Zero-dimensional simulations of RF and ns-pulsed He/CO_2 atmospheric pressure plasma jets

Y. He[1] , F. Verdaguer[1] , R.P. Brinkmann[1] , A.R. Gibson[2] and E. Kemaneci[1]

[1] *Institute of Theoretical Electrical Engineering, Ruhr-University Bochum, Bochum, Germany*
[2] *Research Group for Biomedical Plasma Technology, Ruhr-University Bochum, Bochum, Germany*

Abstract: A zero-dimensional model is developed to study the Radio-Frequency (RF) and nanosecond (ns)-pulsed He/CO_2 plasma jets. The model is coupled with a two-term Boltzmann equation to self-consistently calculate the electron kinetics. 78 species (e.g. CO(v=1-10) and CO_2(v=001-0021)) and 3900 reactions are considered to describe the He/CO_2 chemical model. A good agreement between the measured and simulated conversion efficiency and energy efficiency is obtained.

Keywords: zero-dimensional model, plasma jet, He/CO_2 chemistry and CO_2 conversion.

1 Motivation

Environmental issues related to CO_2 emissions have gained increasing attention in recent years. Plasma-assisted CO_2 conversion is of particular interest due to its efficient excitation of the gas molecules to an energy level near the dissociation limit, at ambient temperature and pressure [**1**]. A large variety of experiments and simulations of CO_2 containing plasmas have been conducted in the literature, a summary is given in [**2**]. These include a number of works in plasma jets, where CO_2 is diluted in noble gases such as He or Ar. While these sources are not necessarily suited for large scale applications, they are of interest for developing a basic understanding of CO_2-related plasma chemical kinetics. For instance, measurements of conversion and energy efficiency in a He/CO_2 Radio-Frequency (RF) atmospheric pressure plasma jet were conducted by Stewig *et al* [**3**], and various species densities as a function of admixture ratio in a similar jet device were measured by Willems *et al* [**4**]. Several time-resolved population measurements of the vibrational levels in a Direct Current (DC) nanosecond (ns)-pulsed He/CO_2 plasma jet at near atmospheric pressure were reported by Du *et al* [**5**]. One of the open problems still to be addressed is a validation between the measurement data and simulation results in a broad range of operation conditions [**2**].

The main goal of this study is to obtain a relative reliable set of chemical kinetics describing the He/CO_2 plasma under extensive validations, and to further carry out detailed investigations, including the formation of CO as a product of interest for the chemical industry.

2 Setup

In this study, RF [**3,4**] and ns-pulsed [**5**] plasma jets are simulated, and their micro-scaled capillary rectangular chamber structures are sketched in **Fig. 1**(a) and (b), respectively. The figures are not drawn to scale. 30 sccm, 250 sccm and 1400 sccm He background gas are fed into the jet chambers diluted with a few percentage of CO_2 admixture near or at atmospheric pressure (see details in **Fig. 1**).

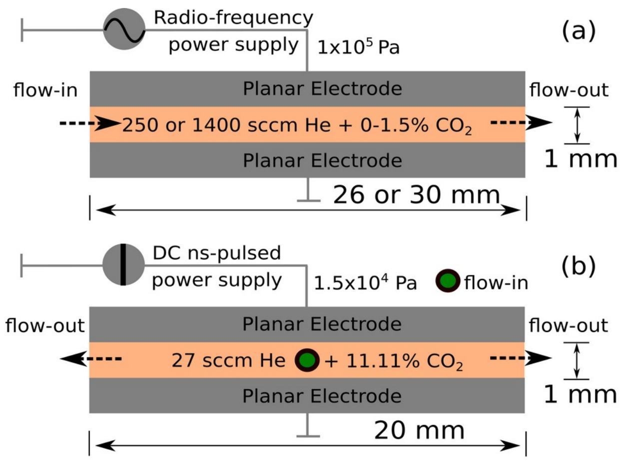

Fig. 1. Sketch of plasma jet chambers

3 Model

The particle balance equations and an electron energy balance equation are implemented by MATLAB to develop a zero-dimensional model calculating the plasma properties: i.e. the species density and the mean electron energy. These equations are defined as [**6**]:

$$\frac{dN_i}{dt} = S_i^V + S_i^W , \quad (1)$$

$$\frac{d}{dt}\left(\frac{3}{2} N_e T_e\right) = P^V + P^W , \quad (2)$$

where N_i is the density of species *i*, *e* represents electron, *T* denotes the temperature, *S* and *P* denote the gain/loss mechanisms of the species density and the power, respectively. The "*V*" and "*W*" denote the plasma volume and the chamber wall, respectively. Both the volume-averaged formalism and the plug-flow formalism of the zero-dimensional modelling approaches are implemented. The former provides volume-averaged plasma property values of the whole chamber region, which typically provides either the values in a series of RF operation conditions or the values in a time resolution during one pulse-modulation power. The plug-flow formalism results in one-dimensional calculation results along the gas flow direction, which is typically prefered to be used for a spatial resolution in a single operation condition.

