

Short communication

A preliminary study of a miniature planar 6-cell PEMFC stack combined with a small hydrogen storage canister

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Abstract

The fabrication and performance evaluation of a miniature 6-cell PEMFC stack based on Micro-Electronic-Mechanical-System (MEMS) technology is presented in this paper. The stack with a planar configuration consists of 6-cells in serial interconnection by spot welding one cell anode with another cell cathode. Each cell was made by sandwiching a membrane-electrode-assembly (MEA) between two flow field plates fabricated by a classical MEMS wet etching method using silicon wafer as the original material. The plates were made electrically conductive by sputtering a Ti/Pt/Au composite metal layer on their surfaces. The 6-cells lie in the same plane with a fuel buffer/distributor as their support, which was fabricated by the MEMS silicon–glass bonding technology. A small hydrogen storage canister was used as fuel source. Operating on dry H₂ at a 40 ml min⁻¹ flow rate and air-breathing conditions at room temperature and atmospheric pressure, the linear polarization experiment gave a measured peak power of 0.9 W at 250 mA cm⁻² for the stack and average power density of 104 mW cm⁻² for each cell. The results suggested that the stack has reasonable performance benefiting from an even fuel supply. But its performance tended to deteriorate with power increase, which became obvious at 600 mW. This suggests that the stack may need some power assistance, from say supercapacitors to maintain its stability when operated at higher power.

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1. Introduction

Recently, numerous researchers have put their effort into fabrication of micro/miniature fuel cells (μ FCs for short) [1–11]. This is because μ FCs are widely considered to be a major candidate for the micro scale power generation with higher energy densities than the most competitive rechargeable batteries, Li-ion and Ni-MH batteries in particular. However, apart from the good designs and advanced materials, miniaturizing fuel cells for micro scale power sources is actually hindered

by the difficulty of reducing its physical dimensions because macro-sized fuel cell components (such as bipolar plates and fuel fed vessels) are often limited by their characteristic fabrication constraints, especially, when it comes to μ FCs. Thanks to Micro-Electronic-Mechanical-System (MEMS) technology, micro channel patterns of μ FCs bipolar plates into which reactants are fed and other components (such as micro fuel processors/reformers [5,12,13] and micro pumps [1]) can be fabricated on a silicon wafer with high resolution and good reproducibility. It has been demonstrated that MEMS is a most promising technology for fuel cell miniaturization and integration. Nevertheless, a lot of work was focused only on using such technology for fabrication of the single cells [3,4,7,9]. In fact, for practical uses, this is not enough and several or even hundreds

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of single cells are needed to be stacked up or integrated to offer the required voltage and power. Up to now, only a few of stacks fabricated by MEMS technology were reported or proposed. For example, Lee et al. [14] reported a micro fuel cell stack in which a planar array of cells was serially connected in a “flip-flop” configuration. The peak power in a 4-cell silicon assembly was reported to reach 40 mW cm^{-2} using hydrogen and oxygen as fuel and oxidizer. Yu et al. [15] fabricated a miniature twin fuel cell connected in series by sandwiching two membrane-electrode-assemblies between two silicon micro machined plates. The electricity interconnection from the cathode of one cell to the anode of another cell is made in the same plane and the interconnection was fabricated by sputtering a layer of copper over a layer of gold on the top of the silicon wafer. The measured peak power density was 190.4 mW cm^{-2} at 270 mA cm^{-2} when operating on dry H_2/O_2 at 25°C and atmospheric pressure.

In this paper, fabrication and performance evaluation of a miniature planar 6-cell PEMFC stack based on MEMS technology is presented. Bipolar plates of the stack and fuel buffer/distributor were fabricated by classical MEMS and silicon–glass bonding technologies, respectively. Performance evaluation of the stack was done by linear polarization and constant power discharging experiments under dry hydrogen, which was originated from a small AB_5 -type hydrogen storage canister and air-breathing conditions.

2. Experimental

The present work was focused on assembling a miniature planar 6-cell stack by mounting the single cells on the surface of the fuel buffer/distributor fabricated by silicon–glass bonding. Each of the 6-cells was fabricated by sandwiching the MEA between the silicon wafer-based flow field plates. The plates were made electrically conductive by sputtering a Ti/Pt/Au composite metal layer on their surface.

