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RESEARCH PAPER

A novel scrape-applied method for the manufacture of the membrane–electrode assembly of the fuel–cell system

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Abstract This study investigates the transfer of the scrape-applied method from the electrodes of a lithium battery to the membrane–electrode assembly of fuel cells, including Proton Exchange Membrane Fuel Cells and Direct Methanol Fuel Cell. Three methods are commonly used to manufacture lithium battery electrodes: the roller-applied method, the spraying-applied method, and the scrape-applied method. This study develops novel scrape-applied equipment for lithium battery electrodes. This method is novel and suitable for producing fuel cell, better than other traditional methods. In this study, the stability of coating process was tested by measuring the weight and thickness of a dry electrode. The stability and reproducibility of electrode fabrication were examined by systematic data analysis. Finally, the study used a specially designed single cell composed of 16 conductive segments, which are insulated locally. The current passing through each segment was measured using Hall Effect sensors connected to the segment compartments. Based on the measured distribution of the local current in a segmented single cell, the influence of flooding and stoichiometry variation of feed gas was discussed in terms of electrochemical reaction rate. The experimental results serve as an important basis for future research in this field, which hold potential benefits to the academia and the industry.

Keywords Fuel cells · Scraper · Electrode · Lithium battery · Reliability

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1 Introduction

Technology transfer plays a pivotal role in the development of fuel cell systems. For instance, the technology used in manufacturing electrodes for lithium battery systems can be transferred to fuel cell systems [1,4]. Fuel cells are manufactured using three methods: the roller-applied method, the scrape-applied method, and the spraying-applied method (Table 1). On the other hand, the technology used to manufacture lithium batteries is very mature. The electrode is the primary part of a lithium battery. The influence of scrape-applied technology and modified equipment on electrode thickness and weight has been previously studied [5]. An improvement in equipment and reliability for scrape-applied technology has also been discussed. Meanwhile, the current work focuses on the scrape technique, which is currently the most common method for manufacturing fuel cells (Table 2).

To make an electrode, the powder must be mixed uniformly in a solution. However, the powder does not typically disperse well and may form larger particles. Therefore, scrape-applied methods are usually preferred.

The roller-applied method involves the use of an up-roller and down-roller (Fig. 1).

The gap between a pair of spinning rollers can control dispersion thickness. However, during the process of rolling coating, a Gas Diffusion Layer (GDL) can be rolled using the roller method. Therefore, this may be a significant problem when utilizing this method.

The scrape-applied method involves the use of a scraper (Fig. 2).

The direction of the scraper controls the gap between the plate and the scraper. However, membrane thickness typically increases due to surface tension and the concentration variation of the solution for the traditional applicator coater of the solution used by conventional applicator

Table 1 Advantages and disadvantages of the coating method

	Advantages	Disadvantages
Roller-applied method	Double-faced spread simultaneously to coating	Catalyst is affected by particle size
Scrape-applied method	Cheap; any particle can be used	Single-faced spread
Spraying-applied method	Fast	Single-faced spread; Catalyst is affected by particle size

Table 2 Three types of electrode technology in this study

	Roller	Spraying	Scrape
Lithium electrode	•		•
PEMFC electrode		•	•
DMFC electrode		•	•

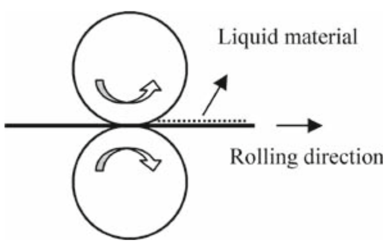


Fig. 1 Roller-applied method

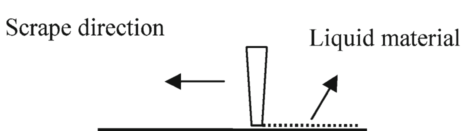


Fig. 2 Scrape-applied method

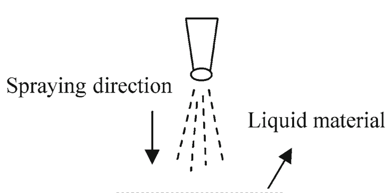


Fig. 3 Spraying-applied method

coaters. Therefore, this work uses a dual scraper to stabilize the manufacturing process. The spraying-applied method involves the use of a sprinkle nozzle (Fig. 3).

