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CHARACTERIZATION OF PLASMA GENERATION IN BUBBLES WITH A PLASMA GUN

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ABSTRACT

Plasma interactions with liquids has drawn much attention over the past few years considering the interest of water decontamination or treatment of pharmaceutical products. In this context, we studied the potential of low temperature plasmas created inside bubbles using a Plasma Gun (PG) from which the capillary exit is immersed into liquids. Plasma generation inside rare gas bubbles, as well as bubble growth and shape modifications were evidenced through time-resolved ICCD imaging. In this work, we performed experiments in distilled and tap water. For the same used gas, results clearly show that the plasma propagation inside the bubbles and the bubbles behaviour are strongly dependent of the liquid conductivity. These observations, not only, show the importance of the characteristics of the liquid on the production of the reactive species, but also the importance of the evolution of these characteristics during the treatment time.

Keywords: Plasma/liquid interactions, non-thermal plasma, plasma in bubbles, ICCD imaging, plasma jet

1. INTRODUCTION

Plasma/liquid interactions are investigated due to the production of reactive species, such as hydrogen peroxide, hydroxyl radicals and oxygen radicals, useful for environmental and biomedical applications [1]. Development of such applications requires a better understanding of in-liquid discharge phenomena, due to the treated liquid impact on the plasma delivery. Only few studies have been dedicated to the direct production of atmospheric pressure plasma in bubbles generated in a liquid (e.g. [2, 3]). In this work, we studied plasmas created inside bubbles using a Plasma Gun (PG) from which

the capillary exit is immersed into distilled or tap water. Series of discharges in the kHz regime are synchronized with the bubble apparition and growth. The train of discharges is stopped when the bubble ends. This allows following the bubble shape evolution and the discharges propagation over the time inside this one, versus discharge parameters and liquid characteristics. ICCD camera was used to observe the plasma propagation and the bubble evolution. The experimental setup used is described in section 2. Results are presented in section 3, followed by a general discussion in section 4.

2. EXPERIMENTAL SETUP

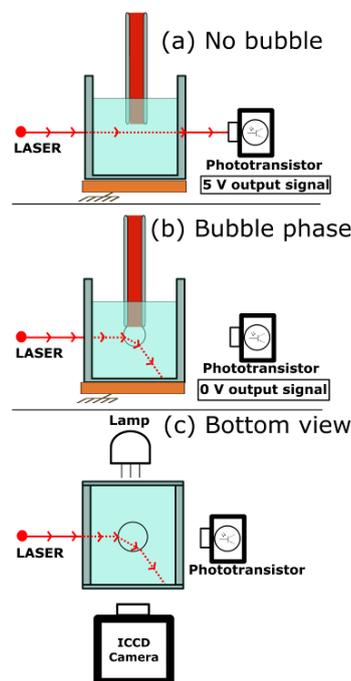


Fig. 1 Experimental scheme of the setup

The plasma jet studied in this work, so called Plasma Gun (PG), was similar to the one used by Robert *et al* [4]. The device was powered by microsecond-duration voltage pulses of 14 kV with a repetition frequency at 2 kHz. Pure neon

(N40), 99,99% purity, was flowing through the device from 12 sccm to 100 sccm flow rate. As shown in Fig. 1, the tip of the PG capillary was immersed, to a depth of 1 cm, into a plastic tank filled with 60 mL of liquid. Due to the gas flow, bubbles are created at the tip of the PG. We used two different liquids: distilled water and tap water. ICCD camera equipped with a 105 mm lens was used to measure the bubble dynamics at the PG tip using an additional backlighting lamp and to characterize plasma generation in the bubbles.

