DC discharges on CO_2/Ar mixtures: modeling and experiment

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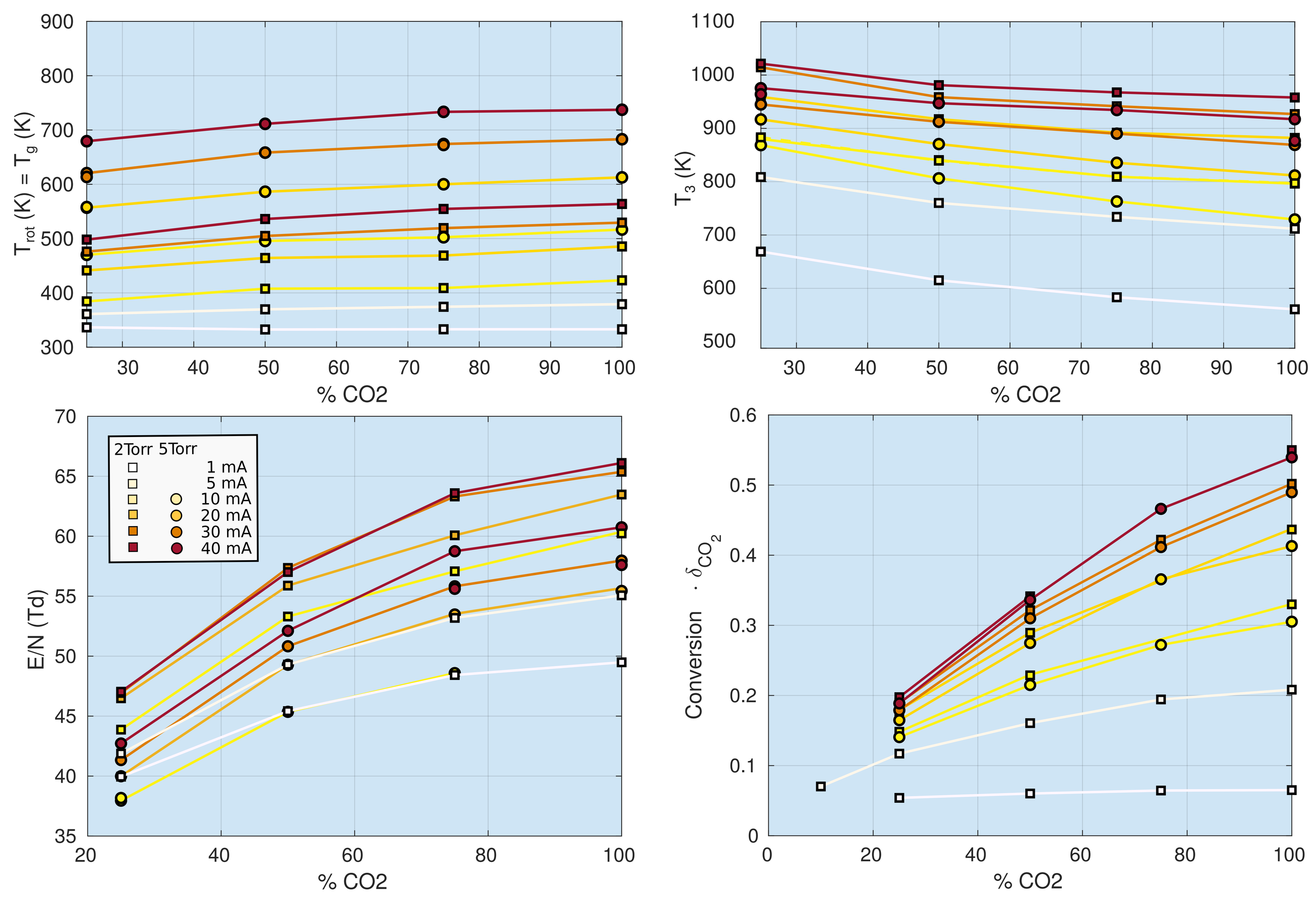
Introduction

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 modeling and experiment**.

