Impact of humidity on the OH distribution in the effluent of an atmospheric pressure plasma jet measured by laser induced fluorescence

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Abstract: To determine the influence of environmental humidity on reactive species, we measured the 2D-distribution of hydroxyl (OH) produced in the effluent of the COST-Jet by laser induced fluorescence. All experiments were performed inside a closed vessel to control humidity of the ambient atmosphere. We compared the measured OH profiles for different water contents in the ambient environment as well as for different water contents in the feed gas to obtain information about the production channels of OH.

Keywords: atmospheric pressure plasma jet, OH, hydroxyl radical, laser induced fluorescence.

1. Introduction

Atmospheric pressure plasma sources are a valuable tool in applications such as materials processing or plasma medicine as they provide a plethora of reactive species while the gas temperature remains close to room temperature. For plasma sources operating in ambient atmosphere, the environmental conditions have a sensitive impact on the reactive species leaving the discharge zone. These effects strongly depend on the type of plasma jet: If the electric field is parallel to the gas flow and reaches into the ambient atmosphere, part of the reactive species is formed in the effluent of the plasma discharge [1]. In contrast, in plasma sources exhibiting an electric field perpendicular to the gas flow (cross-field), reactive species are almost exclusively produced in the plasma zone [2]. Using such a cross-field configuration, the exciting plasma zone can be clearly distinguished from the de-exciting afterglow (effluent). One important parameter is humidity: Water impurities in the feed gas or diffusion of moisture from the ambient atmosphere in the gas stream can contribute to an increase of humidity in the effluent and to the formation of reactive species such as hydrogen peroxide which is relevant in plasma-medical context. To study these effects, OH as a side-product from the dissociation of water by the plasma can be used as a tracer molecule. It is an important precursor for chemical reactions, e.g. for hydrogen peroxide production. We used laser induced fluorescence to measure absolute densities of OH depending on the humidity content of the ambient environment as well as the feed gas. By systematically varying the water concentration of the gas inside the vessel by a bubbler system, the influence of humidity on the OH density profile was studied. These results were then compared to profiles, which were obtained from the variation of humidity in the feed gas providing valuable information about the production channels of OH.

2. Experimental Setup

The plasma source used in this work is the COST-Jet [1]. It is a capacitively coupled plasma, which is ignited by RF-excitation between two plane-parallel electrodes enclosed in quartz panes. The homogeneous plasma forms in a discharge channel with a volume of 1x1x30 mm^3. The feed gas consists of 1 slm helium. The helium is enriched with humidity by passing it through a bubbler system. The whole setup is encased in a vacuum chamber to provide stable environmental conditions and control the relative humidity at atmospheric pressure.

OH is detected using laser induced fluorescence (LIF). A schematic of the setup is illustrated ini Figure 1. We use a frequency-doubled Nd:YAG laser coupled to a dye laser producing ns pulses at 283 nm. Fluorescence is detected using a fiber-coupled photomultiplier at 309 nm to measure lifetimes and fluorenscence intensity. Absolute values were obtained by calibrating the setup using Rayleigh scattering and taking into account quenching by impurities as described in literature [4].

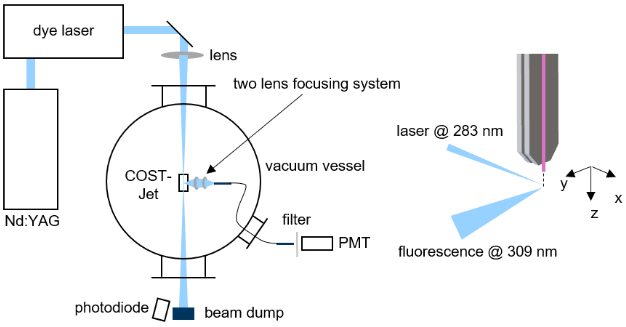

Fig. 1. Schematic illustration of the laser setup and the vacuum chamber (left), as well as the beam and detection setup in the effluent of the COST-Jet (right).