Special care was paid on the synchronisation of plasma ignition, ICCD imaging and bubble generation. A laser beam propagating through the liquid, was detected with a phototransistor. When a gas bubble intercept the laser pathway, the phototransistor signal turns to zero due to light scattering at the liquid-gas interfaces. This technique showed a reproducible behaviour of the phototransistor on/off signal for any gas flow rate used in this work. This signal was used as a trigger for the control of both the PG power supply and the ICCD gating as illustrated in Fig. 2. For the baseline, flow rate of 12 sccm used in this work, the bubbles are detected during about 350 ms. The delay between the voltage pulse and ICCD trigger and the gate duration were adjustable from the camera software. To record the plasma emission during the full duration of one voltage pulse, include the jitter associated with the bubble generation and reduce the stray light from the experimental room and the laser beam, a typical 20 μ s ICCD gating was selected. Light reflections from the PG capillary was prevented by covering this latter with a black dielectric cover.

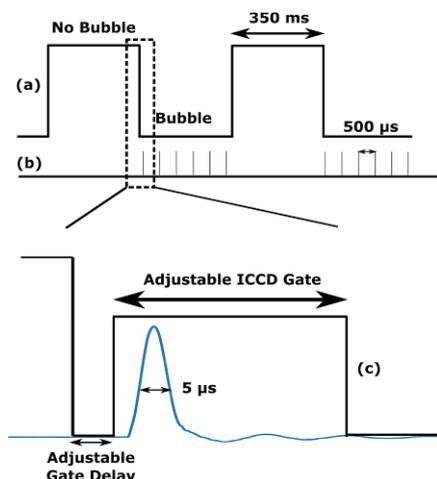


Fig. 2 Illustration of the different signals. (a) Phototransistor signal, (b) Discharge pulse is 5 μ s FWHM, (c) ICCD gating.

3. RESULTS

3.1. Plasma generation in bubbles

Fig. 3 presents plasma emission images obtained 30 ms after bubble emergence at the capillary outlet when the tank is filled with distilled or tap water.

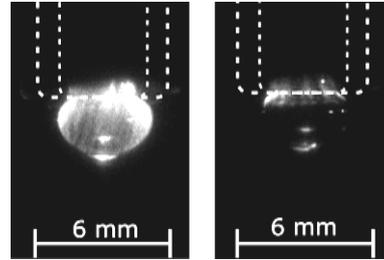


Fig. 3 Plasma emission in neon-bubble generated at 2 kHz with 12 sccm flow rate in distilled water (left) and in tap water (right).

For bubble emerging in distilled (low conductivity ($\sim 4 \mu$ S/cm) water, the plasma expansion from the PG capillary occurs in the whole volume of the bubble and exhibits a rather diffuse pattern with a stronger light emission at the boundaries with the liquid solution. Plasma hugging at the liquid boundary was previously reported and simulated [2, 3]. Conversely, in the high conductivity ($\sim 340 \mu$ S/cm) tap water experiment, the plasma is much more filamentary and propagates only a few mm at the outlet of the PG capillary, most of the gas bubble volume does not radiate any light. The key role of the dielectric or conductive nature of the liquid at the boundaries of the gas bubble is evidenced and correlates with previously reported propagation of plasma over dielectric or conductive but solid targets [5]. It was reported that with conductive target the plasma plume connects on a small size spot while plasma may expand over dielectric targets covered with a rare gas (helium or neon) layer.

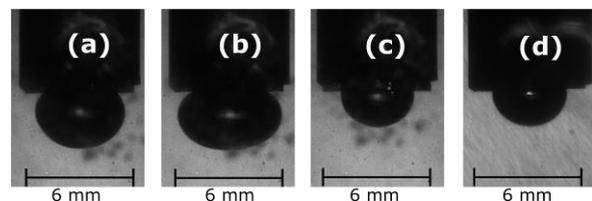


Fig. 4 100 μ s exposure ICCD of the bubble at 12 sccm flow rate with plasma in distilled water (a) and in tap water (b), without plasma in tap water (c) and in distilled water (d).

