DESIGN AND FABRICATION OF
A VACUUM-PACKAGED MICRO FUEL REFORMER

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ABSTRACT: A fully-microfabricated wafer-level vacuum package of a micro fuel reformer was designed, fabricated and tested. For thermal insulation, a high-temperature reactor is suspended by microfabricated tubes, in which fuel and reformed gas flow, and is vacuum-packaged by anodic bonding. Conductive heat loss through air in the package was investigated by making the packaging pressure as a parameter. The measured heat loss is 1.2 W at a reforming temperature of 240 °C for sufficiently low packaging pressure, showing a potential to realize a micro fuel reformer with a thermal efficiency of 75 %. Also, the temperature of the package outside is as low as 60 °C, which is enough to install the micro fuel reformer in portable electronics.

Key Words: vacuum packaging, thermal insulation, fuel reformer, fuel cell, microreactor

1. INTRODUCTION

Recently, increasing demand for the longer operation time of portable devices has made fuel cells attractive as portable energy sources. Hydrogen-fueled polymer electrolyte membrane fuel cells (PEMFC) have higher power density than direct methanol fuel cells (DMFC), but need a compact hydrogen source.

There are several methods to supply hydrogen to fuel cells, e.g. using compressed hydrogen, liquid hydrogen, fuel reforming and chemical hydride. For portable devices, the best method to supply hydrogen in portable devices seems using a micro fuel reformer fabricated by microelectro mechanical system (MEMS) technology. MEMS technology is useful to fabricate and integrate a reforming reactor, a carbon monoxide remover, a catalytic combustor etc. in a small volume [1–3]. Furthermore, excellent thermal isolation of reaction areas is possible using thin film microstructures fabricated by MEMS technology [4–7].

In the previous study, we have demonstrated a high power density of 5.9 W LHV/2.8 cc with a fully-microfabricated fuel reformer [8]. However, the previous device had no thermal insulation structure, and its thermal efficiency was as low as 36 %. In this paper, we propose a vacuum-packaged micro fuel reformer for high thermal efficiency and low package temperature.

2. CONCEPT

For portable applications, methanol is generally used as a fuel, because methanol reforming proceeds at a relatively low temperature around 250 ºC, and is easier than other hydrocarbons. To produce hydrogen from methanol, steam reforming is generally used. The total reaction of methanol steam reforming is given by

$$CH_3OH + H_2O + 49.45 \text{ kJ/mol} \rightarrow 3H_2 + CO_2. \quad (1)$$

The total reaction is endothermic. Thus, supplying heat to a reactor makes the reaction go forward to produce hydrogen.

Figure 1 shows a system diagram of a methanol reformer. The evaporator and the reformer are heated by the heater for the evaporation and reforming of the fuel, respectively. To reduce the heat loss from these high temperature parts to low temperature parts of the reformer, a vacuum packaging is used. The chip-level vacuum packaging of a micro fuel reformer was presented by Casio Computer in 2006 [9]. However, a
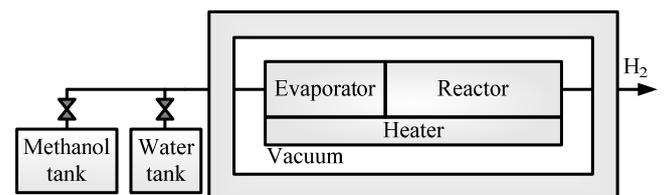

Figure 1: Diagram of a vacuum-packaged methanol reforming system.

wafer-level vacuum package was not reported to the best of our knowledge. In this study, we developed the wafer-level vacuum packaging technology for a fully-micromachined fuel reformer.

3. DESIGN

3.1 Structure and fabrication

Figure 2 shows the cross-sectional and exploded structure of our vacuum-packaged micro fuel reformer. Silicon and Pyrex glass layers are anodically bonded, constituting the micro fuel reformer. The second layer made of a 1 mm thick Pyrex glass substrate has a suspended reactor with a volume of 0.035 cc formed by sandblast from top and bottom side. The glass beams with a microchannel suspending the reactor scarcely contribute to heat conduction from the reaction area to the surrounding package due to a low thermal conductivity of glass.

