Kinetics of atmospheric-pressure discharges in helium mixtures with nitrogen and oxygen

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This work studies different elementary processes controlling the global discharge behavior and the kinetics of the main charged and neutral species in helium / nitrogen / oxygen plasmas. Numerical simulations of the electron impact reactions in helium-based plasmas are performed using the IST-LoKI computational tool. The influence of He metastable states and consequent superelastic and stepwise ionization collisions are documented, as well as the influence of the admixture of air gases with different collision cross sections and ionization thresholds. For a relative density of the molecular gas of only 0.1%, the calculated ionization rate coefficient is shown to be higher than in pure He. Particle balance equations have been solved for a reduced He-N_2 kinetic scheme, considering the local application of a transitory electric field with 50Td maximum amplitude. The key role of the N_2 admixture in He and of various elementary processes, especially Penning reactions, on the ionization degree of the plasma is pointed out.

1. Introduction

In last years, helium and helium-mixture plasmas at atmospheric pressure have received considerable interest due to their potential for biomedical applications. In particular, the propagation of atmospheric-pressure helium microplasma jets in long capillary tubes is being studied for the development of medical devices.

So far, most experiments have been dedicated to the study of the plasma plume. But for endoscopic treatments with jets, it is also important to better understand and optimize the propagation of discharges in long dielectric tubes as catheters. Recently, different experiments have been carried out to study the dynamics of discharges in tubes [1,2].

In this context, mixtures involving small quantities of nitrogen, oxygen or synthesized air (80%N_2-20%O_2) have acquired great importance. The optimization of applications requires identifying and understanding the elementary processes controlling the global behavior of the discharge and the kinetics of the main charged and neutral species in helium / nitrogen / oxygen plasmas. Also, the definition of a reduced kinetic reaction set is relevant for the modeling of helium-based plasma jets, in order to provide a consistent analysis of the plasma ignition and propagation in dielectric tubes.

2. Models and results

In this work, the tool IST-LoKI (LisbOn KInetics) is adapted and used to study the plasma kinetics in the aforementioned conditions. IST-LoKI is a self-consistent numerical code that solves the two-term electron Boltzmann equation together with a system of balance equations describing the creation and loss processes of the dominant species.

2.1. Electron kinetics

The present study focuses first on the electron energy distribution function (EEDF) of helium-containing plasmas, calculated for several values of the reduced electric field (*E/N*). In particular, the effects of small admixtures of N_2, O_2 and synthesized air, and the influence of He metastables (He(2^3S) and He(2^1S)), involved in stepwise ionization and in superelastic collisions, are investigated.

The influence of He metastables on the EEDF is noted mostly at low reduced electric fields. Figure 1 shows the case for 1Td, when the relative density of He(2^3S) is increased from 0 to 10^{-4}. 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 19.82eV, corresponding to the energy gained by one electron in a superelastic collision.

The addition of metastable states has a noticeable effect also in the calculated electron rate coefficients, particularly in the ionization coefficient. Figure 2 shows this effect for several mixtures of He(1^1S) and He(2^3S) over a wide range of *E/N* values, where an evident influence of stepwise ionization reactions is perceived for reduced fields below 20Td.

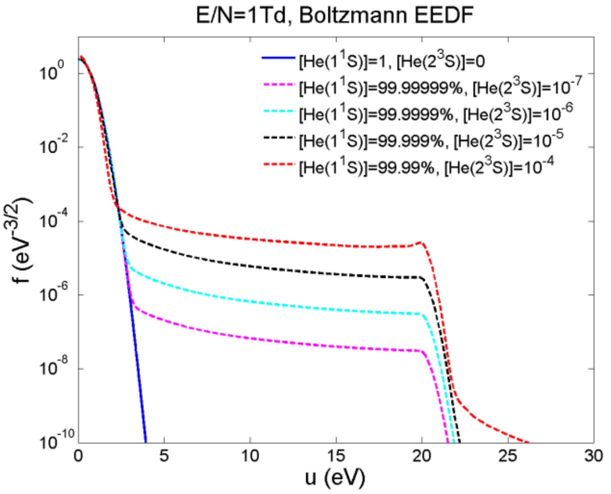

Figure 1: EEDF at *E/N*=1Td for several mixtures of helium ground-state and metastable 2^3S

