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Dependence of discharge ignition on initial condition in atmospheric cascade glow discharges

Ying Guo,1,2,3 Zhengming Shi,1 Qianhan Han,1 Chenyu Wu,1 Jing Zhang,1 and Jianjun Shi1,2,3,a)

AFFILIATIONS
1 College of Science, Donghua University, Shanghai 201620, People’s Republic of China
2 Member of Magnetic Confinement Fusion Research Center, Ministry of Education of the People’s Republic of China, Shanghai 201620, China
3 Shanghai Center for High Performance Fibers and Composites, Center for Civil Aviation Composites of Donghua University, Shanghai 201620, People’s Republic of China

a)Author to whom correspondence should be addressed: jshi@dhu.edu.cn

ABSTRACT

A two-dimensional numerical fluid model was developed to study the effects of initial discharge conditions on the discharge dynamics and characteristics of atmospheric cascade glow discharge including dielectric barrier pulsed discharge and radio frequency (RF) discharge burst. In the dielectric barrier pulsed discharge, the second discharge in the pulse voltage falling phase develops faster than the first discharge in the pulse voltage rising phase, which is demonstrated by the spatiotemporal evolution of discharge and can be attributed to the initial discharge conditions in terms of electron and ion density and their spatial distribution. On the other hand, in the pulse modulated RF discharge coupled with pulsed discharge, the initial conditions of RF discharge can be influenced by the pulsed discharge. The ignition dynamics of RF discharge is compared with and without the pulsed discharge in terms of plasma densities and electric field to show the role of residual plasma species in the initial discharge conditions. It proposes that the discharge ignition both in the pulsed discharge and the RF discharge is dependent on the density and spatial profile of plasma species in the initial discharge conditions, which helps ignite and achieve stable operation of atmospheric glow discharge.

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I. INTRODUCTION

Nonthermal atmospheric plasmas have wide application prospects in material surface modification, film deposition, biological disinfection, ozone generation, etc.1–4 Atmospheric pressure glow discharges (APGDs) generated with excitation frequency from kilohertz to megahertz are employed to produce atmospheric plasmas with different characteristics, such as plasma density and gas temperature, which are determined by the discharge dynamics and mechanism. In order to better apply APGDs in business, research studies on gas breakdown have become hot spots.

For dielectric barrier discharge (DBD) with sinusoidal excitation at a frequency in the kilohertz range, a pulsed discharge event occurs in the voltage rising phase during every half cycle, which limits the discharge stability and plasma density.9 To improve the discharge stability, sub-microsecond voltage pulses were applied to generate pulsed DBD with two pulse discharges occurring in the voltage rising phase and falling phase, whose characteristics were different in terms of discharge intensity and dynamics.10 It is agreed that the second discharge is ignited by the space charge which is accumulated on the surface of the dielectric layer in the first discharge.

On the other hand, in the radio frequency (RF) APGDs with the excitation frequency in the megahertz range, the plasma species in the discharge gap oscillate with the RF electric field,11 which keeps the plasma density and gas temperature higher than those in DBD. It takes several microseconds for RF APGDs to achieve stable operation, as demonstrated by the spatial temporal evolution of
pulse modulated RF APGDs.\textsuperscript{12-14} It was shown that the discharge ignition of RF discharge burst was determined by the residual plasma species from the previous RF discharge burst, which suggested that RF discharge ignition was dependent on the initial conditions of discharge.

In this paper, a two-dimensional fluid model of cascade plasma in atmospheric helium with pulsed discharge and pulse modulated RF APGDs was developed to investigate the dependence of discharge ignition characteristics and dynamics on the initial conditions of discharge. The paper is divided into three parts: the computational model is described in the first part, the different initial conditions of pulsed discharge are studied in the second part, and the comparative investigation of pulse modulated RF discharge burst with and without the assistance of pulsed discharge is presented in the third part.

