18. MODELLING OF PLASMA REACTORS

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18. 1. INTRODUCTION

The research activity in 2005 was fully focused onto the modeling of several configurations of Plasma Reactors (PR), used in different types of industrial chains with electronics, photovoltaic, optics, food industry, or surface modification.

The main objective with this research program is the development of sophisticated simulation tools, to optimize the operation conditions of different PRs, according to the specific needs with each application. The validation of such tools requires a strong interplay between modeling and experiment, without which it is not possible to improve the understanding of the main mechanisms controlling discharge operation.

Several projects have been initiated, pursued or concluded, in the following main fields of investigation:

- Inductively-coupled plasma reactors;
- Microwave plasmas sustained by an Axial Injection Torch;
- Microwave plasma reactor with coaxial geometry at 2.45GHz frequency in argon;
- Capacitively-coupled radio-frequency discharges in hydrogen.

18.2. INDUCTIVELY-COUPLED PLASMA REACTOR FOR THE ETCHING OF METAL OXIDES[1]

We have started the modeling of an inductively-coupled plasma reactor (ICP-R), for the etching of metal oxides by fluorocarbon plasmas.

Standard fabrication techniques of CMOS-base electronic devices generally apply deep etching processes to silicon oxide (with low-dielectric constant). Usually, this plasma processing technique is carried out within ICP-R's, which present the advantage of operating at very low pressures (~mTorr) and high plasma densities (~10^{13} cm^{-3}), thus ensuring both the effectiveness of the etching mechanism and a reduced contamination. The *full description of etching processes within an ICP-R* can significantly contribute to their control and optimization. However, the development of such a global simulation program constitutes a very hard task due to the complex and diverse nature of the phenomena involved: electromagnetic, chemical (either in gas or in surface phases) and transport. The development of such a simulation tool will bring together complementary competences in the domains of Plasma Physics and Engineering, Surface Physics and Material Science. We intend to develop *a multi-scale model of an ICP-R* (Figure 18.1) *for the etching of metal oxides, by fluorocarbon plasmas.*

In 2005, we have concentrated onto the description of the excitation source part of the reactor, by developing a two-dimensional simulation tool that calculates the electromagnetic field distribution therein.

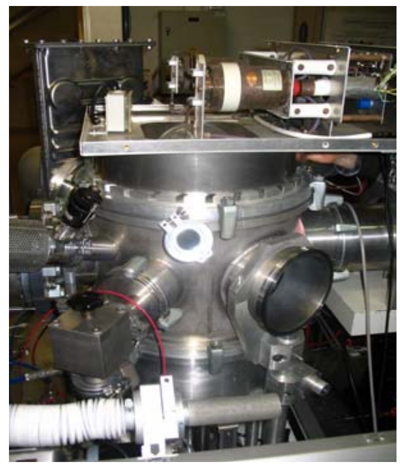

Figure 18.1 - ICP reactor with the LPCM (Nantes, France)

18.3. MICROWAVE PLASMAS SUSTAINED BY AN AXIAL INJECTION TORCH (*TORCHE À INJECTION AXIALE*, TIA)[2]

The disposal of pollutants produced by industrial processes is a mandatory task to ensure an environmental protection within a sustained development framework. Recently, a *microwave plasma torch* was applied *in waste treatment,* to the destruction of dangerous chemical compounds (such as Volatile Organic Compounds (VOCs) and BEXT aromatic hydrocarbons), with great success. Results show that this is a non-expensive and efficient way of neutralizing dangerous products, yielding destruction rates above 99,999% when applied to trichloroethylene and carbon tetrachloride.

The microwave plasma under study is produced by a TIA inside a cylindrical reactor (5.5 cm radius and 15 cm high) – Spanish patents P200201328 and P200302980. The 2.45 GHz discharge runs in atomic (Ar, He) or molecular (N_2, air) gases at atmospheric pressure, for powers ranging from 300 to 3000 W and gas flow-rates between 0.5 and 13 L/min, which shows the versatility of the plasma. The

[1] Work carried out in collaboration with the *Laboratoire des Plasmas et des Couches Minces* (LPCM) of the *Institut des Matériaux de Nantes* (IMN, Nantes, France).

