Mishin, Maxim; Protopopova, Vera; Andey, Uvarov; Sergey, Alexandrov

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Spatial distribution of the electrical potential and ion concentration in the downstream area of atmospheric pressure remote plasma

M. V. Mishin,1,a V. S. Protopopova,2,b A. A. Uvarov,1 and S. E. Alexandrov1

1Department of Physical Chemistry and Technology of Microsystem Devices, Saint Petersburg State Polytechnical University, Saint Petersburg, 195251, Russia
2Department of Materials Science and Engineering, Aalto University Espoo, P.O. Box 16200, Finland

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This paper presents the results from an experimental study of the ion flux characteristics behind the remote plasma zone in a vertical tube reaction chamber for atmospheric pressure plasma enhanced chemical vapor deposition. Capacitively coupled radio frequency plasma was generated in pure He and gas mixtures: He–Ar, He–O2, He–TEOS. We previously used the reaction system He–TEOS for the synthesis of self-assembled structures of silicon dioxide nanoparticles. It is likely that the electrical parameters of the area, where nanoparticles have been transported from the synthesis zone to the substrate, play a significant role in the self-organization processes both in the vapor phase and on the substrate surface. The results from the spatial distribution of the electrical potential and ion concentration in the discharge downstream area measured by means of the external probe of original design and the special data processing method are demonstrated in this work. Positive and negatives ions with maximum concentrations of 10^6–10^7 cm^{-3} have been found at 10–80 mm distance behind the plasma zone. On the basis of the revealed distributions for different gas mixtures, the physical model of the observed phenomena is proposed. The model illustrates the capability of the virtual ion emitter formation behind the discharge gap and the presence of an extremum of the electrical potential at the distance of approximately 10^{-2}–10^{-1} mm from the grounded electrode.

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I. INTRODUCTION

Various types of discharges1–4 have been widely used for the synthesis of different nanomaterials, such as thin films,5–8 nanoparticles,9–11 nanotubes12–14 and nanocomposites.15,16 The applications of atmospheric pressure (AP) discharges conducted at radio frequency (RF) are of high interest due to several advantages, for instance, (i) the convenience of use and the simplicity of hardware design; (ii) high concentrations of precursors in the vapor phase and, as consequence, high product yield; (iii) low thermal input to the substrate due to intrinsic low-temperature nature. A particular feature of AP plasma-assisted processes is that homogeneous reactions promote the nanoparticle formation,9–11 which then can build self-assembled structures.

We have previously demonstrated17,18 the capability of the fractal cluster formation and the regularities of the morphology dynamics of the deposit consisted of SiO2 nanoparticles during atmospheric pressure plasma-enhanced chemical vapor deposition (AP-PECVD) from tetraethoxysilane (TEOS). Using the experimental setup with remote plasma, nanoparticles have been transferred from the syn-

[a]Electronic mail: m.v.mishin.spbstu@gmail.com
[b]This research was performed while V. S. Protopopova was at Department of Physical Chemistry and Technology of Microsystem Devices, Saint Petersburg State Polytechnical University, Saint Petersburg, 195251, Russia.
thesis area to the substrate by a carrier gas containing a significant amount of ions, which finally leads to the acquisition of electrical charge by nanoparticles. Additionally, the deposit surface can be charged by ion bombardment. Electrical forces, which arise owing to the uneven charge distribution of the deposit surface, can lead to self-organization of nanoparticles and rearrangement of the surface morphology.\textsuperscript{17,19}

Inhomogeneity of the electric field spatial distribution in the PECVD reaction chamber can be a crucial factor responsible for agglomeration and coalescence of nanoparticles synthesized from the vapor phase.\textsuperscript{20,21} Interaction of charged nanoparticles and dipoles with uneven electric fields of plasma can change their velocities and alter spatial allocation. Such processes have been thoroughly investigated and covered extensively by articles devoted to dusty plasmas,\textsuperscript{22–26} but most of those results were obtained in low pressure systems.

Novelty of the presented below results is caused by the fact that the electrical characteristics have been studied in the reaction chamber with strongly localized plasma. The localization area was defined by the position of both electrodes. The linear size of plasma zone was much less then the size of the downstream area, \textit{i.e.} the distance between the bottom electrode and the substrate.\textsuperscript{11,17,18} The used chamber and electrode design was entirely different from such well studied systems equipped by remote low-temperature plasma and operated under atmospheric pressure as jet systems,\textsuperscript{27–29} corona discharge systems,\textsuperscript{30–32} microwave discharge systems.\textsuperscript{33–35} In the cases of all above mentioned systems, plasma (i) was spatial inhomogeneous and (ii) freely collapsed in the chamber area between the discharge area and the substrate. In addition, we would like to emphasis on the difference between atmospheric and low pressure discharges. Atmospheric pressure plasma is characterized by the short mean free path of charge carriers, which lead to electron absence outside of discharge zone in comparison to low pressure discharges.\textsuperscript{36–39}

The goal of this work is to determine ion concentration and the spatial distribution of the electrical potential in the downstream area of remote RF plasma between the discharge gap and the substrate in various gas mixtures at atmospheric pressure.

