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Short communication

A tapered serpentine flow field for the anode of micro direct methanol fuel cells

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ABSTRACT

We develop a self-breathing micro direct methanol fuel cell (μDMFC) characterized by a new anode structure with tapered single serpentine flow fields to improve cell performance. Compared with the conventional single serpentine flow field, this new design enhances the methanol mass transport efficiency and the exhaust resultant (CO_2) rate due to the increasing pressure difference between adjacent flow channels. The μDMFCs with two single serpentine flow fields are fabricated using silicon-based micro-electro-mechanical systems (MEMS) technologies and are tested at room temperature. The experimental results reveal that the new tapered single serpentine flow field exhibits a significantly higher peak power density than that of the conventional flow field, demonstrating a substantial increase of 17.9% in mass transport coefficients.

1. Introduction

In recent years, the use of portable and mobile electronic devices, such as laptops or cellular phones, has spread rapidly due to remarkable progress in technology and the information demand of our society. Therefore, the need for more compact power sources with higher power density has increased rapidly. Fuel cells employ hydrogen as fuel to generate power and meet portable requirements. They possess high efficiency and are clean and quiet. Applied electrochemical and new energy source developments have also focused on fuel cells. In particular, micro-electro-mechanical systems (MEMS) technology based on micro direct methanol fuel cells (μDMFCs) has been recognized as a leading candidate to supply power to portable electronics. Considering the potential of μDMFC to overturn the use of conventional power sources, many researchers have paid increasing attention to various aspects concerning μDMFC technology [1–4]. Above all, the anode flow field configuration is a crucial factor for μDMFC applications [5,6].

Basically, a μDMFC is composed of two current collectors (the anode and the cathode) with flow fields sandwiched around a membrane electrode assembly (MEA). In the oxidation process at the anode and deoxidization process at the cathode, the electric power is exported continually [7]. The anode current collector not only supplies a passage for the transport of reactants (methanol) and resultants (CO_2), but it also provides structural support for the weak MEA and collects current. At present, studies of anode flow fields mainly focus on parameter optimizations and new configuration designs. Yang and Zhao [8] investigated the effects of different anode flow fields and parameters on the performance of DMFCs, and the experiments indicated that the single serpentine flow fields performed better than parallel ones. Similarly, Zhao and co-workers [6] fabricated a μDMFC with an active 1.0 cm × 1.0 cm area to study the effects of flow field structures on cell performance and reached the same conclusions. Furthermore, channel depths of the serpentine flow field were optimized experimentally. Oliveira et al. [9] studied the effects of three different serpentine anode flow fields (single serpentine (SFF), multi-serpentine (MSFF) and mixed parallel and serpentine (MFF)) on the performance of DMFCs, and the experimental results showed that the use of MSFF or MFF as anode flow fields yielded a better performance. Moreover, new anode flow fields were also presented to improve the performance of DMFCs [5,7,10].

Single serpentine flow fields are generally applied in the anode configurations of μDMFCs, as shown in Fig. 1(a). Owing to the mass transport shadow region of an under-rib diffusion layer from the channel to the electrode, it results in the fall of methanol transport efficiency to reduce the cell performance. In fact, the pressure difference between adjacent flow channels of the ribs determines the convection and diffusion of the methanol molecules in the electrode. If the transport velocity of molecules increases with increment of the pressure difference, the methanol transport efficiency to the catalyst layer is improved and the resultant (CO_2) is also exhausted from the flow channels more quickly. Based

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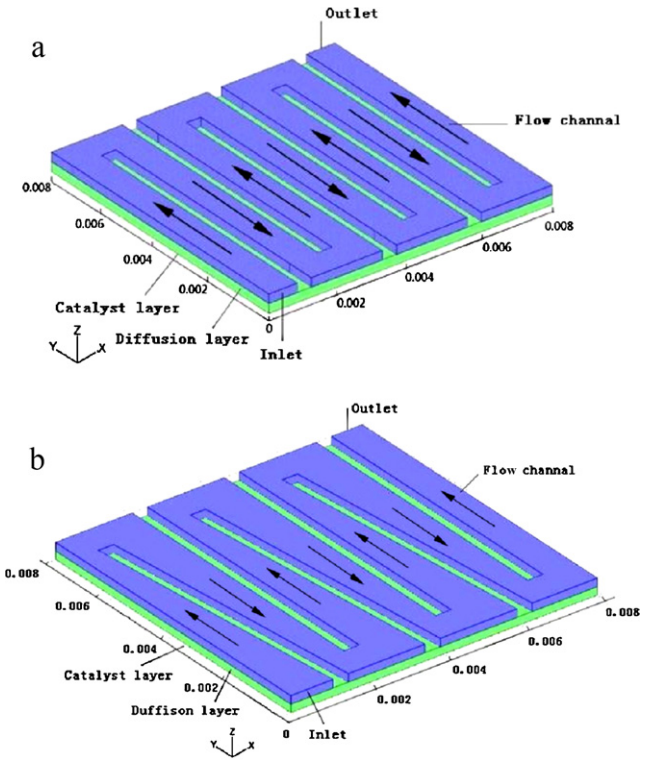

