

## Micro Fuel Reformer Integrated with a Combustor and a Microchannel Evaporator

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### Abstract

This paper describes the development of a micro fuel reformer integrated with a combustor and an evaporator. The reforming test was performed by using a mixture of methanol and water (steam carbon ratio 1.9) as reforming fuel and hydrogen as combustion fuel. It was found that the design of the microchannel evaporator is critical to obtain larger hydrogen output. Hydrogen output and CO concentration were investigated by varying the input combustion power at different fuel feeding rates. 26.1 sccm of hydrogen, which is equivalent to 4.7 W in LHV (low heat value), was produced, when input combustion power was 11 W.

*Keywords: fuel reformer, combustor, evaporator, fuel cell, microreactor*

### 1. INTRODUCTION

A DMFC (direct methanol fuel cell) uses liquid fuel (methanol), which is easy to store and handle. However, it still suffers from low power density due to sluggish electrochemical oxidation of methanol and methanol cross-over, although many researchers are trying to solve these problems [1, 2]. On the other hand, a hydrogen PEMFC (proton exchange membrane fuel cell) has a much higher power density than the DMFC, but hydrogen storage is impractical for portable applications. A micro fuel reformer to convert liquid fuel to hydrogen possibly gives the solution.

Recently, small fuel reformers are under development in the world [3-6]. Casio Computer developed a micro methanol reformer integrated with a combustor, an evaporator and a CO remover [5]. It consists of 13 glass layers stacked by anodic bonding. They connected the methanol reformer to a PEMFC, and confirmed 2.6 W electric output. Pacific Northwest National Laboratory developed a smaller methanol reformer integrated with a combustor, an evaporator and a CO methanizer [6]. It produced 3.55 sccm of hydrogen, which is equivalent to 0.64 W in LHV (low heat value).

This study aims to develop a MEMS-based methanol reformer with higher power density. This paper reports the evaluation results of a prototype integrated with a combustor and an evaporator.

### 2. SYSTEM CONCEPT

Figure 1 shows the system diagram of the fuel cell with a fuel reformer. This system is composed of fuel tanks, micro valves, an air pump, the fuel reformer and a fuel cell. The

fuel reformer is composed of an evaporator, a reforming reactor, a CO remover and a combustor.

Methanol and water are first fed to the evaporator from pressurized tanks through the micro valves. The evaporation of methanol and water need heats of 35 kJ/mol and 40 kJ/mol, respectively, which are supplied from the combustor. The evaporated fuel is converted to H<sub>2</sub> and CO<sub>2</sub> using a catalyst in the reforming reactor. The total reaction of methanol steam reforming is given by



Actually, CO, which is harmful for PEFCs (polymer electrolyte fuel cells), produces in the steam reforming reaction. Thus, a CO remover should be installed after the reforming reactor. Finally, the treated reformed gases are supplied to the fuel cell.

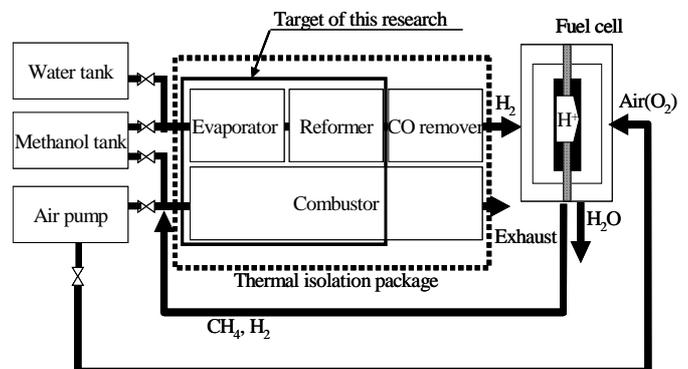
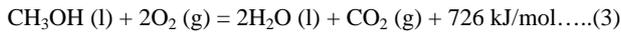


Figure 1. System diagram of the fuel cell system

The heat required for these reactions is generated by the combustor using the following reaction:



Instead of this methanol combustion, residual hydrogen and methane in exhaust gas from the fuel cell can be burned. The total input energy is the chemical energy of supplied methanol, and the total output energy is that of produced hydrogen, which is a part of the total input energy. The other is used to supply the heating energy, latent heat and reaction energy of the fuel, and to heat the fuel reformer. The energy to heat the fuel reformer significantly depends on its thermal isolation performance, and should be minimized for high efficiency.

