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Electrical method for the measurements of volume averaged electron density and effective coupled power to the plasma bulk

M. Henault,[1] G. Wattieaux,[2] T. Lecas,[1] J. P. Renouard,[3] and L. Boufendi[1,a)]
[1]*GREMI, Université d'Orléans, 14 Rue d'Issoudun BP6744, 45067 Orléans cedex 2, France*
[2]*LAPLACE, Université Paul Sabatier, 118 Route de Narbonne, 31062 Toulouse, France*
[3]*Ecole Polytech'Orléans, Université d'Orléans Site Galilée, 12, Rue de Blois, 45067 Orléans, France*

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Nanoparticles growing or injected in a low pressure cold plasma generated by a radiofrequency capacitively coupled capacitive discharge induce strong modifications in the electrical parameters of both plasma and discharge. In this paper, a non-intrusive method, based on the measurement of the plasma impedance, is used to determine the volume averaged electron density and effective coupled power to the plasma bulk. Good agreements are found when the results are compared to those given by other well-known and established methods. © *2016 AIP Publishing LLC*.
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I. INTRODUCTION

Radiofrequency driven capacitively coupled plasmas (CCPs) are widely used in different technologies such as thin film deposition, etching, and surface treatment. In such plasmas take place complex chemical reactions that are behind all the studied and used processes. For these purposes, electron density and temperature as well as effective coupled RF power to the plasma bulk are needed for physical and chemical models to well describe and understand the occurring phenomena.

As shown by Wattieaux *et al.*,[1,2] it is possible to use the plasma and the discharge electrical characteristics to accurately measure the average size and concentration, over the plasma volume, of nanoparticles grown or injected in plasmas generated in CCPs. However, this needs to develop a method and a tool to determine such properties without disturbing the discharge and also in a relatively easy way. It is also important that this method should be easy to be implemented and handled in industrial devices. That is one of the main challenges of this work.

Different techniques have been developed and used such as Langmuir probes, microwave interferometry, microwave resonant cavity (MRC),[3–5] hairpin probes,[6] and self excited electron resonance spectroscopy (SEERS).[7,8] However, in some conditions like when reactive gases are used to deposit insulating thin films or when dust nanoparticles are present in the plasma gas phase, the measurements are very difficult and/or the efficiency of the diagnostic tools is strongly affected.

Many works have been devoted to study theoretically and experimentally the electrical characteristics of CCPs in a wide range of configurations and operating conditions in terms of pressure, RF power, material nature used for the fabrication of the electrodes, etc. Many theoretical models were developed to understand these kinds of discharges. They brought to the fore the key role of the space charge sheaths and especially the nonlinearity of the sheaths that can have strong effects on the plasma and discharge properties. In this work, we used the homogeneous model developed by Lieberman and his coworkers[9] to describe and determine the plasma impedance and then deduce the effective coupled power to the plasma and the electron density. These measurements will be compared to those given, respectively, by the subtractive method[10,11] and the microwave resonant cavity one.

In the first part of this paper, we shall give a description of the experimental set-up that was used to perform this work, then the model we used in the second part. The third part will be devoted to the presentation of the results and their interpretation, and finally we will conclude.

II. DESCRIPTION OF THE EXPERIMENTAL SET-UP

The experimental set-up was described in details in previous articles.[1] The RF discharge is produced in a grounded cylindrical box (13 cm inner diameter) equipped with a showerhead-type powered electrode (Fig. 1). The bottom of

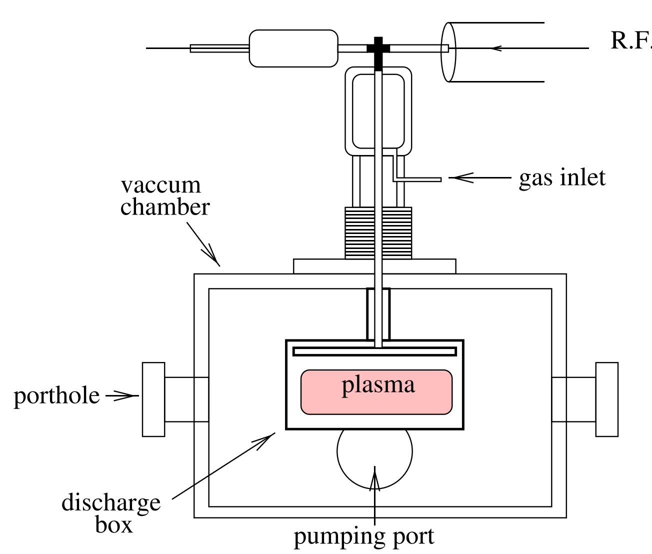

FIG. 1. Schematic of the reactor.

