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 H2 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 H2 flow rate compared to the smooth PCAFSF. In case of the PCAFSF, the methanol conversion and H2 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 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
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]:

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

(1)
where $V$ is the volume of PCAFSF (cm$^3$), $M$ is the mass of PCAFSF (g), $\rho_{Cu}$ is the density of copper material (g/cm$^3$) and $\rho_{Al}$ is the density of aluminum material. The different proportions of PCAFSF with different porosities are shown in Table 1.

**Loading procedure of catalyst**

Before loading the catalyst, the PCAFSFs were cleaned with ethanol in an ultrasonic bath for 5 min to remove any organic matter.
substances. In this study, a two-layer impregnation method was employed to load the catalyst on the PCAFSFs. Therefore, (capital) Cu(NO₃)₂, Zn(NO₃)₂, Al(NO₃)₃ and Zr(NO₃)₄ were mixed in the molar ratio 11:6:4:1 in distilled water with copper ion concentration of 4.6%, and then the solution was further mixed with an Al₂O₃ colloidal solution to prepare the catalyst precursor. Zr in the Cu-based catalysts not only enhanced the deoxidization of the copper, but also increased the surface area and the decomposition capability[17,24]. In the loading process of the catalyst, the PCAFSFs were sufficiently impregnated using the catalyst precursor solution, and subsequently dried in an oven. The above mentioned steps were repeated until the loading of the precursor solution was finished. In this way, the PCAFSFs loaded with Cu/Zn/Al/Zr catalyst could be obtained[24]. The amount of the catalyst on each piece of PCAFSFs was approximately 0.3 g. In order to activate the catalyst, the loaded PCAFSFs were calcined at 400 °C for 2 h under N₂ at a flow rate of 100 mL/min. Then, they were calcined at 300 °C for 1 h in a mixture of N₂ (100 mL/min) and H₂ (50 mL/min). The morphology of the naked and loaded Decafs were observed using scanning electron microscopy (SEM) (SU-70, Hitachi, Japan). The ultrasonic water bath vibration method was used to evaluate the adhesive strength of the catalyst. The catalyst-loaded PCAFSF was placed in glassware filled with distilled water, which was then placed in the ultrasonic vibration device (No: KQ5200DB, Anshan Ultrasonic Instruments Co., Ltd, China). After emulsification for several minutes with an input power of 200 W, the PCAFSF was dried in a blast oven and then weighed on electronic balance.

**Design of cylindrical microreactor**

Fig. 3 shows the diagram of the cylindrical laminated methanol steam reforming microreactor for hydrogen production. The microreactor consists of an evaporation chamber, a reaction chamber, heating rods, thermocouples, and inlet and outlet tubes. Three grid trays were used in the evaporation chamber to increase the amount of water vaporization and maintain flow uniformity. A mixture of methanol and distilled water (1:1.3 mol ratio) was pumped into the evaporation chamber through the inlet tube with an injection pump [25].

<table>
<thead>
<tr>
<th>Number</th>
<th>Materials</th>
<th>Porosity</th>
<th>Materials composition (g)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Type 1</td>
<td>Cu/Al</td>
<td>70%</td>
<td>Cu-3.34/Al-1.02</td>
</tr>
<tr>
<td>Type 2</td>
<td>Cu/Al</td>
<td>70%</td>
<td>Cu-2.24/Al-1.36</td>
</tr>
<tr>
<td>Type 3</td>
<td>Cu/Al</td>
<td>70%</td>
<td>Cu-1.12/Al-1.70</td>
</tr>
<tr>
<td>Type 4</td>
<td>Cu/Al</td>
<td>80%</td>
<td>Cu-3.34/Al-0.34</td>
</tr>
<tr>
<td>Type 5</td>
<td>Cu/Al</td>
<td>80%</td>
<td>Cu-2.24/Al-0.68</td>
</tr>
<tr>
<td>Type 6</td>
<td>Cu/Al</td>
<td>80%</td>
<td>Cu-1.12/Al-1.02</td>
</tr>
</tbody>
</table>

![Fig. 3 – Diagram of the cylindrical laminated methanol steam reforming microreactor for hydrogen production.](image)
After the mixture was removed in the evaporation chamber, the reformate gas was made to react in the presence of the catalyst in the reaction chamber to produce hydrogen-rich gas.