II. EXPERIMENTAL DETAILS

The measurements were performed in the vertical tube reaction chamber.\textsuperscript{11,17,18} Capacitively coupled plasma (CCP) was generated and sustained in the gap between two plane parallel mesh electrodes (24 mm in diameter) made of stainless steel. The electrodes, separated by 2 mm, were placed perpendicular to the gas flow direction. RF power (operation frequency 13.56 MHz) was applied to the top electrode, the bottom electrode was grounded. The RF circuit included a variometer, which was installed between the RF power source and the discharge gap for RF matching. The discharge current and the voltage were acquired by means of a current transformer and a voltage divider, respectively, and then were multiplied with a measured power factor to get a true discharge driving power. In all the experiments the power value was kept at 15 W.

High-purity helium (oxygen concentration of no more than 1 ppm) was used as the plasma-forming gas and the carrier gas. Besides pure helium, the measurements were also performed in mixtures of He–Ar (5%), He–O\textsubscript{2} (1%) and He–TEOS. The silicon-organic precursor TEOS had been widely studied as a reagent for the silicon dioxide film formation for microelectronics applications. TEOS vapor was carried from a temperature-stabilized (25 °C) evaporator to the reaction chamber by helium flow fixed at 100 sccm. The total gas flow was 350 sccm. Under these conditions, TEOS concentration was 5x10^{-5} g/cm\textsuperscript{3} in the reaction chamber. Prior to the experiments, the setup was purged with He flow during 15 min.

The measurements of the electric current $I$ carried by ions from the discharge gap were performed using the specially designed external probe (Fig. 1(a)) placed along the reaction chamber axis. The probe was 6 mm in diameter and mounted inside a semispherical screen (7 mm in diameter) made from stainless steel mesh with a width of 0.7 mm x 0.7 mm and a transparency of 80%. The distance between the grounded electrode and the probe was varied in the range from 10 to 100 mm.

The electronic circuit used for the probe measurements is shown in Fig. 1(b). The probe was biased with the periodic variable sawtooth voltage $U_{\text{ext}}$ with a frequency of 10 Hz and a peak-to-peak
FIG. 1. (a) Probe design and (b) the circuit for the current-voltage curves registration by means of the probe. Notations: EL1, EL2 – plane parallel electrodes, Q – gas flow direction (He), PR – external probe, TrAMP – transimpedance amplifier, OI – optical isolator, AMP1 and AMP2 – voltage amplifiers, SGEN – signal generator, OSC – oscilloscope.

Voltage of ±50 V. This voltage was formed by means of a signal generator (SGEN) and a voltage amplifier (AMP1). The ion current collected by the probe PR was supplied to the input of a transimpedance amplifier TrAMP. The local “ground” and input of transimpedance amplifier were offset relative to the “measuring ground” by the value of the probe bias $U_{\text{ext}}$. The signal from the output of the transimpedance amplifier, which was corresponded to the probe current $I$, was delivered to an oscilloscope (OSC) “Y” channel through an optical isolator (OI) and an amplifier (AMP2), and was registered relative to the “instrument ground.” The instantaneous value of $U_{\text{ext}}$ was also measured by the oscilloscope relative to the “instrument ground” (“X” channel). Thus, the circuit allowed for acquiring the sets of multiple probe current-voltage curves during a single experiment, which were further averaged and processed. A typical waveform showing temporal behavior of the registered probe current $I$ and the bias voltage $U_{\text{ext}}$ are shown in Fig. 2(a).

III. RESULTS AND DISCUSSION

The probe current-voltage characteristics were measured in downstream area at various distances $d$ from the discharge gap in pure He, as well as in He–Ar, He–$O_2$ mixtures and the reaction...
FIG. 3. Schematic representation of the ion current balance near the external probe. Notations: $Q$ – gas flow, $j_{dr}$ – drift ion current density, $j_{diff}$ – diffusion current density, $j_{fl}$ – ion current density transferred by gas flow, $R_1$ – screen radius, $R_2$ – probe radius, $R_{ch}$ – reaction chamber radius.

system He–TEOS. The curves obtained in pure He are displayed in Fig. 2(b). The curves were found to have strong resemblance with that shown in Fig. 2(b) for other gas mixtures.

