Characteristics of hydrogen combustion assisted by an intermittent dielectric barrier discharge

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Combustion characteristics of molecular hydrogen mixed with excess oxygen diluted with molecular nitrogen ($H_2/O_2/N_2$) in the intermittent dielectric barrier discharges (DBD) were investigated to remove safely hydrogen in an off-gas from fuel cell vehicles. The discharge was formed the gap volume between electrodes at an applied peak-to-peak voltage of 12-15 kV at an atmospheric pressure. The discharge power was between 0.3 kW and 2.0 kW. The hydrogen conversion was increased with increasing the applied voltage and the repetition rate corresponding to an increase in the discharge power. A 98.8% hydrogen conversion was obtained at the discharge power of 2.0 kW and the treated exhaust gas temperature of 80 °C. It was experimentally found that the gas temperature and the discharge power mostly contribute for increasing of the hydrogen conversion.

1. Introduction

Modern transportation system depended on petroleum has led to serious energy depletion and environmental problems such as emission of nitrogen oxide and particulate matter. Particularly, emission of substantial greenhouse gas, CO_2, from automobiles has played a major role in the recently observed global warming[1)]. To moderate emission of carbon dioxide and environmental pollutants from automobiles, petroleum electric hybrid vehicles (PEHVs) and fuel cell vehicles (FCVs) have been developed in recent decades. Nowadays PEHVs are becoming popular as environmentally friendly cars having high energy efficiency and CO_2 free.

Development of FCVs in world wide automobile companies have been done aggressively as national research projects and their company research, however, they have historically some barriers such as cost cuts of the vehicle and fuel storage issues in commercialization[2,3)]. Moreover, they have a potential issue that is treatment of off-gas containing high concentration of hydrogen from fuel cell.

The effort of the off-gas treatment by catalytic combustion of hydrogen was first observed in a patent in 1991[4)]. In recent years, dilution technique of off-gas by air[5-6)] and hydrogen combustion method using a small burner[7)] were developed. However, these techniques dose not reach essential solution because costly platinum catalyst is use in catalyst combustion, on the other hand, diluted hydrogen may have risk of an explosion under exceptional conditions. In response to the issues, we propose direct treatment of hydrogen mixed with excess air in an intermittent dielectric barrier discharges (DBD).

Starikovskiil[8)] investigated effect of pulsed nanosecond discharges on ignition delay of mixture of hydrogen and hydrocarbons. It was found that the ignition was accelerated by non-equilibrium excitation, and a dominant reaction path was $H + O_2 \rightarrow OH + O$. This suggests that rapid combustion of hydrogen may be caused in a DBD that can generate H and O radicals. No research has yet been carried out hydrogen combustion in a DBD.

The aim of this study is to obtain combustion characteristics of molecular hydrogen mixed with excess air ($H_2/O_2/N_2$) in the DBD. Behaviour of hydrogen conversion was investigated by change in dominant parameters such as applied voltage, repetition rate, and reaction time.

2. Experimental

The design of DBD for hydrogen combustion is shown in Figure 1. Coaxial electrodes geometry with quartz glass tube was used as a device of an atmospheric pressure DBD. The high voltage electrode of 50 mm in diameter and 280 mm long was inserted into the inner tube, while the grounded electrode (0.1 mm thick and 200 mm long) was wounded around the outer tube. The gap between the outer and inner tube is 1.5 mm. The DBD occurred at the gap volume within the length of the grounded electrode. An intermittent one-cycle sinusoidal (OCS) output was adopted as a power source for generating non-equilibrium plasma. The discharge power of OCS, P, is measured by an electricity meter.

Figure 2 shows a waveform of applied OCS voltage without a load of DBD. The applied voltage V_{pp} which is measured by means of a high voltage probe is defined as peak-to-peak voltage. The duration of one cycle waveform T_0 is approximately 10 μs. The repetition rate R_R is given as the frequency of the repetition time T_1 per second.

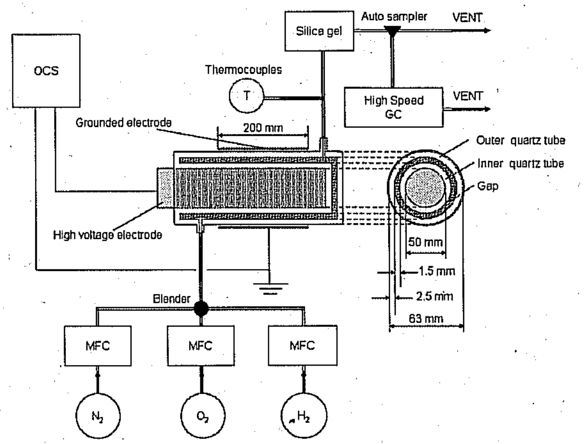

Figure 1. Experimental setup for hydrogen combustion supported by DBD.

