Time resolved diagnostics and kinetic modeling of the ignition transient of a H_2+10%N_2 square wave modulated hollow cathode discharge

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A combined diagnostics and modeling is presented on time evolutions of the concentrations of H_2 and N_2 precursors and NH_3 produced in the plasma during the ignition transient of a low pressure H_2 /(10%)N_2 discharge, generated in a modulated dc hollow cathode reactor. Time resolved emission spectroscopy and quadrupole mass spectrometry are used for the measurements, and a zero order kinetic model is employed to explain the experimental results.

The chemistry in low pressure plasmas containing H_2 and N_2 is of interest in a variety of fields, from material processing to astrophysics. The plasma assisted synthesis of NH_3 [1,2], the inhibition of tritium co-deposits in fusion reactors [3], or the possibility of using N_2H^+ as a tracer of N_2 in astronomical observations [4] are all relevant issues related to plasmas of this kind, but the complete understanding of these media is hindered by the concurrence of a large number of interconnected gas-phase and surface reactions.

As a contribution towards the investigation of relevant processes in these plasmas, we performed a detailed diagnostics of low pressure H_2 + 10% N_2 dc hollow cathode discharges in a previous study, using electrical probes for the estimation of electron temperatures and densities, and mass spectrometry to determine the stationary concentration of ionic species and stable neutrals [5]. A zero order kinetic model was developed, based on a set of time dependent differential equations from the ignition of the discharge to the attaintment of the steady state. The analysis of the measurements with the model allowed the identification of the main physicochemical mechanisms responsible for the observed distributions of neutrals and ions, and for their dependence with discharge pressure.

With the aim of getting further insight in the plasma kinetics, modulated discharges are considered often helpful, since specific plasma processes occurring at different time scales can be evinced by studying the transient behaviour of the different species during the first stages after the ignition of the discharge [6]. In this work, this procedure is followed in H_2 + 10% N_2 plasmas by using low frequency square wave modulation and time resolved diagnostics, and comparing the results with our previously published model [5]. Visible emission spectroscopy and quadrupole mass spectrometry with electron impact ionization are used as complementary detection techniques (Fig.1).

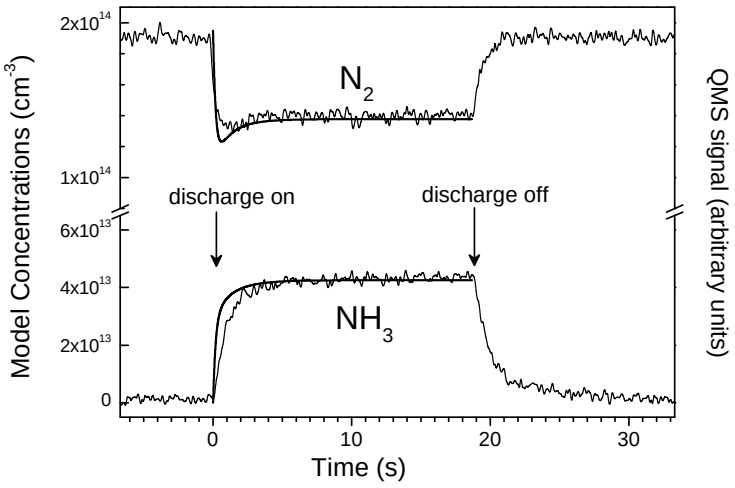

Fig. 1: Temporal evolution of N_2 and NH_3 in a H_2 + 10% N_2 modulated discharge. Thick lines: Model predictions. Narrow lines: mass spectrometric results, in arbitrary units, scaled to the predicted steady state concentrations.

The results confirm globally the validity of the previous model and, at the same time, allow to clarify some of the assumptions for which the stationary conditions were not sensitive, and to refine some of the proposed coefficients (mainly pertaining to heterogeneous processes), for which there were not formerly published data.

References

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