Optical Emission and Mass Spectra Observations during Hydrogen Combustion in Atmospheric Pressure Microwave Plasma*)

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(Received 12 December 2011 / Accepted 11 April 2012)

We experimentally investigated hydrogen combustion by atmospheric pressure plasma generated by a 2.45 GHz microwave discharge. Small amounts of hydrogen and oxygen were mixed in the operational argon gas during discharge. To clarify the details of combustion, optical emission was measured. The constituents of combustion-processed gases were observed by a quadruple mass spectrometer. The degree of hydrogen oxidation, the so-called conversion rate, increased with input microwave power. The maximum hydrogen conversion rate was greater than 80% under these experimental conditions. During discharge, an optical emission peak due to OH radicals was observed.

Keywords: atmospheric pressure plasma, hydrogen combustion, optical emission spectroscopy, mass spectroscopy, tritium

DOI: 10.1585/pfr.7.2401075

1. Introduction

Recovery of tritium in a nuclear fusion reactor building is an important concern. Current tritium removal systems remove tritium from a gas by cracking the tritium-containing components on a heated precious metal catalyst. The tritium combines with oxygen in the air stream to form tritiated water. Then, the tritiated water contained in the air stream is removed by a molecular sieve bed [1]. Although this system offers adequate efficiency, problems such as a high pressure drop, the use of a large amount of precious metals, and inefficient heating occur when the processing throughput is quite large.

To resolve these problems, we have proposed hydrogen isotope oxidation in atmospheric pressure plasma [2]. Atmospheric pressure plasmas have been widely applied in a variety of fields such as chemical processing and medical and material engineering in recent years [3]. These plasmas have advantages such as low cost and low energy consumption because they can be produced without vacuum equipment. In addition, hydrogen and oxygen radicals are easily generated by high energy electron and ion impacts in the plasma. These radicals play an important role in hydrogen oxidation in gas phase reactions [4–7]. Therefore, it is expected that highly effective oxidation without precious metals can be developed using atmospheric pressure plasma.

In this paper, we report experimental results of optical emission spectroscopy and mass spectroscopy during hydrogen combustion in an atmospheric plasma discharge.

2. Experimental Setup

Figure 1 shows a schematic of the experimental apparatus for hydrogen combustion using a microwave discharge at atmospheric pressure. A 2.45 GHz microwave power supply was connected to a plasma source through an automatic matching box. The maximum output power was 150 W. Note that the effective microwave power that reached the antenna in a plasma source is less than 50% of the output of the power supply. A glass vessel was connected to the plasma source by an NW flange. A moisture absorber was placed upstream of a quadrupole mass spectrometer (QMS) to remove the background moisture and water molecules generated during discharge. Hydrogen, oxygen and argon were used as sample combustion gases. A mixture of these gases was fed into the plasma source through a buffer tank. Each gaseous flow was controlled by mass flow controllers. QMS was used primarily to measure time evolution of hydrogen, oxygen, and argon gases. Simultaneously, visible light emissions from the plasma were observed by a spectrometer through a biconvex lens and an optical fiber. In this experiment, the flow rates of argon, oxygen, and hydrogen were set to obtain a total flow rate of 2.7 ℓ/min, as shown in Table 1. Thus, the concentrations of hydrogen and oxygen were as high as 1.5 vol% and 14.8 vol%, respectively. Microwave power for the discharge

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*) This article is based on the presentation at the 21st International Toki Conference (ITC21).

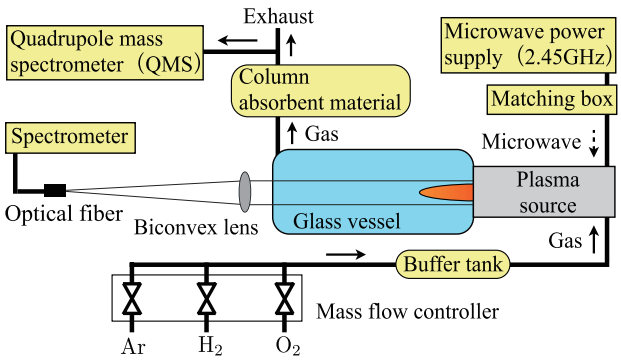

Fig. 1 Experimental setup for hydrogen oxidation using an atmospheric pressure microwave plasma torch.

