

Optimizing the Anode Structure of a Passive Tubular-Shaped Direct Methanol Fuel Cell to Operate With High Concentration Methanol

Jing Huang

Travis Ward

Amir Faghri¹

e-mail: faghri@enr.uconn.edu

Department of Mechanical Engineering,
University of Connecticut,
Storrs, CT 06269

In order to take full advantage of the high energy density available in methanol fuel, one must use high concentration methanol in direct methanol fuel cells (DMFCs). However, this causes severe methanol crossover and leads to low power density and fuel efficiency. In this work, a tubular shape is adopted to generate higher volumetric power density; porous polytetrafluoroethylen (PTFE) membranes at the anode are used to control methanol transport with a high concentration fuel. A novel passive tubular-shaped DMFC is improved to achieve stable operation with methanol concentrations up to 20 M. It is observed that a balance between fuel transport resistance, power density, energy density, and fuel efficiency exists. Increased resistance enhances fuel efficiency, hence, energy density, but limits the fuel supply and causes low power density. With the improved anode structure and higher concentration fuel (1 M to 15 M), the energy output of the tubular DMFC increases 591%, from 0.094 Wh to 0.65 Wh with 2 ml fuel. The power density maintains the same level as 16 mW/cm². For different fuel concentrations, there exists an optimum structure to generate the highest power density, which is a result of minimizing the methanol crossover while also providing sufficient fuel. The discharge characteristic at constant voltage and its effect on fuel efficiency are also discussed. [DOI: 10.1115/1.4007274]

Keywords: DMFC tubular, passive, fuel efficiency, high concentration methanol, performance

1 Introduction

As one of the most promising portable power supply solutions, direct methanol fuel cells (DMFCs) have attracted tremendous research and industrial interest in the last decade. One of the most extraordinary merits of DMFCs is their potential of providing high energy density. In the premise of complete electro-oxidation, a liter of methanol can produce 4820 Wh of electricity, which is much higher than its competitors, such as Li-ion batteries (250~620 Wh L⁻¹) and hydrogen polymer electrolyte membrane (PEM) fuel cells (1556 Wh L⁻¹ at 700 bar). However, in current applications, the performance of DMFCs is impaired by a low power density, which is caused by slow electrochemical kinetics and methanol/water crossover through the electrolyte membrane.

Traditional polymer electrolyte membrane fuel cells (PEMFCs) have a planar shape because of the straightforward structure design. Several research efforts, however, have shown that a tubular-shaped PEMFC would provide a larger active area without increasing the overall volume occupied by the fuel cell [1–3]. For a DMFC, this is especially important because one of the problems of a regular DMFC is its relatively low power density.

A tubular DMFC has several other significant advantages compared to a planar-shaped one: (1) it can operate in all orientations without the lack of methanol contacting the anode catalyst layer; (2) because of a decreased volume, the fabrication cost is reduced;

(3) it has the same shape as existing AA, AAA, D, and C batteries, which would allow for easier conversion between batteries and fuel cells in the future, and (4) with the elimination of flow fields at the cathode, uniform pressure can be applied across the membrane electrode assembly (MEA) [3].

The tubular direct alcohol fuel cells can be divided into two categories. The first is a tubular-cast membrane type, and the second is a wrapped MEA type. For the first type, the membrane is cast into a tubular shape and the catalyst is then sprayed onto the inner and outer sides of the membrane [4–6]. However, this type of tubular DMFC usually has a very poor performance (10–15 mW/cm²) because of its low conductivity and high contact resistance. The second type uses a regular MEA wrapped onto a tubular shaped frame, which serves as the fuel reservoir and as the anode current collector [1,2,7]. An outer frame presses the MEA and the other layers together. In 2007, Cao put forth a conceptual design of a fuel cell stack composed of tubular single cells [8]. Some theoretical evaluations indicated that the stack design may potentially increase the reactive surface area by more than 10 times. Thus, with the same power density per unit active area, tubular DMFCs have a higher volumetric power density; hence, a higher total power output with the same volume.

Another issue for the DMFC is methanol crossover. In most existing works on tubular DMFCs, dilute methanol was used to alleviate this problem. However, it decreased the energy density, which should be one of the most attractive characteristics of the DMFC. Recently, the concept of a passive vapor feed DMFC has attracted more attention because of its potential to directly utilize high concentration methanol and its elimination of the issues related to CO₂ bubble removal. A simple vaporizer was used to passively convert liquid methanol solutions stored in a fuel tank

¹Corresponding author.

