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Effects of HF frequency on plasma characteristics in dual-frequency helium discharge at atmospheric pressure by fluid modeling

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Abstract

On the basis of the fluid theory and the drift–diffusion approximation, a numerical model for dual-frequency atmospheric pressure helium discharge is established, in order to investigate the effects of the high frequency source (HF) on the characteristics of dual-frequency atmospheric pressure helium discharge. The numerical results showed that the electron heating rate increases with enhancing HF frequency, as well as the particles densities, electron dissipation rate, current density, net electron generation and bulk plasma region. Moreover, it is also observed that the efficient electron heating region moves when the HF frequency has been changed. The plasma parameters are not linear change with the HF frequency linearly increasing.

Keywords: dual-frequency, numerical study, electron energy mechanisms

(Some figures may appear in colour only in the online journal)

1. Introduction

It is well known that the atmospheric pressure gas discharge plays an important role in materials processing. It has been widely applied, such as cleaning, depositing as well as materials surface treatments [1–4]. Low-pressure discharge plasmas also can realize these materials surface treatments in semiconductor industry. However, the capital costs associated with the equipment of vacuum systems and non-continuous manufacturing methods have limited their development. Because of not required vacuum systems and scan accomplish continuous operation, there has been a rapid growth in the development of atmospheric pressure plasma technologies.

Normally, generation of atmospheric pressure glow discharge is driven by single frequency source which is from kilohertz to megahertz. In the range of kilohertz, the dielectric barrier is applied to obtain stable discharge plasmas [5, 6]. In the range of megahertz, a high density plasma can be obtained in radio frequency (RF) discharge which is generated between two bare electrodes at atmospheric pressure [7, 8]. However, to improve the surface processing or etching, the high plasma density and the ion energy are both required. Recently, a dual-frequency (DF) discharge plasma technology has been received extensive attention in atmospheric pressure discharge. For low pressure discharges, this technology, one RF of high frequency (HF) is much greater than that of low frequency (LF), and HF source determines the plasma density and LF source controls the ion energy [9–13]. For atmospheric pressure discharges, DF technology is taken to realize controlling plasma discharge parameters independently. Kong et al 2009 [14] studied a DF atmospheric pressure plasma jet. They found that the HF at 5.5 MHz and LF at 30 kHz could obtain long plume length and high plasma density. Wang et al 2010 [15] studied the atmospheric pressure helium discharge driven by DF sources by numerical simulation. It can be found that the short pulses can assist to obtain a better power...
efficiency and high plasma density. Gans et al. 2012 [16] numerically investigated an atmospheric pressure DF driven source. The results showed that coupling of HF and LF was not only a simple linear superposition, but also a nonlinear effect between them. Huang et al. 2015 [17] reported an DF discharge in atmospheric helium. It was shown that the discharge current is mainly influenced by HF source and the applied voltage is mainly influenced by LF source.

In this paper, a numerical model for DF atmospheric pressure discharge is presented in section 2. We discuss the effects of HF frequency on the helium plasma characteristics of DF atmospheric pressure discharge in section 3. The characteristics of plasma density, electron energy mechanisms, ionization rate and so on are investigated in detail. These works are summarized and conclusions are drawn in section 4.

2. Numerical model

We shall consider a discharge generated by two parallel plate electrodes, as shown in figure 1, where L denotes the distance of the two plates. The left electrode (at x = 0) is driven by two sinusoidal RF voltages $V = V_0 \sin(2\pi f_d t) + V_L \sin(2\pi f_L t)$, and the right electrode (at x = L) is grounded, where the subscript H represents the HF source and the subscript L represents the LF source.

