A new flow field design for polymer electrolyte-based fuel cells

C. Xu, T.S. Zhao *

Department of Mechanical Engineering, The Hong Kong University of Science and Technology, Clear Water Bay, Kowloon, Hong Kong SAR, China

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Abstract

We present a new flow field design, termed convection-enhanced serpentine flow field (CESFF), for polymer electrolyte-based fuel cells, which was obtained by re-patterning conventional single serpentine flow fields. We show theoretically that the CESFF induces larger pressure differences between adjacent flow channels over the entire electrode surface than does the conventional flow field, thereby enhancing in-plane forced flow through the electrode porous layer. This characteristic increases mass transport rates of reactants and products to and from the catalyst layer and reduces the amount of liquid water that is entrapped in the porous electrode, thereby minimizing electrode flooding over the entire electrode surface. We applied this new flow field to a single direct methanol fuel cell and demonstrated experimentally that the new flow field resulted in substantial improvements in both cell performance and operating stability as opposed to the conventional serpentine flow field design.

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1. Introduction

Polymer electrolyte membrane (PEM)-based fuel cells, including hydrogen fed PEMFCs and direct liquid methanol fuel cells (DMFCs), offer the tantalizing promise of cleaner electricity with less impact on the environment than traditional energy conversion technologies. However, the commercialization of PEM fuel cells is still hindered by several technological problems, among which severe water flooding of the cathode and the induced mass transport losses are critical [1–3]. Over recent decades, different fundamental issues of the PEMFC system, including the water management of the cathode, have been studied extensively [1–31].

The flow field is one of the key components of a PEMFC, which serves as both the current collector and the reactant distributor. The reactants, as well as the products, are transported to and from the cell through the flow channels. The essential requirements for the flow field are uniform distribution of reactants over the entire electrode surface and effective removal of products from the cell, to minimize the concentration polarization. To this end, different flow field configurations, including parallel, serpentine, interdigitated, and many other combined versions, have been developed [3]. It has been understood that the flow field design has a deterministic role on mass transport and water management, and thus great efforts have been made for the optimal design of flow field such that high and stable cell performance can be achieved [3–28].

Yamada et al. [4] investigated the occurrence of water flooding in the gas diffusion layer (GDL) of a PEMFC, and their results showed that it was water flooding in the GDL that directly caused the concentration polarization. They also found that liquid water accumulated more rapidly under channel ribs with the increase in current density, because liquid water under ribs was harder to be expelled due to the longer transport route. Turhan et al. [12] studied liquid water build-up and distribution in a PEMFC using the neutron imaging method, and their results showed that liquid water build-up decreased with increasing inlet gas flow rate. They also showed that due to restricted mass
transport, there was liquid water under ribs even at very high flow rates and low humidity. Nguyen [21] designed the interdigitated flow field, with which gas was forced to flow through the GDL and the shear force of this gas flow helped blow out the liquid water that was entrapped in the inner layers of the electrodes. As a result, mass transport rates of the reactants from the flow channel to the inner catalyst layer could be improved, and the water flooding problem at the cathode could be significantly reduced. Williams et al. [24] experimentally found that convection under ribs also occurred in a serpentine flow field due to the pressure difference between two adjacent channels, and this convection improved oxygen transport and helped blow out liquid water in the GDL, reducing transport losses from flooding. Jang et al. [25] studied a baffle-blocked flow field, and found that gas convection in the GDL could be enhanced due to the baffle-blockage effect, which enhanced the reactant transport to the inner electrode and benefited water removal and flooding prevention.

Our literature review indicates that water flooding in the GDL under ribs is usually more serious than elsewhere at the cathode. The previous studies also demonstrated that channel-to-channel convection under ribs can facilitate both mass transport into the electrode and removal of water flooding. Therefore, enhancing convection through the GDL by optimizing the flow field is an effective way to reduce water flooding at the cathode, enhance the mass transport, and thus improve both cell performance and operating stability. Based on this understanding, we designed a new flow field for PEM based fuel cells, which was obtained by re-patterning conventional single serpentine flow fields. We showed theoretically that the new flow field induces enhanced gas flow through the GDL without increasing the compressor power. We applied this new flow field to an in-house fabricated DMFC and demonstrated the marked features of the new flow field over the conventional serpentine flow field design.

