Study Of The Optimum Conditions And Parameters To Perform Simulation Of Low Temperature Capacitive Radio Frequency Argon Discharge

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Abstract. Capacitively coupled plasma (CCP) is used extensively in plasma processing, such as, plasma etching, deposition, and sputtering. Particle-in-Cell (PIC) is used to explore the discharge dynamics microscopically. Considering all species in PIC simulation is challenging. Also, considering various atomic transitions, e.g., ionization, recombination, excitation, and deexcitation is not doable. The state of the art is to carry out fluid simulation. We compare between PIC and fluid simulation for radio frequency CCP to reveal the effect of various approximations as assuming constant temperature, simplifying Navier-Stokes equations in terms of particles mobility and diffusivity, and considering artificial boundary conditions at the electrodes. The fluid model predicts qualitatively PIC results in few minutes. For Argon discharge in geometrically symmetric CCP, the plasma bulk is quasineutral. Over the electrodes, two sheaths are built up due to the difference between ion and electron fluxes. The dynamics of two sheaths are out of phase. In the fluid model, chemical reactions and atomic processes can be considered. Argon metastable states density is maximum in the plasma bulk. In semi dark sheaths, metastable states concentration is small. The proposed fluid model could be used as a simulation platform to find the optimum conditions and to interpret experimental results.

1. Introduction

Capacitively coupled plasma reactors are characterized with simple geometry. They are simply a two planar, cylindrical or spherical electrodes. The discharge takes place between the electrodes. The potential difference between the electrodes may be driven by DC or/and Ac sources [1-3]. To generate high density plasmas in the bulk, the discharge is driven by radio frequency (RF) sources. The plasma bulk density increases by increasing the frequency [4]. Also, radio- frequency discharges allow the efficient processing of dielectric substrates as well as conductors. There are different configurations to
allow control of the ion flux and ion energy over substrates. The two electrodes may be different in area, this kind of reactors called geometrically asymmetric capacitively coupled plasma reactors, where the sheath potential is inversely proportional to the electrode area [5]. Also, asymmetry between the sheaths could be achieved electrically when multiple harmonics sustain the discharge [6, 7]. Recently, magnetic fields are found to alter the discharge symmetry [8-10].

Argon is an inert gas. Its chemistry is simple. Therefore, it is used extensively in radio frequency capacitively coupled plasma reactors to understand the discharge dynamics experimentally and theoretically [11,12,13,14]. Also, in many industrial plasma applications, gases like O₂, N₂, CH₄, CF₄ are used with Argon to optimize the sputtered, deposited, or etched layers and profiles [15,16,17,18]. Of course, adding contaminations to Argon will change the chemical composition of plasma species, their concentrations, and the plasma density. In this manuscript we will model the Argon discharge. The model will be utilized as a simulation platform to add other gaseous in the future. It is not feasible to understand the plasma dynamics via solving the equation of motion of all plasma particles; the number of plasma particles is huge, and the motion of each charged particle affects the electric and magnetic fields distribution. Also, the general and exact solution of Boltzmann equation is not present [19]. Therefore, approximations have to be done to simplify the problem and at the same time to give acceptable physical description. The state of the art is to carry out a Particle-In-Cell (PIC) simulation [20, 21]. The equation of motion of superparticels is solved in a self-consistent way with Maxwell’s equations. Each superparticle presents the physics of 10³ to 10⁶ of real particles. Although PIC simulation provides a microscopic description, the simulation is computationally expensive. One run may take few hours to few weeks depending on the underline problem and the available computer facilities. It is mandatory to find other methods to describe the plasma dynamics acceptably but in a proper time. One of these models is the lumped element circuits [22-24]. The discharge dynamics is calculated from the equivalent circuit. Resonances, harmonics generation, and power dissipations are well calculated in efficient time. Also, fluid models give a macroscopic description in a proper time. There are different approximations considering fluid models [25-29]. Here, we will assume drift-diffusion (DD) approximation at low pressure to investigate how far the results from PIC simulation. The fluid simulation assuming the DD approximation are in a good qualitative agreement with kinetically self-consistent simulation, however, the code is timely efficient. The long-time of PIC simulation is reduced to few minutes.

