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Time-Resolved Optical Emission Spectroscopy of Microwave Bubble Plasma in Liquid
マイクロ波励起液中気泡内プラズマの時分解発光分光計測

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A system to produce stable microwave plasma inside size-controlled bubbles in liquid was developed. Time-dependent optical emissions from He I (587.6 nm) and H_α (656.3 nm) those were originated from feed gas and the water vapor, respectively, were investigated varying bubble size, especially focusing on the early stage of the discharge ignition. Rapid increase in the He I emission was observed and was almost the same irrespective of the bubble size. In the case of the H_α emission, however, emission increase was slow compared with those of the He I emissions and the rise time was strongly influenced by the bubble size. This result suggests the bubble size strongly influences the water vapor pressure increase inside the bubbles.

1. Introduction

To date, much attention has been given to plasma-liquid interactions, due to its potential applications to enhance chemical reactions in high density media, and various kinds of plasma production technique in liquids have been studied using dc, ac, rf, and microwaves.

So far, we have developed microwave discharge plasma source under water and have demonstrated effective decomposition of various kinds of organic materials dissolved in the water [1,2]. We have also indicated importance of the water temperature and the water pressure to improve of the discharge efficiency as well as the organic decomposition efficiency [3,4].

To improve the efficiency of plasma decomposition of organic materials inside the bubble of the water, understanding of the reaction processes inside the bubble is indispensable. However, due to difficulty in diagnosing the plasma inside the unstable and fluctuating bubbles, basic research to understand the reaction processes in the bubble plasma is still open problem. To solve this, we have developed a system to ignite plasma inside the bubble under a controlled environment. In this study, temporal variations of optical emissions from the plasma are compared at different bubble sizes.

2. Experimental

Figure 1 shows schematic of experimental set up. Pulsed microwave (peak power<1 kW, pulse width: 6 μs, repetition frequency: 1 Hz) is introduced to a coaxial waveguide with a small hole (1.2 mm in diameter) at the end of the waveguide. A quartz cell of 4.4×4.4×4.0 cm^3 is mounted on the waveguide and is filled with de-ionized water. Discharge gas (He) is introduced into the waveguide through a Piezo valve driven by a pulse generator. By controlling the open duration of the valve, a small bubble is produced on the discharge hole. Temporal variation of the bubble shape is monitored by a CCD camera with an imaging intensifier. The gate width of the image intensifier is 0.6 ms. Atmospheric-pressure microwave plasma is produced between a grounded outer electrode (discharge hole) and an inner needle electrode. Discharge is ignited at various bubble sizes by controlling the discharge timing. Optical emission from the plasma inside the bubble is also measured by a monochromator using a photon counting technique.

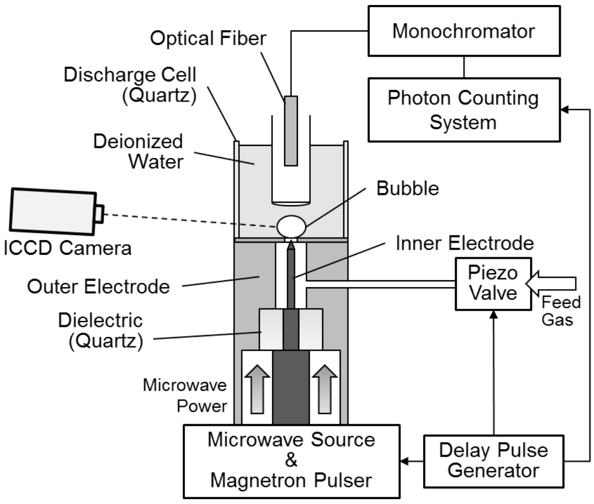

Fig. 1. Schematic of experimental apparatus.