2. Theoretical

Consider a conventional single serpentine flow field (SFF), shown in Fig. 1a, in which one continuous channel that proceeds through a series of alternating 180° turns and 14 ribs with the same length of 29.0 mm are formed. When a fluid is pumped to flow through the channel from the inlet to the outlet, the local pressure in the channel can be approximated by:

\[ P(x) = P_{\text{inlet}} - P_{\text{loss}}(x) = P_{\text{inlet}} - \frac{128\mu Q}{\pi D_{\text{eff}}^4} x \]  

(1)

where \( Q \) is the mass flow rate of the fluid, \( D_{\text{eff}} \) is the effective dynamic diameter of the fluid flow, and \( x \) is the channel distance measured from the inlet [32]. Eq. (1) indicates that the pressure decreases with \( x \) downstream due to viscous losses, resulting in a pressure difference across each channel rib. For example, the pressure at point M \((x_M)\) is higher than that at point N \((x_N)\). It follows from Eq. (1) that the corresponding pressure difference across the rib, i.e., between points M and N, is given by:

\[ \Delta P = P(x_M) - P(x_N) = \frac{128\mu Q}{\pi D_{\text{eff}}^4} (x_N - x_M) \]  

(2)

This pressure difference tends to lead to an in-plane “short-circuit” flow through the porous GDL under ribs, referred to as under-rib convection hereafter [24,26–28], with its average artificial velocity represented by Darcy’s law:

\[ v = -\frac{K}{\mu} \nabla P = \frac{128KQ}{\pi D_{\text{eff}}^4 W} (x_N - x_M) \]  

(3)

where \( K \) is the fluid permeability through the GDL and \( W \) is the rib width. Eq. (3) indicates that for a given mass flow rate \( Q \), the strength of under-rib convection between points M and N depends upon the geometric dimensions of channel and GDL, such as the rib width, \( W \), and fluid permeability through the GDL, \( K \). It is also clear from Eq. (3) that the strength of under-rib convection is not uniform, varying linearly from zero to the maximum value according to channel length \( x \). We now show that the strength of under-rib convection near each rib head (see Fig. 1a) is nearly zero but it reaches a maximum near each rib root. We first look at the strength of under-rib convection in the channel inlet region (the upper-left corner). As illustrated in Fig. 1a,
near the rib head, the distance between points B and C ($x_C - x_B$) is rather small, and thus the pressure difference between B and C is nearly zero, i.e.:

$$\Delta P_{BC} \approx 0$$  \hspace{1cm} (4)

On the other hand, near the rib root (see points A and B in Fig. 1a), the distance between points A and B is $x_B - x_A = 2L_r$, with $L_r$ representing rib length, and thus the pressure difference near the rib root reaches the maximum value, i.e.:

$$\Delta P_{AB} = \frac{256\mu Q}{\pi D_{eff}^4} L_r$$  \hspace{1cm} (5)

We now look at the strength of under-rib convection in the channel outlet region (the lower-right corner). In a similar manner, we can show that the pressure difference near the rib head (between points D and E) is nearly zero, i.e.:

$$\Delta P_{DE} \approx 0$$  \hspace{1cm} (6)

whereas the pressure difference near the rib root (between points E and F) is

$$\Delta P_{EF} = \frac{256\mu Q}{\pi D_{eff}^4} L_r$$  \hspace{1cm} (7)

Eqs. (5) and (7) implies that for the same GDL, at the same fluid flow rate, and at the same cross sections of channel and rib, a longer rib will result in a larger pressure difference between adjacent channels, and thus stronger under-rib convection. A more detailed analysis of under-rib convection in the serpentine flow field can also be found elsewhere [28].

Based on the above understanding, we re-patterned the conventional single serpentine flow field and obtained a new flow field, as shown in Fig. 1b. The basic idea is to force the fluid to flow toward the outlet region first, then retrace to the inlet region, and finally retrace back to the outlet region again. As shown in Fig. 1b, this re-patterning results in 6 channel ribs only, as opposed to 14 ribs in the SFF, among which 4 ribs have nearly the same length as those in the SFF, whereas the other 2 serpentine ribs are about 5.5 times longer, i.e., 159 mm. We now examine the strength of under-rib convection of the new flow field with longer ribs in the inlet region and outlet region. In the inlet region (the upper-left corner of Fig. 1b), since the rib of the new flow field is about 5.5 times longer than that of the SFF, according to Eq. (5), the pressure difference near the rib root (between A' and B') is

$$\Delta P_{A'B'} = 5.5\Delta P_{AB}$$  \hspace{1cm} (8)

