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Planar array stack design aided by rapid prototyping in development of air-breathing PEMFC

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Abstract

The polymer electrolyte membrane fuel cell (PEMFC) is one of the most important research topics in the new and clean energy area. The middle or high power PEMFCs can be applied to the transportation or the distributed power system. But for the small power application, it is needed to match the power requirement of the product generally. On the other hand, the direct methanol fuel cell (DMFC) is one of the most common type that researchers are interested in, but recently the miniature or the micro-PEMFCs attract more attention due to their advantages of high open circuit voltage and high power density.

The objective of this study is to develop a new air-breathing planar array fuel cell stacked from 10 cells made by rapid prototyping technology which has potential for fast commercial design, low cost manufacturing, and even without converters/inverters for the system. In this paper, the main material of flow field plates is acrylonitrile–butadiene–styrene (ABS) which allows the fuel cell be mass-manufactured by plastic injection molding technology. The rapid prototyping technology is applied to construct the prototype and verify the practicability of the proposed stack design. A 10-cell air-breathing miniature PEMFC stack with a volume of 6 cm × 6 cm × 0.9 cm is developed and tested. Its segmented membrane electrode assembly (MEA) is designed with the active surface area of 1.3 cm × 1.3 cm in each individual MEA. The platinum loading at anode and cathode are 0.2 mg cm^{-2} and 0.4 mg cm^{-2}, respectively.

Results show that the peak power densities of the parallel connected and serial connected stack are 99 mW cm^{-2} at 0.425 V and 92 mW cm^{-2} at 4.25 V, respectively under the conditions of 70 °C relative saturated humidity (*i.e.*, dew point temperature), ambient temperature and free convection air. Besides, the stack performance is increased under forced convection. If the cell surface air is blown by an electric fan, the peak power densities of parallel connected and serial connected stack are improved to 123 mW cm^{-2} at 0.425 V and 105 mW cm^{-2} at 5.25 V, respectively. The forced convection air can not only increases the oxygen diffusion rate at the air-breathing surface, but also enhance the uniformity of output voltage distribution. The performance obtained in this work reaches to the state-of-the-air of air-breathing planar PEMFC stack comparing to recent literatures.

In this study, the different behavior of output performance at water-rich region and water-lean region is also discussed.

Keywords: PEMFC; Air-breathing; Miniature fuel cell; Array stack design; Rapid prototyping

1. Introduction

The PEMFC is one of the most interesting alternative clean power sources for portable applications and distributed power systems. It might also be a viable alternative in many special applications where a highly reliable source of electricity is needed; *e.g.* in the power system of hospital and submarine. At the same time, the miniature PEMFC will be a good electricity source of micro-aerial vehicles (MAVs), robots and many small power computer, communication and consumer electronics. To make fuel cell viable for small power applications, both the fuel cell and its system solutions need to be developed further. The major challenges to develop the miniature PEMFC stack are multi-cell stacking to obtain high voltage

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output, assembly of stack with small volume, fabrication of membrane electrode assembly (MEA) and integration of fuel cell and sub-systems.

Many authors pay more attention to the development of miniature or micro-fuel cell by MEMs technologies [1–4] and alternative stack design concepts as well as manufacturing technologies [5–9]. The supply of reactive gas to cathode in the miniature fuel cell is usually adopted by air-breathing concepts to reduce the auxiliary oxygen supply system. Recent literatures show the state-of-the-art of air-breathing planar PEMFC provide the power density in the range of 100–120 mW cm^{-2} [1,5–9].

Nowadays, three major types of stack design are bipolar stack design, pseudo-bipolar stack design and mono-polar stack design [10]. The bipolar stack design is suitable for the high power stack (from 100 W to 1 MW), and it needs highly efficient water and heat management to stabilize system and to optimize output performance. The pseudo-bipolar stack design is suitable for the moderate power stack (from 20 to 150 W), and it needs an extra electrical connection to link each bi-cell. The mono-polar stack design is suitable for the low power stack, and it has highest resistance among these designs.

In this work, a fuel cell test station is set-up and utilized to measure polarization curve of overall stack and individual cells by measuring overall current, voltage of stack and voltage of individual cell. The performance tests of the parallel connected stack and the serial connected stack have been completed, and the analysis of polarization curves of the parallel connected, the serial connected stack and the individual cell can provide the useful information to the future design of air-breathing miniature PEMFC stack.

2. Design concept of the miniature planar array PEMFC stack

The original design concept of a miniature planar array PEMFC stack is to design it with a small volume and high voltages output in a planar form. The specification of output voltage is 6 V for a general purpose electronic power source. There are many advantages like less stack layers, and less rely on the converter/inverter which may increase the power efficiency to the final utilization.

