MEMS based micro-fuel processor

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1. Introduction

Hydrogen is the best fuel for getting high performance of fuel cell. In order to implement it in micro-fuel cell for application in portable electronics (like mobile phone, notebook etc), there is a need of hydrogen storage. The catch is that it is difficult to find a way to store or produce enough pure hydrogen in the laptop. One could imagine putting a tank of very pure hydrogen in the laptop and refilling it at filling stations, rather like those proposed for cars. But in practice, the explosive nature of hydrogen rules that possibility out. One could not, for example, countenance taking a laptop with a hydrogen cylinder on to an aeroplane. In fact, of possible fuels, so far only formic acid and methanol have been approved by the dangerous-goods committee of the International Civil Aviation Organization for use in aeroplanes and storage in luggage. Therefore two good options are the installation of micro-fuel processor before micro-fuel cell which provides the facility to select the fuel (like methanol, ethanol) other than hydrogen without changing the feed (i.e. hydrogen) for fuel cell or direct use of methanol, ethanol and formic acid in fuel cell. In direct alcohol fuel cell system, the main studies center around methanol (direct methanol fuel cell, DMFC) though there is recent trend of studying with ethanol keeping in mind of lower toxicity and higher power density. DMFC has main drawback of high crossover rate and low rate of oxidation. Reformed hydrogen fuel cell (RHFC) is expected to become a more compact system compared to a DMFC system, because a RHFC has approximately one order of magnitude higher power density than a DMFC system. The advantages of using silicon wafer as micro-reformer is its matured MEMS technology and reduction of the reactor's size to very small.

There is growing consensus for RHFC with methanol feed that a combination of a micro fuel cell system and a rechargeable battery will be the most applicable, in which the fuel cell continuously recharges the battery and the battery in turn provides electricity to the portable device. It should also be noted that incorporation of polybenzimidazole (PBI) based high temperature tolerating proton exchange membrane facilitates the design of the micro fuel processor.

In the literature, the use of micro-packed bed reactor for the reforming of methanol to produce hydrogen has been focused recently. Pattekar and Kothare [1] presented a silicon reactor fabricated by deep reactive ion etching (DRIE). Most recently, Pattekar and Kothare [2] developed a radial flow micro-packed-bed reactor which posseses less pressure drop compared to conventional one due to variable (increasing) flow cross section along the reaction path.

The different workers were tried with the catalyst coated micro-channel reactor and plate type reactor expecting low pressure drop, less channeling of gas and better response in transient behavior. Details are given in Kundu et al. [3].

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2. Objectives

In the present work, MEMS based micro-reformer with micro vaporizer has been designed which can be integrated with PEM fuel cell. Two types of channels are made for micro-reformer and the performance has been compared. The durability test for each type of micro-reformer was performed.

3. Experimental Set-up

Figure 1 shows the experimental set-up. It mainly consists of liquid feed pump, compact reactor unit comprising vaporizer chamber followed by reactor chamber and temperature indicator. The reactor was heated through electrical heating system by on chip platinum lines deposited on the rear side of the silicon wafer patterned or by heating through the electrical hot plate. Micro-channeled plate made of silicon wafer has been used as a reactor considering its mature technology and considerable reduction in size. Standard photolithography steps followed by deep reactive ion etching were used for etching on a silicon substrate to form micro-channels, vaporizer and filter. The split type channels were made in the micro-vaporizer region in order to reduce the back pressure at the inlet port which helps to get more uniform flow of fluid. Before packing inside the channels, the catalyst beads were crashed by mortar and pestle and the particle size of 50-100 μm are separated by sieving prior to use.

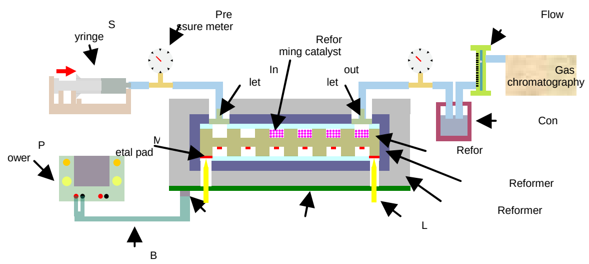

Figure 1. Experimental set-up

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Figure 2. Catalyst loaded MEMS reactor

The catalysts are packed thorough port “p” (Figure 2) by injecting the catalyst slurry made with DI water. The maximum amount of catalyst loaded is 140 mg. The feed rate of the liquid mixture was varied from 0.01 ml/min to 0.02 ml/min. The water and methanol ratio in the feed was 1.2:1 for getting low concentration of CO. The total volume of micro-reformer is 0.9 cc in which 30% of the total volume is utilized for reaction and vaporization zone.

The produced gas stream was directed through a cold trap to remove liquid components and then passed to an on-line micro-gas chromatograph or to bubble flow meter for measuring flow. The micro-gas chromatograph (Varian CP-4900) was equipped with a Molecular Sieve 5A and Porapak Q column and a TCD detector using Argon as the carrier gas.

