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NUMERICAL SIMULATION OF THE BEHAVIOR OF NEGATIVE IONS IN A HIGH-FREQUENCY CAPACITIVE DISCHARGE IN CARBON DIOXIDE

V. Lisovskiy, S. Dudin, A. Shakhnazarian, P. Platonov, V. Yegorenkov
V.N. Karazin Kharkiv National University, Kharkiv, Ukraine
E-mail: lisovskiy@yahoo.com

In the present work, the dynamics of axial profiles of negative ions and the degree of electronegativity $a = N_n / N_e$ of different parts of the RF capacitive discharge in carbon dioxide are investigated using the SIGLO-rf fluid code. Time-averaged axial profiles of densities of electrons N_e, positive N_p and negative N_n ions are also obtained in a wide range of gas pressures and applied RF voltages. It is shown that at a sufficiently high gas pressure (1 Torr and higher), the quasi-neutral plasma consists predominantly of positive and negative ions. The degree of electroneutrality a is about several tens and decreases with increasing RF voltage. Because of oscillations in the RF electric field, electrons are ejected every half-period from a near-electrode sheath to the sheath near the opposite electrode being the temporary anode. This leads to a significant increase in the degree of electronegativity in the sheath abandoned by the electrons. At low carbon dioxide pressures (below 0.17 Torr), the plasma consists predominantly of electrons and positive ions with a very low negative ion density, and the degree of electronegativity is significantly less than unity.

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INTRODUCTION

Discharges in electronegative gases are used to generate negative ion flows [1–4], heat thermonuclear plasma [5], for plasma etching of semiconductor materials and deposition of thin films [6, 7]. In the etching and deposition technologies, the radio-frequency capacitive discharge burning in gases containing fluorine, hydrogen, and other electronegative components is most widely used. Therefore, much attention is paid to both experimental and numerical studies of its properties [7–22].

In electronegative gases, either direct capture of a free electron by a gas molecule occurs (for example, in SF_6, SF_6^- ions are formed in this way), or dissociative attachment occurs with the detachment of one of the atoms from the main molecule, which becomes a negative ion: $CO_2 + e^- \rightarrow CO + O^-$.

Free electrons moving in an electric field carry the discharge current, gain sufficiently high energy, and create new ion-electron pairs in ionizing collisions with gas molecules. Electrons attached to gas molecules with the negative ion formation lose the ability to produce ionization. Current transfer by heavy negative ions is usually negligible due to their low mobility in an electric field. Both of the above-mentioned attachment processes lead to the loss of free electrons and, as a result, to the obstruction of burning a discharge.

The accumulation of negative ions in the plasma can significantly change the current transfer processes [23, 24], up to the transition of ambipolar diffusion to free diffusion [24] at high values of the degree of electronegativity $a = N_n/N_e \approx 100…1000$. Therefore, the study of the processes of generation and accumulation of negative ions in the plasma of a radio-frequency capacitive discharge is an important problem.

To solve this problem, we used the SIGLO-rf fluid code, which allowed us to reveal the behavior of negative ions in RF capacitive discharge in carbon dioxide in a wide pressure range. This approach allowed us to calculate the evolution of the axial distribution of a when changing the discharge parameters.

1. DESCRIPTION OF SIGLO-RF CODE

Prof. Boeuf and Prof. Pitchford from the University of Toulouse published papers [25, 26] describing numerical simulation of processes occurring in radio-frequency capacitive discharge. Based on the equations and assumptions described in [25, 26] they developed the fluid code SIGLO-rf. Our group has already used it for numerical modeling of the properties of radio-frequency capacitive discharge in oxygen [19], argon [27], and carbon dioxide [28]. A detailed description of the equations solved by the SIGLO-rf code is given in a recent paper [28]. Therefore, here we limit ourselves to a brief analysis of the processes considered by the code.

