

Radio Frequency Plasmas in Water under Pressurized Condition

圧力負荷を伴う水中高周波プラズマ

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Radio frequency plasmas in water under pressurized condition were investigated. In spectroscopic measurements, the intensity ratio between OH and H α , $I_{OH}/I_{H\alpha}$, decreases with increasing in pressure. Methylene blue was employed for degradation study. As compared with exposure at 1 atm, larger degradation was achieved at 3 atm. Concentration of hydrogen peroxide was 65 mg/L after 3 min exposure at 1 atm while it was 180 mg/L after exposure at 3 atm.

1. Introduction

Plasma in water can be used in advanced oxidation processes (AOPs) since it produces active species such as OH radicals. A number of studies have been conducted on discharges generated in water [1-3].

Recently, the generation of radio frequency (RF) plasmas in water has been reported [4]. RF power is injected into water through an electrode. RF plasma surrounded by a bubble is generated and maintained on the electrode. In ref [4], plasmas are utilized for the degradation of methylene blue dissolved in water. This degradation may be due to OH radical produced by RF plasma. In ref [5], the temperature of OH, electron density, and behavior of bubbles generated by a 27.12MHz in-liquid plasma are investigated in water under pressures ranging from 0.1 to 0.4MPa. The temperature of OH and the electron density increase with increasing in pressure. These changes following after pressurization may influence the generation and activity of OH radicals. Here, in order to discuss the influence of the pressure, we carried out the degradation studies and observation of the formation of hydrogen peroxide under pressurized condition.

2. Experimental Apparatus

RF plasma surrounded with bubbles in water was generated on the electrode in a pressure vessel (Fig. 1). The pressure vessel (ID: 60 mm, height: 100 mm) was made of stainless steel and it can be used below 10 atm. Since the vessel was connected to the ground line, it functioned as a counter electrode. Water or methylene blue solution (250 mL) was

stored in the vessel. Water flowing through a stainless pipe was used to control the temperature of water.

The hot electrode was made of copper; its diameter was 3 mm. Its top was covered with a ceramic pipe (OD: 5 mm, ID: 3 mm). The electrodes were connected to a resonator, and RF power was supplied to the resonator through the tuner from a power supply (T161-6013H, Thamway, $f = 13.56$ MHz). Here, the net input power was fixed at 400 W.

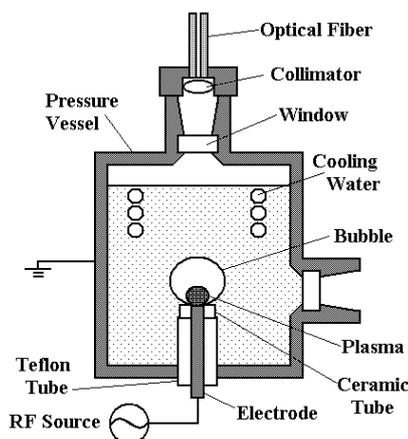


Fig. 1 Schematic view of experimental setup.

3. Experimental Results and Discussions

The plasmas were initiated as follows. Keeping the RF power low, the reflected RF power was reduced by adjusting the tuner. Then, the RF power was increased until breakdown. The breakdown was observed around 600W at an atmospheric pressure. Just after breakdown, the RF power was decreased to 400 W. Pressure was gradually increased to 3 atm for one minute after breakdown.

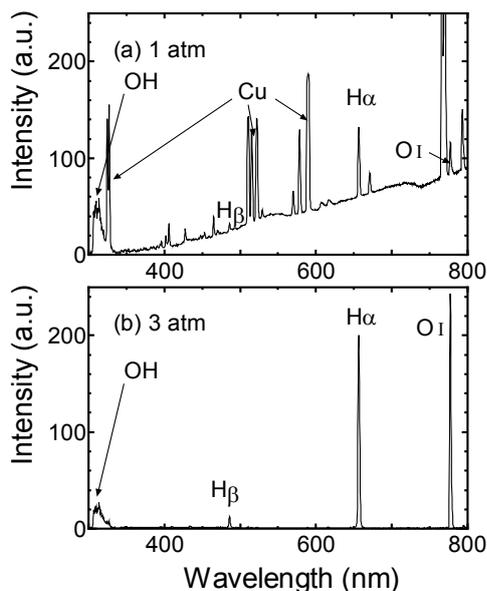


Fig.2 Emission spectra of RF plasma maintained at (a) 1 atm and (b) 3 atm.

Emission spectra of RF plasma generated in water at (a) 1 atm and (b) 3 atm are shown in Fig. 2. The spectral lines, generated due to the radicals, OH, O, and H, were observed. This shows that water molecules are decomposed into OH, O, and H radicals by the RF plasma. In Fig. 2(a), strong continuous spectrum is observed. This is the radiation from the electrode and shows that the electrode is heated by large RF power. Copper lines are also observed. In Fig.2 (b), continuous spectrum is not observed. This suggests that the electrode is cooled enough in high pressure case. The intensity ratio between OH and H α , $I_{OH}/I_{H\alpha}$, decreases with increasing in pressure. Reference [5] shows that temperature of RF plasma in water increases with increasing in pressure. Increasing in temperature leads to decomposition of molecules. The change of the intensity ratio may be induced from increasing in temperature.

RF plasma in water can degrade organic matter. Here, we employed methylene blue solution (5 mg/L). As compared with the solution before exposure, the solution after exposure lightened and its color tone varied. This variation is obvious from the absorbance spectrum. A double-beam spectrophotometer (U-2810, Hitachi) was used for the measurement of absorbance spectra. Typical variation of absorbance spectra with exposure time is shown in Fig. 3. As shown in Fig.3 larger degradation was achieved at 3 atm.

Hydrogen peroxide is also produced by the generation of RF plasma in water through a recombination process: $2OH\cdot \rightarrow H_2O_2$. After pure

water (250 ml) was exposed to the RF plasma for 3 min, the concentration of hydrogen peroxide was determined using a colorimetric method (DPM-H $_2$ O $_2$, Kyoritsu Chemical-Check Lab). It was 65 mg/L after exposure at 1 atm while it was 180 mg/L after exposure at 3 atm. Degradation of methylene blue and generation of hydrogen peroxide suggest that pressurized condition enhances the formation of OH radical. This is in contrast with the fact that the intensity ratio decreases with increasing in pressure.

4. Summary

Radio frequency plasmas in water can be maintained even under pressurized condition. Spectroscopic measurements show that the intensity ratio between OH and H α , $I_{OH}/I_{H\alpha}$, decreases with increasing in pressure. In degradation study, plasmas at 3 atm can degrade more than those at 1 atm. Concentration of hydrogen peroxide was 65 mg/L after 3 min exposure at 1 atm while it was 180 mg/L after exposure at 3 atm.

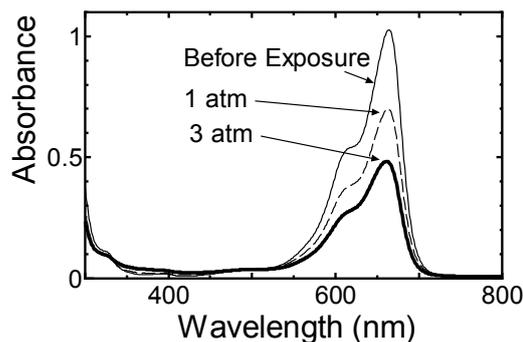


Fig.3 Absorbance spectra of methylene blue solution before (solid line), after exposure at 1 atm (dashed line) and 3 atm (bold line), respectively. The exposure time is 3 min.

Acknowledgments

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