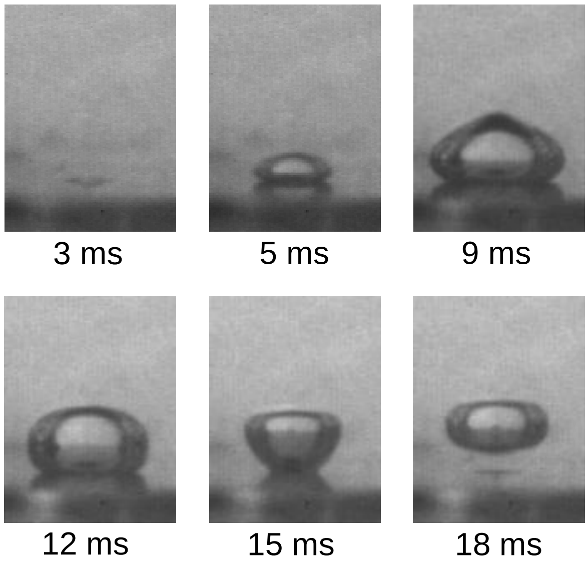

Fig. 2. Temporal variations of bubble size.

3. Results and Discussions

Figure 2 shows temporal variations of the bubble shape after opening the Piezo valve. Here, T_V denotes time after opening the bubble and the valve is closed at T_V=4 ms. Bubble shapes are monitored in every 1 ms using the ICCD camera and typical images at different timings are shown in Fig. 2. First of all, reproducible bubble shape and bubble size are observed from the ICCD measurement. He bubble starts to increase from T_V=4 ms owing to the delay of the pressure increase inside the waveguide. The size of the bubble increases monotonically from 4 to 12 ms and the maximum bubble size is 6.8 mm in diameter and 3.6 mm in height at T_V=12 ms.

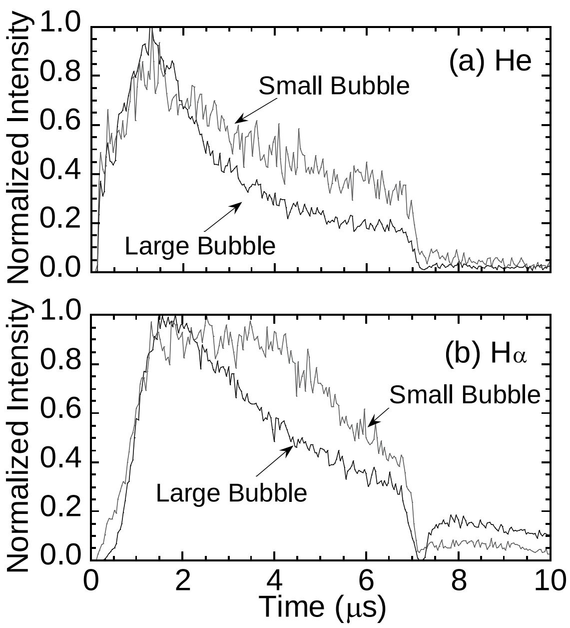

Fig. 3. Temporal variation of (a)He I and (b)H_α emission from the plasma for the cases of small and large bubbles.

To investigate the influence of the bubble size on the discharge characteristics, temporal variations of He and H_α emissions from the bubble plasma at two sizes, i.e. small (T_V =5 ms) and large (T_V =12 ms) was measured. Figure 3 shows He I (587.6 nm) and H_α (656.3 nm) emission intensities normalized by each maximum values during the discharge. Rapid rise of the He emission at the initial phase after the plasma ignition is observed, and the temporal variations of the He I emission is similar in both bubble sizes. In the case of the H_α emission, however, its intensity increases slowly compared with He I emission. Furthermore, time evolution of H_α depends on the bubble size, showing longer rise time of H_α emission at larger bubble size. This result suggests time-dependent vapor pressure variations in the bubble as a result of water evaporation due to the heating from produced plasma inside the bubble and slow increase in the water vapor pressure in the case of large bubbles.

4. Conclusions

We have developed a system to produce microwave atmospheric pressure plasma inside well-reproduced bubbles under water. Time dependences of He and H_α emissions from the plasma were monitored by time-resolved optical emission spectroscopy. Time variations of He and H_α emissions in the early stage of the plasma ignition were measured. Rapid increase in the He intensity was observed irrespective of the bubble size. In the case of the H_α emission, however, rise time was slow compared with that of the He emission and became slower with increasing the bubble size. This suggests that the time dependence of the reactive-species generation is strongly influenced by the bubble size.

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