Since carbon dioxide is electronegative, in this case, the balance equations for free electrons, and positive and negative ions are solved numerically. For each of the listed types of charged particles, the momentum equations are solved simultaneously, and for electrons, the energy equation is also solved. The Poisson equation is also used to calculate the electric field strength. All the necessary constants of the processes of generation and loss of charged particles are described in [28].

The calculations were performed for the distance between the vertically located electrodes of 2 cm. The RF voltage of 13.56 MHz frequency was applied to the right electrode. The RF voltage amplitude was varied from the minimum voltage of maintaining the discharge (extinction curve) to 1000 V. The left electrode was under the Earth's potential. The calculations were performed in the range of carbon dioxide pressures from 0.07 to 10 Torr. At pressures below 0.07 Torr, the discharge was not maintained, i.e. after the start of the calculations, the plasma density rapidly decreased from the initial value (10^8 cm^{-3}) down to zero. For each case shown in the figures below, the calculations were performed for 100,000 periods of the RF electric field.

Note that after about 20,000 periods a steady state was reached, and the plasma parameters stopped changing during further calculations.

2. RESULTS OF MODELING

Let us first consider the results for a carbon dioxide pressure of 1 Torr. The average axial profiles of the charged particle densities over the RF period, as well as the density ratio $a = N_n / N_e$ at RF voltage amplitudes of 100 and 1000 V, are shown in Figs. 1 and 2, respectively. At such a gas pressure, electron density maxima are visible near the boundaries of the near-electrode layers due to the formation of the so-called “active” double layer in these regions [28]. It is intended to balance the number of charges of opposite signs arriving at the surface of each of the electrodes during the RF period. At an RF voltage amplitude of 100 V, the density of negative ions in the quasi-neutral plasma almost reaches the density of positive ions, and the electron density is almost an order of magnitude lower. Since the electrons enter the near-electrode sheath for a certain part of the period, their average density decreases closer to the electrode surface.

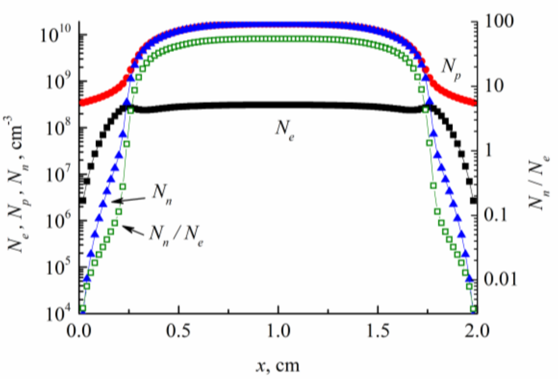

Fig. 1. Time-averaged axial profiles of the density of electrons N_e, positive N_p and negative ions N_n in comparison with N_n / N_e ratio at carbon dioxide pressure of 1 Torr and the RF voltage amplitude of 100 V

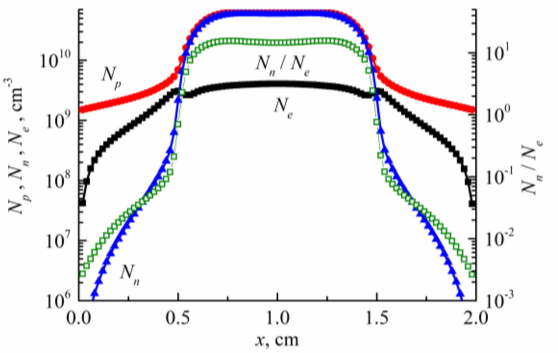

Fig. 2. Time-averaged axial profiles of the density of electrons N_e, positive N_p and negative ions N_n in comparison with N_n / N_e ratio at carbon dioxide pressure of 1 Torr and the RF voltage amplitude of 1000 V

Negative ions are in a potential well formed by the time-averaged electric field, and cannot reach the surface of the electrodes. Negative ions present in the sheaths were formed there due to attachment to molecules of free electrons, which periodically fill the near-electrode sheath. These ions are gradually pushed out of the layers into the plasma volume. The degree of electronegativity $a = N_n/N_e$ under the conditions of Fig. 1 reaches 54 (Fig. 3). When the voltage is reduced to 85 V (the minimum voltage for maintaining the discharge), it increases to a = 92. Increasing the RF voltage amplitude to 1000 V leads to a decrease in the degree of electronegativity a to 15 (see Figs. 2 and 3). The reason for this is the density increase of not only electrons and negative ions but also positive ions. Positive and negative ions recombine with each other in the plasma volume, which reduces the magnitude of the degree of electronegativity.