The third layer is made from a SOI (silicon on insulator) substrate whose device layer is 60 μm thick. On the device layer, a Si_3N_4 insulation layer is deposited by APCVD (atmospheric pressure chemical vapor deposition), and then a heater/temperature sensor made of Pt/Ta is fabricated by lift-off process. The resistance of the heater/temperature sensor is 480 Ω at room temperature.

After anodic bonding with the fourth Pyrex glass layer, the suspended reactor and evaporator are fabricated by RIE (reactive ion etching). The thin silicon with a high thermal conductivity prevents hot spots in the microchannels and the reactor. The first and forth layers package the second and third layers in vacuum. The first layer has a fuel inlet and a product outlet fabricated by

deep RIE. Boron is diffused to the topside, and the diffused layer is used as an etch stop to make a diaphragm by EPW (ethylene diamine pyrocatechol water) etching. The diaphragm is used to monitor vacuum level inside the package. The fourth layer has two holes to contact the inner heater/temperature sensor, which are opened by sandblast.

After the fabrication of each layer, all layers are bonded by anodic bonding in atmosphere. Finally, the vacuum packaging is completed by sputtering metal into the electrical contact holes on the forth layer in vacuum.

3.2 Calculation

To determine the detailed dimensions of the device, heat loss from the reactor to the package is theoretically estimated. Heat losses by conduction, radiation, heat of product gas and convection are considered to estimate the thermal loss of the system. Figure 3 shows possible heat flows in the micro fuel reformer.

The meandered suspending tubes are approximated as straight beams in the calculation of the conductive heat loss of the suspending tubes. The radiative heat loss is estimated on the assumption that the reactor and the package are black-body sources. Product gas conveys heat from the reactor to the surrounding environment, resulting in heat loss. It is assumed that the product gas is composed of 75 % hydrogen and 25 % carbon dioxide, and that the flow rate is 30 sccm. Natural convective heat transfer from the top, bottom and side surfaces are also considered.

Figure 4 shows calculated various heat losses as functions of the temperature of the designed reactor. The total heat loss is 1.2 W, when the reactor is heated at 240 ºC. This result
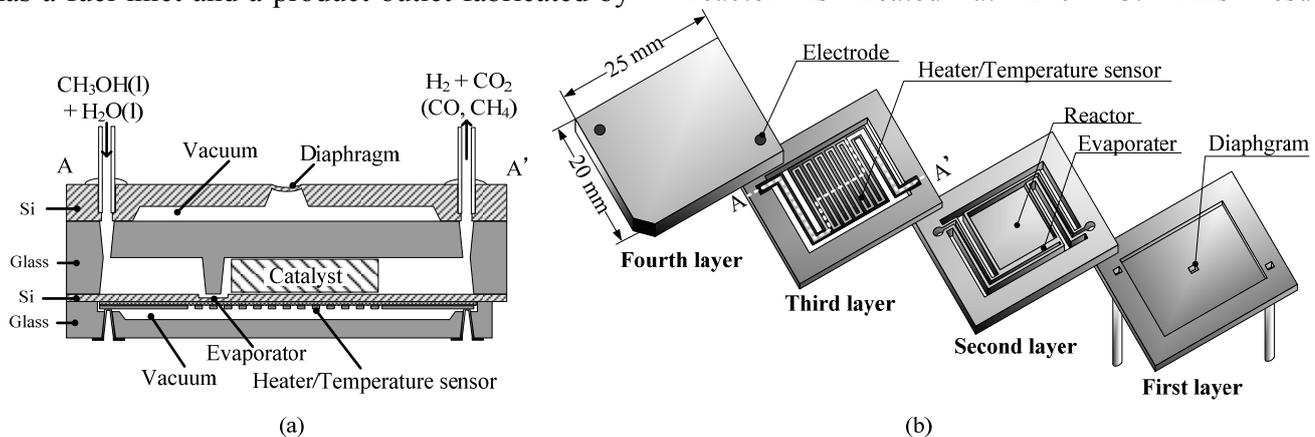

