

Development of High Performance Micro DMFCs and a DMFC Stack

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A silicon-based micro direct methanol fuel cell (μ DMFC) for portable applications has been fabricated and its electrochemical characterization carried out. A membrane-electrode assembly (MEA) was specially fabricated to mitigate methanol crossover. The cell with active area of 1.625 cm² demonstrated a maximum power density of 50 mW/cm² at 60 °C. Since the silicon wafer is too fragile to compress for sealing, and a thicker layer of gold has to be coated on the silicon wafer to reduce contact resistance, further development of micro DMFCs for high power application was carried out using stainless steel as bipolar plate in which flow channels were fabricated by photochemical etching technology. The maximum power density of the micro DMFC reaches 62.5 mW/cm² at 40 °C and 100 mW/cm² at 60 °C with atmospheric pressure. An 8-cell air-breathing DMFC stack has been developed. Mass transport phenomena such as water transport and oxygen transport were investigated. By using a water management technique, cathode flooding was avoided in our air-breathing DMFC stack. Furthermore, it was found that oxygen transport in the air-breathing cathode is still very efficient. The DMFC stack produced a maximum output power of 1.33 W at 2.21 V at room temperature, corresponding to a power density of 33.3 mW/cm². A passive DMFC using pure methanol was demonstrated with steady-state output power of 20–25 mW/cm² over more than 10 h without heat management. [DOI: 10.1115/1.2173668]

Introduction

Compared to hydrogen in proton exchange membrane fuel cells (PEMFCs), methanol is easier to refill and store for direct methanol fuel cells (DMFCs). Due to high theoretical energy density, DMFCs are promising power sources for portable applications such as cell phones, laptops, camcorders, and micropower sources for future integrated micro-systems that enable sensing, computing, actuation, control, and communication on a single chip [1–5].

In order to further improve the performance of DMFCs to compete with lithium-ion batteries, several challenges for DMFC development have to be addressed, such as sluggish anode catalyst activity, methanol crossover, water management, heat management, gas management, and system minimization [6]. So far, Pt-Ru binary catalysts have been extensively used, while the search for more active catalysts for methanol oxidation is still undergoing [4]. Methanol crossover produces mixed overpotential in the cathode thus decreases the cell voltage. In addition, methanol fuel efficiency will decrease because of the methanol crossover effect. Proper increase of total mass transport resistance through the anodic backing material and microporous layer to reduce methanol mass transport from anode to cathode is an alternative method besides using thick membrane to alleviate methanol crossover effect [7,8]. Water management is also an important issue in DMFCs. Due to the electro-osmotic effect, a large amount of water transports from the anode to the cathode. Consequently, water needs to be replenished in the anode. On the other hand, a large amount of water has to be removed efficiently in the cathode to prevent electrode flooding. A new idea for water management is to recycle water from the cathode to the anode directly through the membrane [9–11]. Heat management is an emerging issue in DMFCs. Consider a total efficiency of around 20–25%, a

DMFC system can produce 60–80 W of heat if we design a 20 W DMFC stack [6]. Removal or utilization of this amount of heat calls for innovative heat management technology. Gas management is another challenge in DMFCs. In liquid-feed DMFCs, carbon dioxide bubbles have to be removed efficiently to refresh the surface of the gas diffusion layer for methanol mass transport [8]. Proper gas management should maintain adequate methanol mass transport to the anode catalyst layer for the electrochemical reaction. System minimization is another challenge in DMFCs design.

Micro DMFCs are excellent power sources applied to micro systems. Based on MEMS technology, fabrication of micro DMFCs and electrochemical characterizations of performance have been extensively carried out [5,12–16].

In this paper, we give a review of the recent progress on micro DMFC development in our group. Two methods of fabricating a membrane electrode assembly (MEA) are introduced. A Si-based micro DMFC was fabricated, and its performance was characterized. Further performance improvement of μ DMFC was made by using stainless steel as a bipolar plate and flow channels were fabricated by photochemical etching technology. To reduce the parasitic power for air circulation in the cathode, an air breathing DMFC was developed. Severe cathode flooding in the air-breathing DMFC was observed, indicating that water management is a critical issue. An 8-cell air breathing DMFC stack was investigated. Based on the new water management technology, cathode flooding is fully avoided. A passive DMFC using pure methanol was demonstrated with acceptable performance.