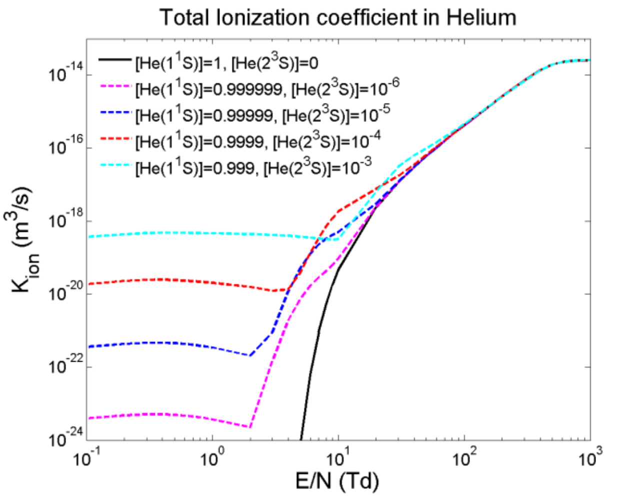

Figure 2: Ionization coefficient in helium, as a function of *E/N*, for different relative densities of He(1^1S) and He(2^3S)

The addition of air constituents to He affects also the EEDF, as shown in Figure 3 for He-N_2 mixtures at *E/N*=50Td, where special attention is paid to scenarios at low N_2 densities. Note that this value of *E/N* – tens of Td – corresponds to the order of magnitude expected for the local reduced electric fields in the propagation front of helium discharges in tubes. Results show that the atomic gas He presents a much more energetic EEDF than the molecular gas N_2. The excited and ionized states of He have very-high energy thresholds, in contrast with the low-energy electronic and vibrational excited states of N_2, leading to a depletion of the EEDF tail when nitrogen is admixed.

The changes observed in figure 3 for the EEDF induce differences also in the calculated rate coefficients, particularly in the global electron-impact ionization coefficient. Figure 4 presents this coefficient for several He-N_2 mixtures, in the range of interest for *E/N*. Results show that the admixture of small percentages of N_2 has little effect on the ionization coefficient, in coherence with the minor changes caused also in the EEDF (see figure 3). However, for the conditions of figure 4, a maximum of the ionization coefficient is obtained for an admixture of only 0.1% N_2, while a large decrease is obtained already at 10% N_2.

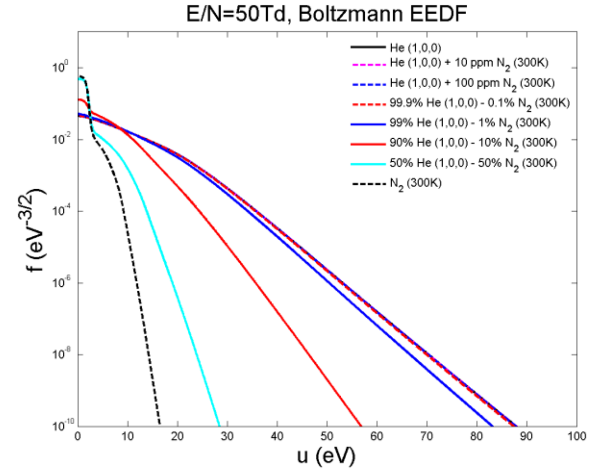

Figure 3: EEDF at *E/N*=50Td for several mixtures of He (ground-state only) and N_2

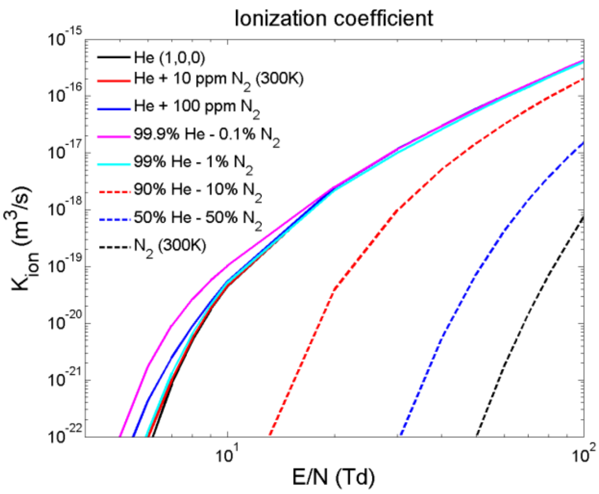

Figure 4: Ionization coefficient, as a function of *E/N*, for different relative densities of He(1^1S) and N_2

The effect in the EEDF of an admixture of O_2 is similar to the one observed for He-N_2 mixtures. However, the electronegativity of O_2 is shown to have an important effect on the plasma behavior, as it adds an attachment reaction to the electron kinetics. Figure 5 depicts the effective ionization coefficient, defined as the difference between the electron-impact ionization coefficient and the attachment coefficient, calculated for several mixtures of He with synthesized air (80%N_2-20%O_2).

