Proceedings of Symposium on Ultrasonic Electronics, Vol. 34 (2013) pp. 441-442
20-22 November, 2013

Effect of superposing ultrasonic wave on microwave plasma under water

マイクロ波水中プラズマにおける超音波重畳の効果

Tomohiro Takahashi[1], Noriharu Takada[2†], and Hirotaka Toyoda[1,3] ([1]Department of Electrical Engineering and Computer Science, Nagoya Univ.; [2]Technical center, Nagoya Univ.; [3]Plasma Nanotechnology Research center, Nagoya Univ.)
高橋朋大 [1]，高田昇治 [2†]，豊田浩孝 [1,3] ([1] 名大院工, [2] 名大全学技術センター, [3] 名大プラズマナノ工学研究センター)

1. Introduction

Recently, plasma production under liquid attracts much attention for several applications such as the production of new materials or the destruction of water pollutant compounds.[1,2] Generally, in liquids such as water, electrical breakdown for the plasma production requires extremely high voltage of dc[3,4] or RF,[5,6] which is applied to metal electrode installed in liquids. Also, in microwave discharge, microwave power fed through a coaxial cable is provided to a metal rod antenna.[7,8] However, these plasma production techniques have a problem of discharge-electrode damage due to sputtering or arcing. In addition, these methods have difficulty in producing wide-area plasmas needed for high performance in practical applications. Therefore, as an alternative of plasma production method under liquid, pulsed microwave excited plasma using a slot antenna covered with dielectrics has been proposed, because the damage to the slot electrode can be minimized and plasma volume can be increased.[9,10] Also, improvement of liquid treatment efficiency with use of reduced-pressure condition during the discharge has been reported.[9,10] Although the physical and chemical reactions during the liquid treatment are complicated, typical reactive species those contribute to decomposition of organic compounds are known to be hydroxyl radical, hydrogen peroxide, oxygen radical and ozone.

It should be noted, in the microwave discharge in water, that the discharge breakdown does not occur in the liquid phase (water) but in the gas phase (bubble). Namely, at the initial stage after the supply of microwave power, a bubble is formed in the vicinity of the slot antenna by microwave heating of water. In the subsequent step, microwave breakdown takes place inside the bubble filled with water vapor and the plasma is maintained by the microwave power. Considering liquid treatment with high performance, lower microwave waste power for the production of the bubble and enhancement of plasma volume for liquid treatment are key parameters. In this work, we have proposed superposition of the pressure change generated by ultrasonic wave to microwave excited plasma production under water. In paticular, we expect that the addition of ultrasonic negative pressure at the slot position causes the assisting effect to the bubble production and the enahcenment of the plasma volume.

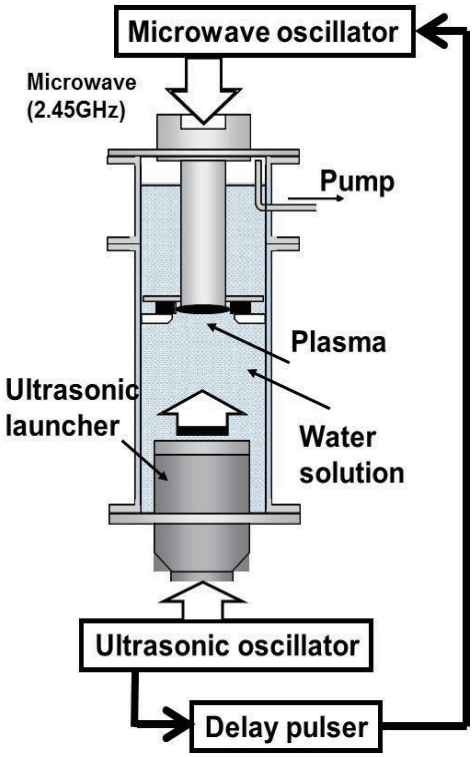

Fig. 1 Schematic view of experimental setup

To date, the influence of ultrasonic wave on chemical reactions in water has been investigated in the field of sonochemistry.[11] It is well known that excitation of acoustic cavitation in water induces the formation of active hydrogen, hydroxy radicals, hydrogen peroxide and enhances chemical reactions.[12,13] Therefore, we also expect the additional effect on the water treatment by the induced active species.

In this study, correlation between the ultrasonic wave and the plasma production and the organic solution decomposition efficiency will be discussed.

2. Experimental

Figure 1 shows a schematic view of the experimental setup. In the experiment, plasma was produced under water in a cylindrical container. Pulsed 2.45 GHz microwave power from a quartz-filled waveguide was injected into water through a slot antenna. Ambient pressure in the discharge chamber was reduced to <10 Pa by a scroll pump. Ultrasonic wave (frequency: 20 kHz) was introduced into the plasma production area under water from an ultrasonic launcher that was positioned facing to the microwave slot antenna. Microwave pulse frequency was synchronized with the ultrasonic frequency by a delay pulser. Phase angle between the microwave pulse and the ultrasonic wave was varied from 0 μs to 50 μs with 5 μs interval (hereinafter these phase angle are referred to as "Delay time"). At each delay time, optical emission intensity from the plasma was measured by an optical fiber spectroscope. Decomposition percentage of organic solution (methylene blue: MB) was evaluated from spectroscopic approaches.

3. Experimental results and discussion

Typical optical emission lines from atom and molecules, i.e., OH (309 nm), H_β (486.1 nm), H_α (656.3 nm) and O (777.5, 844.6 nm) were observed from microwave plasma. It is speculated that these active species are induced by the plasma inside the bubble because the optical emission occurs through electron impact dissociative excitation of H_2O inside the bubble, i.e., $H_2O+e \rightarrow H + OH^* + e$.

Optical emission intensity with superposition of the ultrasonic wave (I_{on}) was compared with the intensity without the ultrasonic wave (I_{off}). In Fig. 2, the intensity ratio ((I_{on} / I_{off}) of OH radical was shown as a function of the delay time. Increase in the ratio was observed at the delay time of 20~30 μs. Furthermore, to understand phase dependence on the optical emission intensity ratio of OH radical, ultrasonic waveform at the discharge point was examined by a broadband pressure sensor (PCB Piezotronics, TOYO Corporation). A sinusoidal wave-like pressure change was measured as shown in Fig. 2. Superposition of negative (reduced) pressure at the delay times of 20~35 μs resulted the increase of the intensity ratio of OH radical. Additionally, enhancement of the MB decomposition percentage was observed at the delay time of 25~35 μs. These results suggest that the superposition of the ultrasonic wave induces the enhancement in the number of OH radical and in the decomposition of the MB.

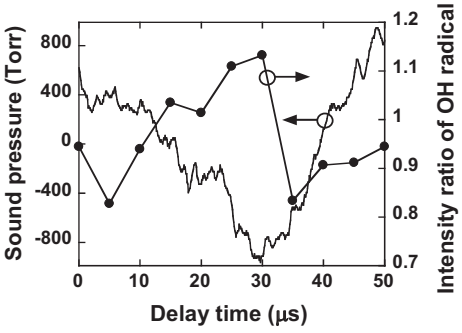

Fig. 2 Optical emission intensity ratio (I_{on}/I_{off}) of OH radical and ultrasonic waveform at the discharge point as a function of the delay time.

4. Summary

Aiming at further improvement of plasma production under water and liquid treatment efficiency using microwave plasma, superposition of the ultrasonic wave was examined. By controlling the discharge timing and the negative pressure phase of the ultrasonic wave, enhancement of optical emission intensity of OH radical and the decomposition of organic compound were confirmed.

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