

Kinetic mechanisms in CO₂-N₂ plasmas

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ABSTRACT

This contribution reports the comparison of simulation results from a 0D self-consistent kinetic model with recent experimental data obtained in low-pressure DC discharges in CO₂-N₂. This comparison allows the development of a new reaction mechanism (i.e., a set of reactions and rate coefficients validated against benchmark experiments) for CO₂-N₂ plasmas. The system of election is a DC glow discharge, operating at pressures in the range $p=0.1-10$ Torr and discharge currents $I=10-50$ mA, in a Pyrex tube of radius $R=1$ cm, which is stable, axially homogenous, and easily accessible to a variety of diagnostics. The set of measurements provides the gas temperature, vibrational temperatures of CO and the various modes of CO₂, reduced field E/N , and densities of O(³P), CO(X¹Σ⁺) and CO₂(X¹Σ⁺_g). The simulation results are obtained with the LoKI (LisbOn Kinetics) [1] simulation tool solving a Boltzmann-chemistry global model.

Investigating the impact of CO and N₂ on the vibrational temperature of the different CO₂ vibrational modes and on the overall conversion is relevant as they can promote CO₂ vibrational excitation. Besides, CO is a product of the dissociation and is therefore always present in the discharge and N₂ can be found as an impurity in industrial CO₂ emissions. It has been shown that the admixture of N₂ has a beneficial impact on CO₂ decomposition [2,3]. Several reasons can be assigned to it, one of them being the near-resonant transfer of vibration quanta from the first vibrational level of N₂ to the asymmetric mode of CO₂ (ν_3) and the fact that vibrationally excited CO₂ can undergo molecular dissociation through the so-called ladder climbing mechanism or by electron impact stepwise processes. Similarly, CO molecules can transfer energy to the ν_3 vibration of CO₂ because the energy difference between the first vibrational level of CO and the first asymmetric stretch vibrational level of CO₂ is only 25meV, which is smaller than the average kinetic energy [4]. However, depending on the CO concentration, the presence of CO can either enhance the dissociation of CO₂ or stimulate the reconversion back to CO₂ through the electronic excited state CO(a³Π) which can have an ambivalent role depending on the CO density [5, 6]. These mechanisms will be discussed in the detail at the conference. Understanding the impact of the different processes on the overall kinetics, along with the validation against experimental data, will contribute to further develop the existing models [3, 7, 8] and to better control and enhance CO₂ conversion.

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