A fluid theory is used to describe the behavior of particles in the plasma. The fluid equations include the continuity, momentum and energy equations. The drift–diffusion approximation is taken to describe the particles fluxes instead of momentum equation at atmospheric pressure [18, 19]. We consider six particles in the model, such as electrons e, helium atoms He, helium ions He$^+$, and metastables He$^*$, and He$_2^*$. The reactions of this model are listed in table 1. There are some assumptions in this model which are in accord with references [20] and [21]. Based on the assumptions, the helical discharge plasma is described as follows. The continuity equation for particles is:

$$\frac{\partial n_{e,i,*}}{\partial t} + \frac{\partial \Gamma_{e,i,*}}{\partial x} = R_{e,i,*},$$

where the subscribes e, i, * describe electron, positive ion and metastable, $n_{e,i,*}$ represents the electron density, positive ion density and metastable density, $\Gamma_{e,i,*}$ denotes the electron flux, positive ion flux and metastable flux, $R_{e,i,*}$ is the electron source, positive ion source and metastable source. The drift–diffusion approximation replaces of the momentum equation, thus the particle flux is

$$\Gamma_{e,i,*} = \mp \mu_{e,i} n_{e,i} E - D_{e,i,*} \frac{\partial n_{e,i,*}}{\partial x},$$

where $\mu_{e,i}$ is the mobility of electrons and ions and $D_{e,i,*}$ is the diffusion coefficient of electrons, positive ions and metastables. These parameters are taken from references [19–21]. The electron energy equation is

$$\frac{\partial}{\partial t} \left( \frac{3}{2} n_e kT_e \right) = - \frac{\partial}{\partial x} q_e - e \Gamma_e \cdot E - \sum_i H_i n_i k_n n_e$$

where the electron energy flux is

$$q_e = - K_e \frac{\partial T_e}{\partial x} + \frac{5}{2} kT_e \Gamma_e$$

and the thermal conductivity of electrons is

$$K_e = \frac{3}{2} kD_e n_e,$$

where $k$, $e$, $N$, $H_i$, and $T_e$ are Boltzmann coefficient, the elementary charge, the background gas number density, the energy-loss coefficient, and electron temperature, respectively. The unit of the electron temperature is eV.

The Poisson equation satisfies

$$\frac{\partial^2 V}{\partial x^2} = \frac{e}{\varepsilon_0}(n_e - n_i).$$

The electric field derives from

$$E = - \frac{\partial V}{\partial x},$$

where $\varepsilon_0$ is permittivity of free space, $V$ is the electric potential, and $E$ is the electric field. In this work, the initial condition is in accord with references [19]. The boundary condition is specified in table 2. In table 2, $k_e$ and $\gamma$ are the electron recombination coefficient and SEE coefficient, respectively, which are taken from references [19]. The numerical method contains a center difference scheme on uniform meshes for space and an implicit scheme for time which is taken to solve all of the equations in this model. In this simulation, we use 400 points in space and 10 000 time steps in one HF period. Thus we get the space-step $\Delta h = L/400$ and the time-step $\Delta t = \frac{1}{10} \times 10^{-6}$ s. In our simulation, the discharge in general reaches steady after 1500 HF cycles when the electron density is approximately constant.

3. Results and discussion

We shall apply the helium as working gas and the gas temperature is a constant of 300 K, the background gas pressure $p = 760$ Torr and the electrode distance $L = 0.2$ cm in all simulations. The applied voltage is $V = V_0 \sin(2\pi f_d t) + V_L \sin(2\pi f_L t)$. Considering frequency as an
The frequency of LF source is electric at atmospheric pressure such as the electron temperature, the plasma characteristics of the dual-frequency helium discharge frequency in order to study the effects of the HF frequency on charge, therefore, in our modeling, we shall consider this 

Figure 2 shows evolution of the maximum electron density during 2000 cycles for the HF frequency of 14, 18, 22, 26 and 30 MHz, respectively. The x-axis is the cycle number.

the large HF frequency leads to the collision between electrons and neutral particles more frequently and results of the more neutral particles ionized. Similar results were reported in low pressure DF discharge [10]. The electron density is dependence of HF frequency.

