Hybrid Model of Parallel Plate RF Discharges in H_2: Effect of DC and Tailored Voltage Waveforms

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Although hydrogen is the main component of many gas mixtures used for plasma processing, the physics and chemistry of even pure hydrogen discharges are not yet fully understood. This paper reports new results concerning the capacitively coupled hydrogen discharge, obtained by a hybrid model. The model is that developed in Bari University improved to include anisotropic collisions and a better treatment of secondary emission. New test cases were considered, concerning DC enhanced RF discharges and plasma production in a RF reactor using tailored voltage waveforms (TVWs). The agreement with experiments in the DC case is quite satisfactory. Work is in progress to validate the model in the TVWs case based on the experiments conducted in Palaiseau.

Hydrogen is an essential component in a variety of gas mixtures employed in plasma enhanced chemical vapor deposition (PECVD). This is mainly due to the high reactivity of H atoms, produced by electron impact dissociation of molecular hydrogen. This work analyzes the results obtained by a numerical model of CCP RF discharges with pure hydrogen feed, based on a fully kinetic plasma model coupled to fluid equations for the chemical kinetics of atoms and vibrationally excited molecules, with reactive boundary conditions. Moreover, the model results were compared to the electron density measured by hairpin resonator probe and the voltage waveforms determined using a novel derivative probe in the case of discharges ignited by tailored voltage waveforms (TVWs) [1] that allow to decouple the plasma density and the energy of the ions bombarding the substrate.

A one-dimensional in space, three-dimensional in velocity, fully self-consistent model has been developed since the late 90s by the Bari University group, to study RF capacitively coupled plasmas (CCPs) in hydrogen [2-5]. In this case, it is necessary to realize a self-consistent coupling of the electron transport with the chemical kinetics of atoms and vibrationally excited molecules, with reactive boundary conditions, specifically to solve at the same time the electron transport and chemical kinetic problems taking into account their coupling.

This model can be defined as a hybrid particle/fluid where the charged species (electrons and five ion species including negative ions) are described as particles propagating under the effect of the self-consistent electric field, while the neutral particles, namely 14 vibrational levels of the electronic ground state of hydrogen molecules, plus H atoms, are described in terms of numeric densities on a mesh which are the solution of a system of rate equations including diffusion and surface reactions.

Special features of the code are the rigorous inclusion of the thermal motion of neutrals in the statistical sampling of the ion/neutral collision frequency, and a special variance reduction technique, based on differential particle weighting, which allows to describe even very low mass fractions of charged particle without loss of accuracy in the collision kinetics.

In addition to these features described elsewhere, the model was augmented to include anisotropic scattering in electron-H_2 elastic collisions and a more detailed description of secondary electron emission by ion impact on the electrodes, based on the methods described in refs. [6, 7] and [8], respectively. A self-adjustment technique was also devised to neutralize the current at the electrodes in order to allow comparison with experiments in a capacitively coupled reactor in the case of non-sinusoidal voltage waveforms.

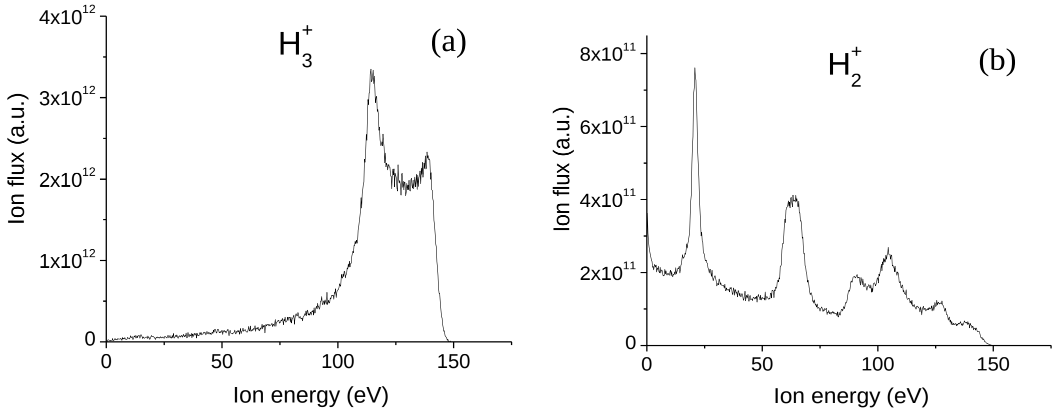

Fig. 1: Ion energy distributions of H_3^+ (a) and H_2^+ (b) ions at the electrodes of a cw RF discharge at 50 mTorr, 6 cm interelectrode gap, T_{gas} = 300 K, V_{RF} = 300 V, ν_{RF} = 13.56 MHz.

As an example of the results obtained, Fig. 1 shows the calculated ion energy distributions of H_3^+ and H_2^+ at the electrodes for the pure RF discharge case. The agreement of these results with recent experiments reported in [9] in similar conditions is very satisfactory.

The model was also employed to study the RF CCP under the influence of an externally applied DC bias voltage and TVWs. Three cases were examined: (a) continuous wave (cw) plasma with a negative DC voltage applied continuously, (b) pulsed plasma with a positive DC voltage applied synchronously during a specified time window in the afterglow, (c) cw plasma produced by TVWs. For the first case, the trends of the electron energy and the plasma concentration with the variation of the DC voltage were in agreement with the behavior observed in experiments in [16] for the first mode of the RF/DC combined discharge, i.e. the RF discharge perturbed by the DC voltage even if these measurements were performed in a different gas (nitrogen). For the case (b) the switching of the DC field produced a transient in species populations, including high lying vibrational levels of molecules, and well-defined peaks in the ion energy distributions. For the last case, work is in progress to compare the model results with the experiments performed in Palaiseau to analyze the effect of “peaks” and “valleys” voltage waveforms on the plasma and the substrate ion bombardment in a large area reactor that has close to ideal geometrical symmetry matching 1-dimensional simulations.

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References

[1] E. V. Johnson, T. Verbeke, J.-C. Vanel and J.-P. Booth, *J. Phys. D: Appl. Phys.* **43** (2010) 412001.
[2] S. Longo, I. D. Boyd, *Chem. Phys.* **238** (1998) 445.
[3] S. Longo, A. Milella, *Chem. Phys.* **274** (2001) 219.
[4] P. Diomede, M. Capitelli, and S. Longo, *Plasma Sources Sci. Technol.* **14** (2005) 459.
[5] S. Longo, P. Diomede, *Plasma Process. Polym*. **6** (2009) 370.
[6] V. Vahedi, M. Surendra, *Comp. Phys. Comm.* **87** (1995) 179.
[7] S. Longo, *Plasma Sources Sci. Technol*. **15** (2006) S181.
[8] R. Krimke, H.M. Urbassek, *J. Phys. D: Appl. Phys*. **29** (1996) 378.
[9] D. O’Connell, R. Zorat, A. R. Ellingboe, and M. M. Turner *Phys. Plasmas* **14** (2007) 103510.
[10] V. A. Lisovskiy, N. D. Kharchenko and V. D. Yegorenkov, *J. Phys. D: Appl. Phys.* **41** (2008) 125207.