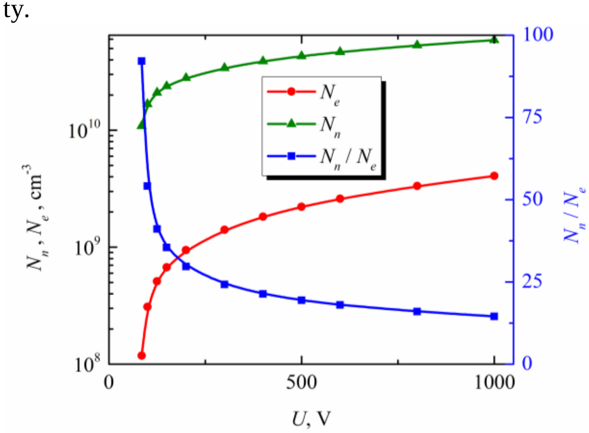

Fig. 3. Time-averaged dependencies of the density of electrons N_e and negative ions N_n in the discharge center and N_n / N_e ratio at carbon dioxide pressure of 1 Torr on the RF voltage amplitude

We have considered the time-averaged axial profiles of the charged particle densities and the degree of plasma electronegativity. Now let us find out how these axial profiles change during the period of the RF electric field. At the initial moment of the period (t/T = 0), when a positive amplitude value of voltage is applied to the right electrode, this electrode is the instantaneous anode and collects the electrons arriving at it (Fig. 4). At this moment, the electrons fill the right sheath, and their axial distribution practically coincide with the distribution of positive ions. Here, the electron density is significantly greater than the density of negative ions. Therefore, in the right sheath, the degree of electronegativity is small, of the order of 1 near the sheath boundary, and then rapidly decreases when approaching the surface of the right electrode.

In the left sheath, near the instantaneous cathode, the situation is the opposite. The electrons are pushed out of the left sheath by the electric field, but there are negative ions there that have not had time to leave the layer. Therefore, when approaching the surface of the left electrode (“cathode”), the degree of electronegativity increases sharply, not due to the increased negative ion density, but due to the absence of electrons in this sheath.

Now let us consider the axial profiles for the moment t/T = 0.35, shown in Fig. 5. This moment of time corresponds to the maximum value of the RF discharge current. The electron profile is close to symmetrical relative to the center of the discharge gap. The electrons are partially ejected from the right sheath and partially fill the left sheath. This profile shows local maxima near the sheath boundaries, which correspond to the formation of passive (near the boundary of expanding right sheath) and active (near the boundary of the left layer, which is in the anodic phase) double layers [28]. Our calculations show that in the region of these double layers, the degree of electronegativity has deep minima with $a < 0.1$. In the quasi-neutral plasma $a \approx 15$, and in the layers, a sharply increases when approaching the electrode surface due to the temporary absence of electrons there.

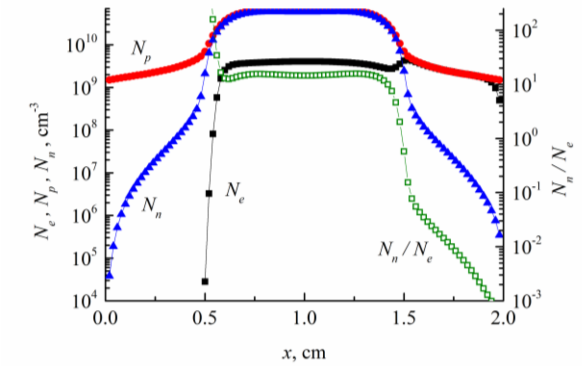

Fig. 4. Axial profiles of the density of electrons N_e, positive N_p and negative ions N_n in comparison with N_n / N_e ratio at carbon dioxide pressure of 1 Torr and the RF voltage amplitude of 1000 V for the moment $t/T = 0$

