Experimental and numerical study of the plasma in coaxial capacitive coupled radio frequency discharge

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Sara Alhomsy-0000-0002-1735-2235

Gérard Bauville

Stéphane Pasquiers

Tiberiu Minea-0000-0003-2886-3492

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Sara Alhoms, , Gérard Bauville, Stéphane Pasquiers, and Tiberiu Minea^{a)}

AFFILIATIONS

Laboratoire de Physique des Gaz et des Plasmas, Université Paris-Saclay, CNRS, F-91405 Orsay Cedex, France

^{a)}Author to whom correspondence should be addressed: tiberiu.minea@universite-paris-saclay.fr

ABSTRACT

A coaxial configuration of discharge is proposed for plasma surface treatment and possibly other applications. The reactor is based on a cylindrical structure, with the inner cylinder radio frequency powered (RF, 13.56 MHz) and the outer cylinder grounded, playing the role of a guard ring. The charged species can escape from the inner cavity through two longitudinal slits made in both cylinders, aligned to each other, and producing a linear slab of plasma. Hence, it is possible to project the plasma directly onto a surface placed under the slits, called external plate. The operation of this device is uniform and stable in argon for a large pressure range (0.8–50 mbar). Furthermore, simulations using the PlasimoTM software package were performed to evaluate the plasma parameters and to explain the experimental results. The ion flux on the surface exposed to this plasma increases when RF power increases, and the pressure or gap distance to the plate decreases. This cylindrical capacitive coupled plasma configuration can be very effective for surface treatment of different materials (conductors or insulators) on large area (when the plate or the system is moving perpendicular to the slits) due to energetic ions and active species released from the plasma.

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I. INTRODUCTION

Radio frequency (RF) glow discharges have been intensively studied starting with the early 1960s essentially driven by the microelectronics research. These plasmas involve a variety of physical and chemical processes, and they are particularly interesting for the large area applications.¹ The biggest impact of RF is still in microelectronics where material processing is essential for very large-scale integrated (VLSI) circuits and high aspect ratio etching to make vias, but also it is extensively used for photovoltaic thin film deposition, for packaging treatment, and other plasma-based technologies.²

The capacitive coupled plasma (CCP) is the most popular RF discharge because the equipment is very simple, typically the one designed for direct current (DC) plasma but runs with radio frequency power.

Generally, the potential structure in the plasma tends to confine electrons and to expel positive ions through the oscillating sheaths,³ since at this frequency, the ions are sensitive only to the average potential due to their inertia.

The classic treatment of sheaths in RF discharges has been well described and understood for more than 50 years.⁴ Koenig and Maissel⁵ were the first to use the capacitive model of the sheaths to explain the electrical behavior of the RF glow discharge systems for RF sputtering. Briefly, the magnitude of the sheath capacitances at the powered electrode (C_p) and the (grounded) wall (C_g) is determined primarily by the relative area of the excitation electrode (A_p) and grounded surfaces (A_g) in contact with the glow discharge.⁷ Depending on the operation pressure, several regimes can be found for the transport of charged species through the sheath. Hence, it was established that the ratio of the DC voltage dropping on each one of the sheaths (facing the two electrodes) is inversely proportional to the electrode area ratio to the power n , where n lies between 1.2 and 4.⁶

Laboratory and industrial plasma sources are often geometrically asymmetric, i.e., the surface areas of the powered and grounded electrodes (A_p and A_g) are not equal, leading to the generation of a DC self-bias voltage to ensure equal fluxes of positive and negative charges in average, over the RF period. For capacitive

coupling, there is no net conduction current flowing through the circuit, neither on the excitation electrode circuit nor on the grounded one. Therefore, the plasma potential must approach the potential of the excitation electrode for a brief period during each RF cycle to allow electrons reaching the excitation electrode. Similarly, the plasma potential must also approach ground potential for a brief period during each cycle. Therefore, the expression of the self-bias (V_{bias}) for capacitively coupled systems⁷ is given by the following equation:

$$V_{bias} = V_{RF} \left(\frac{C_p - C_g}{C_p + C_g} \right), \quad (1)$$

where V_{RF} is the amplitude of applied RF voltage.

Due to extensive use in the semiconductor industry, planar asymmetric plasma reactors are relatively well understood. Early works on the geometrically asymmetric RF capacitive discharges were focused on planar excited electrode and a large area grounded (often the discharge chamber itself), which generally had a cylindrical shape.^{7-9,12-16}

On the contrary, linear RF plasma for surface treatment is much rarer. It has been recently proposed, for instance, by Baránková *et al.*¹⁷ This configuration was successfully exploited for plasma enhanced chemical vapor deposition (PECVD) combined with physical vapor deposition (PVD) and efficiently operated with a hollow powered electrode (target) formed of two parallel (graphite) plates. However, they did not follow or reported on the self-bias voltage on the RF electrode and the role of ions in this type of device. They choose to use a second RF power supply to bias the dielectric (glass) substrate in order to control the ion flux.

We are presently interested in the cylindrical geometry of the electrodes operated with a single RF power supply. Let us note that only a few experiments and models do exist for this case.^{18,19} Recently, Upadhyay *et al.*¹⁹ used an asymmetric non-planar geometry to process the inner walls of superconductive radio frequency (SRF) cavities. They powered the inner cylinder of the cylindrical capacitive coupled discharge (CCCD), while the outer cylinder was grounded, which was, in their case, the cavity wall to be etched. The plasma, in their case, develops between the two cylinders. The surface area asymmetry between inner and outer electrodes (cylinders) creates a negative self-bias potential on the inner electrode, obviously smaller.

In the present work, the novel design of CCCD is proposed, and it succeeded to produce a linear plasma that survives inside the inner (powered) electrode but can also escape through a slit to treat, for instance, an external plate placed under the reactor (facing the slit). In this work, the gas used is argon. The plasma formed in the cylindrical coaxial cavity is stable over a large range of pressures (0.8–50 mbar) and RF powers (20–1200 W). We tested the operation of the initial configuration, called “reactor A,” and aiming its scaling-up, a second configuration has been tested afterward with longer cylinders, called “reactor B.”

