Control of electron heating and plasma chemistry in atmospheric pressure radio frequency plasma jets

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Abstract: Understanding and tailoring the electron power absorption dynamics is the key to optimizing the generation of reactive/excited neutrals in atmospheric pressure plasma jets. Such control is, however, limited in standard single frequency discharges with planar electrodes. Based on experiments and simulations, we demonstrate that enhanced plasma control can be realized by customized driving voltage waveforms and electrode topologies. For $He/N_2/O_2$ gas mixtures we show that a more efficient and tailored production of active species densities such as He metastables, O and N atoms can be achieved in this way.

Keywords: Plasma chemistry control, Voltage Waveform Tailoring, customized electrodes.

1. Introduction

Capacitively coupled micro atmospheric pressure plasma jets (μAPPJs) driven at radio frequencies (RF) are important tools for a variety of applications of high societal benefit such as plasma medicine and surface processing. They are typically operated at a single driving frequency and based on planar electrodes. There is a lack of fundamental understanding of the effects of external control parameters such as the driving voltage waveform and the electrode topology on the spatio-temporal electron power absorption dynamics and the mechanisms of reactive species generation in such plasma sources as a basis for knowledge based plasma process development. Based on a synergistic combination of experiments and simulations, we obtain such understanding for $He/N_2/O_2$ gas mixtures by studying the space and nanosecond time resolved dynamics of energetic electrons and the buildup of neutral species by electron-neutral dissociation from the gas inlet towards the nozzle and into the effluent. We demonstrate that Voltage Waveform Tailoring (VWT) [1-7] and customized electrode topologies [8] provide enhanced control of the Electron Energy Probability Function (EEPF) in μAPPJs. In this way, the generation of selected neutral species such as helium metastables, atomic nitrogen and oxygen is enhanced, controlled, and made more energy efficient.

2. Experiment

Measurements are performed based on a reference microplasma 'COST-jet' [8]. The active plasma volume is confined between two parallel stainless steel electrodes of 30 mm length (see fig. 1). The electrode gap is fixed at 1 mm. The plasma is operated at different mixtures of $He/N_2/O_2$, tailored driving voltage waveforms and electrode topologies. Such voltage waveforms are generated by an arbitrary function generator in combination with a broadband amplifier as a finite Fourier Series of multiple consecutive harmonics of 13.56 MHz with individually adjustable harmonics' amplitudes and phases [1-7]. The electrode topologies include trenches of different shapes and dimensions implemented into both electrodes [8].

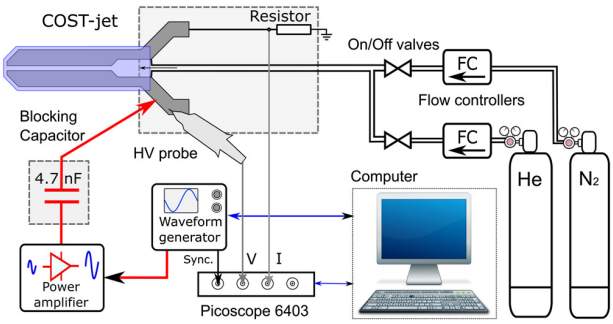

Fig. 1. Sketch of the experimental setup used for He/N_2 gas mixtures [5].

The spatio-temporally resolved dynamics of energetic electrons is measured by Phase Resolved Optical Emission Spectroscopy (PROES) [2,3]. Helium metastable densities are detected by Tunable Diode Laser Absorption (TDLAS) [3], while atomic oxygen densities are measured by Two Photon Absorption Laser Induced Fluorescence (TALIF) [4]. Time resolved voltage and current measurements are performed, too. Finally, the gas temperature is accessed spatially resolved by Optical Emission Spectroscopy (OES).

3. Simulation

Computational investigations are performed based on 1d3v PIC/MCC simulations for He/N_2 [10] as well as 1d and 2d fluid [11] and hybrid fluid/kinetic simulations [6] for various $He/N_2/O_2$ gas mixtures. The simulations use the measured driving voltage waveforms as input and are performed under identical discharge conditions as those used experimentally. In this way a quantitative multi-diagnostic experimental validation of these simulations is performed, based on which the simulations provide space

and time resolved access to plasma parameters that cannot be measured. These include electron densities, EEPFs, and electric fields.

