AC air plasma in contact with water: fundamental investigation and application in water treatment

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Abstract: Plasma-liquid interactions have great potential for numerous applications, including environmental remediation. The simultaneous physical and chemical phenomena, induced by plasma, have synergetic effect and can be used in water treatment, among other applications. In this paper, we study an AC discharge in air using pin-to-water geometry. Our results show that the discharge behaviour is very sensitive to gap distance. Such a sensitivity is also observed when testing the degradation of methylene blue, a standard pollutant.

Keywords: air plasma, pin-to-plate, ICCD imaging, water treatment.

1. Introduction

Plasmas in- and in-contact with liquids have very high potential for environmental remediation. In this context, the simultaneous generation of oxidative and energetic species makes it at the top of the next promising technologies for wastewater treatment, among other applications. As water is involved in the plasma process, water molecules are dissociated, and various species and physical phenomena are produced, such as O, O₃, OH, H_2O_2 , UV radiation, shockwaves, etc. [1]. Plasmas-water interactions have been investigated using various reactor, and mainly direct current (DC) and pulsed discharges [2].

Here, we present some of plasma characteristics and the induced-water modifications using pin-to-water geometry. Particularly, the space- and time-evolution of the discharge is studied using ICCD camera. The current-voltage characteristics will be also provided and discussed. Finally, using methylene blue as standard pollutant, its degradation efficiency is evaluated under various discharge conditions.

2. Experimental Setup

The experimental setup is shown in Figure 1. The plasma is produced in air by applying high-voltage between pin electrode and water. The pin electrode is made of tungsten and has a diameter of 2 mm (it is mechanically polished to have a curvature radius ~50 μ m). The pin is mounted on a micrometer screw positioning system to finely adjust the distance between the pin-tip and the water surface.



Fig. 1. Schematic of the experimental setup used to generate air plasma in contact with water.

A 300 mL of distilled water was filled in a cylindrical container and is connected to grounded by immersing a stainless-steel rod in water. The high-voltage is generated using an arbitrary function generator (Agilent, 33220A), a current amplifier (Crest Audio CC, 4000), and a transformer (Montoux, 600VA60V/9kV). The currentvoltage characteristics were measured using a high-voltage probe (Pearson P6015A) and current coil (Pearson, P2877), respectively, and were acquired using an oscilloscope (Tectronix, DPO5420B). Plasma images are obtained using an Intensified Charge Couple Device (ICCD) camera (Andor Technologies, DH520 18F) with a mounted lens (Tamron, SP 52B). A pulse generator (Quantum Composers Plus, 9518) is used to synchronize the camera with the electric signal and to control the integration time of the camera. To measure the degradation rate of methylene blue (MB), a home-made absorption setup (Fig. 2) was built using a tungsten lamp (400-1000 nm), sample holder (rectangular PE sample), and spectrometer (Avantes AvaSpec, 2084-USB-2). The setup is calibrated to measure MB concentration between 2 µg/L and 15 mg/L, using standard solutions.



Fig. 2. Schematic of the home-made absorption apparatus to measure degradation rate of methylene blue.

3. Electrical and optical characteristics of the discharge First, we studied the effect of the gap, distance between the pin-tip and water surface) on the electrical characteristics of the discharge. Figure 3 shows three typical current-voltage characteristics measured for three gap distances: 1, 3, and 5 mm. In this study, the frequency as well as the applied voltage (peak-to-peak) were kept constant at 10 kHz and 10 kV, respectively. Since the highvoltage is a sinusoidal waveform, the polarity of the pin electrode changes each 50 μ s.

For gap distance of 1 mm, the breakdown (voltage-drop and current-peak) is observed for each half-period, but the measured values depend on the polarity. An increase of the gap distance to 3 mm, the voltage to reach breakdown as well as the current-peak, during the positive half-period, are higher than those reported for 1 mm. In contrast, the measured current during the negative half-period remains comparable to the 1 mm case. This result was already well described by, *e.g.*, Magureanu *et al.* [3].

Further increase of the gap distance to 5 mm induces a drastic change to the electrical characteristics. Indeed, a voltage-drop and current-peak were observed only when pin polarity is negative. Finally, the breakdown of the gas is no longer possible for a gap distance higher than 9 mm.



Fig. 3. Typical current and voltage characteristics for three gaps: 1, 3, and 5 mm.

To investigate the emission morphology of the discharges under various gap distances, we used the ICCD camera that integrates light from \sim 200-1000 nm, and the integration time was fixed to the whole half-period. Typical images for the plasma column, for gap distances of 1, 3, and 5 mm, are presented in Figure 4.

The images clearly highlight various spatial distribution of the emitted light. For gap distance of 1 mm, the discharge is like a triangle (pyramid in 3D) and its basis is localized at the water surface. During positive polarity, the region with high intensity is much larger than that observed during negative polarity; the pin seems to be more illuminated and non-homogeneous distribution can be observed. As the gap is increased to 3 mm, during the positive polarity, the gap is continuously fulfilled by plasma, and two regions can be distinguished: 1) close to the water surface, the triangle shape can be distinguished and its height (~1 mm) seems to be comparable to that identified in the 1 mm-gap case, 2) from the triangle top to the pin-tip, an homogeneous plasma column can be identified. During the negative polarity, although the behaviour of the plasma-column is very similar to that reported in the 1 mm-case, one can identify an illuminated plasma spot on water surface. Further increase of the gap distance to 5 mm induces a drastic change in the emitted light during the positive polarity; the intensity is relatively low as compared with other gap-distances. It is worth noting that even there is no a clear voltage-drop and current peak in the current voltage characteristics, one can still remark a continuous current (few of mA) and a global voltage attenuation (from 5 to 3 kV), explaining thus the detected light in the image. During the negative polarity, as mentioned for other gap-distances, the general plasma shape does not change, but the intensity of the water surface is increased significantly.



Fig. 4. ICCD images (exposure is 50µs) of the plasma column for three gaps: 1, 3, and 5 mm.

4. Water treatment

We used this above setup for water treatment. Adding MB to distilled water at a concentration of 10 mg/L, we have monitored the degradation rate of MB by measuring the absorbance at 633 nm. Figure 5 shows the evolution of MB concentration as a function of treatment time. A simple comparison between the values indicates that the efficiency in removing MB is higher for short gap distance.



Fig. 5. Time evolution of MB concentration during plasma treatment under three gaps: 1, 3, and 5 mm.

5. Conclusion

In conclusion, we presented the results obtained with a pin-to-water geometry working at a relatively low frequency (10 kHz). We investigated the electrical and optical properties of the discharge, and their variations as a function of the gap distance between pin-tip and water surface. At short gap distance (typically < 4 mm), the emitting light from discharge is strong during the positive as well as the negative half-periods. Beyond 4 mm, the discharge light intensity during the negative half-period is much higher than that measured during the positive period. The influence of the gap distance on the removing rate of a standard pollutant, methylene blue, has been conducted, and we observed that the efficiency is higher for shorter gap distance.

6. References

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