a)Author to whom correspondence should be addressed. Electronic mail: laifa.boufendi@univ-orleans.fr

the box is closed thanks to a 20% transparency grid. This allows a vertical laminar gas flow in the experimental conditions used here in this work.

Three vertical slits (2 mm wide, 4 cm high) allow optical access to the plasma at 0°, 90°, and 180° around the chamber. The whole system is enclosed in a vacuum vessel of 30 cm in height and inner diameter. Three optical viewports on the vacuum vessel (5 cm in diameter and 90° apart) are aligned with the slits.

The electron density was measured using the discharge box as microwave resonant cavity (Fig. 2). The resonance mode frequency shift, induced by the plasma, was used to determine the electron density. This frequency shift was measured thanks to a Schottky diode and an oscilloscope (Tektronix DPO 7254), which has a sampling frequency of 40 GS/s and 2.5 GHz bandwidth. As the plasma frequency remains lower than the resonance frequency, a perturbation model gives the relationship between the measured frequency shift and the average free electron density n_e over the plasma volume

$$n_e = A8\pi^2 f_{res0}\Delta f_{res}\frac{m_e\varepsilon_0}{e^2}, \quad (1)$$

where $f_{\mathrm{res}0}$ is the resonance frequency in the absence of the plasma, $\Delta \mathrm{f}_{\mathrm{res}}$ is the frequency shift of the cavity mode when the plasma is switched on. A is a geometrical factor which is introduced in order to take into account the electron density profile N_e and the electric field E inside the cavity. It is given by

$$A = \frac{\iiint N_e dV \times \iiint E^2 dV}{\iiint N_e E^2 dV}. \quad (2)$$

In the present work, TM010 cavity mode is considered. The corresponding resonance frequency of the discharge box is 1.738 GHz in vacuum. According to Eq. (2), $A = 1$ for an uniform electron density distribution and it rises up to 1.38 considering an ambipolar diffusion regime where the electron density profile is given by a Bessel function of the first kind (and so does the electric field). However, in the following, we will take arbitrarily $A = 1$ (uniform electron density), since we have no idea about the true electron density profile. Thus, one needs to keep in mind that even if the reproducibility of the measurement is very good, the average electron density given by the resonant cavity method might be underestimated by a factor reaching up to 40%.

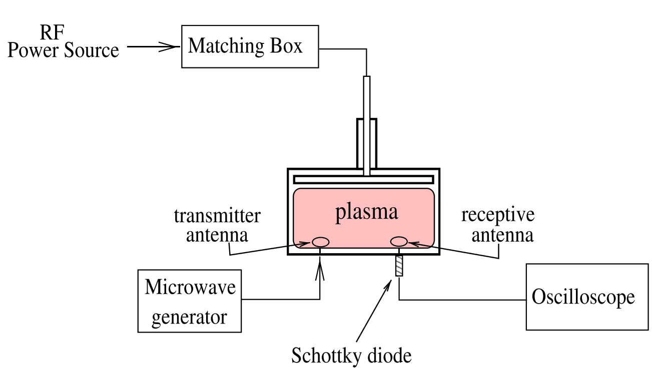

FIG. 2. Schematics of the microwave resonant cavity method.

The RF voltage and total current were measured using homemade probes. The capacitive voltage probe has been designed in order to present an input capacitance of about 3 pF. The current probe (Rogowsky coil) bandwidth is 250 MHz and causes a very low magnetic resistance in the circuit (about 0.1 Ω).