The model is coupled with a Boltzmann solver, the open source MATLAB code LisbOn KInetics Boltzmann (LoKI-B) published by Tejero-del-Caz *et al* [**7**]. A self-consistent calculation of the electron kinetics is obtained by calling LoKI-B to calculate the non-Maxwellian electron energy distribution functions and further update the look-up tables of rate coefficients of electron impact reactions (see more details in [**6**]).

4 Chemical kinetics

A list of the considered 78 species in the He/CO_2 model is reported in **Table 1**, and around 3900 reactions are included in the chemical model. The He/CO_2 chemical kinetics in this study are mainly adopted from those in Koelman *et al* [**8**] and Kozak *et al* [**9**]. Additionally, the chemical sets of He and He/O_2 reactions are adopted from those in our previous study [**1**], where the corresponding simulation results are validated against the diverse species densities measured by various groups.

Table 1. Species included in the He/CO_2 chemical model.

He, He^*, He_2^*, He^+, He_2^+,
C, C_2, O, O_2, O_3, CO, CO_2, C_2O,
O^*, $O_2(e=1)$, $CO(e=1-4)$, $CO_2(e=1,2)$,
C^+, C_2^+, O^+, O_2^+, O_4^+, CO^+, CO_2^+, CO_4^+, $C_2O_2^+$, $C_2O_3^+$, $C_2O_4^+$,
O^-, O_2^-, O_3^-, O_4^-, CO_2^-, CO_3^-, CO_4^-,
$O_2(v=1-3)$, $CO(v=1-10)$, $CO_2(v=a,b,c,d)$, $CO_2(v=1-21)$, e

5 Results & Conclusion

A validation of the conversion and energy efficiency of the He/CO_2 plasma jets is conducted in this study. The conversion efficiency is defined by ($nCO_{2,in}$ - $nCO_{2,out}$) / $nCO_{2,in}$, where $nCO_{2,in}$ and $nCO_{2,out}$ is the density of CO_2 flowing in and out the plasma chamber, respectively. The energy efficiency is a function of the absorbed power in the plasma, the flow rate through the chamber and the conversion efficiency, explicitly given in [**10**]. The simulated conversion and energy efficiency for a variation of the gas admixutre ratio and absorbed power agree well with the measured ones in [**3**], as presented in **Fig. 2** and **3**. Both the measurements and simulations show that the conversion efficiency is enhanced and the energy efficiency is reduced with the increasing absorbed power value, whereas the conversion efficiency is reduced and the energy efficiency is enhanced in the case of the increasing CO_2 admixture ratio. Note that the desired situation is a simultaneous enhanced conversion and energy efficiency. However, this is not available under the variation of the absorbed power and CO_2 admixture ratio, based on the current study. A further analysis of achieving the aforementioned desired situation or finding optimal conversion and energy efficiency under certain condition is of particular interest.

6 Summary & Outlook

Zero-dimensional model is developed to study the RF and ns-pulsed He/CO_2 plasma jets. Validation against experimental measurements carried out in [**3**] shows a good agreement for conversion and energy efficiency over a range of operating conditions. Insight into the chemical pathways towards CO_2 conversion is also revealed and will be discussed further during the conference.

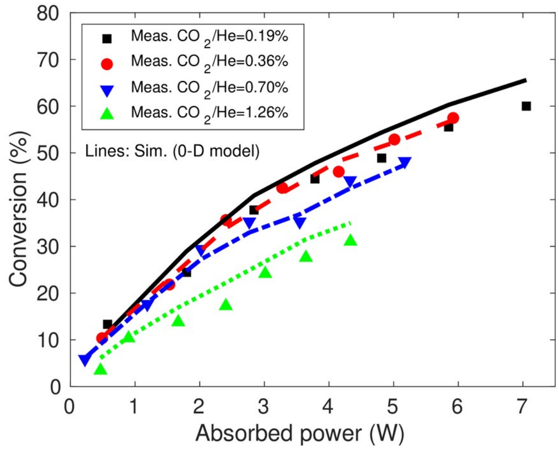

Fig. 2. Conversion efficiency of a RF He/CO_2 plasma jet. The measurement data (points) are taken from [**3**]. The simulation results (lines) are from this study.

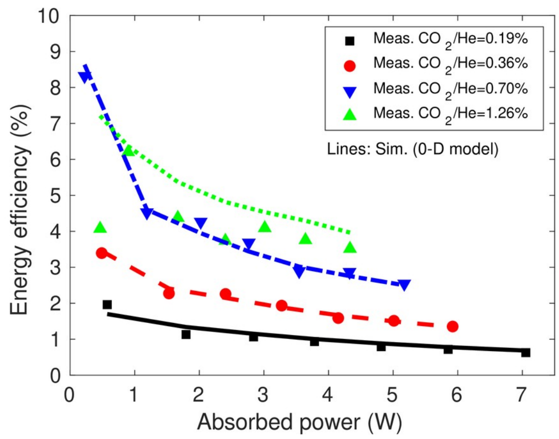

Fig. 3. Energy efficiency of a RF He/CO_2 plasma jet. The measurement data (points) are taken from [**3**]. The simulation results (lines) are from this study.

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