Kinetics in atmospheric-pressure discharges in Helium

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Introduction

In last years, helium and helium-based plasmas at atmospheric-pressure and room-temperature have received considerable interest due to their potential for biomedical applications. They can lead to the production of energetic species when interacting with open air, or to the generation of reactive radicals and ions when admixed with a molecular gas. Recently, different experiments have been carried out to study the dynamics of these plasmas in long capillary tubes, used for the development of medical devices [1]. To optimize these applications one needs to identify and to understand the elementary processes controlling the global behavior of the discharge and the kinetics of the main charged and neutral species in the plasma, produced using mixtures of helium with small quantities of nitrogen, oxygen or synthesized air (80%N_2-20%O_2).

Electron kinetics

In this work, the tool LoKI (LisbOn KInetics) is adapted and used to study electron-impact reactions in the aforementioned conditions. LoKI is a self-consistent numerical code that solves the two-term electron Boltzmann equation (EBE) together with a system of rate balance equations describing the creation and loss processes of the dominant plasma species. Firstly, this study focuses on the electron energy distribution function (EEDF) of helium plasmas, calculated for several values of the reduced electric field, *E/N*, analyzing the influence of He metastables involved in stepwise inelastic and superelastic collisions. From the EEDF it is possible to determine the electron-impact rate coefficients, as function of *E/N* [2]. The electron-neutral scattering cross sections used for the calculations in He include the following mechanisms [3]: elastic collisions; direct and stepwise excitation and ionization collisions, with ground-state He(1^1S) and with metastable excited states He(2^3S) and He(2^1S); de-excitation collisions with metastables He(2^3S) and He(2^1S). The influence of He metastables on the EEDF is noted mostly at low reduced electric fields, as figure 1 shows for 1Td. It is seen that electron-He(2^3S) superelastic collisions turn the EEDF more energetic, creating a plateau up to the He(2^3S) threshold at 19.82 eV. Ionization is obtained for electron energies above 3.97 eV for collisions with He(2^1S), 4.77 eV for collisions with He(2^3S) and 24.59 eV for collisions with He(1^1S). Figure 2 shows the effect of the presence of the metastables, plotting the total ionization coefficient as function of *E/N*.

Helium plasma kinetics

By introducing a kinetic scheme for He, the zero-dimensional simulations of LoKI allow to understand the kinetics in the plasma for given conditions. A detailed kinetic scheme for a pure He atmospheric-pressure discharge is given in [3], providing indication on the most important species and reactions. In the present work, LoKI is used with a reduced kinetic scheme for He heavy species, which can serve as a first step for further study of He-based plasmas. The scheme accounts for the ionic and excited species He^+, He_2^+, He(2^3S), He(2^1S), He(2^3P), He(2^1P), He(3^3S), He(3^3P), He(3^3D) and He_2* and for electron-impact, recombination, dissociation, Penning and associative ionization, charge transfer and diffusion mechanisms. The present scheme contains an improvement with respect to [3] in the rate coefficients for the associative ionization reactions He($3^3S/3^3P/3^3D$) + He(1^1S) → He_2^+ + e^-. The coefficients obtained from [4] and [5] for different gas temperatures between 100 K and 2500 K were fitted to polynomial functions of Tg, resulting in the temperature-dependent coefficients represented in figure 3. The scheme was validated by comparing results with those in [3], at atmospheric-pressure, for different gas temperatures and electron densities. In this work, as in [6], the gas temperature is set to 300 K, the pressure is atmospheric and the tube radius is set to 2 mm [1]. The field is taken as DC and the imposed electron density is varied to cover the range obtained in [6], between 10^9 and 10^{14} cm^{-3}.

The calculated values of *E/N* are in the range 4-6 Td and increase with n_e. He_2^+ is the dominant ion for all the values of n_e considered. Figure 4 shows the densities of the He excited states in the plasma. The synthesis of the analysis made for the charged species can be found in figures 5-8, where the set of reactions having influence on charge balance is represented. From figure 7, we conclude that associative ionization (F), stepwise ionization (B) and Penning ionization (D,E) rates are always very important for charge creation, reinforcing the role of the internal energy contained in the He excited states, transferred to ionization. Among the Penning reactions, those that depend on He(2^1S) or He(2^3P) are especially important. Excimer ionization (C) also proves to be relevant at high n_e. Charged-particle destruction is mostly controlled by diffusion (D'), at low n_e, and by dissociative recombination (A'), at high n_e. In the particular case of the molecular ions, the conversion reaction He^+ + 2 He(1^1S) → He_2^+ + He(1^1S) (G) is a very important creation mechanism, while the corresponding reverse reaction (E') is shown to be irrelevant for the present conditions. Numerical tests show that the species He(2^1P) and the electron-stabilized recombination reactions have a negligible role in the kinetics, for the present conditions, thus suggesting that they can be disregarded in a more reduced kinetic scheme.

Acknowledgements

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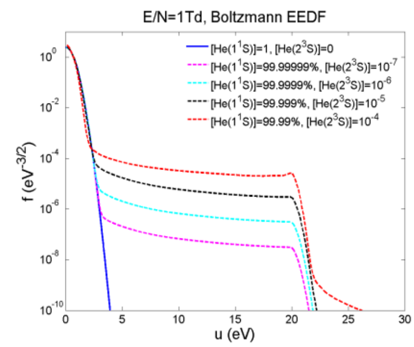

Figure 1: EEDFs at *E/N* = 1 Td, for several mixtures of the ground-state He(1^1S) and the excited-state He(2^3S).

