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Investigation of the liquid velocity gradient induced by a plasma dielectric barrier discharge and its impact on reactive species generation

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In the applications of atmospheric pressure plasma interacting with a liquid phase, ensuring optimal plasma efficiency relies significantly on the effective transport of reactive species from the gas to the liquid and within the liquid itself. In the absence of external forces, and considering the relatively slow diffusion of these species inside the liquid, their movement is predominantly governed by the plasma-induced liquid flow. Depending on input parameters that affect the net force exerted on the liquid phase, two distinct plasma-induced liquid flows can be observed: 1) a linear downward flow occurring along the plasma discharge axis [1], and 2) an upward flow accompanied by the formation of vortices on both sides of the plasma discharge axis [2].

Several potential driving forces contribute to the generation of liquid flow, such as thermal instabilities arising from localized heating at the plasma-liquid interface, electrohydrodynamic (EHD) forces, mechanical coupling with the gas flow, pressure waves as well as the influence of the electric field and electrical surface stresses. Dickenson *et al.* [3] used a 2D-axisymmetric model and validated it through Particle Image Velocimetry (PIV) measurements to characterize the mechanical interaction between plasma and liquid in a pin-water electrode system. Their findings indicate that the driving mechanism for liquid flow is correlated with the charge relaxation time of the liquid. Interestingly, in their argon atmospheric pressure plasma jet (APPJ), Kawasaki *et al.* [1] demonstrate the possibility to switch the direction of the plasma-induced liquid flow simply by changing the frequency. Stancampiano *et al.* [2] investigated the origin of the vortex formation with a helium APPJ, and they conclude that the main causes are EHD forces and gas flow tangential components induced by the plasma jet.

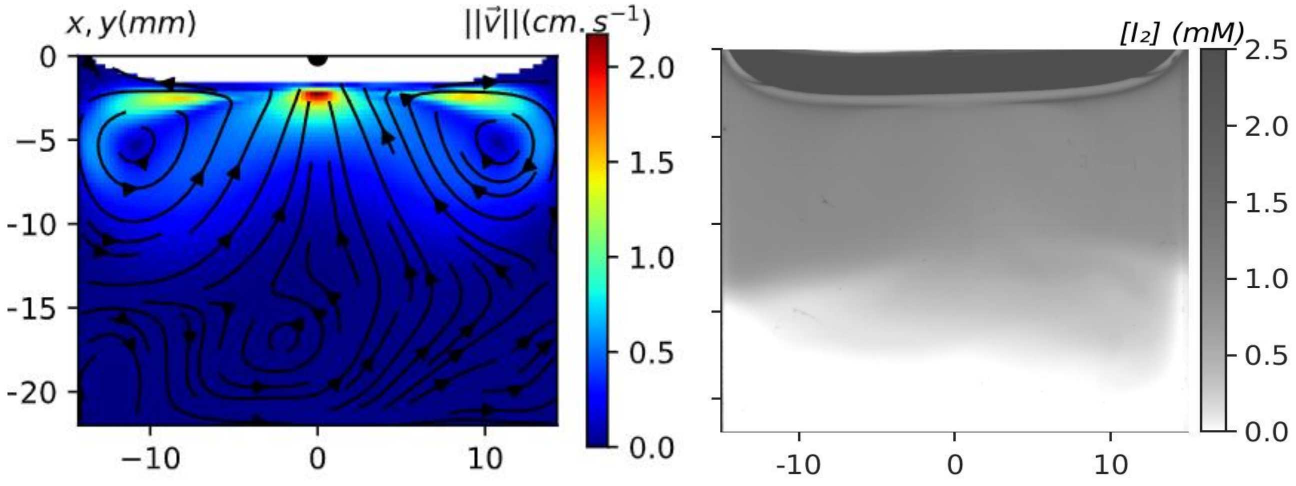
The present study aims to extend our understanding of this phenomenon. In our experimental setup, the plasma discharge is ignited at the tip of a tungsten needle with a curvature radius of 100 µm. The needle is placed above an optical glass tank, filled with 20 mL of the liquid to be treated. The distance between the tip and the liquid surface (the electrode gap) is varied between 2 and 8 mm using a micrometer screw. The tank is placed symmetrically on a piece of copper tape (90 × 50 mm^2), which serves as the grounded electrode. The needle is connected to a high voltage amplifier (Trek 20 kV, 20 mA), which amplifies a low sinusoidal voltage produced by a function generator. The voltage between the needle and ground (discharge voltage) is measured with a high voltage probe (LeCroy PPE, 20 kV, 100 MHz). Note that this voltage is the sum of the voltages across the electrode gap, the liquid phase and the dielectric (the reactor bottom). The current is obtained by measuring the voltage across a shunt resistor. All signals are recorded with a digital oscilloscope (HDO6054, 500 MHz, 5 GS/s).

PIV measurements were conducted during the discharge process to obtain the velocity vector fields in the liquid. For this purpose, 5-µm polyamide particles doped with rhodamine B were used as tracers. A 32-mJ pulsed Nd:YAG laser, operating at 532 nm with a pulse duration of 7 ns, was employed to create two successive laser sheets with an adjustable time delay. Concurrently, a high-resolution camera (Imager pro X 4M, 2048 × 2048 pixels) was utilized to record a single laser pulse in each frame. The

2D velocity vector field for each pair of camera frames was then computed through a cross-correlation procedure.

The initial phase of the study aimed to confirm the key role of liquid flow in the transport of reactive species. To observe the motion of reactive species, we employed a potassium iodide solution. In fact, iodide ions can react with various oxidant species, resulting in the production of iodine that has a color spectrum ranging from yellow to brown, depending on its concentration. Consequently, in situ local iodine concentrations were obtained through grey levels acquired using a black and white camera (Point Grey, FL3-U3-32S2M-CS, 1600 × 1200 pixels). Prior calibration ensured a correlation between grey levels and known iodine concentrations in prepared solutions. In parallel (in a separate experiment conducted under the same conditions), PIV measurements provided information on the flow associated with the discharge over the potassium iodide solution.

As an illustration of the obtained results, Figure 1a displays the mean velocity vector field corresponding to a 5-minute plasma treatment of the potassium iodide solution, while Figure 1b presents the mean grey level image under identical experimental conditions. The formation of two vortices is observed on either side of the discharge axis, extending to a depth of approximately 10 mm below the interface (Fig.1a). Moreover, there is a significant velocity gradient between the plasma/liquid interface and the bulk liquid. The velocity is maximum at the plasma/liquid contact point, reaching 2 cm/s. A notable similarity exists between the higher velocity zones (Fig. 1a) and the regions with elevated iodine concentration (Fig. 1b), highlighting the potential to optimize the generation of reactive species and, consequently, the efficiency of plasma discharge, simply by controlling transport phenomena. To achieve this control, in the second phase of our study, we examined several parameters independently, including frequency, voltage, gap distance (with pure water as the liquid for these three variables), surface tension (using mixtures of ethanol and pure water with varying ethanol percentages), conductivity (using potassium chloride solutions at different concentrations) and discharge duration. The influence of all these input parameters will be discussed during the oral presentation.



a) b)

Fig 1: a) Mean velocity vector field and b) iodine production during 5 minutes of plasma treatment. Gap = 2 mm, frequency = 2 kHz and voltage = 9 kV.

[1] T. Kawasaki et al. , Jpn. J. Appl. Phys. ***62*** *(2023) 060904.*

[2] A. Stancampiano et al., Plasma Sources Sci. Technol. ***30*** *(2021) 015002.*

[3] A. Dickenson et al., J. Appl. Phys. ***129*** *(2021) 213301.*