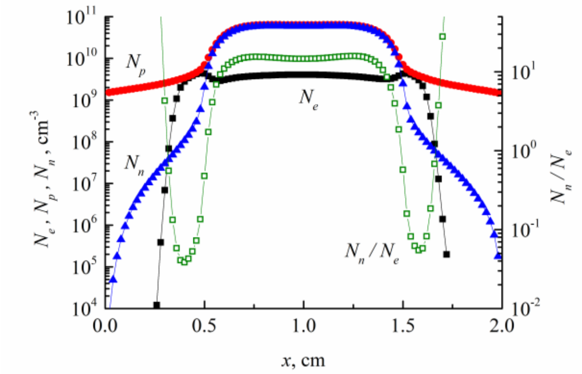

Fig. 5. Axial profiles of the density of electrons N_e, positive N_p and negative ions N_n, in comparison with N_n / N_e ratio at carbon dioxide pressure of 1 Torr and the RF voltage amplitude of 1000 V for the moment $t/T = 0.35$ corresponding to the peak value of RF discharge current

It is not expedient to consider the case for the time moment $t/T = 0.5$, since in this case, the axial profiles are mirror-reflected relative to the center of the discharge gap compared to $t/T = 0$ moment.

During the first half of the RF period, we see three maxima on the axial profile of the electron attachment rate. The first of them is observed in the region of the appearance of the first (passive) double layer (in Fig. 6 its position is shown as DL_1). The second maximum is more extended and located in the bulk of quasi-neutral plasma (PL). The third sharp maximum occurs due to the formation of the active double layer (DL_2) when the electron cloud enters the near-electrode sheath, which is in the anode phase.

In the second half of the RF period, the situation changes to the opposite, the passive and active double layers in the first half of the period are transformed, respectively, into the active and passive double layers in the second half of the RF period (see Fig. 6).

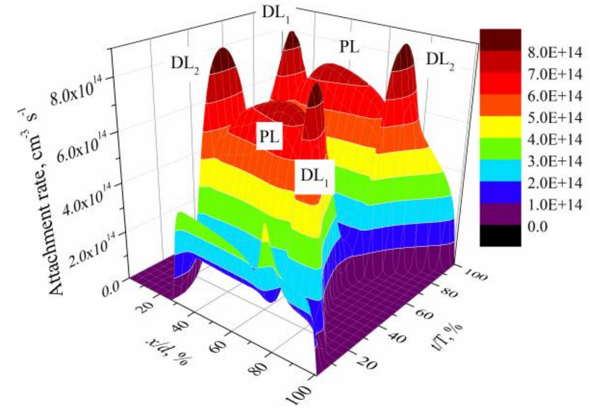

Fig. 6. Spatio-temporal dependence of the electron attachment rate at carbon dioxide pressure of 1 Torr and the RF voltage amplitude of 1000 V

We mentioned above that the axial profile of the negative ion density changes slightly during the RF period at a carbon dioxide pressure of 1 Torr. This is seen in Fig. 7, where the corresponding profiles are given for the moments of time $t/T = 0$ and $t/T = 0.5$. Small differences in these profiles are visible only near the electrode surface, while they coincide in the quasi-neutral plasma. Due to lower mobility at elevated pressure, the negative ions generated due to electron attachment in a sheath do not have time to leave the sheath in half the RF period, even at high values of the RF voltage amplitude.

