



Passive vapor-feed direct methanol fuel cell using sintered porous metals to realize high-concentration operation



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HIGHLIGHTS

- A vapor-feed direct methanol fuel cell with pervaporation membrane is developed.
- A sintered porous metal-fiber plate is used to enhance water back diffusion.
- The fuel cell can be fed with highly-concentrated or even neat methanol.
- The effects of both structural and operational factors are discussed.

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ABSTRACT

The use of methanol vapor is helpful to reduce the effect of methanol crossover in a direct methanol fuel cell. This study develops a passive vapor-feed direct methanol fuel cell (VF-DMFC) based on a pervaporation membrane and sintered porous metals which act as functional layers for effective control of the mass transfer process. The feasibility of using this method is experimentally validated. For the cathode, a sintered porous metal-fiber plate (SPMFP) with great hydrophobicity is used as a water management layer to enhance water back diffusion from the cathode to the anode. Results indicate that the use of a SPMFP promotes a higher cell performance especially when a higher methanol concentration is used. The highest peak power density of 19.3 mW/cm² is achieved at an ambient temperature when 12 M methanol is supplied. Neat-methanol operation is also viable under this condition. The cathode current collector (CC) with a window-like pattern yields a higher cell performance than the parallel-channel. For the anode, the use of a hydrophilic sintered porous metal-powder plate (SPMPP) embedded in the anode CC reduces the cell performance due to limitation of methanol delivery. This work also reports the influences of methanol concentration and vapor chamber temperature.

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1. Introduction

Hydrogen-based polymer electrolyte membrane (PEM) fuel cell is recognized as a promising candidate to replace the traditional fossil energies, which has great potential to be used for vehicular and distributed power generation. However, the use of pressurized gaseous hydrogen must bring about great challenges in fuel storage, transportation and delivery [1]. Although using a fuel reforming reactor seems to be an attractive solution for online hydrogen generation, it must increase the system complexity, cost and parasitic power losses, thereby hindering its real application. In this situation, the fuel cells based directly on liquid fuels face a good opportunity to be put into practical use [2]. In this field, the direct

methanol fuel cell (DMFC) has attracted wide attentions due to its advantages in high energy density, convenient operation and compact configuration, whereas it usually suffers a far lower performance than the hydrogen-based fuel cell because of catalytic inactivity, low-temperature operation and especially methanol crossover (MCO) [3–8].

As is well-known, the presence of MCO inevitably causes severe voltage losses because the permeated methanol at the cathode may directly react with the oxygen and thus lead to a mixed potential. This issue must be well addressed in order to enhance the output of a DMFC [6,7]. To this end, some researchers attempt to feed methanol vapor instead of liquid fuels to reduce the effect of MCO [9]. As a result, the vapor-feed DMFC (VF-DMFC) permits the use of concentrated or even neat methanol, thus increasing its energy density and prolonging its operating time for a run of fueling. An active way to produce methanol vapor is to utilize a vaporizer

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driven by electric heating, which facilitates fast start-up and convenient control of the vaporization process [10–13]. However, this method requires extra power consumption leading to lower system efficiency. To overcome such deficiencies, an alternative means is to introduce produced heat from an external electronic device with the aid of a heat pipe or a heat spreader [14,15]. This sounds a good idea as the waste heat can be recycled for methanol vaporization, but how to integrate the assistant heat sources with the fuel cell system is still a troubling issue. An interesting method for heat collection is to combine a catalytic combustor with the fuel cell [12,14,16]. For this embodiment, the outwall of the combustion chamber can serve as a heat sink which helps transfer released heat from the combustor to vaporize the methanol in the fuel chamber of a DMFC. It is also preferable to reuse the exhaust heat from both the fuel cell and combustion reactions to assist in evaporating the methanol. Although this technique abandons the auxiliary power supply, a more complex thermal management of the fuel cell coupled with a combustor is quite necessary.

The active and semi-passive patterns of methanol vaporization inevitably increase the volume, cost and noise of the DMFC system [17]. In order to find a fully passive manner to generate vaporous methanol for DMFCs, many studies focused on the possibility of using a pervaporation membrane which enables the aqueous methanol to be spontaneously transformed into gaseous phase [17–24]. Evidently, the use of a phase-change membrane for a passive VF-DMFC is more able to weaken its dependence on peripheral apparatus and assistant heat sources with no need for substantial change of its architecture. From the perspective of commercialization, this design is more likely to be implemented for portable products. In view of open literatures, the prototypes based on passive vaporization technology have been developed in laboratory scale, but there are still many critical issues worthy of more investigation, including the reactant (e.g. methanol and water) and product (e.g. water, CO₂, and heat) management, and structural and operational optimization. Typically, we have to face a reality that a passive air-breathing VF-DMFC operating at a higher methanol concentration may encounter reaction distress for lack of water content at the anode. Under this condition, how to recover water for anode reaction must be an intractable issue [19–27]. In addition, the negative effect of MCO still happens to this type of fuel cell. This issue should be optimized especially during high-concentration operation. What is worse, some involved factors interact or even conflict with each other, possibly exerting coupling effects on the cell performance. Therefore, how to make reasonable explanations for relevant phenomena are still in suspense.

