

Short communication

Design and testing of a passive planar three-cell DMFC

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Abstract

A passive (air breathing) planar three-cell direct methanol fuel cell was designed, fabricated and tested. The design concepts for plates, current collectors, seals and membrane electrode assemblies are discussed in this paper. Testing of single cells and stacks with parallel and serial connections were carried out. The results show that a reliable areal power density of 8.6 mW cm^{-2} can be achieved at ambient temperature with passive operation. Stacks with a serial connection of the single cells gave significantly higher performance than a parallel connection. It was also identified that high electrical resistance was the dominant factor in reducing performance. The major causes of the high resistance were the stainless steel hardware used and poor contact between the electrode and current collector. Future work will involve material and design studies of current collectors and seals to minimize the electrical resistance, and architectural design studies to effectively utilize the heat generated to raise operation temperature. © 2006 Elsevier B.V. All rights reserved.

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1. Introduction

Within the last 10 years, there has been dramatically increased interest in direct liquid fuel cells (DFCs) as potential power sources for portable devices. Among all types of DFC, the direct methanol fuel cell (DMFC) has been attracting the majority of research and development efforts, because methanol shows both good electrochemical activity and a high energy density compared with other liquid fuel candidates [1]. Additionally, methanol can be economically generated from a number of different sources such as natural gas, coal, or biomass.

From an operational point of view, passive air-breathing DMFCs can be designed to operate at ambient temperature without a fuel pump and other ancillary devices. The fuel supply relies on the diffusion from an accessible fuel reservoir, while the oxidant is supplied from the ambient air. This arrangement results in the elimination of parasitic power loss from ancillary devices, much simpler structures, and more compact system

designs than active DMFCs. These features can potentially result in higher reliability, lower cost, higher fuel utilization, and higher energy density, which are favourable attributes for power sources of future electronic devices.

Significant work has been reported on passive air-breathing DMFCs, by organizations across the world, on important issues such as fuel concentration, membrane thickness, membrane electrode assembly (MEA) and fabrication [2–6]. However, this work has mostly been based on single cell architectures.

In practice, fuel cell power systems require a stack of multiple cells connected in series to achieve a useful output voltage. In the passive air-breathing operating mode, a DMFC stack can hardly utilize a conventional “bipolar plate” structure. Special stack structure is required for passive diffusion of the liquid fuel and to provide access to ambient air.

Yoshitake et al. reported a DMFC pack with eight passive cells connected in series [7]. This DMFC pack was able to work with a super capacitor to power a cell phone. However, details of the MEA and stack structural design were not released. Some other DMFC stack designs have adopted structures that consist of planar mono-polar cells [8–10]. Even though some impressive performances have been reported, design and fabrication details are still absent in the literature.

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This work studied the design, fabrication, and performance evaluation of a planar multi-cell DMFC stack, as a first stage in the development of power sources for portable electronic applications. The stack was designed to be a passive system that operates at room temperature, without a fuel pump or an air blower. For future development, this design will also allow the stack to be both flexible in form factor and capable of incorporating novel architectures and materials.

2. Cell design

With passive supply of both reactants, the cell design requirements revolved around four main sub-categories: plates, current collectors, gaskets, and the MEA. The following sections describe the design of the various components in further detail.

2.1. Plates

To maintain design flexibility, polycarbonate was chosen for the plate material. Rigidity is typically an important factor in plate design for fuel cells, but a main goal of this work was to produce a flexible design. Flexible design here refers to the flexibility to use a broad range of materials, and to form the fuel cell into non-planar shapes. Polycarbonate provided the potential to conform to many curved shapes and was attractive because plastic components are prevalent in electronics fabrication. With the use of polycarbonate plates, separate electrically conductive current collectors were required. An additional requirement for the current collector was ease of formation into non-planar shapes. Stainless steel mesh was chosen, since it fulfilled the requirements and was readily available. Conformation to non-planar shapes was also required for the gasket and MEA. While gaskets are typically flexible, this requirement for the MEA meant that a final design should incorporate a flexible carbon cloth gas diffusion layer (GDL) rather than a stiff carbon paper.