2. Fluid model

Fluid models provide a macroscopic description to the plasma species based on the averaged values of density, speed, and energy. The fluid model equations could be derived from the averaging of moments of Boltzmann equation. The equations set is closed with Maxwell’s equations and assuming an equation of state. In typical capacitive coupled plasma, the gap size between the two electrodes is small compared to the area of the electrode; therefore, the problem could be simplified into 1D problem. If the distance between the electrodes is along z-axis, then the continuity equation for electrons, ions, and metastable states reads as:

\[
\frac{\partial n_e}{\partial t} + \frac{\partial \vec{v}_e}{\partial z} = K_{ei} n_e n_e + K_{mi} n_m n_e + K_{mn} n_m^2 ,
\]

(1)

\[
\frac{\partial n_i}{\partial t} + \frac{\partial \vec{v}_i}{\partial z} = K_{ei} n_e n_i + K_{mi} n_m n_i + K_{mm} n_m^2 ,
\]

(2)
And
\[
\frac{\partial n_m}{\partial t} + \frac{\partial \Gamma_m}{\partial z} = K_{ex} n_e n_m - K_{mi} n_i n_m - K_{em} n_m n_e - K_{in} n_m n_e - 2K_{m} n_m^2 - K_{q} n_n n_m - K_{q} n_m^2 n_m.
\] (3)

Where \(n_e, n_i, n_m, n_n\) are the density of electrons, Ar ions, Ar metastable states, and Ar neutral atoms, respectively. \(\Gamma_j = n_j u_j\) is the species flux and \(u_j\) is the species speed where \(j = e, i, m, n\) for electrons, ions, metastable, and neutral particles. Different atomic processes are considered as electron impact ionization of neutral species and metastable states, collision of metastable states, excitation of neutral species, and deexcitation of metastable states. The right hand side of equations 1, 2, and 3 are the gain and loss terms for each species with positive and negative signs, respectively. The rate coefficient of each process may be constant or temperature dependent as given in table 1 [30, 31].

The momentum conservation for low temperature plasma can be written as
\[
m_e n_e \left( \frac{\partial u_e}{\partial t} + u_e \frac{\partial u_e}{\partial z} \right) = -e n_e E - \nabla P_e - m_e n_e v_{en} u_e
\] (4)
\[
m_i n_i \left( \frac{\partial u_i}{\partial t} + u_i \frac{\partial u_i}{\partial z} \right) = Z e n_i E - \nabla P_i - m_i n_i v_{in} u_i
\] (5)
And
\[
m_m n_m \left( \frac{\partial u_m}{\partial t} + u_m \frac{\partial u_m}{\partial z} \right) = -\nabla P_m - m_m n_m v_{mn} u_m
\] (6)

Where collisions with the neutral background is dominant. \(v_{jn}\) is the collision frequency of species \(j\) with the background gas. Collisions between electrons, ions, and metastable states are negligible. \(P_j = n_j kT_j\) is the pressure of each species. The momentum conservation (Navier-Stokes equations) can be simplified utilizing the drift-diffusion (DD) approximation. Almost of the distance between the two electrodes are plasma bulk, where dependence on time and position is slowly varying, i.e., \(\frac{\partial}{\partial t} \rightarrow 0\) and \(\frac{\partial}{\partial z} \rightarrow 0\). Within the plasma bulk, the flux of species according to the DD approximation is given as
\[
\Gamma_e = -n_e \mu_e E - D_e \frac{\partial n_e}{\partial x},
\] (7)
\[
\Gamma_i = n_i \mu_i E - D_i \frac{\partial n_i}{\partial x},
\] (8)
and
\[
\Gamma_m = -D_m \frac{\partial n_m}{\partial x}.
\] (9)