Figure 3 gives the spatial profiles of cycle-averaged ion density for different frequencies of the HF frequency $f_{\text{HF}} = 14$, 18, 22, 26 and 30 MHz. It shows that the ion density increases with the increase of HF frequency values. Compared the density of He$^+_2$ (figure 3(b)) and the density of He$^+$ (figure 3(a)), the density of He$_2^+$ is two orders higher than the density of He$^+$, in other words, the main positive ion is He$_2^+$ in atmospheric pressure DF helium discharge. The spatial profiles of cycle-averaged electron density for different frequencies are shown in figure 3(c). With the HF frequency

## Table 1. The processes of helium discharge.

<table>
<thead>
<tr>
<th>No.</th>
<th>Processes</th>
<th>Coefficient (cm$^3$ s$^{-1}$)</th>
<th>$H_i$ (eV)</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.</td>
<td>$e + \text{He} \rightarrow \text{He}^+ + 2e$</td>
<td>$k_1 = 2.584 \times 10^{-12}T_e^{0.68}T_0^{-2.854092 \times 10^{17}/T_e}$</td>
<td>24.6</td>
<td>[22]</td>
</tr>
<tr>
<td>2.</td>
<td>$e + \text{He} \rightarrow \text{He}^+ + e$</td>
<td>$k_2 = 4.2 \times 10^{-9}T_e^{0.31}T_0^{-1.98/T_e}$</td>
<td>19.8</td>
<td>[23]</td>
</tr>
<tr>
<td>3.</td>
<td>$e + \text{He}^+ \rightarrow \text{He}^++2e$</td>
<td>$k_3 = 1.28 \times 10^{-7}T_e^{0.6}T_0^{-4.78/T_e}$</td>
<td>4.87</td>
<td>[23]</td>
</tr>
<tr>
<td>4.</td>
<td>$\text{He}^++2\text{He} \rightarrow \text{He}_2^++\text{He}$</td>
<td>$k_4 = 2.0 \times 10^{-31}$</td>
<td></td>
<td></td>
</tr>
<tr>
<td>5.</td>
<td>$\text{He}^++\text{He}^+ \rightarrow e + \text{He}^++e$</td>
<td>$k_5 = 8.7 \times 10^{-10}$</td>
<td></td>
<td></td>
</tr>
<tr>
<td>6.</td>
<td>$\text{He}_n^+\text{He}_n^+ \rightarrow e + \text{He}_2^+_n$</td>
<td>$k_6 = 2.03 \times 10^{-9}$</td>
<td></td>
<td></td>
</tr>
<tr>
<td>7.</td>
<td>$e + \text{He}_2^+ \rightarrow 2\text{He}$</td>
<td>$k_7 = 4.0 \times 10^{-9}$</td>
<td></td>
<td></td>
</tr>
<tr>
<td>8.</td>
<td>$e + \text{He}^+ \rightarrow \text{He} + e$</td>
<td>$k_8 = 2.9 \times 10^{-9}$</td>
<td></td>
<td></td>
</tr>
<tr>
<td>9.</td>
<td>$2\text{He}^+ + \text{He}^+ \rightarrow \text{He} + \text{He}_2^+$</td>
<td>$k_9 = 1.3 \times 10^{-33}$</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

$^a$Coefficients in cm$^3$ s$^{-1}$ and cm$^6$ s$^{-1}$ for two- and three-body collisions, respectively [23].

$^bT_e$ in eV except for 1, which is in K [22].

## Table 2. The boundary conditions.

<table>
<thead>
<tr>
<th>The boundary conditions</th>
<th>Electron flux</th>
<th>Positive ion flux</th>
<th>Metastable flux</th>
<th>Electron temperature</th>
<th>Potential</th>
</tr>
</thead>
<tbody>
<tr>
<td>$x = 0$</td>
<td>$J_e = -k_en_e - \gamma \sum J_i$</td>
<td>$J_i = \mu_i q_i E$</td>
<td>$n_a = 0$</td>
<td>$T_e = 0.5$</td>
<td>$V = V_H \sin(2\pi f_H t)$ + $V_L \sin(2\pi f_L t)$</td>
</tr>
<tr>
<td>$x = L$</td>
<td>$J_e = k_en_e - \gamma \sum J_i$</td>
<td>$J_i = \mu_i q_i E$</td>
<td>$n_a = 0$</td>
<td>$T_e = 0.5$</td>
<td>$V = 0$.</td>
</tr>
</tbody>
</table>