Table 1 Experimental gas flow rates.

#	Ar [ℓ/min]	O_2 [ℓ/min]	H_2 [ℓ/min]
1	2.29	0.40 (14.8 vol%)	0.01 (0.4 vol%)
2	2.28	0.40 (14.8 vol%)	0.02 (0.7 vol%)
3	2.27	0.40 (14.8 vol%)	0.03 (1.1 vol%)
4	2.26	0.40 (14.8 vol%)	0.04 (1.5 vol%)

was varied from 20 to 100 W by adjusting the output of the microwave power supply. All experimental data shown in this paper were obtained under steady experimental conditions. Generally, steady state can be reached in approximately 15 min after changing conditions such as the input power and gas flow rate.

3. Experimental Results and Discussions

To evaluate the hydrogen combustion performance, the hydrogen conversion rate X_h , is defined as follows [4],

$$X_h = \frac{[H_2]_{in} - [H_2]_{out}}{[H_2]_{in}} \times 100[\%], \quad (1)$$

where $[H_2]_{in}$ and $[H_2]_{out}$ are hydrogen molecule densities that correspond to the ion current of QMS before and after oxidation, respectively.

Figure 2 shows the dependence of the conversion rate X_h on the microwave power. X_h increases with power. Furthermore, X_h appears to saturate at high power. Saturation is attributed to the characteristics of the plasma source because plasma density could also saturate with increasing discharge power under our experimental conditions. A maximum hydrogen conversion rate of more than 80% has been obtained under these experimental conditions.

Figure 3 shows the emission spectra during discharge with a mixture of hydrogen, oxygen, and argon under experimental condition #4 for a 100 W discharge. The optical emission peak from the OH radical was observed. As the input microwave power was increased, the emission intensities of OH radical also increased.

According to Fig. 2, X_h tend to saturate with increasing input power. However, the relationship between argon

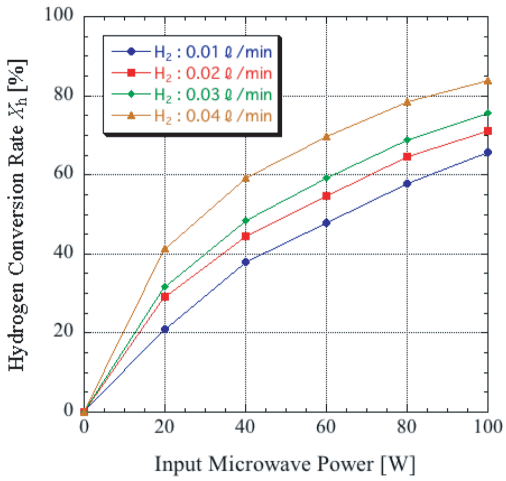

Fig. 2 Dependence of hydrogen conversion rate on input microwave power.

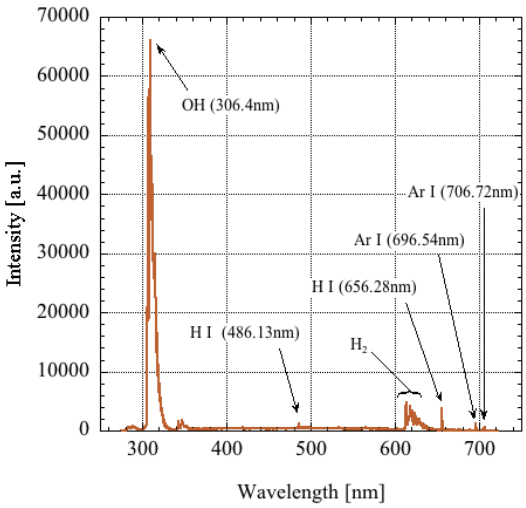

Fig. 3 Optical emission spectra from atmospheric pressure microwave argon plasma with hydrogen and oxygen.

emission intensity and X_h shows a linear dependence that does not saturate, as shown in Fig. 4. Argon intensity is considered to be proportional to plasma density. The observed saturation of X_h might be attributable to the limited plasma density due to the plasma source structure. Hence, if we can increase the plasma density, X_h is also expected to increase.

