Characteristics of the Boiling Phenomena in In-liquid Plasma

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Abstract:
The in-liquid plasma is generated in the bubbles in a liquid by micro wave (MW) or high frequency wave (MW) irradiation. The heat flux is approximately one order larger than commonly reported nucleate boiling heat flux. Since heat generation in bubble occurs by the collision of charge particles accelerated by electric field in the plasma, in-liquid plasma is a new boiling phenomenon that uses plasma itself as a heat-source for heating. The electron temperature, electron density, temperature of OH, and behavior of bubbles generated by 27.12-MHz in-liquid plasma are investigated in water under pressures ranging from 1 kPa to 400 kPa. The excitation temperature decreases as the pressure increases and, conversely, the temperature of OH and the electron density increases.

Keywords: in-liquid plasma, boiling, plasma in water, high frequency, bubble

1. Introduction
The in-liquid plasma method is a technology in which plasma is generated inside bubbles in a liquid [1-3]. The behavior of the bubbles basically resembles the sub-cooled boiling phenomena. The plasma does not exist in contact with liquid but exists envelope by vapor generated from the evaporation of the liquid by the heat of plasma. The temperature of the gas of liquid inside plasma can reach more than thousand K under atmospheric pressure, it is expected that the in-liquid plasma can be used as a chemical reactor.

Generating plasma in organic solvents disposes of waste liquids, creates flammable gases [4,5], and enables the synthesis of diamonds [6,7] and SiC [3]. While this indicates that technologies applying in-liquid plasma are rapidly evolving, there still are many areas that remain unclear, such as the discharge characteristics in the plasma, the formation of the plasma, and the pathways of the chemical reactions.

On the other hand, the reaction mechanisms for plasma in water are much simpler that those found in organic solvents; hence, there has been much research on plasma in water in an attempt to clarify in-liquid physical and chemical characteristics [3,8-10]. Since the in-liquid plasma is generated in a liquid, there is much expectation that low-temperature plasma can be maintained without damaging the electrode or substrate and without introducing gases because the liquid provides a cooling effect.

In this research, we generated in-liquid plasma in a ranging from 1 kPa to 400 kPa using a 27.12-MHz high-frequency wave, and the behavior of a bubbles and containing plasma was investigated by a high-speed camera. Moreover, the electron temperature, electron density, temperature of OH, were determined by using spectrometry.

2. Experimental apparatus and procedure
A schematic of the apparatus used in the experiment is shown in Fig. 1. Two observation windows made of transparent quartz glass were placed in a stainless reaction chamber that has a 63.3 mm interior diameter. The electrode was a 3 mm tungsten rod with a semi-spherical tip. The side surface of the rod was covered with quartz glass to reduce the contact surface area between the liquid and the electrode and to concentrate the high electrical field at the tip of the electrode. An aluminum rod with a 6 mm diameter was used as the counter electrode. The distance between the electrodes was approximately 20 mm. After the vessel was filled with approximately 520 mL of pure water, electric power was supplied from a 27.12MHz high-frequency power source. The reflective energy was adjusted to the lowest possible level and plasma was generated from the tip of electrode. After the plasma was generated at the vacuum conditions, isostatic pressing

![Fig. 1 Experimental apparatus](image-url)
equipment was used to gradually increase the pressure inside the reactor. This pressure was then adjusted to the target pressure.

This experiment was conducted with the electric output power fixed at 150W at pressures ranging from 1kPa to 101kPa, and at 400W at pressures ranging from 101kPa to 400kPa. When the pressure was increased between 101kPa and 400kPa, the reflection power was reduced from 200 to 120W. As a result, the increase in system pressure meant that the net supply power increased. At the time of the experiment, it is not possible to adjust the impedance matching of the device within the range of present pressure conditions. However, due to the characteristics of the present device, impedance matching becomes easier as the pressure increased. As the pressure rises, the reflection output decreases. When the experiment is conducted under constant forward power, the result is that the net output of HF power supply increases as the pressure increases. The plasma could not be maintained if the input power was not increased as the pressure increased. In addition, since thermal damage to the electrode occurs at the net output power of more than 400W, a system pressure of 1-400kPa was used.

3. Experimental Results and Observations

Figure 2 shows the results of the relationship between excitation temperature and pressure in the reactor. If the electrons are assumed to have Boltzmann distribution, it is possible to determine the excitation temperature by using the emission intensity ratio of Hα and Hβ [10,11]. In comparison, at 101kPa, the excitation temperature in 200W output was approximately 5% higher than that in the 150W. However, since the change in excitation temperature in relation to the change in pressure showed the same tendency in the data of both experiments, it is indicated that the excitation temperature is being correctly measured. When the system pressure is increased from 1kPa to 400kPa, the excitation temperature of the plasma drops from approximately 4300 to 3200K.

LIFBASE code, a spectrum simulation program, was used to calculate the spectral lines of OH (A-X) band at various temperatures and was used for comparing the OH spectral lines with the spectrum obtained from the experiment [13]. The rotational temperature and vibrational temperature have been assumed to be equal. The sampling time of the OH emission spectrum is 2s. The red line in Fig. 3 is the result of the LIFBASE calculation. There is a close match with the measurement values at 5080K at 400kPa shown in Fig.3. The rotational temperatures of OH at atmospheric pressure and above are presented in Fig. 4. The temperature of OH increases from 1800K to 5000K as the pressure rises.