Fig. 1. Design of the anode flow fields: (a) conventional single serpentine flow fields; and (b) new tapered single serpentine flow fields.

on the above considerations, we presented a new tapered single serpentine flow field for the anode of a self-breathing μDMFC, as illustrated in Fig. 1(b). The simulation results showed that the pressure difference could be enhanced effectively with the same channel areas and channel length of conventional single serpentine flow fields. Using silicon-based MEMS technology, the self-breathing μDMFC was fabricated and tested to prove the superiority of this new flow field.

2. Experimental

The anode current collectors with conventional and new single serpentine flow patterns were fabricated using normal silicon-based MEMS technology. Two anode flow fields had the same active area (0.8 cm × 0.8 cm), the same open ratio (47.3%) and the same total length of the single channel (63.50 mm). Two $480 \pm 10\,\mu m$ silicon wafers with ⟨1 0 0⟩ crystal orientation were employed as anode current collectors, including conventional single serpentine flow fields and new tapered ones, respectively. First, a 0.8-μm-thick Si_3N_4 layer was deposited on the Si wafers using low pressure chemical vapor deposition (LPCVD). To achieve the selected etching, photolithography was applied to pattern microchannels on the Si_3N_4 layer. The serpentine channels were etched with the depth of 240 μm using an anisotropic etching process, which was performed using a 40% KOH solution at 40 °C. Considering their application for portability, the cathode current collectors with common perforated structure were employed with the same active areas. To fashion the self-breathing openings, circular holes with a radius of 0.3 mm were inserted on the silicon wafer using a laser process. To collect current and minimize contact resistance between the MEA and the silicon wafer, a Ti/Au (0.05 μm/1.0 μm) layer was sputtered on the current collectors.

A 5-layered MEA with an active area of 0.8 cm × 0.8 cm fabricated by the catalyst coated membrane (CCM) method was employed for the silicon-based μDMFCs. The hydrophilic catalyst layer was prepared utilizing the decal transfer method. First, the Nafion®117 membrane (DuPont TM, 175 μm) was ion-exchanged to its Na^+ form by boiling in a 0.5 M NaOH solution at 80 °C for 1 h and then in deionized water at 80 °C for 1 h. The confected catalyst ink (Pt–Ru/Pt black, 5 wt.% solubilized Nafion®, isopropanol, alcohol, glycerol, and deionized water) was uniformly sprayed onto the polytetrafluoroethylene (PTFE) sheets using a spray gun, with the catalyst loadings of 4.0 mg cm^{-2} (anode) and 2.0 mg cm^{-2} (cathode). The thin films were then transferred from the PTFE sheets to both sides of the membrane by hot pressing at 160 °C and 5 MPa for 90 s, thus forming the CCM. Afterward, carbon paper (TGPH-090, Toray, Inc.) was prepared with a hydrophobic (10 wt.% PTFE for the anode and 30 wt.% PTFE for the cathode) and pore-forming (NH_4HCO_3) pretreatment to form the gas diffusion layer (GDL). In the end, the 5-layered MEA was achieved with two GDLs hot pressed on both sides of the CCM at 130 °C and 4 MPa for 120 s.

Utilizing the polydimethylsiloxane (PDMS) packaging process, the MEA was sandwiched between the anode and cathode current collectors to realize a self-breathing μDMFC with an active area of 0.8 cm × 0.8 cm. Experiments using the self-breathing μDMFCs with two anode single serpentine flow patterns were performed under atmospheric pressure at room temperature (i.e., 20 °C). A peristaltic pump was used to deliver a dilute concentration of methanol (1 M) with a feeding rate of 1 mL min^{-1} at room temperature. For performance evaluation of the μDMFCs, an electronic load (N3300A&N3302A, Agilent Technologies) was employed to measure the polarization curves and power density curves.