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into vapor in order to drive the fuel cell [9–11]. Ren et al. used a silicone membrane as a vaporizer in a passive DMFC. Water crossover was controlled by an optimized passive water management system through back diffusion and back convection [12,13]. Guo and Faghri utilized the advantage of a porous wick structure to separate the methanol reservoir from the MEA without the need for a complex microfluidics subsystem. Pure methanol in a fuel tank was wicked to an evaporation pad to produce methanol vapor. Successful thermal fluids management enabled prototype cells to operate for 6 months without obvious degradation [14–17]. Xu et al. studied the effect of the anode structure on the methanol vapor generation rate of a passive vapor-feed DMFC, which had a membrane vaporizer and a hydrophobic vapor transport layer (VTL). The parameters studied included the VTL thickness, the vaporizer open ratio, and the water management layer thickness. The optimized structure enabled the fuel cell to operate with a fuel efficiency of 62% with neat methanol and output a peak power density of 34 mW/cm² [18].

In this work, we investigate the effect of adapting different anode structures to a passive tubular-shaped DMFC for high concentration methanol solutions, which has not been reported in the literature. The knowledge and techniques obtained through optimizing the performance of planar DMFCs are adopted. These include adding microporous PTFE membranes to increase the transport resistance to methanol, using a Nafion[®] 117 membrane between the methanol reservoir and gas diffusion layer to serve as a pervaporation layer and changing the open ratio of the fuel reservoir exposed to the anode electrode. After the optimization of the structure, the passive tubular-shaped DMFC operated stably for over 5 h with 2 ml of a 20M methanol solution. It is worthwhile to note that this is the first experimental work describing the modification of a tubular-shaped passive DMFC to achieve stable, repeatable, high concentration methanol operation.

2 Experiment

2.1 Structure of the Passive Tubular-Shaped DMFC. The structure of the passive tubular-shaped DMFC frame, catalyst coated membrane, and membrane electrode structure (MEA) are the same as our previous work, as described in Ref. [1]. This work focuses on the structural enhancement to the anode side of the fuel cell to improve fuel cell performance and fuel efficiency with high concentration methanol, up to 20M. Similar materials and

structures were applied to the anode to increase the mass transport resistance as previously explained in Ref. [18].

Six different structures were considered at the anode in our experimental work, as shown in Fig. 1. The MEAs used in the tests are provided by BCS Fuel Cells, Inc. Nafion[®] 115 was chosen as the electrolyte membrane with the consideration of the balance between ion conductivity, mechanical strength, and methanol crossover. The active area is 4 × 5.3 cm². The catalyst loadings are 5 mg/cm² Pt and 5 mg/cm² PtRu for the cathode and anode, respectively. Porous PTFE membranes (Saint-Gobain Zitec[®] G-110) were placed on the outside of the anode diffusion layer to increase the mass transfer resistance from the methanol channel to the reaction sites in the catalyst layer. Nafion[®] 117 was used as a pervaporation membrane in structures S4, S5, and S6. To control the pervaporation rate, a layer of regular nonporous PTFE membrane was placed between the fuel reservoir and the pervaporation membrane. A specific number of holes were perforated on this PTFE membrane to achieve different open ratios (3%, 10%, and 50%) and reduce the total mass transport from the methanol channels to the gas diffusion layer. Structure S3 was considered to have a 100% open ratio. It should be pointed out that one problem with a tubular fuel cell is the flexibility of the cell frame. For planar ones, adding more layers to the cell is not difficult while for a tubular one, adding more layers to the cell increases the thickness so that the frame needs to be redesigned and fabricated.

2.2 Testing. The tests were performed using a Scribner Associates 850e test station. This apparatus is equipped with an electronic load that can record the current, voltage, internal resistance, and temperature of the fuel cell. It is capable of applying a variable voltage or current to the fuel cell and recording the resulting polarization curve. All tests were performed in a controlled laboratory setting with ambient temperatures that ranged between 24–28 °C and relative humidities that ranged between 5%–50%.

