Journal of Power Sources 196 (2011) 717–728

Contents lists available at ScienceDirect

Journal of Power Sources

journal homepage: www.elsevier.com/locate/jpowsour

Experimental investigation of the effect of free openings of current collectors on a direct methanol fuel cell

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ARTICLE INFO

Article history:
Received 11 June 2010
Received in revised form 15 July 2010
Accepted 20 July 2010
Available online 30 July 2010

Keywords:
Current collector
Direct methanol fuel cell
Free open ratio
Perimeter length

ABSTRACT

A current collector is one of the key components of a direct methanol fuel cell (DMFC). For a planar-type DMFC, the current collector is usually fabricated from a thin metal that has open holes. The geometry of the current collector may have a significant effect on DMFC performance. Therefore, the design of the current collector is important for DMFC design. The objective of this study is to make a systematic experimental investigation of DMFC performance in the presence of current collectors with different free open ratios and total perimeter lengths of the free openings. Current collectors with 5×5, 7×7, and 10×10 hole arrangements under different total free open ratios of 30%, 40%, 50%, and 60% are investigated. The results show that the total free open ratio can significantly affect cell performance; they also show that decreasing the total free open ratio decreases cell performance, and increasing the total free open ratio increases cell performance. A high total free open ratio affects the total contact area between the membrane electrolyte assembly (MEA) and current collectors. Proper consideration of both the total free open ratio and the total contact area between the MEA and current collectors is necessary for the design of DMFC current collectors. In addition, a longer total perimeter of the free openings yields higher cell performance with the same free open ratio of the current collectors.

1. Introduction

Research on alternative energy sources has increased rapidly in recent years because petroleum reserves will be exhausted in a few decades. A fuel cell is a type of power generation device that directly converts chemical energy stored in fuel into electrical energy with high efficiency [1]. Among different types of fuel cells, the direct methanol fuel cell (DMFC) is suitable for its compact design, and it is prominent in portable applications because it has the major advantages of quick and convenient refueling, near room temperature operating conditions, safety, and uses low cost methanol. It is easy to store and carry, and it converts methanol directly into electricity without a bulky reformer [2]. Therefore, the DMFC is suitable for compact design, and the most promising short-term application of DMFCs is in the field of portable power sources. DMFC systems could possibly replace Li-ion batteries due to the recent advances of minimizing DMFC stacks [3].

The basic electrochemical reactions of the DMFC at the anode, at the cathode, and overall are as follows [4]:

$$\text{Anode}: \quad CH_3OH + H_2O \rightarrow 6e^- + 6H^+ + CO_2 \tag{1}$$

$$\text{Cathode}: \quad \frac{3}{2}O_2 + 6e^- + 6H^+ \rightarrow 3H_2O \tag{2}$$

$$\text{Overall}: \quad CH_3OH + \frac{3}{2}O_2 \rightarrow 2H_2O + CO_2 \tag{3}$$

Liquid methanol solution is usually adopted as the DMFC fuel. The reaction at the anode is an oxidation reaction that converts methanol and water into electrons, hydrogen protons, and carbon dioxide. Then the hydrogen protons on the anode side move though the electrolyte and arrive at the cathode side. The electrons move to the cathode through an external circuit. The reaction in the cathode is a reduction reaction that forms water from oxygen, protons, and electrons.

For polymer electrolyte membrane (PEM) fuel cells, the bipolar plates represent a significant part of the overall cost. A reduction in unit cost of materials or a reduction in the amount of material used could considerably reduce the cost of bipolar plates. Therefore, much effort has been invested in the development of cost-effective materials for bipolar plates, such as graphite materials, polymers

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doi:10.1016/j.jpowsour.2010.07.056

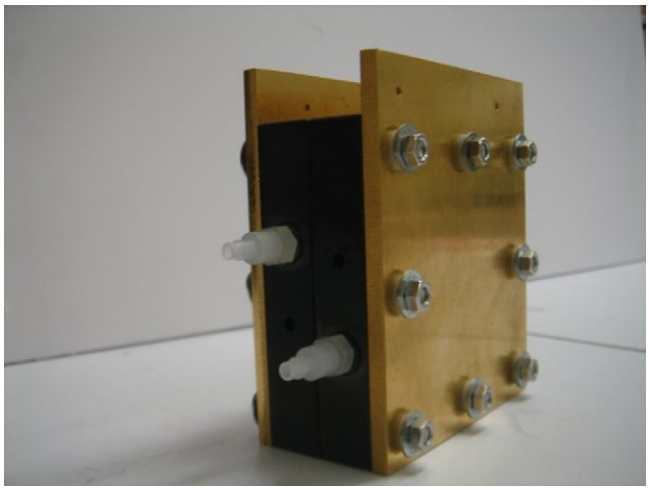

Fig. 1. A traditional single-cell DMFC structure.

mixed with graphite, and metallic materials. In addition, to reduce the thickness of the bipolar plates, it is necessary to reduce the amount of material used, which can also lower the cost of the bipolar plates [3–6]. The bipolar plates collect and carry current from the cells, distribute fuel and oxidant inside the cells, separate the anode and cathode sides from cell to cell in the stack, and remove water for thermal management. Generally, the bipolar plates of the fuel cells should have certain characteristics that allow them to achieve high performance and to maintain stable operation, such as superior capabilities of thermal and electric conductivities, good corrosion resistance, good gas impermeability, low density, and sufficient compressive strength. In addition, the cost of the material and the fabrication of the bipolar plates should be low [7–10].

A single-cell DMFC structure is shown in Fig. 1; the MEA is sandwiched between the anode and cathode polar plates, which are usually made of graphite or metal, and the flow channels are grooved on the surfaces. The polar plates distribute both fuel and oxidants while simultaneously collecting electrons. This type of DMFC is serially interconnected via bipolar plates, and this is referred to as vertical or bipolar plate stacking. Each bipolar plate simultaneously acts as the anode of one cell and the cathode of the neighboring cell. The cells are connected in a back-to-back orientation to form the stack [4]. To shrink the size of the DMFCs and make them more flexible in terms of design, especially for low-power portable applications, a side-by-side configuration that connects the cells in a lateral direction and forms a planar-type DMFC has been intensively explored as an alternative to vertical stacking in recent years. The main side-by-side configurations are the banded membrane concept and the flip-flop planar series interconnection. In the banded configuration, the electrodes overlap such that the cathode of one cell is strapped across the membrane to the anode of the next adjacent cell, and interconnections can be made by edge tabs. The flip-flop configuration adopts one-sided bipolar films, thereby eliminating the need to cross the membrane plane. Such configurations could allow DMFCs to be used in planar array fuel cells [11,12]. In addition, the printed circuit board (PCB)-type fuel cell (PCB-FC) is also a typical planar-type fuel cell. PCB technology is a mature and well-established mass production process. Therefore, the application of the PCB manufacturing process to fuel cells could increase design flexibility, shrink cell size, potentially lead to higher power densities, ease device integration, and improve packaging form factors [13–15].