Fig. 4 shows 100 μ s exposure ICCD images of the bubble in distilled water and in tap water, captured 30 ms after the beginning of the bubble

growth, when plasma is ignited and the bubble images at the same delay when PG is switch off. In the distilled water the bubble appears almost spherical, while in tap water, the bubble exhibits an egg-like shape. The drastic difference between these two shapes must be due to the plasma development and characteristics inside the bubble. Comparison of bubble growth with or without plasma ignition also evidence the strong impact of discharges on bubble dynamics as detailed in next section of this abstract.

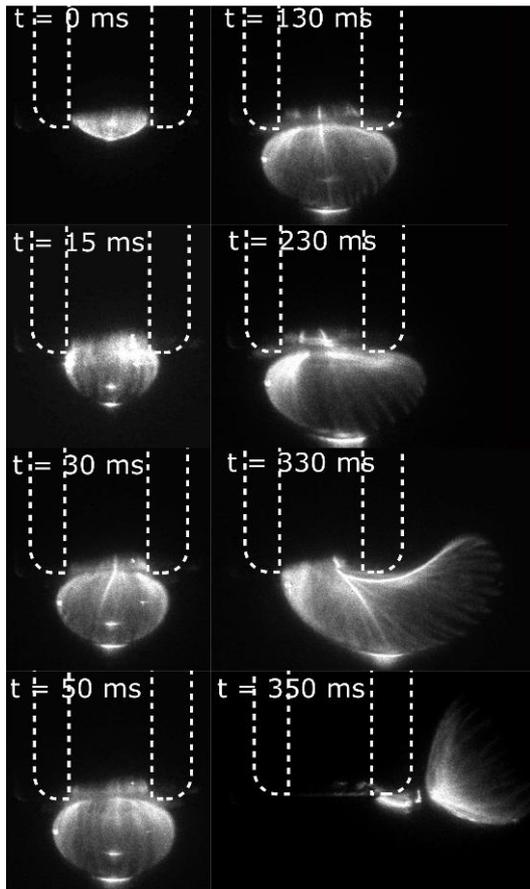


Fig. 5 Evolution of a bubble in distilled water.

Fig. 5 presents time resolved plasma emission during the whole bubble lifetime, from its emergence to its detachment from the PG capillary, in distilled water solution. The simultaneous expansion of the bubble volume and its filling with the plasma is observed during the first 50 ms. Then, bubble growth mainly occurs perpendicularly to the PG capillary axis, and a gradual shifts in the right direction in this case (see Fig. 5) is measured from about 50 to 350 ms after bubble emergence. During this second phase, the plasma still expands in the whole volume of the bubble. While plasma fills the full volume, more or less intense plasma filaments are observed and look to be mainly

present at the boundary with the liquid solution. Finally, bubble detached from the PG capillary and travels upwards in the tank. During the first instants of this bubble detachment, plasma emission occurs at the capillary outlet where neon gas is still flowing, but more surprisingly it has been measured that plasma generation is also triggered in the detached bubble. As shown for 350 ms delay, plasma is ignited in the detached bubble across the thin water layer present between the neon flow tip flowing from the PG capillary and the upwards moving bubble. This plasma ignition may be described as atmospheric pressure plasma transfer across dielectric material [6]. Such plasma expansion in released bubbles was also reported in [3] with a different but helium fed discharge setup.

3.2. Influence of plasma on bubble growth

The bubble growth dynamics was assessed from time resolved ICCD images by reporting the evolution of the on-axis bubble height from the capillary outlet versus time. As the full diagnostics chain is triggered from laser beam probing the bubble emergence, the very first instants of bubble growth could not been measured. Fig. 6 presents the on-axis bubble height time evolution for the baseline 12 sccm gas flow rate (the PG being ignited at 2 kHz and 14 kV peak voltage), and for three 12, 18 and 76 sccm neon flow rates operation. For these three latter operating conditions the PG was not ignited. As previously depicted from ICCD plasma images (Fig. 5), in the baseline setup (12 sccm, PG on), the bubble height rapidly increase during the first 50 ms and then stabilizes around 4 mm until around 300 ms delay from bubble emergence. Then the bubble detached more or less erratically during typical time duration of about 20 ms. For the same neon flow rate but with no plasma ignition, the bubble expansion is drastically more slow and does not reach the same 4 mm excursion from capillary tip during the 300 ms time window. It was measured that increasing the flow rate up 18 sccm (40% increase) allow the bubble to reach the 4 mm excursion but still with a much more slow height increase rate. The fast increase of bubble on-axis expansion measured in the baseline setup was mimicked with no plasma ignition but if neon flow rate was six fold increased. Nevertheless, for such high neon flow rate, the bubble expands even at larger distances from capillary tip reaching nearly 5mm before