[2] Work performed in collaboration with the Physics Department of the University of Cordoba (Cordoba, Spain).

system is also very stable and resistant to the introduction of water-vapour-saturated gaseous samples, unlike the majority of microwave plasmas.

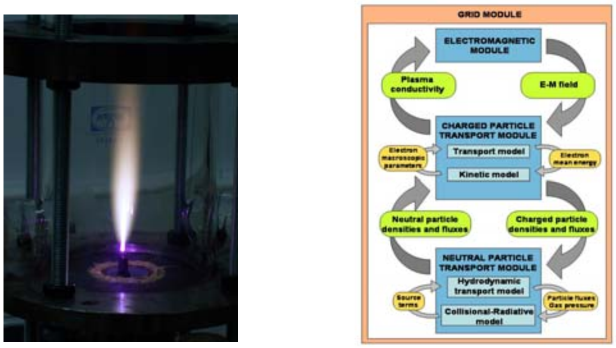

Figure 18.2 - Microwave torch with the Physics Department of the University of Cordoba, Spain (left), and schematic of the self-consistent model (right).

The project intends to develop a two-dimensional, self-consistent model for microwave helium plasmas, created by the TIA inside the reactor chamber. The model will be developed following a modular structure, each module addressing to a specific task (Figure 18.2). This clarifies model's structure, enhancing its versatility as a simulation tool.

The first two modules of the model have been successfully developed:

- *Grid Module.* The grid design follows the reactor geometry, being denser in regions where the plasma parameters are expected to present steep variations. For this two-dimensional model, variable grids have been defined along both axial and radial directions, with a higher point density at the tip of the torch and near the reactor walls. The number of grid points is high enough as to provide both numerical accuracy and a proper description of the plasma processes, while ensuring acceptable running times. These conditions were fulfilled adopting a non-uniform flexible grid, to discretize model equations (Figure 18.3).
- *Electromagnetic Module.* This module solves Maxwell's equations for the two-dimensional distribution of the electromagnetic (e-m) field within the reactor-TIA system, for a given spatial profile of the plasma conductivity. A DC-type description has been adopted to write the time dependence of the e-m field (and thus to calculate the transport features with charged particles), since the electron-neutral collision frequency at atmospheric pressure is higher than the microwave excitation frequency. Since both the TIA and the reactor chamber have axial symmetry, the e-m field components do not depend on the azimuthal angle. Moreover, the plasma dimensions and the coaxial nature of the TIA excitation device justify the assumption of a TM propagation mode, for the e-m field in the reactor-TIA system. Boundary conditions have been carefully chosen, particularly at the interface between the reactor chamber and the coaxial excitation waveguide, which acts as an open boundary. Absorbing boundary conditions have been considered to eliminate non-physical reflections coming from modelling the coaxial waveguide as a limited system.

Globally, a description of the electromagnetic field distribution with this microwave discharge has been successfully achieved.

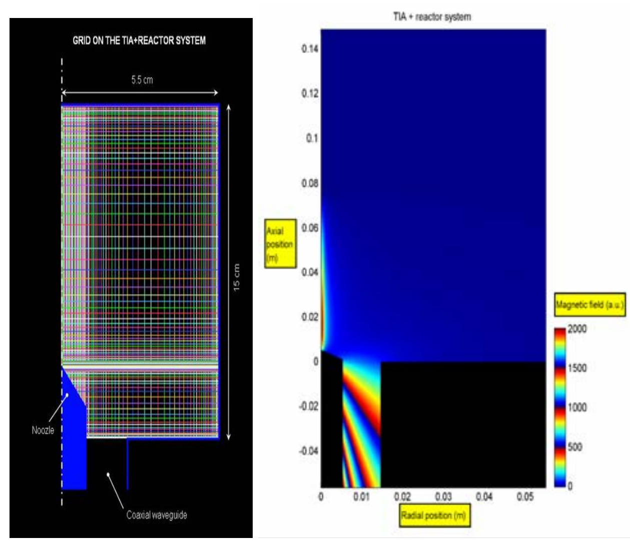

Figure 18.3 - Grid adopted (left) and magnetic field contour plot (right) with the reactor + TIA system.