Experiment

Two kinds of MEA fabrication methods were used in our study. The first one is to directly coat catalyst layer on the backing material such as carbon paper, then sandwich a thin membrane such as Nafion 112 between two catalyst-coated electrodes and hot-press to a MEA [7,8]. In order to alleviate methanol crossover using a thin membrane, a thick carbon paper and a compact microporous layer were used in the anode to increase methanol mass transport resistance. The advantages of using thin membrane in-

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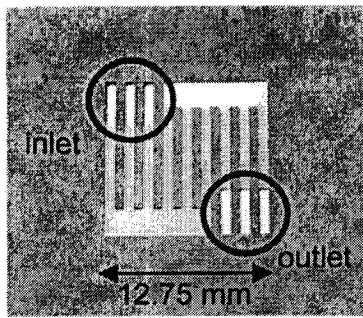


Fig. 1 Picture of Si-plate with flow channels

clude low Ohmic resistance [8] and convenience for water management [11]. The other method of making MEA is the catalyst-coated-membrane (CCM) method [11], usually, a carbon cloth coated with a microporous layer was used as GDL in the cathode. Generally, a MEA using carbon cloth in the cathode has less oxygen transport resistance, which is required in air-breathing DMFCs. The CCM MEAs using carbon cloth as a backing layer in the cathode have been used for the air-breathing cells in this paper. The normal metal loading of catalyst layers is about 4–5 mg/cm² in the anode and around 1 mg/cm² in the cathode for both MEA fabrication methods.

A 2 M methanol solution was used to test the cell performance. An electronic load system (BT4, Arbin) in a galvanodynamic polarization mode was used to measure the polarization curve at a scan rate of 3 mA/s. In addition, a multichannel electronic load system (Arbin) was used to measure polarization curves for the DMFC stack.

Development of Micro DMFCs

A silicon based micro DMFC was developed [5]. The flow channels were microfabricated by MEMS technology on the silicon wafer. Both the flow channel and the rib separating two neighboring channels were 750 μm wide, with a channel length of 12.75 mm, as shown in Fig. 1. There were a total of nine channels with serpentine flowfield, forming a cell with an effective area of approximately 1.625 cm². Figure 2 shows the performance of the Si-based micro DMFC. At room temperature, the maximum power density is only about 12 mW/cm². At 60 °C, the peak power density reaches 50 mW/cm². More details on operation conditions were given in the published paper [5].

A problem emerging in micro fuel cells based on Si wafer is that the Si substrate is quite fragile, making it difficult to compress

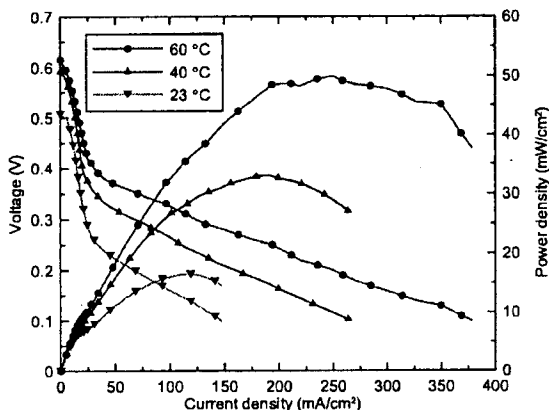


Fig. 2 Performance of Si-based micro DMFC

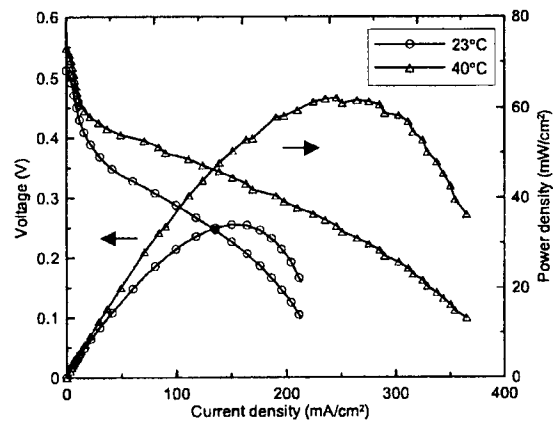


Fig. 3 Performance of SS-based micro DMFC at room temperature and 40 °C

the fuel cell tightly for good seals and for lowering the contact resistance between MEA and Si-based bipolar plates. Pavio et al. instead explored low-temperature co-fired ceramic (LTCC) material as an alternative for the bipolar plate of micro fuel cell systems, and a DMFC prototype, packaged using LTCC, was reported [17]. However, both silicon wafer and ceramic are nearly electrically insulated. Conductivity for current collection fully depends on the thickness of the conductive layer coated on these substrates, which will increase the cost significantly in order to minimize the resistance required in high power application.

