Kinetic mechanisms in vibrationally excited CO_2 plasmas

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Recent measurements of the gas temperature, vibrational temperatures of CO_2 and CO, E/N, $O(^3P)$, $CO(X^1\Sigma^+)$ and $CO_2(X^1\Sigma^+_g)$ and $O(^3P)$ loss frequencies performed in low-pressure DC discharges provide an ideal set of constraints for validating numerical simulations from a 0D self-consistent kinetic model. The dominant elementary processes with impact on the overall coupled kinetics are highlighted with a special focus on the vibrational kinetics of CO_2 and CO.

Investigating the impact of CO on the vibrational temperature of the different CO_2 vibrational modes and on the plasma chemistry is relevant as it is a product of the dissociation. It will therefore be present in the discharge and vibrationally energy transfers between CO and CO_2 are known to be effective [1]. Hence, CO vibrations can 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=1-5 Torr and discharge currents I=20 and 40 mA, in a Pyrex tube of radius R=1 cm, which is stable, axially homogenous, and easily accessible to a variety of diagnostics. A set of experimental data including, the gas temperature, vibrational temperatures of CO_2 and CO, reduced field E/N, densities of $O(^3P)$, $CO(X^1\Sigma^+)$ and $CO_2(X^1\Sigma^+_g)$ and $O(^3P)$ loss frequencies, was recently measured and provided by the Laboratoire de Physique des Plasmas. Our simulation results are obtained with the LoKI (LisbOn Kinetics) simulation tool [2] solving a Boltzmann-chemistry 0D self-consistent kinetic model. The comparison of the model predictions with the experimental data will contribute to further develop the existing models [3-5] and to better control and enhance plasma-assisted CO_2 conversion.

The transfers between vibrationally excited CO and the asymmetric stretching mode (v_3) of CO_2 are very efficient and can promote the ladder climbing mechanism along this mode, with a potential positive effect on CO_2 dissociation. These CO molecules can transfer energy to the v_3 vibration because the energy difference between the first vibrational level of CO and the first asymmetric stretch vibrational level of CO_2 is only 25meV which is smaller than the average kinetic energy [1]. Moreover, due to the resonance effect of short-lived negative ions CO^-, the observed cross sections of electron impact excitation of molecular vibrations of CO are rather large [6]. The process of vibrational transfer between CO and CO_2 and its effect on the overall kinetics will be discussed during the conference.

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