

OH Kinetics in a Shielded Atmospheric Pressure Plasma Jet

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Gas phase non-equilibrium plasmas containing water vapor are of growing interest for many environmental and biomedical applications. Nonetheless the plasma kinetics that is of the utmost importance for these applications remain poorly understood [1]. A He-H₂O reaction mechanism has been compiled and validated for a diffuse atmospheric pressure glow discharge [2]. Models of the more complicated plasma kinetics in high electron density filamentary water containing discharges that are ubiquitous in many applications have currently not been investigated in detail.

In this work, we present a laser induced fluorescence (LIF) study of the OH density in an atmospheric pressure nanosecond pulsed Ar + 0.26% H₂O plasma jet. The plasma jet operates into ambient air but is shielded with a coaxial flow of argon to reduce the diffusion of air into the effluent of the jet. The jet impinges on a metal plate electrode connected to ground through a 50 kΩ resistor to limit the plasma current. The plasma is generated by a 200 ns pulsed voltage with an amplitude of 4.7 kV at a frequency of 5 kHz. A stable plasma filament is formed between the needle electrode in the jet and the metal plate allowing for spatially and time resolved laser diagnostics. The gas temperature (T_g) and electron density are measured by Rayleigh scattering and Stark broadening of hydrogen lines respectively. Electron densities in excess of 10^{22} m⁻³ have been measured. The gas temperature does not exceed 550 K in the core of the plasma.

The strong variation in the time dependent relative OH density as a function of radial distance as obtained by LIF is shown in Fig. 1. This spatial variation in OH kinetics correlates with an increased air and reduced water vapor concentration at a radial position of 1.1 mm compared to 0.4 mm as found by an computational fluid dynamics calculations. The reduced lifetime of OH (~ 10 μs) at 0.4 mm is due to the large density of H atoms in the core of the discharge. In addition the larger concentration of O₂ compared to water at 1.1 mm leads to an OH recycling mechanism as shown in Ref. [3] that can increase the effective lifetime of OH to a few hundred μs.

These results illustrate the sensitivity of ambient gas composition in atmospheric pressure plasma jets on the density and kinetics of reactive species that are important for biomedical applications.

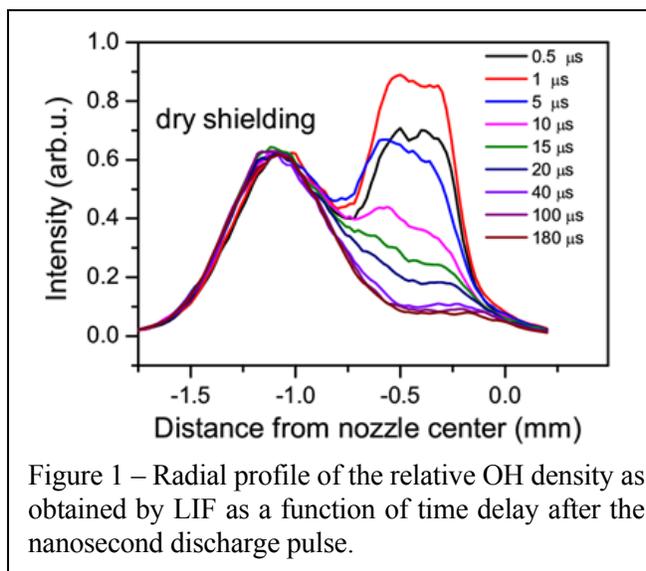


Figure 1 – Radial profile of the relative OH density as obtained by LIF as a function of time delay after the nanosecond discharge pulse.

