Atmospheric Plasmas Induce Electrolytic-Like Flows in Grounded Solutions

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Abstract: The modification of liquid flow induced during atmospheric plasma-water interactions when the liquid is changed from electrically floating to grounding (thus entering a regime of electrolysis) is described. A helium plasma jet is impinged upon a demineralised water target when a ground electrode is both absent from or submerged within the solution, and flow is recorded using particle image velocimetry. Modifications of the applied voltage and the salinity of the grounded solutions are made, and results are given a qualitative comparison to theory. An increase in upwards flow velocity was observed with both grounding and an increase of applied voltage, whereas downwards velocity increased in grounded solutions with increased salinity. This is believed to be due to the electrolysis exerting a net force on the flow, with the direction depending on the mobility coefficients of the ions present.

Keywords: Atmospheric Plasma, Plasma Jet, Liquid Flow, Plasma Electrolysis

1. Introduction

In the applications of atmospheric pressure plasmas where a liquid medium is present, the transmission of reactive species and ions is of paramount importance regarding the efficiency of the plasma usage.[1-3] Due to its governance in the transmission of solutes in liquids, furthering our understanding of which plasma and liquid parameters can affect the fluid flow could potentially lead to methods in controlling this transmission. This would then lead to adding a new modifiable parameter to these widely used systems.

One property that has not been focussed on in flow modification is the grounding of the system. This then changes our "simple" plasma-liquid interactions into plasma electrolysis[4]. The plasma impinged surface acts as an anode or cathode depending on the type of applied voltage, and the ground acts as the opposite. The transmission of positive and negative species within the liquid to the cathode and anode respectively will hypothetically induce a net force on the liquid[5,6], dependant on certain parameters of the ion/liquid system. This force should of course induce a velocity increase in one direction or the other within the flow, when compared to that when the liquid is electrically floating.

The authors compare the flow in both electrically floating and grounded liquids under impingement of a helium atmospheric pressure plasma jet (APPJ). Both the applied plasma voltage and the salinity of the solution are varied, to expand upon observations of how different electrolytic properties manipulate the flow's intensity and direction. A qualitative comparison to theory is then made.

2. Experimental Setup

The pH at the surface was measured to confirm if electrolysis is occurring using a "Mettler Toledo SevenExcellence pH/Cond meter S470". Following this, liquid phase flow was observed using particle image velocimetry (PIV)[7]. A schematic of the experimental setup can be observed in figure 1.

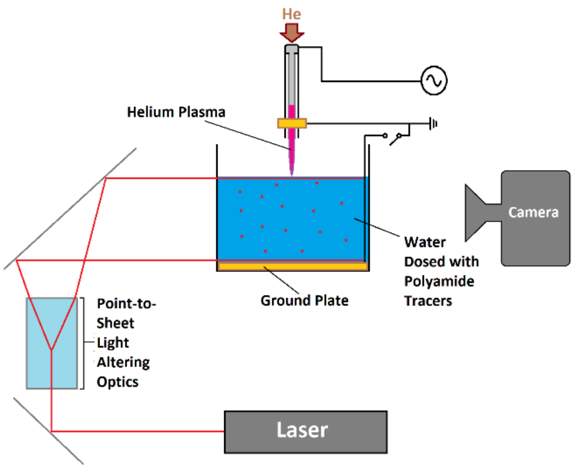

Fig. 1. PIV Experimental Setup

A 1mW red laser beam propagates through optics to generate a light sheet illuminating ~55µm polyamide tracers suspended in demineralised water based solutions. A 50mm square recording window is recorded using a "FastCam Mini AX100" at 60fps for 30 second windows during each experiment.

The APPJ is positioned 1cm above the liquid surface. Helium gas is supplied through at 1slm and formed into a plasma using an AC power supply at a frequency of 30kHz. During grounded experiments, a flat copper plate connected to wiring that travels to ground is added. In this configuration, the plasma-surface interaction zone will act as the anode and the copper plate should act as the cathode during plasma electrolysis.

PIV recordings were analysed using the open source MATLAB plugin PIVLab[8-10]. Recordings were time averaged and the vertical velocity with the largest magnitude was obtained – this would be the point in which the horizontal component would have the lowest influence over the PIV output and thus these were chosen as the data output for each experiment. Positive velocities are where the solution is flowing upwards, and negative velocities are where it's downwards. Experiments were repeated for improved accuracy when attempting to observe trends.

3. Results

As mentioned in the experimental setup section, clarifying if electrolysis is indeed occurring within the system was performed first. The pH was measured at the plasma-water interaction point, due to the increase of OH^- that would be present. An increase in pH at the anode was thought to not necessarily be observable however, as the plasma used is relatively weak to improve reproducibility for flow measuring. Surprisingly, an increase in pH was observed after 1 hour of 2kV plasma impingement, as shown in table 1.