The spraying force and velocity can control the thickness of surface tension. However, in conventional spraying coating, the nozzle design and pressure difference between the aperture size and the quantity of the catalyst cause local current non-uniformity and adversely affect the entire electrode current density output.

These three manufacturing methods are very mature and can easily control electrode thickness and weight. However, only the scrape-applied method is unaffected by particle size. Thus, it is the most popular method for manufacturing electrodes.

The quality of the electrode can be measured by the current distribution. Nevertheless, there are a few studies that examined the current distribution within a single Proton Exchange Membrane Fuel Cell (PEMFC) [6,8]. Thus, directly measuring the local current distribution can provide a reference for the designs of elements, cell stacks, and systems in Proton Exchange Membrane Fuel Cells (PEMFCs). Correspondingly, the purpose of this study is to identify the characteristics of the electrochemical reactions within a single PEMFC under various conditions. A single cell is designed to measure the current distribution within it. Therefore, this study can determine the effects of electrochemical reaction speed on different factors such as the structures of flow channels and operating voltages.

2 Experimental design

Manufacturing a single fuel–cell electrode

The single fuel–cell electrode in this study has a standard dimension of 25 cm^2. The scrape-applied method was used. Figure 4 shows the schematic layout of the schematic apparatus used in the scrape-applied method.