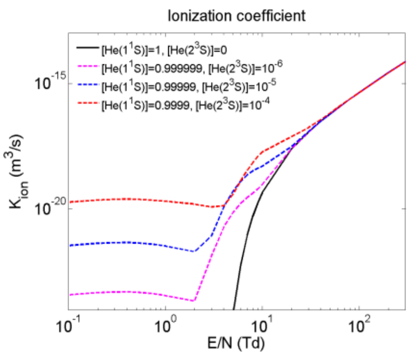

Figure 2: Electron-impact ionization coefficients, as function of *E/N*, for several mixtures of He(1^1S) and He(2^3S).

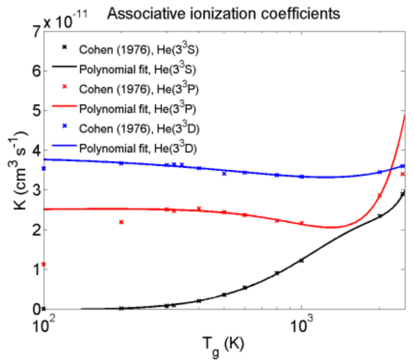

Figure 3: Associative ionization coefficients obtained from [4] and [5] and fitted to the formulae: for He(3^3S), $4.43\times10^{-24}\times T_g^4 - 2.35\times10^{-20}\times T_g^3 + 3.88\times10^{-17}\times T_g^2 - 7.56\times10^{-15}\times T_g + 2.62\times10^{-13}$; for He($3^3P$), $6.73\times10^{-21}\times T_g^3 - 1.49\times10^{-17}\times T_g^2 + 4.70\times10^{-15}\times T_g + 2.48\times10^{-11}$; for He($3^3D$), $-6.57\times10^{-22}\times T_g^3 + 5.16\times10^{-18}\times T_g^2 - 9.76\times10^{-15}\times T_g + 3.86\times10^{-11}$.

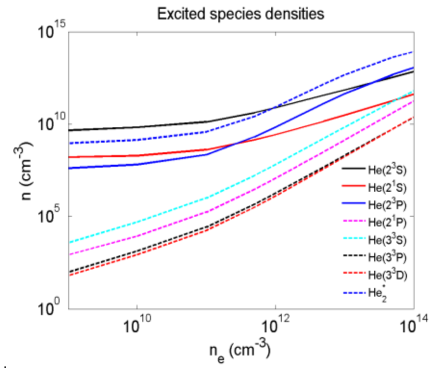

Figure 4: Densities of the Helium excited species in a steady-state He discharge at P_{atm} and T_g = 300 K, as function of the electron density.

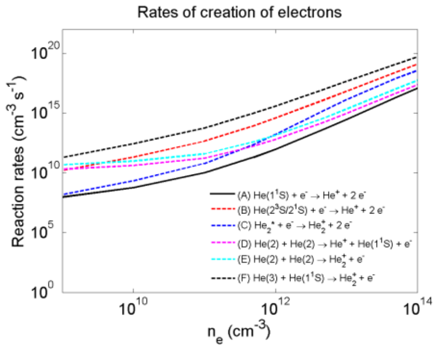

Figure 5: Charged-particle production rates in a steady-state He discharge at P_{atm} and T_g = 300 K, as function of the electron density: A – direct ionization; B – stepwise ionization; C – excimer ionization; D and E – Penning ionization; F - associative ionization.

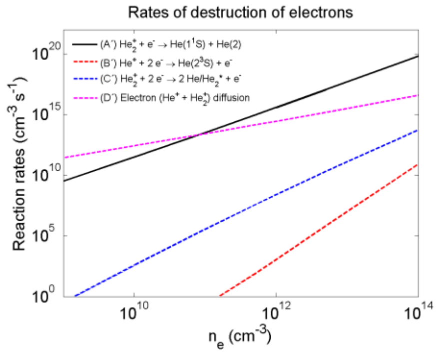

Figure 6: Charged-particle destruction rates in a steady-state He discharge at P_{atm} and T_g = 300 K, as function of the electron density: A' – dissociative recombination; B' and C' – electron-stabilized recombination; D' – diffusion.

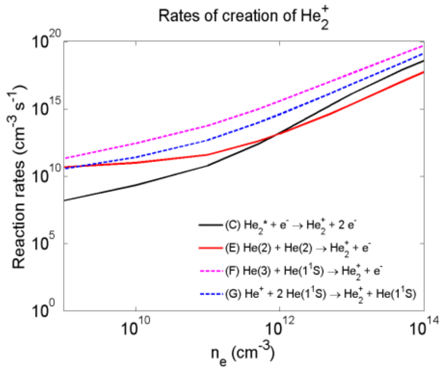

Figure 7: He_2^+ production rates in a steady-state He discharge at P_{atm} and T_g = 300 K, as function of the electron density: C – excimer ionization; E – Penning ionization; F – associative ionization; G – Ion conversion.

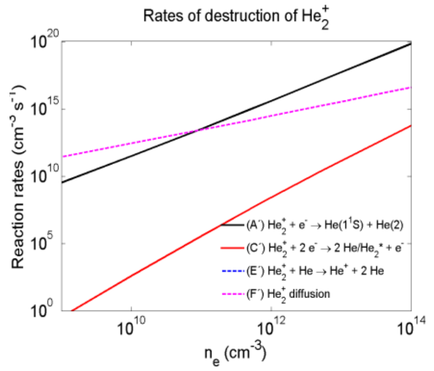

Figure 8: He_2^+ destruction rates in a steady-state He discharge at P_{atm} and T_g = 300 K, as function of the electron density: A' – dissociative recombination; C' – electron-stabilized recombination; E' – ion conversion; F' - diffusion.