Prior to installation on the tubular fuel cell, each MEA was activated by H₂/O₂ and 1M methanol solutions/humidified air in an active mode. Polarization curves were obtained and compared to ensure that all of the MEAs used in the tests had a similar performance. The differences in the peak power density between each MEA were smaller than 5%. The tests that were conducted on each unique fuel cell structure included determining the polarization curve and fuel cell efficiency. The polarization curve was

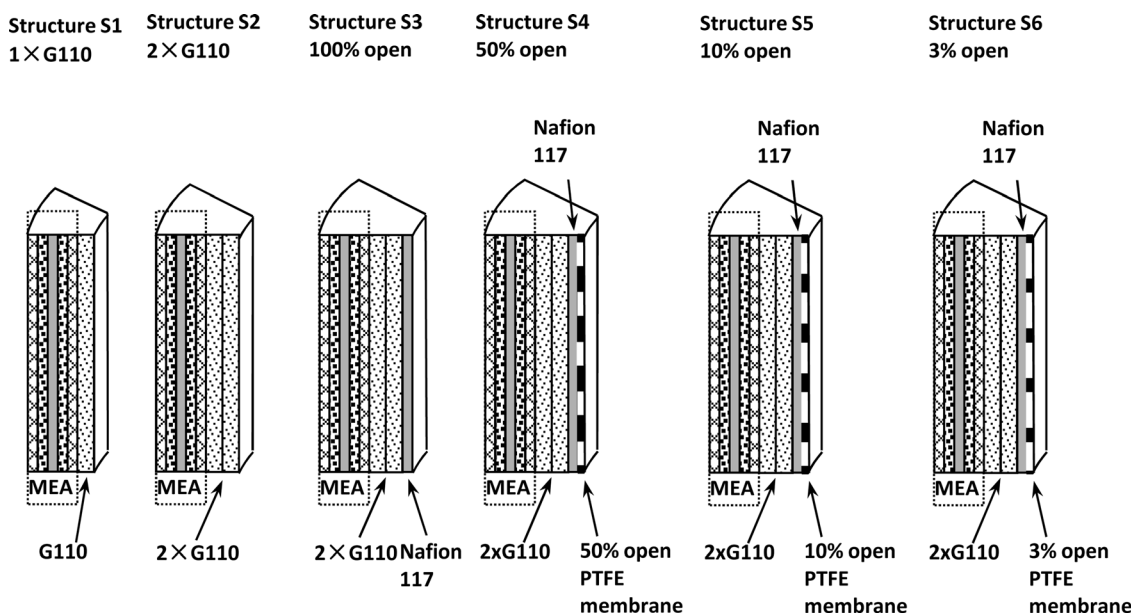


Fig. 1 Different anode structures tested

generated by scanning the voltage from open circuit conditions (OCV) down to 0.05 V, in steps of 0.05 V, and recording the average current density produced by the fuel cell for each voltage. The results were plotted as the voltage versus current density (VI curve) and the power density versus current density to compare the range of voltages, current density, and maximum power density that each cell structure produced. The fuel cell efficiency is a measurement of how much fuel is used by the fuel cell compared to the quantity of fuel provided to the fuel cell, which is useful for concluding how much fuel is lost due to crossover, evaporation, or leakage. It was determined, based on the constant voltage discharge, as follows

$$\eta = \frac{M_{\text{MeOH}} \int_0^T i(t) dt / 6F}{m_{\text{MeOH,sup}}} \quad (1)$$

where t is the time of the discharging process, M_{MeOH} the molecular weight of methanol, T is the total discharging time, $i(t)$ is the transient discharging current density, F is Faraday's constant, and $m_{\text{MeOH,sup}}$ is the mass of methanol provided to the fuel reservoir.

For each structure, methanol solutions with different concentrations, ranging from 1–20 M, were tested. For each case, several polarization curves were obtained until the performance of the cell was stable. Fuel efficiency tests were then carried out. Subsequently, 2 ml of methanol solution was injected into the reservoir. After a stable open circuit voltage was achieved, a constant voltage (0.35 V) discharge was performed until there was no current output in order to determine fuel efficiency.

3 Results and Discussion

3.1 Effect of Structure on Performance. Figure 2 presents the polarization curves for a range of methanol concentrations and structure S4, which has a 50% open ratio pervaporation membrane. For all methanol concentrations of less than 15 M, the peak power density increases with each higher methanol concentration. For low concentrations, such as 3 M and 5 M, the factor limiting the power density is mainly the mass transfer, since the VI curves dropped sharply under high current density. The peak power density reaches 16.5 mW/cm² with the 15 M solution. For the case with the 18 M solution, the peak power density and current density drastically drops, in contrast to the results presented with the 15 M solution. During this test, mass transfer no longer limits the fuel supply for electrochemical reactions; the main reason for the drop in performance is the methanol crossover, which causes parasitic voltage loss on the cathode side and; at the same time, accelerates the depletion of methanol on the anode side. The effect of methanol concentration on the crossover rate can also be seen by the different open circuit voltages, as shown in Fig. 2(b). The OCV for the 3 M solution is 0.545 V, while the OCV for the 18 M solution drops to 0.449 V.