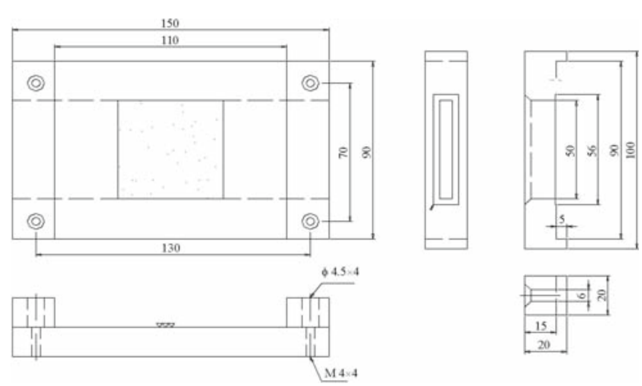


Fig. 4 Schematic apparatus of the scrape-applied method

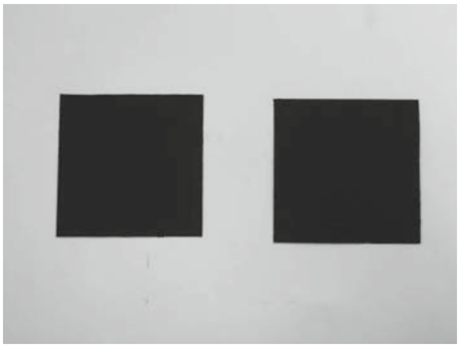


Fig. 5 Finished electrodes of the fuel cell

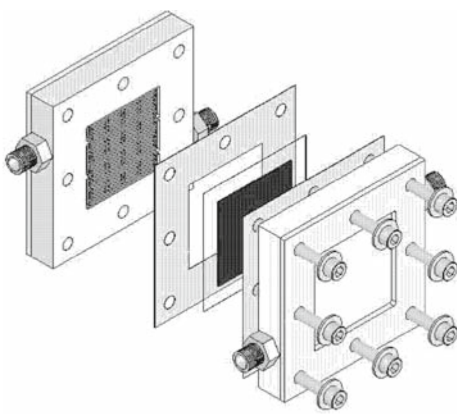


Fig. 6 The schematic assembly of the single fuel cell

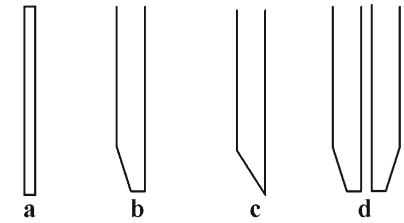


Fig. 7 Traditional types of scraper

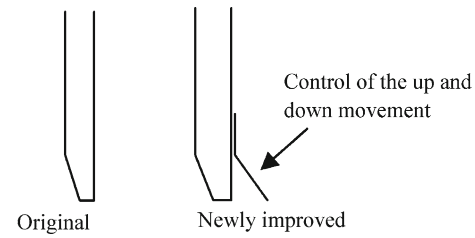


Fig. 8 The original and newly improved scraper

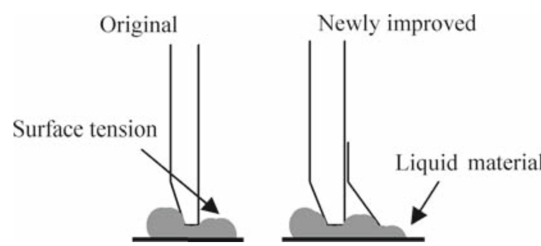


Fig. 9 Flow phenomenon of the coating

The experiment used a standard test area of 25 cm^2 in a single cell, the coating method, and a catalyst coated onto a hydrophilic treated carbon paper with a coating blade (ratio, XC72-Pt/C:Nafion is 1:3). The catalytic XC72-Pt/C was coated onto the hydrophobic carbon paper (or cloth). When the catalytic mixture was too thick, Isopropyl Alcohol (IPA) was added. When the mixture was too thin, the mixing time was extended. After the coating was air-dried, GDL was then dried in a high-temperature furnace. Figure 5 shows the finished products.

After cooling to room temperature, the catalyst on the carbon paper was weighed (mg/cm^2). This electrode and the quality of the leading membrane should pass the hot-pressing (135°C/90s) test. The layers are in the following order: (1) glass fiber, (2) metal bipolar plate, (3) leak-proof spacer, (4) membrane–electrode assembly (MEA), (5) leak-proof spacer, (6) metal bipolar plate, (7) and glass fiber board (6). In this structure, the curing flow system is inside the metal bipolar plate. All metals are stainless steel 316. After carving the curing flow system into the surface of these materials, a layer of titanium oxide (TiO_2) was added to prevent corrosion. The width and depth of the curing flow system are 1mm. The response area is 5cm×5cm.

2.1 The scraper's design

Figure 7 shows several traditional scrapers. Uneven coating thickness and stickiness can usually influence this coating. The purpose of the new scribble is to control mixture thickness.

Figure 8 presents a modified shaves board. An additional slide board, which can move up and down, is attached to the scraper's back. Figure 9 shows the utilization of the flow phenomenon of the fluid and the liquid tension principle. Thus, the mixture thickness can be controlled easily, and the manufacturing quality can be steadily improved as well.

2.2 Data analysis

When the coating dried, the weight of the electrode and the thickness of the electrode membrane were measured. The quality and uniformity of the coating were also examined using the current distribution. Figure 10 shows the 3D images of the single-cell 3D assembly diagram. Finally, the electrical properties of MEA were measured, and its performance was compared with that of traditional MEA.