The plates fulfilled three tasks: providing sufficient contact pressure to the electrode, compression of the gaskets and supply of the reactants. Grooves were incorporated in the plates to house the gasket (described in detail in Section 2.2). Twelve bolts along the perimeter of the cells provided the contact pressure and gasket compression. Aside from compression, the plates are also needed to supply reactants to the MEA. Recesses were cut into the plates to support the mesh along the edges, while keeping the top of the mesh flush with the plate surface. Inserts were used to support the mesh along the middle. A cross-shaped insert was used at the cathode, while a single rectangular support was used on the anode in order to allow convective flow of methanol (or forced flow if desired for testing purposes). Grooves were machined on the inserts to maintain good accessibility of reactants. Fig. 1 shows details of the mesh support features.

For the cathode, the ambient air can be accessed through the open windows with the mesh current collector. For the anode, the design was intended to contain the methanol in a reservoir pocket and allow convective flow, while also maintaining the

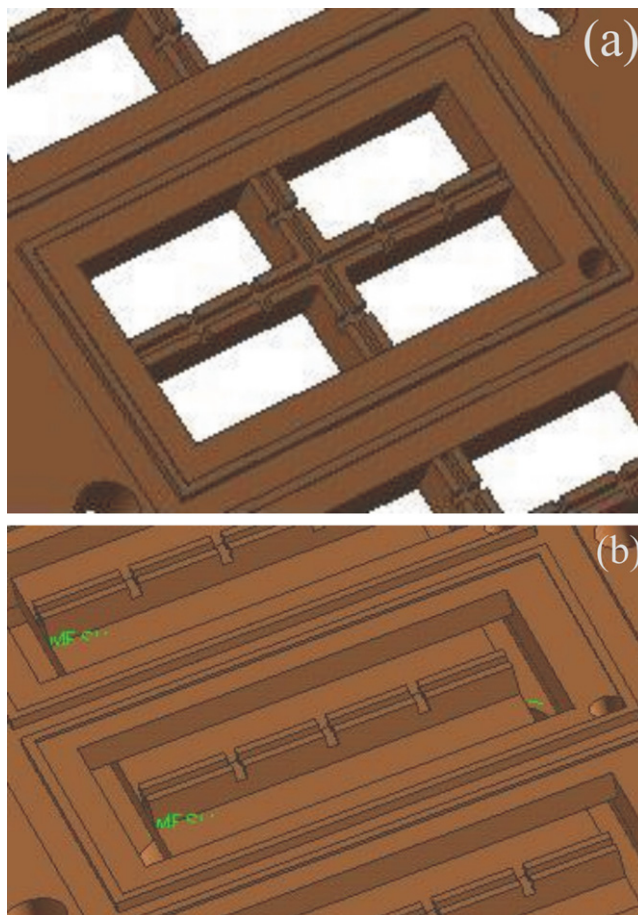


Fig. 1. Mesh support details for the plates: (a) cathode and (b) anode.

option for forced flow. For the purpose of forced flow, the design (illustrated in Fig. 2a) included a single inlet and a single outlet at the bottom and top, respectively, of the middle cell. The inlet led to a manifold channel which was the same length as all three cells, which in turn led to the separate pockets (of the same size as the active area) for each cell. These pockets then led to an upper manifold channel and the outlet, with the same dimensions as the lower channel and inlet. For the passive supply system, Swagelok fittings and Teflon tubing were used to connect the inlet to the outlet to form a closed system. The completed plates are shown in Figs. 2b and 3.

2.2. Current collectors

In the passive air-breathing fuel cell design, 304 stainless steel mesh current collectors from InterNet Inc. were used. The mesh used in this design had a thickness of 0.012 in. (0.30 mm), a strand width of 0.010 in. (0.25 mm), and 55% open area. It was desirable to be able to test the DMFC in different electrical cell configurations (single cell, multiple cells connected in series or in parallel). To attain this desired flexibility, a stainless steel threaded rod was attached to each mesh current collector on the anode and cathode side to allow for external electrical connection. A photo of a completed current collector is shown in Fig. 4.