2.1. MEA

A segmented MEA with 10 individual cells on one membrane was designed and fabricated in this study. Although it is difficult to orientate gas diffusion electrode layers (GDE) on the membrane and to control the hot-pressing parameters, it can minimize the sealing problem and simplify the stack assembly. The MEA is composed of DuPont™ NRE-212, 20% Pt/C catalyst and 30% polytetrafluoroethylene (PTFE) treated gas diffusion layers. Each individual cell has 1.7 cm^2 reactive area with Pt loading of 0.2 mg cm^{-2} for anode and 0.4 mg cm^{-2} for cathode.

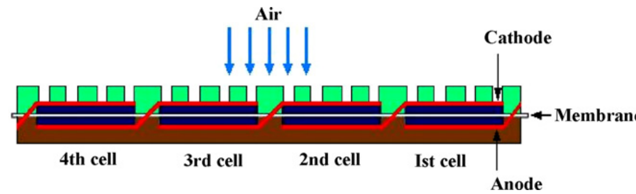

Fig. 1. The schematics of connection of the mono-polar stack.

2.2. Stack design

In this study, mono-polar stack design (Fig. 1) is applied to the miniature fuel cell stack and the planar figure of stack is designed to make cathode ventilated to air. The stack is composed of several layers (as shown in Figs. 2 and 3) including a proton exchange membrane with 10 active areas, sealing layers, collector plates, a ventilation plate, a fuel flow field plate, a fuel distributor and the end plates. Ribs are not only the flow channel walls in flow field plates but also the connectors that conduct electron to the load. Ribs are manufactured by copper and the copper surface is coated with a layer of gold to protect catalyst from poison ion. To make sure 10 individual cells on the same MEA insulating to each other, the flow field plates must be insulating materials. The ABS is selected because of its high mechanical strength, low cost and fabrication convenience both in rapid prototyping and future production. The fuel distributor distributes hydrogen from the upstream hydrogen source into 10 flow channels of 10 individual cells and gathers exhaust from flow channels to the hydrogen outlet. End plates are manufactured by aluminum, which can make collectors clip MEA tightly and uniformly. Proper design of end plates would results in good performance to avoid the high Ohmic resistance between the MEA and collectors, and adequate gas transportation in the gas diffusion layers. Because aluminum has advantages of high mechanical strength and light weight, it is a suitable material for end plates. A common electronic cooling fan (as shown in Fig. 3) was installed to enhance the convection to improve the output performance. The specification of fan in this paper is 6 cm × 6 cm in size, 12 V in voltage and 100 mA in current.

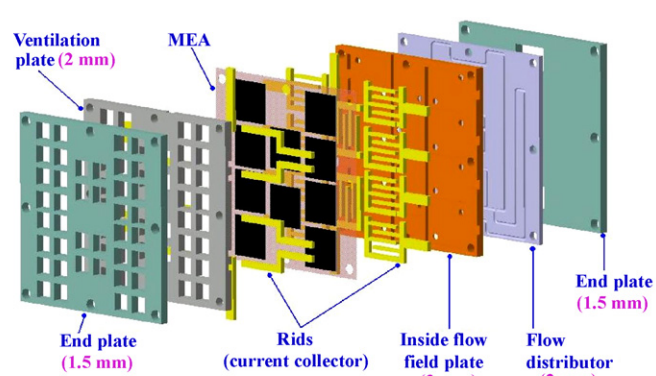

Fig. 2. Construction layout of the stack.

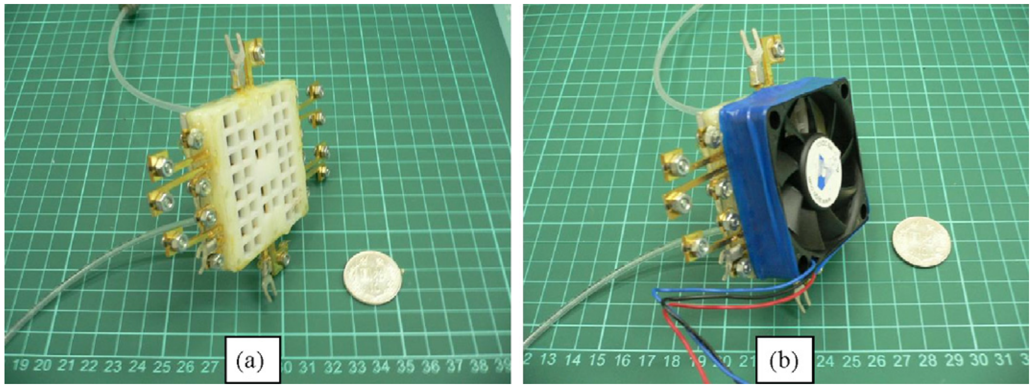

Fig. 3. (a) The 10-cell air-breathing miniature PEMFC stack and (b) the 10-cell air-breathing miniature PEMFC stack with electronic cooling fan.