Figure 2: Structure of the vacuum-packaged reformer. (a) Cross section, (b) Exploded structure.

demonstrates that the heat loss is only 1.2 W during fuel reforming at 240 ºC. If this device achieves a power density identical to the previous work [8], a hydrogen production at 3.6 W LHV is expected, and thus the thermal efficiency becomes as high as 75 %.

At low temperature, the radiative heat loss and the conductive heat loss through the silicon beams occupy most of the total heat loss. At high temperature, the radiative heat loss grows up to the largest one. Further reduction of the radiative heat loss, which is the largest heat loss, is possible by the deposition of a low emissivity material on the hot or cold wall in the micro fuel reformer.

4. EXPERIMENTAL

4.1 Measurement setup

The fabricated device with the diaphragm mechanically penetrated was connected to vacuum pumps, and the inside of the package was pumped out. The reactor inside the vacuum package was heated by applying a constant current to the heater fabricated on the third silicon layer. The temperature of the reactor and the package outside was measured with the embedded temperature sensor and a thermocouple, respectively.

4.2 Effect of packaging pressure

Conductive heat loss through air remaining in the package at various pressures was experimentally measured. Figure 5 shows the conductive heat loss as a function of the chamber pressure at an applied current of 30 mA. As found in the figure, the reduction of the convective heat loss is large from 10^3 Pa to 1 Pa. The kinetic theory of gases predicts that the thermal conductivity of a gas is a function of the pressure, when the mean free path of gas molecules is limited by the size of enclosure. The energy flux of gas conduction is given by

$$Q_{conduction}^{gas} = \frac{1}{3}\bar{c}\lambda C_{V,m}[A]\frac{dT}{dx}, \qquad (2)$$

where $\bar{c}$ is an average gas molecule velocity, λ is a mean free path, $C_{V,m}$ is a volume constant molar heat capacity, [A] is the molar concentration of gas A, T is a gas temperature, and x is the length in a diffusion direction. When λ is limited by the size of enclosure, λ is considered as a constant
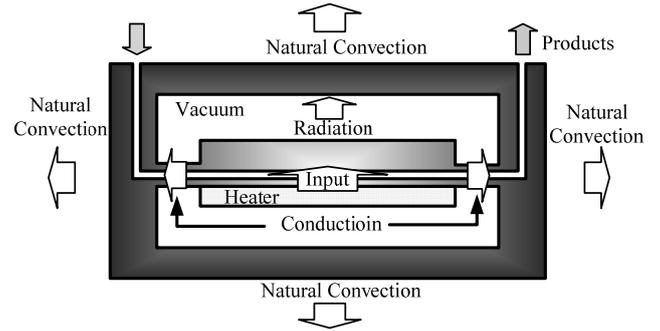

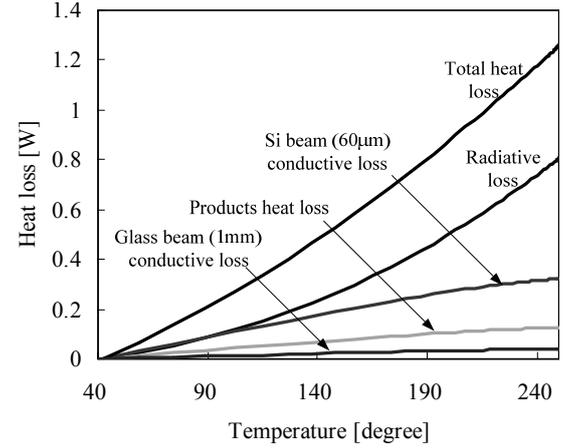Figure 3: Diagram of heat flows in the micro fuel reformer.

