

The development of micro-fuel processor using low temperature co-fired ceramic (LTCC)

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Abstract

A micro-fuel processor system integrating steam reformer and partial oxidation reactor was manufactured using low temperature co-fired ceramic (LTCC). A CuO/ZnO/Al₂O₃ catalyst and Pt-based catalyst prepared by wet impregnation were used for steam reforming and partial oxidation, respectively. The performance of the LTCC micro-fuel processor was measured at various operating conditions such as the effect of the ratio of the feed flow rate, the ratio of H₂O/CH₃OH and the operating temperature on the LTCC reformer and the CO clean-up system. The hydrogen concentration and the methanol conversion were high and stable as a durable micro-fuel processor from the feasibility test which was operated continuously for 12 h. The product gas was composed of 75% hydrogen, 25% carbon dioxide and carbon monoxide (< 50 ppm) at 260 °C, respectively.

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1. Introduction

Development of miniaturized fuel cell-powered systems has been extensively studied because they are 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 [1–3]. One of the most prospecting types of fuel cell is the proton exchange membrane fuel cells (PEMFCs) fueled with hydrogen. Methanol is an excellent hydrocarbon fuel

to obtain hydrogen by steam reforming at much lower temperatures (250–300 °C) with low cost, simple to storage and good miscibility with water. Furthermore, methanol is profitable for environmental as it is rapidly biodegradable in air, soil and water [4].

A diverse of the investigations have developed fuel processors made of stainless-steel [5,6]. Stainless-steel is an attractive material because of its easiness of treatment and high strength. In spite of these advantages, stainless-steel has some problems for micro-scale devices: corrosion resistance decrease in strong acid, heavy and expensive. For installing hydrogen reforming unit to a portable fuel cell, compact design of reformer is mainly the strategic point. To work out these problems, some alternative materials have been suggested,

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i.e., silicon and ceramic monolith based fabricated steam reformers. Silicon micro fabrication technology is promising technology for macro-scale facilities into MEMS devices. For developing a silicon based micro-fuel processor, Si (110) and p-type (100) wafer has been generally used to fabricate micro-channel formed by anisotropic wet etching [7,8]. However, silicon fabrication technology requires accurate and expensive processing step. In other words, wet etching process can easily be influenced by temperature and impurities. In case of ceramic monolith type fuel processors, in order to accommodate a fully integrated design approach, for both thermal transfer and volume efficiency in the design, Low Temperature Co-fired Ceramic (LTCC) has been used to build the demonstration prototype [9,10]. Because LTCC allows a layered construction with embedded micro-channels within the multi-layers LTCC can be used in the fabrication of stacking type ceramic monolithic devices such as capacitors inductors and resistors. LTCC has attractive advantages as a reformer application since LTCC is economic and reliable device for high volumes and high packaging density [11].

In this paper, we developed a micro-scale reformer which generates H₂ rich gas from methanol solution including CO clean-up system (PROX) using LTCC. In order to realize a micro-channel fuel processor, micro-channels were patterned on LTCC tapes and separated from substrates using PCB milling machine. Vaporizer and combustor which were fabricated by stacking LTCC substrates were integrated. The size of the reformer was $W: 40 \times D: 75 \times H: 10$ mm. Steam reforming catalyst was filled up inside combustor as a packed bed type. After vaporizing, gas reacts on copper–zinc oxide catalysts through the endothermic reaction. The main products are hydrogen, carbon monoxide, and carbon dioxide [12]. In the steam reforming process, the amount of carbon monoxide in the dry product gas at optimal condition is about 10³–10⁴ ppm. For the operation of PEMFCs, in order to prevent poisoning of the anode catalyst, the carbon monoxide concentration of the hydrogen-rich gas (ca. 40–75% H₂) must be reduced below 100 ppm. The selective carbon monoxide oxidation is occurred in CO clean-up system using supported Pt catalyst.

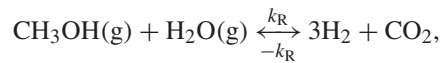
2. Experimental methods & materials

2.1. Methanol reforming reactions

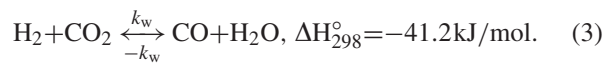
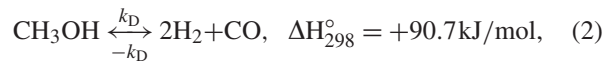
For converting methanol to hydrogen, there are two processes available: (i) steam reforming (SR); and (ii)

partial oxidation (PROX). It was assumed that methanol was initially decomposed to produce carbon monoxide and hydrogen, and then the carbon monoxide reacted with water to produce carbon dioxide and hydrogen in early studies. Recently it was proposed that methanol reforming reaction occurred directly with water to produce carbon dioxide and hydrogen [13].

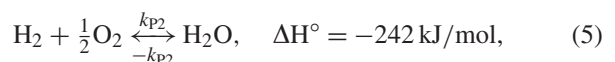
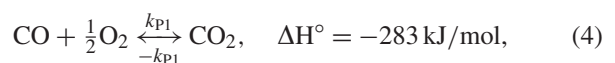
Methanol is converted to hydrogen for CuO/ZnO/Al₂O₃ catalyst during the reforming process that can be written expressed by following equations:



$$\Delta H_{298}^\circ = +49.5 \text{ kJ/mol}, \quad (1)$$



Steam reforming of methanol (Eq. (1)) is slow, endothermic and developed completely over commercial catalyst. Methanol decomposition (Eq. (2)) is highly endothermic process and the most simple conversion method as only methanol is used in the fuel. The decomposition of methanol yields product gas containing up to 67% hydrogen and 33% carbon monoxide. PEMFCs is highly sensitive to carbon monoxide, which poisons the platinum catalyst on the anode. Carbon monoxide has detrimental effects on the performance of the fuel cell. Water gas shift reaction (WGS) (Eq. (3)) reduces the carbon monoxide content of the hydrogen rich gas stream about 0.5 ~ 1% (5000–10,000 ppm) while increasing the hydrogen content in the product stream and low temperature favors the forward reaction at equilibrium conditions [14]. For the purpose of fuel cell application, concentration of carbon monoxide should be maintained below 50 ppm by adding a CO clean-up step after reformation. Noble metals (Pt, Rh and Ru) were found to be the most appropriate catalysts for the PROX reaction: carbon monoxide was totally converted to carbon dioxide in a wide temperature range (100–180 °C). Optimal reaction temperature was 100 °C for rhodium (Rh), ruthenium (Ru) and 170 °C for platinum (Pt) but noticeable amount of hydrogen was also simultaneously consumed in the presence of excess oxygen [15,16].



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