The efficiency of the methanol steam reforming,  $\eta$ , is defined by

$$\eta = \frac{\Delta H_c(\text{H}_2)}{\Delta H_c(\text{CH}_3\text{OH}) + Q(\text{Heating energy})} \dots (4)$$

Here,  $\Delta H_c(\text{H}_2)$  is the chemical energy of produced hydrogen in HHV (high heat value),  $\Delta H_c(\text{CH}_3\text{OH})$  is that of methanol fed to the reforming reactor, and  $Q$  is the heating energy generated by the combustor, that is, the chemical energy of a fuel fed to the combustor. To reduce  $Q$  for high  $\eta$ , the thermal isolation package is used as shown in Fig. 1.

In this study, we fabricated the micro fuel reformer integrated with a microchannel evaporator and a catalytic combustor, which is the core element of the system.

### 3. STRUCTURE

Figure 3 shows the structure of the micro fuel reformer. It is composed of the reforming chamber, the catalytic combustor and the microchannel evaporator. The combustion chamber is fabricated in a glass substrate by sandblasting, and covered by silicon substrates from both sides. The inside of the combustor is coated with a Pt catalyst loaded on a  $\text{TiO}_2$  support made by a sol-gel method. The reforming chamber is also fabricated in a glass substrate, and bonded to the silicon substrates. The reforming catalyst is specially made from a commercially-available catalyst made of  $\text{CuO}/\text{ZnO}/\text{Al}_2\text{O}_3$  (Süd-Chemie Catalysts Japan, Inc.). The microchannel evaporator is fabricated in the silicon substrates. There are 20 microchannel evaporators positioned just before the reforming chamber.

Figure 4 shows the SEM image of the microchannel evaporators. We fabricated six kinds of microchannel evaporator shown in Table 1. Narrower channels give larger heat-transferring surface and higher pressure loss to prevent liquid-phase fuel from entering the reaction chambers. However, too narrow channels can limit the output. Thus, the better size of the channel is found by comparing the different channels.

Figure 5 shows the photograph of the micro fuel reformer. The outer size of the combustor is 25 mm × 20 mm × 5.6 mm.

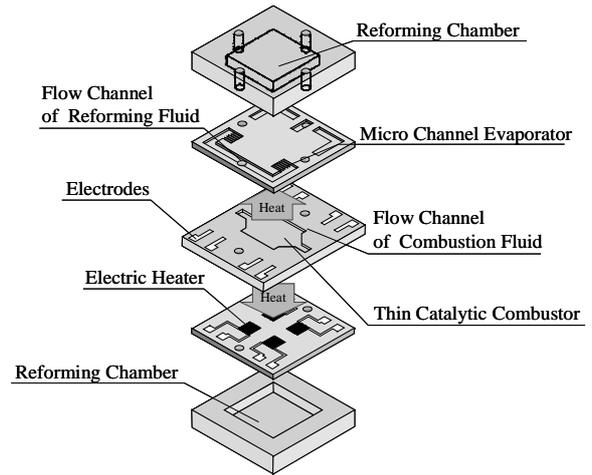


Figure 3. Structure of the fuel reformer

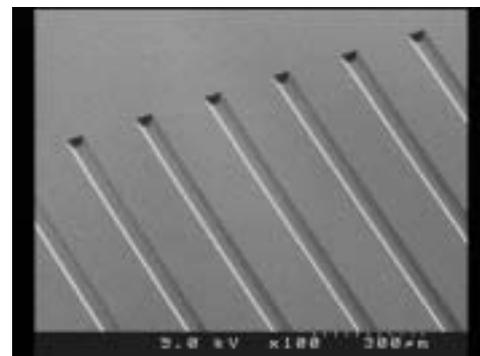


Figure 4. SEM image of the microchannel evaporator

Table 1 Size of microchannel evaporators

Sample	Width	Depth	Length
1	50 mm	30 mm	4 mm
2		19 mm	1 mm
3		31 mm	
4		51 mm	
5		85 mm	
6	2 mm	100 mm	