With the above background, the present work aims to gain a deeper insight into the structural and operational aspects of a fully passive VF-DMFC that uses a pervaporation membrane as the vaporizer. The effectiveness of constructing a mass-transfer-controlling layer made of sintered porous metals was experimentally validated. Information provided in this study may facilitate a more in-depth understanding of major influence factors, and give guidance for design, fabrication and optimization of the passive VF-DMFC.

2. Experimental

2.1. Design and fabrication of the passive VF-DMFC

A schematic view of the in-house VF-DMFC based on the passive pervaporation mechanism is illustrated in Fig. 1. The anode compartment contained a built-in methanol vapor chamber (MVC) located under the current collector (CC). A fuel storage chamber (FSC) filled with liquid methanol was machined in the bottom block. A commercial membrane, i.e. Nafion® 117 (Dupont,

Inc.) with a thickness of 183 μm was sandwiched between the MVC and FSC, acting as a self-governing vaporizer to feed vaporous methanol continuously to the anode of a VF-DMFC. The above two components were both made from polycarbonate. According to our previous experiences [28], we created a circular-hole array pattern with a lower open ratio (28.3%) for the anode CC. For the cathode, two opening patterns were prepared for comparison, namely the parallel through channel (58%) and open window (100%). In this study, all the CCs were made of SUS316L sheet which owns good electrical conductivity and corrosion resistance [29]. Silicon rubber gaskets were used to seal the gap between the neighboring components.

A hydrophilic sintered porous metal-powder plate (SPMPP) was embedded in the anode CC, while a sintered porous metal-fiber plate (SPMFP) was used as the water management layer (WML) included in the cathode CC. This design was intended to establish a hydraulic pressure to force the produced water to flow backward in the cathode-to-anode direction [30]. How these two functional layers affected the mass transfer process and with it cell performance was a focused problem in this study. Their structures and morphology were characterized by scanning electron microscopy (SEM), as shown in Fig. 2. The wettability of the SPMFP was evaluated using an optical contact angle measurement platform (OCA20, Dataphysics, Inc.). Two specimens were selected and three points for each were picked for sessile droplet tests. Results indicate that the contact angles are in the range of 122–134°, which mean that the SPMFP is absolutely hydrophobic (see Fig. 3). In this work, copper was used as the raw material to make the SPMFP/SPMPP with a porosity of 70%. The copper powders were commercially available. The fabrication process of SPMFP could be briefly described as follows. Firstly, the metal fibers were produced through multi-tooth cutting on a horizontal lathe, and then cut into segments with a certain average length, for example, 10–15 mm in this work. After being cleaned, the fibers were put into a mold cavity, flattened and sintered in a programmable sintering furnace. When these processes were completed, the SPMFP samples with prescribed sizes and shapes could be taken out from the mold assembly.

2.2. Preparation of the membrane electrode assembly (MEA)

As shown in Fig. 1, the Nafion® 117 (Dupont, Inc.) was also used to make the membrane electrode assembly (MEA) which had an effective area of 9 cm². The diffusion layer (DL) was made up of a carbon-paper (TGP-H-060, Toray, Inc.) backing layer (BL) and an additional micro porous layer (MPL). On the anode side, the carbon paper was untreated, and the MPL was composed of carbon black, 10% Nafion solution and isopropyl alcohol. Differently, the cathode used wet-proofed carbon paper as the BL and 15% PTFE latex to make the MPL hydrophobic to enhance water back diffusion. A mixture of catalyst powders (Johnson Matthey, Inc.), 20% Nafion and isopropyl alcohol was attached onto the MPL to form a catalyst layer (CL). The catalyst loadings for the anode and cathode were 2 mg/cm² Pt–Ru (1:1, a/o) and 4 mg/cm² Pt, respectively. The last step was to bond the membrane and catalyst-coated DLs together by hot-pressing at an appropriate compaction force of 120 kg/cm² for 2 min [31].

2.3. Testing conditions

Before the fuel cell started, the MVC and FSC were both cleaned by deionized water so as to remove the residual chemicals on their inner walls. In order to achieve more reliable data record, the fuel cell electrode was activated at a constant load for half a day. For each run of test, we injected a dose of methanol compatible with the volume of the FSC. The cell performance was mainly characterized by

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