A major difference between the results presented in figures 4 and 5 is that the addition of O_2 (hence of an attachment mechanism) is responsible for the appearance of negative values in the effective ionization coefficient. These negative values are obtained at low reduced electric fields depending on the mixture composition (see the interrupted curves in figure 5), and they suggest enhanced difficulties in the breakdown of oxygen-containing plasmas.

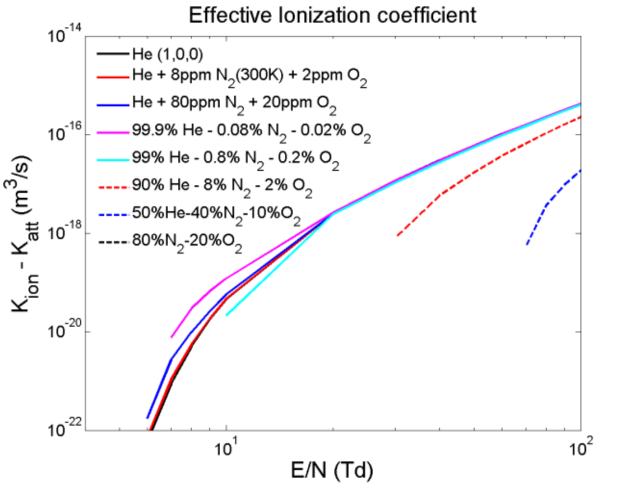

Figure 5: Effective ionization coefficient, as a function of *E/N*, for different relative densities of He(1^1S) and dry air

2.2. Heavy species kinetics

The study of electron reactions alone, although important, is insufficient to provide a detailed description of the plasma, revealing its dominant species and their creation and loss mechanisms. For this purpose, reactions between heavy species have to be considered also, which demands solving the global system of rate balance equations for the different plasma constituents. At present, this task is done using the ZDPlasKin [3] and Bolsig+ [4] solvers.

A detailed kinetic scheme for a pure helium atmospheric-pressure discharge is given in [5], accounting for He^+, He_2^+, He($n^{2s+1}l$) and He_2^*, and providing indication on the most important species and reactions. In the present work, the definition of a collisional-radiative model using a reduced kinetic scheme for He heavy species is put forward, considering the conditions of interest for He plasma jets.

In [6], a kinetic scheme has been derived from the detailed experimental study of an atmospheric-pressure discharge in helium with a small admixture of N_2. In this kinetic scheme, the importance of three-body Penning ionization and of charge transfer reactions was evidenced. In the present work, that kinetic scheme is reduced by taking into account 4 positive ions (He^+, He_2^+, N_2^+, N_2^+(B)), He excited species and electrons.

Here, the influence of nitrogen admixtures on the discharge kinetics is studied. Figure 6 shows the temporal evolution of the electron density behind the discharge front of a typical atmospheric-pressure discharge, propagating in a tube as in [7]. Calculations use as input the time evolution values of the local transitory electric field, also presented in figure 6.

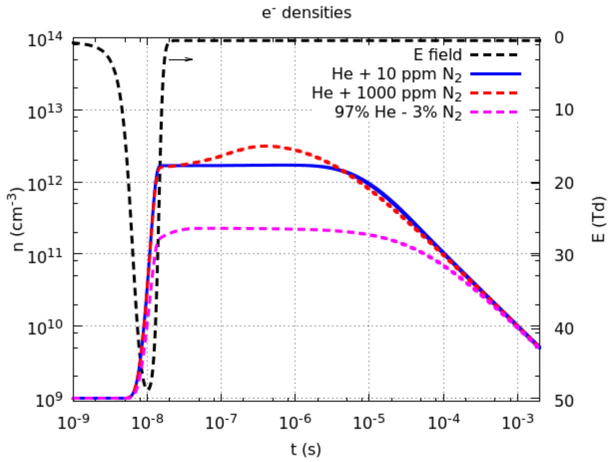

Figure 6: Time evolution of the local electron density, for local transitory pulsed field with 50Td maximum amplitude

Figure 6 shows that the admixture of low concentrations of N_2 leads to an increase in the ionization of the post-discharge behind the discharge front. The highest ionization coefficient is attained at 1000 ppm N_2. However, higher concentrations of N_2 (e.g. 3%) lead to much lower ionization levels due to the decrease in the initial electron-impact ionization coefficient, in accordance with the results shown in figures 3 and 4. In the particular case of atmospheric-pressure discharge propagation in tubes, the ionization level behind the discharge front affects the conductivity of the plasma in the tube, hence the discharge dynamics and structure. The kinetic effects will therefore influence the jet propagation length and velocity, with higher velocities observed in the case of 99.9%He-0.1%N_2.