An alternative method used to fabricate the bipolar plate is photochemical etching of thin stainless steel plates (500 μm or thinner) for μDMFCs instead of silicon wafer and ceramic [18]. Stainless steel has much higher conductivity and mechanical strength. A thin layer of gold coating on stainless steel plate is sufficient to prevent corrosion and improve the electrical contact. Photochemical etching is a high-quality, fast-turnaround, low-cost method for machining flat metal parts. For microfabrication of flow channels in the stainless steel bipolar plate, this offers precision and accuracy unavailable in other milling processes.

Stainless steel plates with a thickness of 500 μm have been used as bipolar plates for current collection and flow distribution. The flow channels were photochemically etched in the stainless steel plates, and the flow pattern was same as the Si-based μDMFC [5]. In order to minimize contact resistance between the MEA and the substrate, a gold layer with a thickness of 0.5 μm was deposited on the front side of each stainless steel plate. Figure 3 shows the performance of SS-based micro DMFC. The peak power is 34 mW/cm² at room temperature and 62.5 mW/cm² at 40 °C with the air flow rate of 161 mL/min. Figure 4 shows the performance at 60 °C. At large air flow rate, the maximum power density is as high as 100 mW/cm² at atmospheric pressure.

Development of an Air-Breathing DMFC

In order to reduce the parasitic power to drive the air fan in the cathode, an air-breathing DMFC is of interest. In the air-breathing DMFC, the cathode is directly exposed to the atmosphere. To minimize mass transport resistance, carbon cloth was used as gas diffusion layer due to its large porosity and gas permeability. A 5 cm² stainless steel air-breathing DMFC was developed with parallel flow channels.

Unlike the large flow rate used in the micro DMFCs for evaluating the maximum power density, the air-breathing DMFC has a limited air supply. On the other hand, the flow rate of the methanol solution in the anode in this air-breathing DMFC was con-

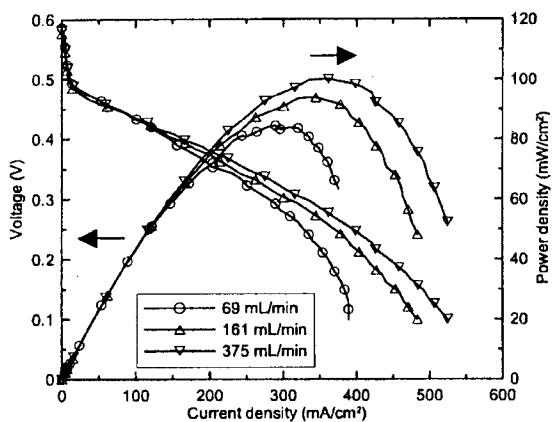


Fig. 4 Performance of SS-based micro DMFC at 60°C

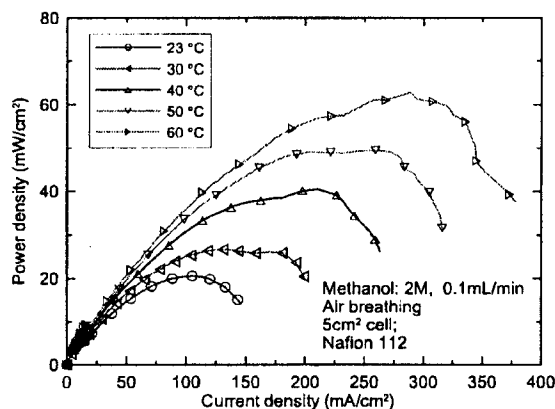


Fig. 6 Power density curves of the air-breathing DMFC at different operating temperatures

trolled to be at low flow rate to reduce the methanol crossover effect as well as to reduce the parasitic power to drive the liquid pump.