Fig. 2 is a planar-type PCB-DMFC module; it is different from the convectional DMFC in that the flow channel and current collector are fabricated separately. The method used to make the correct free

Fig. 2. A planar-type PCB-DMFC module.

openings of current collectors is important for planar-type DMFCs; however, there are very few related studies of this method. Huang et al. [16] adopted a computational fluid dynamics (CFD) simulation to discuss the effect of breathing-hole size on the electrochemical performance of a free-breathing cathode on a DMFC. Their simulation results showed that a larger breathing hole increased the ohmic resistance penalty, and the increased area in contact with the fresh air reduced concentration polarization. However, they did not include the anode reaction, and the numerical results were not compared with the experiments. Chang et al. [17] performed systematic experimental studies on the free opening effects of current collectors on DMFC performance. Their investigation was accomplished using a single-cell DMFC in which the current collectors had Sierpinski carpet fractal holes. They found that the total free open ratio and the total perimeter length of the holes are two important factors, but the total perimeter length of the holes has a greater effect on DMFC performance than the free open ratio of the holes. In addition, a longer total free opening perimeter led to better cell performance under the same total free open ratio of current collectors. The total free open ratio of the current collector was only 30%, following the Sierpinski carpet rule. Later, Kuan et al. [18] adopted the Hilbert curve continuous-type fractal geometry in the design of the current collectors for the DMFC, which increased the total free open ratio of the current collector up to 50%; the total perimeter length of the openings was also significantly increased. The experimental results showed that current collectors with more uniform opening distributions and higher total free opening perimeter lengths could reduce the effect of anode flow rate on cell performance. In addition,

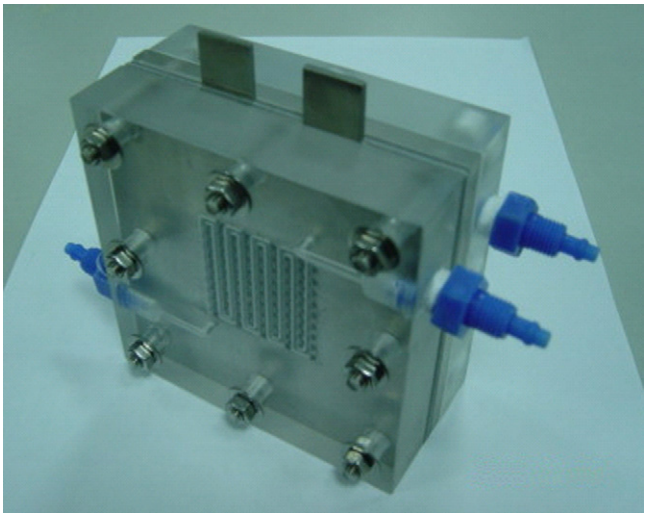

Fig. 3. A single-cell DMFC test fixture.

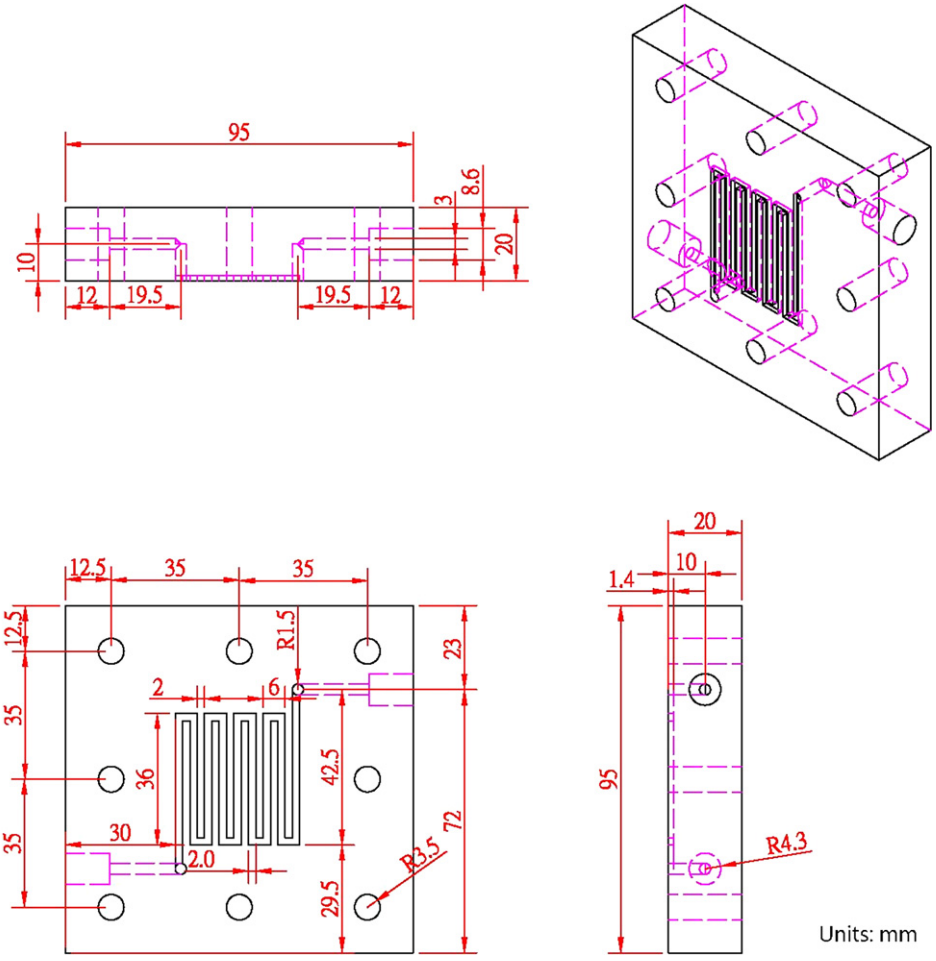

Fig. 4. The geometry and dimensions of the flow board.

the higher total free open ratio and total free opening perimeter length of the current collectors could increase cell performance. Therefore, an investigation of the effect of current collectors with higher total free open ratios and longer total perimeter lengths of openings is necessary. The purpose of this study was to conduct further study on how the openings on the electric collector affect DMFC performance. The current collectors were drilled as different arrays of rectangular openings with different sizes. The total free open ratio varied from 30% to 60%, and the total perimeter lengths of openings were changed via the number and size of the openings.

2. Experimental setup

This study evaluates the effect of opening parameters in the current collectors on the performance of a fuel cell. To simplify the experiments and ensure cell stability throughout the research, a DMFC with stainless steel 316L (SS316L) current collectors was adopted to alternate the metal films on the current collectors of the PCB-DMFC. Although the SS316L has lower electric conductivity than graphite and some metal materials, such as gold or copper, the SS316L was chosen as the material for the current collectors in this research because it has the advantages of low cost, wide availability, light weight, high mechanical strength, and easy machining. The corrosion resistance of SS316L is not as good as graphite, but this can be avoided by surface modification methods. Because the length of the experiments conducted in this research was not long enough to create surface corrosion on the surfaces of the SS316L current collectors and affect performance, it was not necessary to use surface treatments on the current collectors used in this study [17–22]. A single-cell DMFC test fixture was adopted in this paper, as shown in Fig. 3. The membrane electrolyte assembly (MEA) is sandwiched between the anode and cathode current collectors.

Both anode and cathode flow boards were made of polymethylmethacrylate (PMMA) and attached to two end sides of the DMFC test fixture. Each anode and cathode flow board had on its surface a single, grooved, serpentine flow channel; the geometry and detailed dimensions of this channel are shown in Fig. 4. The construction of the single-cell test fixture is shown in Fig. 5. To prevent leakage, a polytetrafluoroethylene (PTFE) gasket was placed between the two layers. The MEA was composed of Nafion® 117 as

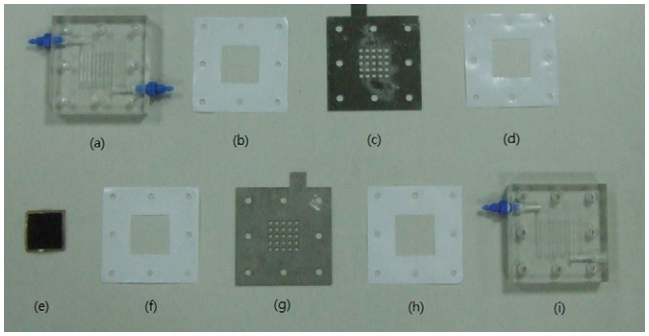

Fig. 5. Construction of the single-cell DMFC test fixture: (a) anode flow board, (b) gasket, (c) anode current collector, (d) gasket, (e) MEA, (f) gasket, (g) cathode current collector, (h) gasket, and (i) cathode current collector.