2.1. Fabrication of flow field plates for the single cell on a silicon wafer

A 4 in., $525 \pm 20 \mu\text{m}$ thick N-type (100) silicon wafer with resistance ratio ranging from 1.5 to $2.5 \Omega \text{ cm}$ was used as the

original material in this work. The fabricating processes was briefly introduced as follows: (1) heat deposit of a silicon dioxide layer on the wafer used as a silicon etch mask; (2) photolithography of the mask windows on the back of the wafer; (3) heat deposit of a silicon dioxide layer again to form the different thick layers on the back of the wafer; (4) photolithography of the mask windows on the front of the wafer; (5) etch of the front wafer in 40 wt% KOH solution at 50°C and the etching depth was controlled at $100 \pm 2 \mu\text{m}$; (6) wash the wafer in BOE solution (4 portions of 40% NH_4F and 6 of 49% HF mixture solution); (7) double sides wet etch the wafer in KOH solution again to form the pin-type flow field patterns with two square through holes for the hydrogen plates and the through holes flow field patterns for the oxygen plates; (8) remove the residual silicon dioxide on the surface of the wafer; (9) heat deposit of continuous silicon dioxide layers for electrical insulation; (10) sputtering a Ti/Pt/Au composite metal layers on the wafer surface to act as the conductive layer and ensure the metal layers would bond well in contact with the silicon wafer. The thickness of Ti/Pt/Au layer was controlled at 0.1, 0.3 and $0.5 \mu\text{m}$, respectively. SEM pictures of the final flow fields of anode and cathode fabricated were shown in Fig. 1. The flow field patterns of the anode are an array of pin-type isolated ‘isles’ and two through pseudo-square holes for fuel in and out. The isle is about $293 \mu\text{m} \times 293 \mu\text{m}$ on top and $300 \mu\text{m}$ in depth. The through hole for hydrogen in is about $1.5 \text{ mm} \times 1.5 \text{ mm}$ and that for hydrogen out is about $0.5 \text{ mm} \times 0.5 \text{ mm}$, respectively. The flow field pattern of the cathode is an array of pin-type isolated through ‘isles’ using for air in and out. The through holes are about $997 \mu\text{m} \times 997 \mu\text{m}$ on the plate’s back. The dimension of the flow field plate is about $1.6 \text{ cm} \times 1.8 \text{ cm}$ with active area of $1.2 \text{ cm} \times 1.2 \text{ cm}$. Note that width and length of the two plates is not equal and about $0.2 \text{ cm} \times 1.6 \text{ cm}$ free spaces was used for spot welding between the adjacent cells. In total, 10 anode flow field plates and 10 cathode flow field plates were formed on the silicon wafer.

2.2. Make the fuel buffer/distributor for the 6-cells using silicon–glass bonding technology

To make the fuel buffer/distributor, a MEMS classical wet etching and bonding technologies were applied. First, using the

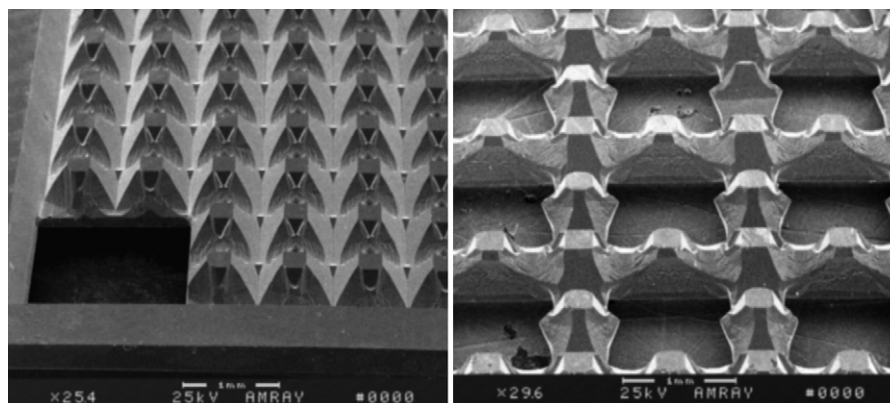


Fig. 1. SEM pictures of the final flow fields of anode and cathode fabricated.