Similarly, in the outlet region (the lower-right corner of Fig. 1b), the pressure difference near the rib root (between E' and F') is

$$\Delta P_{E'F'} = 5.5\Delta P_{EF}$$  \hspace{1cm} (9)

Both Eqs. (8) and (9) indicate that under-rib convection in the inlet and outlet regions of the new flow field is 5.5 time stronger than that in the SFF. Therefore, for the same width and depth of both channel and rib, it is safe to conclude that on average the strength of under-rib convection over the new flow field is much stronger than in the conventional SFF. It is also of significance that, since the open ratio and the total length of the single channel are the same for the two flow fields, the pressure drop between the inlet and the outlet is nearly the same for the two flow fields, indicating that the new flow field will not cause an increase in the pumping power.

In summary, the distinctive feature of the new flow field is that the strength of channel-to-channel under-rib convection is substantially enhanced as opposed to the conventional SFF but the pumping power remains almost the same. Because of this feature, we refer to the new flow field as convection-enhanced serpentine flow field (CESFF) hereafter, and in the remainder of this paper we shall show that the application of this new flow field in a DMFC results in alleviated water flooding and enhanced mass transport in this type of PEM based fuel cells. Herein, it may be worth mentioning that the concept of under-rib convection is the primary motivation for another successful type of flow field design, called the interdigitated configuration [21].

3. Experimental

3.1. The DMFC

The in-house fabricated DMFC consisted of a MEA, with an active area of 3.0 cm × 3.0 cm, sandwiched between two bipolar plates, which were fixed by two fixture plates. The MEA consisted of a Nafion® 115 membrane and two E-TEK single-side ELAT electrodes. The carbon cloth (E-TEK, Type A) with 30% PTFE wet-proofing treatment was used as the backing support layer both in the anode and in the cathode electrodes. 4.0 mg cm$^{-2}$ with unsupported [Pt:Ru] Ox (1:1 atom%) and 2.0 mg cm$^{-2}$ using 40% Pt on Vulcan XC-72 were used as the catalyst loading on the anode and cathode side. Furthermore, 0.8 mg cm$^{-2}$ Nafion® was applied onto the catalyst layer surface of each electrode. The membrane was pretreated by a well-known standard procedure [2]. Finally, the MEA was formed by hot pressing under the condition of 135°C and 4 MPa for 3 min.

3.2. The flow field fabrication

To avoid corrosion, the flow field plates, shown in Fig. 1, were made of 316L stainless steel plates with a thickness of 1.0 mm, into which the flow fields were fabricated by wire-cut technique. To reduce the electrical resistance, the flow fields were sputtered with a thin layer of Pt (0.2 μm). Two different flow fields, a SFF and a new CESFF flow field, as shown in Fig. 1, were tested on both the anode and the cathode side. The open ratios of all the flow fields are 0.5 and both the channel width and the rib width are 1.0 mm.
3.3. The test rig

The experiments were carried out in the test rig detailed elsewhere [2]. On the anode, aqueous methanol solution was fed by a digital HPLC micro-pump (Series III). Before entering the cell, methanol solution was pre-heated to a desired temperature by a heater connected to a temperature controller. On the cathode, 99.999% high purity oxygen or synthetic air was supplied without humidification. A mass flow meter (Omega FMA-7105E), along with a multiple channel indicator (Omega FMA-5876 A), was used to control and measure the gas flow rate.

3.4. Measurement instrumentation and test conditions

In this work, the Arbin BT2000 (Arbin Instrument) electro-load interfaced with a computer was employed to control the cell operation and measure voltage–current (polarization) curves with a potentiostat discharging mode. In detail, the cell voltage was decreased from 0.65 V to 0.10 V step by step, with the duration of 1 min for each step. Unless mentioned else, all the experiments were performed at temperature of 70 °C and used the same anode flow field having the same configuration and geometry as SFF and fed with 1 M methanol solution with the flow rate of 1.0 mL min⁻¹. Such a methanol solution feeding ensured that no evident concentration polarization occurred caused by the mass transport limitation of methanol[26]. Gas flow rate of oxygen or air was varied from 20.0 standard cubic centimeters per minute (sccm) to 400.0 sccm at atmospheric pressure.