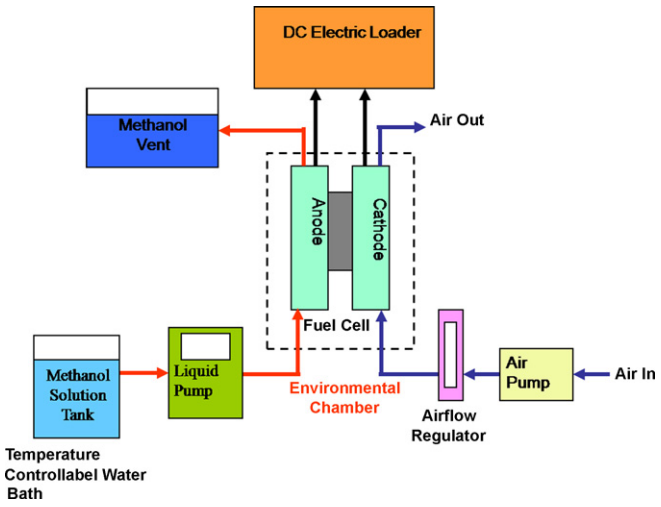

Fig. 6. Schematic illustration of the experimental setup.

the electrolyte and carbon cloth for the diffusion layers. The catalyst load at the anode was 4 mg cm^{-2} Pt–Ru, and the catalyst loading at the cathode was 4 mg cm^{-2} Pt. The active area of the MEA was 35 mm × 35 mm.

Fig. 6 shows a schematic illustration of the experimental setup for measuring the DMFC polarization curves. The DMFC was placed in an environment-controllable chamber. A beaker held the methanol solution that was preheated by a temperature-controllable water bath. The methanol solution was pumped into the DMFC anode by a squire pump. The airflow was driven and pumped into the DMFC cathode by an air pump with an airflow regulator. The DMFC was loaded by a DC electric loader.

To evaluate DMFC performance, the polarization curves for the DMFC with different free openings on the current collectors were determined. The polarization curve shows different limiting mechanisms occurring during the operation of a fuel cell at zero current, low current density, and high current density ranges [4].

3. Current collectors with different hole arrangements

In this research, a series of SS316L current collectors with rectangular holes in different arrangements were designed and machined. The free open ratio and the total perimeter of the free openings are two important factors for the design of the current collectors. To investigate the effect of these two important factors, rectangular holes were designed as 5 × 5, 7 × 7 and 10 × 10 arrangements with 30%, 40%, 50%, and 60% total free open ratio, which was achieved by using different hole sizes. Therefore, the current collectors could be designed to possess different free open ratios with different total perimeter lengths of the holes. A summary of the geometric information regarding hole arrangement, total free open ratio, and total perimeter length for the current collectors is shown in Table 1.

The size of each current collector is 95 mm × 95 mm, and its thickness is 2 mm. The reactive area of the MEA is 35 mm × 35 mm. The free open ratio is referred to as the reactive area (35 mm × 35 mm), in other words, the total free open area divided by the reactive area, as shown in Fig. 7. Fig. 8a–d shows the current collectors with a 5 × 5 hole arrangement with 30–60% total free open ratios. The total perimeter length of the openings of the current collectors with 30%, 40%, 50%, and 60% total free open ratios were 383, 443, 494, and 541 mm, respectively. Fig. 9a–d shows the current collectors with a 7 × 7 hole arrangement with 30–60% total free open ratios. The total perimeter length of the openings of the currents with 30%, 40%, 50%, and 60% total free open ratios were 537, 619.4, 693.8, and 758.5 mm, respectively. Fig. 10a–d shows

Table 1
Geometric information for different circular-type bipolar plates.

Reactive area	Hole arrangement	Total perimeter length of openings (mm)	Total free open area (mm^2)	Total free open ratio
35 mm × 35 mm	5 × 5	383.0	366.7	30%
	7 × 7	537.0	367.9	
	10 × 10	768.0	368.6	
35 mm × 35 mm	5 × 5	443.0	490.6	40%
	7 × 7	619.4	489.3	
	10 × 10	884.0	488.4	
35 mm × 35 mm	5 × 5	494.0	610.1	50%
	7 × 7	693.8	614.0	
	10 × 10	988.0	610.1	
35 mm × 35 mm	5 × 5	541.0	731.7	60%
	7 × 7	758.5	733.9	
	10 × 10	1084.0	734.4	

current collectors with a 5 × 5 hole arrangement and 30–60% free open ratios. Detailed dimensions are also shown in Figs. 8–10. The total perimeter length of openings of the current collectors with 30%, 40%, 50%, and 60% total free open ratios were 768, 884, 988, and 1084 mm, respectively.

4. Results and discussion

In all of the experiments, the DMFC was placed into a environment-controllable chamber, and the environmental conditions were as follows. Environmental temperature was 55 °C, and the relativity humidity was 60%. The anode was supplied with a 2 M methanol/DI water solution, and the flow rate was maintained at 15 cm^{-3} min^{-1}. The cathode was supplied with air, and the airflow rate was maintained at 1000 cm^{-3} min^{-1}.

In the design of the current collectors throughout the research, the total free open ratio of the current collectors determines the size of the space for the reactants to flow into the diffusion layers on both the anode and cathode sides, and it also determines the space used to remove the products produced from the elec-

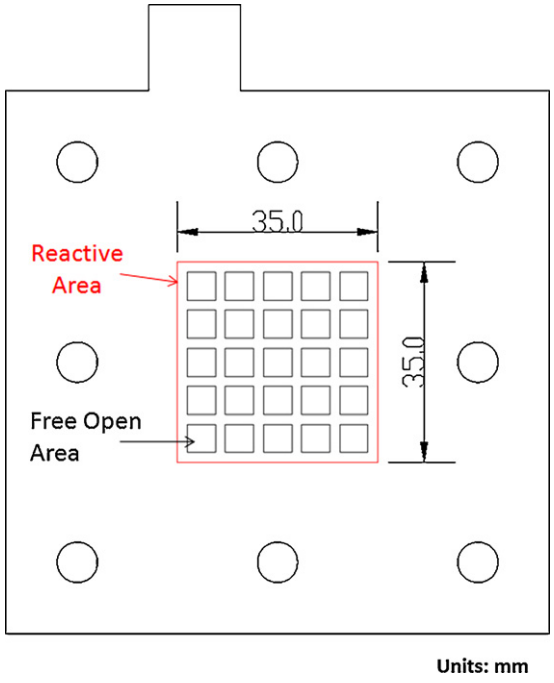

Fig. 7. Illustration of the free open area of the current collectors.

