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Performance of PEMFC stack using expanded graphite bipolar plates

Xiqiang Yan [a,b,*], Ming Hou [a], Haifeng Zhang [a], Fenning Jing [a], Pingwen Ming [a], Baolian Yi [a]

[a] *Fuel Cell R&D Center, Dalian Institute of Chemical Physics, Chinese Academy of Sciences, Dalian 116023, China*
[b] *Graduate University of the Chinese Academy of Sciences, Beijing 100039, China*

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Abstract

The bipolar plates are in weight and volume the major part of PEM fuel cell stack, and also a significant effect to the stack cost. To develop the low-cost and low-weight bipolar plate for PEM fuel cell, we have developed a kind of cheap expanded graphite plate material and a production process for fuel cell bipolar plates. The plates have a high electric conductivity and low density, and can be stamped directly forming fuel cell bipolar plates. Then, 1 and 10 kW stacks using expanded graphite bipolar plates are successfully assembled. The contact resistance of the bipolar plate is investigated and the electrochemical performances of the fuel cell stacks are tested. Good fuel cell performance is obtained and the voltage distribution among every single cell in the stacks is very uniform.

Keywords: PEMFC; Expanded graphite; Bipolar plate

1. Introduction

The proton exchange membrane fuel cell is an optimal alternative in future vehicle application owing to its properties of high power density and energy conversion efficiency. In addition, it is an environment-friendly power source. Presently there are still some challenges for the commercialization of PEMFC, one of the main obstacles is the quite high manufacture cost of the fuel cell stack. The two key components of fuel cell stack are bipolar plates and MEAs that cost more than 90% of total cost. Therefore, greatly improving performance of these two components and decreasing the cost of manufacture materials are the crucial problems of fuel cell commercial application. The cost target of bipolar plates for fuel cell vehicles is established to be US$ 10 kW^{-1} [1].

Currently, challenges for commercial applications of PEMFC include reducing the cost and weight of the fuel cell stack [2]. An integrant part of the PEM fuel cell stack is the bipolar plates, which accounts for about 80% of total weight and 45% of stack cost [3]. So the critical topic of bipolar plate study is to develop the lighter and lower cost bipolar plates. There are many kinds of bipolar plates for fuel cells, the most commonly used bipolar plate materials are metal and carbon. Sheet metal is a potential candidate for bipolar plate material since it has good mechanical stability, electrical conductivity and thermal conductivity and can be easily stamped to desired shape to accommodate the flow channels. However, as bipolar plates, they are exposed to an operating environment with a pH of 2–3 at temperatures of around 80 °C [4], and prone to corrosion or dissolution. On the other hand the dissolved metal ions may result in the poisoning of PEM membranes and hence lower the ionic conductivity. Moreover, a corrosion layer on the surface of a bipolar plate increases the electrical resistance and decreases the power output of the cell. To solve these issues, metal plates coated with a protective layer have been studied to improve the properties in the environment of PEMFC operating [5–8]. As the other candidates to meet the requirements of PEMFC bipolar plate, carbon composites have been developed extensively [9–16]. Graphite plate that based on carbon material is widely used in fuel cell bipolar plates because it has excellent chemical stability in the fuel cell environment. However, the cost of material and manufacturing of traditional graphite bipolar plate is very high and there is no method to decrease it obviously [17,18].

Because the material and manufacturing costs of the traditional graphite plates are not the best choice at least for automotive applications, extensive efforts have been made to develop alternative bipolar plate materials, including metallic

* Corresponding author. Tel.: +86 411 84379587; fax: +86 411 84379185.
E-mail address: xqyan@dicp.ac.cn (X. Yan).

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and graphite-based composite bipolar plates [12,13,19–21]. Graphite-based composite bipolar plates are made from a combination of graphite or carbon powder filler and a polymer resin with conventional polymer processing methods like compression molding or injection molding. They offer the advantages of lower cost, higher flexibility and greater ease of manufacturing than graphite plates. They are also light in weight compared to metallic and graphite plates. The gas flow channels can be molded directly into the plate, eliminating the need for the costly machining step.

In this article, we have developed a kind of bipolar plate made of expanded graphite plates. This expanded graphite plate material is porous and sandwich structure and it also has the properties of low cost, lightweight, good corrosion resistance and high electrical conductivity. The raw graphite ore is rapidly expanded when the expansion reagent joins, it can be pressed to different densities of expanded graphite plate [22]. Because the expanded graphite plate has not to be pretreated at high temperature the material manufacturing cost can be decreased much more, its price is US\$ 4–5 kg^{-1} in China market. On the other hand, the expanded graphite bipolar plate has predominance in the power density and the endurance of impacting vibration because it has lower density and high flexibility. Therefore, the expand graphite bipolar plates are worldwide investigated by fuel cell automotive developers. Ballard Power Systems Inc. has firstly investigated expanded graphite bipolar plates and applied in fuel cell buses. We have also developed the manufacture technology of expanded graphite bipolar plate, but our manufacture technique and material [23] is different from that of Ballard Power Systems Inc. [24]. We have successfully assembled 1 and 10 kW stacks using expanded graphite bipolar plates.