3. New design and fabrication process for fuel cells—rapid prototyping

According to earlier studies reported in literatures, there are two major design and fabrication processes for micro to miniature planar air-breathing PEMFCs, *i.e.*, MEMs technology [1–4] and PCB technology [5–9]. In this study, a new design and fabrication process for this planar array of PEMFCs is proposed, that is rapid prototyping (RP) technology. This miniature planar array PEMFC is the first try in both academic and industrial area worldwide. After modifying the preliminary design in 2005 [11], its volume has been reduced from 6 cm × 6 cm × 3 cm to 6 cm × 6 cm × 1 cm and its performance become more and more stable.

Regarding the manufacturing time for the geometry of flow field plates in this work, the rapid prototyping takes around 1 h to finish, but MEMs is estimated to take 12–36 h and the conventional computer numerical control (CNC) machining is estimated to take more than 2 h. From the process time, one can find that rapid prototyping technology can accelerate the fuel cell fabrication at the early development stage. Besides, if the flow field plate is designed more and more complicated, CNC may not work on this complicated flow field, however, the rapid prototyping will take advantage in this manufacturing without any difficulty. Therefore, the successful development of RP-based fuel cells provides another practicable choice according to customer's requirement to develop a planar air-breathing fuel cell very quickly.

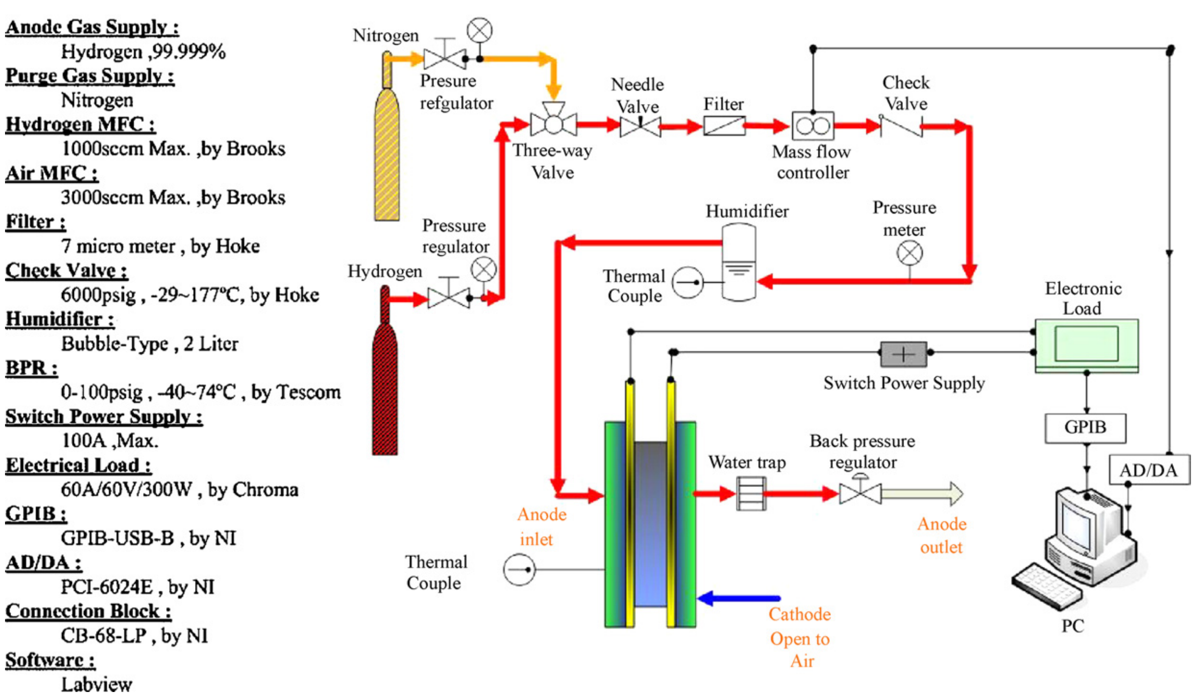

Fig. 4. Anode gas supply: Hydrogen, 99.999%; purge gas supply, Nitrogen; Hydrogen MFC, 1000 sccm max., by Brooks; air MFC, 3000 sccm max., by Brooks; filter, 7 μm, by Hoke; check valve, 6000 psig, −29 ∼ 177 °C, by Hoke; humidifier, bubble-type, 2 l; BPR, 0–100 psig, −40 ∼ 74 °C, by Tescom; switch power supply, 100 A, max.; electrical load, 60 A/60 A/300 W, by Chroma; GPIB, GPIB–USB–B, by NI; AD/DA, PCI-6024E, by NI; connection block, CB-68–LP, by NI; software, Labview.