Important parameter for the DF atmospheric pressure discharge, therefore, in our modeling, we shall consider this frequency in order to study the effects of the HF frequency on plasma characteristics of the dual-frequency helium discharge at atmospheric pressure such as the electron temperature, the electric field, discharge current, the particle density and the mechanisms of power. The frequency of LF source is unchanged but the frequency of HF source is varied in this simulation. The voltage of HF source is $V_H = 600$ V, the voltage of LF source is $V_L = 300$ V, and frequency of LF source is $f_L = 2$ MHz. According to the results shown in our previous work reference [24], the waveforms of the discharge current and the applied voltage of DF discharge are mainly presented by sinusoidal, moreover the current waveform leads the voltage waveform by less than $\pi$ in phase, which is consist with the experimental result of reference [17]. This also can verify that the model is reasonable for simulating DF discharge at atmospheric pressure. The effect of LF source frequency is less than HF source frequency on the electron density and electron temperature according to our previous work [24].

Figure 2 shows evolution of the maximum electron density versus cycles for different HF frequencies during 2000 HF cycles. It shows that the larger HF frequency is, the longer time is required of reaching discharge steady stage. According to the rate of electron growth with time, for the large HF frequency $f_H = 22$, 26, and 30 MHz, it is divided into two stages during the discharge process, the growing fast stage (This stage means that the electron increases with time rapidly) and steady stage. For the small HF frequency $f_H = 14$ and 18 MHz, the discharge process can be divided into growing slowly stage and steady stage. The maximum electron density increases with the HF frequency increasing when the discharge reaches steady. The possible reason is that
increasing from 14 to 30 MHz, the electron density increases from \(1.3 \times 10^{12}\) to \(7.01 \times 10^{12}\) cm\(^{-3}\).

The cycle-averaged He\(^+\) density profiles for different HF values are shown in figure 4(a). The peaks of metastable density appear in the plasma/sheath interface and increase with the increase of HF frequency values. The term of the right hand of equation (1) \(R_n\) is the source of metastables by electron impact excitation of ground state helium, electron re-excitation to the resonant levels, destruction of metastables by step-wise ionization, metastable quenching, and metastable pooling, respectively. Thus, these processes determine the profile of metastable density. With the case of high HF values at 22, 26, and 30 MHz, the peaks of metastable density increase quickly. Moreover, the sheath thickness decreases and the bulk region enlarge. Figure 4(b) gives the cycle-averaged He\(_2^+\) density profiles for different HF values. From figure 4(b), it can be observed obviously that the density of He\(_2^+\) increases with HF frequency increasing. Compared with He\(^+\) density shown in figure 4(a), He\(_2^+\) density is two orders of He\(^+\) density, which is consist with the numerical results of references [25].

The electron impact excitation process not only relies linearly on the electron density but also exponentially on the electron temperature. The power absorbed per electron determines the excitation process. In order to study the power absorbed by electron, we plot the cycle-averaged electron temperature and electric field in figure 5. In the discharge space, the electric field is significant to characterize the behavior of the discharge, and especially the spatial distribution of electric field can reveal the discharge mode. Figure 5 shows that the cycle-averaged electric field profiles at different HF frequencies under the considered conditions. It illustrates that the maximum electric field increases as the HF frequency increasing at the electrode surface, and the value of maximum electric field

![Figure 3](image-url). Spatial profiles of cycle-averaged (a) He\(^+\) density; (b) He\(_2^+\) density; (c) electron density cross the electrode gap are shown in HF frequency at 14, 18, 22, 26, and 30 MHz.