18.4. MICROWAVE PLASMA REACTOR WITH COAXIAL GEOMETRY AT 2.45 GHZ FREQUENCY IN ARGON[3]

We have pursued the characterization (both experimental and theoretical) of a microwave plasma reactor with coaxial geometry (corresponding to the sequence metal-air-dielectric-plasma-metal of propagation media, within a ~1cm tube radius), operating at 2.45GHz frequency in argon, for 10-100mTorr pressures and 100-1000W coupled powers. The experimental analysis is essentially developed at the LPGP (Figure 18.4), whereas numerical simulations are carried out at the CFP.

A self-consistent one-dimensional (radial) fluid-type code has been used to demonstrate and to analyze the development of an electron-plasma resonance, associated with the strong density gradients that occur within space-charge sheath regions, at discharge boundaries. The code couples the transport equations (continuity and momentum transfer) for electron and ion particles and for the electron mean energy, Poisson's equation for the space-charge field associated with charge separation regions, and Maxwell equations for the adequate surface-wave (SW) propagating mode. Results reveal a strong increase in the SW electric

[3] Work carried out in collaboration with Dr. C. Boisse-Laporte (with the *Laboratoire de Physique des Gaz et des Plasmas* (LPGP), Orsay, France).

field, at positions where the plasma frequency equals the microwave excitation frequency (resonance effect), thus evidencing the strong plasma-wave coupling responsible for discharge maintenance.

In 2005, the work was focused mainly on the following topics.

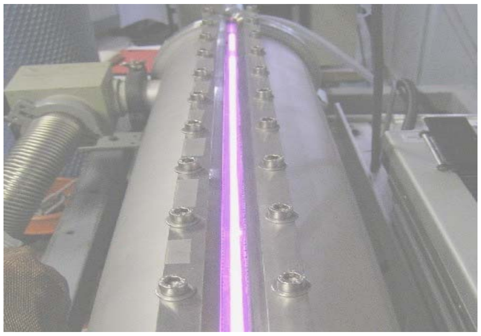

Figure 18.4 - Coaxial microwave plasma reactor with the LPGP (Orsay, France).

(i) Systematic runs of a numerical simulation code describing the microwave discharge, in view of characterizing the electron-plasma resonance as a function of the operating conditions (pressure and power coupled). The results obtained revealed that, at low pressures, there is a strong cross-linking between resonance and space-charge sheath, thus making impossible to separate the regions associated with these two phenomena.
It was also possible to define a similarity curve for the average power density deposition, for various electron densities, pressures and radial dimensions (Figure 18.5).

(ii) The experimental detection and characterization of the electron plasma-resonance, by means of directional measurements using a planar Langmuir probe. Results confirm the existence of strong deformations, associated with I-V characteristics obtained at different probe orientations. These deformations can be related to the presence of a supra-thermal flux of electrons, following their collisionless heating within resonance region.
Comparisons between calculated and measured values for the electron density and the wave attenuation constant were also carried out.

(iii) The analysis of the strong anisotropies associated with the presence of electron-plasma resonances, comparing model predictions with experimental measurements. For this study, the stationary fluid-type electron transport equations were updated, to include some extra non-linear wave-plasma coupling terms. The latter introduced an axial electron flux, hence perpendicular to the model's one-dimensional (radial) direction, which is to be compared to the relevant experimental information. In particular, the latter is to be obtained under stationary-wave conditions, for which the presence of extraordinary fluxes can be detected as perturbations to the electric field distribution.