Table 1. Surface pH following 1 hour of 2kV APPJ impingement

Electrical System Configuration	pH
Floating	6.178±0.002
Grounded	7.218±0.002

The results of PIV experiments where the applied voltage is varied during APPJ-demineralised water interactions can be observed in table 2.

Table. 2. Vertical liquid flow velocity underneath APPJ impingement for varying applied voltages where the system is electrically floating or grounded

Applied Plasma Voltage (kV)	Vertical Flow Velocities Under Plasma Jet (mms^{-1})	
	Floating	Grounded
0 (He Gas)	0.46±0.07	0.46±0.07
1.5	1.42±0.20	3.03±0.20
2.0	3.17±0.17	9.04±0.32
2.5	6.96±0.65	14.44±0.93

The flow velocity increased when the ground electrode is added and electrolysis can be expected to occur. During the ion liquid phase transmission, it can be expected that a net force would be added to the system that does not exist in the floating version, thus explaining the modified velocity. The electrohydrodynamic (EHD) force F induced on a fluid by a type of ion is given as[6]

$$F = \frac{Id}{k} \tag{1}$$

where I is the electric current transmitting through the fluid, d is the distance between the electrodes (in our experiments, d is taken as the depth of the liquid) and k is the mobility coefficient of the ion in question. k is obtained from [11] for both H^+ and OH^- due to their relevance in electrolysis and are displayed in table 3.

Table 3. H^+ and OH^- mobility coefficients in water

Ion	k ($10^{-8}\ m^2V^{-1}s^{-1}$)
H^+	36.23
OH^-	20.64

Due to the larger mobility coefficient of H^+ in comparison to OH^-, the resultant induced force would be in the upwards direction. This would of course cause a consistent increase in upwards flow velocity, as observed in the experiments.

To further solidify our hypothesis of electrolytic-driven flow modification, the salinity of the solution was increased. This addition of table salt dissolving and separating into both Na^+ and Cl^- will let us further understand the influence of ions on the resultant flow. This is especially true as the values of k for the salt ions[11] are the opposite to those in table 3, with the positive ions mobility instead being lower than the negative ions.

Table 4. Na^+ and Cl^- mobility coefficients in water

Ion	k ($10^{-8}\ m^2V^{-1}s^{-1}$)
Na^+	5.19
Cl^-	7.92

We can therefore expect the resultant flow to shift downwards the more salt is added. The salinity was increased from the previous 0 to 300gl^{-1}, and PIV measurements were taken for the 2kV plasma jet interacting with these solutions, as can be seen in table 5.

Table 5. Vertical liquid flow velocities underneath APPJ impingement for varying salinities where the system is electrically grounded

Salinity (gl^{-1})	Vertical Flow Velocities Under Plasma Jet (mms^{-1})
0	9.04±0.32
10	1.90±0.40
30	-0.18±0.80
50	-2.77±0.43
100	-3.20±0.91
200	-0.89±0.88
300	-3.63±0.54

The introduction of table salt gradually causes the flow direction to indeed shift downwards, as expected.

We must also consider the possibility that ions generated by the plasma in the gas phase before transitioning through the surface and into the liquid phase may also induce an EHD force onto the solution. Mass spectrometry results, however, revealed that the total count of ions generated by the plasma were lower than those of NaCl within the 10gl^{-1} salinity solution by a factor of 10^{15}, as can be seen in table 6.

Table 6. Ion counts from different sources. Plasma generated ions were obtained by mass spectrometry of the same plasma jet used during PIV and NaCl ions were dissolved at $10gl^{-1}$ prior to impingement

Ion Type	Counts
Plasma Generated Positive Ions	8.53×10^{5}
Plasma Generated Negative Ions	1.45×10^{7}
Pre-Dissolved NaCl Ions	3.40×10^{22}

Thus it was concluded that any force induced by these generated ions onto the liquid would be negligible.

4. Conclusion

To improve the efficiency of plasma-liquid applications, the effect of liquid flow can be used to influence the transmission of ionic and reactive species. We have discussed the effect of grounding a liquid target for plasma jet impingement, and the resultant changes in flow that occur. The effect of electrolysis within grounded demineralised water drives the flow upwards. The effect of then increasing salinity in the grounded water influences the flow to instead increase the downward flow velocity and thus switch direction. The resultant flow direction should rely on the mobility coefficients of the ions present, with a lower coefficient increasing the size of the flow velocity in the direction of that ions path.

The project will continue to investigate the effect of ions, in particular those generated by the plasma within the liquid phase. These will be investigated along with other electrical properties to gain a larger understanding of how these are also affecting the induced flow. In addition, there will be an expansion on numerical velocity modelling of the system to give the output of these experimental studies a more quantitative theoretical comparison, as opposed to the qualitative one described here.

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6. References

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