4. Results and discussion

4.1. Increased mass-transport limitations with the new flow field

We first tested the application of the new flow field to the anode of the DMFC and made comparison with the conventional flow field design. The experiments were performed with dilute methanol concentration (0.25 M) at the flow rate of 1.0 mL min⁻¹ and at 75 °C. In order to ensure that the mass transport limitation was caused by the transport of methanol at the anode only, pure oxygen was fed to the cathode flow field (SFF) at a sufficiently high flow rate. The measured polarization curves are presented in Fig. 2. Clearly, the new flow field yielded a substantial (35%) increase in the limiting current density over the conventional design. Our previous study [26] showed that the overall mass transport coefficient from the channel to the electrode could be determined by measuring the limiting current density for a given flow rate Q based on:

$$k_{\text{tot}} = \frac{i_{\text{lim}}}{C_{\text{in}}} \frac{Z_{\text{n}} A}{F Q}$$

where $C_{\text{in}}$ represents the methanol concentration at the channel inlet, $i_{\text{lim}}$ is the mass transport controlled limiting current density, $F$ is the Faraday constant, and $A$ is the area of electrode. We measured the limiting current densities for the both flow fields at different flow rates $Q$ while keeping $C_{\text{in}} = 0.25$ M and calculated the corresponding mass transport coefficients using Eq. (10). The results are presented in Fig. 3. For all the tested methanol solution flow rates, it is found that the overall mass transfer coefficients were increased by about 45% when the conventional flow field was replaced with the new flow field. These experimental results 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, as theoretically demonstrated in the preceding section.
4.2. Reduced water flooding problem with the new flow field

We now present the tested results of the new flow field application in the cathode of the DMFC and made comparison with the conventional flow field design. Fig. 4 shows the polarization curves of the DMFC with its cathode flow field consisting of either the SFF (Fig. 4a) or the CESFF (Fig. 4b). Each curve was obtained by continuously measuring the discharging current density while decreasing the cell voltage step by step from 0.65 V to 0.10 V, lasting for about 23 min. The measurements were made for different oxygen flow rates, while keeping all other operating parameters the same. It can be seen from Fig. 4a that, for the SFF oxygen flow rate had a significant effect on cell performance and operating stability. When the oxygen flow rate was very low, e.g. 20.0 sccm, the cell could not show the normal polarization behavior, i.e., generated current density became unstable and suddenly swung after stable discharging for about 8 min (at 0.475 V). During the following 14 min of discharging, although the cell voltage was further deceased step by step, the generated current density kept fluctuating seriously with much smaller values than the normal ones. Previous studies [14,16] had shown that this unstable discharging behavior was caused by severe water flooding at the cathode. When the oxygen flow rate was low, the strength of gas sweeping liquid water at the cathode became weaker, thus leading to liquid water buildup within the catalyst layer. As a result, the effective area of the electrode was reduced and the total current generated declined. Another possible reason of severe current fluctuation is the occurrence of spontaneous hydrogen evolution in flooded regions [29,30]. Methanol at the cathode that permeated from oxygen reduction and thus reduces the total current. Generally, water flooding will hinder the transport of oxygen to the electrochemical active sites, thus leading to current decline and fluctuations. When the oxygen flow rate was increased to 50.0 sccm, the flooding-induced current density fluctuation occurred later (at a higher current density) and lasted much shorter, i.e., the fluctuation occurred after stable discharging for 11 min (at 0.4 V) and lasted for about 8 min. Further increasing the oxygen gas flow rate to 100.0 sccm, only slight fluctuation occurred after stable discharging for 12 min (at 0.375 V) and lasted for less than 2 min. This is because when oxygen gas flow rate was increased, the gas flow could take away liquid water faster, thereby lowering liquid water buildup rate and mitigating water flooding. When the oxygen flow rate was further increased to 200.0 sccm, the cell polarization behavior became normal, i.e., cell current density increased smoothly with decreasing cell voltage, meaning that no severe water flooding occurred as a result of the faster water removal rate associated with the higher gas flow rate.

