

PEMFC 구동을 위한 LTCC Micro-Channel Reformer 의 개발 및 연구

신이나, 형우찬, 함승주*, 정찬화¹

연세대학교, ¹ 성균관대학교

(haam@yonsei.ac.kr*)

The development of the integrated Micro-Channel Reformer using (LTCC)

Yeena Shin, Woochan Hyung, Seungjoo Haam* and Chanhwa Chung¹

Department of Chemical Engineering, Yonsei University

¹ Department of Chemical Engineering, Sungkunkwan University

(haam@yonsei.ac.kr*)

Introduction

The miniature fuel cells recently emerged as a promising power source for applications such as cellular phones, small digital devices, and autonomous sensors to embedded monitors or to micro-electro mechanical system (MEMS) devices. Direct methanol fuel cell (DMFC) is easy to make smaller than polymer electrolyte membrane fuel cell (PEMFC) which includes hydrogen tank. However, it also has a drawback of relatively lower efficiency compared to PEMFC. We designed micro-fuel reformer system which generates H₂ rich gas from methanol solution. A micro-fuel processor system was manufactured with low temperature co-fired ceramics (LTCC) because LTCC is superior to other materials principally due to its high thermal and chemical stability, simpler fabrication processes, and lower materials cost. In order to prevent poisoning of the anode catalyst, the carbon monoxide concentration of the hydrogen-rich gas must be reduced to below 100 ppm, which can be achieved by addition of selective partial carbon monoxide oxidation (PROX).

Theory

There are two processes available for extracting Hydrogen from methanol: a) steam reforming (SR) and b) partial oxidation (PROX). Steam reforming of methanol has received much attention due to the ability to yield a product gas containing up to 75 % hydrogen while maintaining a high selectivity towards carbon dioxide. The main drawbacks of the steam reforming process are slow and endothermic. Pt, Rh and Ru catalysts were found to be the most appropriate for the PROX reaction: carbon monoxide was converted 100 % to carbon dioxide but a noticeable amount of hydrogen was also simultaneously consumed in the presence of excess oxygen. An “optimum” reaction temperature was 100 °C for Rh and Ru, and 170 °C for Pt.

Experiments

LTCC tape was used in the fabrication of ceramic monolithic devices. LTCC tape consists of ceramic particles in a polymer matrix. Prior to the sintering, LTCC tape is very flexible and easily patterned. In green state, various micro channels, reaction cavities and fluid interconnects could be formed by digitally controlled mechanical punching (PCB milling machine, Dong-il Cad system, 1000 H). These patterned layers were stacked and laminated at low temperature (110~120 °C) and pressure (2,000~2,200 psi), still maintaining the structural integrity of the embedded features, to form a monolithic 3D structure. The fabrication process of ceramic monolithic structures from LTCC tape is illustrated in Fig.1

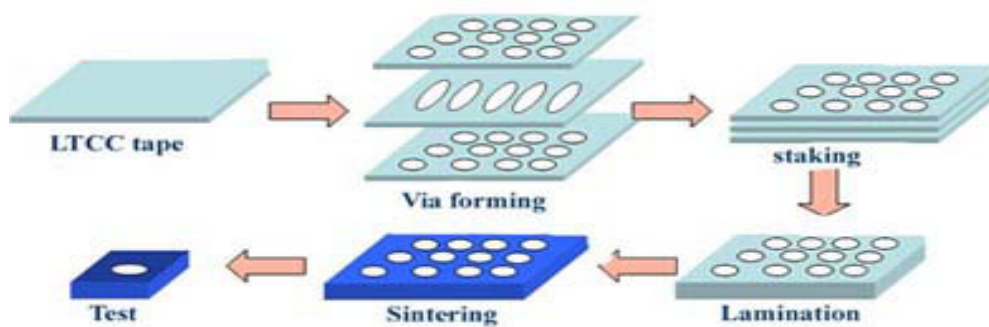


Figure 1. LTCC Process diagram.

For Steam reforming, a commercial Cu/ZnO/Al₂O₃ catalyst from Katalco (33-5) was used. For PROX reforming, Pt catalyst was used in PROX reforming. Pt was prepared using wet impregnation and electroless Pt Plating. According to 4 ml/h and 10 ml/h input flow rates, outlet flow rate was measured by flow meter after condensing un-reacted steam and methanol. Then the total volume of outflow was analyzed by gas Chromatography (Young Lin M600D)



Fig.2 Prox Catalyst by Pt wet impregnation

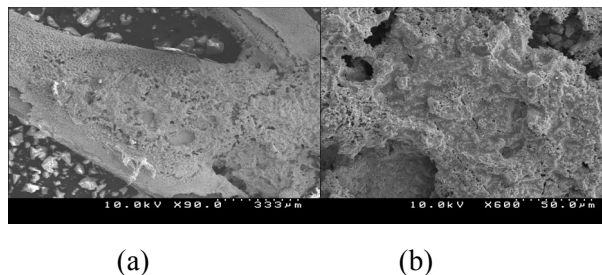


Fig.3 Electroless Pt Plating by scanning electron microscopy (SEM)