3. Results and discussion

Consulting the three-dimensional static anode model of the DMFC built by Yang and Shi [11,12], the new tapered single serpentine flow field was characterized and simulated using COMSOL Multiphysics 3.6. From the results presented in Fig. 2, it was found that the pressure difference between the inlet and outlet pressures increased by about 47% when the conventional flow field was replaced with the new flow field. Due to the increase in pressure drop, the pressure difference between any adjacent flow channels should be improved. We then compared these results with the line pressure drops along the flow directions by randomly selecting two flow channels of single serpentine flow fields, as shown in Fig. 3. As expected, the line pressure drop with tapered single serpentine flow fields was obviously enlarged. Therefore, the methanol transport efficiency in the diffusion layer should be improved, and, simultaneously, the resultant CO_2 exhaust accelerated from the outlet. In addition, the methanol concentration distribution on the catalyst layer is illustrated in Fig. 4. For the new tapered single serpentine flow field, the methanol content on the surface could be obviously increased and distributed more uniformly. Thus, the cell performance could be enhanced, and the concentration polarization could be avoided to a certain extent. Based on the above simulations, the new tapered single serpentine flow field for a μDMFC had remarkable advantages over the conventional one.

To ensure that the mass transport limitation was caused by the transport of methanol at the anode in different serpentine flow channels only, all the tests in this work were operated synchronously using the same conditions, including the cathode structure and environment. From the measured polarization curves illustrated in Fig. 5, the open circuit voltage (521.1 mV) of the μDMFC with the tapered single serpentine flow field was a little lower than that of its conventional counterpart (535.7 mV). However, the new tapered flow field (15.4 mW cm^{-2}) yielded a substantial (35.3%) increase in peak power density over the conventional design.

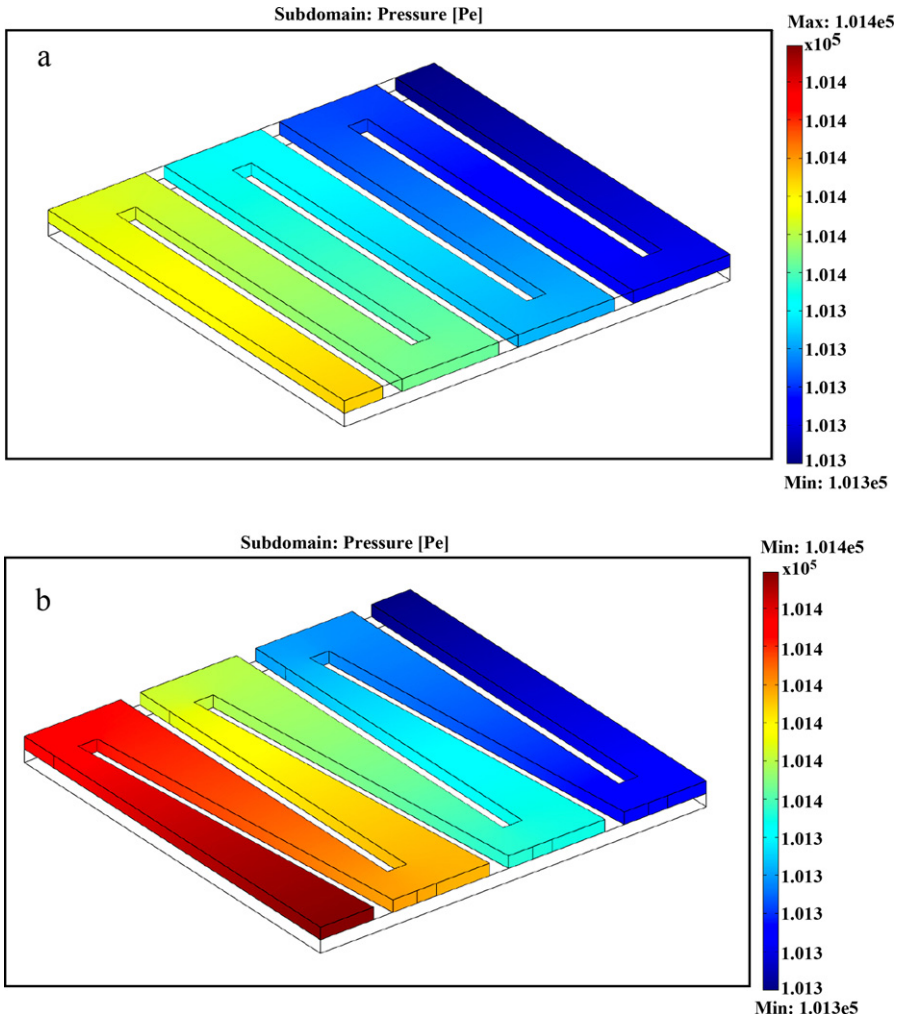

Fig. 2. Pressure distributions in two types of single serpentine flow fields: (a) conventional single serpentine flow field; and (b) new tapered single serpentine flow field.