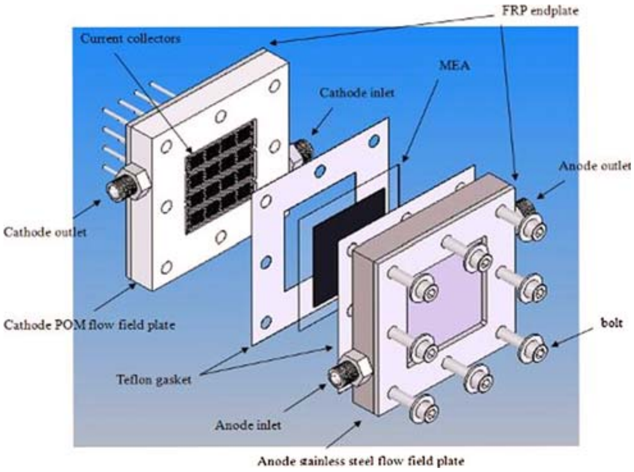


Fig. 10 Single-cell 3D assembly diagram

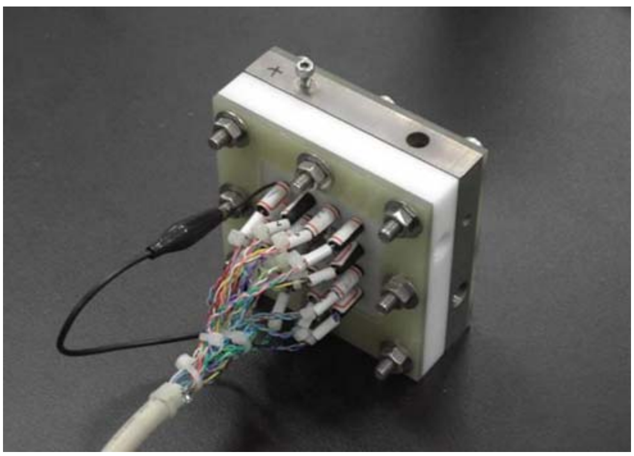


Fig. 11 Local current density distribution measure mental

The assembled single cell was installed on a test bench. Conducting wires, which were prepared in advance, were connected to the current collector composed of the 16 stainless steel. Finally, the current is transported to the device for measuring the current comprised of 16 Hall ampere meters (Fig. 11).

3 Reliability analysis

After assembling the fuel cell electrode, the average values of the newly improved scraper were obtained. Figures 12 and 13 show the calculation results for stability, weight, and thickness. Figures 14, 15 and 16 show the characteristic curves for the PEMFC system, while Fig. 17 shows the characteristic curves for the DMFC system.

The formulas for calculating the mean value ($\bar{w}$) and root mean square (σ_x) are

$$\bar{w} = \frac{\sum_{j=1}^{n} w_j}{n}, \tag{1}$$

$$\sigma_x = \sqrt{\frac{(w^1 - \bar{w})^2 + (w^2 - \bar{w})^2 + \cdots + (w^n - \bar{w})^2}{n}}. \tag{2}$$

Figure 12 shows the performance data for the electrical properties of the traditional scraper. Figures 13, 14 and 15 present the data for the electrical properties of the modified scraper.

4 Results and discussion

This experiment measures the variations in cell performance and current distribution by changing the operational coefficients of the single cell and the flow rates of the reactive gases. Additionally, the experiment determines the current distribution in the flow field and the effects of the liquid coating over

Fig. 12 Comparison of coating weight. **a** Original; **b** newly improved

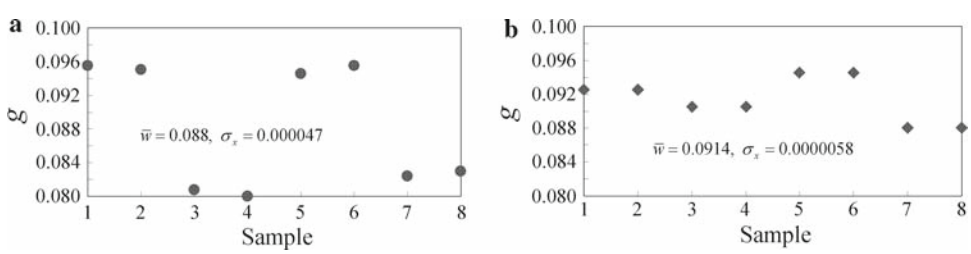
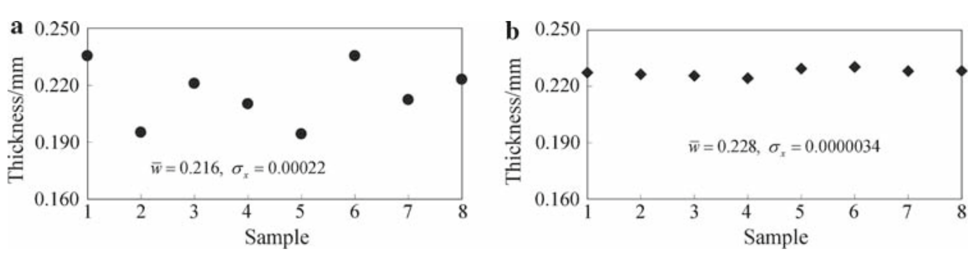


Fig. 13 Comparison of coating thickness. **a** Original; **b** newly improved



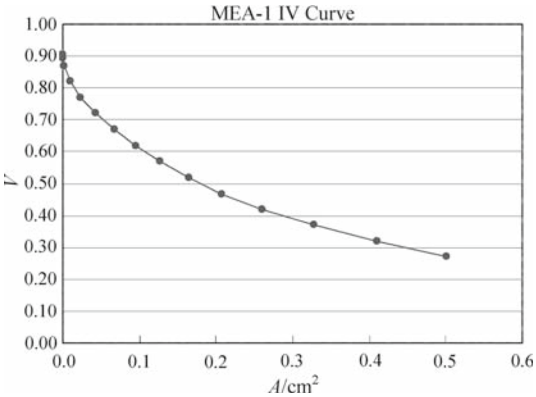


Fig. 14 IV Curve of the PEMFC electrode with the original scraper (Gas: H_2/O_2)