### II. COMPUTATIONAL MODEL

In this study, a two-dimensional self-consistent fluid model was developed. The atmospheric discharge is generated between two parallel plate electrodes covered with dielectric layers, the pulse voltage is applied on the top electrode, and a modulated RF voltage is applied on the bottom electrode. The amplitudes of pulse voltage and RF voltage are 1 kV and 0.5 kV, respectively. The RF frequency is 13.56 MHz. The gas gap between two dielectrically insulated electrodes is fixed at 2.5 mm. The thickness and the relative permittivity constant of dielectric layers are 1.0 mm and 10, respectively. Helium (He) gas is at a pressure of 760 Torr and the temperature is 300 K. Six species, namely, electrons (e), helium ions (He\textsuperscript{+}), dimer helium ions (He\textsuperscript{2+}), excited helium atoms (He\textsuperscript{*}), excited helium dimer (He\textsuperscript{2*}), and background helium atoms (He), are considered in the model, whose densities can be evaluated using the following continuity equations:\textsuperscript{16}

\begin{align}
\frac{\partial n_e}{\partial t} + \frac{\partial \Gamma_{e,x}}{\partial x} + \frac{\partial \Gamma_{e,y}}{\partial y} &= S_e, \quad (1) \\
\frac{\partial n_i}{\partial t} + \frac{\partial \Gamma_{i,x}}{\partial x} + \frac{\partial \Gamma_{i,y}}{\partial y} &= S_i, \quad (2)
\end{align}

where \( n \) and \( \Gamma \) are the density and flux of species, respectively, \( S \) is the sum of loss and source of the specific species,\textsuperscript{16} and the subscripts + and e represent ions and electrons, respectively. The subscripts \( x \) and \( y \) represent the axial and radial directions, respectively. The flux equations of ions and electrons are given with the diffusion-drift approximation,

\begin{align}
\Gamma_{i,x} &= +\mu_i E_n n_i - D_i \frac{\partial n_i}{\partial x}, \quad (3) \\
\Gamma_{i,y} &= +\mu_i E_n n_i - D_i \frac{\partial n_i}{\partial y}, \quad (4) \\
\Gamma_{e,x} &= -\mu_e E_n n_e - D_e \frac{\partial n_e}{\partial x}, \quad (5) \\
\Gamma_{e,y} &= -\mu_e E_n n_e - D_e \frac{\partial n_e}{\partial y}, \quad (6)
\end{align}

where \( D \) and \( \mu \) are the diffusion and mobility coefficient,\textsuperscript{17} respectively. The electrical field \( E \) can be obtained by solving the following equation:\textsuperscript{18}

\begin{equation}
\varepsilon_0 \left( \frac{\partial E_x}{\partial t} + \frac{\partial E_y}{\partial t} \right) = e \left( \sum_i \Gamma_{i,p} - \Gamma_e \right), \quad (7)
\end{equation}

where \( \varepsilon_0 \) and \( e \) are the permittivity and electron charge, respectively, \( p \) represents different ions considered in the model. The boundary condition for electrons and ions is

\begin{equation}
\Gamma_e = -\gamma \sum_p \Gamma_{e,p}. \quad (8)
\end{equation}

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig1.png}
\caption{(a) Waveforms of the applied voltage of pulsed discharge and RF APGDs and (b) waveforms of the applied voltage and current density of pulsed discharge; waveforms of RF (c) discharge voltage and (d) current density; the red line indicates that the current density has been stabilized.}
\end{figure}
The secondary emission coefficient $\gamma$ is 0.01. The flux of neutral species, ions, and metastable species at the electrode surface is dominated by drift, and the diffusive flux is negligible. The set of equations is solved using a time-dependent solver in COMSOL Multiphysics 5.4. The elementary reactions among the six plasma species mentioned above and their reaction coefficients follow the data in the literature. \cite{13, 16–18}

