Communication—Patterned Electrodes to Increase Water Back-Diffusion in Hydroxide Exchange Membrane Fuel Cells

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Hydroxide exchange membrane fuel cells (HEMFCs) support non-precious-metal catalysts and have demonstrated high performance, but suffer from flooding on the anode and possible drying on the cathode. We present a novel electrode patterning approach for redistributing water within the cell. Bare membrane channels provide additional pathways for water back-diffusion from anode to cathode, yielding modest performance improvements in the mass transport polarization regime.

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Experimental

High-performance MEAs were fabricated with a robotic sprayer (Sono-Tek ExactaCoat) by a catalyst-coated membrane (CCM) method as described elsewhere. Electrolytes (A201/A901 membranes and AS-4 ionomer solution) were supplied by Tokuyama Corp.; 50% Pt on high-surface-area C catalyst, by Tanaka Kikinzoku Kogyo (TKK); unwetproofed TGP-H-060 carbon paper gas diffusion layers, by Toray; and the fuel cell enclosure, by ElectroChem.

MEAs were tested with a fuel cell test system (Scribner 850e). Regardless of electrode type, the cell was always assembled with the same gaskets (window size: 5 cm²). The flow rate of the H₂ and O₂ feeds was 0.2 mL/min, the cell temperature was 60 °C, and the humidifier temperature for both feeds was 70 °C. Performance is reported on the basis of the active electrode area, excluding any channels of bare membrane.

Safety note: all tests were conducted at standard pressure. To avoid explosive crossover, exercise extreme caution when testing patterned MEAs at elevated pressure.

Results and Discussion

MEAs were fabricated with masks based on the designs in Figure 1 and tested under identical conditions. To ensure the best possible comparison, four of the five masks were applied together and sprayed simultaneously with the same batch of ink.

The results suggest a modest relationship between electrode perimeter and peak power density (PPD), a measure of performance. With a thick membrane (A201, 28 μm) no relationship is visible (Fig. 2, black circles). However, as expected, with the thinner A901 membrane (10 μm) a significant trend emerges (red triangles). Representative polarization curves are shown in Figure 3. Whereas all of the MEAs behave similarly at low current, at high current in the mass transport regime the patterned electrodes generally perform better, indicating that the water diffusion channels are effective in controlling flooding and/or drying.

Interestingly, the trend is not monotonic: increasing the degree of patterning beyond 2 x 2 hurts performance. While this trend should be verified with additional experiments, the error bars are sufficiently tight that some further speculation is warranted. The decrease in performance for the 4 x 4 patterned cell could represent gas crossover losses, an issue specific to thin membranes like A901. Just as the CLs act as a barrier to water transport, it also supplements the membrane in separating H₂ on the anode from O₂ on the cathode. Removing part of the CL in a PEMFC might have little effect on the water distribution if a thick membrane already acts as a significant barrier to transport.
of the CL is expected to increase crossover. The trouble with this explanation is that crossover losses are normally visible at open circuit, yet for these data the patterned and unpatterned MEAs behave identically until a relatively high load is applied. Further, it is unclear why redistributing a given amount of channel area should affect crossover losses; intuitively, many small channels should result in the same gas crossover as few large ones. Another possible explanation is that due to crossover losses the feed flow rate is inadequate to fully fuel the electrode area at the end of the flow path, but this seems unlikely with the relatively high flow rate employed (200 mL/min for 5 cm² electrode area).

Despite these concerns, as additional evidence of greater gas crossover for 4 × 4 vs. 2 × 2 patterning, we measured the “initial” open-circuit voltage (IOCV) immediately after connecting the

![Figure 1](image1.png)

**Figure 1.** Illustration of enhanced water diffusion in patterned MEAs. The face view shows mask designs used to prepare a series of MEAs. A photograph of an MEA fabricated with the 4 × 4 pattern from (c) is also shown. The edge view shows the anode side of the fuel cell sandwich. The gasket and gas diffusion layer (GDL) are not to scale; in reality each is about 20 times thicker than the membrane or CL. Dashed blue arrows indicate transport routes whereby water on the anode must pass through both the CL and membrane to reach the cathode side. Solid arrows indicate preferred routes that utilize water channels (white), macroscopic gaps in the CL. In (a), there are no such gaps because the electrode is too large for the gasket opening. In (c-e), channels are introduced deliberately. The total electrode area of 3.5 cm² excludes these channels. Under the hypothesis that the water channels improve performance, after normalizing to the active electrode area (i.e., the total black area contained within the dashed line in the face view), (a) should perform worst and (c) best.

![Figure 2](image2.png)

**Figure 2.** Peak power density of conventional and patterned MEAs made with thin A901 or thick A201 membranes. Error bars indicate ±1 standard error (n = 3). The electrode perimeter-to-area ratio is defined as the amount of perimeter length bordering the active electrode patches divided by the total active electrode area within the 5 cm² window. The five specific x-values correspond to five different patterns as illustrated at the top of the plot (see also Fig. 1). For the large (≥5 cm²) electrode, the perimeter-to-area ratio is zero because there is no exposed membrane in the active 5 cm² gasket window.

![Figure 3](image3.png)

**Figure 3.** Polarization of conventional and patterned MEAs made with thin A901 membranes. The data are taken representatively from the same tests as Figure 2.
Figure 4. Initial open-circuit voltage (IOCV) of patterned MEAs before testing. Error bars indicate ±1 standard error (n = 3). The data are from the same tests as Figure 2.

cell to the H₂ and O₂ feeds. This IOCV measurement differs from the normal OCV point at the beginning of a polarization scan in that the cell has not yet been conditioned. No water has been generated inside the cell and too little time has passed for the humidified feeds to wet the electrolyte. Without the swelling that results from wetting, the volume of hydrophilic regions in the membrane and CLs is lower, so hydrophobic regions occupy a larger fraction of the total volume. Thus, both because the total volume is lower and because transport of nonpolar gases is faster through hydrophobic media, conductance of H₂ and O₂ through the electrolyte should be highest just after the feeds are connected. Therefore, IOCV can be thought of as an OCV measurement that is especially sensitive to gas crossover. Indeed, under these conditions the IOCV is much lower for highly patterned A901 MEAs (Fig. 4), consistent with the hypothesis that crossover is more significant at high levels of patterning. As expected, when A201 is used instead, patterning makes no difference because the thick membrane already blocks virtually all gas crossover.

Summary

Overall, electrode patterning does appear to slightly increase cell performance as long as the membrane is thin (or has a high water diffusivity) compared to the CLs. However, at high levels of patterning the tradeoff between enhanced gas crossover and enhanced water diffusion leads to a performance optimum. At best, the electrode patterns proposed here could increase cell performance by several percent. For most applications, such a small improvement is unlikely to be practical when the water channels could instead be filled in with additional electrode area. Speculatively, electrode patterning could offer an advantage in scenarios where the gas permeability of the membrane is low, fuel efficiency matters more than capital cost, and/or the catalyst is especially expensive compared to other device components.

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