**Methanol steam reforming test**

Fig. 4 shows the schematic diagram of the testing system for the methanol steam reforming microreactor for hydrogen production.
production. This system consists of a fuel tank, injection pump, evaporator, reformer, thermostat, data collector, condenser, dryer, gas chromatograph, computer, mass flow controller, and soap bubble flow meter. During the experiment, the temperature of the microreactor was controlled by the thermostat. A mixture of methanol and water (1:1.3 mol ratio) was pumped into the evaporation chamber. Next, the hydrogen-rich gas was passed through the 0 °C cold trap to condense any remaining unreacted methanol and water. An on-line gas chromatograph (No: GC1690) fitted with a TCD detector was then used to analyze the composition of the hydrogen-rich gas. And we have detected the CO percentage of rich hydrogen gas is about 2%. The flow rate of the hydrogen-rich gas was indicated by a soap bubble flow meter.

Methanol conversion $X_{\text{MeOH}}$ and hydrogen flow rate $n_{\text{H}_2}$ were calculated as follows [26].

$$X_{\text{MeOH}} = \frac{F(y_{\text{CO}} + y_{\text{CO}_2})}{22.4 \times n_{\text{MeOH.in}}} \times 100\% \quad (4)$$

$$n_{\text{H}_2} = F \times y_{\text{H}_2} \quad (5)$$

where $F$ is the normal flow rate of effluent gas, $y$ is the volumetric fraction, and $\text{MeOH.in}$ is the molar flow rate of methanol fed into the microreactor.

Results and discussion

Microstructure and EDX test of Cu-Al PCAFSF

Fig. 5 shows the SEM images of PCAFSF with 80% porosity (Type 6). It can be seen that the PCAFSF has a three-dimensional reticulated structure, and interconnected pores with a relatively uniform distribution of copper fiber and aluminum fibers, as shown in Fig. 5a. These microstructure characteristics are beneficial for a catalyst coating. The SEM result of the combination of copper and aluminum fibers, copper and copper fibers, aluminum and aluminum fibers is shown in Fig. 5b, c, and d, respectively. It is also found that the cross-connection points and parallel-connection points between the metal fibers were observed. The EDX testing results of PCAFSF on the four positions corresponding to
Point 1, 2, 3, 4 in the Fig. 5 is shown in Fig. 6. It is evident from Fig. 6.1 and 2 that when the aluminum fiber surface was near the copper fibers, the percentage of aluminum weight was decreased while the percentage of copper weight was increased. Especially, the percentage of weight of copper was increased to 48.7%, as shown in Fig. 6.2. From Fig. 6.3 and 4, when the copper fiber surface was closer to the aluminum fibers, the percentage of copper weight was decreased and the percentage of aluminum weight was increased. These results indicate that there is a migration of elements between the copper and aluminum fibers takes place in the sintering process. This phenomenon may promote the formation of a stable sintering point between the copper and aluminum fibers. The distribution of the surface elements over the Cu catalyst were characterized using the EDX, as shown in Fig. 7. The elemental distribution indicated that the Cu element has been highly dispersed on the surface of the PCAFSF.

Catalyst loading performance

Fig. 8 shows the effective catalyst loading amount of smooth and rough PCAFSF, PCFSF and PAFSF with same porosity (80%). The amount of loaded catalyst for different supports was approximately 0.3 g, which was closed to the theoretical amount. The difference in effective loading among fiber sintered materials was not significant. It is indicated that the two-layer impregnation method is good way to load the catalyst. After ultrasonic vibration for 1 min, the effective catalyst loading amount of the fiber sintered materials were found to differ, with the PCFSF exhibiting the largest effective catalyst loading amount. It was also found that the PAFSF showed the smallest effective catalyst loading amount, followed by that of smooth PCAFSF. These results may be attributed to the fact that an aluminum oxide layer is fairly easily formed on the surface of PAFSF during the sintering process, such that a sintering neck between fibers is difficult to be produced. In contrast to the smooth PCAFSF, surface

![Graph](image1)

**Fig. 8** — Effective catalyst loading amount of smooth PCAFSF, rough PCAFSF, PCFSF and PAFSF with same porosity (80%).

![Graph](image2)

**Fig. 9** — Reaction performance of PCAFSF, PCFSF, PAFSF as catalyst support with same porosity under different GHSVs at 300 ℃ reaction temperature: (a) Methanol conversion; (b) H2 flow rate.