Figure 3 presents the influence of the anode side structures on the fuel cell polarization curves during operation with a 15 M methanol solution. With the increase of the open ratio from 3% to 50%, the mass transport resistance from the fuel reservoir to the catalyst layer is reduced, which permits a better fuel supply to the catalyst layer and leads to higher power and current density. When the open ratio is higher than 50%, however, the methanol concentration at the catalyst layer is too high and, as a result, methanol crossover impairs the performance. This is reflected in Fig. 3 with anode structure S3, in which the power and current both decrease due to significant methanol crossover.

During the experiments, we tested each of the six structures with different fuel concentrations ranging from 1 M–20 M. To simultaneously compare the results, Fig. 4 shows the peak power density achieved, considering each methanol concentration and structure. Generally, for each anode structure, increasing the concentration of the methanol solution increases the power density

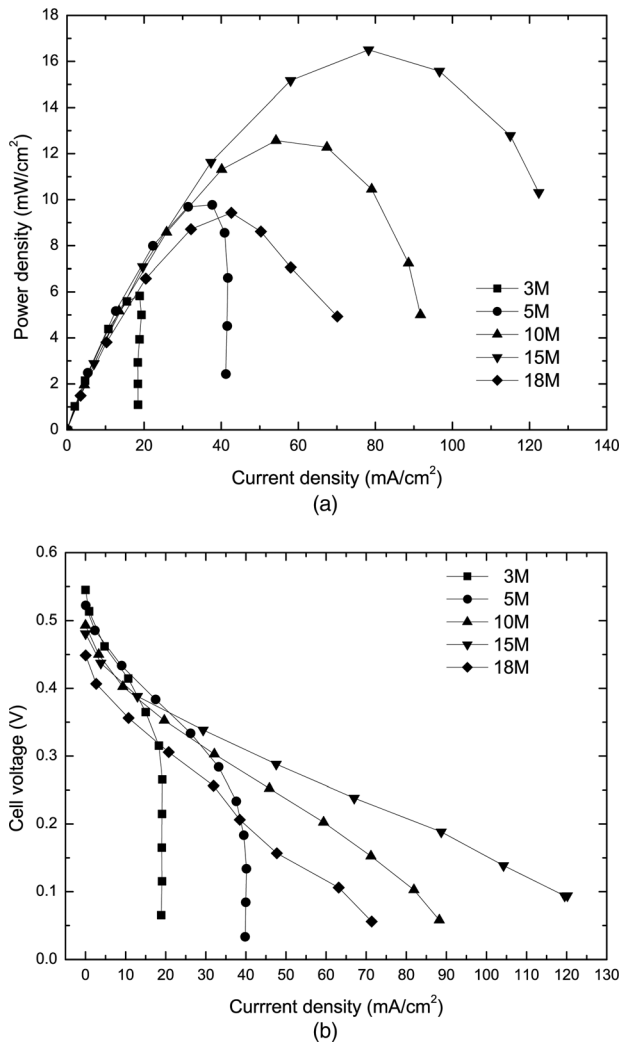


Fig. 2 Polarization and power curves for different concentrations of methanol using structure S4: (a) power density, and (b) current density

until a peak value and then it reduces due to increased methanol crossover. This is the point at which a given structure cannot operate with any higher methanol concentration since the methanol permeates through each of the layers and crosses over the membrane, reducing the performance of the fuel cell. For structure S1, the maximum power density is 18.99 mW/cm², which occurs at a concentration of 5 M. The performance then sharply deteriorates with fuel of a higher concentration. For structure S4, the maximum power density occurs at 15 M, which is 16.5 mW/cm². One can conclude, thus, that by optimizing the mass transfer at the anode side, the methanol crossover can be well controlled and the ability of the fuel cell to handle high concentration methanol is enhanced without significantly impairing the performance. With structure S6 (3% open ratio), the tubular fuel cell is operated in a stable condition for over 5 h with 2 ml of the 20 M methanol solution. The power output, however, is less than 5 mW/cm², which is caused by a limited fuel supply.

