Advanced investigation of electrical and fluid-dynamic parameters on a nanopulsed plasma jet impinging on a liquid substrate

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Abstract: The contribution reports on the characterization of a cold atmospheric pressure plasma jet driven by high-voltage pulses with rise time and duration of few nanoseconds. iCCD imaging, Schlieren imaging, Optical Emission Spectroscopy (OES) are adopted for the source characterization while impinging on a liquid substrate at different distances and in different operating conditions.

Keywords: cold atmospheric pressure plasma jets (APPJs), plasma diagnostics, material and biomedical applications

1. Introduction

Non-equilibrium atmospheric pressure plasmas are widely investigated for their use in biomedical applications and therapies [1-5]. Plasma jet sources are able to generate a plasma plume propagating in the surrounding air and producing reactive oxygen and nitrogen species (RONS) that have a central role in the interaction with biological substrates.

Recently, it was demonstrated that the nature of the target influences plasma characteristics, such as dissipated power, reactive species generated and discharge structure [6-7].

In this study, we investigated the characteristics of a plasma jet driven by nanosecond pulses [8] impinging on a liquid substrate placed at different distances from the source outlet. The aim of this work is to use a multi-diagnostics approach to evaluate the influence of electrical and fluid dynamic parameters (peak voltage, pulse repetition frequency, gas flow rate and gap width between the jet outlet and the liquid surface) on the interaction between the plasma plume and the liquid surface. In particular, the structure of the plasma discharge, the fluid dynamics of the gas effluent and the chemical composition of the plasma plume in free flow conditions and when impinging on the liquid surface are investigated for a wide range of electrical and fluid dynamic parameters.

Since plasma treatment alters the electrical conductivity of a liquid target [9] and which in turn affects plasma characteristics [7], a buffer solution, whose conductivity (114 µS/cm) and pH are unaffected by the plasma treatment, is used as a substrate. A Schlieren imaging setup in Z configuration has been adopted to investigate both the fluid dynamic behaviour of the plasma gas mixing with the surrounding ambient air in the region downstream the plasma source outlet and its interaction with the liquid substrate.

iCCD imaging is used to investigate the temporal evolution of the plasma discharge and also to compare its discharge structure with the fluid dynamic behaviour acquired through Schlieren imaging.

Through spatially resolved optical emission spectroscopy (OES), the influence of operating conditions on the chemical composition of the plasma plume impinging on the liquid surface is explored.

2. Experimental setup

The plasma source adopted in this work is a single electrode plasma jet developed by the authors and previously reported in [6, 10-12], as shown in Fig. 1. The high voltage single electrode is a stainless steel sharpened metallic needle with a diameter of 0.3 mm; the electrode protrudes from a quartz capillary (outer diameter of 1 mm) by 3 mm. The plasma is ejected through a 1 mm orifice [6, 12] and helium (He) is used as plasma gas.

The plasma source is driven by a commercial pulse generator (FID GmbH - FPG 20-1NMK) producing high voltage pulses with a slew rate of few kV/ns, pulse duration around 30 ns, a peak voltage (PV) of 7-20 kV and an energy per pulse of 50 mJ at maximum voltage amplitude into a 100-200 Ω load impedance.

A phosphate buffer solution was prepared dissolving potassium dihydrogen phosphate (KH\textsubscript{2}PO\textsubscript{4})/ disodium hydrogen phosphate (Na\textsubscript{2}HPO\textsubscript{4}) in deionized water and adopted as a liquid substrate. The conductibility of the solution was 114 µS/cm and any increase of conductibility was observed after 30 minutes of plasma treatment. As schematically shown in Fig. 2, 120 ml of buffer solution was contained in a borosilicate glass reservoir with a grounded plate placed at its bottom.

A Schlieren imaging setup in a Z configuration [9], composed of a 450 W ozone free xenon lamp (Newport-Oriel 66355 Simplicity Arc Source), a slit and an iris diaphragm, two parabolic mirrors with a focal length of...
Fig. 1. 3D cross-section representation of the plasma source adopted in the experiments [6].

Fig. 2. Setup configuration used during the experiments.

1 m, a knife edge (positioned vertically) and a high-speed camera (Memrecam K3R- NAC Image Technology, operated at 8000 fps and 5 µs shutter time), has been adopted to visualize refractive-index gradients generated in the region downstream the plasma source outlet due to plasma gas mixing with the surrounding ambient air and to impingement on a buffered solution.

An iCCD camera (Princeton Instruments PIMAX3) was used to investigate the temporal evolution of the plasma discharge. A pulse generator (BNC 575 digital pulse/delay generator) has been used to synchronize the generator, the oscilloscope (Tektronix DPO 40034) and the iCCD camera. Two configurations of the iCCD camera have been adopted: in the first configuration, a single intensified frame (gain 50) with exposure time of 35 ns covering the entire voltage pulse has been acquired with the aim of comparing the discharge structure; in the second configuration, several sequential frames at time steps of 5 ns and with an exposure time of 10 ns (30 accumulations collected on the CCD sensor for each frame with an intensification gain of 75) have been acquired in order to track the temporal evolution of the plasma discharge during the high-voltage pulse.

A CCD camera (PIXIS400B, Princeton Instruments) mounted on a 500 mm spectrometer (Acton SP2500i, Princeton Instruments) has been adopted to collect spatially resolved optical emission spectra in the ultraviolet (UV), visible (VIS) and near infrared (NIR) regions with an exposure time has been set at 32 ms.