The probes were calibrated (gain and phase shift) on a 50 Ω calibrated resistance (gain) and a standard capacitance (phase shift).

III. MODEL

In order to reach the desired plasma and discharge parameters, we considered an electrical model. In fact, the acquired current signal is made of a capacitive part mainly due to the stray capacitance of the reactor and a discharge part due to the plasma. The plasma impedance has been decomposed into the series associations of the sheath impedances with the impedance of the plasma bulk.[12] In a first approximation, the electrostatic sheath was considered non-collisional meaning that their impedance can be modeled by a single capacitance C_{sheath}.[9] The plasma bulk was considered homogeneous with impedance given by

$$Z_p = (jC_0\varepsilon_r\omega)^{-1}, \quad (3)$$

where C_0 is the capacitance of the inter-electrode gap under vacuum, ω is the angular frequency of the electrical current, and ε_r is the cold plasma dielectric constant

$$\varepsilon_r = 1 - \frac{\omega_p^2}{\nu^2+\omega^2} - j\frac{\omega_p^2\frac{\nu}{\omega}}{\nu^2+\omega^2}, \quad (4)$$

where ν is the electron-neutral collision frequency, and ω_p is the plasma electron angular frequency.

According to the previous set of equations, the bulk plasma impedance Z_p corresponds to a capacitance C_0 in parallel with an inductance L_p in series with a resistance R_p (see Fig. 3). L_p and R_p representing, respectively, the plasma inductance that is related to the electron inertia and the ohmic resistance related to the electron-neutral elastic collisions are given by

$$L_p = (C_0\omega_p^2)^{-1}, \quad (5)$$

$$R_p = \nu \times L_p. \quad (6)$$

Finally, a stray capacitance C_{stray} mainly due to the counter-electrode has to be taken into account (see Fig. 3).

The overall impedance measured between the powered electrode and the grounded one is thus

$$Z_{tot} = (jC_{stray}\omega + ((jC_{sheath}\omega)^{-1} + Z_p)^{-1})^{-1}. \quad (7)$$

This impedance can also be determined experimentally

$$Z_{tot} = \frac{V_{max}}{I_{max}} \times e^{j\varphi} = |Z_{tot}| \times e^{j\varphi}, \quad (8)$$

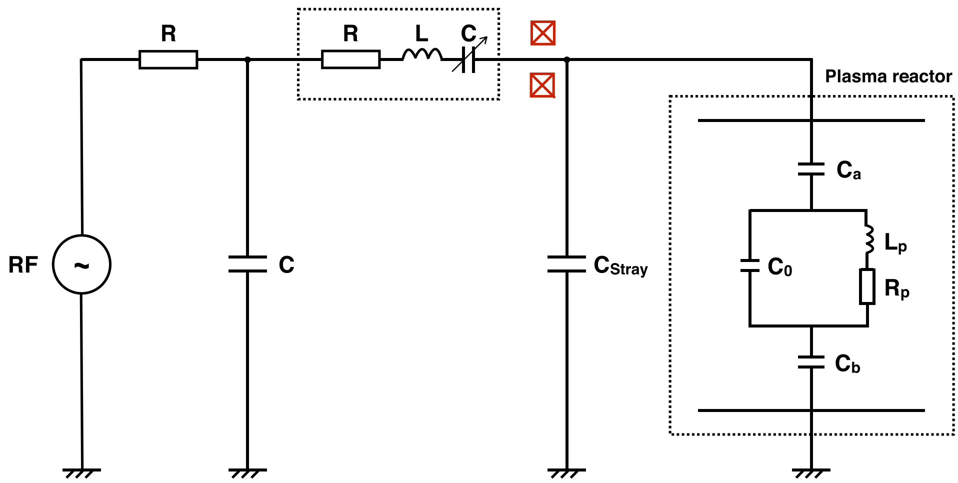

FIG. 3. Equivalent electrical circuit of the experimental set-up.

where V_{max} and I_{max} are the amplitude of the AC part of the voltage and the current at the powered electrode, and φ is the current/voltage phase shift.

C_{stray} can be determined from the measurement of $|Z_{tot}|$ when there is no plasma between the electrode since $C_{stray} \gg C_0$.