Figure 5 depicts the polarization curves of the air-breathing DMFC at different operating temperatures with a constant stoichiometry of 2.5 at 150 mA/cm² in the anode. Shown in Fig. 5, a significant voltage drop appears at low current density, indicating that activation overpotential is very large. From the profiles of the polarization curves shown in Fig. 5, no limiting mass transport phenomena can be observed even at the large current density. This indicates that the oxygen transport in the air-breathing cathode is still efficient. On the other hand, although the feeding rate of methanol in the anode was only with the stoichiometry of 2.5 at 150 mA/cm², which theoretically gives the maximum limiting current density of 375 mA/cm², the actual maximum limiting current density at 60°C shown in Fig. 5 is larger than this value. This is due to the parallel flow channels in the cell design. The total cross-sectional area of the parallel flow channels is too large, which makes a large amount of methanol solution remaining stagnant in the flow channels. The residual methanol solution in the channels provides an additional mass source for the electrochemical reaction, although with the low methanol feeding rate from the methanol tank. As a conclusion, for the small liquid flow rate, the parallel flow channel design is not suitable for decreasing the actual methanol feeding rate, which in turn decreases the methanol crossover rate. One way to get a real limiting current density

in the polarization curve is to measure the current density by fixing the cell voltage and operate the cell for an extend period until a steady state is reached.

Figure 6 shows the power density curves at different operating temperatures. The maximum power density reaches about 40 mW/cm² at 40°C and 61 mW/cm² at 60°C. By fixing the operating current density at 50 mA/cm² at room temperature, we measured the variation of the voltage to examine performance stability. In the experiments, we found the cathode was very likely to suffer from flooding. As shown in Fig. 7(a), small droplets initially appeared on the surface of the GDL in the cathode. Gradually the droplets enlarged, as shown Fig. 7(b) and extended their surface to the edge of channels. Eventually, the droplets collapsed and formed a liquid film covering the surface of the GDL, as shown in Fig. 7(c). This resulted in the reduction of the mass transport effective area in the cathode and cell performance. Figure 8 shows the variation of the voltage with time by fixing the current density at different temperatures. At room temperature (23°C), the voltage is eventually decreased, as shown in Fig. 8, which is thought to be caused by the cathode flooding effect. At elevated temperature, such as 60°C, a large amount of water can be evaporated and therefore cathode flooding is alleviated. The voltage is relatively stable with no significant degradation. The small fluctuation of voltage at 40°C and 60°C shown in Fig. 8 may be caused by several effects, such as intermittent methanol

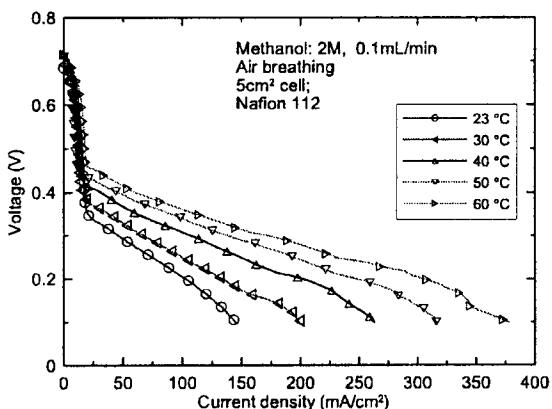


Fig. 5 Polarization curves of an air-breathing DMFC at different temperatures

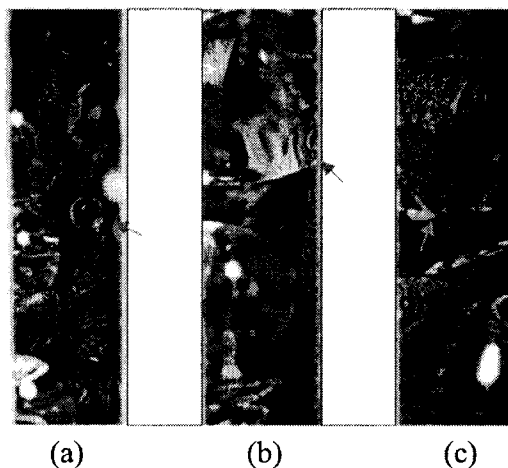


Fig. 7 Flooding at the GDL surface at room temperature

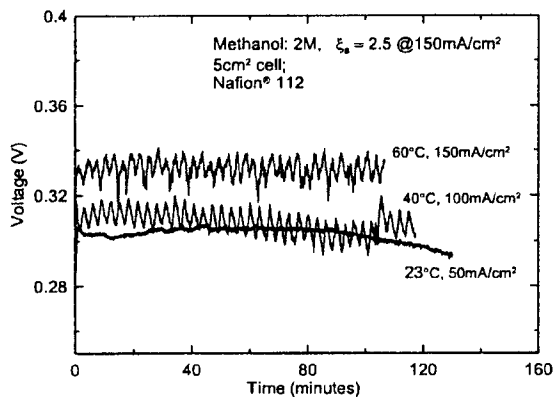


Fig. 8 Durability of cell performance

flow due to the nature of methanol pump, periodical blockage of flow channel by the CO_2 bubble, and temperature fluctuation of the cell. The steady-state power output for this air-breathing DMFC is about 50 mW/cm^2 at 60°C .