The maximum current density follows a similar trend as the peak power density and is plotted in Fig. 5. The decreased current density during the higher concentration operation is caused by water crossover from the anode to the cathode. The mechanism of water crossover is quite similar to that of methanol. It is also caused by electro-osmotic drag during proton transport, diffusion by water concentration gradients, and convection by hydraulic pressure gradients. To achieve a higher current density, more

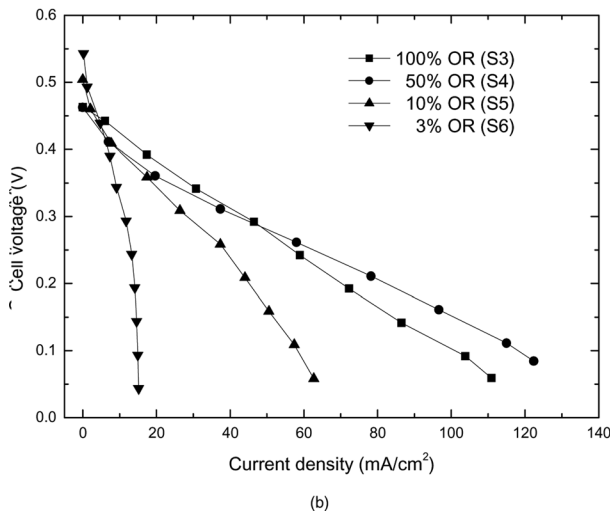
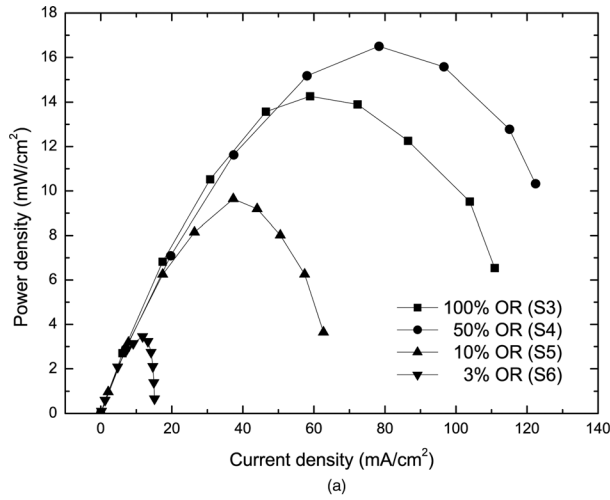


Fig. 3 Polarization and power curves for different anode side structures with a 15M methanol solution: (a) power density, and (b) current density

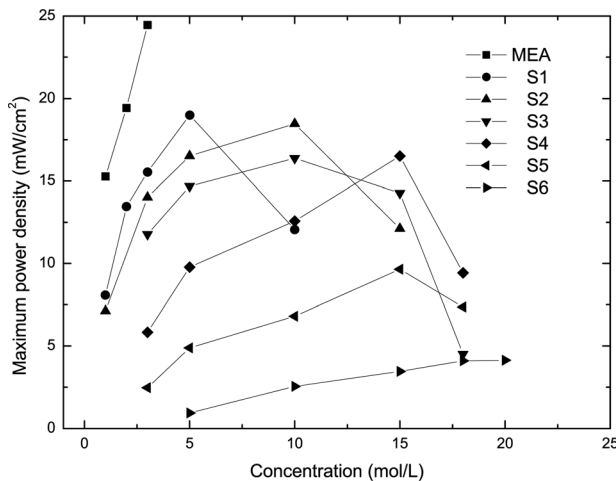


Fig. 4 Peak power density versus methanol solution concentration for different anode structures

protons need to be transported through the Nafion[®] membrane, which causes more water crossover. This impairs the cell performance in two ways. First, more water crossover from the anode to the cathode causes flooding on the cathode side, which limits the oxygen supply. Second, water crossover depletes the water supply