We considered the balance of the current components contributing to the measured current $I$ (Fig. 3). Ion entry to the volume surrounded by the semispherical screen can be caused by transfer with gas flow $Q$ and diffusion through the screen. Ion removal from this volume occurs due to the drift in the electric field created by the potential difference between the probe and the screen.

Our consideration proceeded on the assumption that the positive current was caused by helium ions $\text{He}^+$, whereas the negative current was determined by oxygen ions $\text{O}_2^-$, which were generated by low-energy electrons attachment to oxygen impurity molecules. Oxygen molecules originated from residual gases in the experimental setup volume after purging and from impurities of used He (less than 1 ppm according to the specifications).

Preliminary experiments demonstrated that the negative current was caused only by negative ions, but not by electrons. Mobility of negative charge carriers was estimated by response time of the negative current on the voltage impulse with short front. The found mobility value was several cm$^2$/V s units, which is three order of magnitude less then electron mobility in considered conditions.

Initially, we made an assumption that concentrations of the charge carriers were low, which allowed us to neglect ion interactions.

Ion removal from the volume surrounded by the semispherical screen is induced by the drift ion current density $j_{dr}$ (1a) to the probe.

$$j_{dr} = \pm qn_\pm \mu_\pm E,$$

where $q$ – charge carried by ions, $n$ – concentration of positive or negative ions, $\mu$ – mobility of positive or negative ions, and $E$ – electric field between the screen and the probe, which is defined as the electric field of a semispherical capacitor according to (1b).

$$E = \frac{\pm U_{ext} R_1 R_2}{R_1 - R_2} \frac{1}{r^2},$$

where $R_1$ – radius of the outer hemisphere (the screen), $R_2$ – radius of the inner hemisphere (the probe), and $r \in [R_2, R_1]$. 
Thus, combining (1a) and (1b) the expression for the drift current density on the probe is rewritten as:

\[ j_{dr} = \pm q \mu_\pm n_\pm \frac{U_{ext}}{R_1 - R_2} \]  

(1c)

Next, we evaluated how ions could enter the volume surrounded by the screen. One way is diffusion through the screen. With the assumption of low concentration of the charge carriers, ambipolar diffusion can be neglected. Therefore, the diffusion current density \( j_{diff} \) is presented as:

\[ j_{diff} = \pm q D_\pm \frac{dn_\pm}{dx}, \]  

(2a)

where \( D_\pm \) – diffusion coefficient of positive or negative ions given via expression (2b):

\[ D_\pm = \frac{1}{3} \bar{v} \lambda_\pm = \frac{1}{q} \mu_\pm kT, \]  

(2b)

\[ \bar{v} = \sqrt{\frac{3kT}{m}}, \]  

(2c)

where \( \bar{v} \) – average thermal velocity, \( \lambda_\pm \) – mean free path of positive or negative ions, \( k \) – Boltzmann constant, \( T \) – gas temperature, and \( m \) – molecule mass.

Substituting (2b) and (2c) for (2a) and using finite difference instead of concentration derivative one can get \( j_{diff} \):

\[ j_{diff} \approx \pm \mu_\pm kT \frac{n_\pm}{R_1 - R_2}. \]  

(2d)

The other way for ions to enter the screen volume is transfer by gas flow. The corresponding current density \( j_{fl} \) is described as follows:

\[ j_{fl} = \pm q n_\pm v_{fl} \frac{S_{scr}}{S_{ch}} = \pm q n_\pm Q \frac{S_{scr}}{S_{ch}} \frac{S_{scr}}{S_{ch}}, \]  

(3)

where \( v_{fl} = Q/S_{ch} \) – linear flow velocity, \( S_{scr} \) – cross-section of the screen (normal to the gas flow direction), and \( S_{ch} \) – cross-section of the reaction chamber.

We estimated the contribution of the evolved current components to the probe current \( I \). The ratio of the diffusion current density \( j_{diff} \) to the ion current density transferred by gas flow \( j_{fl} \) is considered as:

\[ \frac{j_{flip}}{j_{diff}} = \pm q n_\pm Q \frac{S_{scr}}{S_{ch}} \frac{S_{scr}}{S_{ch}} = \pm q \mu_\pm \frac{R_1 - R_2}{S_{ch}}, \]  

(4')

The values of helium ion mobility in their own gas are calculated as the first Chapman–Enskog approximation, taking into account the charge exchange effect:

\[ \mu_\pm = \frac{3\sqrt{\pi} e}{16N\sqrt{2kTm}}, \]  

(5a)

where \( T \) and \( m \) – gas temperature and helium molecule mass, respectively.

