

Particle-in-cell Simulations of Atmospheric Pressure He/H₂O Discharges

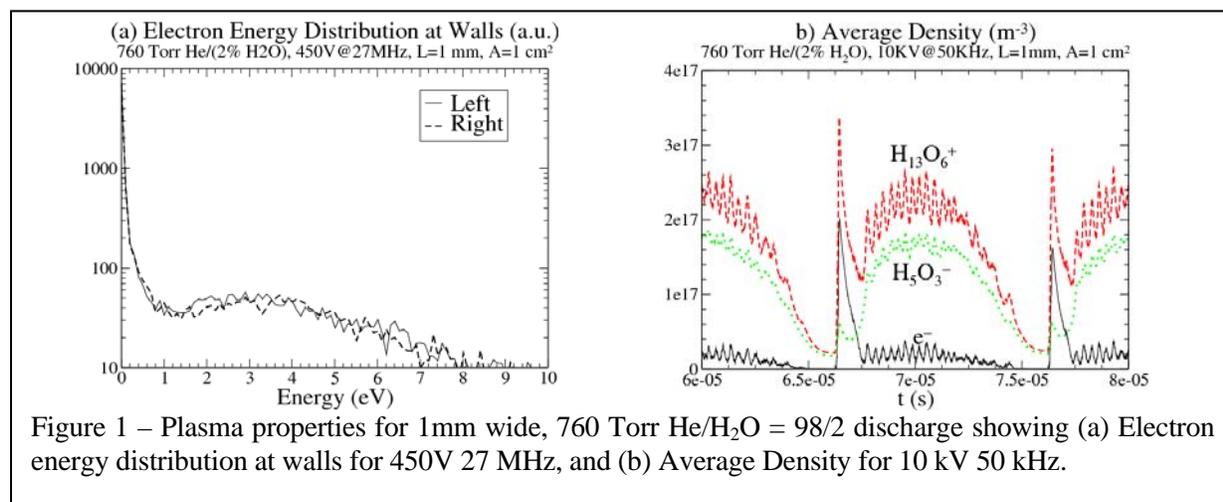
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Atmospheric pressure micro-discharges in contact with liquid surfaces are of increasing interest, especially in the bio-medical field. We conducted 1D3v Particle-in-cell (PIC) simulations of a voltage-driven 1 mm wide atmospheric pressure He/H₂O = 98/2 plasma discharge in series with a 0.5 mm thick liquid H₂O layer and a 1mm thick quartz dielectric layer. We chose a 2% H₂O mixture as it is the approximate saturation level of water vapor in air at room temperature (300 K). The H₂O and dielectric layers are modeled as a capacitor with $C_T \approx 3.46$ pF being equal to the series combination of $C_{H_2O} = \kappa_{H_2O} \epsilon_0 A / d_{H_2O}$ and $C_D = \kappa_D \epsilon_0 A / d_D$, where $\kappa_{H_2O} = 80$, $d_{H_2O} = 0.5$ mm, $\kappa_D = 4$, $d_D = 1$ mm, and electrode area $A = 1$ cm². A previously developed two-temperature hybrid global model of atmospheric pressure He/H₂O discharges [1] was used to determine the most important species and collisional reactions to use in the PIC simulations. We found that the H₁₃O₆⁺, H₅O₃⁻, and electrons were the most prominent charged species while the metastable helium density was found to be negligible as most of the He* were quenched via Penning ionization. So, the PIC simulations followed the H₁₃O₆⁺, H₅O₃⁻, and electron species' trajectories amid a uniform constant temperature neutral background of atmospheric pressure He with 2% H₂O trace gas. The ion-induced secondary emission coefficient γ_1 was assumed to be 0.15 at all surfaces. A series of simulations were conducted at 27 MHz with $V_{rf} = 350$ -600 V corresponding to $J_{rf} \approx 500$ -2100 A/m². As shown, in Fig. 1a, we find that the H₂O rotational and vibrational excitation losses are so high that electrons reach the walls at thermal temperatures. We also simulated a much lower frequency case of 50 kHz with $V_{rf} = 10$ kV. In this case, the discharge runs in a pure time-varying γ -mode, and as shown in Fig. 1b, the average density of electrons in the discharge peaks when the positive ions hit the walls and induce secondary electron emission and then rapidly dissipates.

References

[1] Ke Ding, M.A. Lieberman and A.J. Lichtenberg, J. Phys. D: Appl. Phys. **47**, 305203 (2014).