The selected material of the flow field plates provided from rapid prototyping is acrylonitrile–butadiene–styrene (ABS) which can be extruded by the injection molding technique for customer design, and that will reduce the cost of product significantly. This successful verification of rapid prototyping fabricated miniature air-breathing fuel cells also implies that the future electronic prototypes even with very complicated geometry can integrate with fuel cell power module with rapid prototyping process. The plastic material lets the mass production of stack possible by plastic injection molding technology in the future.

Nowadays, computer integrated manufacturing (CIM) technology can transform quickly from a design concept to CAD data format, and rapid prototyping further utilize these CAD data to forming the three-dimensional model to accelerate the new product development. The rapid prototyping, according to a computerized free surface solid model either formed by forward or reverse engineering, will stack the material through a two-dimensional slice section layer by layer to form a three-dimensional solid model. This method distinguishes itself from conventional machinery processing (*e.g.*, CNC method) which applies subtractive method or removing unneeded parts from the original material, by applying additive method instead. This technology reduces producing and processing difficulties, and allows manufacturing to proceed as long as three-dimensional CAD design file has been completed. Rapid prototyping technology fast connects product design and manufacturing, without conventional numerical control code editing and considerations in finding available fixtures [12,13].There are six major rapid prototyping methods presently. The fused deposition modeling (FDM) method is selected to apply to the fabrication of flow field plates in this work because the operation temperature of PEM fuel cell is considerably low, and the material of ABS can not only sustain corrosion of hydrogen proton and operation temperature of fuel cells, but also can provide a high stiffness structure.

4. Experimental set-up

In this study, a fuel cell test station (as shown in Fig. 4) was established by authors themselves to measure the polarization curve of the stack and each individual cell. This test station can measure voltage, current and power through the electronic load, and can acquire data through data acquisition (DAQ) system. The volume flow rate of hydrogen is controlled by a hydrogen flow controller which is acquired and controlled by LABVIEW software through an analogy to digital converter. The bubbling type humidifier is used in this test station. A heating system is designed and controlled for the humidifier to increase the relative saturated humidity of reactant gas through increasing water temperature (*i.e.*, dew point). The accuracy of the temperature control is within ±1 °C.

In this work, the heating on the fuel cell is not necessary because the fuel cell stack works at room temperature. Extra-heating can enhance the performance of fuel cell because of higher activeness of catalyst. Commonly, 60–80 °C is the best operation temperature range of PEMFC. For miniature PEMFC, setting the heating system into integrated power generation system will substantially increase the system volume, and this will limit practicability of miniature PEMFC stacks. Therefore, extra-heating in fuel cell is not a good way to enhance the miniature PEMFC stack performance.

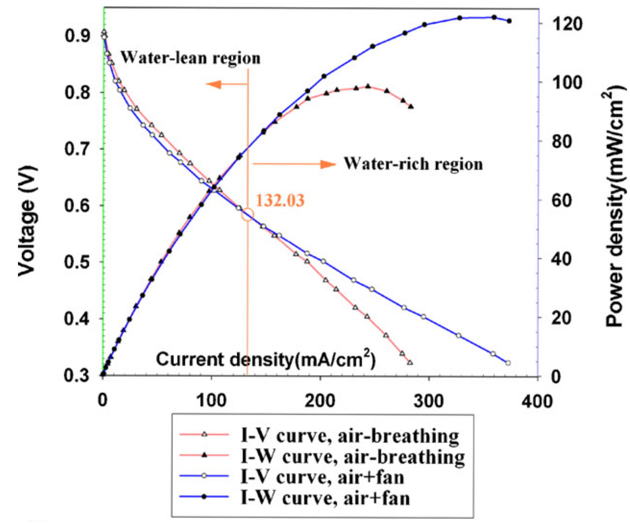

Fig. 5. Performance curves of parallel connected fuel cell at 25 °C, in 70 °C relative saturated humidity of hydrogen and different air supply conditions.

5. Results and discussion

5.1. Performance test in parallel-connection

There are 10-segmented reactive areas in one proton exchange membrane. The anode is on the same side of the membrane, and the cathode is on the other side. Parallel-connection of this stack works like a single cell with 17 cm^2 reactive area.

Fig. 5 shows the current density and power density curves of parallel connected stack operating at room temperature (25 °C), 70 °C relative saturated humidity of hydrogen and different air supply conditions (free and forced convection). In the free convection condition, the peak power density reached 99 $mW\,cm^{-2}$ corresponding to the current density of 233 $mA\,cm^{-2}$ at 0.425 V. While applying forced convection air, the peak power density was improved to 123 $mW\,cm^{-2}$ corresponding to the current density of 289 $mA\,cm^{-2}$ at 0.425 V. There is about 25% power increase from the free convection to the forced convection.