Table 1. Atomic processes considered in the simulation [30, 31]
### Equation of Reaction Rate of Reaction Coefficient

| Reaction                                      | Rate of Reaction Coefficient |
|-----------------------------------------------|------------------------------|
| $e + \text{Ar} \rightarrow \text{Ar}^* + 2e$ | $K_{ei} = 1.253 \times 10^{-7} \exp(-18.618/T_e)$ cm$^3$/s |
| $e + \text{Ar} \rightarrow \text{Ar}^* + e$ | $K_{ex} = 3.712 \times 10^{-8} \exp(-15.06/T_e)$ cm$^3$/s |
| $e + \text{Ar}^* \rightarrow \text{Ar} + 2e$ | $K_{mi} = 2.05 \times 10^{-7} \exp(-4.95/T_e)$ cm$^3$/s |
| $e + \text{Ar}^* \rightarrow \text{Ar}^* + e$ | $K_{em} = 1.818 \times 10^{-9} \exp(-2.14/T_e)$ cm$^3$/s |
| $\text{Ar}^* + \text{Ar} \rightarrow \text{Ar}^* + \text{Ar} + e$ | $K_{r} = 2 \times 10^{-7}$ cm$^3$/s |
| $\text{Ar}^* + \text{Ar} \rightarrow 2\text{Ar}$ | $K_{mm} = 6.2 \times 10^{-10}$ cm$^3$/s |
| $\text{Ar}^* + 2\text{Ar} \rightarrow \text{Ar} + \text{Ar}$ | $K_{3q} = 1.1 \times 10^{-31}$ cm$^6$/s |

The fluxes are calculated based on the species mobility $\mu_j = e/m_j v_{jn}$ and diffusivity $D_j = T_e/m_j v_{jn}$ [32]. For electrons, the momentum transfer collisional frequency and the electron plasma frequency are larger than the driven radio-frequency, inertia of electrons may be neglected so that the mean velocities respond instantaneously to the electric field. Therefore, the DD approximation for electrons may be valid in the plasma sheath. On contrary, ions are heavier than electrons. Their mobility and diffusivity are small compared to electrons’ mobility and diffusivity. Therefore, in the sheath, for ions we replace the instantaneous field $E$ in equation 8 with the time averaged field; i.e., $\bar{E} = \int_0^t E dt/t$. In the intermediate regime when the ion plasma frequency is comparable to the driven frequency, the DD approximation or its modification may be not accurate because ions respond partially to the instantaneous field.

The electric field $E$ and the potential $\phi$ are calculated from

$$E = -\frac{\partial \phi}{\partial z}$$

and

$$\frac{\partial^2 \phi}{\partial z^2} = \frac{-e(n_i - n_e)}{\epsilon_o}.$$  

In order to solve the equations’ model numerically, the boundary conditions are as follows:

At left electrode

$$V_p = V_{DC} - V_{RF} \sin(\omega_{rf} t)$$  

and at right electrode

$$V_G = V_{DC} - V_{RF} \sin(\omega_{rf} t + \pi).$$

The electron density and ion density between the electrodes, $0 < z < L$ are

$$n_{e,l} = n_0 \sin(\pi z/L).$$  

At the electrodes the electron density is assumed to be inertial-less, so $n_{e,P,G} = n_0 \exp(eV_{P,G}/kT_e)$; the ion density is calculated from the balance between the ion flux and the electron flux to the electrodes and ensures the equality

$$V_{DC} = \sum_{k=1}^{\infty} \sum_{l=1}^{\infty} \left(n_e(k,l) - n_i(k,l)\right) \Delta z \Delta t + V_f,$$

where $k$ and $l$ are the time and position indices on the time-space mesh, the area of each cell on the mesh is $\Delta z \Delta t$. $V_f$ is the floating potential.

### 3. Results and discussion
An Argon gas with a pressure of 10 mTorr is considered. The distance between the two electrodes is 5 cm. The electron temperature is 1.5 eV and the ion temperature is 0.025 eV. Fluid simulation is iterated for 150 RF periods. The distance between the two electrodes is divided into 250 grids and each RF period is divided into 50 time steps. The traditional RF frequency of 13.65 MHz is used. The two electrodes are with the same area and a single frequency is driven. This is the geometrically and electrically symmetric discharge, where the dc self-bias of both sheaths is equal in magnitude and out of phase. Due to the high mobility and diffusivity of electrons with respect to ions, the electrode is always negatively biased with respect to the plasma.