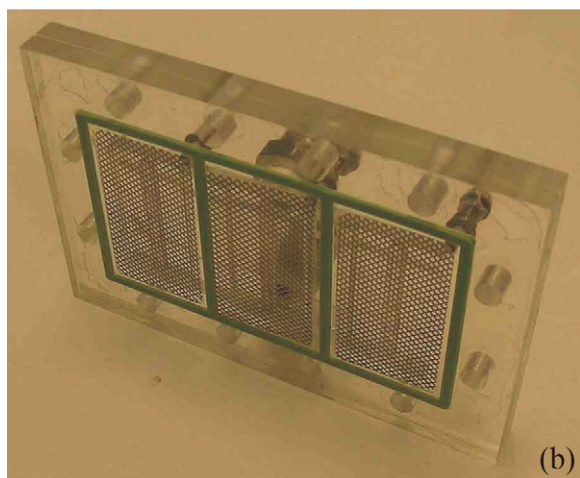
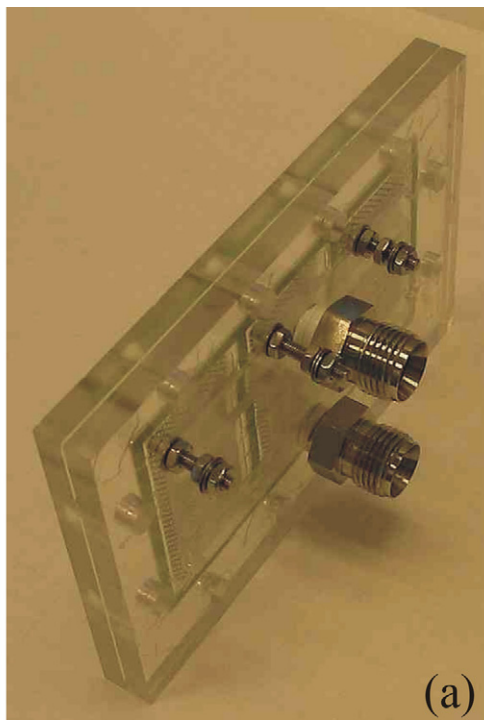


Fig. 2. Anode plate: (a) inlet and outlet connections and (b) completed anode plate.

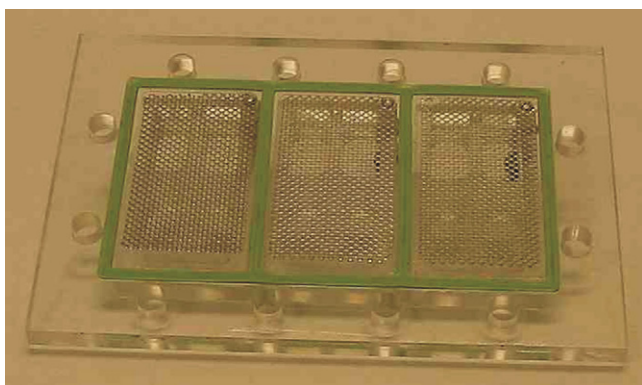


Fig. 3. Completed cathode plate.

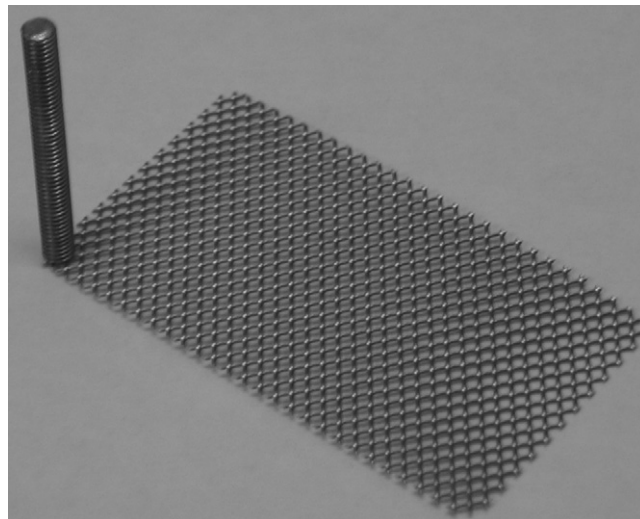


Fig. 4. Completed mesh current collector.

