

A LOW-TEMPERATURE POM MICRO METHANOL REFORMER WITH HIGH FUEL CONVERSION RATE AND HYDROGEN PRODUCTION YIELD

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ABSTRACT

In this article, a low-temperature partial-oxidation-mode (POM) micro methanol reformer with the performance of **high durability and hydrogen productivity** is introduced. Our previous researches [1, 2] have demonstrated a POM type micro reformer with an operation temperature at 220°C and fuel conversion rate and hydrogen selection rate higher than 90%. In this study, a much improved micro methanol reformer, combining nano-catalyst dispensing technique with serpentine micro flow structures, can further reduce the operation temperature down to 180 °C but still carry out a comparable performance. Experimental results showed that the hydrogen yield can be obviously increased **2.6 times** with increased concentration of methanol from 1.3×10^{-5} to 3.4×10^{-5} (mole/min) with conversion rate near **98% at 180°C**. The ratio of hydrogen yield to feed rate also approaches as high as **0.051** in this study at **180°C**, superior to most of the reports in the literature [3].

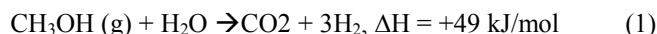
KEYWORDS: Hydrogen, methanol, reformer, micro channel,

INTRODUCTION

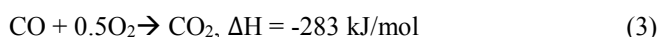
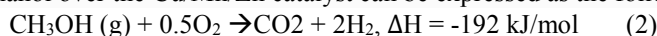
To solve the crisis of energy utility, fuel cells have emerged as alternative power sources for a myriad of applications in transportation, portable electronics, and residential power supplies. However, the major issues encountered by hydrogen fuel cell are the generation, transportation, and storage of fuel. Among many potential solutions, production of hydrogen directly from hydrocarbon fuels, such as methanol, has attracted plenty of attentions recently for its ease on storage/transportation and applications. For portable electronic applications, a small size methanol reformer is highly desired. However, the design and implementation of such a miniaturized device require the incorporation of MEMS and nanotechnologies in a highly integrated manner.

THEORY

Methanol has gained interest due to its availability and high hydrogen-to-carbon ratio. Generally the micro methanol reformer produces hydrogen via catalytic action of the Cu/Mn/Zn catalyst coated in the micro-channel of reformer. So far, there are several reactions that can efficiently transform methanol into hydrogen, and among all the reactions steam reforming reaction (SRM) and partial oxidative reforming reaction (POM) are mostly attracted attentions owing to their characteristics. SRM can theoretically produce a hydrogen-rich gas with a composition of 75% H₂ and 25% CO₂, but it's an exothermic reaction:



So, SRM will increase the needs of power via needing an extra heating resource to reach a suitable operating temperature. The POM reaction of methanol over the Cu/Mn/Zn catalyst can be expressed as the following reactions:



Eq. (2) shows POM is a highly exothermic reaction, which can which can be used to construct highly dynamic and fast reforming systems; the preferential oxidation reaction (PROX) (Eq. (3)) and the water gas shift reaction (Eq.(4)) are clean-up steps to reduce the CO concentration in the productive gas. In methanol reformer, its performance depends on many factors, such as temperature, pressure, catalyst type, water to methanol ratio, reactor geometry, flow pattern, etc. but the conditions that decide whether micro-PEMFC will be successfully commercialized is not only the performance of methanol reformer but its hydrogen yield in the long run.

In this work, we adjust the variation of the concentration of reactant gas in the mixed gas, including steam methanol, oxygen and argon which is as a carrier gas to solve the problem of the hydrogen yield of the micro reformer.

EXPERIMENTAL

This study incorporates serpentine type micro flow channel with large cavity well for containing high density nano-catalysts, as shown in Fig.1, to carry out high performance micro methanol operated in low temperature. The main test apparatus is composed of the reactant supply system, integrated reactor, a hotplate to control the operating temperature, data acquisition system with a gas chromatograph unit for product analysis, and K-type thermocouples for detecting the temperature. The experimental setup for the reformer testing is shown in Fig.2. For the reactant supply system, argon, as the carrier gas, oxygen and liquid methanol will be used as the reactant fuel, and the feed rate of them are set as 81.7 and 6.1 ml/min through the mass flow controller (the Brooks instrument, 5850E), and 0.02 ml/min via pump to obtain the steam flow rate of 12.2 ml/min after evaporating, respectively, therefore the composition of the reactant gas will follow the stoichiometric of the POM reaction. The reactant gas will be plenary premixed and preheated in the mixing chamber of 90

°C before being delivered into the micro-methanol reformer at designated feed rate, 2ml/min, to carry out the reforming reaction. The methanol conversion, hydrogen yield and selectivity can be obtained based on the measured compositions in the reformed gas. The explicit fabrication process of the reformers is similar to our previous reports [1, 2]. The silicon-based micro channel was made through the standard lithography process, then bound with a Pyrex glass plate opened with inlet/out holes, as shown in Fig. 3. Finger-shaped grooves were first fabricated on silicon (100) wafer by photolithography and deep silicon reactive ion etching (DRIE) process. Then, hydrophilic groove-

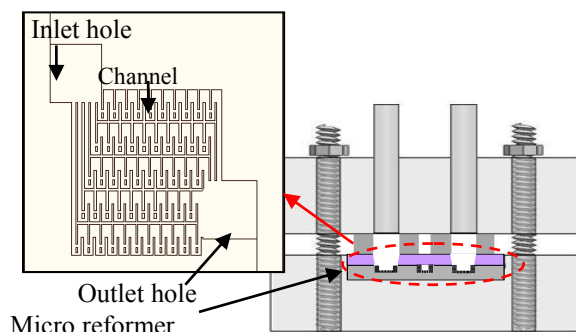


Figure. 1 The image of the design of the whole device

walls were obtained by oxygen plasma manner. Pyrex glasses with inlet/outlet holes were machined by laser dripping process and then bonded on the chips to obtain micro-channels by using anodic bonding process.