For the particular case of 1000 ppm of N_2, the influence of some species and reactions is also studied. In particular, the importance of Penning reactions (He(2^3S) + N_2 $\rightarrow$ He + N_2^+ + e^-) on the discharge structure is quantified. Figures 7 and 8 compare the plasma composition when Penning reactions are included and discarded.

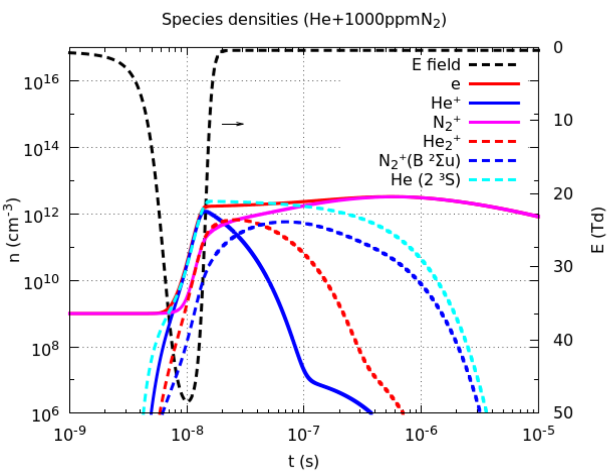

Figure 7: Time evolution of the local density of species, for local transitory pulsed field with 50Td maximum amplitude. Simulation results considering the full kinetic scheme

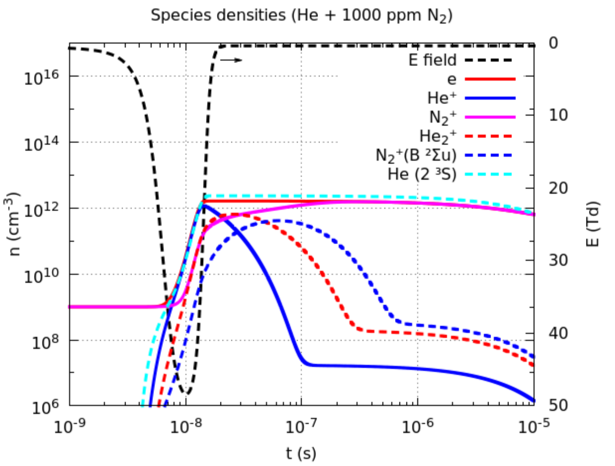

Figure 8: Time evolution of the local density of species, for local transitory pulsed field with 50Td maximum amplitude. Simulation results discarding Penning reactions from the kinetic scheme

These figures reveal that Penning reactions are essential to describe the ionization level in the post-discharge behind the discharge front, showing the importance of both He(2^3S) and N_2 species on the discharge dynamics and further explaining the relevance of N_2 admixture.

3. Conclusions

The importance of electron-impact reactions in helium-based plasmas has been analysed with numerical simulations, using either the IST-LoKI tool or the ZDPlasKin and Bolsig+ solvers. The influence on the EEDF of the He metastable states causing superelastic collisions was documented. Metastable states were proven to affect also the global electron-impact ionization in the discharge, depending of their densities, due to stepwise ionization reactions.

EEDFs have been compared for different mixtures of He with the atmospheric gases N_2 and O_2, exhibiting higher tails in the pure noble gas. The effect of these mixtures on the calculated electron-impact rate coefficients has also been registered, with emphasis on the global ionization coefficient. The cases of low molecular gas densities, around 0.1%, were shown to provide higher ionization coefficients than the case of pure He, due to the low ionization thresholds of N_2 and O_2. The electronegativity of O_2 was shown to affect the electron kinetics, as a result of the electron attachment mechanism. The analysis of the effective ionization rate coefficient shows that discharge breakdown is more difficult in the presence of O_2.

Particle balance equations have been solved for a reduced He-N_2 kinetic scheme, considering the local application of a transitory electric field with 50Td maximum amplitude. The key role of kinetic processes, especially Penning reactions, on the ionization degree of the plasma has been put forward. Moreover, the relation between nitrogen admixture and the different elementary processes participating in the kinetics has been studied and discussed, revealing the influence of N_2 on the plasma ionization level and its implications in the discharge structure.

4. Acknowledgements

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5. References

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