The oxygen supplement of cathode in the free convection condition depends only on the oxygen diffusion driven by concentration gradient. However, in the forced convection condition, oxygen diffusion is driven by concentration gradient and pressure gradient. Therefore, the fuel cell in forced convection condition has better performance than that in free convection apparently.

5.2. Performance test in serial-connection

To connect cells serially, the cathode of former cell must connect with the anode of latter cell sequentially. Serial-connection

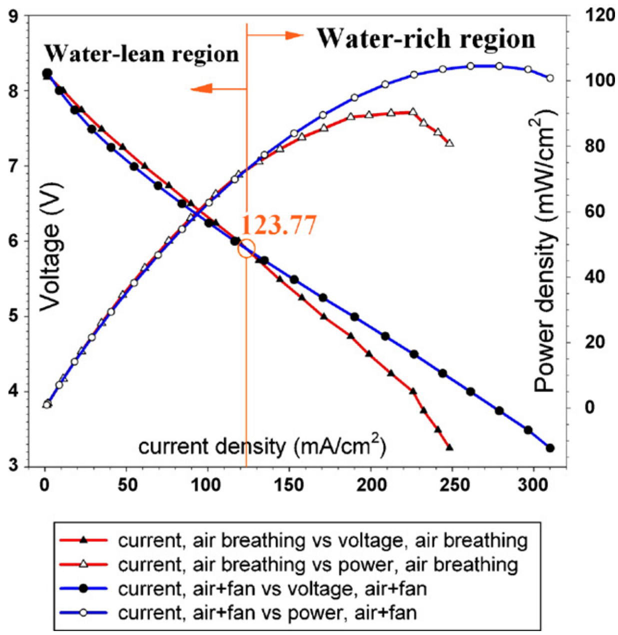

Fig. 6. Performance curves of serial connected fuel cell at 25 °C, in 70 °C relative saturated humidity of hydrogen and different air supply conditions.

is easier to be achieved for a bipolar stack, but for a mono-polar stack, serial-connection needs more careful connection solutions to reduce redundant Ohmic resistance from the connection wires.

Fig. 6 shows the current density and power density curves of serial connected stack at room temperature (25 °C), 70 °C relative saturated humidity of hydrogen and different air supply conditions (free and force convection). In the free convectional air condition, the peak power density reached 92 mW cm^{-2} corresponding to a current density of 216 mA cm^{-2} at 4.25 V. In the forced convectional air condition, the peak power density also increases with increase of humidity and reach 105 mW cm^{-2} corresponding to a current density of 200 mA cm^{-2} at 5.25 V.

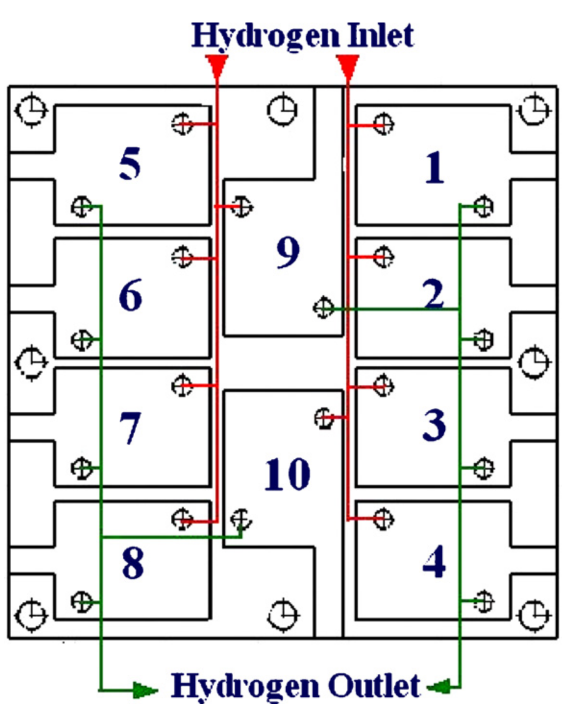

Fig. 7. Schematic drawing of sequence numbers of individual cells.

