A computational chemical kinetics study of a supersonic microwave plasma for CO_2 dissociation

V. Vermeiren, A. Berthelot, A. Bogaerts

Research group PLASMANT, Department of Chemistry, University of Antwerp, Belgium

Past experiments have reported[1] a record high energy efficiency (up to 90%) for plasma based CO_2 dissociation by means of a supersonic microwave discharge. So far, no detailed description of the chemical processes, occurring in such reactor, has been reported. In this work, we study these processes by means of a chemical kinetics model, elucidating the crucial role of the asymmetric vibrational modes of CO_2. This model uses flow values, calculated by the commercial software package COMSOL, as input parameters for the chemical kinetics model. The study is performed over a range of specific energy input values, by varying both the flow rate and the applied microwave power.

1. Introduction

Microwave sustained discharges have gained increasing interest as a possible pathway in the reduction of anthropogenic CO_2 emission[2]. Their non-equilibrium nature allows for a very efficient excitation of the asymmetric vibrational modes, leading to dissociation[3,4].

Very promising experimental results have been reported with a supersonic microwave discharge, in which the flow passes through a Laval nozzle[1]. In this setup, the flow passes through a convergent-divergent nozzle, which creates a desired pressure drop in addition to a supersonic flow velocity.

So far, no computational chemical kinetics study has been reported, explaining the underlying chemistry for such type of discharge, using a pure CO_2 gas.

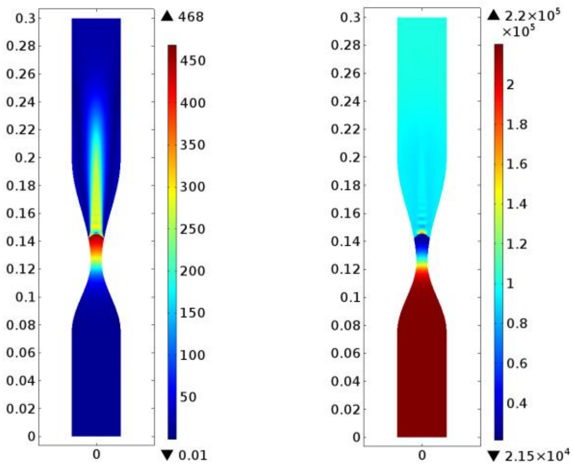

Figure 1: Calculated velocity magnitude [m/s] (left) and absolute pressure [Pa] (right) profiles for a total input pressure of 2 atm.

2. Methodology

This computational study is performed by a 0D chemical kinetics model, using the code ZDPlaskin[5]. The model solves various balance equations for the different plasma species, providing the evolution of the species densities through the reactor. The Electron Energy Distribution Function is calculated at every computational point by a built-in Boltzman solver, called BOLSIG+[6].

The chemistry set which is used in this work is based on the work of Kozák et al[3,4]. It takes into account all the CO_2 asymmetric mode levels up to the dissociation energy of 5.5eV, together with 4 effective low-lying symmetric stretching and bending mode levels.

3. Results

In figure 1, we show the calculated values for velocity and pressure when applying a total pressure of 2 atm on the inlet. The results show the characteristic pressure drop after the nozzle, followed by a shockwave, as is also experimentally observed for a similar setup[1,4].

4. References

[1] Asisov R. I., Vakar A.V., Jivotov V.K., Proc. of the USSR Academy of Sciences, **271** (1983).

[2] Fridman A., Plasma Chemistry, Cambridge University Press (2012).

[3] T. Kozák and A. Bogaerts, Plasma Sources Sci. Technol. **23**, 4 (2014).

[4] T. Kozák, A. Bogaerts, Plasma Sources Sci. Technol. **24**, 1 (2015).

[5] Pancheshnyi S., Eismann B., Hagelaar G. J. M. and Pitchford L. C. Computer Code ZDPlasKin www.zdplaskin.laplace.univ-tlse.fr (2008).

[6] Hagelaar G.J.M. and Pitchford L.C., Plasma Sources Sci. Technol. **14**, 722 (2005).