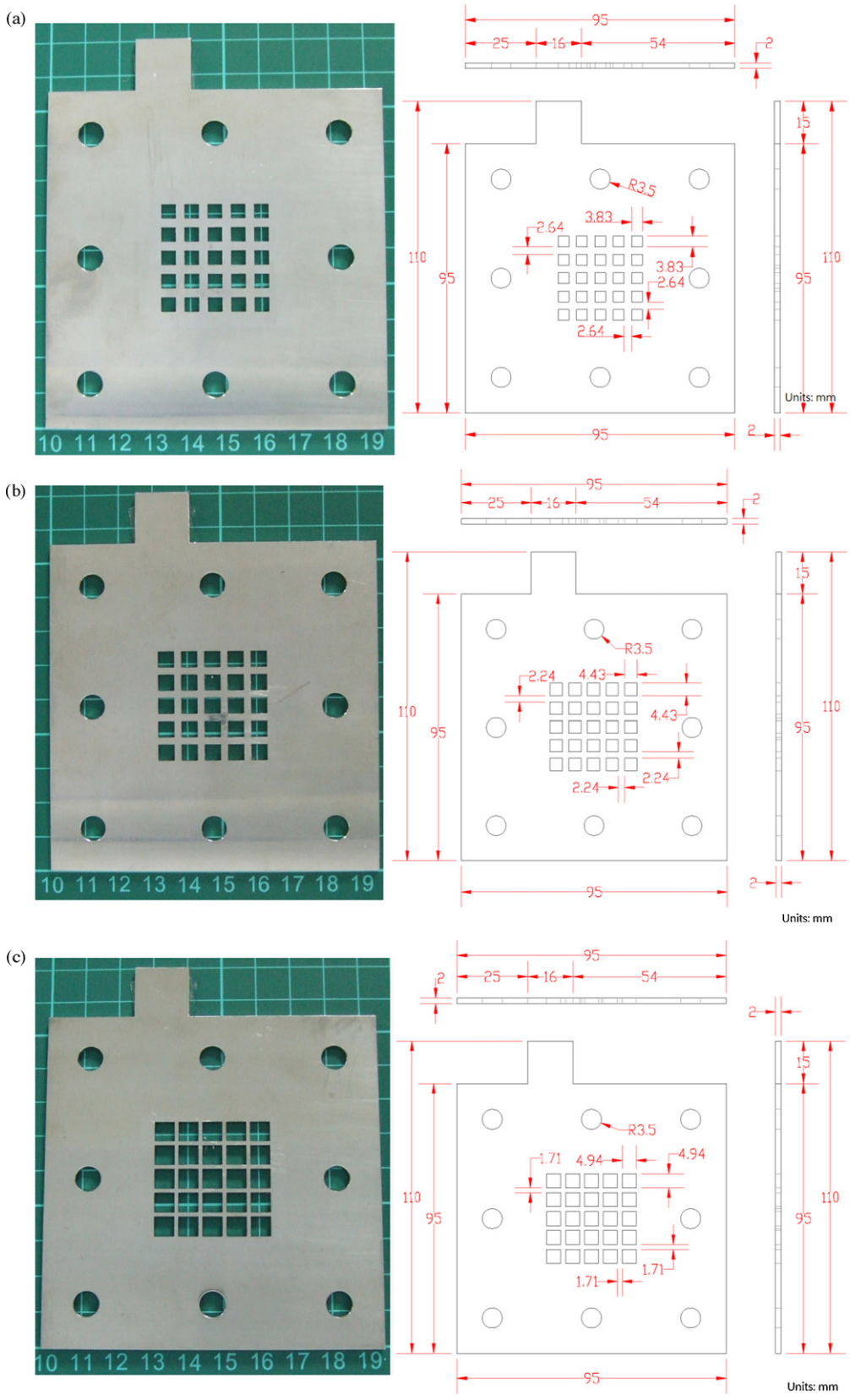

Fig. 8. Current collectors with a 5 × 5 hole arrangement: (a) 30% total free open ratio, (b) 40% total free open ratio, (c) 50% total free open ratio, and (d) 60% total free open ratio.

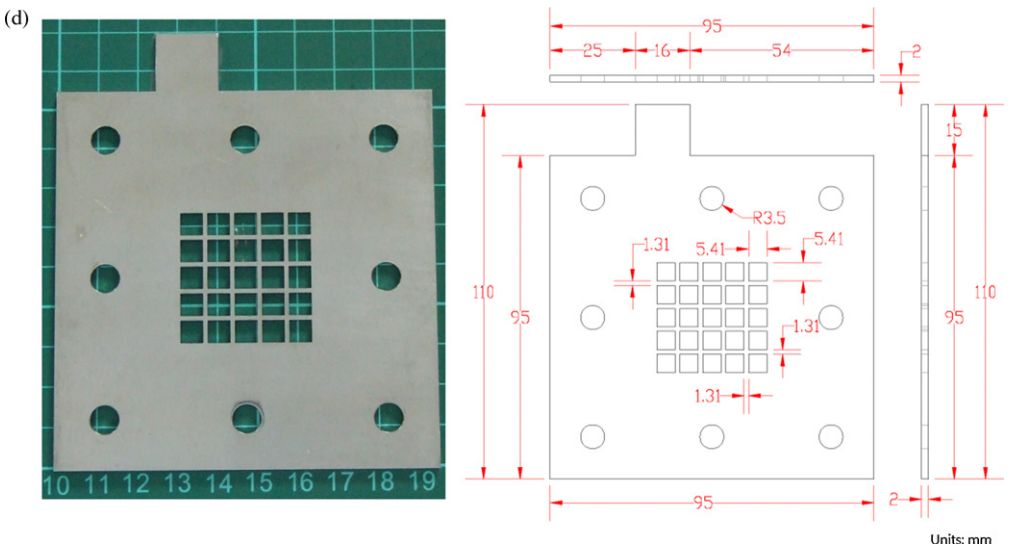

Fig. 8. (*Continued*)

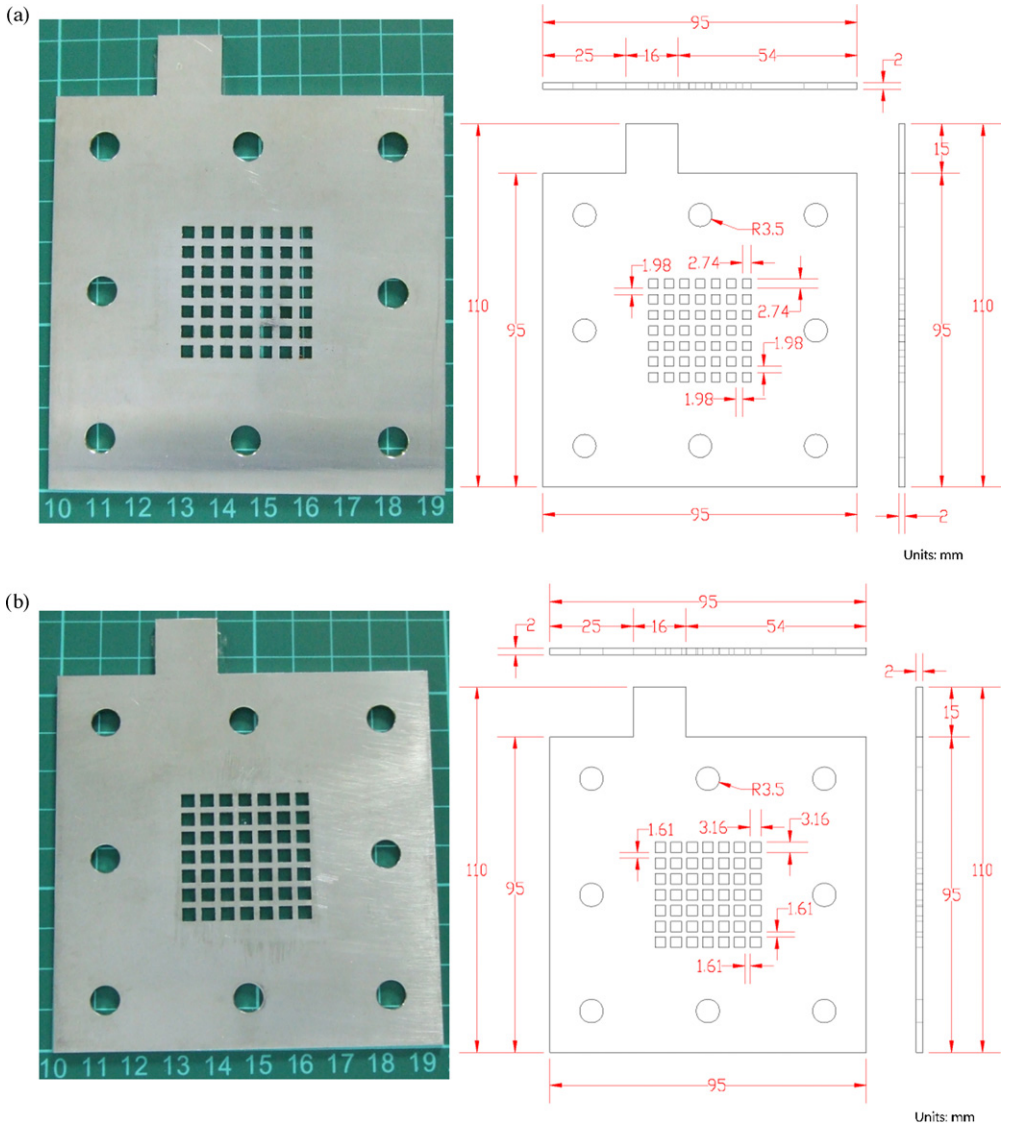

Fig. 9. Current collectors with a 7 × 7 hole arrangement: (a) 30% total free open ratio, (b) 40% total free open ratio, (c) 50% total free open ratio, and (d) 60% total free open ratio.