This study, the feed rate of reactant gas was set at 2ml/min, including argon as the carrier gas, oxygen and steam methanol as the fuel gas. For investigating the influence of concentration of fuel gas in reactant gas, fuel concentrations were varied from 1 to 3.2 folds.

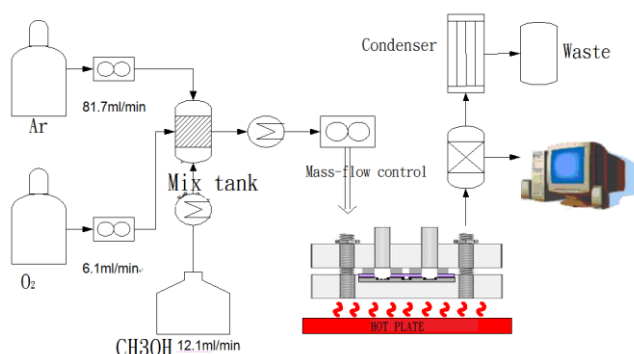


Figure. 2. The experimental setup for micro reformer testing.

RESULTS AND DISCUSSION

According to our previous work we know that it's necessary to provide enough content and thickness of catalyst in the micro reformer to have better performance while operating in the lower temperature, and to have potential to sustain higher concentration of the reactant gas. The Fig. 4 shows that the hydrogen yield and performance of the micro reformer are increased with the increasing of fuel concentrations, and the hydrogen yield increases 2.6 times from 1.3×10^{-5} to 3.4×10^{-5} (mole/min) with a high fuel conversion rate near 98% at 180°C. 3 fold concentration is the optimized value in this study and no more hydrogen can be generated beyond this point. The micro reformer carries out very high hydrogen-yield/feed-rate ratio at $Y=0.051$ and demonstrate superior performance than others in the literature in terms of operation temperature, conversion rate, and Y factor.

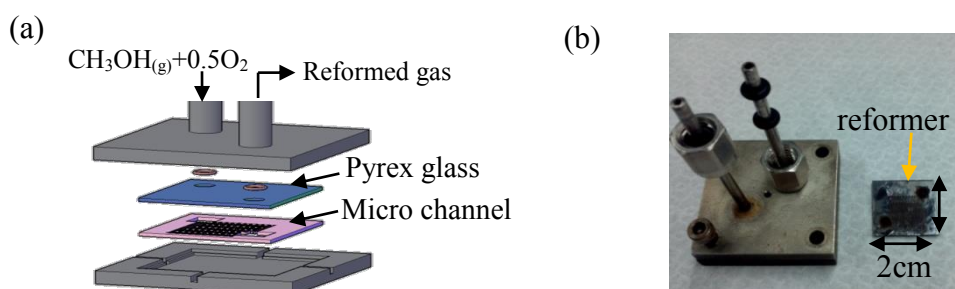


Figure 3. The micro reactor. (a) Schematic of the reactor design. (b) Image of the packaged reactor utilized in this study

CONCLUSION

In order to improve the hydrogen yield, one of methods is to increase the concentration of the reactant gas. According to the experimental result, the hydrogen yield has been modified 2.6 times from 1.3×10^{-5} to 3.4×10^{-5} (mole/min) by adjusting the concentration of the reactant gas, and it also keep a high performance of the 95% of the conversion rate, and the 96% of hydrogen selectivity even the testing temperature was in the lower condition, 170 °C.

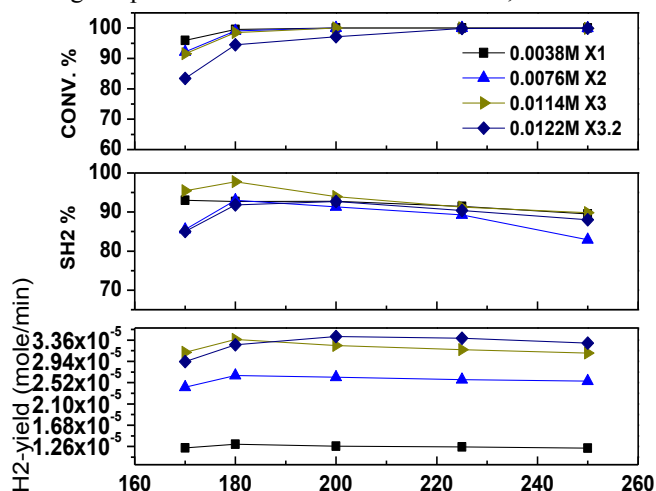


Figure 4. The performance in fuel conversion rate, hydrogen selection rate, and hydrogen production yield of the micro reformer at different testing temperatures.

ACKNOWLEDGEMENTS

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