![Figure 1](image1.png)  ![Figure 2](image2.png)

**Figure 1.** The potential between the electrodes at three phases, $\tau$ is the periodic time of the radio frequency voltage.

**Figure 2.** The electric field between the electrodes at three phases, $\tau$ is the periodic time of the radio frequency voltage.

![Figure 3](image3.png)  ![Figure 4](image4.png)

**Figure 3.** Electrons and ions fluxes between the two electrodes.

**Figure 4.** Time averaged metastable states density.
Figure 1 displays the potential between the electrodes. The potential at the electrode is more negative compared to the plasma bulk. There is a phase difference $\pi$ between the two sheaths. The collapse of a sheath is accompanied with an expansion of the other sheath. Figure 1 provides the potential at three phases ($t = \tau$, $t = 3\tau/5$, and $t = \tau/5$) within the RF voltage signal. When the time equals the periodic time $t = \tau$, the left sheath is very small and its potential equals the floating potential, i.e., the potential due to the thermal motion of electrons. At $t = 3\tau/5$, the right sheath is collapsed with respect to the left sheath. Moreover, when most of the RF source voltage goes to a sheath, the potential on the opposite sheath is around the floating potential. The corresponding electric field between the two electrodes is shown in Figure 2. The sheath electric field accelerates positive ions to hit the substrate or the electrodes. From equations 7, electrons follow the applied field instantaneously. The electron fluxes at the three phases are shown in Figure 3. The time averaged electron flux is shown with a dotted line. The ion flux depends on the time averaged field and it is shown by the dashed line in Figure 3. To avoid misleading from Figure 3, the ion flux is not zero within the sheaths. But the time averaged electron flux is larger than the time averaged ion flux with the order of $10^3$. This is a self-consistent with the results in Figure 1 and this is the reason behind the generation of the self-biased $V_{DC}$. The density of metastable states is shown in Figure 4, the maximum density of the metastable states are at the plasma bulk, where the brightness is also maximum. In order to check the validity of the DD approximation, the fluid results are compared with PIC results. In PIC simulation the distance between the two electrodes is discretized into 129 grids. The RF period is discretized into 600 time steps. To get convergence, the PIC simulation is iterated for 5000 RF periods. In fluid and PIC simulations, numerical instabilities are diminished. The time averaged density and field are shown in Figure 5 and Figure 6. This is a good qualitative agreement. As known in typical RF capacitive discharges, the plasma bulk is quasineutral. In the sheath, the ion density is larger than the electron density. The discrepancies between the fluid simulation and the PIC calculations could be minimized when the exact Navier-Stokes equations for ions are solved. Also, when the energy conservation and the variations of the particles mobility and diffusivity with temperature is considered. However, here, we got a macroscopic description in few minutes which is qualitatively agree with the computationally expensive PIC results. Our model can include different species of important gaseous as $O_2$, $N_2$, $CH_4$, $CF_4$ and, consequently, it may be used extensively in industrial applications. In addition, when the fluid model is coupled with a Monte Carlo scheme, the ion energy and the ion angular energy may be calculated in a hybrid manner [33] or fast self-consistent calculations could be carried out as in Ensemble-in-Space time (EST) model [34].
4. Conclusion

In this manuscript, a symmetric radio frequency capacitively coupled plasma has been simulated using Particle-in-Cell and a fluid model. Both models are in a qualitative agreement: The plasma bulk is quasineutral, two sheaths -where the electron density is smaller than the ion density- are created, and within sheaths a large field toward electrodes is generated. However, the fluid model is computationally efficient. In the fluid model, atomic processes are considered. The Metastable states of Argon are calculated between the electrodes. For high radio frequency regime; i.e., frequencies greater than 10 MHz, the assumption of constant electron temperature and the drift-diffusion approximation are valid. The proposed fluid model can be implemented by adding the chemistry of different species of important gaseous as O\(_2\), N\(_2\), CH\(_4\), and CF\(_4\). The proposed model can be used as a simulation platform to interpret and optimize etching, deposition, and sputtering experiments. Also, the fluid model could be coupled to a Monte Carlo scheme or to Ensemble-in-Space time model to calculate the particles distribution over the electrodes.

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