The preceding theoretical analysis demonstrated that the mass-transport rates from the channel to the electrode could be significantly enhanced with the new flow field due to the enhanced under-rib convection as a result of the increased pressure difference between adjacent flow channels. The overall mass transport coefficient from the channel to the electrode could be calculated by measuring the limiting current density [13]:

$$k_{tot} = \frac{(i_{lim}/6F)}{C_0 - (i_{lim}A/12Fu_0)} \quad (1)$$

where C_0 represents the methanol concentration at the channel inlet (1 M), i_{lim} is the mass transport controlled limiting current density (120 mA cm^{-2} for the conventional flow field and 140 mA cm^{-2} for the new flow field), F is the Faraday constant (96,495 C mol^{-1}), u_0 is the given inlet flow rate (1 mL min^{-1}), and A is the area of the electrode (0.64 cm^2). The corresponding mass transport coefficients were calculated using Eq. (1), achieving 2.18×10^{-6} m s^{-1} for the conventional single serpentine flow field and 2.57×10^{-6} m s^{-1} for the new one. An increase of 17.9% showed that methanol mass transport rates on the diffusion layer obviously improved when the conventional flow field was replaced with the new flow field. Compared with its conventional counterpart, the new flow field had the significant advantage of enhanced mass-transport rates from the channel to the electrode. Because of the increased pressure difference between adjacent flow channels, the methanol crossover increased with the small current density, but the performance of the self-breathing μDMFC with the tapered single serpentine flow field was improved effectively as a whole.

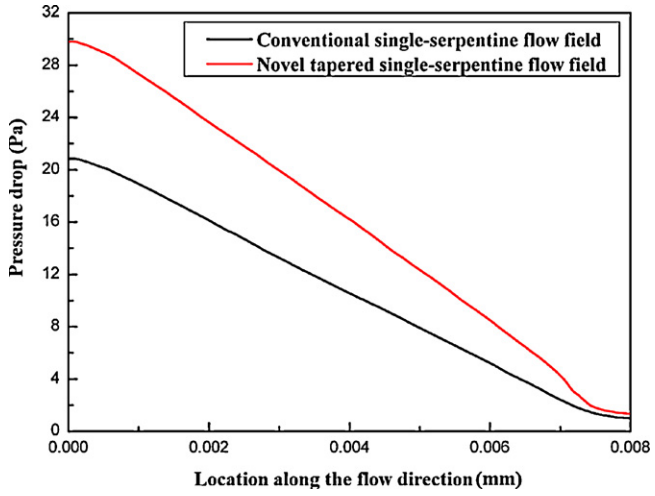

Fig. 3. Comparisons of the line pressure drop at the interfaces between the diffusion layers and two single serpentine flow fields.