2.3. Gaskets

Due to the properties of polycarbonate, the seal design was based on the assumption that compressibility of the seal was one of the most important parameters. The allowable bolt torque was lower with this polycarbonate design (5–7 in-lbs were applied) than typically used in fuel cells. It was also determined that, due to the low rigidity of the polycarbonate, evenly distributed contact pressure would be difficult to achieve.

Cured SILASTIC J (Dow Corning) was used as the seal material, which can be designed to any desired dimensions by using a machined mold. In this design, 8–9% Dow Corning 200[®] Fluid was added to the SILASTIC J to lower the durometer of the cured seal to close to 50 A (similar to that of EPDM rubber).

When designing the gaskets, it was important to account for any variations of thickness within the MEA to ensure both proper sealing and good contact pressure. Fig. 5 shows compressed half views of two example MEA-gasket configurations (not to scale). The first step in the gasket design was to determine the size of the groove in the plate. For simplicity, the width and depth of the groove were chosen to be 3 mm and 1 mm, respectively. The second step was to determine the required compressed gasket thickness (CGT, refer to Fig. 5). Previously, 0.006 in. (0.15 mm) of compression has been used for our standard PEM fuel cell designs. With the lower allowable bolt torque in the polycarbonate DMFC, it was found that the gaskets with 0.004 in. (0.10 mm) of compression provided superior contact pressure without a reduction in sealing. Since the compressed and uncompressed cross-sectional areas should be equal and the width and height of the compressed cross-section and the height of the uncompressed cross-section were known, the uncompressed width could be calculated.

2.4. Membrane electrode assembly

Commercial electrodes from E-TEK were selected to make the MEAs used in this study. The E-TEK A-11 electrodes are

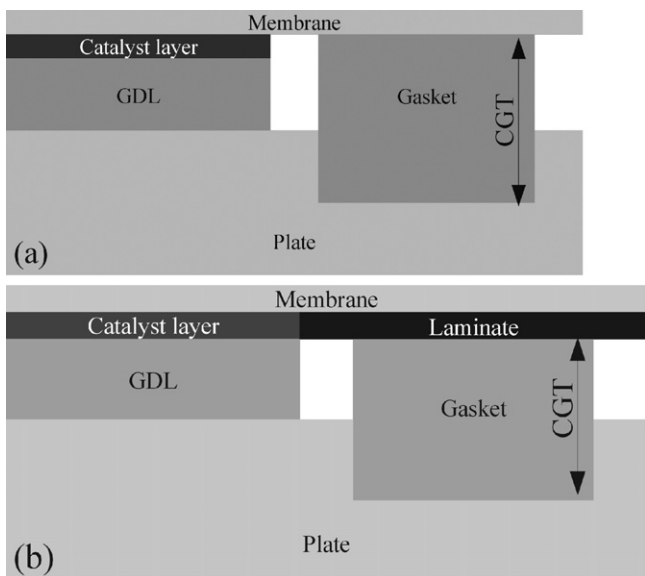


Fig. 5. MEA-gasket configurations: (a) conventional MEA and (b) laminated MEA.

optimized individually for the anode and cathode for use in direct methanol fuel cell applications. The anode utilizes a plain weave carbon cloth with a weight of 116 g m^{-2} and a thickness of 0.35 mm . The catalyst loading is 4 mg cm^{-2} and consists of an 80% Pt:Ru alloy on optimized carbon. A satin weave carbon cloth with a weight of 289 g m^{-2} and a 0.65 mm thickness is used at the cathode. Unsupported Pt black is used for the catalyst layer, with a 4 mg cm^{-2} loading. The electrodes were hot-pressed and bonded to the Nafion[®] 117 membrane at $125 \text{ }^\circ\text{C}$, under 100 atm pressure for 5 min . In order to give the membrane some extra support, the part that is not covered by the electrodes on the membrane was laminated with Mylar. The completed MEA, including the Mylar laminate for membrane support is shown in Fig. 6 and the fully assembled fuel cell is shown in Fig. 7.