The active power P (or coupled power) is given by

$$P = \frac{U.I}{2}\cos(\varphi), \tag{9}$$

where I and U are, respectively, the amplitudes of the current and the voltage measured at the powered electrode.

It is straightforward that C_0 and L_p do not significantly affect the discharge impedance when

$$\frac{\omega_p^2}{\upsilon\omega} \gg 1\,;\, \frac{\omega_p^2}{\upsilon\omega} \gg \frac{C_s}{C_0}\,;\, \frac{\omega_p^2}{\omega^2} \gg \frac{C_s}{C_0}. \tag{10}$$

These conditions are usually met for laboratory CCP devices operating below 100 Pa. Then, we have

$$\frac{I}{U} = (C_s + C_{stray}).\omega, \tag{11}$$

$$I_D = C_s.\omega.U, \tag{12}$$

$$R_p = \frac{2P}{I_D^2}, \tag{13}$$

$$\omega_p^2 = \frac{\upsilon}{C_0.R_p}, \tag{14}$$

where C_s is the sheath capacitance, and C_{stray} is the stray capacitance. I_D is the amplitude of the discharge current, $R_p = \nu.(C_0.\omega_p^2)^{-1}$ is the bulk plasma resistance, ω_p $(\mathrm{rad\cdot s^{-1}}) \approx 56.4 \times n_e$ $(\mathrm{m^{-3}})$ is the plasma angular frequency, $\nu = n \times \sigma \times v_{the}$ is the electron/neutral elastic excitation collision frequency with $n = p/(k_B T)$ the gas density (with p the pressure, k_B the Boltzmann constant, and T the gas temperature), $\sigma(\mathrm{m^2}) \approx 5 \times 10^{-20}$ is the associated cross-section, and v_{the} $(\mathrm{m\cdot s^{-1}}) \approx 5.9 \times 10^5.\ \sqrt{T_e\,(eV)}$ is the electron thermal velocity.

According to the previous set of equations, we will separate the parameters in two families. The first one corresponds to the parameters which do not fluctuate but for which a systematic error δ may be done on their estimation (T, T_e, σ, C_0, C_{stray}, and ν). The second family concerns the parameters, which may randomly fluctuate by a quantity Δ but for which the systematic error is negligible thanks to an accurate calibration (U, I, and φ). Thus, P_{mes} the measured active power and nmes the measured electron density are given by

$$P_{mes} = P + \Delta P + \delta P, \tag{15}$$

$$n_{mes} = n_e + \Delta n_e + \delta n_e. \tag{16}$$

As P depends on parameters that are supposed to be determined with a negligible systematic error, the active power is evaluated with a very good accuracy

$$\frac{\delta P}{P} \approx 0\ . \tag{17}$$

Usually, $|\varphi|$ is very close to $\pi/2$ in CCP discharge. The uncertainty of this parameter is by far the most influent one in the determination of ΔP and Δn_e. By considering that the random relative variations of U, I, and φ do not exceed a few percent corresponding to what we observed experimentally

$$\frac{\Delta P}{P} \approx \frac{\Delta n_e}{n_e} \approx \tan(\varphi)\Delta\varphi. \tag{18}$$

Finally, if we assume that the relative systematic error of ν is much more important than the relative systematic error of C0

$$\frac{\Delta n_e}{n_e} \approx \frac{\Delta\upsilon}{\upsilon}\ . \tag{19}$$

In the following, we consider a $\pm 1\%$ uncertainty for U and I, $\pm 0.3\%$ for φ, and systematic errors of $\pm 50\%$ for T_e, $\pm 20\%$ for σ, and $\pm 10\%$ for T. We also assume $|\varphi| = 1.55$ rad. In these conditions

$$\frac{\Delta P}{P} \approx \frac{\Delta n_e}{n_e} \approx 22\%, \tag{20}$$

$$\frac{\delta n_e}{n_e} \approx 47\%. \tag{21}$$

We developed a numerical code to determine φ with a very good accuracy, and we find out then that the experimental variation of φ from one measurement to another in the same working conditions is $\pm 3\%$ when the plasma is on and less than $\pm 0.01\%$ when the plasma is off. Consequently, by averaging 100 successive measurements, the uncertainty of the average value of φ is $\pm 0.3\%$ when the plasma is on. This averaging limits the temporal resolution of the determination of P and n_e to about 10 μs.