quickly detaching about 100 ms after its emergence. These measurements enlighten that plasma ignition has a key influence on bubble growth, the action being partly associated with a gas flow rate modification but also probably associated with pressure burst or small gas heating and consecutive neon density modulation following plasma ignition. The first effect was previously reported as the channelling action of plasma and recently confirmed for helium, neon or argon feeding gas [7]. The second action is a new observation, which may explain the different bubble shapes measured (Fig. 4) in different waters, associated with different plasma developments in bubble (Fig. 3).

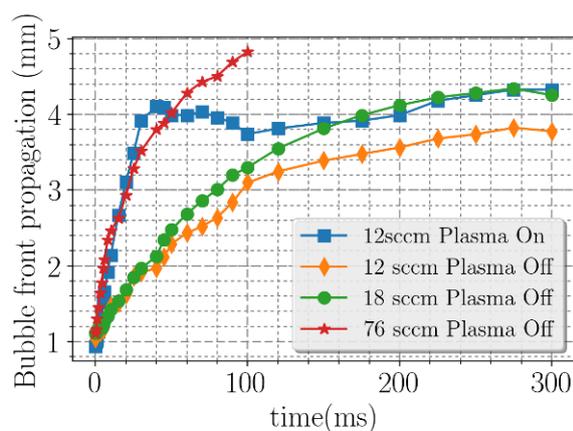


Fig. 6 Evolution of the bubble growth at 12 sccm neon flow rate with plasma versus 18 sccm and 76 sccm flow rate without plasma.

4. DISCUSSION

The generation of plasma in bubbles emerging from the plasma gun capillary flushed with very small gas flow rate of neon and immersed into liquid solution as been demonstrated and characterized by ICCD imaging. The key role of water solution conductivity was analyzed and results in different plasma expansion in gas bubble. Plasma expansion in bubble appears to be much stronger at the gas-liquid interface and much more filamentary and in smaller volume when liquid solution is highly conductive. The strong influence of plasma generation on bubble growth dynamics has also been documented. It is suggested that plasma has partly action on the gas flow features at the capillary outlet but also probably induces heat or pressure modulation of gas inside bubbles in comparison with bubble generated with no plasma ignition. More work is in progress to analyze these phenomena. This would be important to consider developing plasma chemistry models in plasma activated

solutions. Finally, it was shown that plasma may be ignited not only in bubbles still connected to the capillary feeding gas but also inside detached bubbles. This open opportunity to activated micro bubbles when large enough bubble density per volume unit is available. Dielectric barrier discharge propagation from each individual bubbles will be involved for such liquid targets activation. This setup, where gaseous bubbles in a liquid buffer are excited, will be almost the reverse with that encountered in sprays activated devices where liquid bubbles are flowing in plasma gaseous buffer.

These fundamental features should be considered when trying to analyze plasma activated liquids properties obtained in different solutions or for different treatment duration. With our setup, treatment of distilled water solutions over few minutes is associated with a drastic evolution of the liquid conductivity and consequently of plasma generation in bubble during plasma delivery. The plasma generation on the boundaries of gas bubble in liquid is of interest for the transfer of plasma excited species from the gaseous phase to the adjacent liquid phase and consecutively efficient activation of liquid solutions.

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