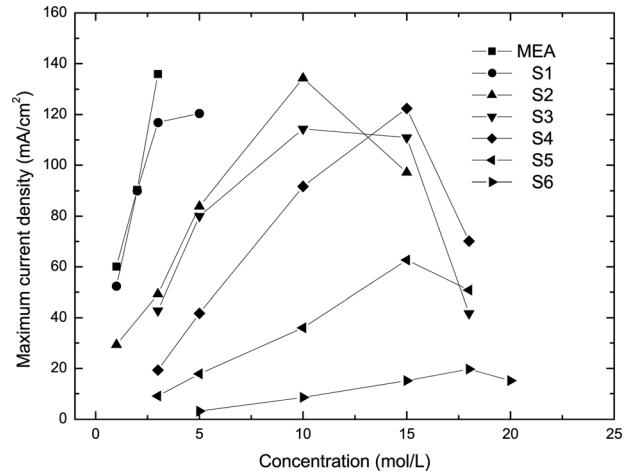


Fig. 5 Maximum current density versus methanol solution concentration

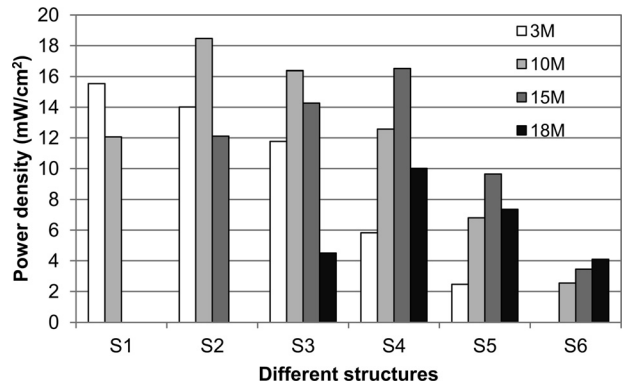


Fig. 6 Maximum power density versus anode structure

on the anode side and; consequently, decreases the methanol oxidation rate. These problems can be alleviated by introducing a water management layer on the cathode side to optimize water distribution across the entire cell.

Figure 6 provides a comparison of the peak power densities for different anode structures with the same methanol concentration. For low concentrations, such as 3 M, the power density monotonically decreases from structure S1 to S6 because of the decreasing methanol supply. For higher concentrations, such as 18 M, the peak power density is achieved with the 50% open ratio pervaporation membrane (structure S4). When the open ratio is higher than 50%, the methanol crossover dominates and impairs the fuel cell performance. Therefore, for each specific methanol concentration, there exists an optimum structure to achieve the highest power density. For higher concentrations of methanol, a higher mass transport resistance is needed at the anode in conjunction with a layer added at the cathode to provide back flux of water from the cathode to the anode.

The temperature of the fuel cell rises as a direct result of methanol crossover. Due to the existence of Pt at the cathode, methanol that crosses over reacts with oxygen, exothermically giving off heat and causing the cell temperature to rise. During our test, the cell temperature is recorded by a thermocouple placed in the cathode air channel and in contact with the cathode diffusion layer. The original temperature of the cell is the same as the ambient air present at the start of each test. The highest temperature is usually measured after 30 min of the fuel efficiency testing since the concentration of fuel in the anode catalyst layer is highest at this point and most of the methanol is likely to cross over to the cathode. Following this point, the concentration of fuel in the anode gradually drops along with the current density, both resulting in reduced

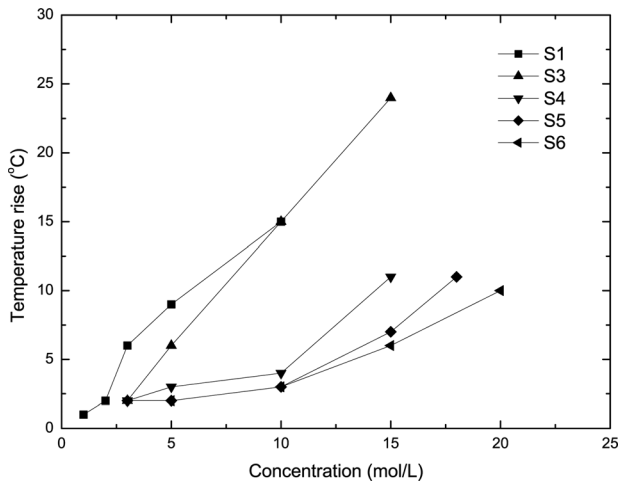


Fig. 7 Maximum temperature rise with different methanol concentrations and structures

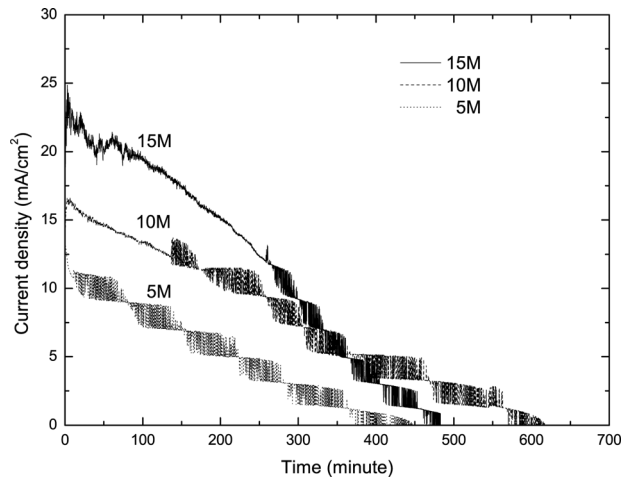


Fig. 9 Constant voltage discharge with the same structure under different methanol concentrations