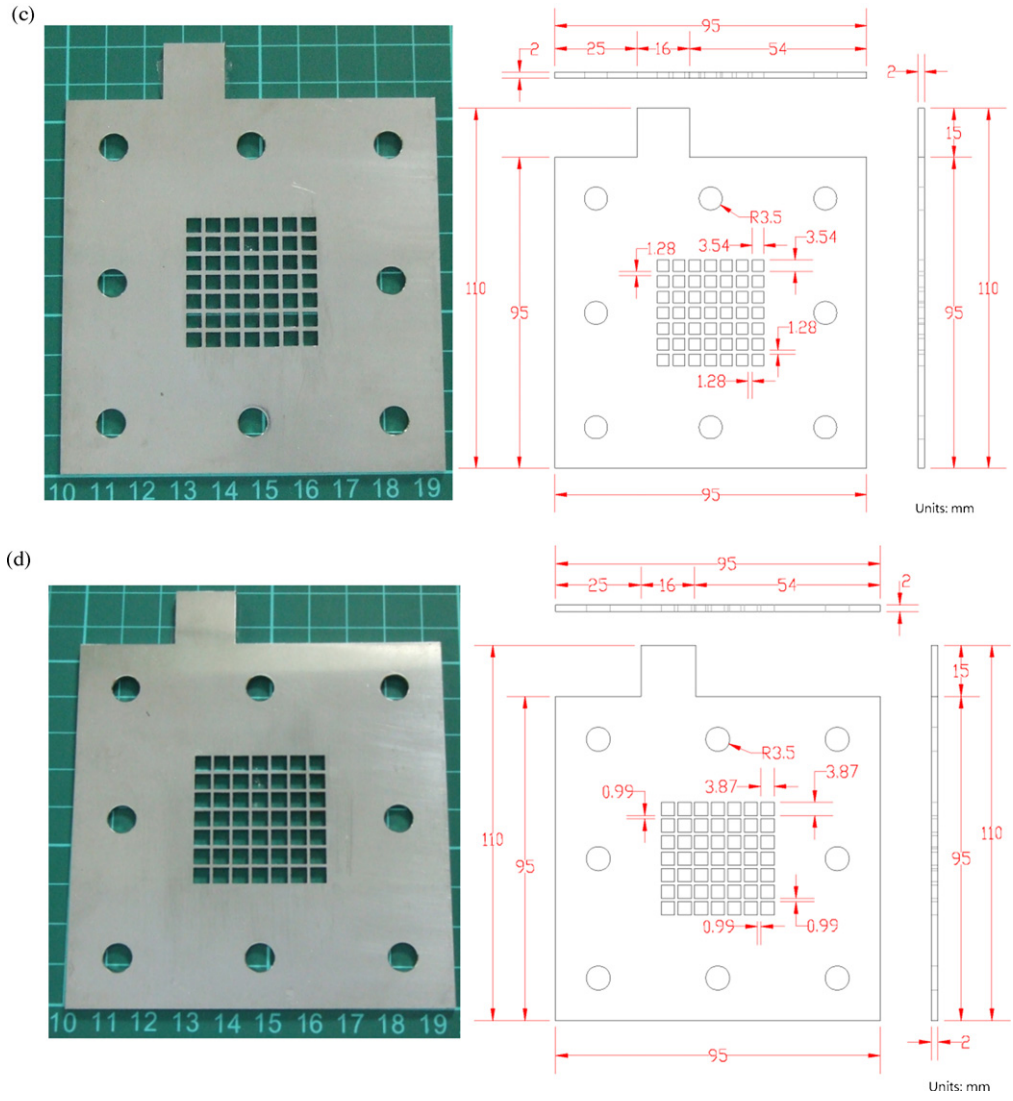

Fig. 9. (*Continued*)

trochemical reactions. However, a larger total free open ratio also means that the contact area between the MEA and current collectors is smaller. Higher total free area ratios have more space for reactants to flow and products to discharge. The total perimeter length of the openings on the current collectors determines the diffusion distances of the reactants and products from the closed areas behind the current collectors to the free open areas. Under the same total free area ratio of the current collectors, the longer total perimeter length of openings allows shorter lengths for the reactants diffused from the free open areas to the closed area behind the current collectors and shorter lengths for the products diffused from the closed areas behind the collectors to the free open areas on the current collectors.

4.1. Current collectors with the same hole arrangement but different total free open ratios

Fig. 11a and b shows a comparison of current density vs. cell voltage (*I–V* curve) and current density vs. power density (*I–P* curve) for DMFCs with different total free open ratios on the current collectors with the same 5 × 5 hole arrangement. The results show that the order of cell performance is 60% > 50% > 40% > 30% with respect to the total free open ratio of the current collectors. Current collectors with a larger total free open ratio and a 5 × 5 hole arrangement also have longer total free opening perimeter length. Therefore, the current collectors with higher free open ratios and a longer total perimeter length of openings have better cell performance.

Fig. 12a and b shows a comparison of the *I–V* and *I–P* curves for a DMFC with different total free open ratios in current collectors with the same 7 × 7 hole arrangement. The results show that DMFC performance with a 50% total free open ratio on the current collectors was similar but slightly higher than the one with a 60% total free open ratio on the current collectors. The DMFC with a 40% total free open ratio on the current collectors presented the third best cell performance, and the DMFC with 30% total free open ratio on the current collectors had the worst cell performance.

Fig. 13a and b shows a comparison of the *I–V* and *I–P* curves for the DMFC with different total free open ratios on the current collectors with the same 10 × 10 hole arrangement. Similar to the 7 × 7 hole arrangement, the results show that DMFC performance with a 50% total free open ratio on the current collectors is similar but slightly better than the one with a 60% total free open ratio on the current collectors. The DMFC with a 40% total free open ratio on the current collectors presented the third best cell performance, and the DMFC with a 30% total free open ratio on the current collectors had the worst cell performance.