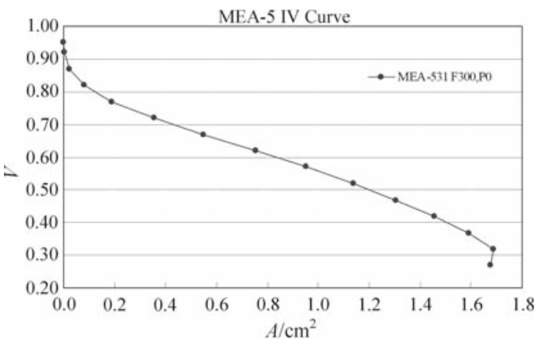


Fig. 15 IV Curve of the PEMFC electrode with the newly improved scraper (Gas: H_2/O_2)

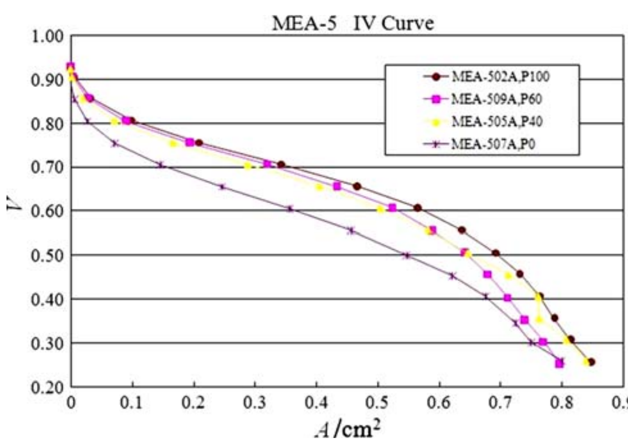


Fig. 16 IV Curve of the PEMFC electrode with the newly improved scraper (Gas: H_2/air)

time. Figures 18, 19, 20 and 21 show the 2D pictures of the measurement results.

The flow-field plate at the cathode is made of Polyoxymethylene (POM) infixed 16 isolated stainless-steel current collectors, each 9 mm × 9 mm × 12 mm (L × W × H). These stainless-steel current collectors are inserted into an engineering plastic frame, each separated by 2.5 mm. An

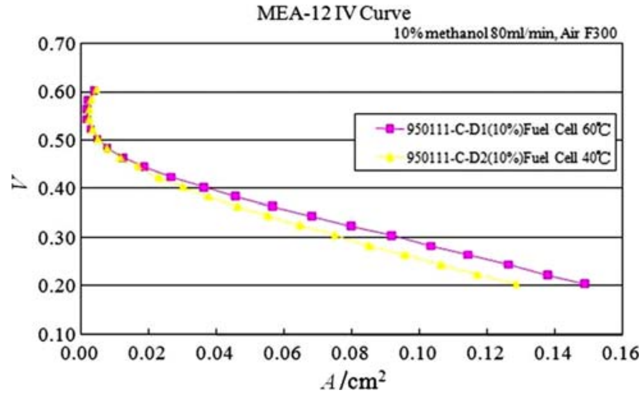


Fig. 17 IV Curve of the DMFC electrode with the newly improved scraper

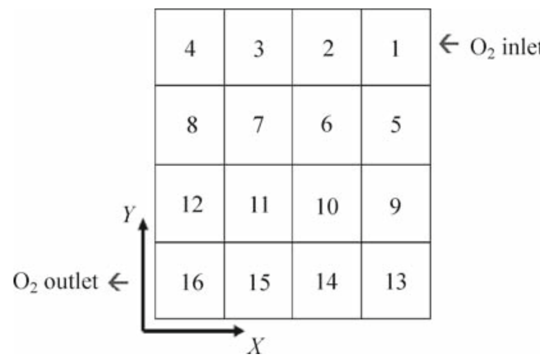


Fig. 18 Measurement position diagram

anaerobic adhesive fills the gaps between the frame and the stainless steel collectors for leak prevention. When the adhesive was dry, the flow channels were carved into the surface of the flow field plate using a Computer Numerical Control (CNC) machine. The flow channels were 1 mm wide and deep, and the reactive surface was 5 cm × 5 cm = 25 cm^2.