An 8-Cell Air-Breathing DMFC Stack

An 8-cell air breathing DMFC stack was developed aiming to power cell phones [19]. Stainless steel plates with a thickness of $500 \mu\text{m}$ were used as substrate plates to collect current. Single-pass serpentine flow channels in the anode were fabricated on the plates by photochemical etching method. The effective area of each cell is 5 cm^2 and the total gross area is $33 \text{ mm} \times 33 \text{ mm}$. In order to minimize contact resistance and prevent corrosion, a gold layer with a thickness of 300 nm was deposited on the front-side of each stainless steel plate. A rectangular polycarbonate plate ($145 \text{ mm} \times 33 \text{ mm} \times 5.6 \text{ mm}$) was fabricated with liquid feeding channels to support 8 individual cells (4 cells on each side). The gross volume of this 8-cell stack is about 34 cm^3 . Figure 9 shows a picture of the 8-cell air breathing DMFC stack. Figure 10 shows the flow paths arrangement for the 8-cell stack. The flow paths are parallel with the length of each flow channel being equal to achieve uniform distribution of the flow through each cell.

Water management is a key issue in DMFCs. 1 mol of water reacts with 1 mol of methanol at the anode, and 3 mols of water is produced at the cathode. Water is also transported from the anode to the cathode by electro-osmotic effect through the membrane, about 3×6 mols per mol of methanol if we assume the electro-osmotic coefficient to be 3 per proton for thick membrane such as Nafion 117 at 60°C [20]. Therefore, a total of 21 mols of water per mol of reacting methanol accumulate at the cathode if there is no water backflow effect. This excessive amount of water results in cathode flooding. Traditionally, larger air flow rate was used to prevent cathode flooding in DMFC. Apparently, this will not be possible for air breathing DMFC. A new water management technique was to promote the water backflow from the cathode to the anode through the thin membrane under the hydraulic pressure difference in the hydrophobic microporous layer in the cathode [11]. By properly choosing the contact angle and the micro pore

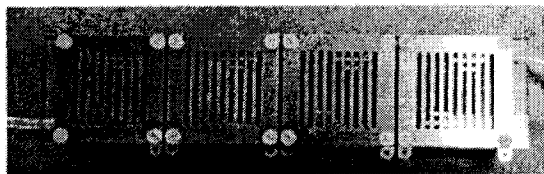


Fig. 9 A picture showing the 8-cell air-breathing DMFC stack

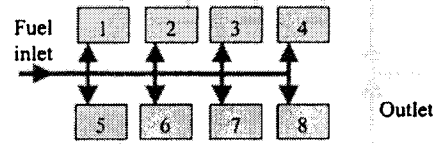


Fig. 10 Parallel flow paths for 8 individual cells

radius size of the microporous layer, the net water transport coefficient given in Eq. (1) can be reduced remarkably. The total rate of water entering and produced at the cathode is given as follows:

$$j_{\text{H}_2\text{O}} = \left(\alpha + \frac{1}{2} \right) \frac{I}{F} \quad (1)$$

where I is total current, F is Faraday's constant, and α is the net water transport coefficient which is a combined result of electro-osmotic drag, diffusion, and hydraulic permeation through the membrane. At 30°C , the net water transport coefficient, α , for our newly developed MEA based on our understanding of the water transport mechanism in DMFC, is only 0.12 at an air stoichiometry of 4@150 mA/cm^2 [11] compared to the electro-osmotic coefficient of about 2.25 [20]. Thus the total water flux at the cathode is only $j_{\text{H}_2\text{O}} = 0.62(I/F)$ for our new MEA, which is only 22.5% of water flux of $j_{\text{H}_2\text{O}} = 2.75(I/F)$ in the traditional MEA where the net water transport coefficient is almost same as the electro-osmotic coefficient. It is anticipated that the net water transport coefficient of our MEA could be lower, perhaps even negative, in air-breathing DMFC since the larger air flow rate will increase the net water transport coefficient [21]. Using this advanced MEA, less amount of water accumulated at the cathode and flooding was not observed in the 8-cell DMFC stack.