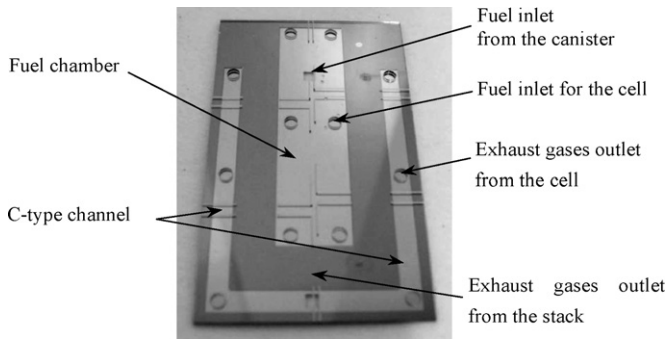


Fig. 2. The picture of the final bonded fuel buffer/distributor.

similar wet etching method described above, a strip chamber with dimensions of about 43 mm long, 11 mm wide and 100 μm deep was formed on the front side of another piece of silicon wafer with 350 μm thickness. This chamber will act as fuel (hydrogen) buffer. AC-type channel, a fuel feed hole and a square outlet hole were fabricated at the same time. The C-type channel is about 3 mm wide and 100 μm deep. The C-type channel will collect exhaust gases from the 6-cells and drain them out from the outlet hole. The fuel feed hole and outlet hole are 2.5 mm wide. Second, a single side polished. A piece of Pyrex glass with 500 μm thickness was ultrasonically drilled to form 12 through holes with 2 mm in diameter. The silicon–glass bonding was applied then in the bonding machine with 400 V working voltage for about 15 min, and the bonding temperature was controlled at about 800 $^{\circ}\text{C}$. Note again that 6 of the 12 through holes were designed to act as the single cells' feed hole and the remains outlet holes, respectively. The picture of the final bonded fuel buffer/distributor is given in Fig. 2 for reference.

2.3. Assembly the cells and mount them on the fuel buffer/distributor

Prior to assembly the single cells, a homemade membrane-electrode-assembly (with Pt loading 0.3 mg cm^{-2} for anode and 0.5 mg cm^{-2} for cathode, respectively; Nafion112 was used as separate membrane) was cut into 6 pieces with the dimension of 1.4 $\text{cm} \times 1.4 \text{ cm}$. In order to prevent short-circuited, the cut MEAs were ultrasonically agitated in deionized water for several seconds to remove the residential conductive materials on the cut sides.

To assembly the cell, a MEA was sandwiched between the anode and cathode plates and pressed for about 12 h by an electronic spiral micro meter. The Epoxy resin was used to seal the cell and act as a bonding agent between the MEA and the plates.

The assembled cells were then mounted one by one on the fuel buffer/distributor with the cathode facing up. All the void space between the cells and the glasses was filled with Epoxy resin seals to guarantee no gas leakage and good sealing. Work of assembling the 6-cell stack was finally completed by spot welding the adjacent cells to make them electrically conductive. The stack is about 4.0 cm^3 in volume and 8.5 g in weight. The entire assembly was then housed in an organic glass box with a ceramic plate cover. Again the Epoxy resin was filled in the void

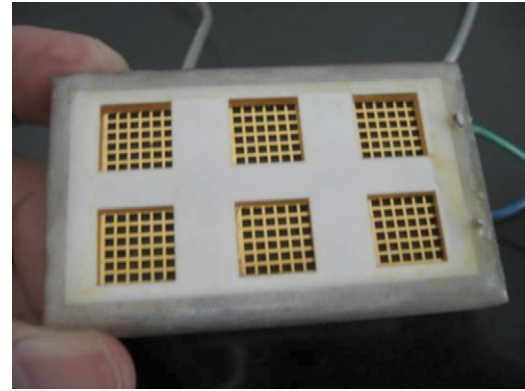


Fig. 3. The picture of the assembled 6-cell stack.

space to ensure the good bonding. The picture of the assembled stack is shown in Fig. 3.

2.4. Performance evaluation of the stack

Performance evaluation was done using Arbin FCT's fuel cell instrument (Arbin Instrument Co.). Before test, a homemade AB₅-type hydrogen storage canister was coupled with the stack (the canister's total volume was about 8 ml and hydrogen stored was about 4 l at room temperature). Hydrogen stored in the canister was used as the original hydrogen source for the stack without any humidification under atmospheric pressure. The composition of the AB₅ hydrogen storage material used was MmNi₅ and the hydrogen release was achieved by putting the canister in warm water bath at about 60 $^{\circ}\text{C}$. The flow rate of hydrogen was regulated at about 40 ml min^{-1} . Before collecting data, the stack was allowed to be activated for at least 1 h at a current density of 35 mA cm^{-2} . The stack was operated at 20 ± 3 $^{\circ}\text{C}$ and 50 $\pm 3\%$ relative humidity.