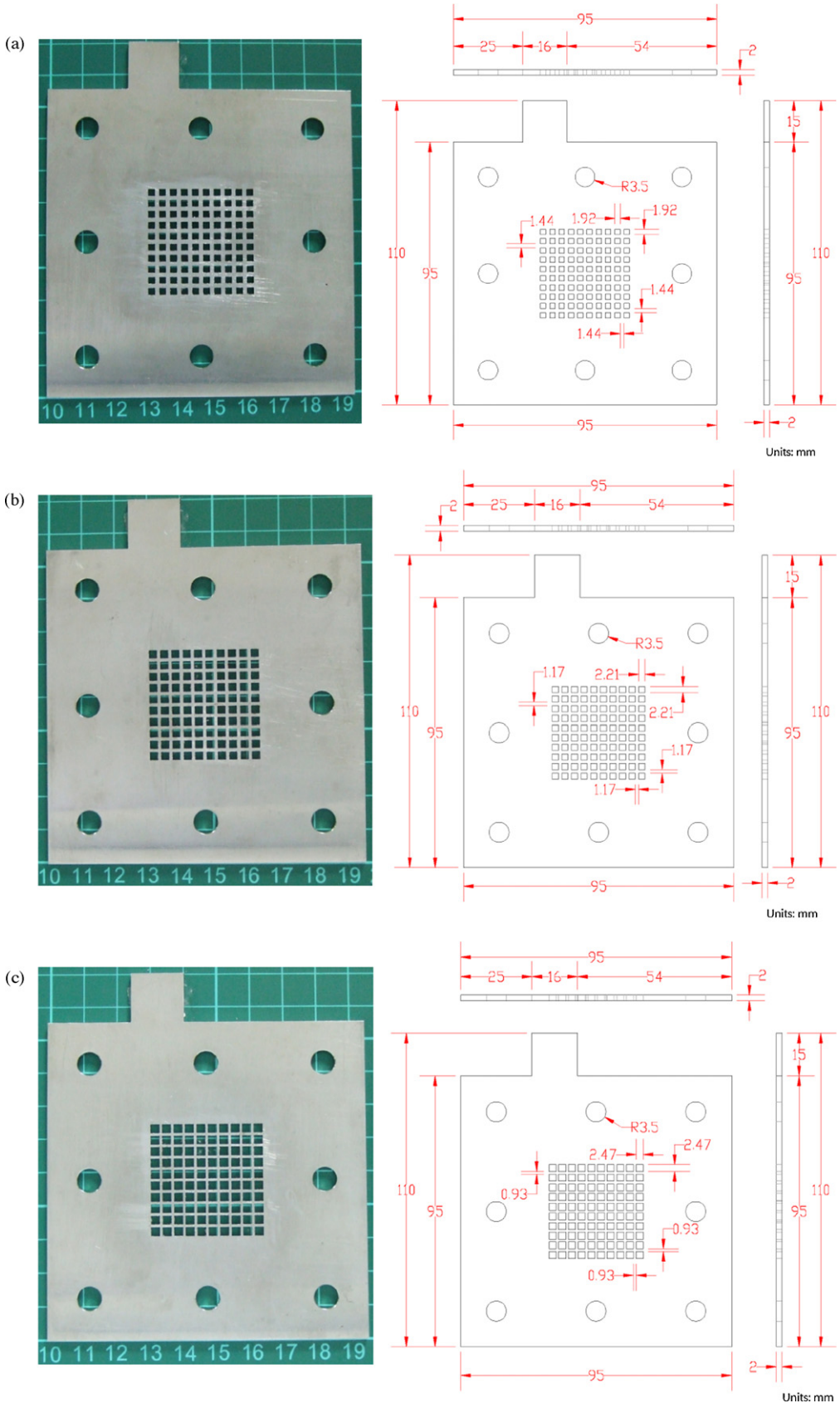

Fig. 10. Current collectors with a 10×10 hole arrangement: (a) 30% total free open ratio, (b) 40% total free open ratio, (c) 50% total free open ratio, and (d) 60% total free open ratio.

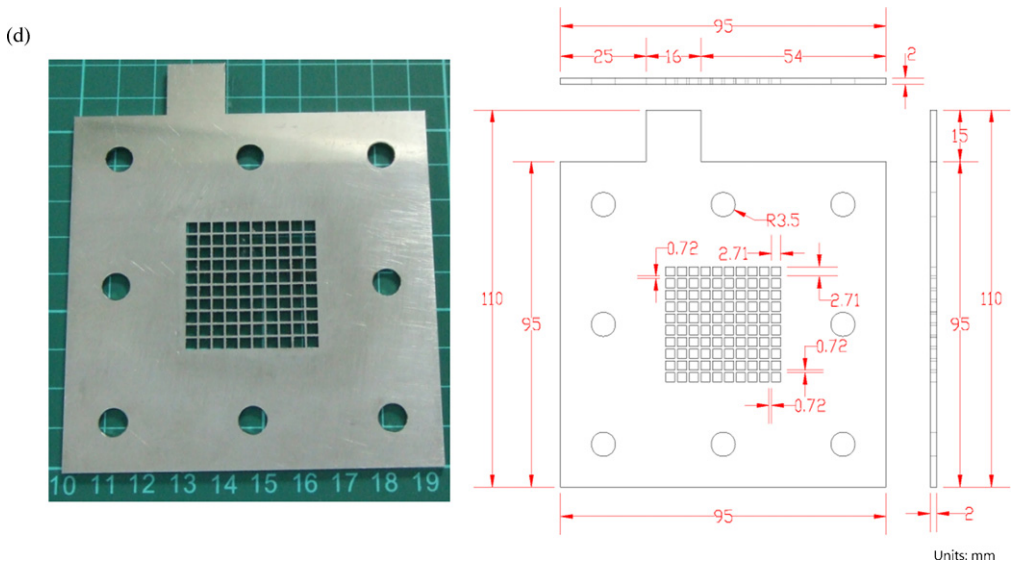

Fig. 10. (*Continued*)

Based on the above observations, several conclusions can be made about the design of current collectors. When the total perimeter length of openings on the current collectors is short, the diffusion distances from the free open areas to the closed area behind the current collectors are longer the replenishment of reactants and the discharge of products are thus slower. For this reason, the cell performance is low. Increasing the total free open ratio of the current collectors would help to increase cell performance because a higher total free open ratio could increase the mass trans-

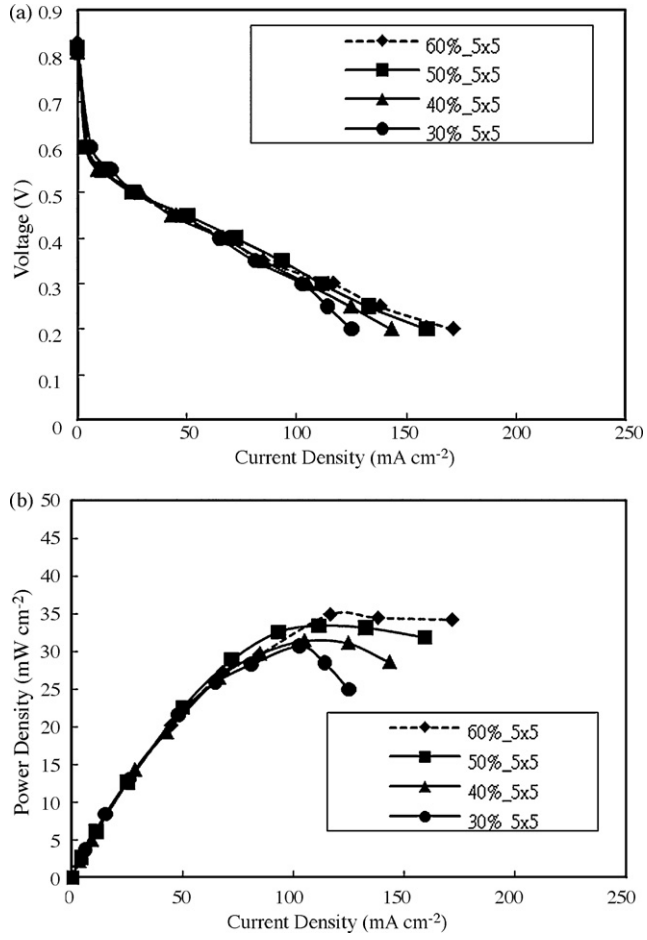

Fig. 11. Performance comparison of the DMFC with different total free open ratios on the current collectors with the same 5 × 5 hole arrangement: (a) *I–V* curves and (b) *I–P* curves.

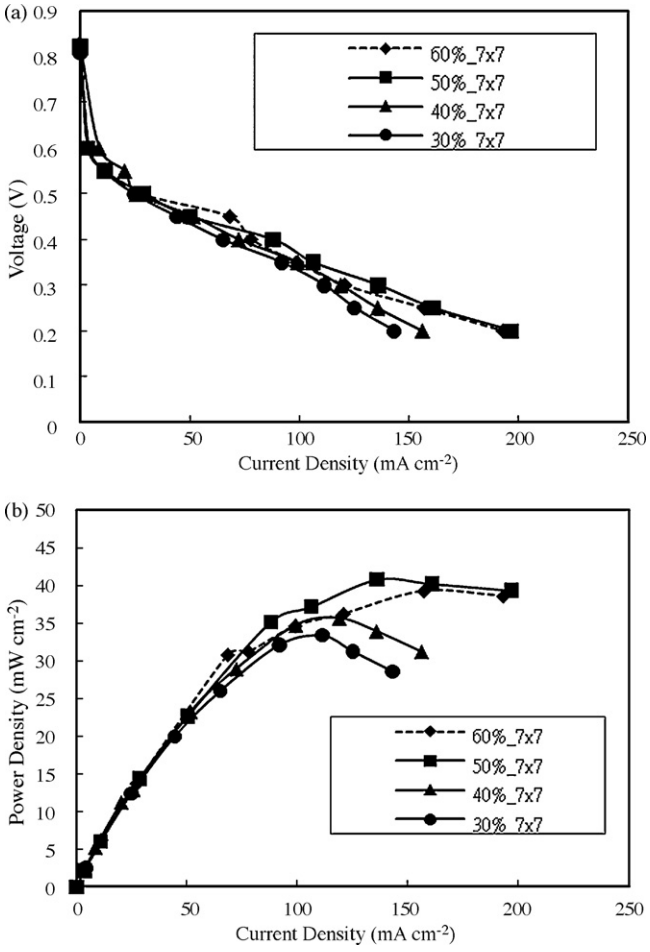

Fig. 12. Performance comparison of the DMFC with different total free open ratios on the current collectors with the same 7 × 7 hole arrangement: (a) *I–V* curves and (b) *I–P* curves.