![Figure 4](image-url). Spatial distributions of cycle-averaged density (a) He\(^+\), (b) He\(_2^+\) in the electrode gap.
field varies from 7.1 kV cm$^{-1}$ at 14 MHz to 16.7 kV cm$^{-1}$ at 30 MHz. In addition, we found that the sheath thickness shrinks with increasing HF frequency. It also can be clearly seen that the electric field nearly zero in the bulk plasma. Moreover, the positive electric field appears in the grounded sheath region, which increases with varying the HF frequency. Conversely, the negative electric field appears in the powered sheath region, which decreases with the increase of HF frequency. The electric field presents symmetrical distribution.

The spatial distributions of cycle-averaged electron temperature are shown in figure 6(a) for different HF frequencies. The maximum electron temperature $T_e$ appears in the sheath region which is near the electrode surface, and the value of maximum electron temperature $T_e$ increases with the increase of HF frequency from 8.2 eV at 14 MHz to 13.8 eV at 30 MHz. The spatial distribution of electron temperature is symmetric and it has minimum value in the center of the electrode gap, besides the minimum value of electron temperature changes little as HF frequency increasing. It is also indicated that the sheath thickness shrinks with increasing HF frequency.

The electron temperatures in the two sheath regions and in the bulk plasma region are presented in figure 6(b) for different HF frequencies. It shows that the electron temperatures are the same in the sheath regions, which increase slowly with HF frequency increasing. In the bulk plasma, the electron temperatures are nearly unchanged. Besides, the peaks of electron temperature linearly increase with increasing the HF frequency.

The second term on left-hand side of equation (3) denotes electron heating rate, it can be denoted by $P_c$:

$$P_c = -e\Gamma_e E = e\mu_e n_e E^2 + eD_e \frac{\partial n_e}{\partial x} \cdot E.$$  

(8)

The electron heating is the sum of ohmic heating and collisionless heating. The electron heating $P_c$ is strongly on the basis of drift–diffusion approximation which is instead of electron momentum balance equation [26, 27]. The electrons gain energy from electric field and then they are ionized. The electron heating rate $P_c$ plays an important role in discharge process. Figure 7 shows the spatial profiles of cycle-averaged electron heating rate $P_c$ for different HF frequencies, the electron heating rates resemble some other simulation at low pressure [28, 29]. With the increase of HF frequency, the electron heating rate $P_c$ significantly increases in the sheaths. In the bulk plasma, the electron heating rates are all constant because of the zero bulk electric field. Moreover, it can be clearly seen that the sheath thickness reduces and the electron heating rate peaks shift from the electron heating rate profiles. The efficient electron heating region is denoted by the position of the peaks of electron heating rate. The shift of peaks of the electron heating rate means along with the HF frequency change, the efficient electron heating region can have the change.

The last term on left-hand side of equation (3) indicates the electron energy dissipation because of inelastic collision between electrons and neutral particles, according to the processes of helium discharge in table 2, which is labeled as $P_L$. 

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**Figure 5.** Spatial distributions of cycle-averaged electric field across the electrode gap for the HF frequency of 14, 18, 22, 26, and 30 MHz respectively, in the 2000th cycle.

**Figure 6.** Spatial distributions of cycle-averaged (a) electron temperature in the electrode gap (b) electron temperature at $x = 0.01, 1, \text{ and } 1.99$ mm, for the HF frequency of 14, 18, 22, 26, and 30 MHz respectively, in the 2000th cycle.
The bigger HF frequency results the more electron energy dissipation in the whole discharge space. Generally, the inelastic collision between electrons and atoms or molecules is primary electron energy dissipation. It increases with the increase of HF frequency, which means the five reaction processes mentioned above increase. The electron energy dissipation is much higher in the sheath region than that in the bulk plasma, which can be interpreted that the source power is coupled to plasma through the mechanism of electron heating rate. Correspondingly the electric field is weak and the averaged electron temperature is high in the sheath region, so the higher electron energy dissipation appears in the sheath region.

