8 Conclusions and outlook

A self-pulsing micro-scaled atmospheric pressure plasma jet (SP-µ-APPJ) has been developed and its dynamic processes are investigated with the help of multiple plasma diagnostics. The SP-µ-APPJ is a modification of the µ-APPJ with a wedged electrode configuration. The SP-µ-APPJ can be operated in two operation regimes, i.e. the cw-operation and the self-pulsing operation regime that each possesses several different discharge modes.

At low applied sender powers the device is in cw-operation. At the threshold of ignition, a homogeneous glow discharge establishes extending to about one half of the electrode length. It is sustained by ionisation processes located in the plasma bulk during sheath expansion and sheath collapse. Therefore, this discharge mode can be identified as 'α-mode' known from its analogue in low pressure capacitive coupled rf-discharges. However, ionisation and electron heating is not caused by the direct interaction with the sheaths (stochastical heating), but by high drift fields in the bulk at the corresponding phase positions within an rf-cycle. Ohmic heating is the dominant mechanism, for which reason this mode has also been introduced as 'Ω-mode'.

With increasing sender power the plasma expands along the electrodes until the complete discharge volume is covered. A second ionisation mechanism appears at the phase of maximum sheath width, whose contribution to the sustainment of the discharge steadily increases. It is characterised by electron avalanches within the sheaths that are seeded by secondary electrons produced via Penning ionisation and pooling reactions among metastable atoms and dimers. As these processes occur close to the surface, where the helium metastable density peaks, this second ionisation mechanism is connected with the γ-coefficient as it is the case for discharges at low pressure. At maximum applied sender power for the cw-regime, both ionisation mechanisms sustain the discharge to a comparable degree and the corresponding discharge mode is titled 'hybrid-mode' or 'α-γ-transition'.

Beyond a certain threshold, the device enters the self-pulsing operation regime with the ignition of a constricted discharge at position of smallest electrode gap distance. The discharge of bright intensity propagates along the electrode configuration and ex-
tincts close to the nozzle, before it reignites at the smallest gap again starting a new self-pulsing cycle. At 2 slm helium feed gas flow a repetition frequency of 1.2 kHz is reached. It consists of intense feet structures close to the electrode surfaces that are connected by a weaker column called central glow. During a complete cycle, the constricted discharge coexists with an homogeneous 'α-mode' glow. This discharge is now dominantly sustained by ionisation and metastable production inside the high field sheaths at time of maximum sheath width. Weak bulk emission and ionisation is attributed to stepwise processes out of the metastable states via low-energetic electrons that have been accelerated in low bulk fields. The constricted discharge could be identified as the known γ-mode-like discharge created in the course of an α-γ-transition instability at atmospheric pressure. The maximum dissipated power is about 18.8 W.

The evolution dynamics of the constricted discharge i.e. its ignition, propagation, extinction and afterglow are mainly determined by helium metastables and gas heating. During the ignition phase within the first 80 µs of the self-pulsing cycle, the 'γ-mode-like' discharge stabilise itself after passing an abnormal regime via a thermal instability. The resulting self-sustaining discharge is characterised by high electron and metastable densities in the order of $5 \cdot 10^{13} \text{ cm}^{-3}$.

Its propagation occurs at gas velocity assuming a constant elevated gas temperature of about 600 to 650 K. Penning ionisation of impurities is a key process for the propagation i.e the subsequent ignition at wider gap distances. It is aided by an increase in the reduced electric field due to the increased temperature. The extinction of the constricted discharge close to the nozzle is influenced by impurities, intruding from the ambient air. Their relatively high concentrations act as electron and metastable sink stopping the discharge and lead to its expiration. A post discharge / afterglow develops, where the dynamics are driven by long lasting high metastable densities produced by the slow recombination processes in helium.

The re-ignition of the 'γ-mode-like' discharge can be described in the same manner than the transition of the operation regimes, when increasing the applied sender power, i.e. by an α-γ instability. The particular discharge volume at the position of smallest gap distance undergoes the different discharge modes known from the cw-regime beforehand.