4. Results

The catalyst loaded in the micro reformer was activated for 1.5 hrs at 220^0C with the water and methanol mixture flow rate of 0.02 ml/min. The performance of parallel and serpentine patterned micro reformer has been shown in Figure 3. The performance in serpentine channeled micro-reformer is always higher than that in parallel channeled micro-reformer. This may be due to shorter residence time of the reacting vapors in parallel channeled micro-reformer. The typical value of residence time is about ~500 ms for sufficient conversion of methanol to hydrogen [4].

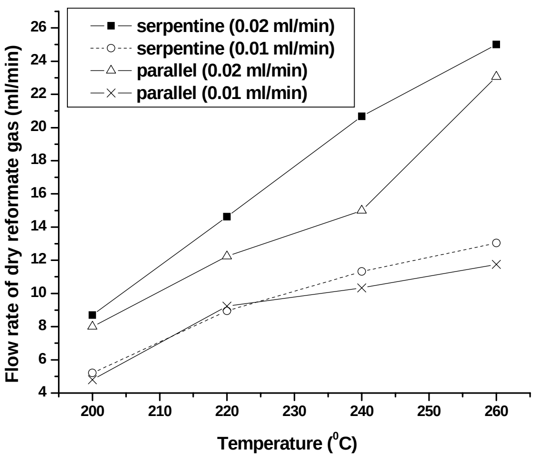

Figure 3. The comparison of the performance of serpentine and parallel channeled micro-reformer in terms of flow rate of dry reformate gas

The GC analysis of the produced gases from steam reforming of methanol shows that the average concentration of H_2 and CO_2 in the produced gas is 75% and 24% respectively, whereas the maximum concentration of CO is 1.5%. Therefore the produced gas from the micro-reformer can be used directly as a feed for micro-fuel cell operated at high temperature with PBI membrane. The flow rate of dry reformate gas is 25 ml/min at 20^0C and atmospheric pressure with serpentine channeled micro-reformer at the feed rate of 0.02 ml/min, whereas that value for parallel channeled micro-reformer is 23 ml/min at the same condition. The durability test of serpentine channeled micro-reformer has been presented in Figure 4. During 8 hrs continuous operation, about 15-20 % conversion decreased in the last 3 hrs, but it regained the original performance after the break.

It also shows that after 25 hrs, the catalyst activity remained same; it only needs some time to regain its original form after a long time continuous operation. The fast deactivation of the catalyst in parallel channeled micro-reformer (Figure 5) may be due to more exposure of temperature in non-uniform behavior and more possibility of maldistribution of the reactant. Parallel channel also creates a lot of problem like leakage of inlet tubing which may be due to higher back-pressure.

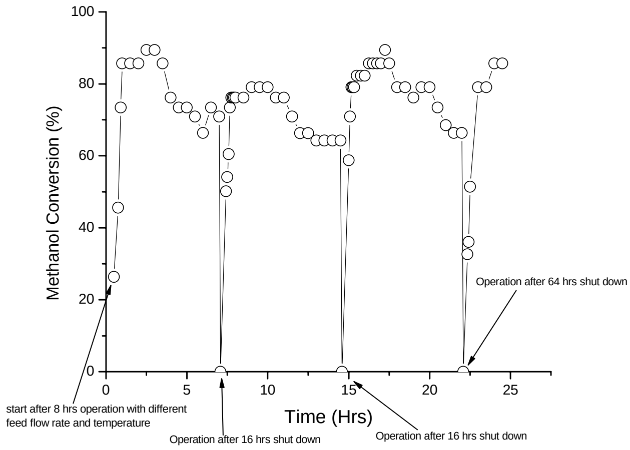

Figure 4. Durability test at 260C and 0.02 ml/min with serpentine channeled micro-reformer

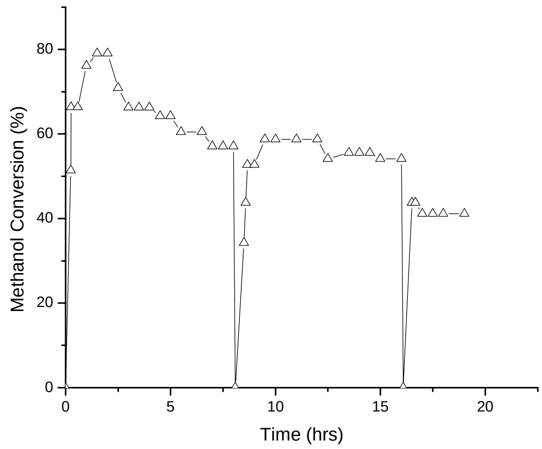

Figure 5. Durability test at 260C and 0.02 ml/min with parallel channeled micro-reformer

5. Conclusions

Silicon based micro-vaporizer plus micro-reformer has been fabricated for reformed-hydrogen fuel cell in the application of cell phone. Average hydrogen production rate is 0.0445mole/hr The performance with serpentine channeled micro-reformer is always superior compared to that in parallel channeled micro-reformer. The durability test also confirms that catalyst in serpentine channeled micro-reformer shows better durability than that in parallel channeled micro-reformer.

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