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# Study on an integrated natural gas fuel processor for 2-kW solid oxide fuel cell

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ABSTRACT

A natural gas fuel processor integrated a compact reformer with two heat exchangers was developed and tested as a hydrogen generator for 2 kW distributed solid oxide fuel cell (SOFC) applications. The compact reformer is comprised of a reforming chamber and two non-catalytic combustion chambers, in which the endothermic and exothermic reactions are coupled into a multilayered cylindrical reactor vessel. The integrated fuel processor can be started up quickly by the combustion of methane and run steadily within 30 min under the preferred thermodynamic operating conditions. The higher temperature zone is located at the latter half part of the reforming chamber, where the temperature gradient is relatively small and favorable for hydrogen production. The results show that a high methane conversion and $H_2$ molar fraction (dry basis) can be achieved, irrespective of the hydrogen production capacity. The energy efficiency of the integrated fuel processor can reach 74.11% when producing 2 $Nm^3/h$ of $H_2$. A SOFC system was fueled successfully by the generated hydrogen-rich reformate in the integrated fuel processor.


## Introduction

Nowadays more and more attention are being paid to small and mid-scale distributed power generating systems based on fuel cell, due to their high efficiencies. These systems are often fueled by fuel processors, characterized by low $NO_X$ emissions and fuel diversities. In particular, the distributed power generating systems combined with cogeneration systems are of great interest for residential applications [1,2]. This application offers mainly two benefits [3]: (a) on site and real-time electricity generation with a high efficiency; (b) utilization of the useful thermal energy produced during operation of the whole systems. Aimed at these kinds of distributed power applications, both solid oxide fuel cell (SOFC) and proton exchange membrane fuel cell (PEMFC) can be used as energy conversion devices [4–7]. The SOFC system is the preferred option, because it can convert a wide variety of fuels with high efficiency (40–60% unassisted) [8], and there is almost no limitation on CO concentration. However, a rather low level of CO is required for PEMFC systems, which means additional CO eliminating unit is essential after the reformer [9–11]. Theoretically, the SOFC systems can be fueled directly by

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hydrocarbon fuels. However, the deactivations of catalyst and anode caused by the carbon deposition are still a major bottleneck. Therefore, fuel pretreatment processors are essentially required for SOFC systems, in which the hydrocarbon fuels can be converted to hydrogen-rich mixture.

The fuel processors have to be carefully designed to generate a relatively stable hydrogen rich gas for a given FC system [12,13] from the wide range of secondary fuels such as natural gas, propane, gasoline, diesel, methanol/ethanol and bio-fuels. Among these hydrocarbon fuels for hydrogen generation, natural gas (mainly composed of methane) is one of the preferred fuels since it is clean, abundant, and well-distributed [14]. At present, several methane reforming schemes are available. While steam reforming is the most widely employed process owing to its high $H_2$/CO ratio in the hydrogen-rich reformates [15]. Besides, the methane steam reforming (MSR) is an endothermic process. It is crucial that how the external heat supply can satisfy the requirements of the methane steam reforming. The optimum scheme is to achieve the autothermal operation by coupling of the endothermic and exothermic processes. Several operation units can be integrated into a compact reactor, in which the autothermal operation and high efficiency can be achieved [16,17]. In view of heat coupling methods, there are mainly two types of autothermal compact reactors. One type is that the endothermic reactions and exothermic reactions react simultaneously at the same catalyst zone in the same chamber. The other type is that the endothermic reactions and exothermic reactions react simultaneously in two separate chambers. By integrating the two chambers into one reactor, the heat required in the endothermic reactions can be self-sufficient without external heat source. Among these two types of reactors, the latter one seems more suitable for generating continuously hydrogen-rich gases through MSR process.

Many pioneering studies were done on the process coupling of the exothermic and endothermic reactions by indirect heat transfer [12,18]. Additionally, various autothermal reactors, such as plate reactor [19–24], tube reactor [25–27], multilayered cylindrical reactor [3,14,28–32] and microchannel reactor [33–36], were coupled and studied experimentally and numerically. The vital improvements on the integrated fuel processor have been summarized [12]. In addition, the process optimization has also been reported based on numerical simulation/calculation, mass and energy management, parametric optimization. A process coupled MSR with the combustion of methane has been investigated, in which the countercurrent operation was adopted in a fixed bed monolith reactor [37,38]. Coupling MSR with the combustion of methane, a multilayered cylindrical metal monolithic reactor was also integrated and studied numerically by Mei H [30], in which the catalysts were deposited on the channel walls, thus promoting the heat transfer on the metal support. A two-dimensional model was developed by Fukuhara and Igarashi [39] to analyze the operation of the coupling methanol decomposition and methane combustion. They compared the performance of a wall-type reactor with a fixed-bed one, in which the exothermic and endothermic reactions proceeded simultaneously. The steady and dynamic behaviors of the reactor were analyzed using a one-dimensional pseudo-homogeneous plug flow model [40]. Using a one-dimensional heterogeneous model, M. Bayat [41] compared and predicted the performance of two different configurations of a thermally coupled reactor with two-layered cylindrical structure. Recently, the issues and challenges of the systemic integration of fuel processors and FC systems have been summarized [15]. The current state of engineering knowledge in fuel processor-FC integration and the future prospects for achieving more efficient fuel processor-FC systems has been reported.

Although many experimental and numerical works have been done on the compact reactors, some technical bottlenecks are still yet to be solved. For example, the reliability of system needs to be improved during the frequent startup and shutdown operations, and the relationship between the reactor configuration, the temperature profiles of the catalyst bed and the system efficiency also needs to be investigated in detail. In the present work, a MSR fuel processor with a capacity of 2 $Nm^3/h$ $H_2$ was developed for a SOFC system, in which a multilayered cylindrical structure reformer and two heat exchangers were integrated. The performance of the integrated fuel processor was investigated by monitoring the spatial temperature distributions and the reformate compositions in the processor. Meanwhile, the validity and practicability of the fuel processor was approved by the joint-test experiments, in which the processor is coupled with a SOFC system.

## Experimental

### *Operation procedure of the integrated fuel processor*

Fig. 1 shows the schematic of the fuel processing procedure of the integrated fuel processor. Firstly, air and $CH_4$ were introduced into the combustion chamber of the compact reformer at room temperature, and lighted directly by an electronic igniter. The temperature of the combustion gas can reach around 1100 °C quickly, which makes the temperature of the compact reformer ramp simultaneously. Meanwhile, $N_2$ was put into the reforming chamber of the compact reformer for protecting catalyst and increasing the heat transfer rate in the catalyst bed. Passing through the combustion chamber-2, the hot combustion tail gas ran into the heat exchangers to preheat the reforming reactants and then was released into the atmosphere. When the temperature in the reforming chamber reached the desired value, $H_2O$ was vaporized and superheated, and $CH_4$ was also preheated by the combustion tail gases in the heat exchanger-1 and the heat exchanger-2, respectively, and then injected into the reforming chamber. Feeding $N_2$ was stopped when the reforming reactions started. When the reforming system ran steadily, the hydrogen-rich reformates can be fed to a SOFC system directly as fuel to produce electricity.

The gas flow rates were controlled by mass flow controllers (MFC, Brooks, 5850E series) and the water was fed by a pump (Micro Pump, INC, America). The products were analyzed by an online gas chromatograph (Agilent 7890 B, America) equipped with a TCD detector, using argon as the carrier gas. The temperature signals were collected at the interval of 1 s using temperature measurement software ($H_2$-Tech Contr.,