The relationship between OH emission intensity and X_h also exhibits a linear dependence, as shown in Fig. 5. However, unlike the case in Fig. 4, this dependence is not affected by the hydrogen gas flow rate. OH radical formation is believed to be limited by the amount of oxygen because the flow rate was fixed in these experiments. This dependence suggests that hydrogen oxidation depends strongly on OH radical density. Hence, the OH emission intensity could be used as an indicator of the degree of hydrogen oxidation in a certain quantity of oxygen.

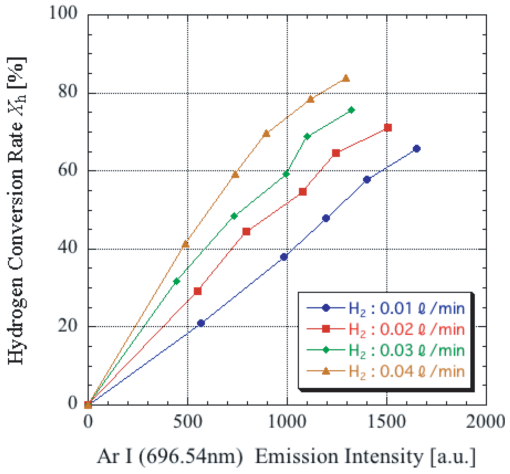

Fig. 4 Dependence of hydrogen conversion rate on ArI emission intensity.

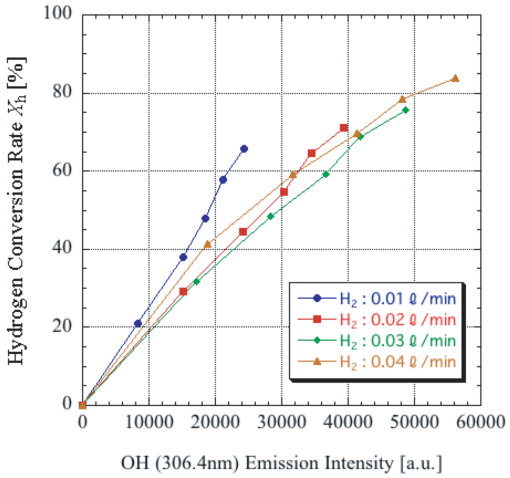

Fig. 5 Dependence of hydrogen conversion rate on OH emission intensity.

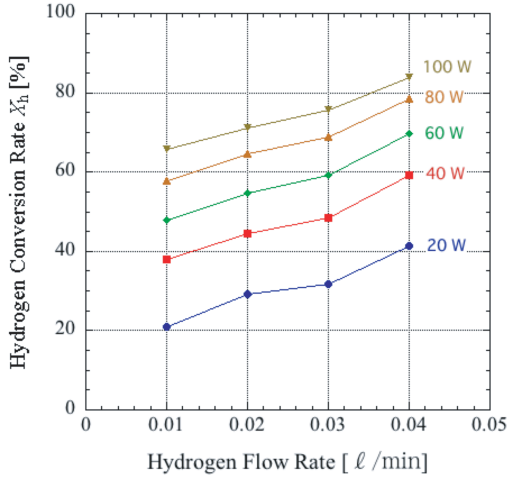

Fig. 6 Dependence of hydrogen conversion rate on hydrogen gas flow rate.

