Effects of pre-treatment of inject gas on OH production in gas-liquid phase plasma

気液混相プラズマを用いたOHラジカル生成における投入ガスの放電前処理効果

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In order to enhance the effect of a gas-liquid interface discharge to water purification, it is important to increase hydroxyl radicals (OH) produced in water, which have a higher oxidation potential than other oxidative species. We have developed a new reactor utilizing two kinds of discharge. Injection gas was pre-treated to generate excited molecules by a dielectric barrier discharge (DBD), and then it was introduced into a gas-liquid interface to generate streamer corona discharge, resulting the production of OH. The concentration of OH was evaluated by that of H_2O_2 per pulse, and it increases with the DBD pre-treatment compared with that without DBD. In addition, the effect of the DBD pre-treatment appeared with the increase of the repetitive frequency of the nanoseconds pulse discharge.

1. Introduction

Atmospheric pressure non-thermal plasmas have attracted significant attention in the research field of water purification due to their good performance of the degradation of persistent organics and the sterilization of bacteria [1]-[3]. The plasma can generate high energy electrons on gas-liquid interface and it produces reactive species such as hydroxyl radicals (OH), ozone (O_3), and atomic oxygen (O). Specifically OH has a higher oxidation potential compared with the other oxidative species. On the other hand, the very short lifetime of OH is a restriction for their application. Therefore, it is important to generate OH on the gas-liquid interface and to interact directly with target substances.

In recent researches some reactive species have been directly measured in an atmospheric pressure plasma by laser induced fluorescence method [4] and their reaction mechanisms have been gradually elucidating. However, the behavior and reaction mechanisms of many reactive species are still unknown. Long-lived excited molecules such as $N_2(A)$, $O_2(a)$, $N_2(v)$ and $O_2(v)$ have attracted researchers' attention in the atmospheric pressure plasma applications [5,6]. For example, it has been reported that the lifetime of $N_2(v)$ is 1~10 ms in dry air [7] and a dissociation process by multiple electron collision via $N_2(v)$ is proposed as a source reaction of nitrogen atom [8]. Therefore, the long lived excited molecules may help to increase the amount of the reactive species at an optimum operation condition.

In this study we have fabricated a new reactor, where two kinds of discharge are utilized, a dielectric barrier discharge (DBD) and a gas-liquid discharge.

In the reactor gas is pre-treated to generate excited molecules by a dielectric barrier discharge (DBD), and introduced into a gas-liquid interface to generate streamer corona discharge, resulting the production of OH. The effects of the pretreatment on OH concentration are studied.

2. Experimental setup

Figure 1 and 2 show the schematic diagram of electrical circuit and the waveform of high voltage pulse, respectively. The circuit consists of a capacitor (C), gap switch (GS), inductor (L), and semiconductors opening switch (SOS) diodes (K25UF). The capacitor is firstly charged up by using a DC high voltage power supply. When the gap switch is closed, the charge stored in the capacitor is released and current starts to flow through the inductor and the SOS diodes. When the direction of the oscillating current reversed, it is interrupted by the diodes with the delay of 100ns. The sudden interruption of the inductor current results in the generation of high voltage pulse V_O with a short pulse duration of several tens of nanoseconds (around 40 nsec) .

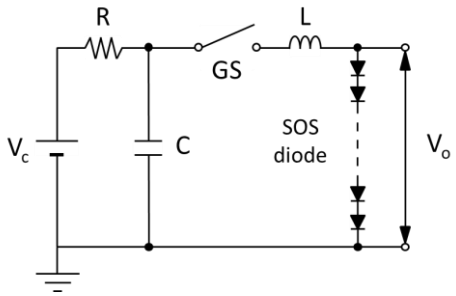

Fig. 1 The schematic diagram of the pulsed high-voltage circuit.

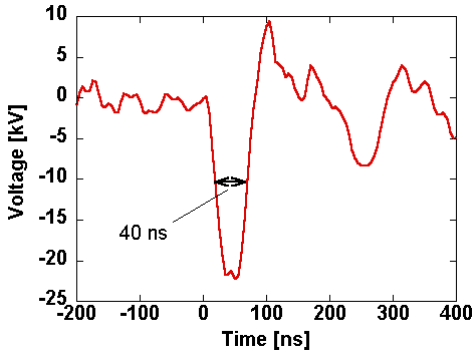

Fig. 2. A typical waveform of the high-voltage pulse.

Figure 3 shows the schematic of a newly developed multistage discharge reactor. A wire electrode is set through the glass tube and the wire tip is projected from the glass tube end. The glass tube with the wire electrode is immersed into water. A grounded electrode is set in the water. When the pulsed high-voltage is applied between the electrodes with injecting dry air gas into the glass tube, barrier discharge is generated inside of the glass tube and streamer corona discharge is generated in the gas bubble appeared at the glass tube end. The spatial length of the barrier discharge region can be changed by changing the length of the grass tube into water.

3. Result and discussion

The production of OH radicals in a discharge reactor is evaluated from the concentration of hydrogen peroxide (H_2O_2) produced in the treated water from absorbance of a light (λ=540nm). The calibration is performed using H_2O_2 solution whose concentration is already known.

Figure 4 shows a concentration of OH as a function of pulse interval time. The concentration of H_2O_2 per pulse increases in the new reactor with the DBD pre-treatment compared without the pre-treatment. The pulse interval time of 3.3 ~ 10 ms (pulse repetition frequency : 100~300 Hz) roughly corresponds to the lifetime of $N_2(v)$ in dry

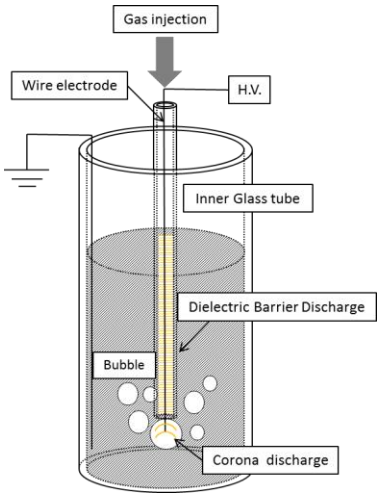

Fig. 3. The schematic of multistage discharge reactor.

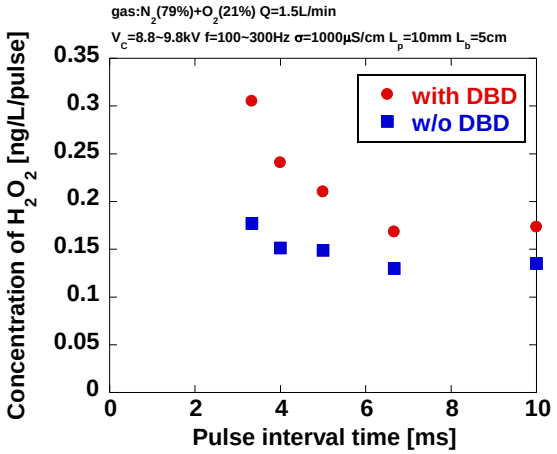

Fig. 4. Concentrations of hydrogen peroxide as a function of pulse interval time.

air. This indicates that $N_2(v)$ generated in the pre-discharge affects the OH production in this reactor.

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