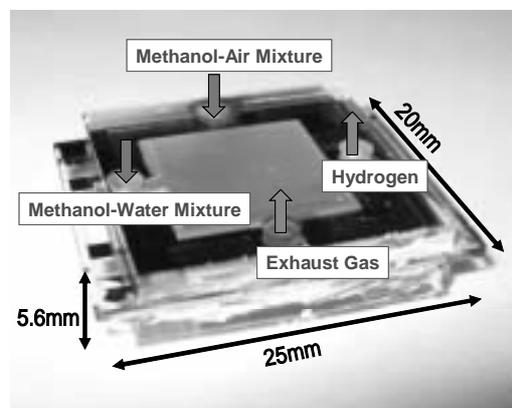


Figure 5. Prototyped fuel reformer

## 4. EXPERIMENT

### A) Experimental Setup

Methanol steam reforming test was performed using the setup shown in Fig. 6. Hydrogen and dry air are supplied to the combustor from gas cylinders through mass flow controllers (MFC) after premixed. The outlet of the combustor is connected to a gas chromatography (GC) system (GC-14B with SHINCARBON-ST column, Shimadzu), only when the combustion efficiency is measured. In other cases, it is opened to atmosphere. The surface temperature of the fuel reformer is measured using an infrared thermometer (TVS-8500, Avionics), and the temperature inside the reforming chamber is measured using a thin film resistive temperature sensor fabricated on the silicon substrate.

A pressure sensor is set before the inlet of the combustor. To measure the pressure loss of the reforming chamber, the air gas line and the pressure sensor are temporarily connected to the reforming chamber.

Methanol and water to be reformed are premixed at a S/C (steam/carbon) ratio of 1.9, and stored in a syringe. The mixture is supplied using a syringe pump at constant feeding rates. The outlet of the reforming chamber is connected to a film flow meter, when the flow rate of reformed gas is measured. To measure the composition of the reformed gas, a gas chromatography system is connected to the outlet of the reforming chamber.

### B) Pressure Loss Measurement

First, the pressure loss of the flow channel through the inlet, microchannel evaporator, reforming chamber and outlet was measured using air. The results for the 6 different microchannel evaporators are shown in Fig. 7. The pressure loss varies from 5.3 kPa/scm to 0.002 kPa/scm, and hereafter these samples are called as Samples 1 to 6, respectively.

Next these samples with different pressure losses at the microchannel evaporator were compared about hydrogen production performance at a S/C of 1.9 and a fuel flow rate of 10  $\mu\text{l}/\text{min}$ . The result is shown in Fig. 8. The data plotted in Fig. 8 are the average in 20 measurements. Each error bar indicates that there was the fluctuation of fuel flow rate due to the pulsation of fuel evaporation.

The hydrogen flow rate increases with decrease in the pressure loss from 5.3 kPa/scm, and saturates around 0.6-0.2 kPa/scm, although the fuel flow rate is constant at 10  $\mu\text{l}/\text{min}$ . This can be because the fuel enters the reforming chamber partly in liquid phase by capillary force when the depth of the microchannel evaporators is too small. Sample 6 with the pressure loss of 0.002 kPa/scm produces little hydrogen, because the liquid-phase fuel enters the reforming chamber by the vapor pressure of the fuel. The design of the microchannel evaporator is critical to maximize hydrogen production. We found the optimized pressure loss of the microchannel evaporator of 0.2-0.6 kPa/scm in this experiment, but the fluctuation of fuel flow must be solved.