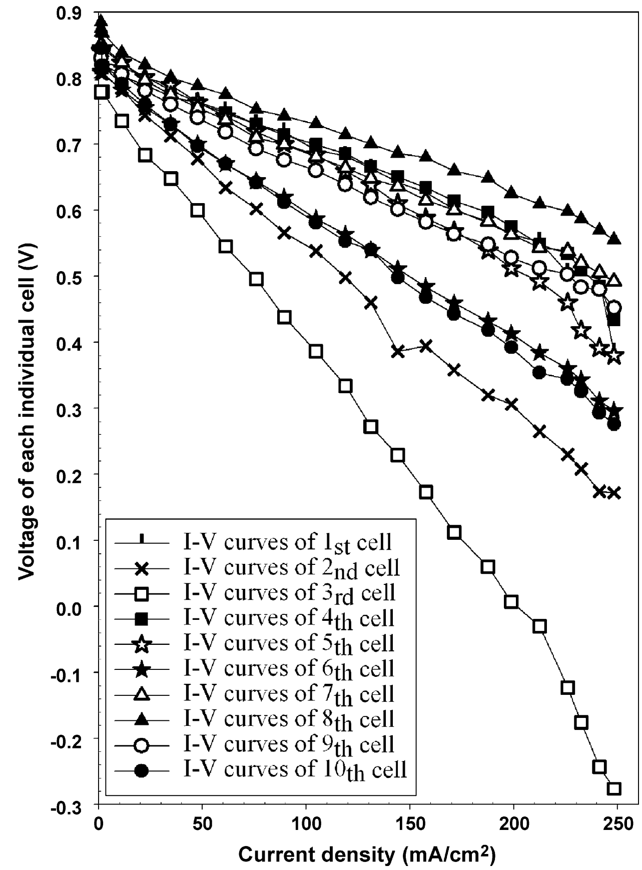

Fig. 8. Polarization curves of the stack and individual cells at 25 °C and 70 °C relative saturated humidity of hydrogen, free convection air.

From Figs. 5 and 6, forced convection can improve the power density in the water-rich region, but it contrarily has negative effect in the water-lean region. In the water-rich region, the better performance in forced convection condition is predictable. However, in the water-lean region, lower performance is out of our expectation. Detail examination of the operation mechanism, it seems still follow the physical phenomenon. It is explained that although forced convection can enhance the oxygen diffusion rate, it will increase the water evaporation rate simultaneously, so that the water produced by electrochemical reaction is insufficient relatively at lower current density. Finally, the combined effect of the high water evaporation rate and low water production rate results in the decreasing ion conductivity of membrane, and the performance drop down accordingly.

The authors further notice that there are two are the two critical points that separate the water-rich and water-lean region, *i.e.*, in this study, 132.03 mA cm^{-2} for parallel-connection and 123.77 mA cm^{-2} for serial-connection. At these two points, the water evaporation rate and production rate will reach an equilibrium condition, and two curves coincide which are shown in Figs. 5 and 6, respectively.

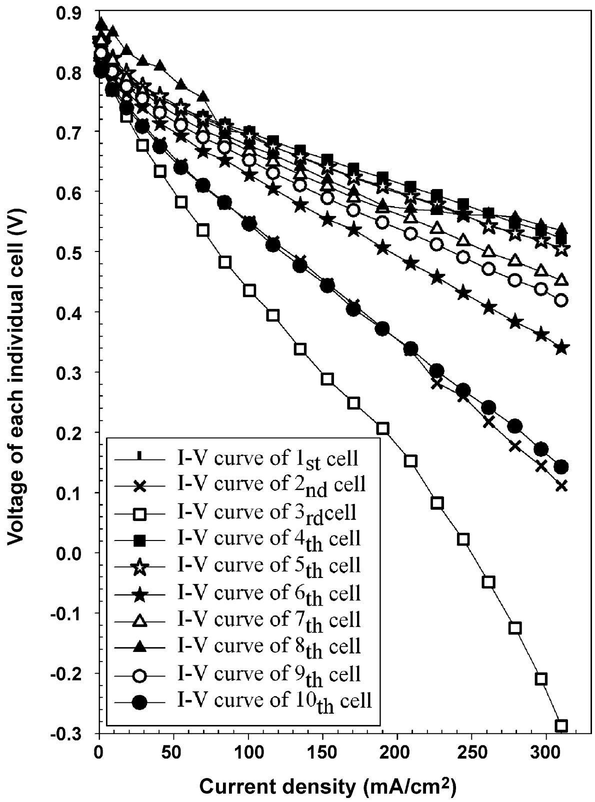

Fig. 9. Polarization curves of the stack and individual cells at 25 °C and 70 °C relative saturated humidity of hydrogen, forced convection air.

5.3. *Polarization curve of each individual cell in serial-connection*

In this work, sequence numbers were assigned to 10 individual cells (Fig. 7). Measurement of current of stack and voltage of overall stack and individual cells has been completed simultaneously to analyze the performance of each individual cell and its influences on the stack.