The mean free path of helium ions in their own gas \( \lambda_\pm \) is described as (5b).

\[ \lambda_\pm = \frac{1}{N \sigma_{res}}, \]  

(5b)

where \( N \) – concentration of gas and \( \sigma_{res} \) – cross-section of the resonant charge exchange.

The \( \sigma_{res} \) value for He in the case of a weak electric field is derived by the approximation approach suggested in the work:

\[ \sigma_{res}(E) = \sigma_{res}(E_1)[1 + a \ln(E/E_1)]^2, \]  

(5c)

where \( \sigma_{res}(E) \) – cross-section of the resonant charge exchange for ion energy \( E \) and \( a \)– constant. For \( E_1 = 1\text{eV} \), those values are \( \sigma_{res}(E_1) = 27.9 \times 10^{-16} \text{cm}^2 \) and \( a = 0.0557 \).
According to the equation (5c), the $\sigma_{res}$ value is equal to $4 \times 10^{15}$ cm$^2$ at room temperature for He, and the value of mobility of He$^+$ ions in He is $\mu_{He^+-He} = 5.7$ cm$^2$/V s.

The value of mobility of negatively charged oxygen ions O$_2^-$ can be estimated with the use of the expression with only polarized cross-section:

$$\mu_i = \frac{36\sqrt{1 + \frac{m_i}{m_t}}}{\sqrt{(a/a_0^3)Ap}},$$

where $A$ – molar mass, $p$ – vapor pressure, $m/m_t$ – ratio of atom and ion masses, and $\alpha/a_0$ – molecule polarizability in the ground state $\alpha$ in relation to the Bohr radius $a_0$.

Using the expression (5d), the value of O$_2^-$ mobility in He is $\mu_{O_2^-He} = 5.76$ cm$^2$/V s.

For the determined mobility values, the current density ratio (4′) is found:

$$\frac{j_{fl}}{j_{diff}} \approx 10^{-1}.$$

Thus, the ion current density transferred by gas flow $j_{fl}$ to the volume surrounded by the screen is negligible in comparison with the diffusion current density $j_{diff}$ and may be excluded from further consideration.

We estimated the ratio of the drift $j_{dr}$ and the diffusion $j_{diff}$ currents densities:

$$\frac{j_{dr}}{j_{diff}} = \frac{\pm q \mu_{\pm} U_{ext} R_1}{\pm \mu_{\pm} (kT) \frac{R_1}{R_2} Z} \approx \frac{q R_1}{kT Z} U_{ext} \approx 10^2 U_{ext}.$$ (6)

The performed estimation shows that the drift ion current density $j_{dr}$ would be limited by ion diffusion through the mesh screen and would have saturation at low values of the probe bias voltage. However, the saturation region is not observed in the obtained current-voltage curves (Fig. 2(b)), indicating that ion concentration is high enough inside the screen and collective phenomena had to be taken into account. Thermalized plasma exists near the probe, where the positive and negative charge carriers are due to ions. Therefore, the space charge region exists in the vicinity of the probe. This region limits the probe current according to the Child–Langmuir law for charged ions in gas. We would like to emphasize on the fact that we take into account numerous collisions with molecules of neutral gas in contrast to the classical view of Child–Langmuir law (7a). Pressure dependence was defined implicitly by ion mobilities, which are the function of the mean free path. Thus the maximum current density corresponding to ions injected through the screen is expressed via (7b).

$$j = \frac{4}{9}\varepsilon_0 \left(\frac{2e}{m_t}\right)^{\frac{1}{2}} U_{ext}^{\frac{3}{2}} Z^{\frac{1}{2}},$$ (7a)

$$j = \frac{9}{8}\varepsilon_0 \mu_{\pm} U_{ext}^2 Z,$$ (7b)

where $\varepsilon_0$ – dielectric constant, $U_0$ – plasma potential, and $Z$ – thickness of the space charge region (sheath), to which the probe bias voltage is applied.

Considering that the sheath thickness is much less than the probe radius, it is assumed that the space-charge cloud is quasi-planar. Then, the ion current is described as follows:

$$i_{\pm} = \pm j s = \frac{9}{8}\varepsilon_0 \mu_{\pm} Z U_{ext}^2 s,$$ (7c)

where $s$ – area of the probe. It can be rewritten in the following manner:

$$i_{\pm}^\frac{1}{2} = \pm \frac{3\sqrt{\pi}}{2} \left(\frac{\varepsilon_0 \mu_{\pm} Z^3}{Z^3}\right)^\frac{1}{2} U_{ext}.$$ (7d)

The factor in front of the probe voltage $U_{ext}$ is the slope of the line in the current-voltage curve plotted in coordinates $i^{1/2} = f(U_{ext})$, example of which is shown in Fig. 4. It is shown from
A typical probe current-voltage curve plotted as $i^{1/2} = f(U_{ext})$, showing the straight line dependence at high values of the probe bias voltage $U_{ext}$.

