

Free Radical Chemistry of a Plasma-water Interface

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Abstract

Low-temperature plasma sources can be used as an alternative to radiation for driving free radical chemistry in water. Recent applications include nanomaterial and chemical synthesis, wastewater treatment, and medical treatments. The exact chemical pathways in these systems have proven difficult to measure, as most of the free radical chemistry occurs within ~ 10 nm of the plasma-liquid interface. To solve this, we developed a highly precise optical absorption technique to measure solvated electrons in the interfacial region [1], as shown in Figure 1. For a negatively biased DC plasma, free gaseous electrons are driven into the solution where they solvate and diffuse up to 100 nm into the solution with an interfacial concentration ~ 0.1 mM. Our results also indicate that they obey the same reaction kinetics as bulk solvated electrons, recombining to form $\text{OH}^-_{(\text{aq})}$ or reacting with scavengers like nitrate.

Our current work is focused on the opposite DC polarity, where positive gas ions impact the liquid with close to 100 eV of energy and ionize the liquid water molecules. This configuration—often referred to as an “electrolyte cathode glow discharge” (ELCAD)—yields complex chemistry with H_3O^+ , $\text{OH}_{(\text{aq})}$, $\text{H}_{(\text{aq})}$, and $\text{e}^-_{(\text{aq})}$ all produced simultaneously at the interface. Despite several decades of study by many researchers, there are a number of open questions regarding the interfacial chemistry of this system. For example, free electrons necessarily must escape from the liquid surface into the gas phase to sustain the plasma, but there is no consensus on the mechanism responsible for this process. Additionally, there is little quantitative information about the ionization process at the liquid surface, such as the penetration depth and free radical yields of the ~ 100 eV ions. This presentation will overview our efforts to identify and quantify the radical chemistry at the plasma-liquid interface and highlight opportunities to utilize these systems for various applications.

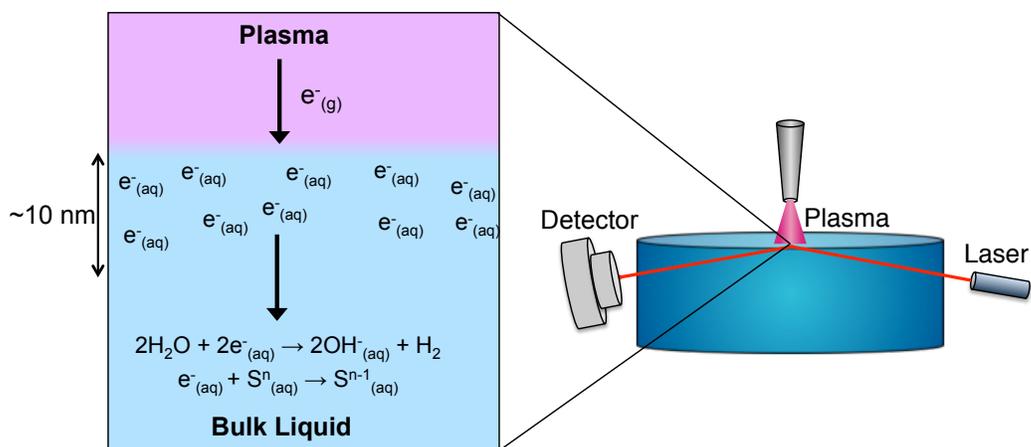


Figure 1. (Left) Gas phase electrons in a low-temperature plasma solvate into an aqueous solution and quickly react in the interfacial region. (Right) A total internal reflection absorption spectroscopy (TIRAS) technique is used to directly observe solvated electrons in the interfacial region.

References

- (1) Rumbach, P., Bartels, D. M., Sankaran, R. M., & Go, D. B., The solvation of electrons by an atmospheric-pressure plasma. *Nature Communications*, **2015**, 6, 1-7.