<table>
<thead>
<tr>
<th>Num</th>
<th>Fiber surface</th>
<th>Materials composition</th>
<th>Porosity</th>
<th>Specific surface area (×1000 m²/m³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Rough Cu</td>
<td>Cu-4.47</td>
<td>80%</td>
<td>155</td>
</tr>
<tr>
<td>2</td>
<td>Rough Al</td>
<td>Al-1.36</td>
<td>80%</td>
<td>87</td>
</tr>
<tr>
<td>3</td>
<td>Smooth Cu/Al</td>
<td>Cu-1.1/Al-1.02</td>
<td>80%</td>
<td>216</td>
</tr>
<tr>
<td>Type 4</td>
<td>Rough Cu/Al</td>
<td>Cu-3.3/Al-0.34</td>
<td>80%</td>
<td>386</td>
</tr>
<tr>
<td>Type 5</td>
<td>Rough Cu/Al</td>
<td>Cu-2.2/Al-0.68</td>
<td>80%</td>
<td>426</td>
</tr>
<tr>
<td>Type 6</td>
<td>Rough Cu/Al</td>
<td>Cu-1.1/Al-1.02</td>
<td>80%</td>
<td>441</td>
</tr>
</tbody>
</table>
microstructures can be observed on rough PCAFSF and these characteristic are beneficial for catalyst loading. For these reasons, the catalyst loading on rough PCAFSF after ultrasonic vibration for 1 min was found to be much large in comparison to other samples.

**BET analysis**

The specific surface areas of the fiber sintered materials were measured by the Brunauer-Emmett-Teller (BET) method using a Micromeritics Tristar 3020 surface area and porosity analyzer. Prior to the measurements, all samples were degassed at 150 °C for 6 h to remove impurities. The specific surface area of copper PCFSF, FAFSF, smooth PCAFSF, and rough PCAFSF (Type 4, 5, 6) with 80% porosity are given in Table 2. It was found that the rough PCAFSF had the larger specific surface area. Among the single metal materials, the specific surface area of PCFSF was larger than that of PAFSF. It was interesting to note that after aluminum fibers were added to the copper fibers, the specific surface area of PCAFSF became larger. It is possible that copper and aluminum fibers mixed with each other in favor of joining each other, the surface of fiber sintered materials produces more small pores and larger specific surface area.

**Hydrogen production of microreactor with fiber sintered materials**

**Different fiber sintered materials**

Fig. 9 shows the comparison of reaction performance of PCAFSF (Type 6), PCFSF (80%) and PAFSF (80%) as catalyst support with different GHSV. From Fig. 8, compared to the PCFSF and PAFSF, the highest methanol conversion and H2 flow rate were observed when the reactant was fed into PCAFSF at different GHSVs. It is abutted that the addition of aluminum fibers to copper fibers improves the specific surface area of PCAFSF. The uniformity of the distribution of the catalyst on the PCAFSF can be improved to increase the effective contact area between the catalyst and reactant. Fig. 10 shows a comparison of the reaction performances of PCAFSF (Type 6), PCFSF (80%) and PAFSF (80%) as catalyst supports at different reaction temperatures. Compared with the PCFSF and PAFSF, PCAFSF also showed the highest methanol conversion and H2 flow rate when the reaction temperatures was below 340 °C. These results may be explained by the fact that methanol steam reforming is an endothermic reaction and the addition of aluminum fibers to copper fibers to improves the heat and mass transfer performance of PCAFSF which results in improved catalytic activity.

Fig. 10 – Reaction performance of PCAFSF, PCFSF, PAFSF as catalyst support with same porosity under different reaction temperatures at 9029.1 mL/g h GHSV: (a) Methanol conversion; (b) H2 flow rate.

Fig. 11 – Comparison of reaction performance of surface morphology with rough PCAFSF and smooth PCAFSF as catalyst support with different GHSV at 300 °C reaction temperature: (a) Methanol conversion; (b) H2 flow rate.

Owing to its larger specific surface area, the hydrogen production performance for this material was higher, especially at lower reaction temperatures. However, at temperatures above 340 °C, the hydrogen production performance resulting from PCFSF was the highest in comparison with PCAFSF and PAFSF, owing to its superior catalyst loading performance and higher thermal conductivity.

Different surface morphology

Fig. 11 shows a comparison between the reaction performances of rough PCAFSF (Type 6) and smooth PCAFSF as catalyst support with different GHSV. Compared with the smooth PCAFSF, higher methanol conversion and H₂ flow rates was obtained when the rough PCAFSF was used as the catalyst support under the same GHSV. This was because the surface microstructure of rough PCAFSF facilitated a more effective catalyst loading compared to that on smooth PCAFSF, and consequently a lesser amount of the catalyst was lost during the reaction. Fig. 12 shows the comparison of the reaction performances of these two catalyst support at different reaction temperatures. It was found that the rough PCAFSF led to a higher methanol conversion and H₂ flow rate at all tested temperatures. This may be explained by the better dispersion of the catalyst on the surface of rough PCAFSF. This material has a large specific surface area, and consequently, the contact area between the catalyst and reactant is larger. For these reason, much higher reaction efficiency and performance relating to hydrogen production on rough PCAFSF is observed.

