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Hydrogen production from cylindrical methanol steam reforming microreactor with porous Cu-Al fiber sintered felt

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ABSTRACT

In this study, the porous Cu-Al fiber sintered felt (PCAFSF) was fabricated by low temperature solid-phase sintering method. The laminated PCAFSF as the catalyst support was used for cylindrical methanol steam reforming microreactor for hydrogen production. The two-layer impregnation method was employed to coat the Cu/Zn/Al/Zr catalyst on the PCAFSF. The material composition, specific surface area and catalyst loading of PCAFSF were also measured. The effect of the fiber material, surface morphology and porosity on the reaction performance of methanol steam reforming microreactor for hydrogen production was further investigated. Our results show that the PCAFSF demonstrated much higher methanol conversion and H_2 flow rate compared to the porous Cu fiber sintered felt (PCFSF) and porous Al fiber sintered felt (PAFSF) having the same porosity. Furthermore, the rough PCAFSF showed much higher methanol conversion and H_2 flow rate compared to the smooth PCAFSF. In case of the PCAFSF, the methanol conversion and H_2 flow rate were increased with the decrease of Cu fiber weight and the increase of Al fiber weight. The best reaction performance of microreactor for hydrogen production was obtained using the three layer PCAFSFs with 80% porosity and 1.12 g Cu fiber/1.02 g Al fiber.

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Introduction

Proton exchange membrane fuel cells (PEMFCs) have received significant attention from scholars worldwide as devices for transforming chemical energy into electrical energy, primarily owing to their high energy intensity and because they produce no pollution and work at low noise levels [1–3]. PEMFCs have been widely used in electrical devices for cars, small

unmanned aerial vehicles, power station in remote area, and so on. Since hydrogen is a key fuel in the operation of PEMFCs, it is important to develop the reaction device to produce the hydrogen sources. On-site hydrogen production using mobile microreactor is considered as an effective means to provide hydrogen sources for PEMFCs, because they are highly safe, small, and convenient to carry [4–6]. Thus, the microchannel reaction systems have been widely used for on-site hydrogen production. Many previous studies have been focused to

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investigate the microchannel structure, catalyst component and its adhesion process, reaction conditions, as well as the integration of the reaction system.

The traditional microchannel structure is composed of a metal plate that is fabricated via mechanical processing and etching [7–9]. Its use as a catalyst support has been researched extensively. For example, Pan et al. [10] studied the performance of microchannel reaction systems in methanol conversion, H₂ flow rate, and gas selectivity by employing microchannels with different cross section shapes, aspect ratios, and spacing dimensions. The results show that parallel and uniformly distributed microchannels lead to higher methanol conversion and H₂ flow rate. Mei et al. [11] fabricated a microchannel reactor with micro-pin-fin arrays, and then studied the effect of structural parameters on the energy transfer efficiency for hydrogen production. It was found that the micro-pin-fin arrays with uniform distributions led to better heat and mass transfer between substances. Kuhn et al. [12,13] used a designed microchannel and corning microchannel as examples to study the single-phase liquid-liquid and two-phase gas-liquid dynamic flow and mass transfer processes with a particle image velocimetry system. It was found that the single-phase flow region distributions were uniform and prevented the large stagnant flow region in the microchannel. Recently, Yaseen et al. [14] used a 3D-2D symmetric model to design a microchannel and studied the diffusion process of the catalyst layers in the microchannel reaction. The thickness of the catalyst in the microchannels could be controlled by applying a carrier coating, and thus the reaction could be optimized by changing the flow velocity in the microchannels.

Porous metal fiber material are a new type of material that can be fabricated, have the three-dimensional reticulated structure, interconnected pores, high porosity, and large specific surface area [15,16]. Porous metal fiber sintered felt (PMFSF) can be fabricated by a low temperature solid-phase sintering procedure. It has been previously proposed that a single porous metal fiber material can be integrated into a microreactor and used as a catalyst support. Owing to their high specific surface area and better heat and mass transfer performance, the use of PMFSFs as catalyst support was found to effectively improve the loading performance of the catalyst, and also enhance the methanol conversion and H₂ flow rate [17-19]. Even though PMFSFs have many advantages, their low thermal stability and quickly decreasing activity of the catalyst supported on the PMFSF generally led to a decrease in the hydrogen production performance. In order to address those problems, several studies have focused on improving PMFSFs. Kiwi-Minsker et al. [20] developed the sintered metal fibre filters for efficient structured combustion catalysts. The enhanced overall catalytic performance was observed in adiabatic catalytic reactor during propane combustion. Bryan et al. [21] designed a "sandwich" microreactor with sintered metal fibers as catalyst support. It is found that the high thermoconductivity of sintered metal fibers improves the heat transfer, avoiding hotspot formation during exothermic reactions. Zhao et al. [22] deposited Au onto a thin-sheet microfibrous structure using Ni fiber with high heat conductivity. The optimized microfibrous structure was obtained for low-temperature gas-phase alcohol oxidation.

To date, research work has been primarily focused on microchannel structure design and the stability of the catalyst. Furthermore, there are only a few reports that describe the use of porous Cu-Al fiber sintered felt (PCAFSF) as a catalyst support [23]. It is already known that PCAFSFs play an important role in catalyst coating, and affect the specific surface area and heat and mass transfer efficiency [19]. In this study, a novel PCAFSF was developed and used as a catalyst support in a cylindrical methanol steam reforming microreactor for hydrogen production. Additionally, the effect of the fiber material, surface morphology, and different proportions of Cu and Al fiber of PCAFSF on the reaction performance was investigated in detail.

Experimental procedures

Fabrication process of PCAFSF

According to a previously reported sintering method, the fabrication of PCAFSF mainly includes three steps, namely, the fabrication of cutting fibers, the multi-teeth mold pressing, and solid phase sintering at an appropriate temperature and under a protective gas atmosphere [18]. The schematic diagram of the fabrication process of PCAFSF is shown in Fig. 1. In the first step, the continuous copper and aluminum fibers were fabricated using a multi-tooth tool on a common horizontal lathe. The diameter of the metal fibers was about 100 µm. In order to create beneficial conditions for mold pressing, the copper and aluminum fibers were chipped into short fibers of length ranging from 10 to 20 mm. Later, according to the requirements of porosity, the as-prepared copper and aluminum fibers were mixed and randomly packed into a predetermined packing chamber of mold pressing equipment, and then a pressure was applied to the metal fibers from the bolts. In this way, the semi-finished PCAFSF with the same shape as that of the predetermined packing chamber was obtained. Subsequently, sintering process was carried out in a box-type furnace (No: FXL-12-11) that provided a hydrogen gas atmosphere with a constant pressure of 0.3 MPa. The sintering temperature and holding time for PCAFSF were 630 °C and 30 min, respectively. When the sintering process was completed, the sample was removed from the furnace and cooled in air to room temperature. Finally, the mold pressing equipment was disassembled and the PCAFSF was ready for loading with the catalyst. The optical images of PCAFSF produced following this manufacturing procedure are shown in Fig. 2.

Calculation of porosity for PCAFSFs

In this work, the PCAFSF samples were 40 mm in diameter and 2 mm in height. Since the obtained PCAFSF has a regular geometric shape, its average porosity could also be calculated using the mass-volume method with the following formula [18]:

$$E(\%) = \left(1 - \frac{M}{\rho V}\right) \times 100 \tag{1}$$

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