COUPLED KINETICS IN CO₂-N₂ PLASMAS

C. FROMENTIN¹ and T. C. DIAS¹ and T. SILVA¹ and V. GUERRA¹ and E. BARATTE² and O. GUAITELLA²

¹Instituto de Plasmas e Fusão Nuclear, Instituto Superior Técnico, Universidade de Lisboa, 1049-001 Lisboa, Portugal

> ²Laboratoire de Physique des Plasmas, Ecole Polytechnique-CNRS 91128 Palaiseau, France

Abstract. CO_2 plasmas are interesting for a wide variety of applications, including CO_2 reforming for the production of solar fuels and added-value chemicals, polymer deposition, spacecraft reentry and in-situ resource utilization on Mars (Pietanza et al 2021). Control and optimization of the different applications requires a deep understanding of the energy transfer pathways and of the coupling of the different kinetics at play, such as the electron, vibrational, chemical and surface kinetics. Of major interest are the processes of conversion of CO_2 into CO and back reactions reconverting CO back into CO_2 .

Experiments are performed in DC glow discharges operating at pressures around 1 Torr and discharge currents of the order of tenths of mA, measuring by Fourier Transform Infra-Red spectroscopy (FTIR) the vibrational temperatures of CO and of the three vibration modes of CO₂, as well as the dissociation fraction, $[CO]/([CO_2]+[CO])$. To interpret and analyze the experiments, a self-consistent kinetic model is developed, that solves the electron Boltzmann equation in the usual 2-term expansion in Legendre polynomials (Tejero-del-Caz et al 2019), coupled with a system of rate-balance equations describing the creation and loss of the most important neutral and charged heavy-particles.

It is verified that the dissociation fraction increases upon addition of N_2 into a CO_2 discharge, in line with previous measurements by Grofulović et al 2019 and Terraz et al 2019. The enhanced dissociation fraction is the outcome of a combination of different effects, such as the modifications in the Electron Energy Distribution Function (EEDF) induced by a different mixture of gases, with associated modifications in the electron impact dissociation rate coefficients, both from ground-state and from vibrationally excited CO_2 , and the dilution of CO_2 in N_2 , associated with a smaller influence of back reaction mechanisms.

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