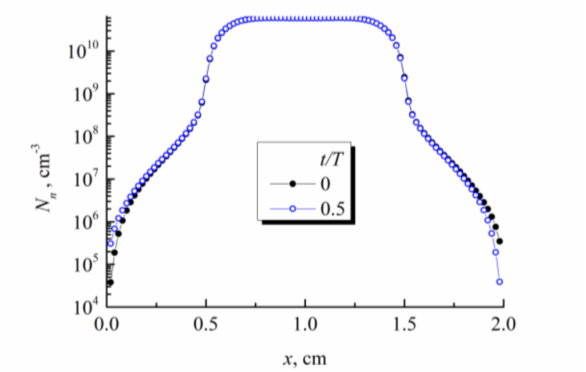

Fig. 7. Axial profiles of the density of negative ions N_n at carbon dioxide pressure of 1 Torr and the RF voltage amplitude of 1000 V for the moments $t/T = 0$ and $t/T = 0.5$

Now let us consider the case of low carbon dioxide pressure $p = 0.07$ Torr. In this case, the near-electrode sheaths are thick and overlap in the center of the discharge gap. The electron cloud runs successively from one electrode to another, which we see in Figs. 8 and 9 for the moments $t/T = 0$ and $t/T = 0.245$. The electron density profile tends to the profile of positive ions, maintaining quasi-neutrality in this region.

Since the gas pressure is low, this leads, firstly, to the fact that the density profile of positive ions becomes almost uniform throughout the gap between the electrodes. Near the electrodes, $N_p = 2.4 \cdot 10^8$ cm^{-3}, and in the central part of the discharge $N_p = 4 \cdot 10^8$ cm^{-3}, i.e. the density of positive ions changes less than 2 times over the discharge gap. At the 1 Torr pressure, this difference was greater than 40 times.

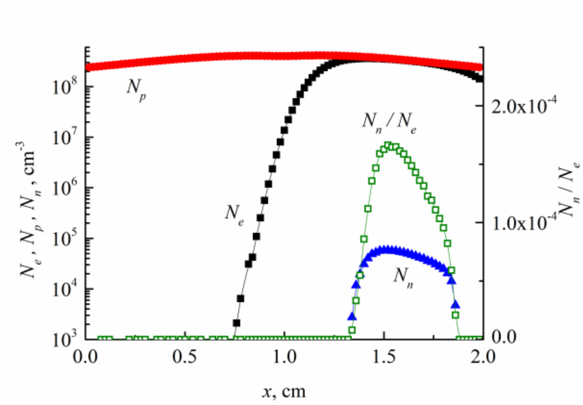

Fig. 8. Axial profiles of the density of electrons N_e, positive N_p and negative ions N_n, in comparison with N_n / N_e ratio at carbon dioxide pressure of 0.07 Torr and the RF voltage amplitude of 500 V for the moment t/T = 0

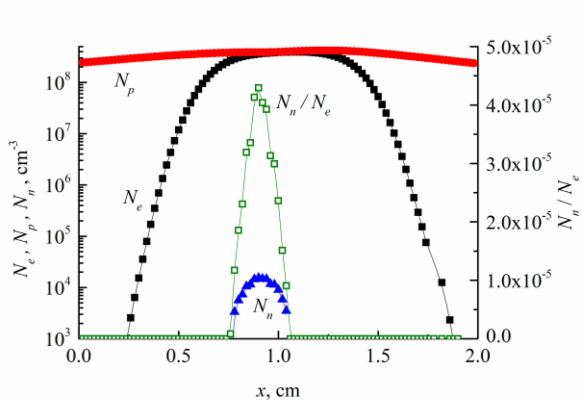

Fig. 9. Axial profiles of the density of electrons N_e, positive N_p and negative ions N_n, in comparison with N_n / N_e ratio at carbon dioxide pressure of 0.07 Torr and the RF voltage amplitude of 500 V for the moment t/T = 0.245 corresponding to the maximum value of RF discharge current