3. Results

Concerning iCCD acquisitions, firstly, the influence of the jet/liquid gap distance on the structure of the plasma discharge has been investigated. For a fixed set of operating conditions (peak voltage 15 kV, pulse repetition frequency 125 Hz and mass flow rate 3 slpm He) the investigated gaps were 5-10-15-20 mm and free flow (no substrate).

In Fig. 3 are presented different pairs of acquisitions (with 10 ms and 35 ns exposure time) for different gap distances. The discharge appears wider and more luminous when the liquid target is closer to the plasma source. Moreover it is observed that the plume can extend for a bigger length when impinging on a target if compared to the freeflow condition.

Fig. 3. iCCD frames of plasma discharge at different gaps both with long exposure time (10 ms, left) and fast gating (35 ns, right).

Secondly, the temporal evolution of the discharge has been studied setting the camera to scan the voltage pulse with an exposure time of 10 ns. The iCCD gate opening for the first recorded frame of each scan was set at the start of the voltage pulse and subsequent frames were recorded at fixed time steps of 5 ns; therefore two consecutive frames overlap for 5 ns.

It was observed that for the cases with 5 and 10 mm
gap, the discharge covers the source-substrate distances in a shorter time than for the other cases. Moreover, the discharges for 10, 15, 20 mm and free flow conditions reach their maximum intensity during the peak of the voltage pulse while for the 5 mm case this maximum is reached during the voltage rise.

Finally, the role of the pulse repetition frequency and the peak voltage on the discharge characteristics has been also investigated varying their values from 125 Hz to 50 Hz and from 15 to 10 kV respectively, while the gap was fixed at 10 mm. In this analysis, it is shown that the applied voltage plays a more relevant role in determining the intensity and propagation of the discharge than the pulse repetition frequency.

Schlieren high-speed recordings of the fluid-dynamic behaviour of the plasma jet have been acquired for the different electrical operating conditions and for different outlet-surface distances. The results, here reported (Fig. 4) for 3 slpm of helium flow rate, 15 kV of peak voltage and 125 Hz of pulse repetition frequency, show the spatial-temporal evolution of the turbulent front generated after the discharge ignition. Since the duration of the high-voltage pulse driving the plasma source is several orders of magnitude smaller than the time span of each Schlieren high-speed camera frame (0.125 ms at 8000 fps), the fluid-dynamic phenomena a relevant time delay after the plasma discharge propagation is observed. In Fig. 3 the turbulent front evolution is presented.

![Fig. 4. Temporal and spatial evolution of turbulence front. Black arrows indicate the propagation front. (He flow rate 3 slpm, PRF 125 Hz and PV 15 kV, gap 10 mm).](image)

From Schlieren images, it can be noted that the turbulent front appears 125 μs after the voltage pulse, propagates axially down to the liquid surface and, after hitting onto the substrate, expands radially over the liquid surface. Furthermore, the depth of the dimple produce by the gas flow on the liquid surface is reduced by the turbulent front impact.

Optical emission spectra in the UV-VIS-NIR regions as a function of both the wavelength and the gap are shown in Fig. 5 for the plasma jet operated with a peak voltage of 15 kV, a pulse repetition frequency of 125 Hz and a He gas flow rate of 3 slpm. The optical fibre was pointed at the outlet of the source (position 0 mm).

![Fig. 4. Optical emission spectra in UV-VIS (top) and VIS-NIR (bottom) range as a function of the wavelength and the gap. PV 15 kV, PRF 125 Hz, He flow rate 3 slpm, Position 0 mm.](image)

Optical radiation is emitted mainly in the VIS region, dominated by the He excitation, when the gap is smaller (5 and 10 mm of gap). In the UVB region, the intensity of the excited radicals and molecules as OH, N\(_2\), N\(_2^+\) is comparable for all tested cases.

An axial OES acquisition, fixing the gap at 10 mm and varying the position of the optical fibre from the source outlet (position 0 mm) to the liquid surface (position 10 mm) has shown that the highest intensity of the plasma plume is related to the position at 5 mm (in agreement with previously published studies [6]). The only exception to the previous statement is represented by the He bands, which results higher near the source outlet where the working gas concentration is higher. The reactive species of oxygen (ROS) and nitrogen (RNS) observed are widely known to play an important role in bacterial decontamination or in the interaction with living tissue [13, 14].

4. Conclusions

The discharge generated by an APPJ developed by the Authors and impinging on a liquid surface has been investigated. Depending on the operating conditions and the distance between the jet outlet and the substrate, the iCCD imaging has presented a plasma plume with different structure and intensity.

From Schlieren results, it was observed that a turbulent front appears several tens of microseconds after the end of the plasma discharge and propagates downstream the gas flow. When the turbulent front hits the liquid surface a reduction of the dimple depth is observed.

The plasma plume spectrum, measured by OES, was characterized by N\(_2\), N\(_2^+\), NO, OH spectral bands as a consequence of mixing with ambient air. The bands intensity appeared higher for the smaller gaps mainly in the VIS region. For all species, except helium, the
maximum lines intensity was detected 5 mm away from the plasma source outlet.

5. Acknowledgments
Work partially supported by COST Action MP1101 “Biomedical Applications of Atmospheric Pressure Plasma Technology” and COST Action TD1208 “Electrical discharges with liquids for future applications”.

6. References