheath model; all walls are assumed to be electrostatically floating and hence the potential difference \(\Phi_t\) between the plasma bulk and the wall is given by [7]

\[
\Phi_t = \frac{k_B T_e}{2} + k_B T_e \ln \left( \frac{M}{2 \pi m} \right)^{1/2}, \tag{10}
\]
where \(k_B\) is the Boltzmann constant and \(M\) is the mass of Cl\(^{+}\). The electrons whose kinetic energy \(\varepsilon_\perp\) are higher than \(|q|\Phi_t\) are let to pass through the sheath and be absorbed on the wall. Here the kinetic energy \(\varepsilon_\perp\) is \(\varepsilon_\perp = mv_\perp^2/2\), where \(v_\perp\) is the component of electron velocity perpendicular to the wall. Electrons that are incident on the wall are removed if \(\varepsilon_\perp > |q|\Phi_t\) and specularly reflected if \(\varepsilon_\perp < |q|\Phi_t\). The potential difference \(\Phi_t\) is determined by the electron temperature \(T_e\) in the cell that is next door to the wall.

After solving the equation of motion with time step \(\Delta t\), collisions \(e - \text{Cl}_2\) and \(e - \text{Cl}\) are treated, where \(e\) denotes electron. The collisional probability of event \(k\) in time \(\Delta t\) is

\[
P_k = n_g \sigma_k(\varepsilon) \left( \frac{2\varepsilon}{m} \right)^{1/2} \Delta t, \tag{11}
\]
where \(\sigma_k\) is the collision cross section for event \(k\) and \(n_g\) is the number density of Cl\(_2\) or Cl. We use a set of collision cross sections for \(e - \text{Cl}_2\) and \(e - \text{Cl}\) reported by Morgan et al. [8] The number of \(e - \text{Cl}_2\) collisional events is 9, that is, elastic collision, ionization, vibrational excitation, four electronic excitations, dissociation, and dissociative attachment. The number of \(e - \text{Cl}\) collisional events is 11, that is, elastic collision, ionization, and nine electronic excitations. The total number of collisional events is 20. Both the occurrence of collision and the collisional event are determined by Nanbu’s method [4]. The densities of Cl\(_2\) and Cl, which are obtained in the flow simulation, are used to determine the collisional probability, which is also a kind of coupling of plasma and flow.

RESULTS AND DISCUSSION

First we carried out the electron Monte Carlo simulation at fixed the density ratios \(n_{\text{Cl}_2}/n_{\text{Cl}}\) as preliminary calculations. In the calculations, the background gas is assumed to be uniform and at rest and the gas temperature is set 400 K and the gas pressure is set 0.5 Pa. We calculated two cases: (a) \(n_{\text{Cl}_2}/n_{\text{Cl}} = 1/0\), and (b) \(n_{\text{Cl}_2}/n_{\text{Cl}} = 0/1\). The driven frequency \(f = \omega/2\pi\) of the coil current is 13.56 MHz and the coil current \(I_c\) is 6 A. Amplitudes of electric field and current density are shown in Figures 2 and 3. The mean electron number densities for cases (a) and (b) are \(2.21 \times 10^{16} \text{ m}^{-3}\) and \(6.74 \times 10^{16} \text{ m}^{-3}\). The amplitude of current density for case (b) is 2.5 times as large as that for case (a) at \(r = 0.096 \text{ m}\) and \(z = 0.45 \text{ m}\). The electric field and the current density peak near the quartz wall and the power is mainly deposited in a narrow skin region. The skin depth \(\delta\) for cases (a) and (b) is about 0.042 m and 0.025 m respectively.
m at $z = 0.45$ m. Note that we defined the skin depth by $e^{-1} = |\hat{E}_\theta|/(R - \delta)/|\hat{E}_\theta|(R)$, where $R$ is the radius of quartz cylinder. The maximum of power deposition for case (b) is larger than for case (a). However, the skin depth for case (b) is thinner than for case (a) and hence the total power deposition into the plasma for cases (a) and (b) result in 454 W and 385 W, respectively. The electron temperature in Figure 4 shows that near the quartz wall $T_e$ is much higher for case (a) than for case (b), consistently with the total power deposition. On the contrary, inside the diffusion chamber, $T_e$ is lower for case (a) than for case (b). The reason is as follows. The electron energy is low in the diffusion chamber because of almost no power deposition; low energy electrons can still have their energy by vibrational excitations for case (a) but cannot do electronic excitations of case (b), where threshold energy is about 9.0 eV, and hence there is no loss of electron energies in case (b).