In this paper, we present the proposed configuration of CCCD, powered by a radio frequency power supply operating at 13.56 MHz, and the optimization of the system in terms of the production of charged particles close to the plate. We report the main

plasma parameters obtained by combining the experimental and numerical approaches.

Section II describes the experimental setup and the numerical approach. The experimental results for both reactors A and B are presented in Sec. III. In Sec. IV, the numerical findings are presented, and the experimental results are discussed in light of these simulations. Finally, Sec. V summarizes the main achievements and the conditions for optimizing the operation of the proposed CCCD configuration.

II. EXPERIMENTAL SETUP AND NUMERICAL APPROACH

A. Experimental setup and diagnostics

A new reactor has recently been developed to generate a linear slab of plasma with the control of the ion species (energy and density) using a single RF power supply (see Figs. 1 and 2). The CCCD reactor is formed by two coaxial tubes of different radii, each one equipped with a longitudinal slit. The edges were closed by two stainless-steel supports. They ensure perfect parallelism between the two cylinders; the spacer they contain is made of dielectric material (alumina) and the RF is fed by one side. Only the inner cylinder is powered, the outer being grounded.

The first characterization was performed with an initial configuration, the so-called reactor “A.” This configuration has the RF electrode made of stainless-steel (90 mm long, $r_{in} = 5$ mm inner radius and $r_{out} = 6.4$ mm outer radius) with the slit area of 50×2 mm² (length $\times$ width = $L_{in} \times w_{in}$). The outer cylinder was made of copper (120 mm long, $R_{in} = 8$ mm inner radius and $R_{out} = 10$ mm outer radius), with a slit area of 60×4 mm² (length $\times$ width = $L_{out} \times w_{out}$).

The essential choice for our work was the geometry of metal electrodes, but the materials itself does not have a significant effect on the various experimental studies carried out.

The distance between these tubes is $\delta = 1.6$ mm, and the two slits are aligned facing each other (see Fig. 2).

The role of the external cylinder is to avoid the plasma formation on the outer surface of the powered electrode, forcing the operation of the system in a hollow electrode mode. The distance δ between the two cylinders is one of the critical parameters and must be made as small as possible. Hence, the outer cylinder operates as a guard ring, and no plasma (or a very weak one) develops between the two cylinders.

When RF power is applied, the plasma glows inside the inner tube cavity, but it leaks out through the slits and spreads onto the surface of the external plate, here copper, placed just under the slits. The gap separating the outer cylinder and the plate is denoted as g , with $g = 8$ mm (see Figs. 1 and 2). This plate was also grounded, but it is possible to bias it as desired, positively or negatively, depending on the followed purpose.

This system is very compact, and the plasma stays confined in a limited volume between the CCCD reactor and the plate. The system was placed in a vacuum chamber of 45 cm height and 35 cm diameter, made of stainless-steel. Prior to each experiment, the primary vacuum was obtained via a Pfeiffer duo 004 (pumping speed 4 m³/h) pump, reaching the base limit pressure of 10^{-2} mbar. Then, the argon gas was injected, over a pressure range lying between 0.8 and 50 mbar. The pressure was given by an absolute

Q8

FIG. 1. Sketch of the experimental setup. $p = 1.6$ mm is the distance between the cylinders, $w_{in} = 2$ mm and $w_{out} = 4$ mm are the widths of the slits for inner and outer cylinder, respectively.

capacitive gauge (Pfeiffer CMR 361). The glow discharge was established by applying the RF delivered by the power supply (COMET, 13.56 MHz, 3 kW) to the inner electrode. It was connected to an automatic impedance matching system to ensure the impedance tuning. So, the potential (boundary conditions) on each electrode is as follows.

On the inner electrode:

$$V_{RF}(t) = V_{bias} + V_{MAX} \sin(\omega t). \quad (2)$$

On the outer electrode and the plate:

$$V_g = 0. \quad (3)$$

FIG. 2. Illustration of (a) the experimental arrangement of CCCD and (b) the view showing the slits of the tubes; $w_{in} = 2$ mm and $w_{out} = 4$ mm are the widths of the slits for inner and outer cylinder, respectively.

The circuit includes an RF probe (Vigilant Power Monitor—SOLAYL), which measures the electrical parameters (average power, voltage amplitude, maximum current, phase shift, impedance, harmonics, etc.). It is driven by a software (Vigilant RF Lab) that also allows us to display the measured data. However, this probe is unable to measure the DC voltage (self-bias) developed on the powered electrode because the voltage sensor is “AC coupled” to the RF electrode. For this reason, we measured independently the self-bias voltage by a home-made system described below.

Note that the absolute values of self-bias are displayed on the power supply but not its sign. The measurements were performed using a blocking capacitor, which excludes the effective DC ion current in the circuit. In fact, this capacitor is one of the capacitances of the impedance matching system. To precisely know the sign of the DC self-bias, we also measured this voltage using a voltmeter. The voltage drop onto one of the resistances of a voltage divider circuit is measured placing the divider between the RF fed line and the ground, as close as possible of the powered electrode. Notice that the electrical connection was still out of the vacuum chamber, at atmospheric pressure (see Fig. 1). The divider consists of two huge resistors (R_1 and R_2) in series with $R_1 = R_2 = 10 \text{ M}\Omega$, such as the current leakage through this branch of the circuit could be neglected with respect to the discharge one. The voltage measurement is performed over the second resistor (R_2), and the self-bias voltage is obtained, obviously, multiplying this voltage by 2 (Fig. 1). It should also be noted that the difference between the voltage returned by the generator and the absolute value of the voltage measured by the voltmeter (multiplied by two) is about 14%, this difference being due to the position on the RF circuit between the two measurement points.

A similar configuration was tested but with longer cylinders/slits, reactor B: inner cylinder was made also of stainless-steel 180 mm long, with a slit area of $145 \times 2 \text{ mm}^2$. The corresponding outer cylinder was made of cooper, 210 mm long, and with a slit area of $150 \times 4 \text{ mm}^2$. The plasma develops as expected in a similar way, linearly, all along the slits leading to a plasma slab outside, toward an external plate facing the slits. Beyond the influence of the RF power or the gas pressure on the CCCD, it is very interesting to study the effect of the gap distance (g) to the plate. Hence, the gap for configuration B of this reactor was varied between 1 and 10 mm ($g = 1, 6$, and 10 mm).