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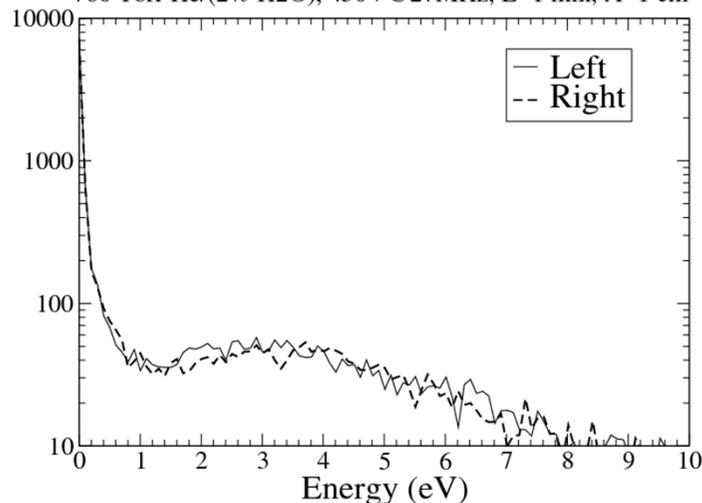
Highlight



PIC SIMULATIONS of ATMOSPHERIC PRESSURE He/H₂O DISCHARGES

- 1d3v PIC simulations were performed of voltage-driven 760 Torr, He/H₂O=98/2, 1 mm discharges in series with a 0.5 mm H₂O layer and 1 mm quartz substrate.
- The H₂O rotational and vibrational losses are high, leading to thermal electrons at discharge walls.
- For a low frequency 50 kHz with $V_{rf} = 10$ kV, the discharge is in a pure time-varying γ -mode and maintained only by secondary electron emission at walls.

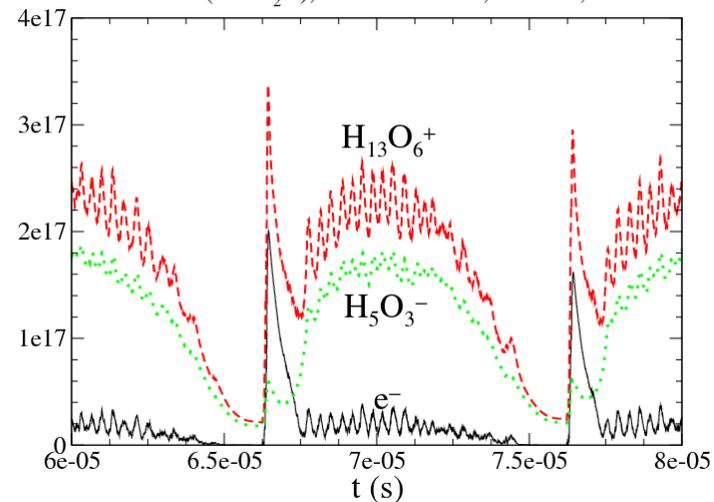
(a) Electron Energy Distribution at Walls (a.u.)
760 Torr He/(2% H₂O), 450V@27MHz, L=1 mm, A=1 cm²



- Wall EEDs for 450 V, 27 MHz

(b) Average Density (m⁻³)

760 Torr He/(2% H₂O), 10KV@50KHz, L=1mm, A=1 cm²



- Average Density for 10 kV, 50KHz

NO₃ Formation on Biomolecular and Polymeric Surfaces after Treatment by Cold Atmospheric Plasma

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Cold atmospheric plasma (CAP) sources are used for a variety of promising applications including thin film deposition, surface decontamination of living and non-living surfaces, and cancer treatment. CAP sources produce many reactive species including O₃, NO_x, and atomic O. Many different plasma sources are being investigated, each generating different amounts of reactive species. The reaction pathways for the formation of these species in the gas phase are also being investigated. At the same time, little knowledge on specific atomistic surface-chemical changes is available.

We have previously reported the formation of surface-bound NO₃ on lipopolysaccharide (LPS), an immune-stimulating molecule, following treatment by an atmospheric pressure plasma jet (APPJ) [1]. As LPS contains both nitrogen and oxygen, it was unclear whether this species originated from nitrogen and oxygen already found in LPS or if it was generated in the gas phase. To study this process, we treated a variety of polymer films and found that NO₃ is present on all surfaces despite the fact that these films have very different molecular structures and chemical compositions. This result indicates that surface-bound NO₃ is generic to a wide variety of organic films after CAP treatment. Furthermore, as films containing neither oxygen nor nitrogen show NO₃ coverage (e.g. polystyrene and polypropylene), this species must originate from the gas phase. While the surface-bound NO₃ did not vary much among the different materials after APPJ treatment, the change in the oxygen composition greatly varied. For example, polystyrene is easily oxidized, but polypropylene and poly(methyl methacryle) strongly resist oxidation. This type of

analysis demonstrates for the selectivity of the CAP treatment to different chemical moieties.