To sum up, the random fluctuations of φ should theoretically lead to a reproducibility of $\pm 22\%$ in the determination of P and $(n_e - \delta n_e)$. The accuracy in the determination of P is very good, whereas it is $\pm 47\%$ for n_e (can be improved if T_e or σ are known with a better accuracy).

Finally, n_e corresponds rigorously to the inverse of the spatial average value of the electron density in the plasma bulk of our reactor. For a 1D cosine profile with a ratio $L_P/L_S = 20.100$ where L_P is the plasma bulk thickness and L_S is the sheath thickness, the electron density provided by our electrical model is underestimated by a factor of about 1.2 to 1.7 compared to the true electron density average value. As for the resonant cavity method, this corresponds to another systematic error that can only be corrected if the electron density profile is known or assumed. Consequently, the value that we obtained by considering that the electron density is constant in the plasma bulk should be understood as an estimation of the electron density average value instead of the exact value of it. This diagnostic should thus be preferentially used when one wants to follow the relative evolution of the average electron density such as it may be useful in dusty plasma applications.

IV. RESULTS AND DISCUSSION

The experiments were first conducted in pure argon plasma and then in argon-acetylene gas mixture (2% C_2H_2). The reason is to operate in a relatively simple situation (Ar) and then to include a dust forming chemistry ($Ar + C_2H_2$). We already know that in this last situation the dust particles induce strong modifications of both plasma and discharge electrical parameters. This is due to electron attachment on the growing nanoparticles.

A. Effective coupled power

Figure 4 gives an example of measurements of the effective plasma coupled power versus the power provided by the power supply. The obtained values using the electrical method are compared to those given by the so-called subtractive one.[10,11] There are two ways to perform the subtractive method. Either by keeping constant the RF voltage at the active electrode or by keeping constant the RF current at the matching box output. We chose to consider the voltage method. This method is carried out as the following. First, the impedance matching is set for a given power P_0 by keeping the pressure as low as possible in the reactor in order to prevent plasma ignition. The RF voltage amplitude V_0 at the powered electrode is recorded in this condition. When Ar is introduced in the reactor, the matching box is tuned after plasma ignition. At this point, the power provided by the generator has to be increased to P_1 in order to set the RF voltage amplitude at V_0. This power increase corresponds to the estimation of the plasma coupled power. However, it is straightforward to demonstrate that the result is overestimated since

$$P_1 - P_0 = R_P I_p^2 + R(I_R^2 - I_{R0}^2), \tag{22}$$

where I_p is the RMS amplitude of the plasma current, R is the series resistance of the matching box. I_R and I_{R0} are, respectively, the RMS amplitude of the current crossing R when the plasma is ignited and when it is off. $R_P I_p^2$