Secondly, the decrease in gas pressure significantly increased the mobility of negative ions and also substantially narrowed the region where they can appear. We see this in Figs. 8 and 9, where the width of the negative ion density peak turned out to be noticeably narrower than for electrons. The electrons most actively stick to gas molecules in the region where there are many electrons themselves and where the rate of the attachment process will be high. Using the BOLSIG+ code [29] with a set of cross sections from [30], we showed that the attachment rate in carbon dioxide is maximum at the reduced electric field value of $E/N \approx 300$ Td. At the gas temperature of 300 K, this corresponds to the value of $E/p \approx 100$ V/(cm·Torr). For the carbon dioxide pressure of 0.07 Torr, the electric field strength should be about 7 V/cm. From Fig. 10 we see that this electric field strength is very low and the region with such a weak field is located near the sign reversal point of the electric field. Therefore, the process of electron attachment to gas molecules occurs mostly in a narrow region, which we see in Figs. 8 and 9. Since the density of negative ions is small compared to the density of electrons, the degree of electronegativity is low during the entire RF period.

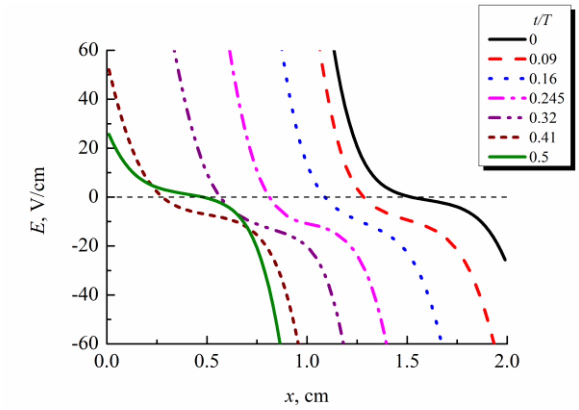

Fig. 10. Axial profiles of electric field strength at carbon dioxide pressure of 0.07 Torr and RF voltage amplitude of 500 V for different moments of time t/T

Note also that the electron attachment region actively moves between the electrodes during the RF period. The corresponding spatio-temporal distribution of the electron attachment rate is shown in Fig. 11. This figure shows that bursts of the electron attachment rate are observed at the moments of expansion of the near-electrode sheath. When one of the electrodes becomes a temporary cathode, it pushes out the electrons that fill the sheath during the anodic phase. In this case, the electrons acquire additional energy, which is spended both on the ionization of gas molecules and on the processes of dissociative attachment. In the next half-period, the opposite electrode (which was previously a temporary anode and attracted electrons to itself) becomes a temporary cathode; now the electrons are pushed out of the sheath near this electrode. In Fig. 11, a peak in the electron attachment rate appears, corresponding to this process.

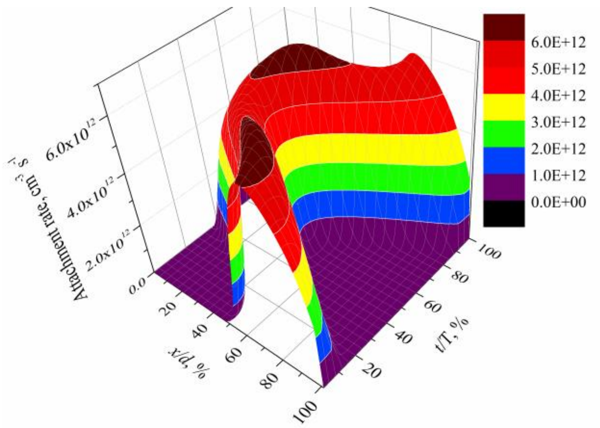

Fig. 11. Spatio-temporal dependence of the electron attachment rate at carbon dioxide pressure of 0.07 Torr and the RF voltage amplitude of 500 V

Now let us reveal the behavior of the charged particle densities and the degree of electronegativity in the center of the discharge gap in a wide range of carbon dioxide pressures (Fig. 12). At gas pressures starting from 0.07 and up to 0.15 Torr, the concentration of negative ions is low, so the degree of electronegativity a is significantly lower than 1. The plasma actually consists of electrons and positive ions. At the pressure of 0.17 Torr, the densities of negative ions and electrons are equalized, and the level of $a = 1$ is reached. With the further increase in carbon dioxide pressure, the densities of negative and positive ions are approximately equal to each other, and the electron density is less than 10% of their value. Thus, in a wide range of pressures, the degree of electronegativity remains approximately in the range of $a = 15…30$ at an RF voltage amplitude of 500 V.

