Is a submerged microwave plasma jet efficient for water treatment?

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Abstract: Microwave plasma jet (MWPJ) in water is a promising setup for water treatment as it is an electrodeless plasma device. Using a surfatron at a 2.45 GHz, plasma is generated in a quartz tube and injected in water. The propagation of the MWPJ in water is highly dependent on the experimental parameters, namely the microwave power, gas composition and flow rate, and water electrical conductivity. The applicability of this plasma for water treatment is evaluated using methylene blue as standard pollutants.

Keywords: Microwave plasma, plasma in liquid, water treatment.

1. Introduction

Interactions between plasma and liquids have gained much interest over the last decade due to their interest in many applications ranging from environmental remediation to material science and health care [1]. The simultaneous generation of physics and chemistry of interest makes plasma at the top of the promising technologies for liquid processing, particularly for water treatment. Numerous plasma sources have been developed and studied for liquid processing. They can be classified into two categories: thermal and non-thermal plasma sources. Non-thermal plasma, including pulsednanosecond discharge, dielectric barrier discharge, and plasma jet, have been extensively investigated for liquid processing. Such plasma has shown great potential for controlling the electron induced chemistry. On the other hand, thermal plasma such as plasma torches and arc jets are known to utilize high temperature chemistry as well as plasma generated reactive species (electrons, ions, radicals, photons, etc.). Most of the plasma sources for liquid processing use electrodes to ignite and sustain the plasma [2]. Unfortunately, such setups suffer from electrode erosion [3], and in the case of water treatment, electrode erosion i) reduces the plasma-device lifetime and ii) pollutes the water with nanomaterials. To overcome such a barrier, electrodeless plasma devices were proposed.

Here, we propose a submerged electrodeless microwave plasma jet (MWPJ) device for water treatment. Such electrodeless setup has the advantages of controlling plasma parameters independently of the liquid, suppressing the electrode erosion, and providing an efficient water treatment.

2. Experimental Setup

A schematic representation of the setup is shown in Fig. 1. A microwave power at 2.45 GHz, using a surfatron device, is applied to a gas flowing in a quartz tube. The quartz tube is connected to a quartz cell filled with 200 mL of water. The outer diameter and inner diameter of the quartz tube are 6.0 mm and 4.0 mm, respectively, and the distance from the bottom of the quartz cell to the quartz tube is 5 cm. The surfatron is positioned closely to the quartz cell.



Fig. 1. Setup to generate MWPJ in water.

We used a high-speed camera to investigate plasmabubble dynamics. Emission spectra are acquired using a monochromator equipped with ICCD camera as a detector. This setup is used to evaluate the applicability of the MWPJ for water treatment by investigating the removal of methylene blue (MB), 50 mg/L, from water.

3. Results and Discussion

Figure 2 shows the plasma-bubble dynamics created by MWPJ in water for two gas compositions: Ar and Ar/N_2 (total gas flow is 2 L/min). In 100% Ar (Fig. 2a), the plasma is visible at an early stage, and its length increases with time. The plasma is about a channel that horizontally crosses the bubble. Adding N₂ to Ar has significantly changed the plasma dynamics, but not the bubble dynamics. The plasma channel appears in the bubble ~6 ms later, and it is thinner than that obtained in 100% Ar. Moreover, the time evolution of the plasma length has an oscillatory behaviour, unlike the case of 100% Ar.



Fig. 2. Plasma and bubble dynamics created by MWPJ in water. a) 100% Ar and b) 98% Ar and 2% N₂; total gas flow is 2 L/min, power is 200 W, and water volume is 200 mL.

The characteristics of the discharge behaviour are determined by processing high-speed images to determine the time evolution of the horizontal lengths of plasma channels and bubbles; the results are presented in Fig. 3. A monotonous increase in bubble length along time is observed, showing a delay in the growth of the bubble as its volume is proportional to its cubic length. When the bubble length reaches a critical length, the bubble gets detached from the side wall and a new bubble starts forming. Unlike the length of a bubble, the plasma length first increases, but then decreases rapidly leading eventually to the total extinction of the plasma. This behaviour is an indication of a complicated phenomenon of plasma dynamics inside the bubble.



Fig. 3. Time evolution of the bubble length and plasma lengths at 100% Ar and at 98-2% Ar-N₂; total gas flow is 2 L/min, power is 200 W, and water volume is 200 mL.

Also, we have studied the influence of N_2 , in Ar- N_2 gas mixture, on the dynamics. The change in the bubble length is negligible for various N_2 percentages. For N_2 lower than 1.5%, the plasma channel increases monotonically with time, but the magnitude of the final length is relatively shorter than that measured in 100% of Ar. For N_2 percentage higher than 1.5%, the monotonous increasing behaviour of the plasma length is not observed anymore, showing "humps and pits" in its trail, characteristic of the oscillation of a discharge channel. The case of 2% of N_2 is presented in Fig. 3 and compared with the 100% of Ar as well as with the bubble length. It is worth noting that no discharge channel is observed for more than 5% of N_2 .

To investigate the potential applicability of the submerged MWPJ to water treatment, we selected MB as a simulated organic contaminant. Figure 4 represents the variations of the absorbance (normalized to the case with 100% Ar, *i.e.*, 0% N₂) as a function of the N₂ percentage in the gas mixture. In this figure, we clearly see that the efficiency of the decolorization process (i.e., MB decomposition) is obtained for a nitrogen percentage of 2.5%. Based on the results obtained with emission intensities at various N2 percentages (not shown here), we postulate that an increase in NH radicals (for small

additions of N2) could play an important role in the decolorization process. In addition to the emitting energetic photons (wavelengths 330-340 nm), NH radicals have a relatively high redox potential (~1 V), and therefore may contribute to the decomposition of MB.



Fig. 4: Absorbance (normalized to the case with 100% Ar) as a function of the N₂ percentage, in an Ar-N₂ gas mixture, after plasma processing of 15 minutes; total gas flow is 2 L/min, power is 200 W, and water volume is 200 mL.

4. Conclusion

The plasma dynamics as well as the bubble dynamics are investigated using high-speed imaging. We find that the plasma length increases with either increased gas flow rate. We also show that the addition of N_2 to Ar highly affects the plasma dynamics exhibiting an extinction of plasma for the N₂ portion is higher than 5% while, at the percentage <5%, an oscillatory behaviour of the discharge channel is found.

A characterization by an optical emission spectroscopy shows that, in the case of Ar plasma, the spectrum is dominated by OH (A-X) band and by Ar I lines, including discernable H (α , β , and γ), O I, and the NH (A– X) band, indicating effective dissociation of water via plasma chemistry. As a result of N2 addition, an intense emission of NH is observed with additional excited N2 and N2⁺ emissions. In order to assess the feasibility of the submerged MWPJ to in-liquid processing, we used methylene blue as a model organic contaminant. We compare the decolorization performance, and we find that adding 1-3% of N₂ to Ar enhances the decolorization of the liquid. This is because the backward reactions of H_2O_2 to produce OH radicals caused by NO and NO2. This result shows that the MWPJ can be used for efficient inliquid chemical processes, avoiding an issue with electrode erosion encountered in other plasma devices.

5. References

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