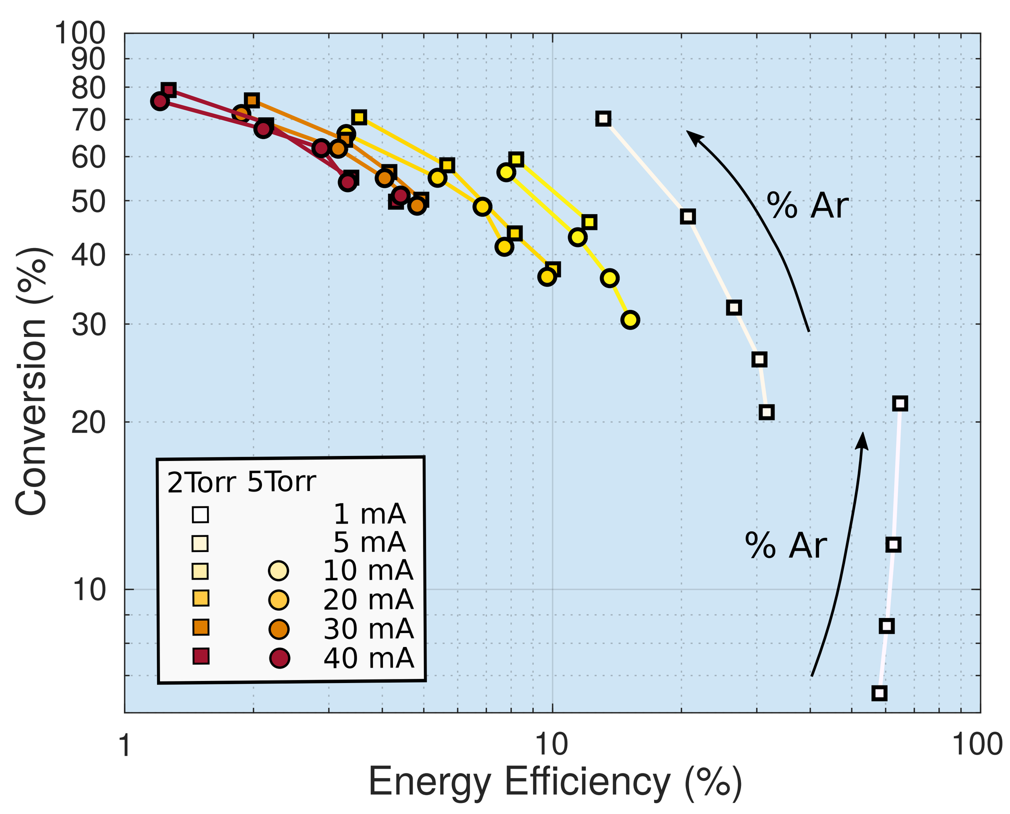
Experimental results

- **Glow discharge**.
- **Cylindrical reactor** (R= 1cm, L =23cm). Flow = 4 sccm.
- **FTIR** spectroscopy. Spectra analysed using the technique described in [3].
- **Rotational** and **vibrational temperatures** of CO_2 in-situ.
- **Conversions of CO_2** into CO measured downstream of the reactor.



- **Gas temperature** drops with Ar content for higher currents.
- **Vibrational temperature of the assymetric stretch mode increases** with Ar. Effect more noticeable at low currents.
- **Reduced electric field** drops significantly with Ar addition.
- **Conversion increases** with increasing Ar content.
- Product of conversion and the initial fraction of $CO_2 \propto$ **net CO produced** drops for all currents. Effect is less noticeable at low currents.