3. Testing results and discussion

Since the planar DMFC was not designed with temperature control capability, the activation (MEA conditioning) was con-

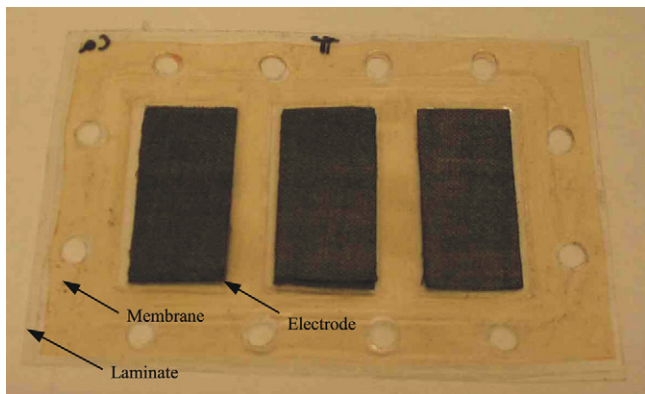


Fig. 6. Completed membrane electrode assembly.

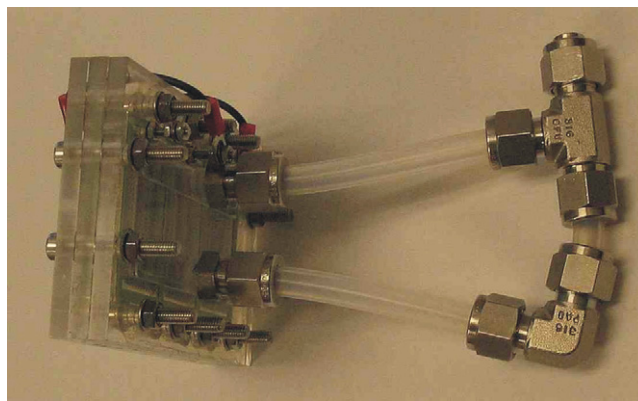


Fig. 7. Fully assembled planar DMFC.

ducted at ambient temperature. However, the procedure was followed as closely to the E-TEK guidelines as possible and the cell was run for several hours at $0.2\text{--}0.3 \text{ V}$. Additionally, before any tests, the cell was run with methanol alternatively under a constant medium load ($0.2\text{--}0.3 \text{ V}$) and at open circuit over 30 min . Polarization curves were obtained by using a 3710 A dc electronic load system from Circuit Specialist Inc. The following sections describe the testing results from different design configurations and test conditions.

3.1. Influence of seal design

Fig. 8 shows a comparison of performance for the first seal design and an improved redesigned seal. The first seal design was based on the thickness of the electrode as received and the thickness of the Nafion[®] 117.

In general, the performance was low given the high activity of commercial DMFC electrodes. The likely reason for the low performance was considered to be the effect of poor electrical contact between the current collectors and the electrodes, in which the seal design may play an important role. Since the first seal design was calculated according to the thicknesses measured prior to hot pressing the electrodes to the Nafion[®] 117. The

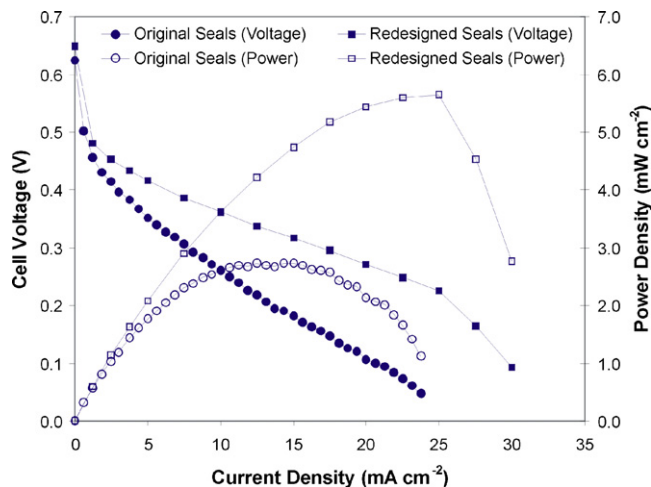


Fig. 8. Performance of single end cell with different seal designs and a fuel concentration of 1 M methanol.