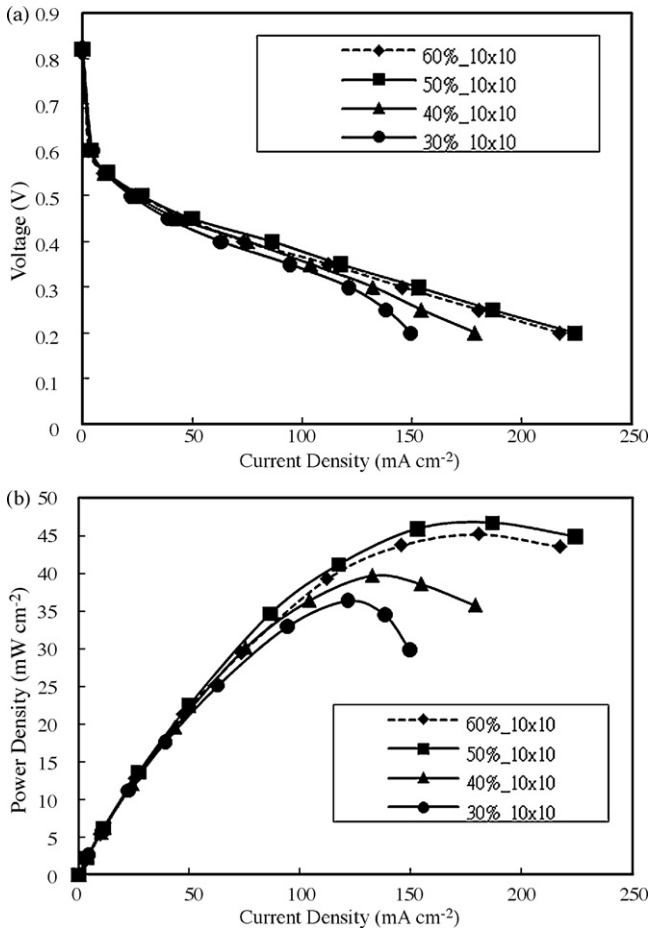

Fig. 13. Performance comparison of the DMFC with different total free open ratios on the current collectors with the same 10×10 hole arrangement: (a) *I–V* curves and (b) *I–P* curves.

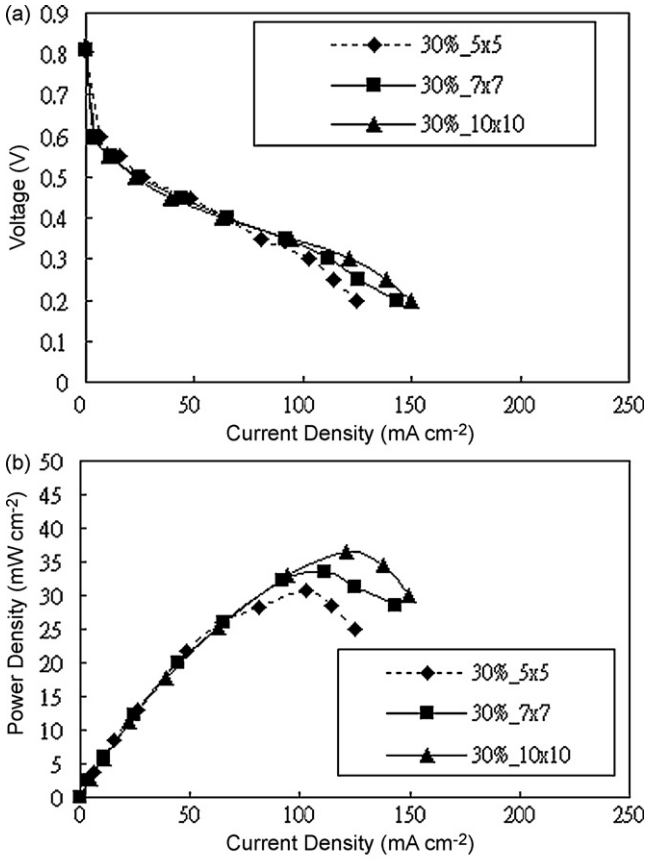

Fig. 14. Performance comparison of the DMFC with different total perimeter lengths of openings on the current collectors with the same 30% total free open ratio: (a) *I–V* curves and (b) *I–P* curves.

fer and thereby facilitate the diffusion of the methanol solution into the anode electrode surface and the diffusion of air into the cathode electrode surface. In addition, a higher total free open ratio of the current collectors provides a more efficient vent path to exhaust the produced CO_2 gas bubbles on the anode side and drain the produced water on the cathode side. However, a low total free open ratio of the current collectors may lead to insufficient diffused reactant at both the anode and cathode sides, bubble blocks at the anode side, and water flooding at the cathode side, thereby degrading cell performance. On the other hand, space for the reactants to flow and the products exhaust of electrochemical reactions at the anode and cathode would strongly affect cell performance when the total perimeter length of the openings of the current collectors is low. A larger space would lead to higher cell performance. Once the total perimeter length of the openings increases to a certain value, increasing the total free open ratio could also increase cell performance in the low total free open ratio range of the current collectors (below 50%). In the high total free open ratio range of current collectors (above 50%), further increases in the total free open ratio would not increase cell performance; on the contrary, it might slightly decrease cell performance. When the diffusion distances of the reactants and products from the closed areas behind the current collectors to the free open areas are sufficiently short, there would no longer be an obvious obstruction of the diffusions of reactions and products. The total free open ratio of the current collectors becomes the most dominant factor affecting cell performance. Although increasing the total free area ratio of the current collectors would increase the mass transfers of the reactants and products, methanol crossover also increases because of the larger permeability of the current collectors, and the contact resistance will increase due to the smaller contact area between the MEA and the current collectors [23]. Therefore, further increases in the total free open ratio under the high total free open ratio range of current collectors (higher than 50%) would not increase the cell performance and might slightly decrease the cell performance.

4.2. Current collectors with the same total free open ratio but different total free opening perimeter lengths

Fig. 14a and b shows the comparison of the *I–V* and *I–P* curves for a DMFC with different total perimeter lengths of openings on current collectors and the same 30% total free open ratio. The results show that the order of cell performance corresponds to $10 \times 10 > 7 \times 7 > 5 \times 5$ hole arrangements of the current collectors. A longer total free opening perimeter length on the current collectors gives better cell performance.

Fig. 15a and b shows the comparisons of the *I–V* and *I–P* curves for DMFCs with different total perimeter lengths of openings on current collectors with the same 40% total free open ratio. The results show that the order of cell performance corresponds to $10 \times 10 > 7 \times 7 > 5 \times 5$ hole arrangements of the current collectors. A longer total free opening perimeter length on the current collectors gives better cell performance.