Globally, an overall description of the microwave discharge under study was successfully achieved.

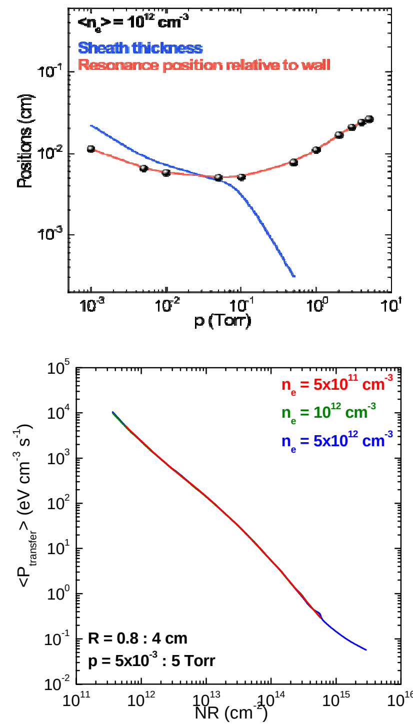

Figure 18.5 - Relative position of resonance and space charge sheath boundary (up) and similarity curve for the average power density (down).

18.5. CAPACITIVELY COUPLED RADIO-FREQUENCY DISCHARGES IN HYDROGEN[4]

We have concluded the *modelling and the characterization* of capacitively-coupled radio-frequency discharges in hydrogen, produced within a cylindrical parallel-plate reactor. These discharges are routinely used in plasma-assisted material processing applications. Model results were compared to experimental measurements (Figure 18.6).

The discharge characterization covered a wide range of excitation frequencies (13.56 – 80.0MHz), gas pressures (0.2 – 6Torr) and applied RF voltages (50 – 800V), and used a state of the art two-dimensional fluid model (to describe the dynamics of electrons, positive ions H^+ H_2^+ H_3^+ and negative ion H^- in the reactor), self-consistently coupled to a homogeneous collisional-radiative model for hydrogen (including a very complete kinetic scheme for $H_2(X_1\Sigma_g^+, v=0..14)$ vibrationally excited ground state molecules and H(n=1-5) electronically excited atoms).

[4] Work performed in collaboration with Dr. G. Gousset (with the *Laboratoire de Physique des Gaz et des Plasmas* (LPGP), Orsay, France), and with Dr. Jacques Jolly (with the *Laboratoire de Physique et Technologie des Plasmas* (LPTP), Palaiseau, France).

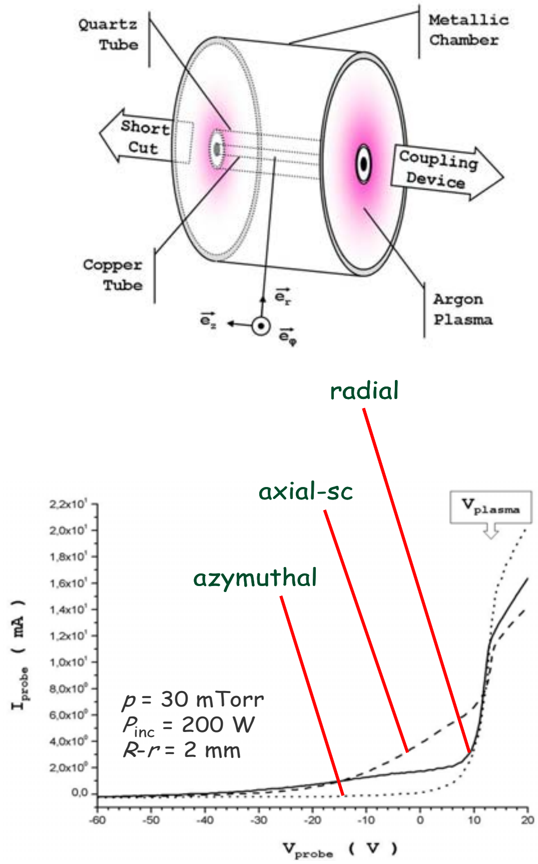

Figure 18.6 - (I,V) direction measurements using a planar Langmuir probe.