References

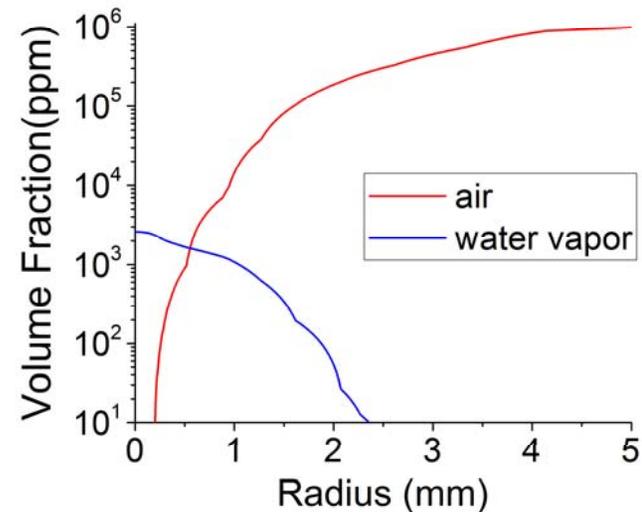
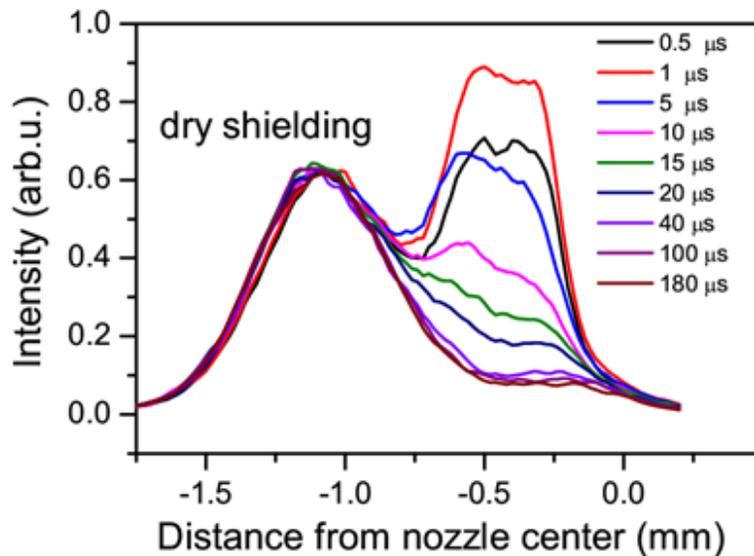
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Highlight

OH KINETICS IN A SHIELDED ATMOSPHERIC PRESSURE PLASMA JET

- OH densities have been measured by laser induced fluorescence in an argon shielded Ar+0.26% H₂O atmospheric pressure plasma jet.
- Strong variation in the time dependent relative OH density is observed as a function of radial distance correlating with air and water vapor concentrations.
- Reduced lifetime of OH (~10 μs) in the core is due to a large density of atomic hydrogen. The constant OH density at 1.1 mm is due to the abundance of O₂ at this location leading to an OH recycling mechanism.



- OH LIF signal (relative density)
- Calculated air and water concentrations

HIGHLIGHT

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Near-Surface Plasma-Water Interactions

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A transient spark discharge in air adjacent to water demonstrates the importance of near-surface reactions in decomposition of indigo carmine dye. Major stable species that are formed in water under these conditions include H_2O_2 , NO_3^- and nitrite NO_2^- . NO_2^- and H_2O_2 are known to react under acidic conditions to form $\text{O}=\text{NOOH}$ which decomposes rapidly to form $\text{OH}\cdot$ radicals, thus decomposing the dye through the bulk of the liquid. By contrast, $\text{OH}\cdot$ arriving from the surface react with indigo carmine in a depth of ~ 10 - 100 nm from the surface, based on comparison of reaction and diffusion rates. A combination of experiments and simple models demonstrate the key role of near-surface reactions compared to bulk reactions. [1]

In a separate study of electrons entering water (acting as anode to a dc discharge in argon), simulation showed that generation of near-surface OH^- following electron-water decomposition in the presence of bulk acid creates a highly basic region very near the surface. In the presence of bulk solution acidity, pH can vary from 2 away from the surface to a 11 over a distance of ≈ 200 nm, as shown in Fig. 1. [2]

In another set of simulations coupling a dc argon discharge with a water solution, we tested the effects of varying surface loss coefficient on gas phase interfacial electrons. The gas phase electron density at the anode can vary by orders of magnitude depending on the assumed value of the water surface recombination probability. We conclude that in order to make more confident model predictions about plasma-liquid systems, finer scale simulations and/or new experimental techniques must be used to elucidate the near-surface gas phase electron dynamics.[3]

