



# Optimizing the porosity configuration of porous copper fiber sintered felt for methanol steam reforming micro-reactor based on flow distribution



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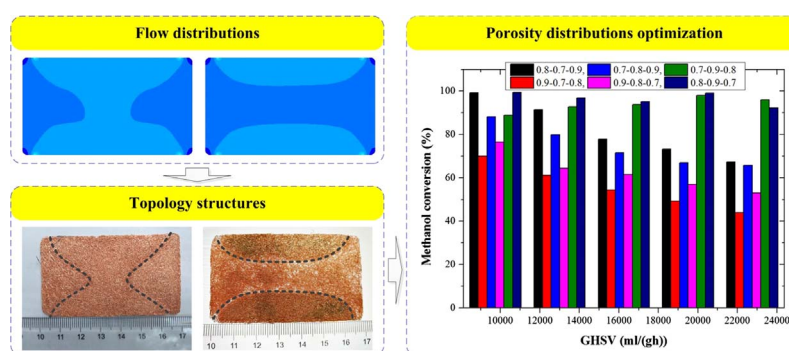
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## HIGHLIGHTS

- A two-step optimization method was proposed for a micro-fibrous MSR catalyst support.
- The support's topology structure was determined based on flow distribution.
- The support's porosity distribution was optimized through experiment investigation.
- The support PCFSF-LRs presented better reaction performance than PCFSF-UUs did.
- The support PCFSF-LR of 0.7–0.9–0.8 presented the best reaction performance.

## GRAPHICAL ABSTRACT



## ARTICLE INFO

### Keywords:

Methanol steam reforming  
Micro-fibrous catalyst support  
Porous copper fiber sintered felt  
Structure optimization  
Porosity configuration

## ABSTRACT

Methanol steam reforming inside micro-reactors is considered as one of the effective approaches for on-board supplying hydrogen for fuel cells. Porous copper fiber sintered felts (PCFSFs) are a new kind of catalyst support for micro-reactors developed in recent years. However, there is a lack of approach to control their porosity configurations due to their random structure. A two-step optimization method was proposed to optimize the PCFSFs' porosity configuration. Firstly, the topology structures of PCFSFs were optimized based on the best flow distributions obtained from macroscopic numerical analyses, and two kinds of PCFSFs with twelve porosity distributions were fabricated through the multi-step mold pressing and solid-phase sintering method. Secondly, the porosity distributions of the semi-optimized PCFSFs were optimized by investigating their reaction characteristics under different gas hourly space velocities (GHSVs) and reaction temperatures. The results indicated that PCFSFs with porosity distribution along the Left-Right direction (PCFSF-LRs) exhibited better reaction performance than PCFSFs with porosity distribution along the Upside-Underside direction (PCFSF-UUs). The methanol conversion and H<sub>2</sub> flow rate for the PCFSF-LRs with porosity distribution of 0.7–0.9–0.8 and 0.8–0.9–0.7 kept on a high level (above 92% and 0.59 mol/h, respectively), regardless of the change of GHSVs and reaction temperatures in most cases. The H<sub>2</sub> selectivity of the PCFSF-LR of 0.7–0.9–0.8 was the highest

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<https://doi.org/10.1016/j.apenergy.2018.02.102>

Received 1 November 2017; Received in revised form 2 February 2018; Accepted 15 February 2018

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under large GHSVs and all tested reaction temperatures. The demonstrated effect of counteracting, even reversing the conventional influence of the GHSV and temperature on the performance of methanol steam reforming may be attributed to the more uniform flow distribution in the two PCFSF-LRs.

## 1. Introduction

Being widely studied and applied in many fields such as vehicle power supply, distributed power station, portable electronics, residential power sources, polymer electrolyte membrane fuel cells (PEMFCs) with hydrogen or hydrogen-rich gas as fuel are considered as one of the best alternative energy conversion devices, which can convert the chemical energy of the fuel directly into electrical energy [1–6]. Since it is well known that hydrogen is an important clean-energy source [7–9], PEMFCs possess advantages not only of quick response, relatively high efficiency and low operating temperature, but also of low/zero emission [10–12]. Nonetheless, direct hydrogen supply is still one of the key barriers hindering the commercialization of PEMFCs, due to the lack of reliable distribution network, storage and safety management handling [6,8,11–13]. This initiates research society towards indirect and on-board hydrogen production reforming from either high-energy liquid fuels or natural gas, such as methanol, methane, ethanol, glycerol, alcohols, dimethyl ether etc. [14–19]. Among them, methanol is considered as one of the most reliable fuel candidates for PEMFCs, due to its readily available, high H/C ration, low cracking and boiling temperatures, and low reaction temperature [19–21].