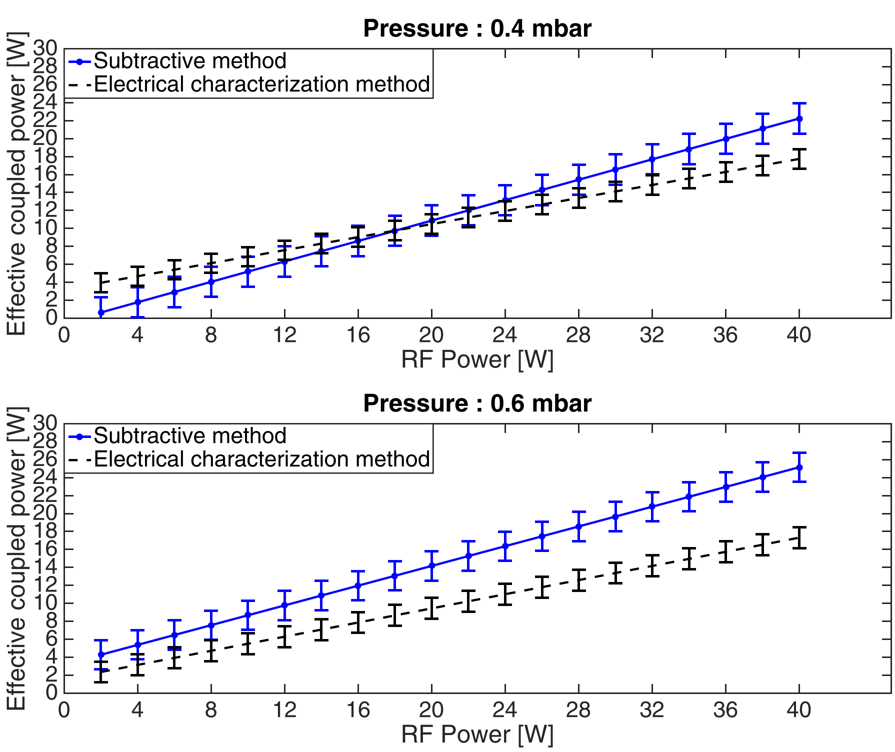

FIG. 4. Comparison of the effective coupled power to the plasma estimated by the electrical method and the subtractive one for two different pressures.

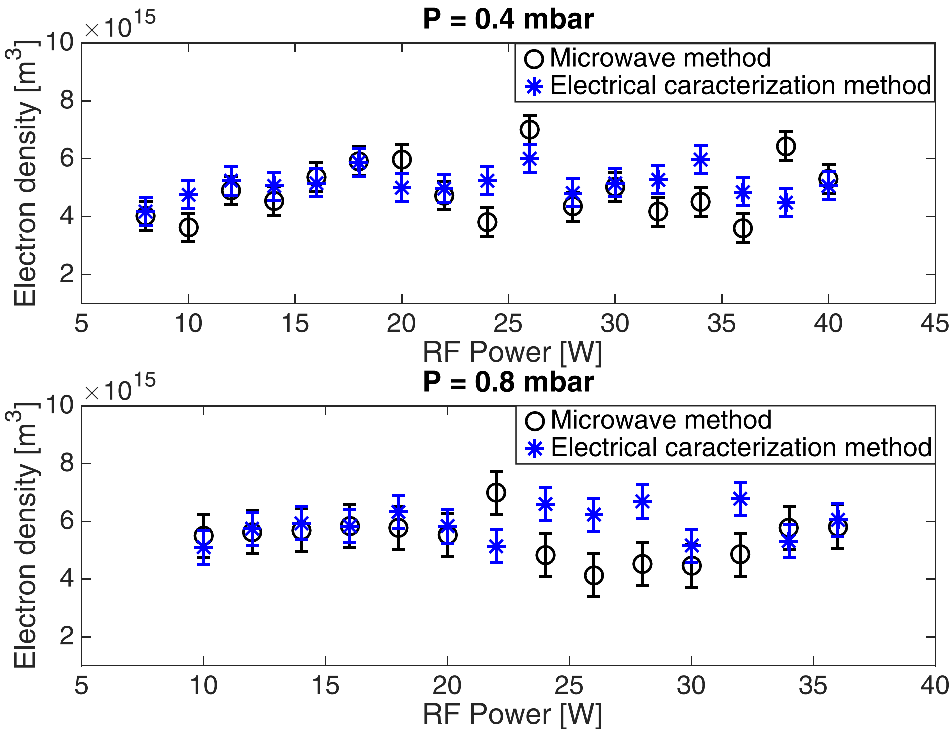

FIG. 5. Comparison of the electron densities given by the electrical method and the microwave resonant cavity one for two different pressures.

corresponds to the plasma coupled power. $P_1 - P_0$ has been computed with the parameters determined in Ref. 2, and we noticed that in this situation, the estimation of the coupled power by the subtractive method is overestimated by a factor of 2. This is coherent with what is displayed in Fig. 4 where there is a rather good agreement between both methods. The fact that the discrepancy increases with the pressure indicate that R_p (or ν/n_e according to the homogeneous model) decreases when the pressure increases but this point is beyond the scope of this publication.