References

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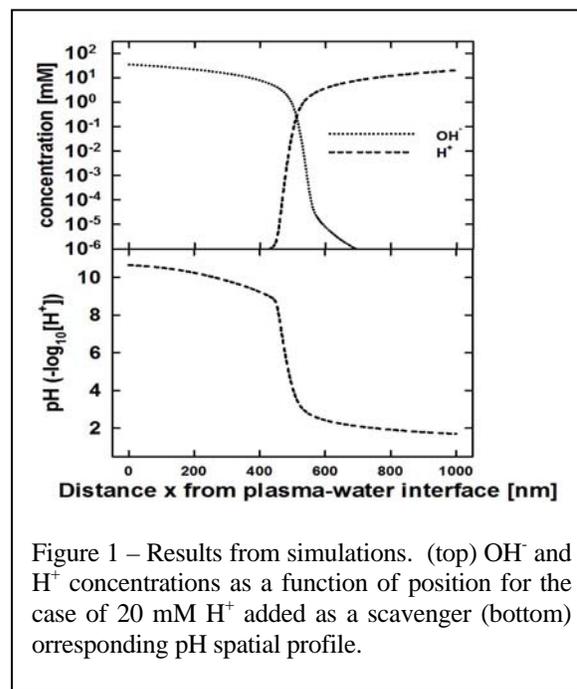
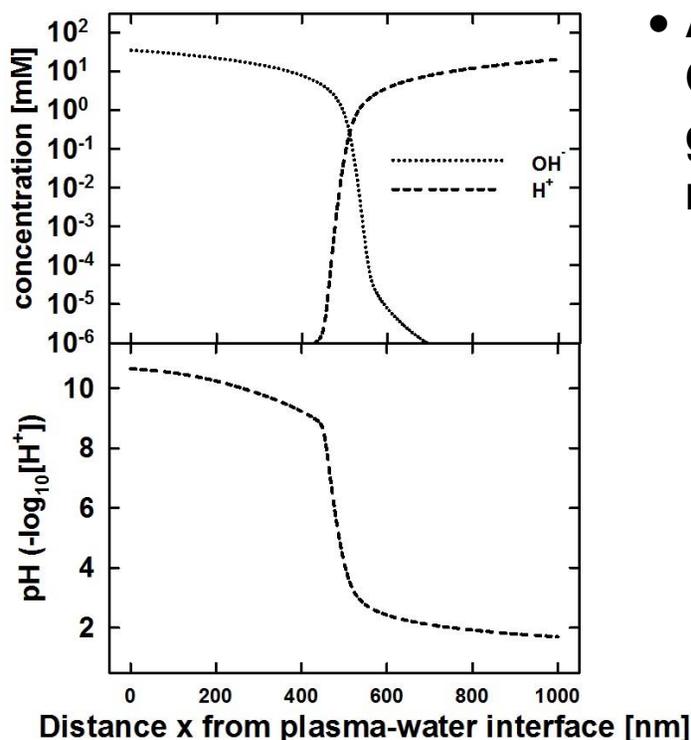


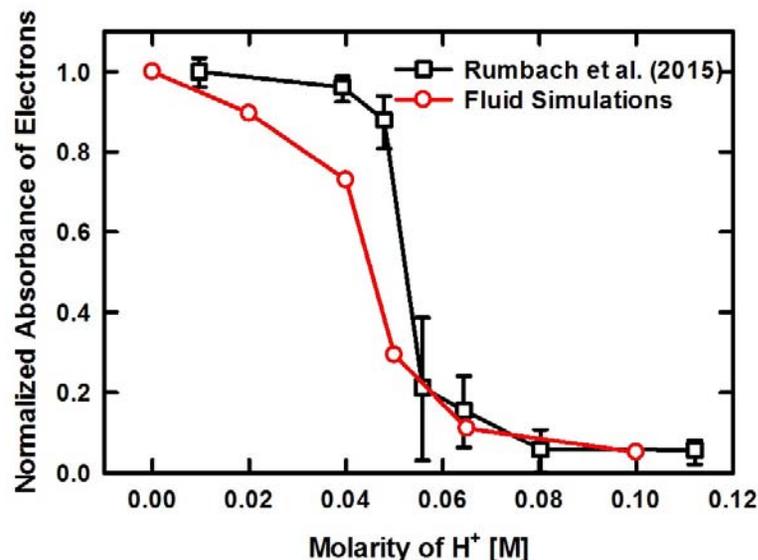
Figure 1 – Results from simulations. (top) OH^- and H^+ concentrations as a function of position for the case of 20 mM H^+ added as a scavenger (bottom) corresponding pH spatial profile.

PLASMA-WATER INTERACTIONS: SCALES MATTER!

- Simulations and experiments of plasma-water interactions show that reactive species can interact over a wide range of length scales from nm to mm.
- When scavengers are used in plasma-activated-water PAW, reactions near surface can dominate.



- As shown here, acid scavenger H^+ reacts with OH^- from e^-_{aq} , creating strong near-surface gradients, complicating the interpretation of the results.



- Spatial profile of pH near plasma-water interface.

- Model-experiment comparison in acid scavenging e^-_{aq}

