

# Fabrication and testing of a miniature H<sub>2</sub>/O<sub>2</sub> and MeOH/O<sub>2</sub> fuel cell

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## Abstract

The construction and performance of a small-scale fuel cell using traditional catalysts and membrane separators is discussed. Cell active area was 1 cm × 1 cm, with a total unit cell volume of 0.3 cm<sup>3</sup> each. Under well-hydrated conditions running under hydrogen and oxygen, these cells demonstrated over 600 mW cm<sup>-2</sup> at 80 °C. Design of flow fields and the requirements for water management showed that flow channels that were too small may have resulted in early channel blockage on the cathode, leading to lower power. Miniature cells suffered from higher than expected interfacial resistance which increased with time and this may be due either to more difficult hydration control in the membrane at smaller dimensions, or less compression in the smaller scale size leading to higher flow field/diffusion layer interfacial resistances. Running single cells in methanol to alleviate hydration issues still showed much higher interfacial resistances, but cell performance was roughly 75 mW cm<sup>-2</sup> at 80 °C in 1 M MeOH with 20 sccm of ambient pressure air, and 140 mW cm<sup>-2</sup> in 1 M MeOH with 20 sccm ambient pressure O<sub>2</sub>. Lower performance in MeOH may be attributed to swelling of the sealing gaskets in methanol, leading to increased interfacial resistances over time.

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## 1. Background and Introduction

Because power demands for portable electronics are increasing rapidly, and run times of portable batteries can become the limiting factor for many portable applications, much research has been focused in recent years on developing longer lasting, higher power density systems. Fuel cells have received attention for these applications due to the potential for fuel cells to be smaller and more efficient than the current battery chemistries that are in use. For example, a good Lithium polymer cell can have energy densities on the order of 800 Wh l<sup>-1</sup>. Methanol fuels have an energy density of ~5000 Wh l<sup>-1</sup>, requiring the fuel cell to be only 16% efficient to be competitive with existing battery systems. However, developing robust, stable fuel cells that func-

tion under the wide variety of operating conditions has proven difficult. Additionally, fuel cells, being energy conversion systems rather than energy storage systems like batteries, must be capable of continuously drawing on a fuel storage tank to continuously provide power. This volume of fuel quickly becomes the single dominant volume in the fuel cell system. Still, miniaturization of the fuel cell allows for the maximum volume to be relegated to the fuel tank.

Several workers have developed miniature fuel cell systems in the past, and continue to develop these systems for small scale applications that will run on MeOH based supply systems. Many workers have focused on using microelectromechanical systems (MEMS) technology by machining Si based devices to replace the flow plates found in larger fuel cells, while others have focused on reducing or eliminating the separator membrane. In the former class, Smyrl has published several papers based upon using a thin Si membrane that has micromachined holes through the thickness of the membrane to act as a catalyst support and gas diffusion layer. Electrocatalysts are subsequently either deposited onto these microporous membranes

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[1], or painted onto the separator directly as has been used traditionally in large-scale fuel cells [2,3]. In either case, these Si end plates are then and then sandwiched on either side of a Nafion 117 membrane. Good performance, approaching that of larger scale fuel cells, is demonstrated, but at very vigorous conditions ( $0.25 \text{ cm}^2$  of active area with  $200 \text{ ml min}^{-1}$  flow rate of MeOH), which indicates a relatively low fuel utilization efficiency within the cell. It is likely that these conditions are necessary in order to overcome the diffusion limitations through the microchannels, where stagnant, diffusion limited transport of methanol from the reservoir to the catalytic surface must occur through these small micropatterned channels, and thus high fuel supply rates are required to overcome this transport. Nevertheless, a methanol/forced air power level of roughly  $40 \text{ mW cm}^{-2}$  is demonstrated at  $0.35 \text{ V}$ , and upwards of  $60 \text{ mW cm}^{-2}$  is demonstrated at maximum power, which is close to the values reported for large cells of  $75\text{--}80 \text{ mW cm}^{-2}$  realized by the Los Alamos team [4]. Kohl et al. [5] have also used a high surface area catalyst in his work with microchannels, but has separated the catalytic layer from the ionic conductive layer for fabrication purposes. He has then replaced the lost ionic contact between catalyst and ionomer by acidifying the incoming fuel stream. While this allowed for excellent miniaturization of the system (allowing for an  $18 \text{ }\mu\text{m}$  wide catalytic area), performance from this type of cell was significantly below that reported for large scale cells, and showed only a  $0.35 \text{ V}$  open circuit response (OCV) with a maximum power delivered of  $15 \text{ }\mu\text{W cm}^{-2}$  at  $80 \text{ }^\circ\text{C}$ .