Fig. 16a and b shows the comparisons of the *I–V* and *I–P* curves for DMFCs with different total perimeter lengths of openings on current collectors with the same 50% total free open ratio. The results show that the order of cell performance corresponds to $10 \times 10 > 7 \times 7 > 5 \times 5$ hole arrangements of the current collectors. A

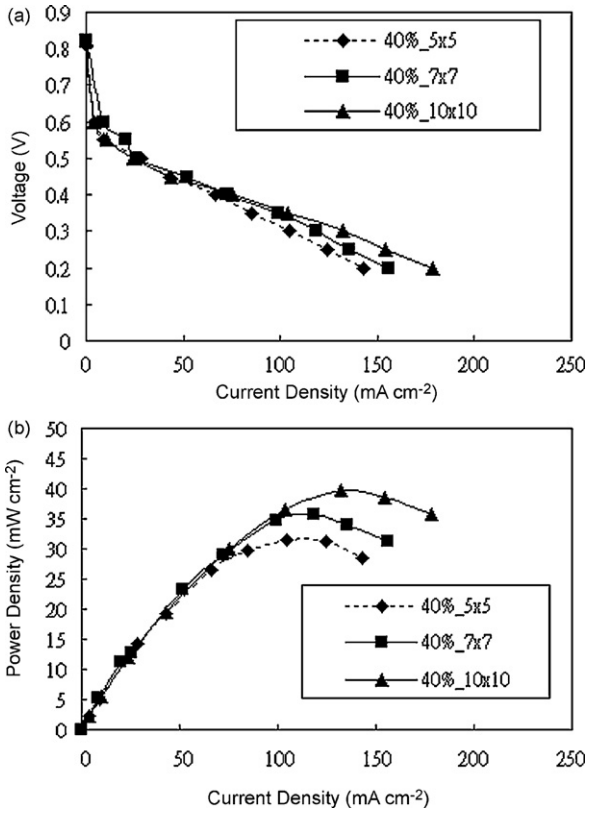

Fig. 15. Performance comparison of the DMFC with different total perimeter lengths of openings on the current collectors with the same 40% total free open ratio: (a) *I–V* curves and (b) *I–P* curves.

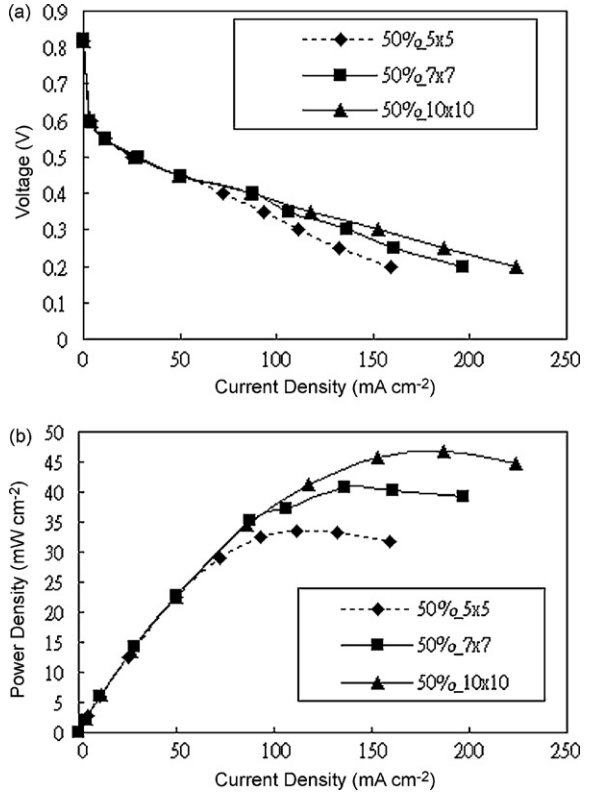

Fig. 16. Performance comparison of the DMFC with different total perimeter lengths of openings on the current collectors with the same 50% total free open ratio: (a) *I–V* curves and (b) *I–P* curves.

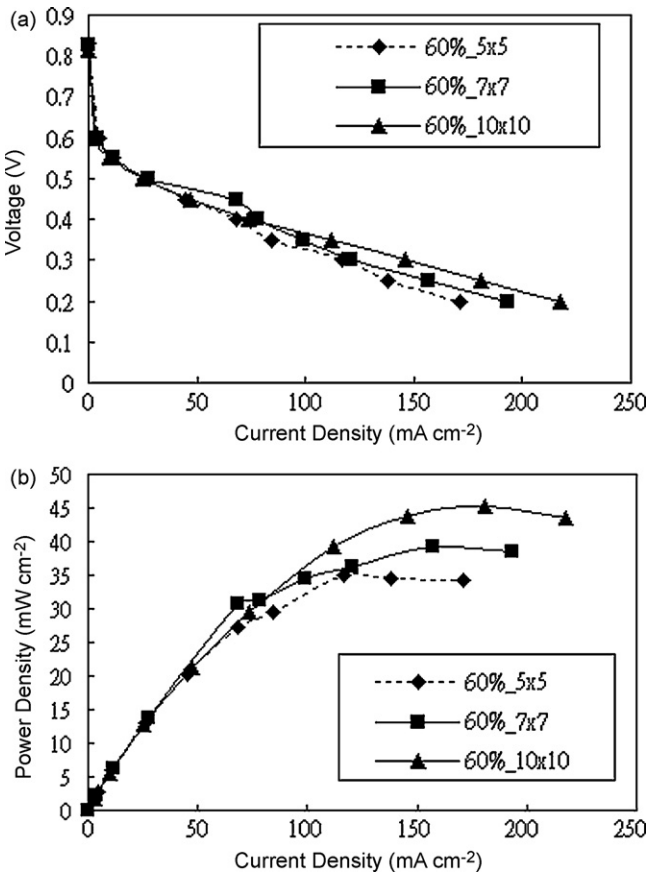

Fig. 17. Performance comparison of the DMFC with different total perimeter lengths of openings on the current collectors with the same 60% total free open ratio: (a) *I–V* curves and (b) *I–P* curves.

longer total free opening perimeter length on the current collectors gives better cell performance.

Fig. 17a and b shows the comparisons of the *I–V* and *I–P* curves for DMFCs with different total perimeter lengths of openings on current collectors with the same 60% total free open ratio. The results show that the order of cell performance corresponds to $10 \times 10 > 7 \times 7 > 5 \times 5$ hole arrangements of the current collectors. A longer total free opening perimeter length on the current collectors gives better cell performance.

According to the above observations, increasing the total perimeter length of openings increases cell performance with the same total free open ratio of the current collectors. The same total free open ratio of the current collectors indicates the same area for the mass transfer of the reactants and products as well as the same contact area between the MEA and current collectors. Increasing the total perimeter length of openings would shorten diffusion lengths from the free open areas to the closed area behind the current collectors for the reactants and products. This aids in the diffusion of the reactants and products and improves cell performance.

Therefore, a proper total free open ratio, such as 50%, of the current collectors and a proper adjustment of the hole arrangement to increase total perimeter length of openings, such as a 10×10 array, are recommend in the design of current collectors for planar-type DMFCs.

5. Conclusions

In this work, the effect of two important factors in the design of collectors for planar-type DMFCs, the total free open ratio and

the total perimeter length of the free openings, was studied by comparing different hole arrangements on the current collectors. In the low total free open ratio range, the cell performance was low, and increasing the total free open ratio significantly improved the mass transfer of reactants and products such that cell performance was increased. In the higher total free open ratio range, the contact area between the MEA and current collectors becomes important, and further increases in the total free open ratio might not increase cell performance because the higher total free open ratio leads to a higher contact resistance due to lower total contact area between the MEA and current collectors. It also increases the methanol crossover due to the larger permeability of the current collectors. Under the same total free open ratio, a longer total perimeter length of openings always gives better cell performance because the longer total perimeter length of openings shortens the diffusion length between the free open areas and the closed area behind the current collectors for the reactants and products, and mass transfers are smoother. The correct total free open ratio and total contact area should be carefully considered in the design of current collectors on planar-type DMFCs. A longer total free opening perimeter length is also suggested. For the present cases, a 50% total free open ratio and a longer total free opening perimeter length with a 10×10 hole arrangement are recommended.

Acknowledgement

The authors would like to acknowledge financial support from the National Science Council of Taiwan, ROC (NSC97-2221-E-167-010 and NSC 98-3114-E-167-001).

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