Introduction

Plasma technology is today seen as a potential route for reduction of CO₂ emissions. However, the elemental kinetic processes occurring in a CO₂ 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 CO₂ 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 CO₂ in-situ.
• Conversions of CO₂ into CO measured downstream of the reactor.

Conversion and energy efficiency

• 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 CO₂. The higher the current the steeper the drop in efficiency with Ar addition.
• At 1 mA there is a slight improvement in energy efficiency due to the faster growth of conversion with Ar added.

Modeling results

• Modeling based on the efforts of N-PRIME group:
  - CO₂ 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 CO₂.
• Routes of dissociation:
  - direct electron impact → 7 eV/mol;
  - vibrational ladder climbing (e-impact vibrational excitation + collisions between two vibrationally excited CO₂ 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 CO₂.

• 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 Tₐ + rise of the rate of excitation of v₃ =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 CO₂ 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 suggest that at low currents the addition of Ar promotes more dissociation via excitation of the assymetric stretch mode of CO₂, although more research is needed to make further conclusions.

References


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