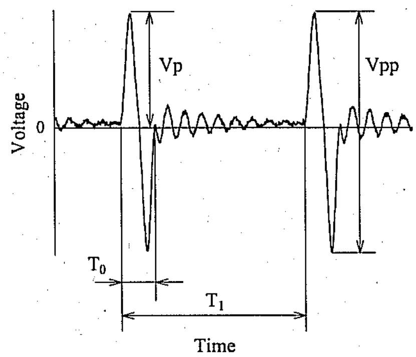

Figure 2. Waveform of voltage supplied from a one-cycle sinusoidal power source.

Table 1 summarizes experimental conditions in this work In order to investigate fundamental characteristics of hydrogen combustion in the DBD, the R_R was varied from 7 kHz to 15 kHz at a fixed V_{pp} of 12 kV or 15 kV. The other experiments under a constant discharge power were examined closely at the extensive R_R in the range between 2.1 kHz and 49.6 kHz corresponding to a discharge power between 0.3 kW and 1.5 kW.

The concentrations and flow rate of the reactant gas ($H_2/O_2/N_2$) were adjusted in a gas blender with mass flow meter. The H_2 and O_2 concentrations were prepared to approximately 2.0 vol% and 19.5 vol%, respectively. For all experimental condition, the flow rate of the reactant gas that is fed into the gap volume was fixed 4.0 l/min at standard conditions for temperature and pressure (SLM). The equivalence ratio φ in the reaction ($H_2 + 1/2O_2 \rightarrow H_2O$) is approximately 0.05, namely, oxygen in the reactant gas is enough excess for hydrogen combustion. The concentrations of H_2 and O_2 in the reactant gas before and after plasma treatments are analyzed rapidly by a micro gas chromatography (Agilent 300A) The hydrogen conversion, X_h, is defined as follows:

$$X_h,\ \% = \frac{[H_2]_i - [H_2]_o}{[H_2]_i} \times 100 \qquad (1)$$

where subscripts i and o denote input and output conditions, respectively, and brackets denote concentrations.

Table 1. Experimental conditions

V_{pp} [kV]	R_R [kHz]	P [kW]
12, 15	7, 10, 15	0.3 - 2.0
12	2.1 - 7.8	0.3
12	4.7 - 14.4	0.5
12, 13, 14	7.4 - 33.6	1.0
12	15.9 - 49.6	1.5

3. Results and discussion

3.1. Fundamental characteristics of hydrogen combustion

Figure 3 shows variation of the concentration of H_2 and O_2, the hydrogen conversion X_h, and the temperature of the exhaust gas T_g versus treatment time at V_{pp} of 15 kV and R_R of 10 kHz. An initial H_2 concentration of 2.0 % is reduced by hydrogen combustion in the DBD, which is decreased to 0.03 % at treatment time of 880 s. The X_h increases suddenly during early treatment time, and achieves finally up to 98.6 %.

As a particular phenomenon of hydrogen combustion by direct treatment in the DBD, an increase of the exhaust gas temperature is observed. It is noted that X_h are closely related to the exhaust gas temperature as shown in Fig.3. Therefore, hydrogen conversion by DBD has to be characterized as a function of the exhaust gas temperature.

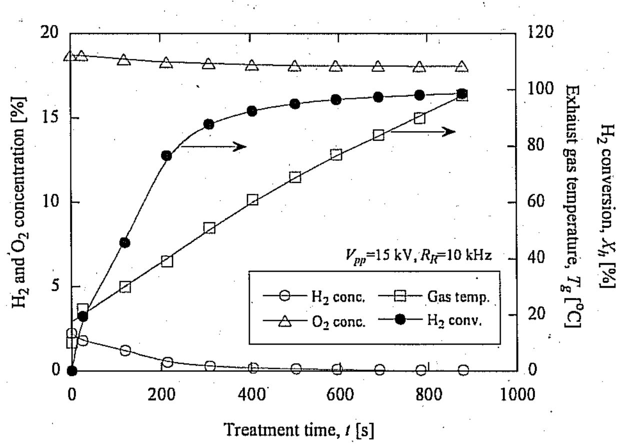

Figure.3 Characteristics of H_2 combustion by direct treatment in the DBD.