In addition, we confirmed the same effect after treatment by two very different sources, the APPJ, which operates with an Ar carrier gas and a surface microdischarge operating with N₂/O₂ mixtures. Thus, NO₃ formation appears to be a generic chemical modification of materials by CAP sources. By comparing results from surface chemical analysis with immunoassays, we found a possible correlation between the biodeactivation of LPS and formation of NO₃ on the LPS surface (see Fig. 1). The highest biodeactivation occurs when NO₃ is highest, not when oxidation resulting from other species is highest.

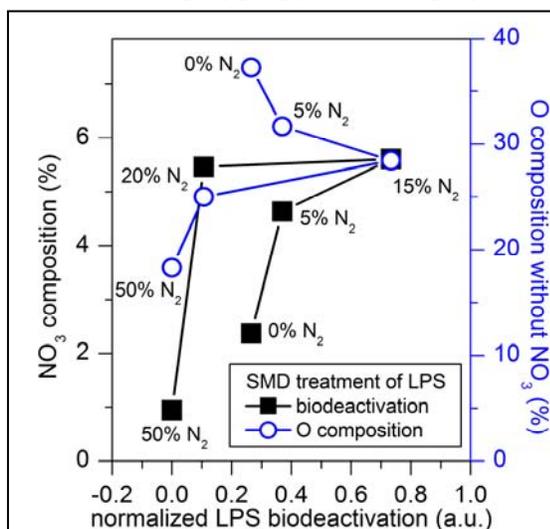


Figure 1 – Surface microdischarges cause the most biodeactivation of LPS when surface-bound NO₃ is highest, not C-O_x.

References

[1] E.A.J. Bartis *et al.* J. Phys. D: Appl. Phys. 46, 312002 (2013)

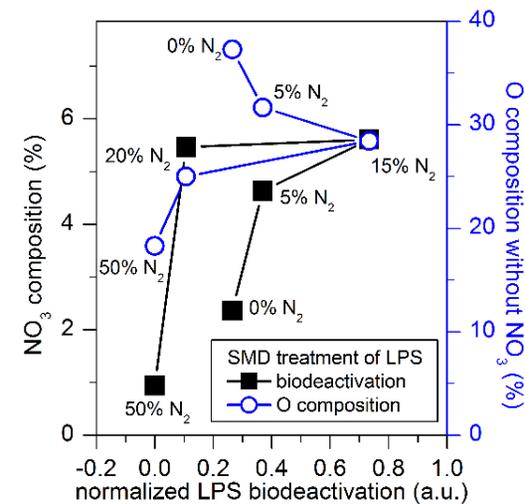
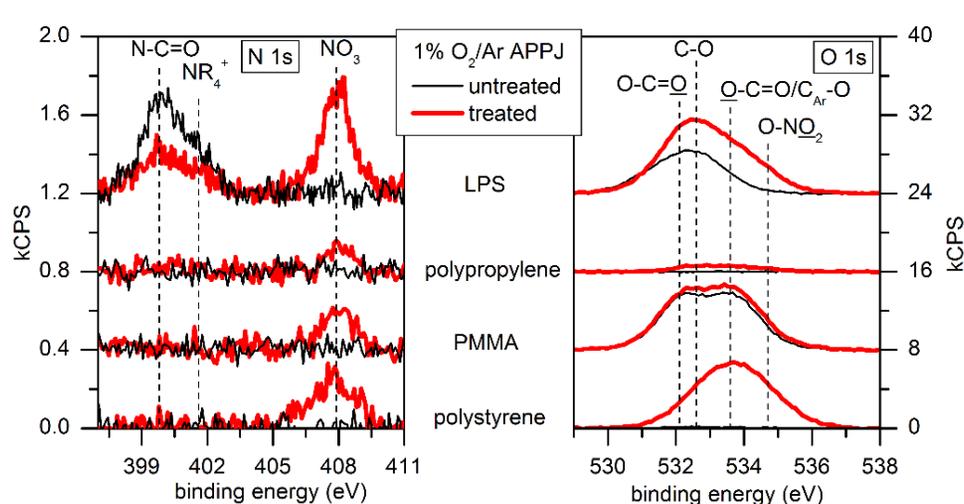
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NO₃ ON BIOMOLECULAR AND POLYMERIC FILMS AFTER COLD ATMOSPHERIC PLASMA TREATMENT

- X-ray photoelectron spectroscopy and enzyme-linked immunosorbent assay characterized atm. pressure plasma treated biomolecular and polymeric films.
- NO₃, forms on various model polymers, including films without oxygen or nitrogen, indicating that the species originates from the gas phase.
- Maximum biodeactivation of lipopolysaccharide occurs when NO₃ coverage is highest, not when oxidation is highest. NO_x is important for many biological processes. The current finding may be important for plasma medicine.



- APPJ-treated films show NO₃ and oxidation on different films

- Biodeactivation correlates with oxidation based on NO₃ rather than other C-O_x species