3. Results and discussion

The linear polarization curve of the stack is shown in Fig. 4. The shape of stack voltage versus current curve is typical and unique for a PEMFC stack. Usually, the curve is divided into three segments by its different voltage drop rates, which corre-

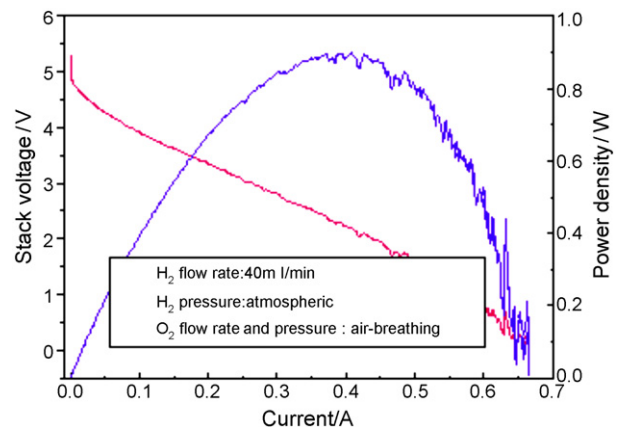


Fig. 4. The linear polarization curve of the stack.

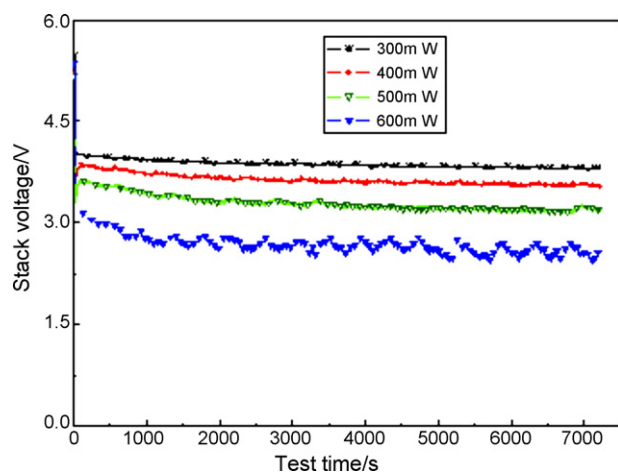


Fig. 5. The stack's constant power discharging performance at 300, 400, 500 and 600 mW.

spond to the different electrochemical processes [16]. The initial drop of the polarization curve at a very low current was due to the electrochemical activation process, which was caused by the sluggish kinetics of oxygen reduction at the cathode. The subsequent linear decrease of the polarization curve was due to an Ohmic loss, which is attributed to the ion flow through the electrolyte membrane, the electron flow through the electrode materials, flow field plates and current collector. The last drop was due to a diffusive loss caused by mass transfer difficulty of reactants, oxygen in most cases. It can be seen that the open circuit voltage of the stack is about 5.3 V and its peak power is about 0.9 W. This could mean that average open circuit voltage and peak power density of each cell approach 0.9 V and 104 mW cm⁻², respectively. Limited current density of the stack was about 650 mA cm⁻². Compared with previous reports [17,18] under similar operating conditions (ambient temperature and atmospheric pressure), we conclude that our miniature stack performed reasonably, which may be attributed to even fuel distribution among the cells thanks to the good design of the fuel buffer/distributor.

Fig. 5 shows the stack's constant power discharging performance at 300, 400, 500 and 600 mW, respectively. In fact, for the practical applications, the fuel cell is often operated in constant power state for the better fuel utilization and performance stability. Clearly, performance stabilities tended to deteriorate with power increase. When the stack was operated at relative high power of 600 mW, this tendency became more obvious. This phenomenon suggests that the stack may need a power assistant device, such as a supercapacitor to maintain its power stability when operated at higher power. This is the further work we are going to carry out.

4. Conclusion

In this paper, the fabrication and performance evaluation of an MEMS-based planar 6-cell PEMFC stack combined with a small hydrogen storage canister, which was used as the hydrogen store are presented and discussed. Operating on dry H₂ with a 40 ml min⁻¹ flow rate and air-breathing conditions at room temperature and atmospheric pressure, the linear polarization test gave a measured peak power of 0.9 W at 250 mA cm⁻² for the miniature stack and average power density of 104 mW cm⁻² for the single cells. At the same time, performance stabilities tend to deteriorate with power increase. This tendency became more obvious when the stack was operated at a high power of 600 mW. This phenomenon suggests that the stack may need power assistant device, such as a supercapacitor to maintain its stability when operated at higher power.

Acknowledgement

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