### C) Reforming Performance of Sample 4

A further methanol steam reforming test was performed by varying the input combustion power from 7 W to 12 W

using the Sample 4 with a pressure loss of 0.6 kPa/scm at the microchannel evaporator.

The feeding rate of the reforming fuel was set at 10, 20, 40  $\mu\text{l}/\text{min}$ . Figure 9 shows the power of produced hydrogen in LHV and the concentration of CO as functions of the input combustion power. Only one point was taken at a fuel feeding rate of 40  $\mu\text{l}/\text{min}$ , because the connection of the fuel tube was broken by the fuel pressure. The hydrogen powers in Fig. 9 are the average in 20 measurements. In every 20 measurements, the concentration of hydrogen measured by GC was almost constant at 75 %, although the flow rate fluctuated as mentioned. Thus, the concentration of CO was measured one on each condition.

At a fuel feeding rate of 10  $\mu\text{l}/\text{min}$  and 20  $\mu\text{l}/\text{min}$ , the quantity of hydrogen increases with increase in the input combustion power until 8 W and 10 W, and saturate around 1.5 W and 3 W, respectively. These values roughly match the theoretically obtainable values, if measurement errors are taken into consideration. From this result, it is found that 8 W and 10 W is the minimum required input combustion power for the fuel feeding rate of 10  $\mu\text{l}/\text{min}$  and 20  $\mu\text{l}/\text{min}$ , respectively, although only 0.36 W is theoretically required for the reforming and evaporation of 10  $\mu\text{l}/\text{min}$  fuel. The other combustion power was mostly consumed to heating the fuel reformer, and must be saved by thermal isolation package.