Fig. 8 shows the polarization curves of 10 individual cells at room temperature (25 °C) in the condition of free convectional air and 70 °C relative saturated humidity of hydrogen. The 3rd cell's performance is worst and the 2nd and 10th cells' performance are slightly lower than others. Fig. 9 shows the polarization curves of 10 individual cells at room temperature (25 °C) in the condition of forced convectional air and 70 °C relative saturated humidity of hydrogen. The 3rd cell's performance is still the worst when forced convectional air has been supplied, but the other cells' performance has been effectively improved by about 20%. The worse performance of 3rd cell can be inferred that the MEA was not treated properly. For the planar array MEA, it is a difficult issue to hot-press 10 gas diffusion electrodes (GDEs) uniformly on one single membrane.

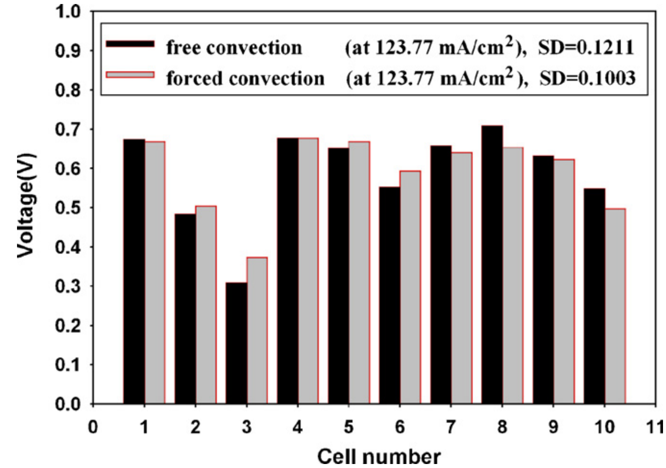

Fig. 11. Comparison of the voltage distributions of each individual cell at 60 mA cm^{-2} using different convection conditions (S.D. = 0.1003).

From the results, one can know one of the most difficult design issues for a planar PEMFC stack is the segmented MEAs treatment. A good treated MEA should not only have good performance but also good uniformity.

Figs. 10–12 show the voltage distributions of individual cells in free convection and forced convection at current densities of 60 mA cm^{-2}, 123.77 mA cm^{-2} and 200 mA cm^{-2}, respectively.

The standard deviation of the air-breathing planar stack using free convectional air at current densities of 200 mA cm^{-2}, 123.77 mA cm^{-2} and 60 mA cm^{-2} are 0.1836, 0.1211 and

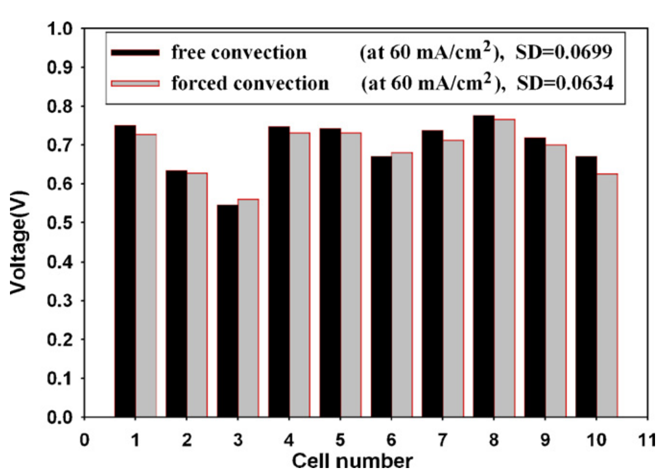

Fig. 10. Comparison of the voltage distributions of each individual cell at 60 mA cm^{-2} using different convection conditions (S.D. = 0.0634).

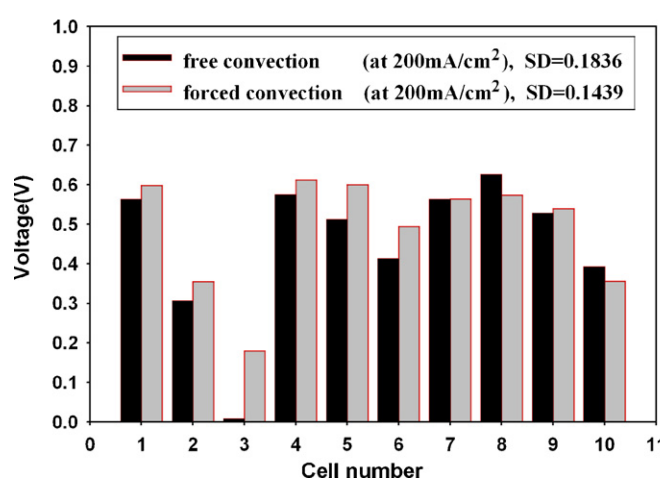

Fig. 12. Comparison of the voltage distributions of each individual cell at mA cm^{-2} using different convection conditions (S.D. = 0.1439).