B. Electron density

Figure 5 gives the evolution of the electron density with the generator output power for two different values of the argon gas pressure in the reactor. Here, we can also see that the values deduced from the plasma impedance and from the microwave resonant cavity method are very similar.

In order to better appreciate the agreement between these methods, Figure 6 shows the evolution of the electron density with the gas pressure for a constant plasma coupled power (10 W).

Finally, the electron density has been monitored during a dust nanoparticles nucleation and growth in an argon-acetylene gas mixture thanks to the electrical method presented in this work (see Fig. 7 where a comparison is carried out with MRC). The reactor pressure was 0.4 mbar, and the RF power from the generator was 10 W at the beginning of the growth.

This evolution is quite similar to those already obtained using the MRC method for the same chemistry or for the silane-based one.[2,5] These measurements were performed in a new reactor dedicated to nanoparticle metrology in which also different methods are being implemented in order to be compared. Coming papers that are under preparation will report on these works.

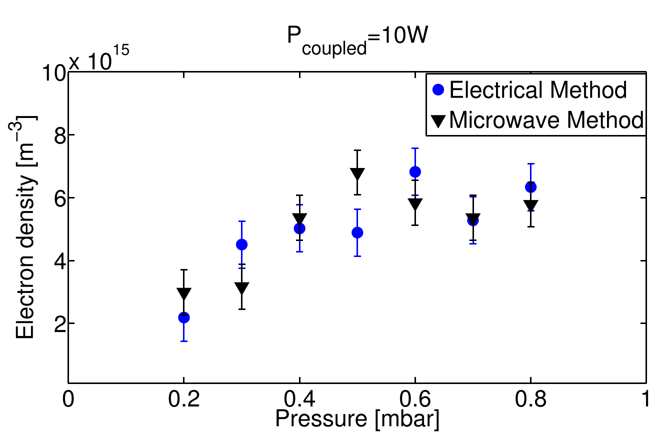

FIG. 6. Evolution of the volume average electron density with gas pressure obtained by the two diagnostic methods for 10 W effective coupled RF power.

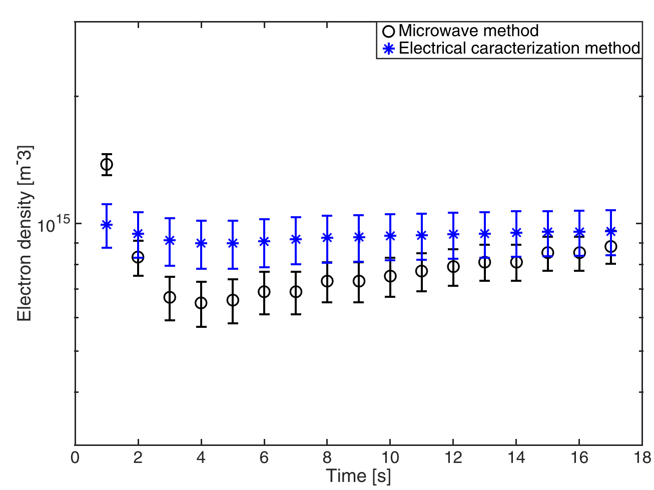

FIG. 7. Time evolution of the electron density for an argon-acetylene gas mixture dust forming plasma.

V. CONCLUSION

In this experimental work, we compared two diagnostic methods that allow the measurement of the volume averaged electron density. For the MRC, we need to install two small antennas in the plasma box, while for the second this very important plasma parameter is deduced from the plasma impedance. This means that this method can be easily implemented and used for industrial devices. Moreover, as it can be monitored through a very fast numerical code, and it can allow monitoring the plasma process with a time resolution of less than 1 ms.

In the chosen RF power and pressure ranges, the two methods used in this work have shown a very good agreement for both the volume averaged electron density and the effective coupled power to the plasma bulk. Even if the MRC method does not disturb the plasma, it is more difficult to handle and is not convenient for an industrial device for the metrology of nanoparticles. The one based on the electrical characterization of the discharge and plasma is a powerful and robust one and more convenient. In forthcoming works, it will be used to sizes grown or injected nanoparticles into the plasma.

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