



Microchannel methane steam reformers with improved heat transfer efficiency and their long-term stability



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HIGHLIGHTS

- The improved heat transfer efficiency was directly reflected in the methane reforming performances.
- Methane conversion has increased by 71.9–86.6 %; at the same supply rate of fuel.
- The long-term stability of microchannel reformers was verified by methane reforming tests for 500 h.

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ABSTRACT

To improve the performance of methane steam reforming as an endothermic reaction, the heat transfer efficiency from a heat source to a catalyst is a much more important factor than the reaction activity of catalyst itself on an industrial scale. In this context, microchannel reactors with combustion reaction (comb) blocks and reforming reaction (reform) blocks assembled by turns attract great attention due to their enhanced heat transfer rate. However, the even heat transfer to a catalyst bed is still a challenging topic. In this study, we improved the heat transfer efficiency of microchannel methane steam reformers by using porous-membrane-type catalysts and shifting a combustion point onto the top of the catalysts. As a result, methane conversion (conv) has increased by 14.7% at the same supply rate of fuel. Surprisingly, $1 \text{ Nm}^3 \text{ h}^{-1}$ of hydrogen has been produced using just 0.58 L (H : 2.9 cm, W : 20 cm, L : 10 cm) of reactors. In addition, the long-term stability of our microchannel reformers was verified by methane reforming tests for 500 h. Therefore, our microchannel reactors are expected to be more suitable for distributed power generation based on fuel cells.

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1. Introduction

As the world's energy demand is continuously increasing, it is necessary to use energy sources more economically and efficiently. In this context, hydrogen and syngas, which can be produced from natural gas, have attracted increasing attention as an energy source for fuel cells and as a building block for production of desired hydrocarbons via gas-to-liquid (GTL) processes in the future [1]. However, about 5 trillion ft^3 (tcf) of associated gas was annually flared, and 12.5 tcf of that was reinjected. In addition, about 6000 tcf of stranded gas is unexploited due to low accessibility. These energy sources are currently wasted without the access to

the energy markets, because the existing large-scale GTL processes are not suitable for the small-scale gas fields such as flaring associated gas, stranded gas and shale gas.

Therefore, small-scale micro-channel reactors (MCR) for methane steam reforming are a challenging topic in distributed energy and GTL industry [2]. Recently, numerous researchers have shown good performance of the steam reforming reaction using MCR. Yu et al. [3] studied the effects of temperature, space velocity, molar ratio of methanol to water, reaction time on the reaction performance of MCR with Cu50/Zn50 [Ce5] catalysts, and the MCR produced hydrogen equivalent to a power output of 11 W. Peela et al. [4] investigated MCR with Rh/CeO₂/Al₂O₃ catalyst for ethanol steam reforming. They demonstrated that the H₂ yield of MCR was $65 \text{ L g}^{-1} \text{ h}^{-1}$, whereas that of the packed-bed reactors was $60 \text{ L g}^{-1} \text{ h}^{-1}$. In addition, MCR with nickel-based catalysts for the methane steam reforming had been studied widely [6–10].

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However, those previous research utilized powder-type catalysts, which could be unfavorable to the enhancement of heat transfer efficiency from heat sources to catalyst bed.

For the performance of methane steam reforming as an endothermic reaction, the heat transfer efficiency from a heat source to a catalyst is much more important than the reaction activity of catalyst itself on an industrial scale. For this reason, microchannel reactors with combustion reaction blocks and reforming reaction blocks assembled by turns attract great attention. Thus, we previously reported that porous-membrane-type catalysts were more favorable to the accomplishment of higher heat transfer rate toward the catalysts, [5,11,12] and designed the MCR with combustion reaction blocks and reforming reaction blocks assembled by turns to be suitable for the shape of the membrane-type catalysts [12]. However, there still remain several challenges to improve its heat transfer efficiency from combustion reaction blocks to reforming reaction blocks, and to confirm its long-term stability. In this study, we report a new concept of MCR designed to position the hydrogen flame right on the porous-membrane-type catalyst, and investigate the effect of flame position on the heat transfer efficiency and reforming performance. In addition, we conducted the long-term test of the MCR for 500 h.