III. INITIAL CONDITIONS OF THE PULSED DISCHARGE

The atmospheric cascade plasma with pulsed discharge and RF discharge burst is excited by a microsecond pulse voltage and pulse modulated RF voltage, whose waveforms are shown in Fig. 1(a). The time interval between the pulse voltage and pulse modulated RF voltage is 10 $\mu$s, while one cycle lasts for 200 $\mu$s with a repetition frequency of 5 kHz. The duration of pulse voltage is 1 $\mu$s at 1 kV amplitude, with the rising time and falling time of 20 ns. The modulated RF voltage lasts for 100 $\mu$s with an amplitude of 0.5 kV. As shown in Fig. 1(b), there are two pulsed discharge events occurring in the rising phase and falling phase of the voltage pulse, which are the typical characteristics of pulsed DBD. \cite{19, 20} The first discharge in the rising phase reaches a discharge current density amplitude of 285.83 A/m$^2$ at a time instant of 0.16 $\mu$s within 0.1 $\mu$s. On the other hand, the secondary discharge in the falling phase reaches a current density amplitude of 645.21 A/m$^2$ at a time instant of 1.11 $\mu$s within 0.01 $\mu$s. It suggests that in the pulsed DBD, the second discharge achieves higher intensity within shorter time than the first discharge. Figures 1(c) and 1(d) show the waveforms of pulse modulated RF applied voltage and discharge current density. In Fig. 1(c), the RF voltage starts to grow at a time instant of 11.1 $\mu$s and reaches a stable amplitude of 500 V at a time instant of 11.6 $\mu$s, which is set to be consistent with that in experiments. \cite{13} The development of RF discharge current density from zero to a stable state is called the ignition phase. As can be seen from Fig. 1(d), the ignition time of RF discharge can be estimated to be about 1.2 $\mu$s (marked by the red line), which is also consistent with the experimental findings. \cite{13}

First, the accuracy of the model needs to be verified. The normalized discharge spatial profile across the discharge gap can be obtained experimentally by accumulating the discharge image intensity along the electrode surface and normalizing to the instantaneous maximum image intensity at each time instant, which is employed to demonstrate the temporal evolution of pulsed discharge. \cite{13, 22} In addition, the maximum particle density at each moment is normalized in simulation. As shown in Fig. 2(a), the first and second discharges in pulsed DBD can be recognized to be ignited above one of the electrodes and transit to another electrode, which happened in the rising phase and falling phase of the voltage pulse. The temporal evolution of the normalized He$^+$ spatial profile is shown in Fig. 2(b), which corresponds to the applied pulse voltage and discharge current in Fig. 1(b). The first discharge occurs at a time instant of 0.16 $\mu$s and is concentrated on the instantaneous cathode surface afterward. As the applied pulse voltage turns off, the second discharge is ignited and reaches a stable amplitude of 500 V at a time instant of 11.6 $\mu$s, which is set to be consistent with that in experiments. \cite{13} The development of RF discharge current density from zero to a stable state is called the ignition phase. As can be seen from Fig. 1(d), the ignition time of RF discharge can be estimated to be about 1.2 $\mu$s (marked by the red line), which is also consistent with the experimental findings. \cite{13}

Second, in order to understand the effect of difference initial conditions on the first discharge and second discharge in pulsed DBD, the spatiotemporal profile of electron density is presented in Fig. 3 to demonstrate the discharge dynamics. \cite{23} In the first discharge, as shown in Fig. 3(a), the sheath region is formed above the bottom electrode at 1.0 mm, in which the electrons achieve a maximum magnitude of $2.2 \times 10^{17}$ m$^{-3}$. In Fig. 3(b), with the second discharge, the sheath region is formed above the top electrode at 3.5 mm with a maximum electron density of $3 \times 10^{17}$ m$^{-3}$, which is higher than that in Fig. 3(a). It can also be seen from Figs. 3(a) and 3(b) that the initial conditions before the ignition of two discharges are different. Before the ignition of first discharge in Fig. 3(a), electron density is uniformly distributed in the discharge gap with a
FIG. 3. Spatiotemporal profile of electron density of (a) first discharge and (b) second discharge in pulsed DBD.

Before the ignition of the second discharge in Fig. 3(b), electron density is inhomogeneously distributed in the discharge gap with a higher density of \(1.5 \times 10^{17} \text{ m}^{-3}\) above the bottom electrode than the density of \(1.0 \times 10^{17} \text{ m}^{-3}\) in the plasma bulk. It suggests that the difference in the discharge initial conditions in terms of electron density magnitude and spatial profile in the discharge gap may be attributed to the difference between the two pulsed discharges. Given that the time interval between the pulsed discharges is about 1 \(\mu\)s, the initial condition of the electron density and its spatial profile in the second discharge will be influenced by the first discharge in which the electrons are concentrated in the regime close to the bottom electrode at 1.0 mm. With the ignition of the second discharge, the applied pulse voltage at the top electrode drops rapidly from 1000 V to 0 V within 20 ns, during which the electric field is reversed due to the space charges in the discharge gap, and the bottom electrode acts as the anode. The electrons in the discharge gap are driven toward the bottom electrode by the electric field, which is shown in Fig. 3(b) with the hump of electron density at the time instant around 1.1 \(\mu\)s. With the building-up density of \(0.3 \times 10^{17} \text{ m}^{-3}\), the electrons accumulated near the bottom electrode are extinguished and the generated electrons in the boundary regime between the sheath and plasma bulk are elevated. On the other hand, in the first discharge, due to the initial conditions with much fewer space charges in the discharge gap, the pulse voltage is applied directly on the top electrode and the sheath region is established adjacent to the bottom electrode with the generation of space charges by ionization. It explains that a higher discharge current density is achieved in the second discharge than the first discharge, which can be attributed to the residual plasma species, especially the electrons. These electrons are considered to change the initial conditions of the subsequent discharge.