B. Numerical approach

For the numerical study of the geometrically asymmetric discharge system, the MicroDischarge model of Plasimo package software has been used.³² It is based on the fluid approach to model the plasma. A detailed description of the model can be found in Ref. 20 and references therein. Hereafter, only the basic features of this model, in particular, the species and reactions, are presented as well as the numerical approximation for this specific discharge geometry.

C. Model equations

Following the conventional fluid approach, the MicroDischarge model of Plasimo package solves the conservation equations system, derived from the moments of the Boltzmann

transport equation, and coupled with the Poisson equation for the charged species. These balance equations were solved for a number of species using the drift-diffusion approximation. The species included in the model, such as electrons, ions, and excited atoms, are described in more detail in Sec. II B. The conservation equations for the different species are

$$\frac{\partial n_p}{\partial t} + \nabla \cdot \Gamma_p = S_p, \quad (4)$$

with Γ_p being the drift-diffusion flux,

$$\Gamma_p = \pm \mu_p E n_p - D_p \nabla n_p, \quad (5)$$

where n_p is the number volume density of species p , S_p is the net source term of species p due to reactions induced by the plasma kinetics, μ_p and D_p are the mobility and the diffusion coefficient of species p , respectively, and E is the local electric field.

In order to specify the local variation of the reaction rate coefficients and electron transport coefficients as a function of the mean electron energy, an additional balance equation for electron energy was included:

$$\frac{\partial (n_e \epsilon_e)}{\partial t} + \nabla \cdot \Gamma_{\epsilon_e} = S_{\epsilon_e}, \quad (6)$$

in which ϵ_e is the average electron energy and n_e is the electron number volume density. The source term of this equation, S_{ϵ_e} , represents the net source term of energy for the plasma electrons, i.e., the energy gained by the electric field and lost by the collisions. The electron energy flux Γ_{ϵ_e} is described in Ref. 20,

$$\Gamma_{\epsilon_e} = -\frac{5}{3} \mu_e E n_e \epsilon_e - \frac{5}{3} n_e D_e \nabla \epsilon_e. \quad (7)$$

The electron transport and reaction rate coefficients were pre-calculated by a Boltzmann solver—Bolsig+,²¹ for an equilibrium electron energy distribution function at different reduced electric fields. The resulting data set was used to create a lookup table to be used as input for the Plasimo model. The transport coefficients for ions were extracted from the LXCat Database²² and were used as input for the model as a function of the reduced electric field. For the charged particles, the diffusion coefficients were calculated from the mobilities using Einstein's relation.³¹

The constant diffusion coefficient for the excited species was taken from Ref. 23.

Finally, in addition to the balance equations discussed above, the Poisson equation was solved,

$$\nabla \cdot (\epsilon_{pl} \nabla \varphi) = -\nabla \cdot (\epsilon_p E) = -\sum_p q_p n_p, \quad (8)$$

in which φ is the electric potential as a function of position and time, ϵ_{pl} is the permittivity of the medium, and q_p is the charge of the species p . From this, the electric field and potential distribution in the discharge were determined in a cross section (2D) perpendicular to the cylinder's axis. All equations of the model are solved

TABLE I. Species and reactions included in the model, with references to the original literature from which the transport coefficients for particles or rate coefficients were considered.

Species	Transport coefficient (Ref.)	Reactions	Rate coefficient (Ref.)
E	Bolsig+ (Ref. 21)	$\text{Ar} + e \Leftrightarrow \text{Ar}^+ + 2e$	Bolsig+ (Ref. 21)
Ar+	LXCat (Ref. 22)	$\text{Ar} + e \Leftrightarrow \text{Ar}^* + e$	Bolsig+ (Ref. 21)
Ar*	(Ref. 23)	Elastic energy loss	Bolsig+ (Ref. 21)

by the finite difference method, where all quantities are represented by their values at a discrete set of points in time and space.²⁰

D. Species and reactions

In this work, we run the code for argon gas, by using the corresponding reaction rates and transport coefficients of particles, as described in Subsection II C. Three species have been considered in the model: electrons (e), single charged positive ions (Ar^+), and one effective excited state (Ar^*).

At the walls, the incoming heavy particles were either neutralized if they are charged or de-excited if they are excited (i.e., the wall is fully absorber in terms of charge or energy). The resulting ground state atoms are assumed to feed the background gas, which is thus constant (unaffected by the plasma species). However, the energetic ions impinging the surfaces can yield to secondary electron emission at the wall (especially RF powered electrode, as detailed in Secs. III and IV). The secondary emission coefficients for Ar^+ depend strongly on the properties of the surface and are not exactly known. We used the value of 0.04 for Ar^+ induced secondary electron emission. More details on the boundary conditions of the model can be found in Ref. 20.

The transport coefficients for particles and the set of reactions between the species that were included in the model are summarized in Table I.

Note that we focused here on the spatial distribution of the density of charged particles, the electric field and potential, but not particularly on the detailed description of the excited states and gaseous kinetics. The latter are interesting particularly in the case of PECVD use of the CCFD, which is out of the scope of the present work. Hence, for the sake of simplicity, the excited levels are all reduced to one effective state to account on the energy loss of electrons in their inelastic path, considering the sum of the cross sections of all excited states.

A second approximation used in these calculations is to neglect the ionization from excited state. After several test simulations, it comes out that the average electrons energy is always greater than 4 eV, which confirms our second approximation.

E. Discharge geometry

The discharge structure is assumed to be infinite long, such as it could be reduced to 2D simulation geometry, with the cross section corresponding to the plane perpendicular to the cylinder's axis. The geometry was kept as close as possible to the real discharge system, with inner tube 5 and 6.4 mm inner and outer radii, respectively, with 2 mm width slit (w_{in}) (blue tube in Fig. 3) powered by the RF signal (10 MHz in simulation). The outer tube

has 8 mm inner radius and 4 mm width slit (w_{out}) (cyan in Fig. 3), and it is connected to the ground together with the plate under the slits. With these dimensions, the space between the two cylinders $\delta = 1.6$ mm, like in the experiment.

