

## **Keep-in-Touch meeting (February 15, 2021, 2.00pm)**

### **Non-equilibrium kinetics in CO<sub>2</sub>-N<sub>2</sub> plasmas**

Chloé Fromentin

*Instituto de Plasmas e Fusão Nuclear, Instituto Superior Técnico, Universidade de Lisboa, Portugal*

Carbon dioxide is among the main products of anthropogenic activity. It is a greenhouse gas, with impact on global warming and climate change. A promising option is to create artificial solar fuels using CO<sub>2</sub> as raw material, using intermittently available renewable energy surpluses, which can easily be deployed within the present infrastructure for conventional fossil fuels. The critical step is CO<sub>2</sub> dissociation. The strong non-equilibrium conditions provided by non-thermal plasmas offer the opportunity to increase the energy efficiencies via preferential excitation of molecular vibrational modes and reducing the energy cost of CO<sub>2</sub> dissociation. A potential way to favor the asymmetric vibration mode is the addition of molecular nitrogen, as it is known in the context of CO<sub>2</sub> laser technologies. Indeed, the first excited level of N<sub>2</sub> has an energy very close to the first asymmetric level of CO<sub>2</sub>, allowing nitrogen to exchange easily vibration quanta with the asymmetric mode of CO<sub>2</sub>. At low pressure and low temperature, the vibration-vibration (V-V) exchanges are faster than the relaxation (V-T) reactions and, therefore, nitrogen molecules can transfer vibration quanta to the v<sub>3</sub> mode of CO<sub>2</sub> before thermalization occurs. In such plasmas, during a time scale of a few milliseconds, the vibrational distribution function of the molecules shows a higher tail in the highly excited levels than in a classic Boltzmann distribution. The presence of these highly vibrating molecules makes non-equilibrium or non-thermal plasmas suitable environments to reach an efficient conversion. In addition, nitrogen is very likely to be present in CO<sub>2</sub> streams coming from industrial emissions, so that understanding its influence on the overall CO<sub>2</sub> decomposition kinetics is mandatory.

The aim of this work is precisely to investigate the kinetics of CO<sub>2</sub>-N<sub>2</sub> plasmas. A zero-dimensional kinetic model for glow-type discharges in CO<sub>2</sub>, CO<sub>2</sub>-O<sub>2</sub> and CO<sub>2</sub>-N<sub>2</sub> is being developed. The model couples the electron kinetics, described by the electron Boltzmann equation, to a system of rate balance equations describing the creation and loss of vibrational and electronic excited states, different radicals and ions. The simulation results are compared with recent experimental data obtained in low-pressure DC discharges. This joint theoretical and experimental effort will allow us to better understand the underlying mechanisms of CO<sub>2</sub> dissociation and will contribute to further refine the existing kinetic schemes and thus to better control and optimize CO<sub>2</sub> conversion.

In this seminar, a general overview of the project and of the global approach adopted is presented. The LoKI modelling tool and its specificities in the context of the current investigation are introduced. The focus is on the refinement of the  $O_2$  chemistry set, with a comparison between the simulation results and experimental data points. The  $CO_2$  chemistry is also discussed, along with the impact of the presence of  $O_2$  on the  $CO_2$  dissociation and the importance of the electronically excited state  $CO(a^3\Pi_r)$ . The implementation of the  $CO_2$  vibrations to the model are briefly summarized. Finally, recent results obtained at the Laboratoire de Physique des plasmas (LPP) using Fourier Transform InfraRed (FTIR) spectroscopy on  $CO_2$ - $N_2$  DC glow discharges are presented and discussed.