Other workers [6] have opted to use direct evaporated catalysts in an effort to make the process monolithically integrated fuel cells, but have shown drastically reduced performance as a result ( $0.78 \text{ mW cm}^{-2}$ ). The fuel supply rate on this system were quite low, only  $10 \text{ }\mu\text{l min}^{-1}$ , but the active area over which the flow was passed was also low, at  $0.018 \text{ cm}^2$  of active area. Scaled to consider the flow as a supply per square centimeter of active area, the supply of fuel here is roughly  $500 \text{ }\mu\text{l min cm}^{-2}$  of active area.

Still other attempts to miniaturize fuel cells capitalized on the concept that flow through channels at the microdimension is almost always laminar [7] to remove the ionic separator within a fuel cell and replace it with a flowing separator of acid or alkaline media to replace the ionic conduction path. Kenis [8] and Whitesides [9] have used this laminar flow separation between the anode and cathode to good effect in microchannels, where a moving channel of acid resulted in a much higher ion exchange capacity when compared to an ionomer. However, power from these cells was not as high as would be expected from the loss of the crossover losses and the increased ion exchange capacity, with power levels in the  $20\text{--}30 \text{ mW cm}^{-2}$  range.

Since the majority of the volume of a fuel cell is not contained within the electrode structures or the separator membrane, targeting these structures for miniaturization will not yield a significant loss in volume of the cell. If, however, the membrane, electrode, and gas diffusion layers of the fuel cell remain identical in structure and thickness to large-scale fuel cells, performance within these layers commensurate to larger scale fuel cells can still be reasonably expected. What remains, then, is to miniaturize the largest volume portion of the fuel cell, which would be

the flow fields, seals, and current collectors. These components are typically made of graphite or stainless steel in large cells, with graphite being preferred due to the electrochemical inertness of the material under harsh fuel cell operating conditions. We have focused on developing robust, reliable miniature fuel cell end plate hardware from graphite that is compatible with traditional membrane electrode assembly (MEA) architectures. In this work, we report on the design and fabrication of the miniature fuel cell hardware that is designed for hydrogen/oxygen performance, as well as some data on the performance of these miniature cells using methanol and oxygen.

## 2. Design and experimental

Most large-scale fuel cells use either coated stainless steel or graphite for the end plate material. These materials are selected due to their good electrical conductivity and relative electrochemical inertness. These plates are relatively thick, however, and comprise up to 95% of the volume of larger fuel cells. For large-scale fuel cells, this large thickness ensures that even compression pressure can be applied on the seals and active area of the fuel cell with little mechanical deformation of the end plates. In a miniature cell, however, mechanical deformation can no longer be ignored. Stainless steels, though more compliant than graphite, has been shown to suffer from corrosion issues under long operational times, and requires an inert coating (such as Au) in order to avoid this. This can cause problems in the conductivity of the cell, which would be exacerbated in the small fuel cell size. For these reasons, graphite end plates were chosen for this application.

For all of the experimental methods described below, only a single fuel cell was being assembled, although the technique could be applied to the building of a cell stack, as well. Thinned plates ( $0.5 \text{ mm}$  thick) of nonporous, high purity graphite were obtained from POCO, Inc. Three designs were fabricated into the plates. The first was an internally manifolded, straight channel plate with  $250 \text{ }\mu\text{m}$  wide channels separated by  $250 \text{ }\mu\text{m}$  wide ribs. These ribs were recessed into the plate by  $100 \text{ }\mu\text{m}$ , to allow space for a traditional  $175 \text{ }\mu\text{m}$  thick gas diffusion layer. In the second design, the channel and rib widths were widened to  $1 \text{ mm}$  each, and the center recess to accommodate the gas diffusion layer was removed, with the GDL accommodated by gaskets. In the final design, a single channel serpentine design with  $1 \text{ mm}$  wide channel on  $0.5 \text{ mm}$  wide ribs was fabricated and matched to the same basic header as was used in the previous two designs. An optical image of each of these designs is shown in Fig. 1.

Machining of the flow field into these plates was accomplished by ultrasonic milling at an offsite facility (Bullen Ultrasonics, OH). To fabricate these plates, a negative master mold of the pattern to be machined into the plates is made through plunge wire electric discharge milling (EDM). This negative master is then mounted to an ultrasonic horn; a slurry of  $0.3 \text{ }\mu\text{m}$  alumina slurry is passed over the graphite plate, and the negative master is pressed, under ultrasonic vibration, against the graphite. The vibration causes the alumina slurry to grind away the graphite, resulting in a positive pattern being etched into the graphite plate by the ultrasonic grinding action. This process

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