The axes y and z are defined as follows (see Fig. 3). The origin (0,0) is the point on the plate crossed by the normal to it passing by the center of the slits (and the axis of the two cylinders). y axis is parallel to the plate surface and perpendicular to the cylinder's axis. Positive and negative values describe the position on each side of the slit (center). z axis represents the vertical axis from the center (0,0) toward the coaxial electrodes structure, on which the distance between the plate and the outer tube represents the gap g .

The sketch of the simulation geometry is shown in Fig. 3. The initial condition for the seed electrons and ions considers a homogeneous density of 10^{15} m^{-3} . The metastable state (effective excited state) density has been set to an initial density of 10^7 m^{-3} . These input values have been changed several times to check their impact on the final solution, and they were the highest without any impact on the final result. So, they have been further systematically used because the highest the input the faster the convergence of the code. The numerical convergence had been considered achieved after more than $60 \mu\text{s}$ of real time simulation (600 RF periods), and the average plasma parameters are certainly not evolving (moment

FIG. 3. Sketch of simulation geometry. Plasma volume is the black region. Inner electrode is the blue region and outer the cyan region (only the inner radius of the outer electrode is important). Cell size (in red) was $0.2 \times 0.2 \text{ mm}^2$, $\delta = 1.6 \text{ mm}$, $w_{in} = 2 \text{ mm}$, $w_{out} = 4 \text{ mm}$, and $g = 8 \text{ mm}$.

FIG. 4. Ar plasma images taken with the reactor A in operation for (a) 15 and (b) 30 mbar. Plasma power was kept constant at 200 W ($g = 8$ mm).

A. First characterization (reactor A)

This section presents the first experimental investigations of the novel configuration. The typical operation has been found for a pressure range between 15 and 50 mbar. For this set of results reported here, the gap between the reactor and the plate was fixed ($g = 8$ mm). Systematic measurements have been performed varying the RF power (P_{RF}) and the gas pressure (p) for argon plasma. A linear plasma was observed all along the slit.

As said, when applying the RF power to the system, a discharge glows and escapes out of the slits. Striations have been observed along the slit. They are mobile in both directions between the tube edges. The velocity and the number of striations strongly depend on gas pressure (see Fig. 4) and RF power. Further studies of these structures are very desirable, as recently reported by Bera *et al.*⁴² The striation discharge pattern is characterized by several plasma “bubbles,” alternating bright and dark regions, formed outside the slit, in the gap between the electrodes and the plate. The formation of such striations was already reported in direct current (DC) glow discharges.^{24,26–28} For RF capacitive coupled discharges, striations have also been observed, in electronegative gases²⁵ and in argon, when plasma is created between two wires wrapped around a long dielectric tube, over a certain range of frequencies and gas pressures.^{29,30} The non-linear dependence of the ionization rate on the electron density is shown to be the main underlying mechanism of the striation phenomena.³⁴

1. Effect of gas pressure and RF power

In this section, the effect of the gas pressure and RF power on discharge parameters are presented, such as the voltage and current amplitude (V_{MAX} , I_{MAX}), the phase between voltage and current (ϕ), the discharge impedance (Z), and the self-bias voltage (V_{bias}). They were measured for three pressures: 15, 30, and 50 mbar. As expected, the gas pressure influences the electrical characteristics of the RF discharge, as found before.

Figure 5(a) shows the temporal evolution of V_{MAX} and I_{MAX} for each operation pressure. The discharge is stable for different pressures and one can see that V_{MAX} and I_{MAX} are the highest for the lowest pressure, here 15 mbar.

FIG. 5. (a) Temporal evolution of the voltage and current amplitude (V_{MAX} , I_{MAX}) for 15, 30, and 50 mbar. (b) Variation of the phase (ϕ) and the impedance (Z) with gas pressure. Plasma power was kept constant at 200 W.

FIG. 6. (a). Temporal evolution of the voltage and current amplitude (V_{MAX} , I_{MAX}) for 200, 400, 800, and 1200 W. (b) Variation of the phase (ϕ), impedance (Z), reactance (X), and resistance (R) of plasma with RF power. Gas pressure was kept constant at 15 mbar.

For 15 mbar, the slow drift of V_{MAX} and I_{MAX} from 0 to 15 s is related to the heating of the electrodes, which is less important at higher pressure, 30 and 50 mbar.

Figure 5(b) shows the variation of the phase and the impedance measured with the gas pressure. Obviously, the impedance and the phase (in absolute value) decrease with the gas pressure.^{35,38} The same effect of gas pressure on electrical characteristics is widely found for RF discharges. The discharge current decreases with increasing of pressure due to the increasing of electron-neutral collision frequency. The discharge voltage varies almost proportionally to the discharge current, and, therefore, the impedance remains in the narrow range contained in the interval (27.5–33 Ω). The higher collision rates for higher pressures cause the increase of the ohmic (or resistive) component of the discharge, which yields to the decrease of the phase.^{35,38}

The plasma impedance is the complex sum of resistive (R , real) and capacitive reactive (X , imaginary) parts. Figure 6 shows the time evolution of different electrical characteristics (V_{MAX} , I_{MAX} , ϕ , Z , X , and R) but this time for several RF powers for a fixed operation pressure (15 mbar). It should be noted that X is negative, but the absolute values are presented.

Figure 6(a) shows the temporal evolution of V_{MAX} and I_{MAX} for different RF powers 200, 400, 800, and 1200 W. The discharge is stable for different powers and one can see that V_{MAX} and I_{MAX} increase with RF power, as expected.

But, as shown in Fig. 6(b), the increase of the power is not only a result of the increasing voltages and currents but also due to the decreasing phase angles (ϕ), which means that the discharge becomes more resistive, and more power is dissipated by ohmic heating. This increase of the real part of the impedance turns the phase angle to smaller values. The physics of the increasing resistivity can be understood considering two different power dissipation channels in the discharge, following the model presented in Refs. 39 and 40: plasma resistivity is the sum of the ohmic resistance in the bulk (electron-neutral collisions) and of the resistance that models the power dissipation in the sheath (by acceleration of ions by the electric field in sheaths). The first decreases but the second increases more quickly with the RF power.

