

Optical Diagnostics of Plasma-Liquid Interactions

Abstract

Plasma–liquid interactions give rise to chemically enriched solutions known as plasma-activated water (PAW), primarily through the transfer of reactive oxygen and nitrogen species (RONS) from the gas phase into the liquid. PAW holds considerable applications across sectors such as biomedicine, agriculture, and food safety. Beyond PAW generation, plasma–liquid interactions are also critical in direct plasma applications, including wastewater treatment and biomedical interventions, where plasma interfaces with biological fluids and tissues. These complex systems involve concurrent physical and chemical processes, making it essential to understand the reaction pathways, species stability, and strategies for tailoring PAW composition for specific applications. Self-pulsing direct current (DC) transient spark (TS) discharge, employed in a pin-to-plate configuration, served as the cold plasma source. A key feature of TS is its ability to simultaneously generate electrospray (ES) microdroplets via the same high-voltage (HV) nozzle, significantly enhancing the gas–liquid interface and facilitating efficient RONS transfer. The study examined both direct and indirect TS–ES interaction modes using gases such as N_2 , O_2 , and synthetic air (dry and humidified), each yielding PAW with distinct chemistries. Direct TS–ES contact produced PAW enriched in H_2O_2 (aq) and NO_2^- (aq), while indirect interactions favored the uptake of gas-phase species like NO , NO_2 , and HNO_2 , resulting in PAW dominated by NO_2^- (aq). Further experiments combined TS-generated NO_x species with O_3 to synthesize reactive nitrogen species (RNS) like N_2O_5 and HNO_3 . Gas-phase products were analyzed via FTIR spectroscopy, while their dissolution and conversion to aqueous NO_3^- (aq) and NO_2^- (aq) were tracked using in-situ UV/VIS spectroscopy. Varying the NO/O_3 ratio enabled deeper insights into plasma-assisted nitrogen fixation and control over RNS formation. Overall, this work identifies critical reaction pathways and demonstrates the tunability of PAW chemistry based on discharge configuration and gas composition. These findings support the targeted application of PAW in diverse fields and contribute to the development and scale-up of TS-based plasma systems for efficient and sustainable nitrogen fixation.