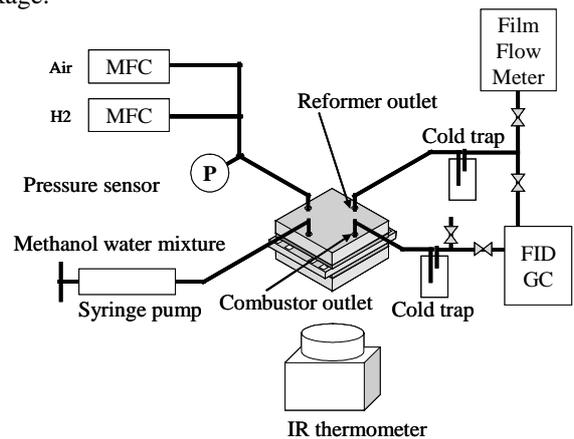


Figure 6. Experimental setup

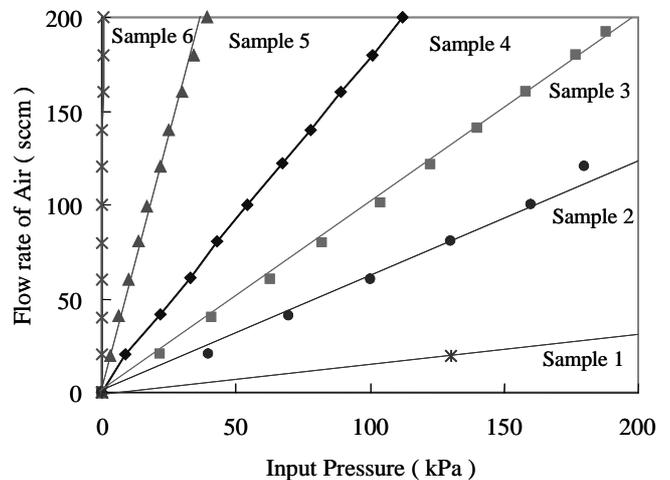


Figure 7. Pressure loss of the reforming flow channel

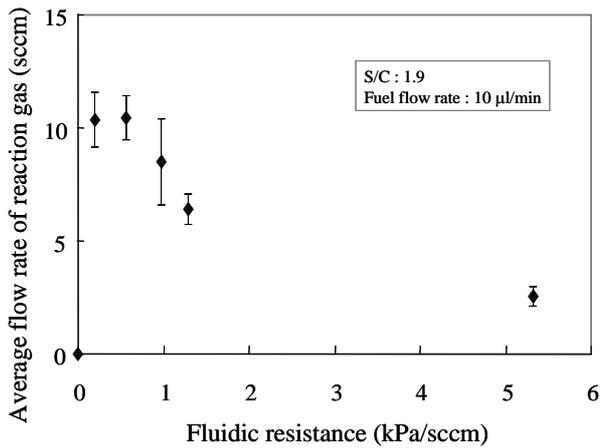


Figure 8. Relationship between pressure loss and hydrogen production performance

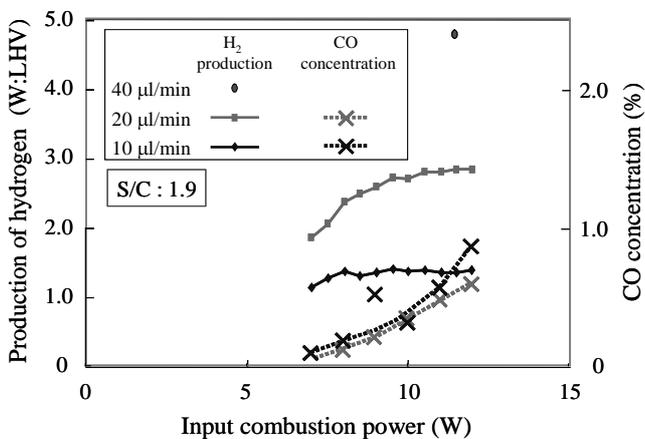


Figure 9. Hydrogen production performance and CO concentration in reformed gas

The CO concentration monotonously increases with increase in the input combustion power as shown in Fig. 9. On the other hand, the hydrogen output saturates at a certain input combustion power as mentioned. Thus, not only to raise the efficiency but also to reduce CO, the input combustion power must be controlled at the optimal value of 8 W and 10 W for a fuel feeding rate of 10 µl/min and 20 µl/min, respectively. The CO concentration of the as-reformed gas is too high to supply to PEMFCs. A methanizer, which converts CO to methane, will be connected to the reforming chamber. The maximum flow rate of hydrogen of 26.1 sccm was obtained, when the input combustion power was 11 W and the fuel feeding rate was 40 µl/min. The equivalent power of the produced hydrogen is 4.7 W in LHV. The maximum efficiency of 24.8 % given by Eq. (4) was obtained, when the combustion power was 9.5 W and the fuel feeding rate was 20 µl/min.

The hydrogen generation rate has already reached the level for practical uses, so the micro fuel reformer becomes practical if it is connected to the methanizer and packaged for thermal isolation.

## 5. CONCLUSION

The micro fuel reformer is composed of a combustor, an evaporator, a reforming chamber and a temperature sensor. The reforming test was performed by using mixture of methanol and water (steam carbon ratio 1.93) as a reforming fuel and hydrogen as a combustion fuel. Comparing different depths of the microchannel evaporator found that the design of the microchannel evaporator is critical to obtain larger hydrogen output.

Hydrogen output and CO concentration were investigated by varying the input combustion power at the fuel feeding rates of 10, 20 and 40 µl/min. The concentration of hydrogen in the reformed gas measured by gas chromatography was almost 75 mol % on each condition. 26.1 sccm of hydrogen, which is equivalent to 4.7 W in LHV (low heat value), was produced, when the combustion power was 11 W. The maximum efficiency of 24.8 % was obtained, when the combustion power was 9.5 W and the fuel feeding rate was 20 µl/min. The CO concentration of the as-reformed gas is as high as 3000-5000 ppm, which must be reduced by CO methanization.

## ACKNOWLEDGEMENT

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