Fig. 4 as well, that the curve is characterized by linear regions within the bias voltage range $|U_{ext}| = 20 – 40$ V. Thus, it can be concluded that the sheath thickness $Z$ is changed insignificantly at this voltage range and can be inferred from the line slope:

$$Z = \sqrt{\frac{9}{8} \pi \frac{e_0 \mu R_1^2}{\left( \frac{d(i^{1/2})}{dU_{ext}} \right)^2}} \approx 3.42 \text{ mm}$$  \hspace{1cm} (8a)

The obtained sheath thickness coincides reasonably well with the distance $(R_1 - R_2)$ at which the voltage $U_{ext}$ is applied. This result confirms that the drift current is limited by the space charge in the sheath and a low value of its thickness.

The sheath thickness $Z$ is of the same order of magnitude as the Debye length of ion plasma $\lambda_{Di}$; however, the electric field penetrates much deeper. As it is demonstrated in the work, $\lambda_{Di}$ can be expressed as:

$$Z = \lambda_{Di} \left( \frac{e U_{ext}}{kT_i} \right)^{1/2},$$  \hspace{1cm} (8b)

$$\lambda_{Di} = \left( \frac{e_0 kT_i}{4\pi e^2 n_x} \right)^{1/2},$$  \hspace{1cm} (8c)

where $T_i$ – ion temperature.

The values of the probe voltage, which are corresponded to linear on the regions current-voltage curve (Fig. 4), change approximately twice as much; consequently, the thickness $Z$ is varied by no more than a factor of $\sqrt{2}$.

The values of ion concentration are inferred from the slope of the current-voltage curves with the use of the expression (8b). The results from determination of the concentration values of positive and negative ions on various distances $d$ from the discharge gap and for all sets of gas mixtures are presented in Fig. 5.

The fact that the current-voltage curves of the probe are obtained at atmospheric pressure allows for assuming the equality of the temperature values of positive, negative and neutral components of the gas flow. Under those conditions, the offset in relation to zero of the maximum position of the first derivative of the value of the ion current with respect to the voltage $dI/dU_{ext}$ corresponds to the spatial potential $U_{sp}$ in the probe area to an accuracy of $kT_i/e \sim 10^{-2}$ V and is equal to approximately several volts (Fig. 6). The obtained $U_{sp}$ values on various distances $d$ behind the plasma zone are shown in Fig. 7 (set of curves 1).

In order to reveal an interrelation between the electrical characteristics of the downstream area of the plasma zone and the discharge gap properties, the plasma parameters were measured by...
FIG. 5. Distributions of positive (a) and negative (b) ion concentrations for all set of gas mixtures in the downstream area of the plasma zone.

FIG. 6. A typical current-voltage curve (curve 1) and its derivative $dI/dU_{ext}$ (curve 2) with the Gauss approximation (curve 3).

means of Langmuir probe diagnostics. The current-voltage curves of the Langmuir probe were processed according to the method proposed in the work.\textsuperscript{47} The obtained value of electron temperature $T_e$, the plasma potential $U_0$ and concentration of the charge carrier $n_0$ for various gas mixtures are summarized in Table I.

The measured values of electron temperature are close to the data presented in the works;\textsuperscript{47–49} however, the concentration values appear to be overestimated. Those high $n_0$ values are possibly due to neglecting of a probe emission current caused by the de-excitation of metastable atoms of helium.\textsuperscript{50} It is demonstrated\textsuperscript{48,49} that such current could exceed the measured ion current by more than two orders of magnitude. Recombination phenomena related to the negative ion presence are not taken into account as hardly possible effect.\textsuperscript{51} However, likely error in obtained value of charge carrier concentration gives insignificant inaccuracy (less then one order of magnitude) in further consideration.

The electric field distribution along the reaction chamber axis behind the discharge gap is affected by the location of mesh wires of the grounded electrode. In order to simplify the analysis, three extreme cases are considered as illustrated in Fig. 8:

1. The grounded electrode is placed perpendicular to the $x$ axis and intersects it at $x_0 = 0$. The electrode is separated from the quasi-neutral plasma zone by the sheath with the thickness of approximately $\lambda_D$. If we consider the electrode is a solid equipotential surface, the electric field is absent behind it. In the case of the mesh electrode, such field distribution is attributed to the region located exactly opposite to the mesh wires (Fig. 8(a)).

