Fundamental Aspects of Filamentary Jet Discharges Interacting with Biologically Relevant Liquids and with Cells

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Plasma jets interacting with liquids are studied for biomedical effects in plasma medicine. While many processes induced in the liquid can be explained by equilibrium chemistry, plasma liquid interaction and plasma biological effects are strongly influenced by non-equilibrium chemistry and by radiation energy transfer processes. The work presents the role of metastable species and VUV radiation in energy dissipation processes relevant for plasma medicine. The complex challenges of diagnostics in filamentary plasma jets, usually studied in many plasma medical investigations are presented at the example of planar laser induced fluorescence spectroscopy and laser absorption spectroscopy. Finally, standard chemical methods are used to correlate liquid to gas phase chemistry and correlate the response of biological systems to plasma treatment.

1. Introduction

Cold atmospheric pressure plasma jets have emerged in the 1990s [1] and sparked the new field of plasma medicine ([2] and references therein). In recent years the interaction of plasma with biological targets was investigated by many groups. Since biological systems are usually in a wet or liquid environment, the effect of plasmas onto liquids has gained increasing attention in the research field of plasma medicine e.g. [3-7]. Fundamental pre–requisite for these plasma-liquid-biological cell systems is the operation of the discharge at atmospheric pressure. Many of these plasma sources are plasma jets, which at atmospheric pressure often are filamentary with plasma channel dimensions of few tens of micrometres. These small dimensions result in high gradients in space but also in time.

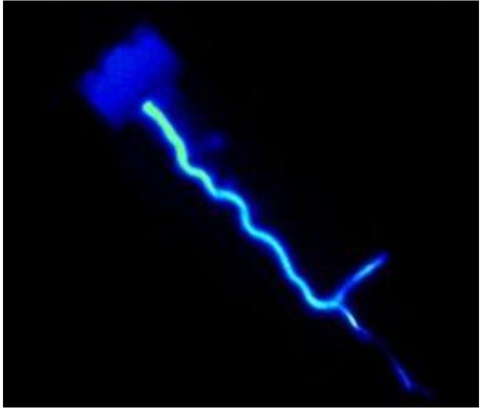

Fig. 1: filamentary plasma jet effluent splitting at the tip

Filamentary plasma jets exhibit dynamic processes that were termed plasma bullet, but are streamer like ionization waves with velocities of several ten thousands m/s [8]. The pathways that these so-called guided streamers take are determined by the surrounding atmosphere [9].

2. Plasma Source

In this work, investigations on a cold atmospheric plasma jet (CAPJ) featuring guided streamers operating with helium (He) or argon (Ar) at a frequency of about 1 MHz are presented. The surrounding gas of the CAPJ is controlled using a shielding gas device, which allows creating a defined gas curtain around the plume of the jet. Shielding gas nitrogen (N_2 purity 99.999 %) and oxygen (O_2 purity 99.995 %) in varying ratios with water admixtures was applied.

3. Particle and Discharge Dynamics

Reactive species can be transported to the liquid [10,11], can be generated by radiation impinging on the liquid surface or can be generated in secondary reactions in the liquid phase [12].

Especially the surrounding atmosphere influences the species composition and excitation dynamics drastically. Using helium as feed gas, metastable helium species play a dominant role in energy dissipation. In case of the gas curtain containing no oxygen, metastable $He(2^3S_1)$ species are not to be found outside the jet discharge zone [13]. A dominant role can be attributed to negative oxygen ions. Apart from forming a storage for negative charges over one excitation period [14], they might be one responsible factor for actually guiding the streamer inside a noble gas channel surrounded by ambient air impurities [9].

For an energy transport and dissemination, metastable species play a major role in noble gas atmospheric pressure plasma jets.

Diagnostics of these species is intricate, especially in the case of filamentary argon plasma jets. The filaments are arbitrarily traveling through the gas stream [9]. A tuneable diode laser absorption spectroscopy measurement reveals this by absorption peaks attributed to Ar($1s_5$) metastables traveling through the probing laser beam (Fig. **2**).

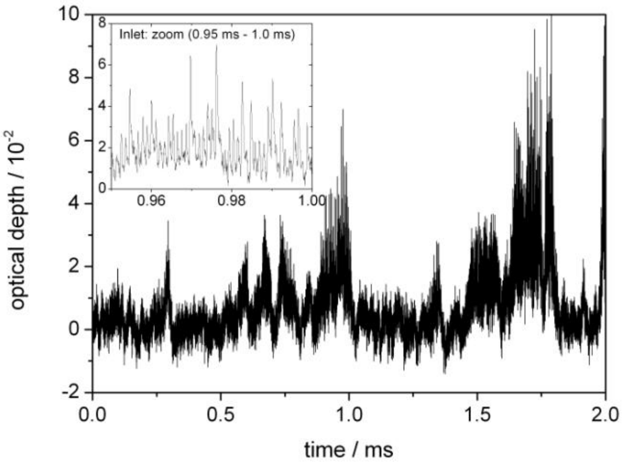

Fig. 2: Ar($1s_5$) temporal optical depth measured at a wavelength of 811.5 nm by tuneable AOM laser absorption spectroscopy in the effluent of an atmospheric pressure argon plasma jet

2. Liquid interaction

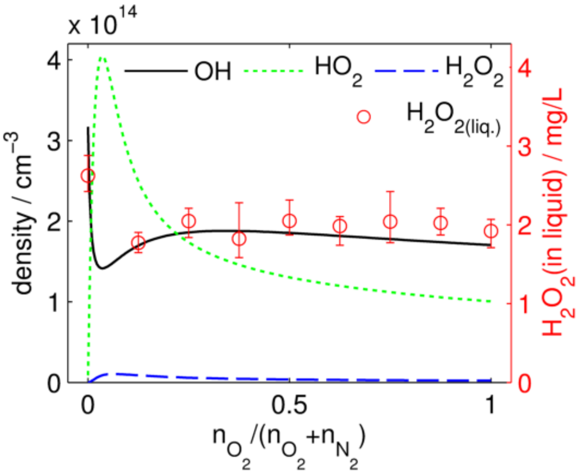

Fig. 3: H_2O_2 concentration measured in plasma-treated liquid and densities of OH, HO_2 and H_2O_2 obtained from a kinetic model for shielding gas compositions ranging from pure N_2 to pure O_2.

It is observed that the gas-phase $^\bullet$OH densities expected from kinetic modelling correlate with the H_2O_2 densities measured in plasma-treated liquids by a commonly used colorimetric assay as shown in Fig. **3**. Hydrogen peroxide, as an example for reactive oxygen species relevant for mammalian systems, detected in the liquids can arise from different origins. One source will be the directly in the gas phase formed H_2O_2, which can be increased by raising the working gas humidity and be dissolved in the solution. Also CAPJ's vacuum ultraviolet (VUV) radiation will contribute to hydrogen peroxide concentration since VUV will dissociate water molecules to form hydroxyl radicals, which quickly recombine to hydrogen peroxide.

4. Conclusion

While equilibrium chemistry can explain many RONS generation and transfer processes, non-equilibrium processes involving electrons, metastables, and radiation contribute significantly to reactive species production in plasma medicine.

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