DC discharges on CO_2/Ar mixtures: modelling and experiment

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The effects of Ar addition on the dissociation of CO_2 are studied in low pressure glow discharges through a modelling and experimental effort. We study the influence of pressure, flow, current and Ar content in the plasma in the dissociation of CO_2. Lower pressure and higher Ar content yield higher dissociation ($\approx$ 80% at 40 mA).

Plasma technology is today seen as a potential route for reduction of CO_2 emissions. However, the elemental kinetic processes occurring in a CO_2 plasma are not yet completely understood and there is still work to be done towards the goal of high conversion and energy efficient processes. In this work we show the benefits of the addition of argon on CO_2 dissociation through modelling and experiment. Argon is known to allow higher energy efficiency and conversion, lowering the breakdown voltage of the plasma, bringing new dissociation pathways to the table and modifying the electron distribution function. The modelling effort, validated by experimental results, helps to better understand the relevant kinetic processes that take place in a CO_2/Ar plasma, in particular the ones that might be related to CO_2 dissociation.

The modelling is based on the efforts of N-PRiME group, namelly using the CO_2 kinetic models of PREMiERE project [1] and the LisbOn KInetics (LoKI) simulation tool [2]. The latter self-consistently couples the solution of the homogeneous two-term Boltzmann equation for electrons with the solution to the system of 0D rate balance equations for the other species in the plasma. In figure 1 we show some results obtained for the electron kinetics considering different CO_2/Ar mixtures.

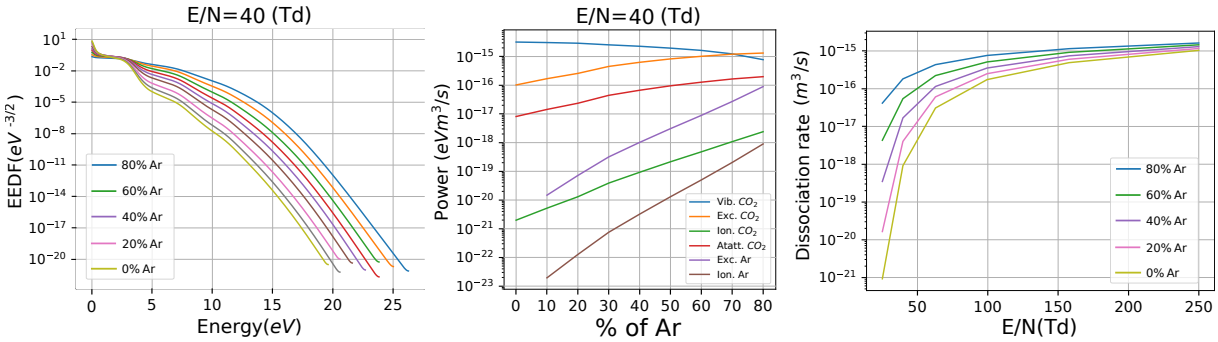

Fig. 1: Results obtained with LoKI for the electron kinetics of a CO_2/Ar mixture

Ar addition modifies the tail of the distribution fuction, with a larger electron population for higher Ar concentrations. Also, due to the high energy threshold for excitation and ionization of argon, these electrons will be preferentially interacting with CO_2 molecules at the typical values of reduced electric field achieved in low presssure discharges. These effects influence the kinetics of CO_2, with higher Ar concentrations yielding higher electron impact dissociation rates of the molecule.

The experiments were performed in a cylindrical plasma reactor made of Pyrex with 1 cm inner radius and 23 cm length. The working conditions vary from 10 to 40 mA applied current, 1 to 10 sccm of total gas flow, 1 to 5 Torr pressure and percentages of argon from 0% to 75%. IR absorption measurements

are preformed downstream of the reactor. We use an FTIR spectometer and analyse the spectra using the technique described in [3]. In figure 2 we show some of the values obtained for the conversion factor:

$$\alpha = \frac{[CO]}{[CO_2] + [CO]} \tag{1}$$

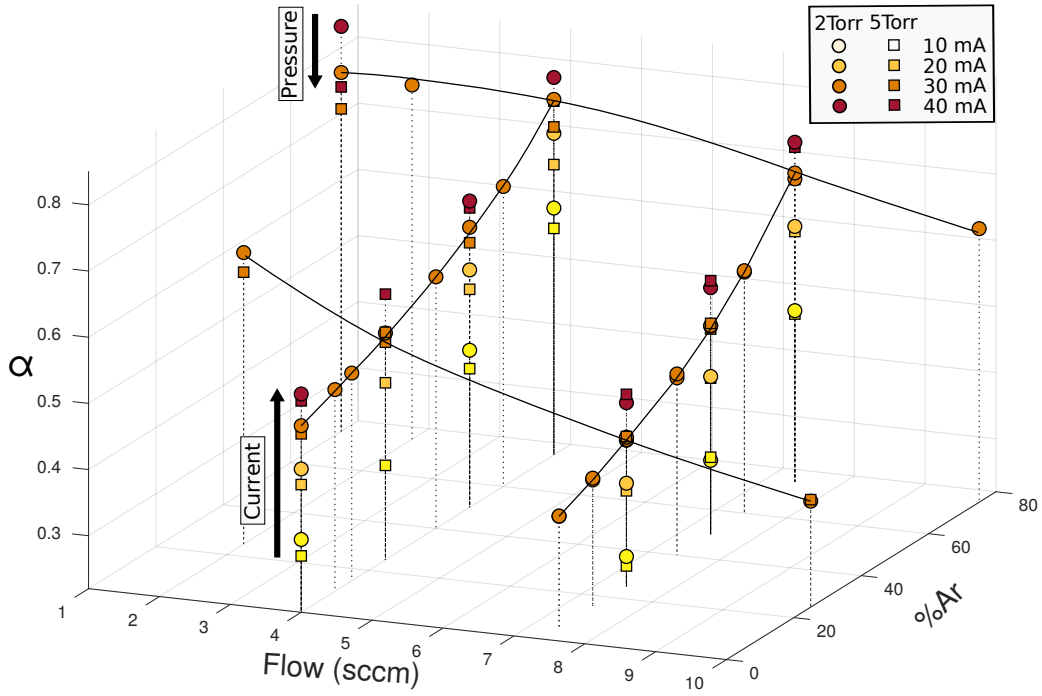

Fig. 2: Experimental results for the CO_2 conversion factor varying applied current, flow, pressure and Ar content. The solid lines link the points obtained at 2 Torr and 30 mA.

Conversions as high as 80% were achieved for the lowest flow and pressure and highest current and precentage of Ar used. The addition of Ar seems to allow a faster establishment of a steady state inside the reactor, since for 75% Ar there is a stabilization of the conversion factor for flows lower than 4 sccm i.e. residence times above ≈ 0.7 s. Additional measurements are performed *in situ* to get vibrational temperatures' evolution and laser absorption techniques are also used to determine the metastable Ar density to study the quenching of this atomic state by CO_2 and its effect on CO_2 dissociation.

Acknowledgments

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