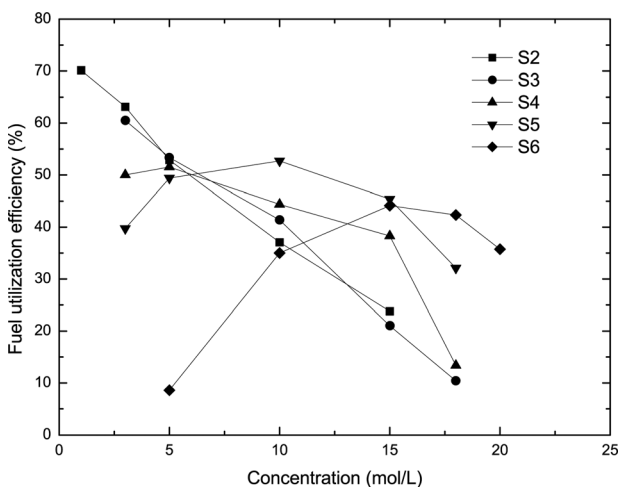


Fig. 8 Fuel efficiency versus methanol solution concentration

methanol crossover, so that the cell temperature reduces back to the ambient value. Figure 7 provides a comparison of the highest temperature rises for each cell structure. For the 10 M fuel, structure S3 shows a maximum temperature rise of 24 °C, while for structure S6 it is only 6 °C. This trend is more evident with higher methanol concentrations, which indicates that, by applying a higher mass transfer resistance, the methanol crossover can be well controlled for high concentration tubular-shaped DMFCs.

3.2 Effect of Structure on Fuel Efficiency. One of the main advantages of DMFCs over hydrogen PEM fuel cells is the high energy density of DMFCs, which is due to the fact that methanol exists as a liquid at normal pressure/temperature conditions. In real applications, this advantage is compromised due to many limiting factors with the DMFC. The theoretical energy density of a DMFC cannot be achieved because of the necessity of using dilute methanol, fuel loss due to methanol crossover, fuel leakage from the gas outlet, etc. To study the influence of the anode structure on fuel utilization, fuel efficiency tests are performed with different structures and fuel concentrations. The results are summarized in Fig. 8.

For structures S2 and S3, in which the anode does not have a high mass transport resistance, higher methanol concentrations always cause a lower fuel efficiency, which has also been reported elsewhere [18] and can be explained by a higher

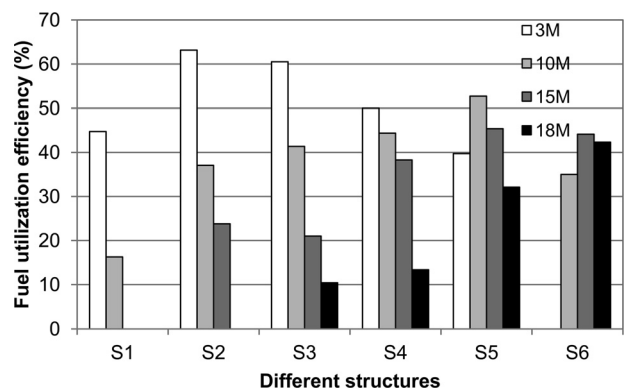


Fig. 10 Fuel efficiency versus anode structure

methanol crossover. However, when the pervaporation open ratio is reduced, the fuel efficiency for dilute methanol is not always higher than that at a high concentration, as shown by the curves for structures S5 and S6 in Fig. 8. This is surprising since it proved to be contrary to what is expected. An explanation might be that the dilute solution leads to a longer operating time because of a lower discharging current, so that fuel escapes by evaporation over a long period of time and reduces the fuel efficiency. However, this is not always true. As shown in Fig. 9, with lower concentrations, such as 5 M, the current is very low, as expected, and the discharge process is still the shortest. Combining this result with Fig. 4, we can find that, when the cell operates at a very small power density, hence, low current, the fuel efficiency is always low. This may be caused by the incomplete oxidation of methanol due to the very sluggish methanol oxidation reaction. In other words, the methanol is not completely converted to carbon dioxide in the anode catalyst layer and does not release the expected 6 protons and electrons for each molecule of methanol. Under this assumption, the fuel efficiency is very low, even when the crossover is not severe. Plotting the data of Fig. 8 in another way to emphasize the effect of the structure on fuel efficiency leads to Fig. 10. It can be seen that for structures with higher mass transport resistance, such as for structures S5 and S6, the fuel efficiency is not as sensitive to the fuel concentration. For structures S1 and S2, however, increasing the fuel concentration sharply decreases the fuel efficiency.