2. When the ion current is not intercepted by the grounded electrode, the maximum density of the positive volume charge has to exist in the plane lying behind the electrode at distance $h$. Ideally,
if the mesh does not collect the ion current at all and the diffusion and recombination processes are neglected, the spatial potential $U_{sp}$ in this plane would be equal to $U_0$ (Fig. 8(b)).

A rough estimation of the distance $h$ was made under the assumption that ions were accelerated by the electric field $E$ from plasma towards the grounded electrode due to the plasma potential drop at the distance of the Debye length. The ion current density in this case is expressed as:

$$j_+ = e n_0 \mu_+ E \approx e n_0 \mu_+ \frac{U_0}{\lambda_D},$$  \hspace{1cm} (9a)

On the other hand, according to the Child–Langmuir law for charged particles in gas,\textsuperscript{45} the maximum density of the injected ion current from the discharge gap through the mesh equals to:

$$j = \frac{9}{8} \varepsilon_0 \mu_+ \frac{U_0^2}{h^3},$$  \hspace{1cm} (9b)

Combining (9a) and (9b) one can evaluate $h$:

$$h = \left( \frac{9}{8} \varepsilon_0 \lambda_D U_0 \right)^{\frac{1}{3}}.$$  \hspace{1cm} (9c)

The obtained values of $h$ for used gas mixtures are shown in Table I.

<table>
<thead>
<tr>
<th></th>
<th>He</th>
<th>He–O\textsubscript{2}</th>
<th>He–Ar</th>
<th>He–TEOS</th>
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<td>$T_e$, eV</td>
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<td>6.09</td>
<td>2.66</td>
<td>2.49</td>
</tr>
<tr>
<td>$n_0$, cm\textsuperscript{-3}</td>
<td>1.4 10\textsuperscript{13}</td>
<td>6.4 10\textsuperscript{13}</td>
<td>1.31 10\textsuperscript{13}</td>
<td>3.84 10\textsuperscript{13}</td>
</tr>
<tr>
<td>$U_0$, V</td>
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<td>27.16</td>
<td>7.45</td>
<td>10.03</td>
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<td>$h$, mm</td>
<td>0.14</td>
<td>0.24</td>
<td>0.09</td>
<td>0.06</td>
</tr>
</tbody>
</table>
On basis of the performed estimation, it is possible to conclude that the spatial potential $U_{sp}$ is around several volts and tends towards the plasma potential $U_0$ at the distance of one-tenth of a millimeter behind the grounded electrode. Taking into account that charge carrier concentration in the plasma zone are likely overestimated, this distance would be several times higher.

In order to reveal the potential distribution along the reaction chamber axis, we propose that the positive space charge $Q_{sp}$ is localized in an infinitely thin cylinder of the radius $R_{el}$. Consequently, the potential distribution is expressed as follows:

$$U_{sp}(x) = \frac{Q_{sp}}{2\pi\varepsilon_0 R_{el}^2} \int_0^{R_{el}} \frac{rdr}{(x^2 + r^2)^{3/2}} = \frac{U_0}{R_{el}} \left[ \left( x^2 + R_{el}^2 \right)^{1/2} - x \right].$$ \hspace{1cm} (10)

3. The grounded electrode totally intercepts all charge carriers. However, it does not cut off the field from the positive plasma charge because the Debye length exceeded the distance between the mesh wires. In the considered case, it happens near the mesh wires (Fig. 8(c)). The axial potential distribution originating from the plasma layer with the finite thickness $L$ and the radius $R_{el}$ is described by the equation:

$$U_{sp}(x) = \frac{Q_{sp}}{2\pi\varepsilon_0 R_{el}^2 L} \int_0^{R_{el}} \int_0^{-L} \frac{dy}{\sqrt{(x + y)^2 + r^2}}.$$ \hspace{1cm} (11)

In spite of the fact that the integral calculation of the expression (11) would give more specified solution in comparison to the expression (10), the estimated potential distributions are similar in the range of the used $L$ and $R_{el}$ values (Fig. 9). The results related to a finite layer thickness are presented for two cases: the thick space charge layer $L = R_{el}$ (curve 2) and the layer with the experimental $L$ and $R_{real}$ values (curve 3). The maximum difference of the inferred spatial potential distributions is 5% at the distance up to 10 cm. Therefore the potential pattern along the reaction chamber axis in the downstream area can be accurately described by the expression (10).