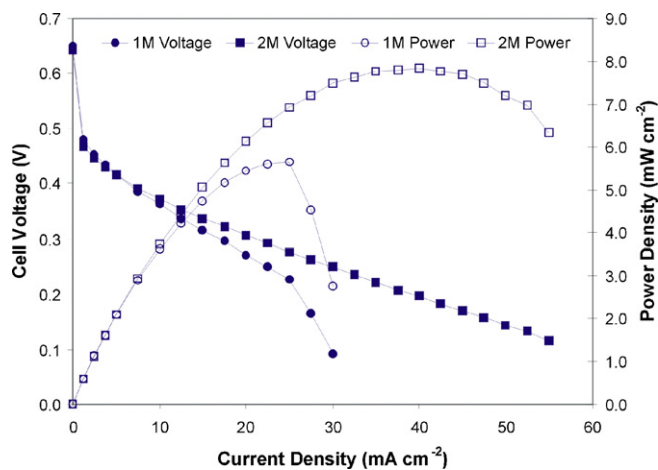


Fig. 9. Performance of single end cell with redesigned seals at 1 M and 2 M methanol concentrations.

combined thickness of the electrodes, as received, was between 0.25–0.35 mm thicker than the thickness after hot pressing. Pressure sensitive paper was used to verify that there was poor contact between the electrodes and mesh.

After replacing the seals with thinner seals, the contact and cell performance was significantly improved as shown in Fig. 8. Due to the different carbon cloths used for the electrodes, a 1.29 mm thick seal was used at the anode and a 1.22 mm thick seal was used at the cathode.

Through the use of thinner seals, the maximum power density achieved with the E-TEK electrodes in single cell was 5.7 mW cm^{-2} , which corresponded to a 108% increase over the previous cell performance. However, one notable feature of the testing at 1 M methanol concentrations with the redesigned seal was that the performance decrease was no longer linear for the entirety of the testing range. In this case the limiting current appeared to be mass transport controlled and not ohmically controlled as with the original seals. Additionally, during testing it was not possible to obtain stable performance at maximum load currents (voltage slowly but consistently decreased) again indicating mass transport limitations likely due to the diffusion rate of methanol to the catalyst layer.

3.2. Influence of testing conditions

Fig. 9 shows a comparison of 1–2 M methanol for the end cell with the redesigned seal. By using 2 M methanol, maximum power density of the specific single cell was increased from 5.7 mW cm^{-2} to 8.0 mW cm^{-2} , which correlates with the literature data [11]. Previously, single cell testing had always been performed on the same cell, which was located on one of the ends. In addition to the higher methanol concentration, more tests were conducted to investigate the performance variations for different single cells and for cells in series configurations. As shown in Fig. 10, performance in the parallel configuration was always significantly lower than the best single cell or the serial configuration. Further investigation showed that the performance was significantly lower than the estimated parallel performance based on the combined single cell performances of the three indi-