Obviously, the impedance decreases as the power increases [Fig. 6(b)]. This decrease is due essentially to that of the absolute

value of the capacitive reactance X , i.e., a compression of the sheaths thickness.

On the other hand, Fig. 7 shows the variation of the self-bias (in absolute value) with the pressure at 100 and 200 W. For all pressures, the measured self-bias was negative. One can see that for both RF powers, the self-bias is the highest for the lowest pressure, here 15 mbar, in line with the finding for V_{MAX} [Fig. 5(a)]. As expected, its value increases with the power, V_{bias} being higher at 200 than at 100 W. Similar variation trends of the absolute value of V_{bias} with RF power and gas pressure were reported in many references, see for instance Refs. 10, 11, and 19.

As already known, the increase of self-bias with RF power is essentially due to the increase of the RF voltage [see Eq. (1)], at constant power.

Hytry and Boutard-Gabillet¹⁰ and Upadhyay *et al.*¹⁹ have explained the increase of the negative self-bias when decreasing the pressure by the expansion of the plasma volume toward the

FIG. 7. Self-bias voltage (absolute value) variation with pressure, at 100 and 200 W RF power ($g = 8$ mm).

grounded area. They verified experimentally that the sheath thickness increases also with the pressure decrease.¹⁰ For our design of CCCP, this plasma feature is discussed in detail in Sec. IV A.

B. Operation with long cylinders (reactor B)

According to the results obtained with the initial configuration of the reactor (Subsection III A—reactor A), a similar one has been built and tested, but with about twice longer slit (reactor B, Fig. 8). Hence, it is expected to enlarge the active region of the plasma. This allows the treatment of larger surfaces, keeping in view its possible application.

However, as the RF power is injected at one end of the inner cylinder, it is necessary to first check if the discharge glows all along the slit and not only close to the feeding side. As expected, the plasma glows in a similar way as for the shorter electrodes system (reactor A). Therefore, the scaling-up of the system seems well possible, and larger systems can be imagined, depending on the requirements.

Let us mention that the striation phenomenon was found again. However, one can easily remark the almost homogeneous light emission all along the slit at the highest pressure tested here, 9 mbar, especially in front of the plate where no variation of the emission light intensity could be recorded.

Concerning the parametric study, the same trends have been observed, as for the configuration A, for pressures between 15 and

50 mbar. However, this range has been extended toward the lower pressures with the reactor B, and a second study was carried out for $p < 15$ mbar.

Another interesting parameter varied in this section was the reactor/plate gap distance ($g = 1, 6$, and 10 mm).

1. Effect of the reactor/plate gap

The self-bias is plotted for different values of g for Ar plasma ($g = 1, 6$, and 10 mm, see Fig. 9) vs the operation pressure. The remarkable difference induced by the gap on the self-bias voltage starts for pressures higher than 3 mbar. The self-bias voltage increases, in absolute value, with the reduction of the gap, being the highest for $g = 1$ mm. For lower pressures (< 3 mbar), the self-bias voltage seems to be independent of the gap separating the CCCD from the plate, which is interesting, again, for applications.

One expects that getting closer the plate to the slits of tubes, the plasma density increases bringing more energetic particles onto the plate. However, the recorded results seem to be consistent in light of numerical simulations, presented and discussed in Sec. IV B.

Figure 10 shows the maxima of RF voltage, RF current, and self-bias voltage measured for 0.8 and 5 mbar ($g = 6$ mm) and for 5 mbar ($g = 1$ mm) at 100 W. One can see that the pressure immobilizes the values of V_{MAX} and V_{bias} independent of the gap value. This indicates that the coaxial structure plays a major role in the establishment of the plasma properties, much more than the grounded external plate placed behind the slits. This information is very important, since the discharge can be (almost) independently adjusted with respect to the surface to treat.

To study the effect of pressure and gap theoretically, we numerically simulated the plasma discharge corresponding to these three cases, further presented and discussed in Sec. IV.

FIG. 8. Ar plasma images taken with the reactor B in operation for different pressures ($P_{RF} = 20$ W, $g = 10$ mm).

FIG. 9. Self-bias variation (absolute value) with gas pressure, for Ar plasma at 100 W ($g = 1, 6$, and 10 mm).

FIG. 10. Time stability of the discharge throughout the maxima of voltage, current (V_{MAX} , I_{MAX}) and self-bias voltage (V_{bias}) measured for (a) 0.8 mbar/g = 6 mm, (b) 5 mbar/g = 6 mm, and (c) 5 mbar/g = 1 mm (reactor B: Ar plasma, $P_{RF} = 100$ W).

IV. MODELING RESULTS AND DISCUSSION

Numerical simulations were performed with the Plasimo software package, introduced in Sec. II B. The aim is to study this asymmetric CCCD, especially to evaluate the density and the flux of charged particles, the average electric potential, and the power dissipated by impinging ions striking the external plate, but also to get a deeper knowledge about the electron energy and other discharge parameters in order to optimize the system. Hence, the previous experimental results become easy to explain. This model has been run with several input parameters such as gas pressure, RF voltage, and geometrical parameters taken from the experiment. It should be noted here that for the Plasimo package, there is no difference between the geometry of the two reactors A and B because its geometry is reduced to the 2D cross section in the plane (y, z) perpendicular to the cylinders axis (x). The main numerical results are presented and discussed below. It should be noted that simulations of plasma self-organization in the axial direction are highly desirable to understand the nature of cluster formation, but this issue is out of the scope of the present paper.

A. Effect of the gas pressure

To study the effect of the gas pressure on discharge parameters, two simulations were performed for 0.8 and 5 mbar, using the experimental parameters shown in Fig. 10.

The input parameters were

- (1) $V_{MAX} = 245$ V, $V_{bias} = -155$ V, $p = 0.8$ mbar, and $g = 6$ mm [experimental parameters Fig. 10(a)],
- (2) $V_{MAX} = 176.5$ V, $V_{bias} = -77$ V, $p = 5$ mbar, and $g = 6$ mm [experimental parameters Fig. 10(b)].

