Keep-in-Touch meeting
(December 12, 2022)

Influence of N_2 on the CO_2 dissociation fraction and vibrational kinetics in low temperature CO_2-N_2 plasmas.

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Fighting climate change is one of the greatest challenge Humanity has to tackle in the twenty-first century and to achieve the goal of zero net emissions by 2050 the development of new technologies capable of reducing anthropogenic CO_2 emissions is required [1]. The conversion of CO_2 into chemicals and energy products via the Carbon Capture and Utilisation (CCU) method is a promising way to not only decrease the CO_2 emissions, but also generate more economic value and reduce the dependence of fossil fuels while closing the carbon cycle [2].

A very encouraging approach is the use of non-thermal plasmas (NTP) for CO_2 conversion [3-6] benefiting from the non-equilibrium conditions in these plasmas where high energy electrons coexist with cold ions and neutrals. NTPs are favorable to achieve efficient dissociation of CO_2, as they offer a selective activation of certain chemical processes with lower energy when compared to conventional methods [7].

Several reasons can motivate the study of CO_2-N_2 low temperature plasmas. On the one hand, N_2 is the main impurity in most industrial flue gas [8], so the influence it may have on the kinetics and chemistry of a plasma must be investigated. On the other hand, the admixture of N_2 has a beneficial impact on CO_2 decomposition [9-13]. Several reasons can be assigned to this effect: the promotion of CO_2 vibrational excitation, the modification of the EEDF, the presence of metastable states of N_2 and the dilution with N_2 limiting the influence of back reaction mechanisms producing CO_2 from CO.

The aim of this work is precisely to investigate the kinetics of CO_2-N_2 plasmas. A zero-dimensional kinetic model for glow-type discharges in CO_2-N_2 is being developed using LoKI (LisbOn Kinetics) which solves a Boltzmann-chemistry global model [14, 15]. 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 and different radicals and ions.

The simulations results are compared with experimental data measured in a low-pressure DC glow discharge. Indeed, a set of measurements providing the gas temperature, vibrational temperatures of CO and the various modes of CO_2, reduced field E/N, and densities of $O(^3P)$, $CO(X^1\Sigma^+)$, $CO_2(X^1\Sigma^+_g)$ and $NO(X^2\Pi_r)$ was recently obtained at the Laboratoire de Physique des plasmas (LPP) using in situ time-resolved step-scan Fourier transform infrared (FTIR) spectroscopy as described in detail in [16] or by actinometry [17].

This joint theoretical and experimental effort allows us to better understand the underlying mechanisms of CO_2 dissociation and will contribute to further refine the existing kinetic schemes. In particular, the influence of N_2 on the CO_2 dissociation is discussed with a special focus on the electronically excited states of N_2. Finally, the implementation and validation of the CO_2, CO and N_2 vibrational kinetics are also summarized.

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