As compared to other methanol reforming processes used for hydrogen production, methanol steam reforming (MSR) is considered to be the most attractive for PEMFC, because of its higher yield of hydrogen and lower generation of CO that is known as the poison of the fuel cell [22–25]. Accordingly, numerous catalytic MSR reactors have been developed. In general, they can be categorized into packed-bed reactors and micro-structured reactors [26,27]. The former is widely used in industry and has the advantages of moderate cost, better catalyst availability and reproducibility and greater understanding of catalyst performance [28]. However, they suffer the problems such as low axial and radial heat conductivity, pronounced axial temperature profiles, high-pressure drop, and slow dynamic response, thus resulting into the crucial inefficiency of the catalytic reforming process [21,26,29,30]. Compared with the conventional packed-bed reactors, micro-structured reactors have the advantages of compactness, rapid

startup and high safety performance [30]. More importantly, the mass and heat transfer in the micro-structured reactors can be enhanced due to their high specific surface area and short conduction paths [11,26,30]. This makes the micro-structured reactors be especially favored by transportation and portable power applications [26].

Catalyst support/substrate is one of the key structures in these micro-structured reactors and can be used to classify these reactors [26,27]. It can greatly improve the performance of catalyst loading and reforming reaction, by optimizing the contact time of the reactants and avoiding the formation of undesired by-products [31,32]. Typical catalyst supports are composed of micro-channels with an equivalent diameter in the range of 10–500  $\mu\text{m}$  [33–35]. In recent years, to develop micro-structured reactors with higher performance of MSR reaction, novel catalyst supports have been intensively studied, including, for example, innovative micro-channels [36–38], foam [11,39], micro-pin-fin arrays [40,41], cube-posts [9], string [26,42,43], fibrous fibers [29,44,45], and etc. Among them, porous copper fiber sintered felts (PCFSF) is a novel catalyst support. Compared with other structures, PCFSFs have advantages mainly in two aspects. Firstly, producing by cutting method and solid sintered process [45], PCFSFs inherently have three-dimensional porous structure and multi-scale morphology [46] with larger superficial roughness ( $R_a$  is 5–20  $\mu\text{m}$ ,  $R_y$  15–60  $\mu\text{m}$  [47]) and specific surface area (0.832  $\text{m}^2/\text{g}$  for a single fiber and 0.789–2.032  $\text{m}^2/\text{g}$  for a PCFSF with 80% porosity [42]). This makes a PCFSF be uniformly coated with enough catalyst [48], thus favoring better adhesion of catalytic layer and extending catalyst lifetime [13,27]. More importantly, additional surface pre-treatment procedures, such as anodic oxidation, thermal oxidation, chemical treatment, primer deposition, etc. [27], needed by other metal catalyst supports to enhance the specific surface area for MSR reaction can be avoided. In this sense, the cost of producing PCFSFs will be reduced. Secondly, PCFSFs have the potential to simultaneously act as different conventional components of a PEMFC such as catalyst support [45,48,49], flow field plate (gas channel) [50], gas diffusion layer [51], etc. Consequently, the PEMFC's structure can be greatly simplified and thereby effectively reducing the cost [51].

As is pointed by Wang and Wang [30], besides the catalyst

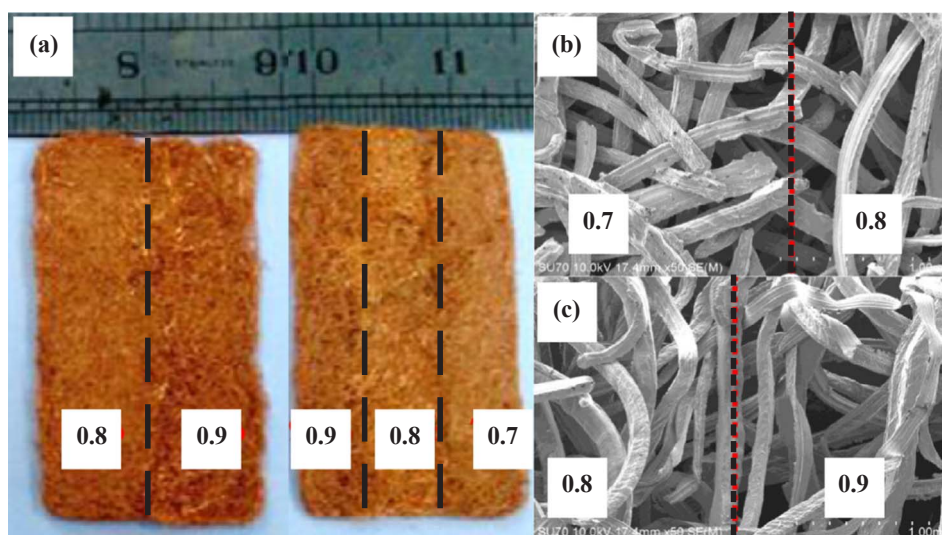


Fig. 1. Appearance and SEM micrograph of gradient PCFSFs: (a) two examples of gradient PCFSFs; (b) between 0.7 and 0.8 porosity portion; (c) between 0.8 and 0.9 porosity portion [31].

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