5 Conclusions

The technology of a newly improved scraper was successfully transferred from a lithium battery electrode to the MEA of fuel cells, including PEMFCs and Direct Methanol Fuel Cell (DMFCs).

(1) This process can solve the liquid surface tension problem and improve the stability of the manufacturing process.
(2) As we can see from Figs. 19, 20 and 21, the current distribution is different.
(3) The catalyst is making substantially great difference in electric current (Fig. 21). Among electric current causes, some hot reactions have very obvious

Fig. 19 Roller-applied method of the measurement position 3D diagram

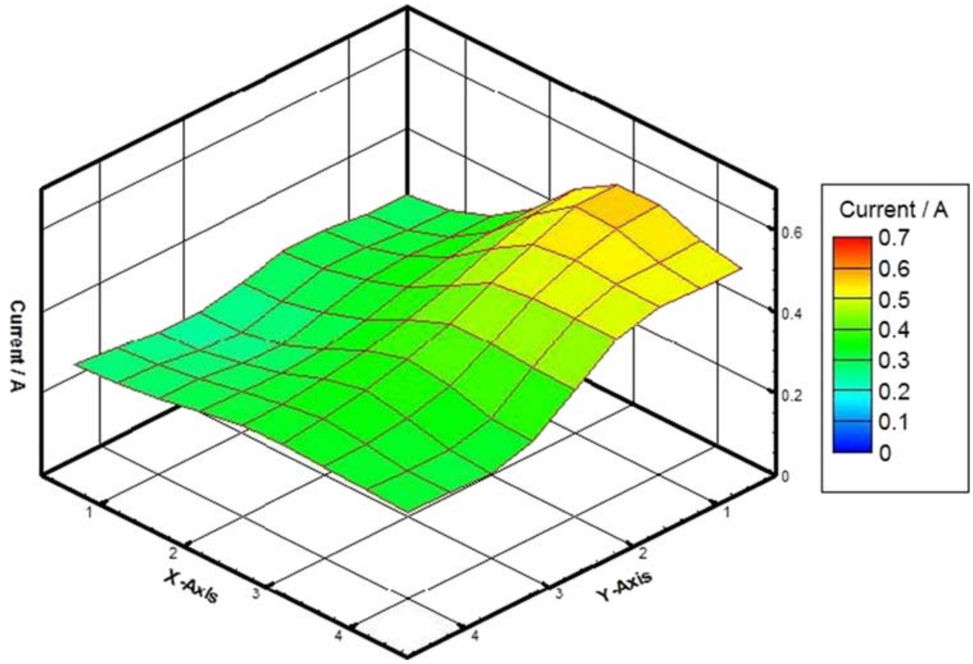
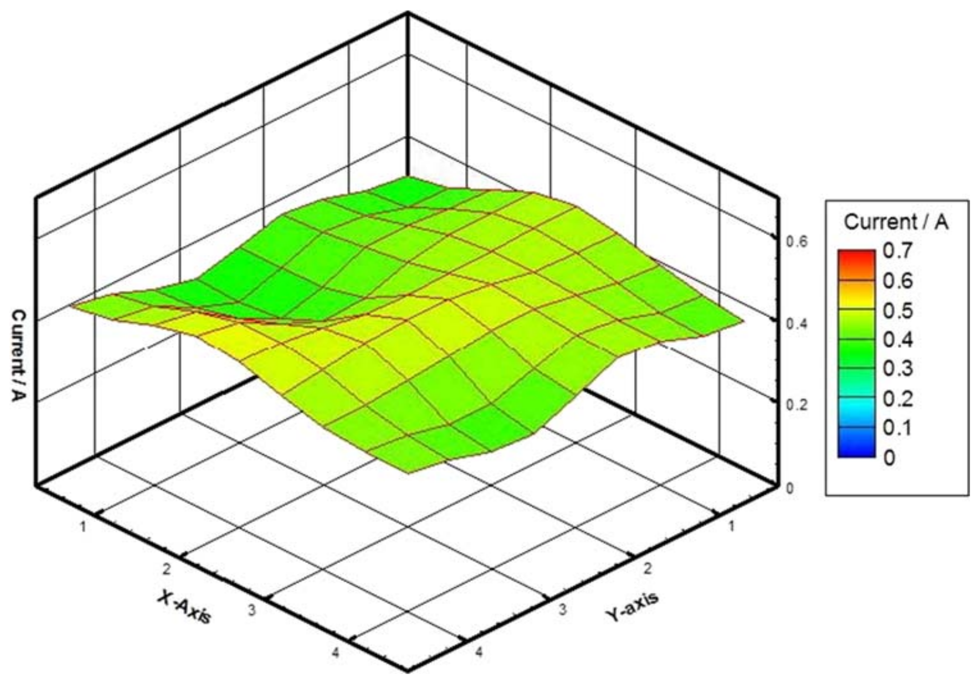