Figure 11 shows the 2D spatial distribution of the density of ions for the two pressures, namely 0.8 and 5 mbar. One can see that

for both cases, a low density of ions is present between the tubes, on each side of the center of the slits, but away from the slits, the density of the plasma vanishes between the two cylinders (average density of ions and electrons being close to zero). As we have already explained, the two cylinders are very close to each other, which limits the formation of a dense plasma in the space separating them ($\delta = 1.6$ mm). The outer cylinder operates as a guard ring and should force the plasma to form essentially inside the cavity created by the inner electrode and outside the slits toward the external plate. So, between the two tubes, there is a negligible loss in terms of power balance, which was the initial goal when this geometry has been designed.

To analyze the ion effect on the plate, 1D curves of different plasma parameters are plotted vs y axis for the experimental

FIG. 11. 2D density of Ar^+ maps in the plane (y, z) normal to the cylinders axis for (a) 0.8 and (b) 5 mbar, calculated by Plasimo ($g = 6$ mm).

FIG. 12. Spatial distribution of (a) the density and (b) the flux of Ar⁺ at the plate for 0.8 and 5 mbar, calculated by Plasimo. Same conditions as in Fig. 11.

position of the plate ($z = 0$, on each side of the center of the slits, corresponding to $y = 0$, see Fig. 11).

Figures 12(a) and 12(b) show the spatial distribution of the density and the flux of Ar⁺, respectively, for the two pressures on the plate, just below the slit. In Fig. 12(a), the width of the plasma spreading onto the plate d is defined for each pressure. The density and the ion flux are both higher for the lower pressure, here 0.8 mbar. Notice also that the ion flux is sharper at low pressure [Fig. 12(b)], its maximum being four times higher at 0.8 mbar compared to 5 mbar.

To better understand the pressure effect on the discharge parameters close to the plate, Figs. 13(a) and 13(b) show the spatial distribution of the dissipated power by ions on the plate and their local velocity each side of the slit, for both pressures. It should be noted here that the power dissipated by ions is defined by the scalar product of the ion current density and the local electric field for different positions (y , $z = 0$), and the velocity of ions at the plate is calculated as the ratio of ion flux by the ion density at the plate.

As shown in Fig. 13, the power dissipated by ions and the velocity of ions at the plate are both higher for 0.8 mbar. The dissipated power density is very similar to the ion flux, being sharper at 0.8 mbar and having a maximum almost four times higher compared to 5 mbar. This can be explained by the increased collision frequency for higher pressures, so that an important fraction of the power goes to the neutral gas and consequently the ion flux reaching the plate is reduced. The ion mean velocity [Fig. 13(b)] clearly indicates a collisional regime at 5 mbar with a maximum velocity at the center of the slits and a ratio between the maxima of about 3, and not 4 as found for the fluxes [Fig. 12(b)] and power density [Fig. 13(a)]. That means the plasma density slightly increases with the pressure, but the heating efficiency is definitely higher at low pressure and it is important if the sheath is collisionless, as discussed in Sec. IV B.

Let us discuss the sheath formation on both sides, the inner electrode and the plate (grounded). As said, for our configuration, the powered electrode is the inner tube; its area (A_p) is proportional

FIG. 13. Spatial distribution on the plate of (a) the power dissipated by Ar⁺ and (b) the mean velocity of Ar⁺ for 0.8 and 5 mbar, calculated by Plasimo. Same conditions as in Fig. 11.

to the internal radius and the length of the slit of the inner tube:
 $A_p = L_{in} \times 2 \times \pi \times r_{in}$.

The effective grounded electrode is the planar surface of the plate seen by the discharge, in the absence of any plasma formed in the space between the electrodes. This area (A_g) is proportional to the distance (d), with d being the width of the plasma spreading onto the plate [see $d_{p=5\text{mbar}}$ and $d_{p=0.8\text{mbar}}$ in Fig. 12(a)]. A_g is also proportional to the length of the slit of outer cylinder (L_{out}):
 $A_g = L_{out} \times d$.

As shown in Fig. 12(a), for 0.8 mbar, $d_{p=0.8\text{mbar}} \approx 62$ mm, but for 5 mbar, d reduces to about $d_{p=5\text{mbar}} \approx 40$ mm, for the same other conditions. The increase of the distance d is then related to the lateral expansion of the plasma out of the slit, which is facilitated by the decreasing the gas pressure.

For reactor B:

$$A_p = L_{in} \times 2 \times \pi \times r_{in} = 145 \times 2 \times \pi \times 5 = 4553 \text{ mm}^2,$$

$$A_g = L_{out} \times d = 150 \times d.$$

On the other hand, Fig. 14 shows the spatial distribution of the average electric potential ($\bar{V}$), along the vertical axis, starting from the plate ($z=0$) up to the internal wall of the outer tube (grounded, $z=24$ mm), for the two pressures. The sheath thickness can be clearly evaluated from the potential drop in front of each electrode. Hence, the sheath thicknesses at the powered electrode ($z \sim 20$ mm) and the grounded plate ($z \sim 0$ mm) are indicated in Fig. 14 denoted by s with a double index, the first one being related to the power “ p ” or ground “ g ,” and the second one to the pressure “0.8” and “5” being separated by a comma. Decreasing the pressure, the sheath width slightly increases at the powered electrode, but it decreases at the grounded one:

FIG. 14. Spatial distribution of the average electric potential, for Ar plasma for 0.8 and 5 mbar, for $y=0$, calculated by Plasimo. Same conditions as Fig. 11.

$p=0.8$ mbar: $s_p=3.5$ mm, $s_g=1.4$ mm, $d=62$ mm. By applying Eq. (1): $V_{bias}=-164.7$ V,
 $p=5$ mbar: $s_p=3.2$ mm, $s_g=1.8$ mm, $d=40$ mm. By applying Eq. (1): $V_{bias}=-70.8$ V.