The results of the preliminary simulations showed that the Cl$_2$/Cl density ratio affects the plasma parameters $n_e$ and $T_e$. This ratio is determined by the flow simulation of Cl$_2$ and Cl. Therefore, it is necessary for a reliable prediction of the plasma parameters to take the flow of Cl$_2$ and Cl into a full consideration. Let us do this way by calculating case (c). The computational conditions for case (c) are as follows. We start from a tentative flow simulation. The initial density ratio is chosen to be $n_{Cl_2}/n_{Cl} = 1/0$ because the discharge is switched on at time zero. We first simulate the steady Cl$_2$ flow for given initial conditions: Cl$_2$ mass flow rate $Q = 50$ sccm, gas pressure $P_k = 0.5$ Pa, gas temperature $T_g = 300$ K, and wall temperature $T_w = 300$ K. $Q$ and $T_w$ are fixed for all succeeding flow simulations, where $P_k$ is a prescribed total pressure at the corner of the diffusion chamber ($r = 0.174$ m, $z = 0.3$ m). The Cl$_2$ pressure at the corner resulted in 0.475 Pa for $P_{pump} = 0.034$ in the initial steady flow. The variation of Cl$_2$ pressure inside the reactor is less than 8%. Using the initial flow field of Cl$_2$, we performed the electron Monte Carlo simulation (EMS). The $E$- and $B$- fields in this simulation are determined for the coil current $I$ of 6 A and its driven frequency of 13.56 MHz as in the case of preliminary calculations. The EMS gives a spatial distribution of the production rate of Cl. Using the data for this rate, we performed the DSMC calculation for a mixture of Cl$_2$ and Cl. The recombination probability of Cl, $P_{rcmb}$ is set 0.0082 on the quartz wall [9] and 0.04 on the metal wall [10]. After completing of this simulation we have the second approximation of the flow field. This field is used in the second EMS, which yields the improved production rate of Cl. We performed the repetitions of a set of DSMC and EMS until the fields of flow and plasma converged.

Figure 5 shows the spatial distributions of the number densities of Cl$_2$ and Cl. The density ratio $n_{Cl_2}/n_{Cl}$ is from 0.32 to 2.30 in the reactor. The Cl$_2$ density is minimal and Cl density is maximal in the middle of quartz cylinder due to the dissociation. The Cl density is about three times larger than Cl$_2$ density in this region. The dissociation of Cl$_2$ mostly occurs in the quartz cylinder and is the dominant process to produce radicals Cl. The rate of dissociative
attachment is much less than that of dissociation. In the diffusion chamber, the molecule Cl₂ is reproduced by the recombination (Cl → 1/2Cl₂) on the chamber wall and there is no dissociation in gas phase. Therefore, the Cl₂ density increases and Cl density decreases there. Figures 6 – 9 show the amplitudes of electric field |\vec{E}_\theta| and current density |\vec{J}_\theta|, electron number density nₑ, and electron temperature Tₑ. The density of Cl is larger than that of Cl₂ in the quartz cylinder. However, the skin depth of 0.042 m agrees with that for case (a), 100 \% Cl₂. Also the distributions of electric field, current density, and electron density for case (c) are close to the results of case (a). The reason is that
the increment of electron density due to the amount of Cl density in case (c) is suppressed by the reduction of total number density at the condition of a constant total gas pressure. The maximum of total gas density \( n_{\text{tot}} = n_{\text{Cl}_2} + n_{\text{Cl}} \) is by 21\% less than that for cases (a) and (b). In addition, the gas densities have spatial distribution in case (c). The electron temperature near the quartz wall is by 10\% higher for case (c) than for case (a) due to the decrease of inelastic collisions \( e-\text{Cl}_2 \).
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REFERENCES