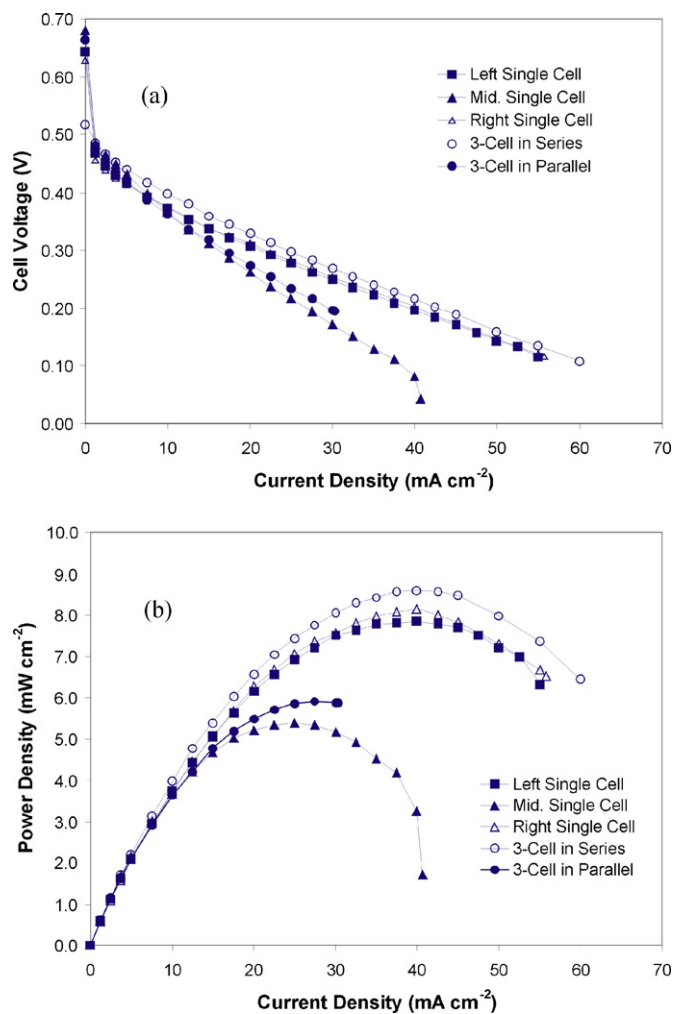


Fig. 10. Performance of different configurations with a fuel concentration of 2 M methanol: (a) polarization curves and (b) power densities. The cell voltage presented for the three-cell in series configuration is the average cell voltage measured (total cell voltage divided by three).

vidual cells. Higher ohmic losses resulting from the high current associated with parallel operation, common to fuel cells [12], were the likely cause of the low performance observed. The electrical resistances at the input and output connections added up to $65 \text{ m}\Omega$. At all but the lowest currents, the average error between the estimated parallel performance (accounting for increased ohmic losses due to the higher currents in the parallel configuration) and actual parallel performance was less than 12%.

In Fig. 10 the results for each individual cell are presented and it is shown that both end cells have similar performance, while the middle cell has poorer performance. The poor performance of the middle cell is largely due to the deformation of the polycarbonate plates resulting in reduced contact pressure on the current collector. This phenomenon was also observed and proven by contact pressure tests using pressure sensitive paper. Distinct marks from the mesh were clearly visible at the edges of the end cells and vague marks could be seen in portions of the centers of the end cells. In the middle cell, however, no marks were present in the middle and few vague marks were visible at the edges.

Cells connected in series showed a marginally better performance than any single cell. The maximum power density achieved in the series configuration was 8.6 mW cm^{-2} , compared with the best single cell (right end) performance of 8.2 mW cm^{-2} . A possible explanation for this is the increased heating of the methanol in the serial configuration (higher power output) resulting in a higher cell temperature. Only one of the cells was active when tested in the single cell configuration, while all three cells were active in the serial configuration. Meanwhile, the methanol reservoir was the same size for all tests. The heat generated by three cells is higher than one cell and it is well documented that the performance of a DMFC increases with operating temperature.

It was not possible to accurately assess the performance of the serial configuration in the design presented. While significantly higher areal power densities for passive air-breathing stacks at ambient temperature have been presented in the literature, they did not use commercially available membranes. Previous work using commercial E-TEK electrodes with Nafion[®] 117 at 25°C in a single 5 cm^2 DMFC (5 M methanol, supplied at 2 mL min^{-1} and 1 bar, oxygen supplied at 1 bar 20 mL min^{-1}) produced a maximum power density of 13 mW cm^{-2} [13]. Future designs will incorporate design features that will allow accurate comparisons with data available in the literature.

4. Conclusions and future work

A passive planar three-cell direct methanol fuel cell was designed, fabricated and tested. The results show that a power density of 8.6 mW cm^{-2} can be achieved at ambient temperature and passive operation. Stacks with a parallel connection of the

single cells showed significantly lower performance than when in a serial configuration. It was also identified that high electrical resistance was the dominant factor in the low performance as a result of the stainless steel hardware and poor contact between the electrodes and current collector. Future work will involve material and design studies of plates, current collectors and seals to minimize the electrical resistance and increase flexibility in form factor and stack architecture design studies to effectively apply the heat generated to raise the cell operation temperature.

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