Figure 4: Calculated heat losses from the reactor to the package as functions of the temperature of the reactor.
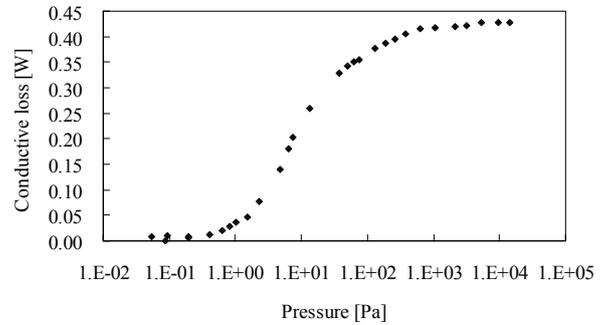

Figure 5: Conductive heat loss through air in the package as a function of the pressure. Heating current is constant at 30 mA.

value, whereas the molar concentration of the gas is proportional to the pressure. Therefore, the conductive loss is proportional to the pressure below 10^3 Pa. At higher pressure above 10^3 Pa, the pressure has little influence on the conductive loss, because decrease in the molar concentration of the gas and increase in the mean free path cancel each other. This experiment found that the gas conductive heat loss becomes below 0.1 W, if the packaging pressure is below 1 Pa. By packaging below this pressure, the conductive

heat loss is practically negligible in comparison with solid conductive and radiative losses.

4.3 Comparison of experiment and calculation

The reactor started to be heated using the inner heater at a constant power of 1.2 W, and the temperatures of the reactor and the package were measured. The packaging pressure was 7×10^{-3} Pa. Figure 6 shows the experimental result in comparison with a calculation result. The calculation took account of heat losses by conduction, radiation, convection and gas flow. The result shows that the calculation well agrees with the experiment. When a power of 1.2 W was applied to the heater, the reactor reached a reforming temperature of 240 ºC in 3 min, while the temperature of the package outside was as low as 60 ºC.

5. CONCLUSION

Vacuum packaging technology for thermal insulation of a micro fuel reformer was developed. A high-temperature reactor is suspended by microfabricated tubes, in which fuel and reformed gas flow, and is vacuum-packaged by anodic bonding. The fabrication and vacuum packaging of the micro fuel reformer are fully based on MEMS technology, and thus wafer-level.

Thermal insulation performance was evaluated by making packaging pressure as a parameter. Conductive heat loss through air in the package decreases from a packaging pressure of 10^{3} Pa to 1 Pa. At a packaging pressure of below 1 Pa, the total heat loss including heat losses by conduction,

radiation, convection and gas flow was as low as 1.2 W, when the reactor reached a reforming temperature of 240 ºC. This suggests a potential to realize a micro fuel reformer with a thermal efficiency of 75 %. At that time, the temperature of the package outside was as low as 60 ºC, which is low enough to install the micro fuel reformer in portable electronics.

AKNOWLEDGEMENT

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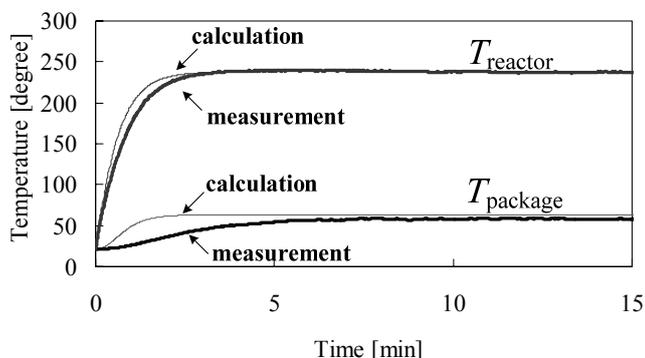
Figure 6: Measured and calculated temperatures of the reactor ($T_{reactor}$) and the package ($T_{package}$) at a heating power of 1.2 W. In the steady state, $T_{reactor}$ and $T_{package}$ are 240 ºC and 60 ºC, respectively.