3. Results

To estimate the influence of surrounding humidity on the production of OH, we first performed a measurement at ambient atmosphere (relative humiditiy at 22 %) with no additional humidity admixed to the feed gas. Figure 2 shows the LIF signal of the effluent of the COST-Jet. In this case, the COST-Jet is pointed downwards. The upper edge of the image is measured at a distance of 1.47 mm from the point where the effluent is leaving the discharge configuration. Surprisingly, the graph shows that the OH density is lowest in the very center of the effluent. When measuring further away from the center of the effluent, the signal first increases to a maximum and then decreases again. Overall, the density is maximum where the ambient atmosphere mixes into the effluent. The results did not meet our original expectations: We expected a maximum density in the center of the effluent due to dissociation of humidity impurities in the plasma discharge. This is observed when humidity is intentionally admixed to the feed gas (not shown here). However, these results suggest another production channel of OH from ambient humidity and independent of charge carriers as these are confined to the discharge channel due the cross-field configuration.

This is also underlined by the decreasing lifetimes of OH further away from the center of the effluent due to quenching (not shown here).

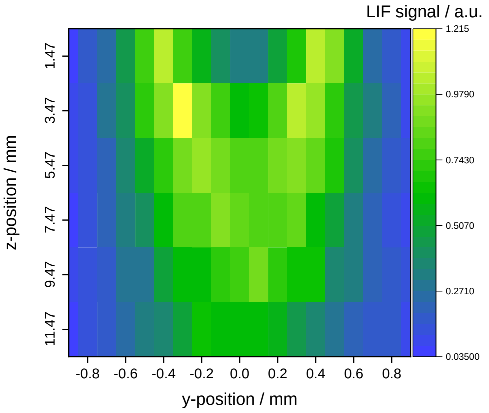

Fig. 2. Relative OH density in the effluent of an atmospheric pressure plasma jet in ambient conditions at a relative humidity of 22 %.

In order to understand the production pathways, we systematically varied the humidity content in the ambient atmosphere. Figure 3 shows perpendicular profiles through 2D-maps as shown in Figure 2 for different humidity contents in the ambient atmosphere around the effluent. The overall LIF signal increases, indicating a higher OH density with higher humidity in ambient atmosphere. Additionally, the distance between the two maximum peaks is decreased indicating that the overall OH maximum is moving towards the center of the effluent. This observation might be due to a higher amount of water diffusing into the effluent which is subsequently dissociated into OH and H. The mechanism of production is not fully understood yet. The OH production and destruction processes inside the plasma discharge are well known [5]: OH is mostly produced by electron impact dissociation of water molecules. Subsequently, OH is consumed by reactions with OH to form H_2O_2, HO_2 or H_2O. However, in our case, electron impact processes can be excluded as the charge carriers are confined to the plasma channel due to the cross-field configuration of the plasma source. A second pathway is dissociation by Penning reactions with excited metastable helium atoms and molecules via formation of H_2O^+ and subsequent reaction with water [6]. However, the lifetime of helium metastables in atmospheric pressure plasma jets is rather short due to quenching [7]. Considering the reduced lifetime of ~2µs, the extension of the OH into the ambient atmosphere seems to be too strong. In the future, metastable measurements in the effluent could confirm or reject this hypothesis.

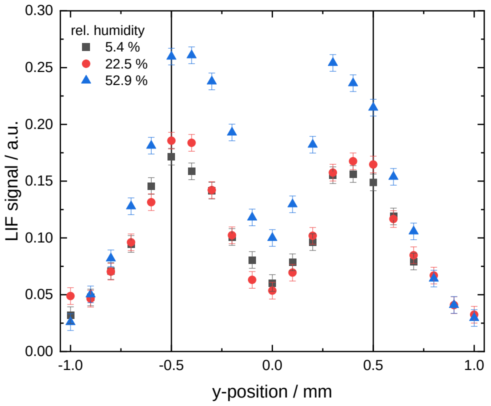

Fig. 3. Relative OH density profile in the effluent (z-position = 1.47 mm) of an atmospheric pressure plasma jet in ambient conditions different relative humidity.

4. Conclusion

In this work, we used LIF diagnostics to measure the spatial distribution of OH radicals in the effluent of an atmospheric pressure plasma jet with a cross-field configuration operated in helium. To estimate the influence of humidity in the ambient atmosphere on the plasma chemistry in the effluent, we varied the humidity of the ambient atmosphere using a closed vessel. Surprisingly, our measurements revealed that the OH density in the effluent increases slightly with increasing amounts of humidity despite the use of a cross-field configuration. This might be due to water diffusing into the effluent. The generation process is most likely due to excitation by helium metastables via production of H_2O^+. However, this hypothesis still has to be confirmed in the future.

5. Acknowledgements

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6. References

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