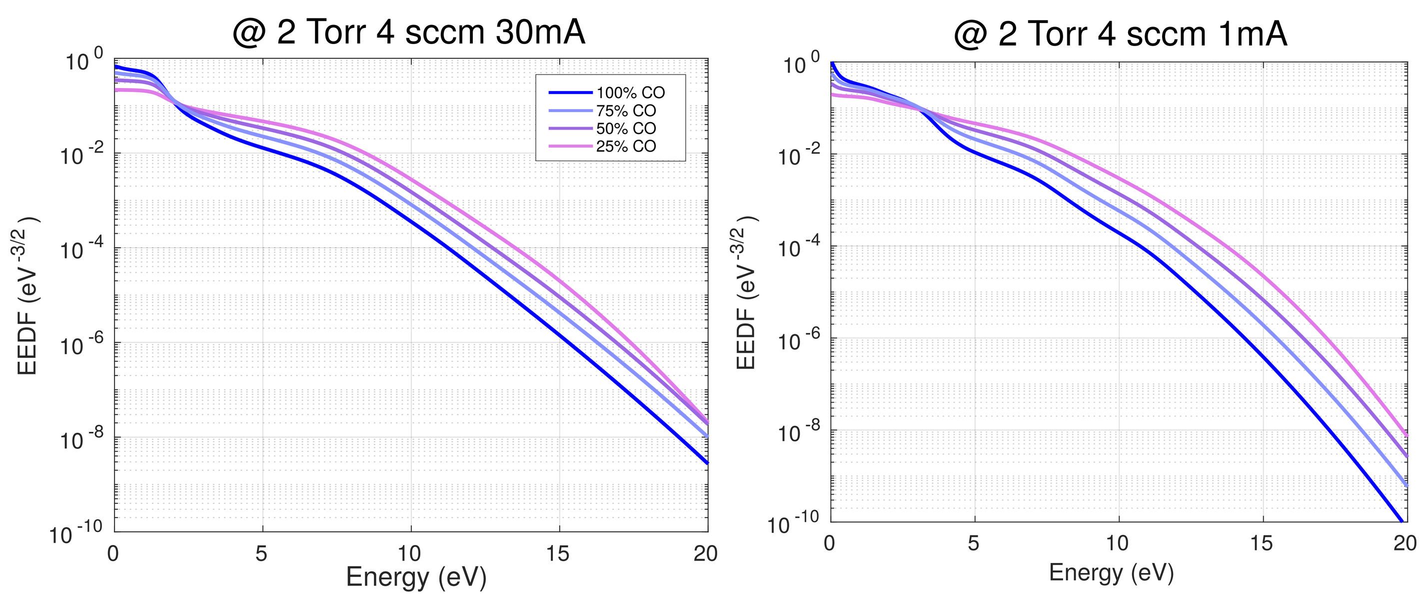
Conversion and energy efficiency



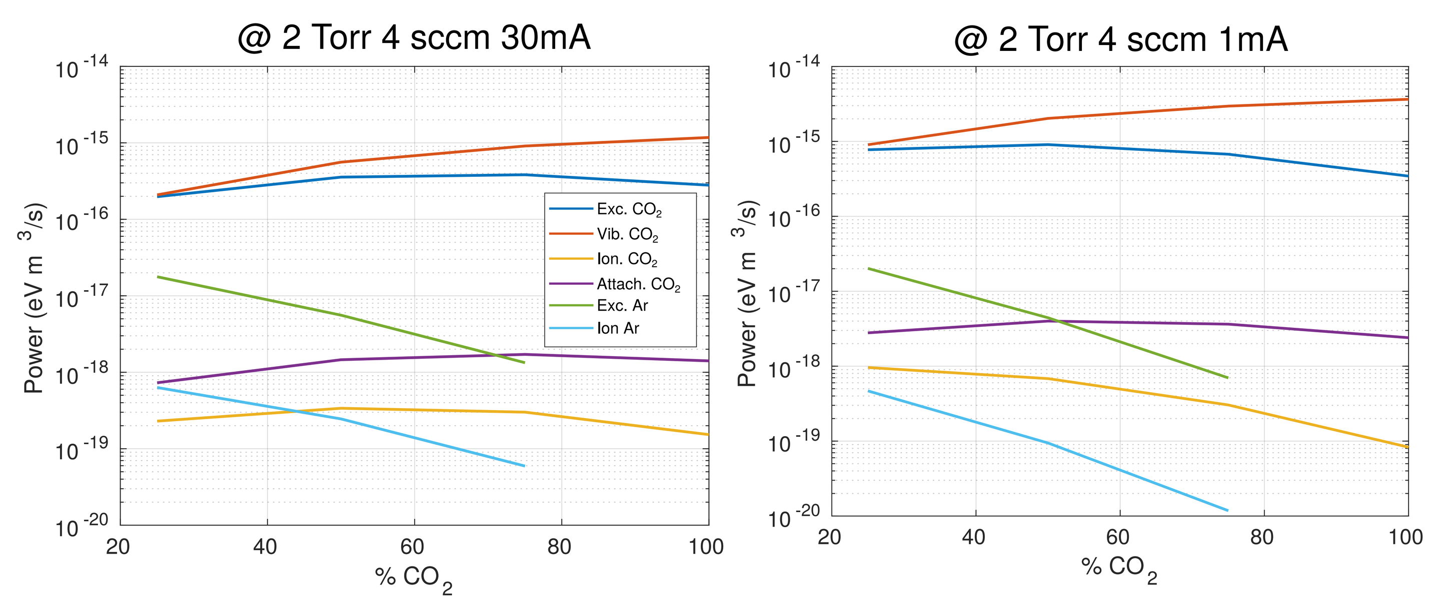
- Compromise between **conversion** and **energy efficiency**.
- The addition of Ar **improves conversion** in all working conditions.
- This comes at the cost of **lower energy efficiency**. The higher the current the steeper the drop in efficiency with Ar addition.
- At **1 mA** there is a slight **improvement in efficiency** due to the faster growth of conversion with Ar added.