Different Cu-Al metal fiber mixed ratios

Different Cu-Al metal fiber mixed ratios with 70% porosity. Fig. 13 shows the methanol conversion and H₂ flow rate obtained with 70% porosity PCAFSF as catalyst support under different GHSVs. With decreasing weight of copper fibers and increasing aluminum fiber weight, there was a gradual increase in methanol conversion and H₂ flow rate. This was mainly because of the increased specific surface area of the material owing to its conversion from a single-metal fiber to one comprised of mixed metals, which is favorable for catalyst loading and strengthens the reaction process, thus, methanol conversion and H₂ flow rate are higher. Fig. 14 shows the methanol conversion and H₂ flow rate obtained with 70% porosity PCAFSF as catalyst support under different reaction temperatures. The PCAFSF (Type 3) with 70% porosity led to the best methanol conversion and H₂ flow rate. This was mainly because the increase in aluminum fiber weight and decrease in copper fiber weight improved the catalyst loading performance and specific surface area of this material. Furthermore, the best reaction performance in the
micromer reactor under different GHSVs and reaction temperatures was achieved with this material over PCAFSF (Type 3).

Different Cu-Al metal fiber mixed ratio with 80% porosity. Fig. 15 shows the methanol conversion and H₂ flow rate obtained with 80% porosity PCAFSF as catalyst support under different GHSVs. When the reaction temperature was 300 °C, methanol conversion was decreased while the H₂ flow rate increased with increasing GHSV. This is mainly due to the resident time of reactant is decreased, and then the reforming reaction cannot be fully carried out on the surface of catalyst, the similar result was also reported in Ref. [17]. In addition, at 80% porosity, methanol conversion and H₂ flow rate increased when the copper fiber weight was decreased and the aluminum fiber weight was increased. This was because the catalyst loading performance and specific surface area of PCAFSF had been improved with addition of aluminum fibers, which proved favorable for catalyst loading and heat and mass transfer of the catalyst [27,28]. Thus, higher methanol conversion and H₂ flow rate are obtained.

Catalytic stability of catalyst test

The PCAFSF (Type 6) was used as a catalyst support in order to evaluate the stability of the methanol steam reforming reaction at 300 °C, using a methanol-to-water molar ratio of 1:1.3 and a GHSV of 9029.1 mL/g h. It can be easily seen in Fig. 17 that methanol conversion and H₂ flow rate decreased slightly with increasing reaction time. In the first 12 h, the catalytic activity remained high with the methanol conversion at 90%, and the H₂ flow rate at 0.43 mol/h. As the reaction time increased, the methanol steam reforming and H₂ flow rate decreased slightly. This was because the catalyst was deactivated over time owing to some carbon deposition and catalyst loss, resulting in decreased catalyst activity. Nevertheless, even after 32 h, the methanol conversion and H₂ flow rate remained at above 60% and 0.32 mol/h, respectively.

The laminated PCAFSF was used as a catalyst support in a cylindrical methanol steam reforming microreactor to increase hydrogen production. The specific surface areas and catalyst loading strengths and hydrogen production of PMFSF were measured. In comparison with PCFSF and PAFSF, the PCAFSF had the highest specific surface area and catalyst loading strength. Different PMFSF materials were used as catalyst supports for the methanol steam reforming reaction for hydrogen production. It was found that rough PCAFSF had a large specific surface area and high catalyst affinity, and as it showed better low-temperature activity, the reaction performance was also better. Compared to smooth PCAFSF with same porosity (80%), the rough PCAFSF demonstrates much higher methanol conversion and H₂ flow rate. Lastly, the weight ratios of the copper and aluminum fibers were changed under the same porosity, and our results shows that at the same porosities of 70% and 80%, the methanol conversion and H₂ flow rate increased as the weight of copper fibers was decreased, and that of the aluminum fibers increased. The use of the PCAFSF(Type 6) as a catalyst support can produce the best reaction performance for hydrogen production.

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**References**


**Fig. 16** — Comparison of reaction performance of PCAFSF with 70% porosity as catalyst support with different reaction temperatures at 9029.1 mL/g h GHSV: (a) Methanol conversion; (b) H₂ flow rate.

**Fig. 17** — Stability of PCAFSF with Type 6 as catalyst support loaded with Cu/Zn/Al/Zr catalyst of methanol steam reforming reaction with same reaction temperature (300 °C) and GHSV(9029.1 mL/g h).