The real pattern of the potential distribution in the downstream area is based on the superposition of three considered cases, complicated via diffusion processes, electrostatic repulsion, and edge effects. There are multiple field perturbations in the plane parallel to the grounded electrode, where...
FIG. 9. The spatial potential distribution from an infinitely thin charged disk (curve 1), from the cylindrical space charge layer of the thickness \( L = R_{el} \) (curve 2) and from the cylindrical space charge layer with the experimental \( L \) and \( R_{real} \) values (curve 3).

the potential lies between the plasma potential and zero. Consequently, multiple potential wells of various depth exist behind the mesh electrode at the distance \( d = [0; h] \) in the plane perpendicular to the chamber axis.

With the use of the expression (10) and the obtained values of \( U_0 \) and \( h \), the potential distribution along the reaction chamber (Fig. 7, set of curves 2) were calculated for various gas mixtures. Satisfactory matching of the experimental and the calculated curves is found for used gas mixtures, except the He–O\(_2\) mixture. The discrepancy in this case is most likely caused by an increase in the contribution of the positive and negative charges recombination under relatively high concentrations of oxygen (1%) in the gas phase. Neglecting the recombination processes would lead to overestimation of both the plasma potential and the space charge potential in the downstream area.

IV. PHYSICAL MODEL

On the basis of the presented data we have proposed the model describing the observed phenomena, which is schematically illustrated in Fig. 10. Positive ions (preferably of He\(^+\)) situated in the near-electrode area (II) are injected to the area (III) placed behind the discharge gap due to acceleration by the voltage difference between the area of the undisturbed plasma (I) and the grounded electrode. Negative ions are formed in the process of electron attachment to molecules of impurity oxygen. This process occurs near the grounded electrode where electrons appear due to the de-excitation of metastable He atoms.

The maximum density of the positive space charge builds up in the plane separated from the mesh electrode by the distance \( h \). The maximum value of the space charge potential tends toward

FIG. 10. Schematic representation of the reaction chamber (a) and the potential distribution along it (b).
the value of the plasma potential $U_0$. A potential $U'$, which are in the plane of the grounded electrode, is determined by positive ions going through the mesh cells and its value is lower than $U_0$. Thus, a potential well of depth $U_0 - U'$ exists near the grounded electrode. Negative ions are generated in this potential well and then enter the positive space charge area. With increasing the distance $x$ from the mesh electrode in the area (IV) the potential decreases. Such a potential decay originates from the drift of the positive and negative ions. The potential of the electrical field decreases in the area (IV) with an increase in the distance $x$ from its maximum value in plane $h$. Negative ions are generated in the area of this potential well and can be received in the area of positive spatial charge.

The area of decay of the positive potential (IV) corresponds to drift of the positive and negative ions.

Ion concentration in the downstream area changes approximately by an order of magnitude at a distance $X$, which is of one centimeter order (Fig. 5). Assuming that the Debye length equals to $10^{-4} \text{m}$ in our conditions (ion concentration $\approx 10^7 \text{cm}^{-3}$) and thus having $(\lambda_D/X) \ll 1$, we conclude that ambipolar drift plays an important role in the ion flux transport, i.e. positive ions leaving the space charge area due to the electric field draw the negative ion current. Therefore the injection of negative ions into area (IV) can be caused by ambipolar drift. Thus, found concentration of negative ions outside the discharge gap is comparable with concentration of positive ions and weakly depends on negative ion concentration in the plasma.

The maximum of ion concentration in the downstream area is of several orders of magnitude lower than in the discharge gap and is around $10^6 - 10^7 \text{cm}^{-3}$. In other words, a virtual emitter is formed at the distance $h$ from the grounded mesh electrode, behind which the ion flux drifts through the positive space charge. Existence of the virtual emitter is the main limiting factor for the ion entrance from the discharge gap to the drift area and causes the low values of their concentration in this area.

Further we evaluate the ion flux decay in the drift area due to the virtual emitter presence. We assume that $\text{He}^+$ ions born in the discharge area are accelerated by the plasma sheath voltage towards the grounded mesh electrode, leave the discharge area through it and enter the drift area, which is formed by the electrode and the probe screen. Here ions form the space charge, which imposes the potential bump.

The sheath voltage $U_0$ is about 10 V, and the distance between the electrode and the probe is $d = 10 \text{mm}$. The potential at the electrode and the probe locations is zero, because they are grounded (refer to Fig. 10).