In the air-breathing DMFC, oxygen was passively transported to the cathode from the environment. To evaluate whether the air breathing DMFC suffers from the oxygen mass transport limitation in the cathode, we measured the limiting current density. Figure 11 shows the polarization curve of cell #1 supplied with 2 M methanol solution. Figure 12 shows the polarization curve of cell #1 using fully humidified hydrogen with very large flow rate. The cell temperature was measured to be around 30°C due to natural cooling for both cases, as given in Figs. 11 and 12. The limiting current density for cell #1 using 2 M methanol is around 160 mA/cm^2 , as shown in Fig. 11, while that using humidified hydrogen is 723 mA/cm^2 , as shown in Fig. 12. Because hydrogen is much more active than methanol with minimum activation loss

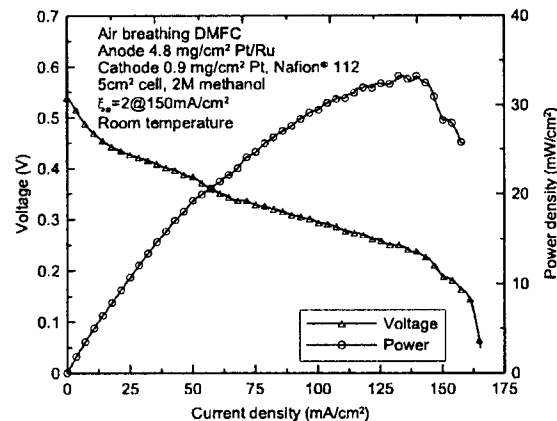


Fig. 11 Polarization curve of cell #1 in the stack

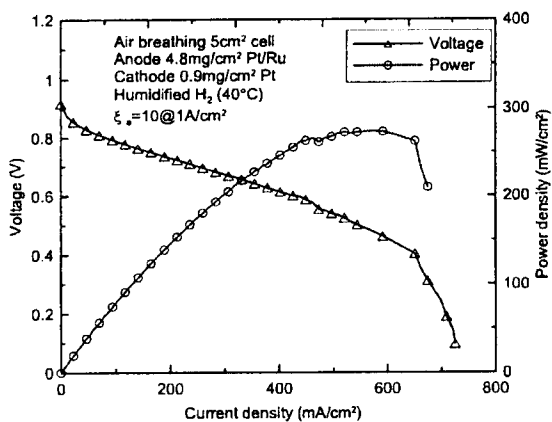


Fig. 12 Polarization curve of cell #1 using humidified hydrogen in the anode

at the anode and because the large hydrogen flow rate used in our experiments guarantee minimum mass transport loss at the anode, the limiting current density obtained with supply of humidified hydrogen mostly accounts for the mass transport limitation at the cathode in the air breathing cell. This limiting current density due to oxygen transport limitation at the cathode is about 4.5 times higher than that obtained for cells supplied with 2 M methanol solution. We can conclude that a sufficient amount of oxygen was supplied to the cathode in the air breathing DMFC. Furthermore, the limiting current density using 2 M methanol is mainly due to the anode mass transport limitation, where we confined the anode stoichiometry of $2@150 \text{ mA/cm}^2$ to reduce the methanol crossover rate.

Methanol crossover at open circuit voltage (OCV) was measured by immersing the air-breathing stack in a pool of DI water or shielding the cathode by a film. Transient voltage scanning from 0 to 1.2 V, having the same polarity as an operating DMFC, was applied to each individual cell from an external power source (multichannel fuel cell test system, Arbin). Figure 13 depicts the distribution of transient crossover rates for the 8 individual cells at open circuit voltage (OCV). The average transient crossover rate is about 159 mA/cm^2 at OCV.

Figure 14 shows the polarization curve for the 8-cell stack. The cells were connected in series. The stack produced 1.28 W at 2.4 V and the maximum output power was 1.33 W at 2.21 V, corresponding to a power density of 33.3 mW/cm^2 .