For this study, one of the most important goals is to emphasize the high energy density of DMFCs by utilizing higher concentration methanol solutions. Therefore, we compare the work done by

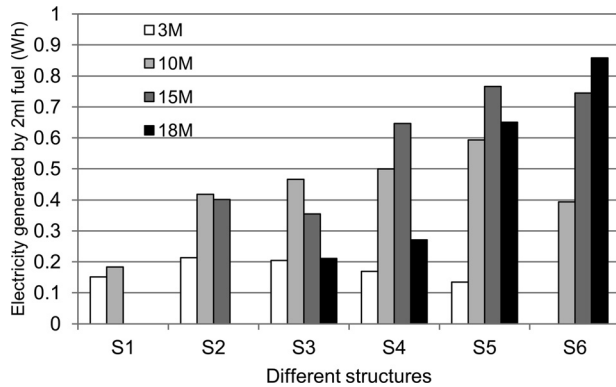


Fig. 11 Electricity generated by 2ml methanol solution at 0.35 V

the same volume (2 ml) of methanol solution under the same discharge voltage for different structures and concentrations. The result is plotted in Fig. 11. Comparing this result with Fig. 10, we can conclude that, although a lower open ratio leads to higher mass transport resistance and, subsequently, lower power density even with high concentrations of fuel, the higher fuel concentration provided a large quantity of total work produced by the fuel cell. Dilute solutions, such as 3 M, exhibit very low electricity generation, while structure S6 (3% open ratio) generates up to 0.86 Wh of electricity at 0.35 V by utilizing an 18 M methanol solution, which is much higher than the results provided by low concentration fuels. Compared with the original structure (MEA without extra layers) using the 1 M methanol solution [1], structure S4, using the 15 M methanol solution, increases the energy output of the tubular DMFC by 591%, from 0.094 Wh to 0.65 Wh, with 2 ml fuel. The power density remains at the same level as 16 mW/cm².

3.3 Future Improvement to Operate With Neat Methanol.

The focus of this work is to improve the anode structure of the DMFC. The goal is to enhance methanol transport resistance enough to prevent methanol crossover, while also allowing sufficient power generation by the fuel cell. However, methanol crossover is only part of the problem associated with DMFCs. As discussed in Sec. 3.1, water management is equally important, especially for the high concentration operation where less water is available at the anode for methanol oxidization. By providing water management layers on the cathode side to force some water back to the anode side, the methanol crossover problem can be further prohibited and the DMFC can operate stably with high concentrations of methanol.

4 Conclusions

In this paper, the anode structure of a DMFC is improved to allow the tubular-shaped DMFC to operate with high concentration methanol, up to 20 M. By adopting the passive vapor feed concept to the anode, including a methanol-impermeable PTFE membrane with different open ratios and porous PTFE membranes, the methanol transport resistance from the fuel reservoir to the anode catalyst layer can be controlled. By comparing the polarization curves and fuel efficiency of six different structures with different methanol concentrations ranging from 1 M to 20 M, the following conclusions are made:

- (1) There exists an optimum fuel concentration to achieve the highest power density for a certain cell structure. Cells with higher methanol transport resistance operate best at a higher fuel concentration. For structure S4, the maximum power density of 16.5 mW/cm² is obtained with a 15 M methanol solution.

- (2) For a specific methanol concentration, there exists a best structure to achieve the best power density. For higher concentrations, higher mass transport resistance is needed. For a 3 M methanol solution, structure S1 achieves the highest power density while, for the 15 M solution, structure S4 achieves a higher power density.
- (3) Although higher mass transport resistance may impair the power density, the fuel efficiency is higher. The higher concentration compensates for the total electricity generation for a specific amount of fuel supply. With an 18 M methanol solution, structure S6 generates the most electricity at 0.86 Wh while, for a 15 M methanol, structure S5 generates the most electricity, which is 0.75 Wh.
- (4) The aim of the paper is to improve energy output without impairing power density. Compared with the original structure (MEA without extra layers) using 1 M methanol solution, Structure S4, using the 15 M methanol solution, increases the energy output of the tubular DMFC by 591%, from 0.094 Wh to 0.65 Wh, with 2 ml fuel. The power density remains at the same level as 16 mW/cm².

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