In order to evaluate the current density of charge carriers in the drift area, the Child-Langmuir equation was modified for the ion current density in gas ambient: \[ J = \frac{9}{8} \varepsilon_0 \mu_+ U^2 \frac{U^2}{X^3} = \chi \frac{U^2}{X^3}, \] (12a)

where $J$ – current density, $\varepsilon_0$ – dielectric constant, $\mu_+$ – ion mobility, $U$ – accelerating voltage and $\chi = 9/8\varepsilon_0 \mu_+$ – constant.

Having the ion current density $J_0$ though the electrode, we can evaluate the location $h$, where the ion density reaches its maximum value:

\[ h = \left( \frac{U_0^2}{\chi J_0} \right)^{\frac{1}{3}}. \] (12b)

At this point the virtual emitter is formed and induces the electric field, which decelerates ions injected from the discharge gap and drives them back. The virtual emitter also creates the electric field, which accelerates ion towards the probe.

In our conditions ($U_0 \approx 10 \text{V}$, $J_0 = 0.2 \text{A/cm}^2$, $\mu_+ = 5.7 \text{cm}^2/\text{V s}$) $h$ is equal to 0.14 mm, which is nearly two orders of magnitude lower than $d = 10 \text{mm}$. Hence, the virtual emitter parallel and very close to the grounded electrode is obtained, from where almost all the injected ion current returns back into the plasma zone. The potential of the virtual emitter is determined by the plasma sheath voltage and equals to $U_{\text{emitter}} \approx U_0 \approx 10 \text{V}$.
Than we evaluate the ion current density, which flows between the virtual emitter and the probe, which is also determined by Child-Langmuir law:

\[ J_1 = \chi \frac{U_0^2}{(d-x_0)^3}. \]  

(12c)

The ratio of the probe current \( J_1 \) to the current injected from plasma region \( J_0 \) is:

\[ \frac{J_1}{J_0} = \frac{\chi \frac{U_0^2}{(d-h)^3}}{\chi \frac{U_0^2}{h^3}} \approx \left( \frac{h}{d} \right)^3, \]  

(12d)

which is in our case:

\[ \frac{J_1}{J_0} = \left( \frac{0.14}{10} \right)^3 = 2.74 \cdot 10^{-6}. \]  

(12e)

The expression (12e) reveals that the current density drops significantly (six orders of magnitude) behind the virtual emitter. It is highly likely the ion density drops too (also approximately in six orders of magnitude).

Considering that all injected ions have similar energy, velocity of ions is \( v \approx 0 \) in the virtual emitter plane. Hence the ion density tends to infinity: \( \rho = \frac{J}{v} \to \infty. \)  

(12f)

In reality the injected ion flows in non-monokinetic and there is some discrepancy in ion energy values, allowing for evaluation of the virtual emitter thickness (the length scale of ion density variation). Thus, the emitter thickness depends on \( kT_i \) for Maxwellian ion energy distribution:

\[ \Delta x = \left( \frac{U_0 + \frac{kT_i}{e}}{J_0} \right)^\frac{1}{2} - \left( \frac{U_0 - \frac{kT_i}{e}}{J_0} \right)^\frac{1}{2}. \]  

(12g)

In our case, ion gas temperature is much less compared to the accelerating voltage (12h):

\[ \frac{kT_i}{e} << U_0. \]  

(12h)

And the emitter thickness is much less than its location coordinate value \( \Delta x << h = 0.14 \text{ mm}. \) Therefore, within our experimental conditions, the probe location (±0.5 mm at \( d = 10 \text{ mm} \)) was far from the sharp concentration gradient. So, location precision was sufficient to get data with reasonable accuracy. The error of experimental values (less then 25%) proves the validity of performed estimations.

V. SUMMARY

As result of this study, the external probe of original design was developed, allowing for determination of the ion flux characteristics in the downstream area of atmospheric pressure remote capacitively coupled plasma in various gas mixtures of He. Analysis of the current-voltage curves obtained with the use of the probe showed the presence of positive and negative ions behind the discharge gap. The spatial distributions of ion concentration and the electrical potential along the reaction chamber axis outside the discharge gap were obtained.

The possibility of the virtual emitter formation at the distance \( 10^{-2} \text{–} 10^{-1} \text{ mm from the grounded electrode was demonstrated. The dramatic decline of ion concentration (six orders of magnitude) occurred near the virtual emitter plane. The slight reduction of ion concentration was observed at longer distance of several centimeters from the discharge gap. Maximum concentration was approximately } 10^6 \text{–} 10^7 \text{ cm}^{-3} \text{ in this area. It was demonstrated that the gas flow containing ions going through the volume was characterized by decreasing of the positive electrical potential, with a maximum near } +(10 \text{–} 20) \text{ V.} \)