The decreasing of the pressure causes the expansion of the plasma volume toward the grounded area, which is experimentally verified for capacitive coupled discharge.^{10,19}

According to the self-bias voltage formula [Eq. (1)], we found values very close to those measured experimentally ($\pm 6\%$ to 8%). Therefore, this legitimates the good choice of the grounded electrode surface and sheath thicknesses values estimated from the simulations, for each case. This is taken as a first rough validation of the model. In other words, the guard ring (outer cylinder) plays a minor role in the discharge energy balance, but it keeps the maximum ionization (plasma density) in the slit's region. Indeed, from Fig. 14, one can undoubtedly see that the outer electrode plays well the role of guard ring since the powered sheath thickness is larger than the distance between the cylinders ($s_p > \delta$); That means it is not possible to form a plasma in the space between the two cylinders, consistent with our initial design.

In addition, Fig. 15 shows the temporal evolution of the average electron energy during some RF periods. The average electron energy for 0.8 mbar is $\epsilon_e \sim 10.2 \pm 3.5$ eV, which is higher than that for 5 mbar, where it is only $\epsilon_e \sim 4.3 \pm 0.5$ eV.

From these two well-chosen simulations, for two pressures (0.8 and 5 mbar) corresponding to experimental conditions, the values of the surface seen by the discharge on the grounded electrode (external plate) and the sheaths' thicknesses are estimated through the spatial distribution of the average density of ions and electrons; we verified the expansion of the plasma toward the grounded electrode with the decrease of the pressure, which is in

FIG. 15. Variation of the average electron energy for Ar plasma, for 0.8 and 5 mbar during some RF periods, calculated by Plasimo. Same conditions as in Fig. 11.

FIG. 16. 2D density of Ar^+ maps in the plane (yz) normal to the cylinders axis, for (a) $g = 1$ and (b) $g = 6$ mm, calculated by Plasimo. ($p = 5$ mbar).

Figure 16 shows the 2D spatial distribution of the density of ions for $g = 1$ and 6 mm. Similarly, away from the slits, the plasma does not form between the tubes. One can see that the increase of the gap from 1 to 6 mm decreases the width of the plasma spreading onto the plate: the increase of the distance d is then closely related to the lateral expansion of the plasma, which is facilitated by the available drift space when increasing the gap.

As shown in Fig. 16, for the region of plasma bulk (between the two slits), the density of ions is higher for the gap of 6 mm than that for the short gap of 1 mm. The increase of plasma density with increasing of the gap was verified for capacitive coupled plasma:³³ the increase of the gap from 1 to 4 cm markedly increases the density of electrons, corresponding to an obstructed discharge.⁴¹ The discharge glows easier with larger gaps.

As previously, to compare the effect on the plate, the curves are plotted vs one coordinate perpendicular to the slits, y (at $z = 0$). Figures 17(a) and 17(b) show the spatial distribution on the plate of the density and the flux of Ar^+ for each considered gap. The density and the flux are much higher (about three times) for the lower gap, $g = 1$ mm, but the treated area is much narrower, in the ratio of the distance d seen by the plasma, $30/7.2 = 4$ times larger in terms of ion flux.

In addition, Fig. 18 shows the spatial distribution of the power density dissipated by ions and their mean velocity at the plate. One can see that both plasma parameters are higher and sharper for the shortest gap.

By calculations, the integral of each curves of Fig. 18(a) divided by the width at half maximum (which represents the width over which the energy is deposited), we notice a ratio of four times greater for the smallest gap (1 mm); the calculations are presented on the figure for each case. There is a huge interest to reduce the gap which act as reducing the width of the plasma slab and made the process as a linear plasma curtain lying on a surface.

To evaluate the model capabilities, an estimation of the sheaths is made similar to the one performed in Sec. IV A. Figure 19 shows the average potential along the vertical axis from the plate ($z = 0$) to the inner (upper side) of the RF electrode (to $z = 16$ mm for $g = 1$ mm and to $z = 21$ mm for $g = 6$ mm), for both

line with other experimental observations for a capacitive coupled discharge.^{10,19}

At this stage, it clearly appears the system is more efficient for the surface treatment if the working pressure is the lowest possible. In our case, reducing the pressure from 5 to 0.8 mbar enhances the plasma effect onto the plate in terms of density, flux, ion mean velocity, and power density dissipation at the surface, by at least a factor of 3, at the same RF power.

B. Effect of the reactor/plate gap distance

To study the effect of the gap distance between the reactor and the external plate, simulations were performed for a fixed pressure $p = 5$ mbar, but for two well-chosen gap values $g = 1$ and 6 mm. The input parameters were

- (1) $V_{MAX} = 176.5$ V, $V_{bias} = -77$ V, $p = 5$ mbar, and $g = 6$ mm [experimental parameters Fig. 10(b)],
- (2) $V_{MAX} = 180$ V, $V_{bias} = -87$ V, $p = 5$ mbar, and $g = 1$ mm [experimental parameters Fig. 10(c)].

FIG. 17. Spatial distribution of (a) the density and (b): the flux of Ar^+ at the plate for $g = 1$ and 6 mm, calculated by Plasimo. Same conditions as in Fig. 16.

FIG. 18. Spatial distribution of (a) the power dissipated by Ar^+ and (b) the velocity of Ar^+ at the plate for $g=1$ and 6 mm , calculated by Plasimo. Same conditions as in Fig. 16.

simulated situations ($g=1$ and 6 mm). One can see that the potential drops over both sheaths, and the plasma potential is higher for $g=1 \text{ mm}$. Through this figure, we can also estimate the sheath thicknesses (s_p and s_g) for each case. The second index of s in Fig. 19 corresponds in this time to the value of the gap.

As shown in Sec. III B (Fig. 9), the self-bias voltage is slightly higher for 1 mm , for pressures above 3 mbar .

From the spatial distribution of the ion density and the average electric potential, the values of d , s_p , and s_g are estimated for each gap value,

$g=1 \text{ mm}$: $d=10 \text{ mm}$, $s_p=6.4 \text{ mm}$, $s_g=0.9 \text{ mm}$. By applying Eq. (1): $V_{\text{bias}} = -72.3 \text{ V}$. To be compared to measured -87 V ,
 $g=6 \text{ mm}$: $d=40 \text{ mm}$, $s_p=3.2 \text{ mm}$, $s_g=1.8 \text{ mm}$. By applying Eq. (1): $V_{\text{bias}} = -70.8 \text{ V}$. To be compared to measured -77 V .

