Investigation of Hydrogen Isotope Combustion Processes in Atmospheric Pressure Plasma

大気圧プラズマによる水素燃焼反応過程の解明

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Atmospheric pressure plasmas have many advantages for widespread applications since no necessary to use vacuum equipments. In this study, preliminary experiments of hydrogen oxidation using an atmospheric pressure plasma have been done with a small amount of hydrogen and oxygen in an argon plasma. As a result of mass spectrometric measurement, we have confirmed that hydrogen could be oxidized during the atmospheric pressure plasma discharge. The hydrogen conversion rates depend on the input power for discharge. Moreover, optical emission spectra measurements indicate that OH radical and O radical would be important role for the oxidation process.

1. Introduction

Recovery of tritium in a nuclear fusion reactor building is one of the important issues. So far, the tritium removal system removes tritium from a 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 has suitable performance efficiency, there are some problems for such as high pressure drop, the utilization of a large amount of precious metals and heating efficiency etc, when the processing throughput is quite huge.

On the other hands, atmospheric pressure plasmas are widely applied in a variety of fields such as chemical processing, medical and material engineering in the recent years [2]. These plasmas have advantages with respect to the cost effect and the low energy consumption since those can be produced without vacuum equipments. Also, hydrogen and oxygen radicals are easily generated by high energy electron and ion impact in the plasma. These radicals play an important role in hydrogen oxidation in the gas phase reactions [3-6]. Therefore, it is expected that a highly effective oxidation process without precious metals can be constructed by using the

atmospheric pressure plasma.

The purpose of this study is to investigate the feasibility of new hydrogen isotope oxidation processing using atmospheric pressure plasma generated by microwave discharge.

2. Experimental Setup

A schematic diagram of the experimental apparatus for hydrogen oxidation using a microwave discharge in atmospheric pressure is shown in Fig. 1. A 2.45 GHz microwave power supply is connected to a plasma source through an automatic matching box. The maximum output power is 150W. Notice that the effective microwave power reached at antenna in a plasma source is less than 50% of the output of the power supply. A glass vessel is connected to the plasma source with a NW flange.

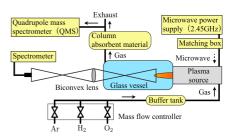


Fig. 1. Experimental setup for hydrogen oxidation using an atmospheric pressure microwave discharge.

A moisture absorbent is put at upstream of a quadrupole mass spectrometer (QMS) to remove the background moisture and generated water molecule during the discharge. H₂, O₂ and Ar were used as the sample combustion gas. The mixture gas of H₂, O₂ and Ar was fed into the plasma source through a buffer tank. Each gaseous flow was controlled by mass flow controllers. The QMS mainly measures the time evolution of hydrogen, oxygen and argon gas. Simultaneously, visible light emissions from the plasma are observed by a spectrometer through a biconvex lens and an optical fiber. In this experiment, the flow rates of H₂, O₂ and Ar are set 0.04 SLM, 0.40 SLM and 1.56 SLM, respectively. Thus, the concentrations of hydrogen and oxygen are corresponding to 2.0 vol% and 20.0 vol%, respectively. The microwave power for discharge are changed from 20 W to 100 W as the output power of the power supply.

3. Results and Discussion

In order to evaluate the performance of the hydrogen oxidation, we introduce the hydrogen conversion rate X_h , defined as follows [3],

$$X_{\rm h}$$
 (%) = $\frac{[{\rm H_2}]_{\rm i} - [{\rm H_2}]_{\rm o}}{[{\rm H_2}]_{\rm i}} \times 100$ (1)

where $[H_2]_i$ and $[H_2]_o$ are the hydrogen molecule densities which correspond to ion current of the QMS before and after oxidation reaction, respectively.

Figure 2 shows the dependence of conversion rate X_h on the microwave power. X_h increase with increasing the power. Furthermore, the X_h seems to saturate in high power region. The saturations would be due to the characteristics of the plasma source because the plasma density could also saturate with increasing the discharge power in our experimental condition. Maximum hydrogen conversion rate has been reached to 85.5% in this experimental condition.

Figure 3 shows emission spectra during the discharge with a mixture of H_2 , O_2 and Ar. The optical emission spectra from OH radical and O radical are observed. It is found that the emission intensities of OH radical and O radical for 100W are higher than 20W. These results indicate OH radical would be key role to oxide hydrogen gas.

4. Summary

A feasibility of hydrogen oxidation process and the fundamental characteristic during atmospheric pressure microwave discharge have been investigated. It has been confirmed that the oxidation is possible under the atmospheric pressure plasma condition. Increasing of the hydrogen conversion rate with increasing the discharge power has been clarified. These results indicate that the rate can be controlled by the discharge power which is related to plasma density and temperature.

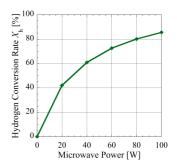


Fig. 2. Dependence of hydrogen conversion rate on the input microwave power.

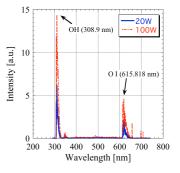


Fig. 3. Optical emission spectra from the atmospheric pressure microwave argon plasma with hydrogen and oxygen.

Acknowledgments

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References

- [1] DOE HANDBOOK: TRITIUM HANDLING AND SAFE STORAGE, DOE-HDBK-1129-99, (1999).
- [2] A. Fridman: "Plasma Chemistry," Cambridge University Press, New York, (2008).
- [3] S. Kambara, R. Kuriyama, T. Osakabe and K. Yukimura, International Journal of Hydrogen Energy, **33**, 6792 (2008).
- [4] 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].
- [5] A. Sarani, A. Y. Nikiforov and C. Leys: Physics of Plasmas, **17**, 063504 (2010).
- [6] S. Li, Q. Y. Wu, J. Zhang, D. Wang and H. S. Uhm: Physics of Plasmas, 17, 063506 (2010).