4. Results

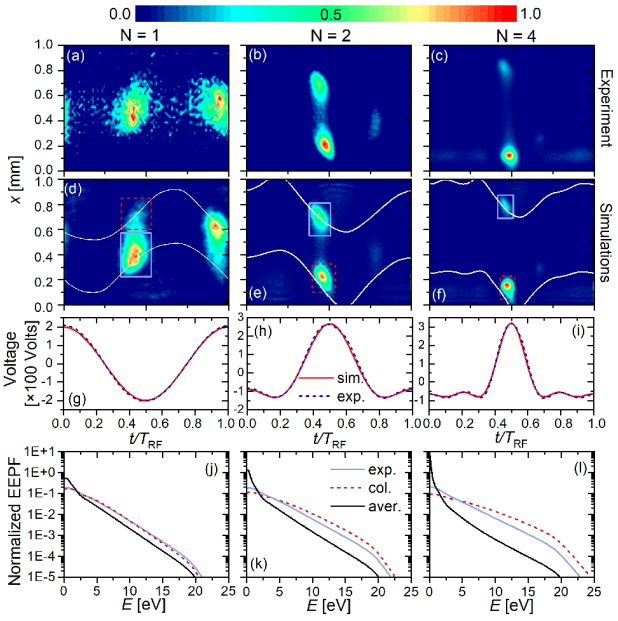

Fig. 2. Spatio-temporal plots of the electron impact excitation rate from the ground state into He-I $^3S_1$ for different peaks driving voltage waveforms constructed from N consecutive harmonics of 13.56 MHz. EEPFs averaged over different spatio-temporal regions of interest marked in row 2 (obtained from PIC/MCC simulations). Discharge conditions: 400 V peak-to-peak driving voltage, gas flow ratio He/N_2 = 1000/1 [2].

Capacitively coupled μAPPJs can be operated in different modes [10]. In the Ω-mode and based on PROES measurements as well as PIC/MCC simulations, we find that electrons absorb power mostly by acceleration in high drift electric fields in the plasma bulk at low driving voltage. In the Penning mode at high driving voltages, the ionization is dominated by secondary electrons and electrons generated as a consequence of Penning ionization inside the sheaths. Recent investigations show that gas heating along the direction of the gas flows can induce heating mode transition. Understanding such fundamentals of electron power absorption is the basis for the knowledge based development of concepts to control the plasma chemistry via tailoring the EEPF by controlling the electron heating dynamics. 2d fluid/kinetic simulations allow tracing the buildup of various neutral species along the direction of the gas flow from the gas inlet towards the effluent.

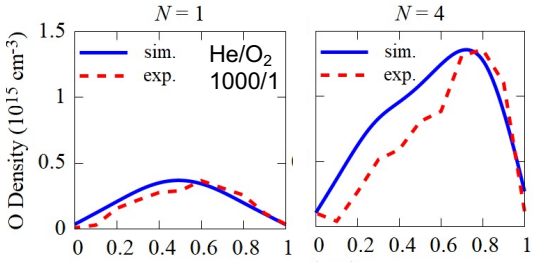

Fig. 3. Simulated and measured time averaged atomic oxygen density profiles as a function of the number of driving harmonics of 13.56 MHz, N, in the center of the active plasma region. Discharge conditions: 500 V peak-to-peak driving voltage, gas flow ratio He/O_2 = 1000/1 [6].

Fig. 2 shows the effects of using multi-frequency peaks driving voltage waveforms synthesized from up to N = 4 consecutive harmonics of 13.56 MHz on the spatio-temporally resolved dynamics of energetic electrons and EEPFs in different regions of interest. The plasma is operated in the Ω-mode. In contrast to the standard single frequency operation, using such tailored voltage waveforms allows to control the spatial and temporal symmetry of the electron dynamics. It allows to dissipate most of the power, that is transferred to electrons, to a few electrons at a specific spatial position and time within the fundamental RF period, namely adjacent to the bottom electrode during the local sheath collapse.

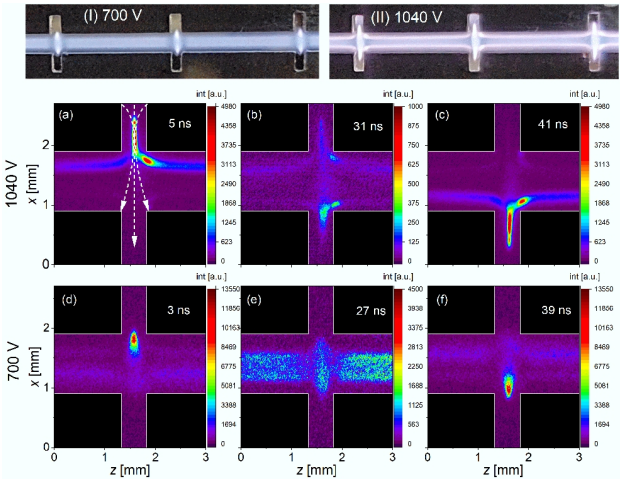

Fig. 4. Top (I, II): Photos of the jet with customized electrode topologies at different peak-to-peak driving voltages. Bottom: Measured 2D space resolved electron impact excitation dynamics within a single trench at different times within one RF period. Discharge conditions: 13.56 MHz, He with 0.5 % O_2 admixture, vertical trenches (1 mm deep, 0.5 mm wide) at both electrodes. The gas flows from the left to the right for all images [8].