FIG. 19. Spatial distribution of the average electric potential along the normal at the plate, for $y=0$, for $g=1$ and 6 mm , calculated by Plasimo. Same conditions as in Fig. 16.

According to the self-bias voltage formula, we found values in the same order of magnitude to those measured experimentally, but the error exceeds 17% for the shortest gap ($g=1 \text{ mm}$). This is an indication that approaching the discharge closer to the plate (ground), the outer slit (grounded as well) starts to play in the RF circuit so its role as electrode cannot be neglected anymore. However, within the error margin, the coarse result legitimates the estimation of the active electrode surfaces and the sheath thicknesses provided by the numerical simulations. We take this as a second rough validation of the model.

To finalize this discussion, Fig. 20 shows the temporal evolution of the average electron energy for these last two cases. This energy is, in average, $\epsilon_e \sim 6.8 \pm 1.1 \text{ eV}$ and $\epsilon_e \sim 4.3 \pm 0.5 \text{ eV}$ for $g=1$ and 6 mm , respectively. A similar result has been already reported, showing that the increase of the gap distance from 1 to 4 cm leads to a decrease of the electron temperature in the plasma.³³

From these two last simulations ($g=1$ and 6 mm , Fig. 20), the values of the surface of the plate playing the role of grounded electrode and the sheath thicknesses have been estimated through the spatial distribution of the ion density and the average plasma potential. In addition, for a short gap of 1 mm , the density, the flux, the velocity of ions, and the energy they dissipate at the plate are all higher than those of the case of 6 mm . Moreover, the plasma slab becomes much narrower, very suitable for localized surface treatment. Therefore, it is better to operate this discharge very close to the surface to treat (external plate, here 1 mm).

At this stage, it is also interesting to validate the effect of gas obtained in Sec. IV A but this time for the shortest gap (1 mm). Hence, the two panels (a) and (b) of the Fig. 21 show, respectively, the spatial distribution of the flux and the power density dissipated by ions, for two pressures 5 and 1.4 mbar , and a fixed short gap $g=1 \text{ mm}$. One can see that both plasma parameters are higher for the lowest pressure (1.4 mbar). So again, the system is more efficient for the surface treatment if the working pressure is kept as low as possible. The lower limit is due to the ability to glow the discharge, and it depends, as experimentally found, on the type of gas used much more than the size of the reactor.

FIG. 20. Variation of the average electron energy for Ar plasma for some RF periods, for $g = 1$ and 6 mm, calculated by Plasimo. Same conditions as in Fig. 16.

assistance. Notice that this CCCD operates with only one RF power supply. Moreover, the ion flux to the plate can be tuned by changing the pressure, or simply the distance between the electrodes and the plate.

Even if the present experimental study has been performed with a metal plate, the discharge is certainly effective on the treatment of dielectric materials, since the RF sheaths drain an equivalent amount of charged particles on each electrode, overall each period. Moreover, this plasma system can be very effective for large area surface treatment when moving one (i.e., either the discharge or the plate) relative to the other in a direction perpendicular to the slits. These characteristics of the proposed CCCD give it a high versatility.

V. CONCLUSION

In this work, a novel design for capacitive coupled argon plasma is presented, formed essentially by two cylindrical coaxial electrodes and called cylindrical capacitive coupled discharge (CCCD). If both cylinders are equipped with a slit, and these slits are aligned, then it is possible to use this device for surface treatment of a flat object or plate placed just under the slits. Two reactor configurations have been designed and characterized, with different lengths (the second configuration was twice longer than the first one), and they operate properly. This can be taken as a first proof of the possible scaling-up of this system.

A linear RF plasma is formed inside the cavity of the inner cylinder, and it spreads out through the slits, forming a plasma slab between the reactor and the plate. It has a stable operation over a relatively wide range of pressures (0.8–50 mbar) or RF power (typically <1.4 kW). Experimentally, the self-bias voltage measured was negative, as expected. Obviously, the self-bias voltage increases, in absolute value, when increasing the RF injected power and decreasing the gas pressure.

Using the fluid modeling (Plasimo package), the values of the surface seen by the plasma on the grounded plate, acting as a ground electrode, could be evaluated. From the model, the lateral width under the slits receiving the charged particles and the sheath thicknesses were estimated for all studied conditions. The optimized

It should be noted here also that for the shortest gap (1 mm), even numerically it is difficult to develop a plasma. Plasimo modeling package failed (could not converge) with RF power of 100 W (same RF power for other simulated cases, Fig. 10) and for the pressures below 1 mbar. To overcome this limitation, we have simulated the case of 1.4 mbar (Fig. 21) and not the exact value of pressure used in the experiment.

As expected by its design, this cylindrical capacitive coupled discharge design produces two important ion sheaths. The one for the powered hollow electrode is usual, but the second one formed in front of the grounded plate appears to be the key for surface treatment. It is governed by the large electron losses imposed to the system by construction. The formation of an important sheath in front of the plate was the initial goal, leading to an efficient acceleration of the ions toward the plate surface and so doing effective ion

FIG. 21. Spatial distribution of (a) the flux of Ar^+ and (b) the power dissipated by Ar^+ at the plate for $g = 1$, and for 1.4; and 5 mbar, calculated by Plasimo, with $V_{\text{MAX}}/V_{\text{bias}}$ are 245 V/–135 V for 1.4 mbar and 180 V/–87 V for 5 mbar.

operation conditions require low gas pressure and the plate very close to the reactor slits.

The present configuration is very useful for the surface heating applications and for specific conditions (non-stationary cluster formation); it can be used for ion assistance on the surface under the slits.

Moreover, in this work, the plate was grounded, but it is possible to bias it as desired, positively or negatively, depending on the followed purpose. Also, it can develop a self-bias voltage if the plate is made in an isolator material. As a perspective, the use of this plasma could be extended following the power transferred from the plasma to the plate, simply by tracking the increase of the plate temperature. This would be an easy means to evaluate the efficiency of this novel discharge configuration in terms of ion flux and surface treatment.

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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