Let us now turn our attention to the polarization behavior of the cell with the new flow field, as shown in Fig. 4b. It is interesting to observe that the polarization curves for different oxygen flow rates that ranged from 20.0 sccm to 200.0 sccm were almost identical. The discharging current density was stable and normal at each given cell voltage even when the gas flow rate was as low as 20.0 sccm, meaning that no severe water flooding of cathode occurred with the application of the new flow field. The comparison between Fig. 4a and b clearly shows that the water flooding problem was mitigated with the flow field. This improvement is primarily associated with the unique feature of the new flow field that is discussed in the preceding section. The enhanced under-rib convection induced by the new flow field makes it easier for the liquid water to be transported out of the diffusion layer, thus resulting in more stable cell discharging behavior. The polarization curves shown in Fig. 4b suggest that with the new flow field a smaller oxygen flow rate (i.e., 20.0 sccm) would be sufficient to avoid severe water flooding of the electrode and
achieve a stable operation. However, with the conventional flow field, a much higher oxygen flow rate (i.e., 200.0 sccm) is required to mitigate the water flooding problem. This increased oxygen flow rate will increase the parasitic power of the gas compressor/blower in the fuel-cell system, thus leading to lower system efficiency.

The better water-removal ability of the new flow field can also be demonstrated in the methanol/air operation. The polarization curves of the DMFC fed with different air flow rates are shown in Fig. 5a for the conventional flow field and in Fig. 5b for the new flow field. It is seen from Fig. 5a that with the new flow field the air flow rate has to be higher than 200.0 sccm, and otherwise lower air flow rates will lead to an abnormal discharging process due to water flooding. However, with the new flow field, an air flow rate higher than about 100.0 sccm will be sufficient to achieve a stable discharging, although slight fluctuations in generated current were found for the air flow rate of 50 sccm. The comparison between Fig. 4a and Fig. 5a also shows that to achieve a stable discharging the required minimum flow rate of air (400.0 sccm) is higher than that of oxygen (200.0 sccm), because oxygen concentration of air is less than unity of pure oxygen.

The more stable cell performance resulting from the reduced water flooding problem with the new flow field can also be demonstrated by observing the constant-current discharging behavior shown in Fig. 6. It is seen from this figure that with the conventional flow field the voltage underwent periodically abrupt increase and decline, because the flow field was unable to effectively transport out liquid water from the porous electrode, resulting in periodical water buildup and subsequent removal. On the other hand, however, since the new flow field provides higher mass transport rates of reactants and products, little liquid buildup occurred in the porous electrode, thus leading to a relatively stable discharging process.

In summary, the experimental results presented above have demonstrated that as a result of the enhanced under-rib convection the new flow field has a better water handling ability than the conventional design. Hence, the minimum gas flow rate required to achieve stable and better cell performance with the new flow field is much smaller than that with the conventional design. The reduced gas flow rate will decrease the parasitic power in the fuel-cell system, thus leading to higher system efficiency.

5. Concluding remarks

The convection-enhanced serpentine flow field (CESFF) developed in this work shows superior performance over the conventional serpentine flow field (SFF), due to its enhanced mass transport capability as a result of the enhanced under-rib convection associated with the increased pressure difference between adjacent flow channels. Another striking feature of this new flow field is that the pressure drop between the inlet and the outlet remains almost the same as the conventional design, because the channel dimensions, including width, depth and total length, are the same for the both flow fields. Hence, the
enhanced mass transport capability in the new flow field design will not lead to an increase in the pumping power. The application of this new flow field to the anode of a DMFC resulted in a substantial increase in the mass-transport limitation as compared with the conventional flow field for the same flow rate of methanol solution. We also applied the flow field design to the cathode of the DMFC and demonstrated that as a result of the improved mass transport capability the new flow field has a better water handling ability than the conventional design, reflected by the fact that the minimum gas flow rate required to achieve stable and better cell performance with the new flow field is much smaller than that with the conventional design. The reduced gas flow rate will decrease the parasitic power in the fuel-cell system, thus leading to higher system efficiency. It should be recognized that the marked features of the CESFF demonstrated in the DMFC in the present work are also applicable to the PEMFCs running on hydrogen/oxygen or hydrogen/air. Finally, it is also worth mentioning that like the conventional serpentine flow field design, the present convection-enhanced serpentine flow field can also be readily extended to the formation of a flow field consisting of multiple serpентines in parallel without losing the feature of enhanced under-rib convection while resulting in a smaller pressure drop compared to the single CESFF.

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