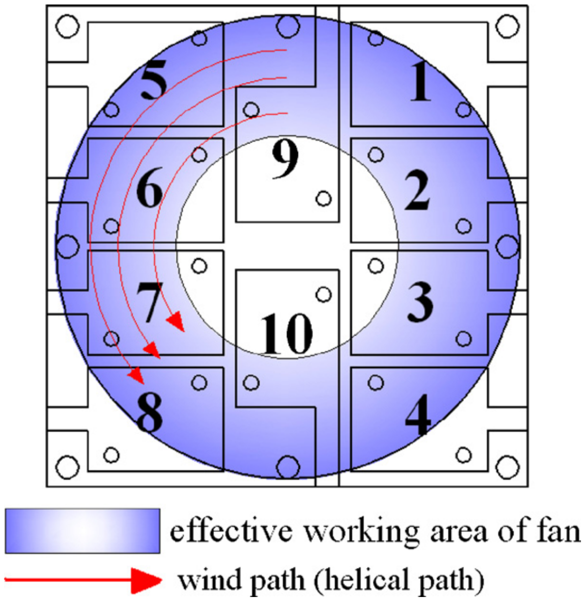

Fig. 13. The schematic of effective area and the wind path of the axial fan.

0.0699, respectively. Obviously, the standard deviation increases with increasing of current output. The higher standard deviation means the lower uniformity of performance distribution. When current increases, the temperature, water production, oxygen requirement and hydrogen requirement increase at the same time. Those factors will all cause the electrochemical reaction distribution to be less uniform.

The standard deviation of the air-breathing planar stack using forced convectional air at current densities of 200 mA cm^{-2}, 123.77 mA cm^{-2} and 60 mA cm^{-2} are 0.1439, 0.1003 and 0.0634, respectively. Apparently, the distribution becomes more uniform when the forced convection mode is applied. From these results, we find that the forced convection air not only enhances the oxygen diffusion rate but also improves the uniformity of the performance distribution in each individual cell.

Comparing serial-connection with parallel-connection, the stack has higher power density in parallel-connection than in serial-connection. The serial connected stack would be affected by certain cells which have worse performance. By contrast, the parallel-connected stack would not. The cells have worse performance means they have lower current density at a certain voltage and it will limit the current density of overall stack. Therefore, improving uniformity of individual cells can improve performance of the stack as well as improving certain cells' performance.

From Figs. 10–12, one can find that at all high current density regions, performance using forced convection is worse than that using free convection at 8th cell and 10th cell. There are two probable reasons for this particular result.

(1) *The distance effect of the axial fan*: In this case, the distance between the stack and the fan is too close to cause a higher field pressure at some locations which will result in lower water evaporation rate.
(2) *Non-uniform working area of axial fan*: According to the reference [14], the axial fan's axial velocity profile is annular (Fig. 13). Therefore, not every area can be affected by the forced convection. From Fig. 13, one can find two central cells and four cells at four corners have less affected area. The cells that have smaller forced convection affect from the forced convection.

Fig. 14. The demonstration of the 10-cell air-breathing miniature PEMFC powered cell phone directly without converters/inverters.

Fig. 14 shows the demonstration of 10-cell air-breathing miniature PEMFC powered cell phone. In this demonstration, no converter/inverter is needed and that is an advantage of this array stack design.

6. Conclusions

In this study, an RP-based 10-cell air-breathing miniature PEMFC stack with a volume smaller than 6 cm × 6 cm × 0.9 cm has been first successfully developed in both academic and industrial area worldwide. Using planar fuel cells in rapid prototyping technology for portable power sources obtained the following conclusions:

(1) Compared to MEMS and conventional CNC fabrication, rapid prototyping is much cheaper and faster in development stage.
(2) The performance of this air-breathing stack reach a state-of-art level compared to former literatures. The peak power densities of the parallel connected and serial connected

stack are 99 mW cm^{-2} at 0.425 V and 92 mW cm^{-2} at 4.25 V, respectively when operating at 70 °C relative saturated humidity, ambient temperature and free convectional air.

(3) In the forced convection, its performance is increased accordingly. The peak power densities of parallel connected and serial connected stack will be improved to 123 mW cm^{-2} at 0.425 V and 105 mW cm^{-2} at 5.25 V, respectively. The performance is also in the state-of-the-art.

(4) From the performance distribution of each individual cell, the forced convection air not only enhance the oxygen diffusion rate but also improves the uniformity of the performance distribution of each individual cell which are shown from the decrease of standard deviation.

(5) There are two critical points found from the *I–V* curves which show a coincide for the free and forced convection conditions. It is found that these critical points separate the water-rich and water-lean region, *i.e.*, 132.03 mA cm^{-2} for parallel-connection and 123.77 mA cm^{-2} for serial-connection. At these two points, the water evaporation rate and production rate will reach an equilibrium condition.

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