Fig. 20 Scrape-applied method of the measurement position 3D diagram



differences. The imbalance heat on the electrode will influence life-span in the fuel cell.

(4) The newly improved scraper is steady in terms of current density distribution (Fig. 20).

(5) The newly improved scraper can stabilize the electrical current, which is compatible with the proposed scrape method (Fig. 20).

(6) The roll and scribble methods are only suited to small areas.

(7) Local current distribution testing adversely affects local temperature and reduces membrane lifetime.

(8) The production equipment will be designed according to the research results in order to enhance MEA performance and reduce production costs.

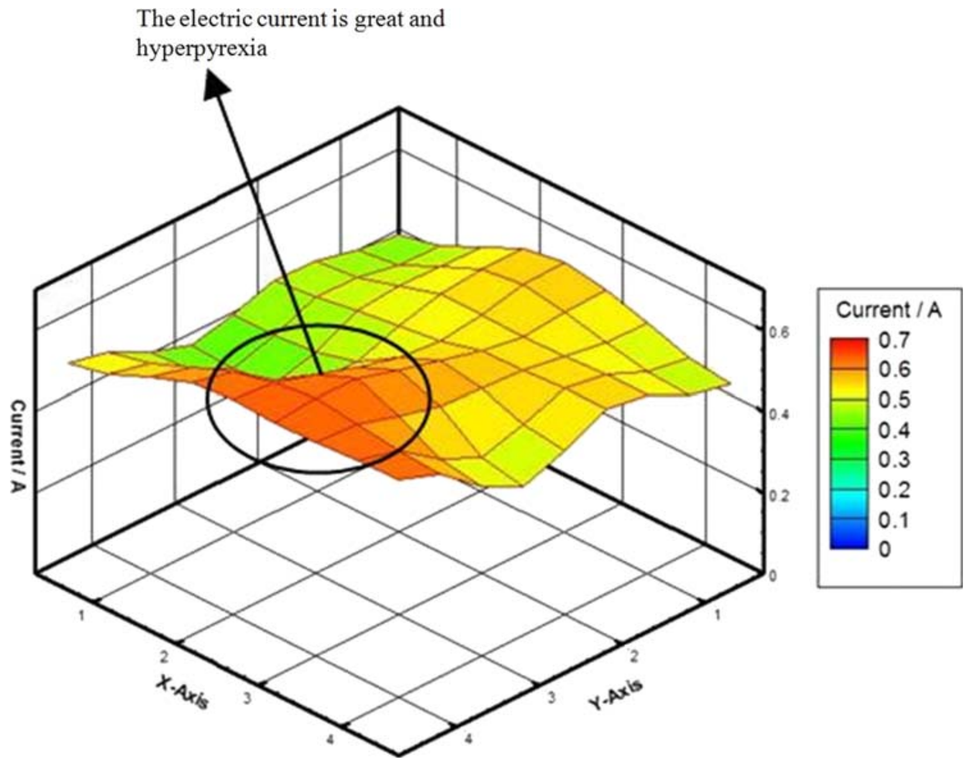


Fig. 21 Spraying-applied method of the measurement position 3D diagram

(9) The good reliability of commercial products can be achieved through the newly improved scraper.

(10) The modified scraper has good stability in the manufacturing process.

(11) The spray coating system will be improved to accommodate different processing designs.

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