2. Experimental methods

2.1. Fabrication of steam reforming catalysts

Nickel powder purchased from Vale Inco Pacific Ltd was used for the preparation of porous-membrane-type catalysts. The average particle size of nickel powder was 3 μm and its purity was 99.7%. To improve the catalytic activity, Pd and Al_2O_3 dipping was carried out by means of wet impregnation. In a typical synthesis, dried nickel powder was impregnated with aluminum nitrate ($\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$, Sigma–Aldrich) and palladium solution of ($\text{Pd}(\text{NO}_3)_2$, Pd 10 wt%, PMRESEARCH) mixture to adjust the final composition of Pd and Al to 0.5 wt% and 0.3 wt%, respectively. The impregnated sample was pretreated with 90% H_2 /10% Ar at 450 $^\circ\text{C}$ for 30 h in order to remove organic compounds. After the pretreatment, the catalyst powder was compressed with metal mold under high pressure (85 mm, 900 MPa). The compressed Ni metal plates were thermally treated at 1100 $^\circ\text{C}$ in H_2 for 2 h in order to improve of its mechanical strength. After the thermal treatment, membrane-type reforming catalysts were successfully prepared. The preparation process of the catalysts is shown in Fig. 1a. The inset of Fig. 1b shows a photograph of the as-prepared catalyst, of which the diameter and thickness are 85.0 and 1.18 mm,

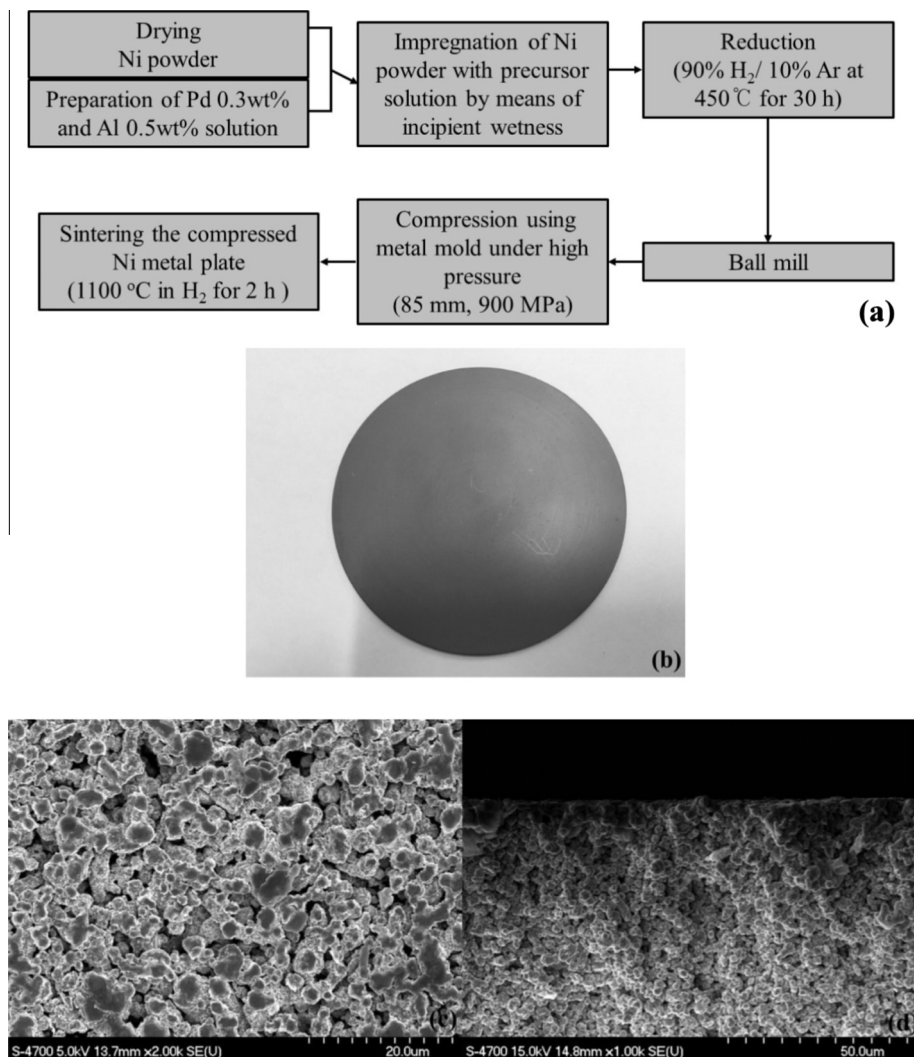


Fig. 1. (a) Preparation process of membrane-type catalysts; (b) photograph of catalyst; SEM images of Pd[0.3]–Al[0.5]/Ni catalyst of catalyst surface (c) and cross-section (d).

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