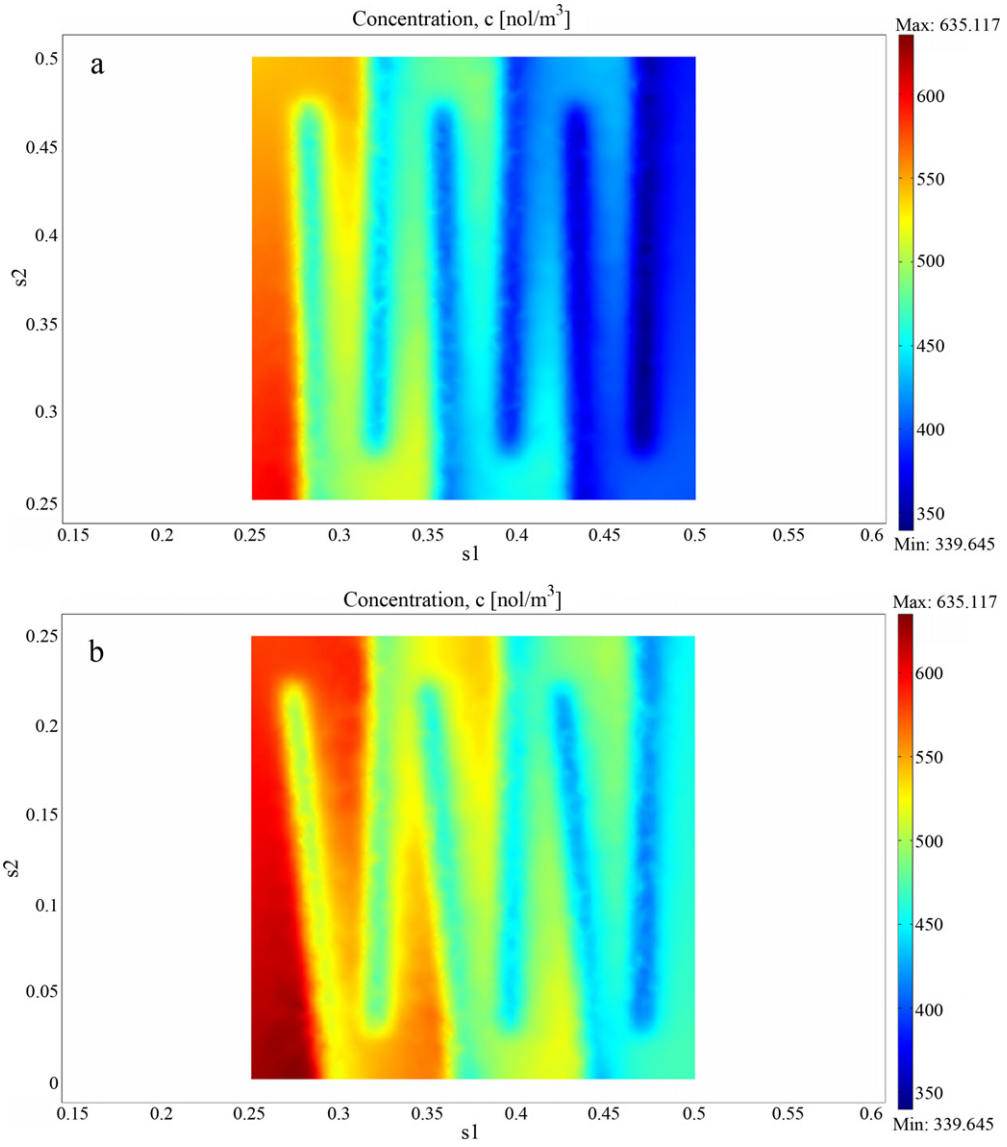

Fig. 4. Methanol concentration distributions of the catalyst surface in two types of single serpentine flow fields: (a) conventional single serpentine flow field; and (b) new tapered single serpentine flow field.

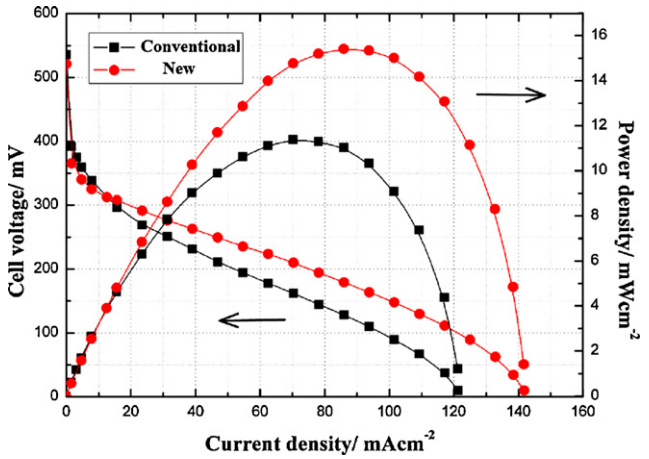

Fig. 5. Performance comparisons of self-breathing μDMFCs with conventional and new single serpentine flow fields.

4. Conclusions

In conclusion, a self-breathing μDMFC with a tapered single serpentine anode flow field was presented. Compared with its conventional counterpart, the new flow field was able to enhance the pressure difference between any of the adjacent flow channels and thus improve the mass transport efficiency and the exhaust resultant rate utilizing the simulation analysis. The self-breathing μDMFCs were fabricated using silicon-based MEMS technology. Due to the increase in methanol mass transport efficiency from the channel to the electrode, the μDMFC with the new single serpentine flow field yielded a maximum power density of 15.4 mW cm^{-2} at room temperature. From the results obtained in this work, it can be concluded that the tapered single serpentine flow field demonstrated in the μDMFC is also applicable to the greater DMFCs or the PEMFCs running on the anode.

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