3.2. Effect of exhaust gas temperature, applied voltage and repetition rate

Figure 4 shows the variation of hydrogen conversion at various exhaust gas temperatures as parameters of the applied voltage and the repetition rate. H_2 and O_2 concentrations were measured when T_g was reached to the designated temperature (20, 40, 60, and 80 °C), and X_h was calculated by equation (1). In Fig.4, it is found that X_h increases with increasing T_g, V_{pp} and R_R.

An increase in temperature of exhaust gas will certainly accelerate combustion rate of molecular hydrogen. An increase in V_{pp} and/or R_R will generate a large amount of H, O and OH radicals in the DBD, which also accelerate the reaction rate of hydrogen combustion.

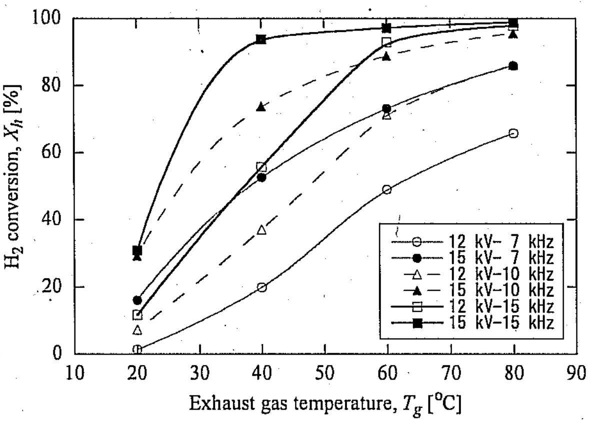

Figure 4. Variation of H_2 conversion with exhaust gas temperature for various V_{pp} and R_R settings.

Figure 5 shows the hydrogen conversion X_h for various exhaust gas temperature as a function of discharge power. For each gas temperature, the hydrogen conversion is related to the discharge power by the following equations under a constant hydrogen concentration in this study.

$$X_h = 1 - \exp\left(-k \times P^2\right) \quad (2)$$

$$k = -0.457 + 2.938 \times 10^{-2} \cdot T_g \quad (3)$$

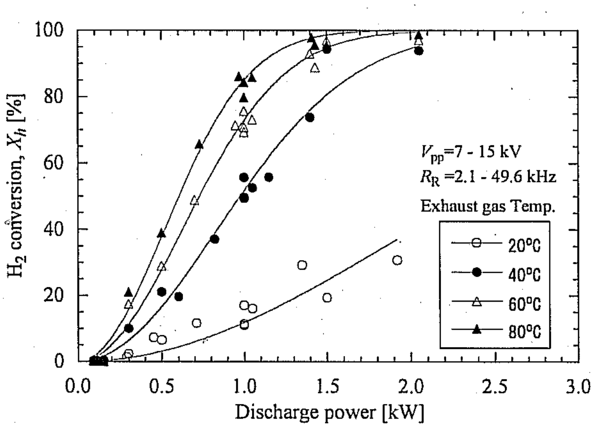

Figure 5. Correlation between discharge power and hydrogen conversion under constant temperatures.

It was found that X_h was directly proportional to the square of the discharge power under these experimental conditions. If the hydrogen concentration becomes higher, the apparent combustion rate constant k is increased. Therefore, the real off-gas containing high hydrogen concentration more than 90 vol% may oxidized completely at lower temperature than the temperature in this study.

Fortunately, detailed mechanisms for H_2/O_2 combustion have been studied by some researchers[9]. It is estimated that O radical is most effective radicals for hydrogen combustion in the DBD from the early studies. Hydrogen combustion mechanisms in the DBD will elucidate by measurement of key radicals by a spectrometer near future.

4. Conclusion

Combustion characteristics of molecular hydrogen in the intermittent dielectric barrier discharges were investigated to remove safely hydrogen in the off-gas from fuel cell. The hydrogen conversion was increased with increasing the applied voltage and the repetition rate. The exhaust gas temperature affects also the hydrogen conversion.

A 98.8% hydrogen conversion was obtained at the discharge power of 2.0 kW. Relation between hydrogen conversion and discharge power was made clear as first-order rate equation.

Eventually, it is found that the direct treatment of hydrogen in DBD is a suitable method for hydrogen treatment in the off-gas.

5. References

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