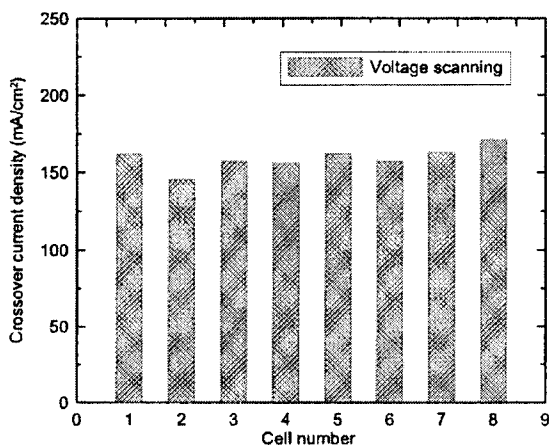


Fig. 13 Distribution of crossover rate at OCV

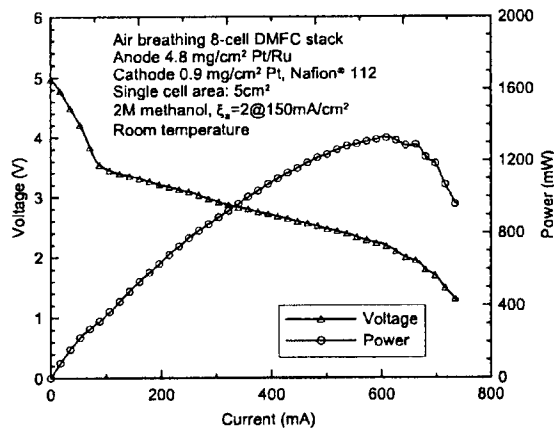


Fig. 14 Polarization curve for the 8-cell air-breathing DMFC stack

A Passive DMFC Using Pure Methanol

In a passive DMFC cell, the cathode is air breathing and the anode runs passively without any liquid pump to circulate the methanol solution. Therefore, the parasitic power is minimized in order to increase the net output of the system. By using innovative water management technology [11], water can back flow to the anode from the cathode through the membrane, as we have addressed in the preceding section. Consequently, it is possible to use pure methanol in the anode. Methanol crossover effect can be used to produce initial water concentration in the cathode at the open circuit voltage. More details on this passive DMFC using pure methanol will be discussed in future publications. Figure 15 shows our preliminary results of a passive DMFC using pure methanol in the anode. The passive DMFC produced a steady-state output power of $20\text{--}25 \text{ mW/cm}^2$ over more than 10 h without any heat management.

Conclusion

A Si-based micro DMFC was developed. Further improvement of micro DMFCs was made by using thin stainless steel plates as bipolar plates and the flow channels were microfabricated by photochemical etching method. The peak power reached 62.5 mW/cm^2 at 40°C and 100 mW/cm^2 at 60°C with atmospheric pressure. An air-breathing DMFC was investigated and it was concluded that cathode flooding is a critical issue. An 8-cell air-breathing DMFC was developed and characterized electro-

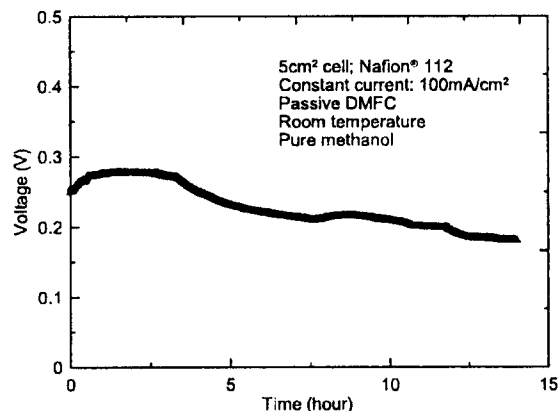


Fig. 15 Variation of voltage with time using pure methanol in a passive DMFC

chemically. By using the water backflow from the cathode to the anode through the thin membrane under the hydraulic pressure difference in the hydrophobic microporous layer in the cathode, flooding in the air-breathing cathode was avoided. It was found that oxygen transport in the air-breathing cathode is efficient. The 8-cell DMFC stack produced a maximum output power of 1.33 W at 2.21 V, corresponding to a power density of 33.3 mW/cm². Preliminary results of a passive DMFC demonstrate that it is totally feasible to use pure methanol. The passive DMFC using pure methanol produced a steady-state output power of 20–25 mW/cm² over more than 10 h.

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