Furthermore, the spatial profiles of electron density, ion density, and electric field across the discharge gap at the time instants of first \(0.16 \mu\)s and second discharge \((1.11 \mu\)s) current peaks are shown in Figs. 4(a) and 4(b), respectively. The ion density includes both the atomic ion density and dimer helium ion density. It is worth noting that the atomic ions are not dominant ions toward the steady-state for high-pressure gas discharges in helium due to the three-body conversion. It can be seen that there is a sheath with a thickness of \(0.3 \text{ mm}\) above the cathode (bottom electrode at 1.0 mm) in the first discharge, and the amplitude of ion density is \(3.32 \times 10^{17} \text{ m}^{-3}\). The maximum magnitude of the electric field is \(13.45 \times 10^{5} \text{ V/m}\), and the peak value of electron density is \(1.61 \times 10^{17} \text{ m}^{-3}\). However, the thickness of the sheath above the...
cathode (top electrode at 3.5 mm) in the second discharge is about 0.5 mm, while the maximum electron density near the sheath boundary is $1.46 \times 10^{17} \text{ m}^{-3}$ and the ion density is $1.56 \times 10^{17} \text{ m}^{-3}$. The maximum magnitude of the electric field is $9.97 \times 10^5 \text{ V/m}$.

The peak densities of electrons and ions are different in Figs. 4(a) and 4(b), which can be attributed to the initial conditions of the second discharge influenced by the first discharge. It is shown in Fig. 3 that the electron intensity in the second discharge is larger than that in the first discharge, while at the time instants of the current peak in Fig. 4, the maximum ion density and electric field in the second discharge are smaller than those in the first discharge. This phenomenon depends on the initial conditions of the existence of plasma species in the discharge gap before the discharge ignition. The charged particles $e$, $\text{He}^+$, and $\text{He}_2^+$ in the discharge gap will be driven by the electric field, which is the sum of the applied electric field and built-in electric field induced by the space charges. When the second discharge occurs, the applied electric field is released because the applied pulse voltage drops to 0 V within 20 ns (from 1.08 $\mu$s to 1.1 $\mu$s), as shown in Fig. 1(b). The electric field in the discharge gap is strengthened because of the built-in electric field by the space charges in the first discharge, which drives the electrons to drift from the top electrode to the bottom electrode. The enhancement of electron density is found above the anode surface of the bottom electrode in Fig. 4(b), which is consistent with that in Fig. 3(b) with the hump of electron density at the time instant of the second discharge. In the regime above the cathode surface, the electrons are depleted, and the sheath region is formed there with an elevated electric field. In the plasma bulk, the electron density increases to around $1.5 \times 10^{17} \text{ m}^{-3}$, compared to that of around $1.0 \times 10^{17} \text{ m}^{-3}$ in the first discharge, which explains the high discharge current density achieved in the second discharge, as shown in Fig. 1(b). This dynamics of electrons in the ignition phase of the second discharge proposes that the residual plasma species in the discharge gap dominate the discharge ignition, which is demonstrated by the reduction in discharge ignition time and sheath strength and the elevated discharge current density. On the other hand, the spatial profiles of electrons and ions and electric field shown in Fig. 4(a) are the typical ignition dynamics of pulsed discharge, in which the initial discharge conditions are considered as the neglection of residual plasma species in the discharge gap. The electrons and ions are mostly generated in the boundary region between the sheath and plasma bulk by the ionization, and electrons are depleted in the sheath region, which induces the enhanced electric field in the sheath region.