A good agreement was found between calculated results and experimental measurements for the self-bias voltage, the plasma potential and impedance and the electrical power coupled to the plasma. However, model predictions for the electron density and the self-bias voltage showed only a qualitative agreement with experiment, with calculated values underestimated with respect to measurements. This qualitative disagreement is only slightly dependent of the kinetic scheme adopted, and probably is a direct consequence of the homogenous model describing the transport of neutral species. To clarify this, *a two-dimensional, time-dependent, hydrodynamic gas model was developed, based on the Navier-Stokes / Saint-Venant equation system plus a multi-component reactive mass transport module*. This innovative model is the key piece of a powerful predictive tool yet under development, to be used in optimizing plasma reactors for material processing.

Calculations of the power absorbed by electrons W_e and the power spent on ion acceleration W_i showed that both these quantities increase with either V_{rf} and f, scaling as $W_e \sim V_{rf}^{1.2} f^{1.6}$ and $W_i \sim V_{rf}^{2.5} f^{1.6}$ at constant pressure. For applications, it is advantageous to operate at low applied voltages (regardless of frequency), in order to maximize the fractional power coupled to the electrons while minimizing that transferred to the ions. Moreover, W_e/W_i varies linearly with p at constant V_{rf}, thus showing that an increase in the pressure favors the transfer of power to the electrons.

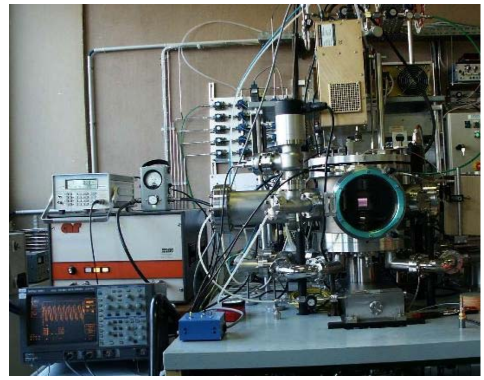

Figure 18.7 - Capacitively-coupled radio frequency reactor with the LPTP (Palaiseau, France)

A good agreement was also found between simulations and measurements (the latter obtained by LIF) for the density of H-atoms, at different discharge operating conditions (Figure 18.8). This corresponds to an improvement with respect to previous model results (failing to predict the correct features with the H-atoms density, even qualitatively), and it was achieved by using recent *in-situ* measurements of the wall-recombination coefficient for hydrogen atoms.

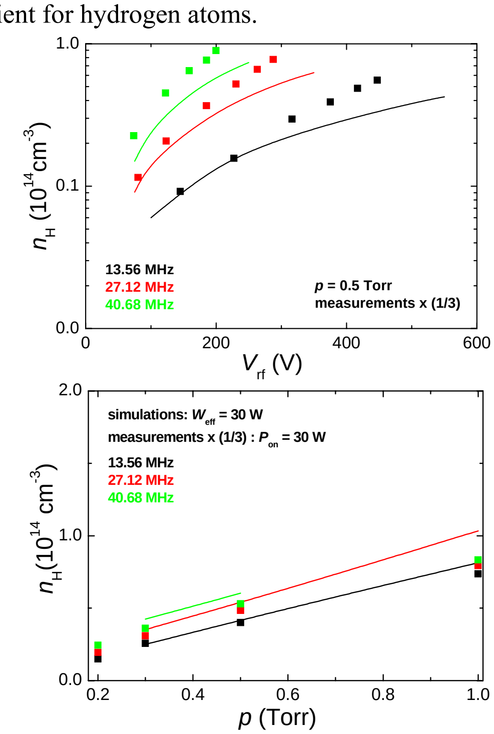

Figure 18.8 - H-atoms density as a function of applied voltage (up) and pressure (down)