(a) Scanning image 333μm, (b) Scanning image 50.0μm

Results & Discussions

The developed micro reformer consisted of steam reforming part and partial oxidation part. Rod-type electric heaters were suitable to control the temperature, respectively. Conversion ratio increased with increase in temperature and decrease in input flow rate, because vaporizing methanol and water require heat to react. Fig.4 illustrates the conversion ratios using different ratios of methanol and water ratio (M:S) as a feed at the input flow rates of 4 ml/h and 10 ml/h. When M:S = 2:3, the conversion ratio is higher than M:S = 1:1 and M:S = 3:2 at all the input flow rates. Fig.5 shows that the increase in temperature increased the mole fraction of carbon monoxide increases by temperature. Sudden increase in the mole fraction of carbon monoxide was observed at the temperature over 300 °C and higher flow rate produced slightly lower mole fraction. In Fig. 6, PROX reforming conversion ratio increases by elevating temperature and decreasing input flow rate. At 4ml/h of the input flow rate, conversion ratio illustrates a slow increase.

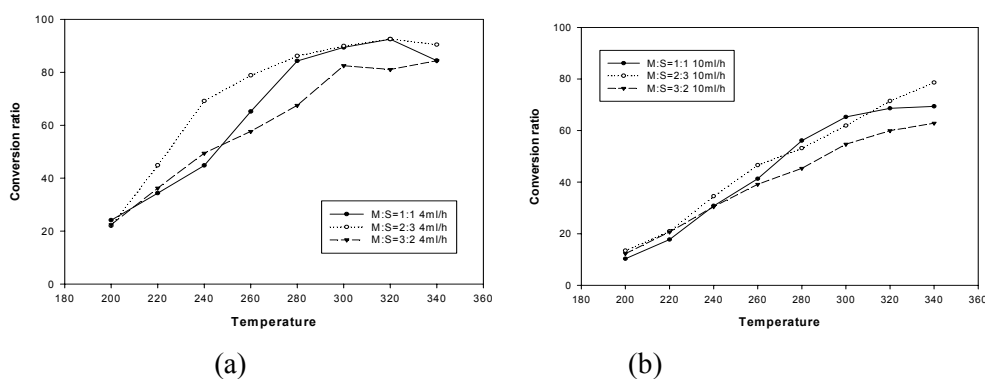


Fig.4 Conversion ratio for various temperature and M:S ratio input flow rates

(a) Input flow rate = 4 ml/h, (b) Input flow rate = 10 ml/h

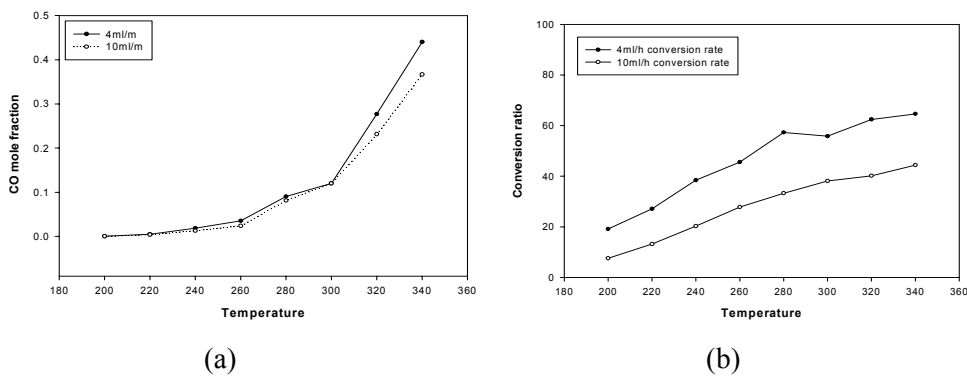


Fig.5 CO mole fraction after PROX reforming (a)

Fig.6 Conversion ratio for various temperature and input flow rates (b)

Acknowledgements

This work was supported by Korea science and engineering foundation (2004-0535-000), Republic of Korea.

Reference

- [1] Andorf, et al., US Patent 5 955 395 (1999).
- [2] Ilaria Rosso , Camilla Galletti , Guido Saracco , Edoardo Garrone and Vito Specchia Applied Catalysis B: Environmental 48 (2004) 195–203
- [3] Guangwen Chen, Quan Yuan, Hengqiang Li and Shulian Li, Chemical Engineering Journal 101 (2004) 101–106
- [4] Fernando Mario, Claude Descorme and Daniel Duprez , Applied Catalysis B: Environmental 54 (2004) 59–66
- [5] Attila Wootsch, Claude Descorme and Daniel Duprez , J. Catalysis 225 (2004) 259–266