## On the in-situ determination of the effective secondary electron emission coefficient in low pressure capacitively coupled radio frequency discharges based on the electrical asymmetry effect

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Radio frequency (RF) driven Capacitively Coupled Plasma (CCP) sources are extensively used in surface modification, layer deposition, and etching applications, particularly in the field of microelectronics. To reveal the fundamentals of such sources and to achieve the most effective processing, the knowledge-based optimization of these plasma sources is necessary, which requires well-defined laboratory experiments and high-fidelity simulations. The latter need a set of reliable input data for both the gas-phase and surface processes taking place in the plasma source. Among these data, the ones that describe the interaction of the plasma species with the surrounding surfaces are often not known with the desired accuracy. Therefore, in modeling studies, the interaction of ions and electrons with the surfaces is often approximated by a simplified approach: the models use (i) an effective ion-induced secondary electron yield,  $\gamma$ , which also includes contributions of species other than ions [1] and (ii) an effective elastic electron reflection coefficient, R, which is a reasonable approximation as long as the energy of the electrons at the surfaces remains low. Even these coefficients are not available to modelers for various gas/electrode material combinations due to the effects of the gas/plasma on the surface [2] and a lack of the in-situ surface diagnostics. The situation is further complicated by the fact that these values may depend on the particle energy distribution functions and electrode surface conditions and that the surface coefficient values available in the literature often originate from surface physics experiments conducted under ultrahigh vacuum conditions with heavily sputtered samples, which scenario differs strongly from those found in practical discharge physics experiments and applications. Therefore, during the past years a number of studies applying various approaches have been carried out to determine the values of  $\gamma$  and R, in situ [3, 4, 5]. In these studies, experimental recordings of some plasma characteristics and computational description have been combined to derive the surface coefficients in CCPs, by fitting the measured and computed data via the unknown surface coefficients.

In our previous work [6] we have explored the possibility to determine the surface coefficients discussed above via measurements of the DC self-bias voltage that develops in a geometrically symmetrical CCP due to the Electrical Asymmetry Effect [7] when the discharge is driven by a base RF harmonic and its second harmonic with a controllable phase angle. In [6] it was found that  $\eta$  is sensitive to  $\gamma$ , but can be taken independent of R, this way offering a way to determine  $\gamma$  based on the measurement of  $\eta$ .

Here we report our combined experimental and simulation studies that utilise this behavior. The operating conditions in the experiment are chosen in a way that (i) a precise measurement of the RF discharge voltage waveform is possible and (ii) the secondary electron emission from the electrodes has a strong effect on the measured DC self-bias voltage. For our investigations, a geometrically symmetric experimental cell is used, which consists of a glass cylinder with an inner diameter of 92 mm and two stainless steel electrode plates facing each other at a distance of L = 27.5 mm. The top electrode of the cell is driven by the RF voltage

$$\phi(t) = \phi_1 \cos(2\pi f_1 t) + \phi_2 \cos(4\pi f_1 t + \theta), \tag{1}$$

while the other electrode is grounded. Base frequency values of  $f_1 = 2$  MHz and 4 MHz and Ar operating pressures of p = 40 Pa and 80 Pa are used, as well as voltage amplitudes  $\phi_1 = 150$  V and  $\phi_2 = 75$ V. By applying a voltage waveform given by eq. (1), the DC bias is controlled by the phase angle  $\theta$ . By scanning  $\theta$  over the  $[0^\circ, 360^\circ]$  interval the  $\eta(\theta)$  function exhibits a nearly triangular shape with relatively flat parts near the maximum and minimum (see figure 1). The match between the measured and computed bias voltage values is searched for phase angles at/near  $\theta = 0^\circ/180^\circ$ .

In the simulations, we use a computational framework that consists of two code modules: (i) a PIC/MCC code that traces electrons and Ar<sup>+</sup> ions (more precisely electron and ion 'superparticles') in the neutral background gas that comprises ground-state Ar atoms and Ar atoms in an number of excited levels, and (ii) a Diffusion-Reaction-Radiation (DRR) module, which computes the spatial density distributions of the Ar atoms in these excited levels, based on the rates of the collision processes computed in the PIC/MCC module and on the rates of the radiative channels (spontaneous emission and re-absorption) within the system of the excited levels and between some of these excited levels and the Ar atom ground-state [8]. The calculations also take into account pooling ionization between the excited atoms, as well as electron-impact stepwise ionization from the excited levels and quenching



Fig. 1: Experimental and computed  $\eta(\theta)$  curves for  $\gamma = 0.07$ and DC self-bias values at  $\theta = 0^{\circ}/180^{\circ}$  for various values of  $\gamma$  (individual points). Discharge conditions: Ar, p = 80 Pa, L = 2.75 cm,  $f_1 = 4$  MHz,  $\phi_1 = 150$  V,  $\phi_2 = 75$  V, R = 0.7.

of the excited levels by neutrals, as well as diffusion losses. The elastic reflection coefficient of electrons is taken to be R = 0.7, based on [4, 5] and the simulations are conducted with various  $\gamma$  values to find the best match with the experimental value of the DC self-bias voltage,  $\eta$ .

Figure 1 illustrates the dependence of the DC self-bias voltage on  $\gamma$  near the extrema of the  $\eta(\gamma)$  curve, based on which a  $\gamma$  value of 0.07 was determined for the actual discharge conditions (Ar, p = 80 Pa, L = 2.75 cm,  $f_1 = 4$  MHz,  $\phi_1 = 150$  V,  $\phi_2 = 75$  V), as well as the full  $\eta(\gamma)$  curve obtained from the calculations using  $\gamma = 0.07$  and the same curve measured in the experiment. These latter two curves show a very good agreement, confirming the in-situ effective secondary electron yield of  $\approx 0.07$ .

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