IV. INITIAL CONDITIONS OF THE RF DISCHARGE

It has been demonstrated that the initial conditions played an important role in the discharge ignition dynamics and discharge characteristics. In cascade discharge with pulsed discharge and pulse modulated RF APGDs, it is proposed that the discharge ignition dynamics will also be influenced by the previous pulsed discharge. Figures 5(a) and 5(b) give the temporal evolution of normalized spatial profiles of $\text{He}^+$ in RF discharge burst without and with pulsed discharge before the RF discharge ignition, respectively, which are employed to compare with the experimental measurements, as explained in Fig. 2. As the RF voltage is applied at a time instant of 11.1 $\mu$s, the RF discharge is ignited in the middle of the discharge gap, whose spatial profile has a bell shape. With the establishment of sheath regions above both electrodes, the RF discharge is stabilized and shows the spatial profile of a double-hump. This evolution of the discharge spatial profile is also demonstrated by the
experimental measurements and recognized as the discharge ignition phase.\textsuperscript{13,14} The distributions of He\textsuperscript{+} density in the discharge space at the time instant of 11.6 μs for RF discharge without and with pulsed discharge are shown in Figs. 5(c) and 5(d), respectively. Helium ion density is evenly distributed in space, with a low density without the assistance of pulsed discharge, as shown in Fig. 5(c). However, in Fig. 5(d), the discharge has gradually stabilized, helium ions are concentrated in the sheath layer near the upper and lower plates, and the density is high (the maximum value reached $3.5 \times 10^{16}$ m\textsuperscript{-3}). The RF discharge ignition time can be estimated to be 0.7 μs and 0.5 μs in Figs. 5(a) and 5(b) by the establishment of the double-hump spatial profile, which suggests that with the assistance of pulsed discharge, the ignition of RF discharge can be shortened. The pulsed discharge occurring previously can change the initial conditions of RF discharge by the residual plasma species in the discharge gap, which is proposed to be the mechanism of the assistant effect of pulsed discharge. The electron density, ion density, and electric field at three typical time instants during the RF discharge ignition phase are further investigated, which are compared in the RF discharge without and with the assistance of pulsed discharge.

In Fig. 6(a), the spatiotemporal evolution of helium ion density during the first 20 RF cycles of the discharge ignition phase with pulsed discharge clearly shows that the discharge spatial profile evolves from the bell shape to the double-hump shape, which is consistent with the experimental findings.\textsuperscript{13,14} The comparison between the spatial distribution of helium ion density across the discharge gap without and with pulsed discharge is presented in Fig. 6(b) at three typical time delays of 0, 10, and 20 RF cycles from the application of RF voltage. The case of the 0 RF cycle is just before the application of RF voltage, which can be considered to be the initial conditions of discharge. The helium ion densities in the discharge gap with and without pulsed discharge are $1.8 \times 10^{16}$ m\textsuperscript{-3} and $5.9 \times 10^{15}$ m\textsuperscript{-3}, respectively, which suggests that the pulsed discharge enhances the ion density in the initial conditions. With a time delay of 10 RF cycles, the ion density in the plasma bulk grows to $1.2 \times 10^{17}$ m\textsuperscript{-3} and $8.1 \times 10^{16}$ m\textsuperscript{-3} in the RF discharge with and without pulsed discharge. This suggests that the development of RF discharge ignition with pulsed discharge is faster than that without pulsed discharge. The ion density in the plasma bulk achieves the same magnitude of $3.7 \times 10^{17}$ m\textsuperscript{-3} with a time delay of 20 RF cycles, which means that the RF discharge operates in the stable state. The spatial profile of ion density across the discharge gap always shows the bell shape, which is due to the fact that the production of ions is mostly located in the boundary region between the sheath and plasma bulk and the ions are trapped in the plasma bulk by the oscillating RF electric field.\textsuperscript{14}

The spatiotemporal evolution of electron density during the first 20 RF cycles of the discharge ignition phase with pulsed discharge is shown in Fig. 7(a). The spatial profile of electron density in the discharge gap has the bell shape with a high concentration of electrons in the middle of the discharge gap. Before the ignition of the discharge, the residual electrons in the discharge gap will travel toward both the electrodes driven by diffusion. As the RF discharge is ignited and sheath regions form above the electrodes, the electrons in the discharge gap will be trapped by the oscillating RF electric field. With the ignition of discharge, the electrons in the discharge gap keep the spatial profile and grow in magnitude.