The investigation of the evolution dynamics of the constricted discharge reveals again the essential character of gas impurities in microplasmas. Their level of concentration determines, by influencing electron and metastable densities, if the discharge ignites and sustains itself, propagates or expires.

Furthermore, the ignition dynamics of the constricted discharge have been investigated in detail by using phase-resolved optical emission spectroscopy. The following
two different ignition situations have to be distinguished: the ignition at position of smallest gap distance and the subsequent ignition of the constricted discharge at wider gap distances responsible for its propagation. It has been revealed that both follow the same physical mechanism and are consequently comparable in nature.

Ignition of the constricted discharge occurs due to a sheath breakdown following the $\alpha$-$\gamma$-instability. As prerequisite, a particular discharge volume must pass the bulk-dominated discharge modes before this ignition is possible. During propagation (ignition at wider gap distances) this is not realised by an increase in applied sender power (as in cw-operation), but by a local increase in metastable density close to the electrodes in the respective volume. This increase in metastable density is initiated by the feet heads of the arriving constricted discharge.

The propagation by subsequent ignition is exploited to investigate the sheath breakdown at atmospheric pressure with high temporal resolution. High energetic electron beams serve as ionisation waves and metastable sources in the bulk. This is a consequence of the sheath breakdown and is responsible for the creation of a stable $\gamma$-mode-like discharge. The increased gas temperature aids this process. The coexistence of the various discharge modes along the electrode configuration (at a distinct time point) could be explained by the measured metastable density distribution in the afterglow of the just passed constricted discharge.

To exploit the self-pulsing operation regime for generating chemically reactive species, the conditions for a stable, long-term operation with oxygen admixtures in the self-pulsing regime have been examined. Critical parameters are gas heating and admixture concentration. If certain thresholds are exceeded, a stationary constricted discharge is formed. For the present SP-$\mu$-APPJ device, stable operation has been achieved for 2 slm helium feed gas flow with 1.5 sccm oxygen admixture, resulting in 490 V maximum voltage amplitude and 600 K gas temperature inside the $\gamma$-mode-like discharge. Electron density and temperature in the central glow are of about $9 \cdot 10^{12} \pm 2 \cdot 10^{12} \text{ cm}^{-3}$ and $1.4 \pm 0.6 \text{ eV}$, respectively.

The measured high atomic oxygen density produced by the $\gamma$-mode-like discharge can be described by a bulk-driven plasma chemistry in the central glow with the help of zero-dimensional chemistry model. Dissociation degrees up to 83% can be reached for the present device after about 400 $\mu$s cycle time. Atomic oxygen production in the sheaths seems to be restricted due to the high metastable density. Phase-resolved emission dynamics of the dominating atomic oxygen lines suggest novel production channels by dissociative excitation in heavy particle collisions between helium and oxygen metastables.
The resulting high density oxygen pulse of a constricted discharge is delivered to a possible target due to its propagation and the gas flow, where the temporal alternation of the coexisting homogeneous glow and the constricted discharge create a selective chemistry for the treatment. Depending one time, high or low fluxes of atomic oxygen can reach the target. A possible drawback may be the increased gas temperature in the effluent, which could be too high for heat-sensitive biomedical applications. Temperature can be reduced by decreasing the self-pulsing frequency by a reduction of the gas flow.

In conclusion, it can be stated that the questions defining the research focus of this thesis (cp. chapter 1) were answered. The SP-µ-APPJ is an adequate tool to study the α-γ-transition instability at atmospheric pressure. It can be operated in a stable repetitive manner to study fundamental processes on the one hand and selective chemical reactions on the other hand.

For future research, it would be interesting to investigate the influence of surface processes on the constricted discharge and especially on the sheath-breakdown dynamics at atmospheric pressure. This can be realised by changing the electrode material. The developed SP-µ-APPJ in combination with the synchronised phase-resolved emission spectroscopy setup offers the opportunity to study the complete evolution of a γ-mode-like discharge by recording only one image while propagating.

From the applicational point of view, the investigation of water and nitrogen admixtures are of great interest. Water is added in the coplanar device for the generation of the hydroxyl radical. As simulations show a high production efficiency for a γ-mode-like discharge, the SP-µ-APPJ could be an ideal source for optimising the hydroxyl output.