

Kinetic mechanisms in CO₂-N₂ plasmas

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Abstract:

Investigating the impact of N₂ on the overall CO₂ conversion is relevant as N₂ can be present as an impurity in industrial CO₂ emission and can be used to promote CO₂ vibrational excitation and further molecular dissociation through the so-called ladder climbing mechanism. 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 homogeneous, and easily accessible to a variety of diagnostics. The set of measurements provides the gas temperature, vibrational temperatures of CO₂, reduced field E/N , and densities of O(³P), NO, NO₂, CO(X¹Σ⁺) and CO₂(X¹Σ⁺_g). Our simulation results are obtained with the LoKI (LisbOn Kinetics) simulation tool [1] solving a Boltzmann-chemistry 0D self-consistent kinetic model. The comparison of the model predictions with the experimental data 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 and provides physical insights into the main mechanisms occurring in these plasmas.

It is shown that the admixture of N₂ has a beneficial impact on CO₂ decomposition, as also pointed out in [2,3]. Several reasons can be assigned to it, one of them being the transfer of vibration quanta from the first vibrational level of N₂ to the asymmetric mode of CO₂ and the fact that vibrationally excited CO₂ can undergo molecular dissociation through the so-called ladder climbing mechanism or by electron impact step-wise processes. Moreover, the dilution with N₂ can also limit the influence of back reaction mechanisms producing back CO₂ from CO. 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 [3-5] and to better control and enhance plasma-assisted CO₂ conversion.

Keywords: glow discharge, CO₂ decomposition, vibrational excitation, 0D model.

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