In the initial conditions with the time delay of 0 RF cycles, as shown in Fig. 7(b), the electron density in the plasma bulk is $3.45 \times 10^{16}$ m\textsuperscript{-3} with pulsed discharge, which is much higher than $0.74 \times 10^{16}$ m\textsuperscript{-3} without pulsed discharge. This difference can be attributed to the electrons generated in the previous pulsed discharge.\textsuperscript{13} With the time delay of 10 RF cycles, the electron density in the plasma bulk develops to $2.51 \times 10^{17}$ m\textsuperscript{-3} and $1.31 \times 10^{17}$ m\textsuperscript{-3} in the RF discharge with and without pulsed discharge, and the sheath region above both electrodes is enhanced, as indicated by the expanding of plasma bulk region. It suggests that with the higher electron density in the discharge initial conditions, the RF discharge develops quicker in the ignition phase. As the time delay evolves to 20 cycles, the spatial profile of electron density in Fig. 7(b) illustrates that the RF discharge both without and with pulsed discharge achieves the stable state with the same operation parameters.

The spatiotemporal evolution of the electric field during the first 20 RF cycles of the discharge ignition phase with pulsed discharge.
FIG. 7. (a) Spatiotemporal evolution of electron density in the RF discharge ignition phase. (b) Spatial profile of electron density with time delays of 0, 10, and 20 RF cycles (solid and dashed lines represent RF discharge with and without pulsed discharge).

discharge is shown in Fig. 8(a). It is worth noting that the electric field is temporally averaged during each RF cycle, which suggests that the electric field is mostly dependent on the built-in electric field in the discharge gap induced by the space charges and can be employed to indicate the regimes of plasma bulk and sheath. The temporal evolution of sheath regions with enhanced electric field above both electrodes is clearly illustrated in Fig. 8(a). In the initial conditions with the time delay of 0 RF cycles, as shown in Fig. 8(b), the electric field in the discharge gap is close to zero except at the surface of electrodes, whose magnitudes of $0.17 \times 10^5$ V/m and $0.03 \times 10^5$ V/m can be recognized in the inset figure in the RF discharge with and without pulsed discharge, respectively. This electric field is induced by the space charge in the vicinity of the electrode because of the higher depletion rate of electrons than that of ions. The higher magnitude of the electric field in the RF discharge with pulsed discharge suggests the much more charged particles in the discharge gap in the initial conditions, which has been demonstrated in Figs. 6 and 7. With the time delay of 10 RF cycles, the sheath regions are formed above both electrodes, as shown in Fig. 8(a). With the pulsed discharge, the electric field on the electrode surface reaches $5.5 \times 10^5$ V/m in RF discharge, which is higher than $3.3 \times 10^5$ V/m in RF discharge without pulsed discharge. The sheath region indicated by the spatial profile of the electric field shows that the sheath thickness is lower in RF discharge with pulsed discharge than that without pulsed discharge. It suggests that the enhancement of the sheath is higher in RF discharge with pulsed discharge than that without pulsed discharge. The enhancement of the sheath is higher in RF discharge with pulsed discharge than that without pulsed discharge. It can also be attributed to be residual plasma species in the discharge gap from the pulsed discharge. As the time delay reaches 20 cycles with the stable operation of RF discharge, the electric field at the electrode surface reaches $6.5 \times 10^5$ V/m in both RF discharges with and without pulsed discharge.
V. CONCLUSIONS

In summary, the discharge ignition characteristics in atmospheric cascade discharge with pulsed discharge and RF discharge burst are determined by the initial conditions in terms of the density and spatial distribution of residual plasma species in the discharge gap. In the pulsed discharge, the second discharge in the pulse voltage falling phase develops quicker with higher discharge current density than the first discharge in the pulse voltage rising phase, which is attributed to the propagation of charged particles (e, He+, and He2+) driven by the reversed electric field in the discharge gap. With the pulsed discharge generated before the ignition of RF discharge, the initial conditions are enhanced in terms of densities of electrons and ions and the electric field above the electrode surface, compared to the initial discharge conditions in RF discharge without pulsed discharge. The enhancement of the initial discharge conditions helps the ignition of RF discharge by developing the sheath regions above both electrodes and helps achieve the stable operation of RF discharge, which is ascribed to the oscillation of residual electrons in the discharge gap upon applying the RF electric field. It demonstrates that the dynamics of residual plasma species, especially the charged particles, in the discharge gap during the discharge ignition phase play important roles in the discharge ignition and discharge characteristics.

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