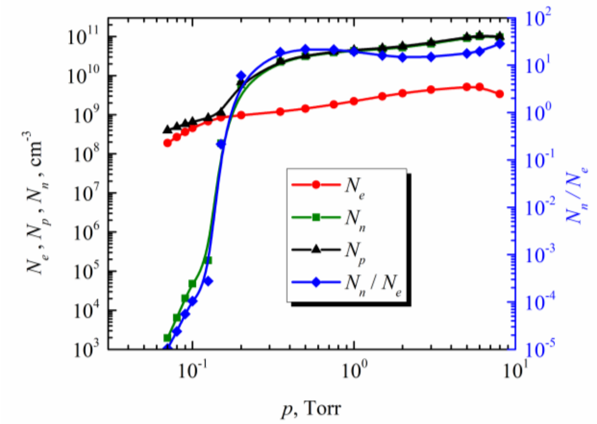

Fig. 12. Pressure dependences of time-averaged densities of electrons N_e, positive N_p and negative ions N_n in the discharge center in comparison with N_n / N_e ratio at RF voltage amplitude of 500 V

CONCLUSIONS

In the present work, the time-average and space-time dependences of the profiles of electrons, positive and negative ions in a wide range of carbon dioxide pressures and applied RF voltages are obtained using the SIGLO-rf fluid code. Thanks to them, the behavior of the degree of electroneutrality $a = N_n / N_e$ is clarified for different parts of the RF capacitive discharge. At a sufficiently high gas pressure (1 Torr and higher), the axial profile of the density of negative ions remains virtually unchanged during the RF period of the electric field. Positive and negative ions dominate in the volume of the quasi-neutral plasma, and the degree of electronegativity a monotonically decreases with increasing RF voltage, but remains in the range of several tens. The time dependences of the density profiles of charged particles showed that electrons are expelled from a near-electrode sheath every half-period of the RF field and fill the sheath near the opposite electrode. Then in the sheath from which the electrons leave, the degree of electronegativity increases sharply. At carbon dioxide pressures below 0.17 Torr, the negative ion density becomes insignificant, and the plasma consists of electrons and positive ions. Because of the high mobility at low pressures, the cloud of negative ions oscillates between the electrodes together with the electrons with a large amplitude.

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ЧИСЕЛЬНЕ МОДЕЛЮВАННЯ ПОВЕДІНКИ НЕГАТИВНИХ ІОНІВ У ВИСОКОЧАСТОТНОМУ ЄМНІСНОМУ РОЗРЯДІ У ВУГЛЕКИСЛОМУ ГАЗІ

В. Лісовський, С. Дудін, А. Шахназарян, П. Платонов, В. Єгоренков

Динаміка осьових профілів негативних іонів та ступеня електронегативності $a = N_n / N_e$ різних частин ВЧ-ємнісного розряду у вуглекислому газі досліджена за допомогою гідродинамічного коду SIGLO-rf. Отримані також середні за часом осьові профілі концентрацій електронів N_e, позитивних N_p і негативних N_n іонів у широкому діапазоні тиску газу та прикладеної ВЧ-напруги. Показано, що при досить високому тиску газу (1 Торр і вище) квазінейтральна плазма складається переважно з позитивних і негативних іонів. Ступінь електронегативності a приблизно дорівнює кільком десяткам і знижується зі зростанням ВЧ-напруги. Через осциляції у ВЧ-електричному полі електрони кожні півперіоду виштовхуються з одного приелектродного шару в протилежний шар біля тимчасового електрода-анода. Це призводить до значного зростання ступеня електронегативності у шарі, який залишили електрони. При низьких тисках вуглекислого газу (нижче 0,17 Торр) плазма переважно складається з електронів і позитивних іонів з дуже низькою концентрацією негативних іонів, а ступінь електронегативності істотно менше одиниці.