Kinetic mechanisms in CO_2-N_2 plasmas

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Abstract:
Investigating the impact of N_2 on the overall CO_2 conversion is relevant as N_2 can be present as an impurity in industrial CO_2 emission and can be used to promote CO_2 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 homogenous, and easily accessible to a variety of diagnostics. The set of measurements provides the gas temperature, vibrational temperatures of CO_2, reduced field E/N, and densities of $O(^3P)$, NO, NO_2, $CO(X^1\Sigma^+)$ and $CO_2(X^1\Sigma^+{}_g)$. 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 CO_2-N_2 plasmas and provides physical insights into the main mechanisms occurring in these plasmas.
It is shown that the admixture of N_2 has a beneficial impact on CO_2 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 N_2 to the asymmetric mode of CO_2 and the fact that vibrationally excited CO_2 can undergo molecular dissociation through the so-called ladder climbing mechanism or by electron impact stepwise processes. Moreover, the dilution with N_2 can also limit the influence of back reaction mechanisms producing back CO_2 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 CO_2 conversion.

Keywords: glow discharge, CO_2 decomposition, vibrational excitation, 0D model.

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