Figure 9 gives the curve of RMS current density with different HF frequencies. From figure 9, we can clearly see that the RMS current is a function of HF frequency. It increases nonlinearly with the increase of HF frequency. The bigger HF frequency produces the larger current density. In accordance with the Child law, the sheath thickness $s$ and the ion current density $J_0$ has the relationship as follow $J_0 = \frac{4s^2}{\pi^2} \sqrt{\frac{2e}{m_i}}$ [29]. Therefore, when the current density is larger, the sheath thickness is the thinner.

The electron as the ‘seed’ in the plasma is the primary parameter of the plasma discharge, thus the net electron generation rate is important as well. According to the table 1, the net electron generation rate can be written as

$$R_e = k_1 n_e N + k_3 n_e n_N + k_5 n_s n_N + k_6 n_s n_N - k_7 n_e n_{He}^*.$$  

Figure 8 shows the different spatial profiles of cycle-averaged electron energy dissipation. Obviously, all the negative electron energy dissipations primarily appear in the sheath regions. The bigger HF frequency results the more electron energy dissipation in the whole discharge space. Generally, the inelastic collision between electrons and atoms or molecules is primary electron energy dissipation. It increases with the increase of HF frequency, which means the five reaction processes mentioned above increase. The electron energy dissipation is much higher in the sheath region than that in the bulk plasma, which can be interpreted that the source power is coupled to plasma through the mechanism of electron heating rate. Correspondingly the electric field is weak and the averaged electron temperature is low in the bulk plasma, therefore it results in the smaller electron energy dissipation. Nevertheless, the sheath electric field is strong and the electron temperature is high in the sheath region, so the higher electron energy dissipation appears in the sheath region.

In order to study the net electron generation rate behavior more clearly, we calculate the spatial and temporal evolution of dimensionless net electron generation rate during the discharge. The spatial and temporal evolution of dimensionless net electron generation rates for 14, 18, 22, 26, and 30 MHz, respectively, are shown in figure 10. By introducing non-dimensional electron density $\hat{n}_e = \frac{n_e}{n_0}$, non-dimensional gas number density $\hat{N} = \frac{N}{n_0}$, non-dimensional reaction coefficient $\hat{k}_i = \frac{k_i n_0}{\bar{v}_i}$ and non-dimensional metastable density $\hat{n}_s = \frac{n_s}{n_0}$, where $n_0$ is the number density $n_0 = 10^{14} \text{ cm}^{-3}$, and then we will get the dimensionless net electron generation rate $\hat{R}_e = \frac{R_e}{n_0}$. With the cycle (discharge time) increasing, the net electron generation rate is increasing. The peaks of net electron generation rate appear on the border of bulk and the sheath. With the increase of HF frequency, the net electron generation rate is increasing. Besides, the peaks of net electron generation rate move toward to the electrode as the HF frequency increasing. It is because the electrons obtain energy mainly from sheath electric field. When the HF frequency increases, the sheath thickness decreases (as shown figure 7).
Figure 10. Spatial and temporal evolution of normalized net electron produce rate in (a) 14 MHz; (b) 18 MHz; (c) 22 MHz; (d) 26 MHz; (e) 30 MHz.
which results the efficient electron heating region moving toward to the electrodes as the HF frequency increasing.

4. Conclusion and perspectives

For a dual-frequency helium discharge, a 1D fluid model is established to investigate the effects of the HF frequency on the DF helium plasma characteristics, particularly on the electron energy mechanism. The numerical results show that the plasma characteristics strongly depend on discharge parameters.

First, the effects of the HF frequency have been numerically studied and a significant effect on the plasma parameters, particularly when the frequency of HF source is high. This is because when the HF frequency is high, the electric field is strong in the sheath, the electrons gain more energy from the sheath electric field to ionize the neutral particles, therefore the particles density, the bulk plasma region, the current density, the net electron generation, the electron heating rate and the electron dissipation rate increase with the HF frequency increasing. Moreover, it is also observed that the efficient electron heating region moves to the electrode when the HF frequency has been increased and the plasma parameters are not linear change with the HF frequency linearly increasing.

Acknowledgments

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