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Electron transport parameters and rate coefficients in $He/CH_4/O_2$ and $He/CH_4/CO_2$ mixtures

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Introduction

The use of non-thermal plasmas in the production of hydrogen, *Syngas*, and more complex hydrocarbons from methane mixtures is an interesting alternative to conventional processes [1]. Mixtures of methane with an oxidant are commonly used, the most common oxidants being O_2 and CO_2 [2]. Recently high conversion rates and selectivities were reported [3] using $He/CH_4/O_2$ and $He/CH_4/CO_2$ mixtures. In this work we present electron transport parameters and rate coefficients in $He/CH_4/O_2$ and $He/CH_4/CO_2$ mixtures as a first step in the development of a model of a dielectric barrier discharge in these mixtures.

Cross Sections and numerical method

The results were computed solving the electron Boltzmann equation for the hydrodynamic regime using the method described in [4]. The cross sections for He, O_2 and CO_2 were taken from [5] while methane cross sections were adjusted to swarm data using as initial set data from [6, 7] complemented by recent results from [8, 9]. The final set includes the momentum transfer cross section, two cross sections for vibrational excitation of modes $\nu_2+\nu_4$ and $\nu_1+\nu_3$, the dissociation cross sections for the formation of CH_3, CH_2 and CH radicals, the formation of H^- and CH_2^- negative ions, and CH_4^+, CH_3^+, CH_2^+, CH^+, C^+, H_2^+ and H^+ ions.

Results

The results were obtained keeping the ratio of concentrations between methane and the oxidant constant at $[CH_4]$:$[O_2]$ = 2.0 and $[CH_4]$:$[CO_2]$ = 1.0, respectively, while the helium concentration varied between 0% and 80%. The reduced electric field, E/n, ranged from 0.5 Td to 500 Td.

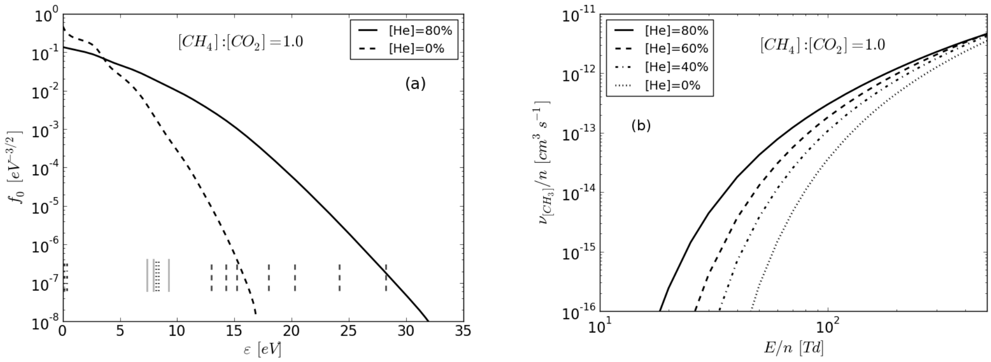

Fig. 1: Results in $He/CH_4/CO_2$ mixtures: (a) Isotropic component of the electron energy distribution function at $E/n = 50$ Td. Vertical lines: threshold values for vibrational excitation, dissociation, ionisation and attachment. (b) Variation of the density-normalized collision frequency for dissociation of CH_4 into CH_3, with E/n for several helium concentrations.

The electron energy distribution function (*eedf*) is shifted to higher values as the helium concentration increases (Fig. 1-(a)). As a result of the increase of the *eedf*, and in spite of the reduction of

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methane concentration, the collision frequencies for methane excitation and ionisation processes increase also with the helium concentration (Fig. 1-(b)). This variation is bigger at low E/n values, and decreases as the reduced field increases.

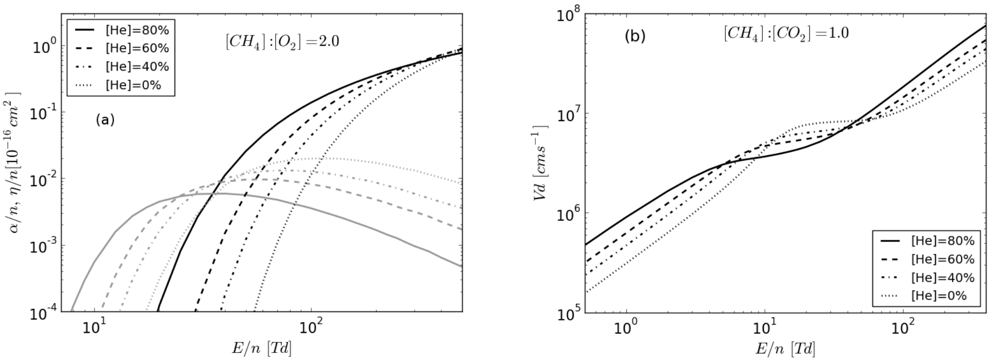

Fig. 2: Effect of helium concentration on (a) the density-normalized ionisation and attachment coefficients in He/CH_4/O_2 mixtures at a constant ratio [CH_4]:[O_2] = 2.0 (black lines: α/n; gray lines: η/n) and (b) the drift velocity in He/CH_4/CO_2 mixtures at a constant ratio [CH_4]:[CO_2] = 1.0

An increase in the helium concentration leads to a shift in the ionisation and attachment coefficients curves to lower E/n values (Fig 2-(a)). The E/n value for $\alpha/n = \eta/n$ decreases from above 100 Td to approximately 30 Td when the helium concentration changes from 0% to 80%.

In He/CH_4/CO_2 mixtures the drift velocity also increases with the helium concentration except in the range 6 to 50 Td where the curves for different helium concentrations cross each other in two points, resulting in an inversion of the relative order (Fig. 2-(b)). In He/CH_4/O_2 mixtures the above double crossing does not occur, only a single crossing is observed.

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