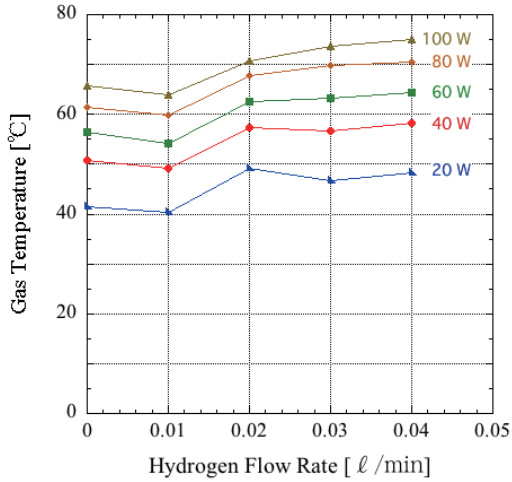

Fig. 7 Dependence of gas temperature on hydrogen gas flow rate.

Under our experimental conditions, X_h increased by approximately 20% as the hydrogen flow rate increased from 0.01 to 0.04 ℓ /min for each input microwave power, as shown in Fig. 6. A higher concentration of hydrogen causes added hydrogen oxidation.

Because hydrogen oxidation is a heating process, gas temperature in the reaction chamber is expected to increase with X_h. Gas temperature, which was measured by a thermocouple located 50 mm downstream from the plasma exit, is shown in Fig. 7. Although X_h increases with discharge power, gas temperatures change slightly with the hydrogen flow rate. Even in the absence of hydrogen, gas temperature became approximately equal to the temperature during discharge with hydrogen. Because gas temperature without plasma is equivalent to room temperature, this result indicates that the increase in gas temperature is mainly due to argon discharge and not oxidation. Although most of the hydrogen is believed to oxidize in the plasma source, the heated gas might already have been cooled at the measured position.

4. Conclusions

Hydrogen combustion process and fundamental characteristics during atmospheric pressure microwave discharge were investigated by optical emission and mass spectroscopy observations. Increases in X_h, each emission intensity, and the gas temperature with discharge power were confirmed. Under these experimental conditions, X_h tends to saturate with increasing input microwave power. The saturation is attributed to the properties of the plasma source because plasma density also saturates with increasing discharge power under our experimental conditions. The hydrogen conversion rate would be increased by using a high density plasma. Because hydrogen oxidation depends strongly on OH radical density, OH emission intensity could be used as an indicator of the hydrogen con-

version rate X_h. Under our experimental conditions, gas temperature changes slightly with changes in the hydrogen flow rate even though X_h increases with discharge power.

To clarify the details of the characteristics of combustion, additional spectroscopic measurements and investigation of the plasma parameters are required. Furthermore, the effect of gas temperature on X_h should be investigated.

Acknowledgments

This study was supported by the NIFS Collaborative Research Program (NIFS09KFSS015, NIFS11KESA005) and the Research Foundation for Electrotechnology of Chubu, Grant-in-Aid (R-21244).

[1] DOE Handbook: Tritium Handling and Safe Storage, DOE-HDBK-1129-99 (1999).
[2] K. Akahane, N. Ezumi, Y. Uesugi, Y. Tanaka, M. Tanaka, and K. Nishimura, Fusion Sci. Technol. **60**, 1343 (2011).
[3] A. Fridman, *Plasma Chemistry* (Cambridge University Press, New York, 2008).
[4] S. Kambara, R. Kuriyama, T. Osakabe and K. Yukimura, International Journal of Hydrogen Energy **33**, 6792 (2008).
[5] T. Osakabe, S. Kambara, R. Kuriyama, A. Koyano, K. Yukimura and H. Moritomi, Journal of the Combustion Society of Japan **50**, 136 (2008) in Japanese.
[6] A. Sarani, A.Y. Nikiforov and C. Leys, Phys. Plasmas **17**, 063504 (2010).
[7] S. Li, Q.Y. Wu, J. Zhang, D. Wang and H.S. Uhm, Phys. Plasmas **17**, 063506 (2010).