Modeling results

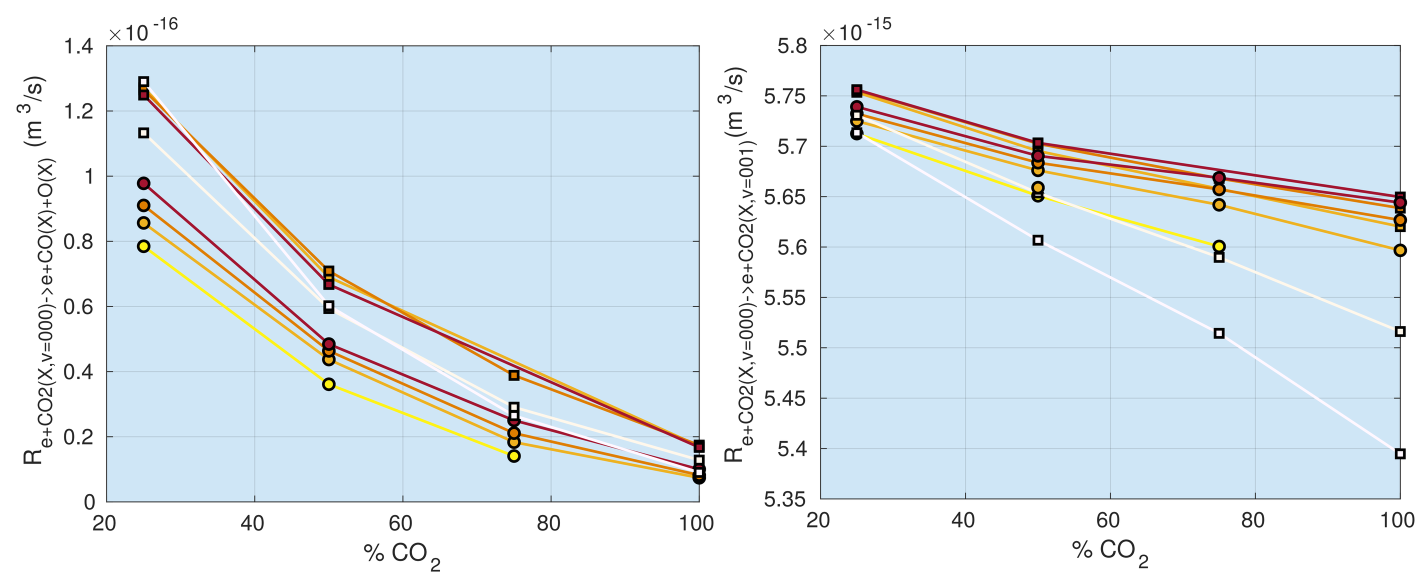
- Modeling based on the efforts of **N-PRiME** group:
  - **-** CO_2 kinetic models of **PREMiERE project** [1];
  - **-** LisbOn KInetics (**LoKI**) simulation tool [2].
- LoKI solves the **homogeneous Boltzmann equation for electrons** in the plasma → Information about **electron kinetics**.



- **Electron distribution function** significantly modified with Ar addition.
- **More populated tail** of the EEDF → More electrons able to dissociate CO_2.
- Routes of dissociation:
  - **- direct electron impact** → 7 eV/mol;
  - **- vibrational ladder climbing** (e^--impact vibrational excitation + collisions between two vibrationally excited CO_2 molecules) → 5 eV/mol;



- For all working conditions the **power spent by electrons on** excitation and ionization of **Ar is negligible** when compared with the power spent on CO_2.



- **Rate of dissociation via electron impact** rises with Ar addition.
  - → direct e^--impact → predominant route of dissociation at **high currents**.
- At **low currents** this rise cannot explain the improvement of experimental values of conversion, but:
  - → **Rise of T_3** + **rise of** the **rate of excitation of v_3** =1 from the g. s.
    - → **Vibrational ladder climbing more important at low currents**.

Conclusions

In a **glow discharge** at pressures of the order of 1 Torr **the addition of Ar to CO_2 greatly improves dissociation without largely deteriorating energy efficiency at low enough input power**. These results can be understood analysing the effect of Ar addition in the electron kinetics. Both the model and the experiment sugest that **at low currents the addition of Ar promotes more dissociaton via excitation of the assymetric stretch mode of CO_2**, although more research is needed to make further conclusions.

References

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Acknowledgements

This work has been done within the LABEX PLAS@PAR project, and received the financial state aid managed by the ANR, as part of the programme "Investissements d'avenir" ANR-11-IDEX-0004-02. This work was partially supported by the Portuguese FCT, under Projects UID/FIS/50010/2013, PTDC/FIS-PLA/1420/2014 (PREMiERE) and PTDC/FISPLA/1243/2014 (KIT-PLASMEBA).

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Europhysics Sectional Conference on the Atomic and Molecular Physics of Ionized Gases Glasgow July 17-21 2018