This happens because of the short local sheath collapse and the requirement to draw sufficient electrons to this electrode during the sheath collapse to compensate the

positive ion flux to this surface on time average. Thus, a strong electric field accelerates electrons towards the electrode and leads to an enhanced high energy tail of the EEPF, since a lot of power is dissipated to a few electrons. As a consequence of this EEPF control by VWT a variety of neutral species generated by energetic electron-neutral collisions is produced more efficiently as compared to single frequency operation. This includes He metastables, N and O atoms (see fig. 3). A detailed analysis of the power dissipation to the plasma shows that the energy efficiency of producing such active neutral species is indeed enhanced by VWT [11].

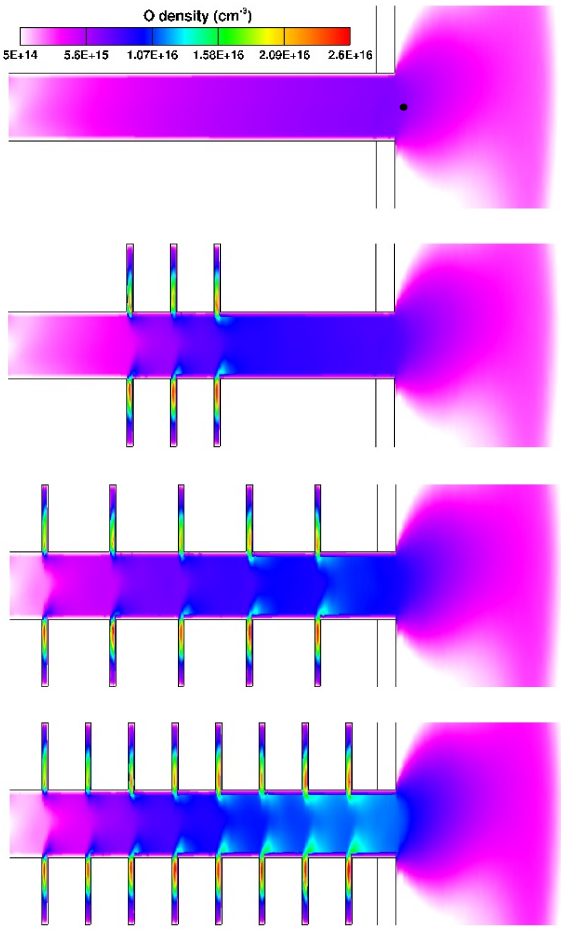

Fig. 5. 2D profiles of the time averaged atomic oxygen densities obtained from fluid simulations for different numbers of trenches. Discharge conditions: 13.56 MHz, 850 V, He + 0.5 % O_2 [8].

Instead of using VWT, the dynamics of energetic electrons can also be controlled by introducing structured electrodes. Fig. 4 shows the effects of vertical trenches (1 mm deep, 0.5 mm wide) on the electron dynamics in a He/O_2 discharge driven at 13.56 MHz. The presence of such trenches leads to a current focusing effect inside these structures and at their orifices during the local sheath collapse, when electrons propagate into such surface structures. Consequently and due to the low electron density inside the trenches, such electrons are accelerated by strong electric fields at times of high current within each RF period. Again, most of the power dissipated to electrons is dissipated to a relatively low number of electrons so that each electron gains high energy.
As shown in fig. 5 this leads to enhanced production of atomic oxygen inside and at the orifice of such trenches. Similarly the generation of He metastables is enhanced. Combining multiple such surface structures allows to enhance the atomic oxygen density at the nozzle of the jet.

5. Conclusions

Based on a synergistic combination of experiments and simulations the electron power absorption dynamics and plasma chemistry in capacitively coupled μAPPJs driven by RF voltage waveforms in He/N_2/O_2 gas mixtures is understood. These fundamental insights provide the basis for enhancing the generation of reactive and excited neutrals by collisions of energetic electrons with the neutral background gas by VWT and structured electrodes. Both methods allow controlling the electron heating dynamics in such a way that a large fraction of the total power dissipated to electrons is transferred to such particles at distinct spatial positions and times within the (fundamental) RF period. In this way the EEPF is tailored so that its high energy tail is enhanced at specific positions and times so that a high density of reactive/excited neutrals, is produced in an efficient way.

6. Acknowledgment

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7. References

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