The emission spectrum in the plasma is affected by the microscopic electric fields from the surrounding ions and electrons, causing Stark broadening. The electron density is calculated by fitting the theoretical profile of the Stark

<table>
<thead>
<tr>
<th>Pressure (kPa)</th>
<th>Electric power (W)</th>
<th>Heat flux (W/m²)</th>
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Figure 2 Measuring excitation temperature

Figure 3 Comparison of the experimental OH (A-X) band and the temperature calculated by LIFBASE code

Figure 4 Rotational temperature of OH (A-X)
broadening of Hβ obtained from a study by Stehlé et al with the data from this experiment [14]. Figure 5 shows the relationship between the electron density and pressure.

The sampling time at each pressure condition was 100s. Doppler broadening was taken into consideration, however, the actual effect was so negligible as to not require consideration in the results. It is assumed that the van der Waals broadening will increase as the pressure rises. Its FWHM is 0.046nm at 5000K. It is speculated that Stark broadening is the dominant pressure broadening at pressures of approximately 400kPa and that the effect of other broadening is small. As the pressure increases, the input power to the electrode must be increased in order to maintain the plasma stably. An absolute minimum power exists for each pressure to stably maintain the plasma. Under stable pressure conditions, even if the net output power of the power supply is increased by approximately 50W, only the plasma generation region will increase; there will be no major effect on the spectrum data of Hβ. Therefore, the increase in electron density is due to the rise in pressure.

When the pressure is increased from 1kPa to 400kPa, the electron density increases from 1.9×10²⁰ to 5.8×10²¹ m⁻³. The number of collisions and separations inside the bubbles increases as the pressure rises. In-liquid plasma can maintain high-density without any special devices, even when the pressure is greater than atmospheric pressure.

Figure 6 shows the relationship between the departure diameter and detachment time at pressures ranging from 101kPa to 400kPa. The volume of the bubbles is calculated from images taken with the high-speed camera, and the departure frequency of bubble f and the bubble departure diameter d when detached from the electrode are investigated. The bubble volume changes periodically. One period, defined as the detachment time (1/f), is considered to be the time from when a bubble detaches from the electrode to the next bubble detachment. The periods were determined by a Fourier transformation of waveforms of the change in bubble volume. The largest diameter in each period was recorded as d.

As the pressure increases, both the detachment diameter and time, corresponding to 1/f, decreases. The value of fd is one of the key parameters in boiling heat transfer [15].

At 101kPa, it is 250mm/s, and at 400kpa, it increases to 350mm/s. While this parameter is used in nucleate boiling, it is more than three to four times larger than the 78 to 90mm/s in Jakob [16] or 111mm/s in Nishikawa [17]. The solid line in the figure is f·(d-2.6) = 142 and all data are on this straight line. If the pressure is increased, the temperature inside the bubbles will increase. Since the degree of superheating increases, the speed of bubble growth increases with the system pressure.

A typical example for in-liquid plasma in water at 400kPa is shown Fig. 7. The entire electrode surface seems to be in contact with the liquid except in the area of the electrical discharge. Since the area where electrical discharge occurs is only a small part of the electrode surface, it is suggested the surface temperature over the entire electrode is not too high. Moreover, the boiling bubbles seem to occur at a location removed from the electrode. Generally speaking, boiling occurs due to heat transfer from a heating surface to a liquid. However, in the boiling phenomenon that occurs in in-liquid plasma, which is a boiling phenomenon in which plasma is the heating source, the liquid is evaporated by the plasma. Heat generation in the bubbles occurs through the collision of charge particles accelerated by electric fields in the plasma.

A typical boiling curve for water at atmospheric pressure is shown in Fig. 8 [18]. While it would not be correct to call the data of this experiment a heat flux because it is not a heat transfer experiment on a heating surface. However, if the value, obtained by dividing the net electric power calculated from the difference between the forward power and the reflected power across the cross section of the electrode, is defined as the apparent heat flux, it would be 2.1×10⁷~4.0×10⁷ W/m², as shown in Table 1. This is a

![Figure 5 Relation between electron density and pressure.](image-url)

![Figure 6 Relation between detachment time and bubble departure diameter.](image-url)
value that is approximately one order larger than commonly reported for nucleate boiling heat flux. Research up to now has reported the gas temperature of plasma to be approximately 3500K under atmospheric pressure. Normally, it would be difficult to reach such a highly superheated state when nucleate boiling occurs on a solid heating surface; however, it is possible to stably generate a boiling high-heat flux with in-liquid plasma. A boiling phenomenon can be maintained utilizing in-liquid plasma that far exceeds the conventional burnout heat flux because heat generation in the bubbles occurs through the collision of charge particles accelerated by electric fields in the plasma.

The degree of ionization of the in-liquid plasma is approximately 0.1%, as estimated from the electron density. However, in high-frequency in-liquid plasma, the electrons remain in the plasma reaction field within the bubbles where temperatures of several thousand degrees are created as a result of the harsh vibration by the high-frequencies. This type of high-temperature reaction field cannot be achieved by pulse discharge, and it is believed that this is a characteristic of high-frequency in-liquid plasma.

4. Conclusions

The following results were attained when the in-liquid plasma in water was generated at pressures ranging from 1kPa to 400kPa.

1. The excitation temperature was reduced from approximately 4300 to 3200K, and the temperature of OH (A-X) was increased from approximately 1800 to 5000K.
2. Increasing the pressure reduces the detachment time and decreases the bubble diameter at departure. The bubble diameter increases linearly relative to the detachment time.
3. In-liquid plasma enables high superheated boiling, which cannot be attained by boiling using a solid heating surface.

Acknowledgements

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References