2. Experimental

2.1. Fabrication of expanded graphite bipolar plate

To prepare expanded graphite bipolar plates, a low viscosity thermosetting resin was used to impregnate the raw expanded graphite plates in vacuum for 10 min to improve the structural rigidity and to reduce gas penetration. Then the impregnated expanded graphite plates were dried in vacuum-stove. The degree and the time of heating will vary with the nature and amount of solvent, and is preferably from about 90 to 120 °C for about 10–30 min for this purpose. The flow channels for gas and cooling water were formed on both major surfaces of an impregnated expanded graphite plate by stamping processes. In this stamping process a hydraulic press machine and two moulds with flow channels were applied, the total time is not more than 2 min for a piece of plate. Finally, two pieces of stamping plates with flow channels were felted together forming an integrated expanded graphite bipolar plate [22]. The size of expanded graphite bipolar plate was 410 mm × 100 mm, the thickness of bipolar plate was about 3.0 mm. The density of bipolar plate is about 1 $g\,cm^{-3}$.

To prepare metal composite bipolar plates, 0.2 mm 316L stainless steel plate with surface treatment was adopted as supporting plate; the expanded graphite plate was adopted as flow field plate which was produced by compressing molding; cooling water flow field was also produced by compressed expanded graphite.

Before single cell tests, electrical contact resistance and water contact angles of graphite, metal composite and expanded graphite plates were measured. For comparison, graphite bipolar plates were prepared by machining flow channels.

2.2. Preparation of membrane electrode assembly (MEA)

For MEA preparation, catalyst ink was prepared by mixing the catalyst powders (40 wt.% Pt/C, E-TEK), Nafion® solution, and *iso*-propyl alcohol. Nafion® 112 was used as the proton exchange membrane and Toray® carbon paper was used as the gas diffusion layers. Then, the prepared catalyst ink was sprayed onto the gas diffusion layers with platinum loading of 0.3 and 0.5 $mg\,cm^{-2}$ for anode and cathode, respectively. Then, MEAs were fabricated by placing the catalyst-coated electrodes on both sides of pre-treated Nafion® 112 membrane, followed by hot pressing at 130 °C and 200 $kg\,cm^{-2}$ for 90 s. The electrode active area was 270 cm^2.

2.3. Tests of single cell with different kind of bipolar plates

Three single cells were assembled by different kinds of bipolar plates and the same MEAs. To operate the single cells, fully humidified hydrogen and oxygen were fed to the anode and cathode, respectively. Stoichiometry of the fuel and the oxidant was 1.5 and 2.5, respectively. Operating temperature and pressure were 60 °C and 30 kPa, respectively. Performances of three single cells were evaluated by measuring *I–V* curves using an electronic load (Arbin Instruments) and shown in Fig. 3. Before the measurements, the single cells were operated for 24 h for activation. To investigate degradation of the bipolar plates, lifetime of the bipolar plates were evaluated by measuring voltage at a current density of 500 $mA\,cm^{-2}$ as a function of operating time.

2.4. The assemblage and testing of a 1 and 10 kW class PEMFC stack

A 1 kW (16 single cells) and a 10 kW (130 single cells) PEMFC stacks were assembled using the expanded graphite bipolar plates and the MEAs which were fabricated by the technology mentioned above. The performance of these two stacks were tested on a high precision PEMFC testing equipment (FCATS-H36000, Hydrogenics, Canada), the experimental parameters of fuel, oxidant, operating temperature and pressure, stoichiometry of hydrogen and air were all same as those of the single cell. The relative humidity of the reactive gases in the PEMFC was accurately controlled by accurately controlled the dew point temperature of the reactive gases. Finally the performances of these stacks were evaluated by the obtained *I–V* curves.

3. Results and discussion

3.1. Material characterization

The contact resistance is an important factor affecting cell performance in fuel cell application since the contact resistance would have stronger effects on cell performance rather than bulk or surface resistance of the bipolar plate [25]. To evaluate electrical resistance of pure graphite, composite and expanded graphite, contact resistance including bulk resistance of a bipolar plate material and two gas diffusion layers and diffusion layers were measured. In this experiment the contact resistances of several kinds of bipolar plates were measured using an electro-omnipotent testing apparatus that was specialized to measure contact resistance all kinds of bipolar plate. To simulate the fuel cell stack condition, the contact resistance was measured by applying various compaction pressures and shown in Fig. 1. The result indicates that the contact resistance between bipolar plates and gas diffusion layers decreases rapidly when the pressure on the two sides of the bipolar plate increases in a relative low ($F < 50\,\mathrm{N\,cm^{-2}}$) scale, but when the pressure reaches to a certain degree ($F > 100\,\mathrm{N\,cm^{-2}}$) this contact resistance was hardly affected by the change of the pressure. As it can be seen in Fig. 1 the contact resistances between carbon paper (diffusion layer) and pure graphite bipolar plate, composite bipolar plate, expanded graphite bipolar plate are 13.94, 21.74, and 31.85 $\Omega\,\mathrm{cm^2}$ at the pressure of 140 $\mathrm{N\,cm^{-2}}$. The contact resistance of expanded graphite bipolar plate was the highest one, this result owing to the macromolecule polymer introduced into the expanded graphite bipolar plate during the preparation of the plate. However, the increase contact resistance did not affect the performance of the fuel cell in Fig. 3. These results show that, at the point of electrical contact resistance, expanded graphite bipolar plate can be applied to bipolar plates of PEMFC.

Surface energy of bipolar plates is another important factor to affect cell performance, particularly at high current densities since water produced by the cathode reaction should be properly removed. The bipolar plate with low surface energy is readily flooded at the cathode side of a cell [26]. To evaluate surface energy of the material, water contact angle is measured. The lower the contact angle is, the lower is the surface energy of the material. Fig. 2 exhibits water contact angle of expanded graphite plates is bigger than that of pure graphite plates. Therefore, this expanded graphite bipolar plate can be applied in PEMFC.

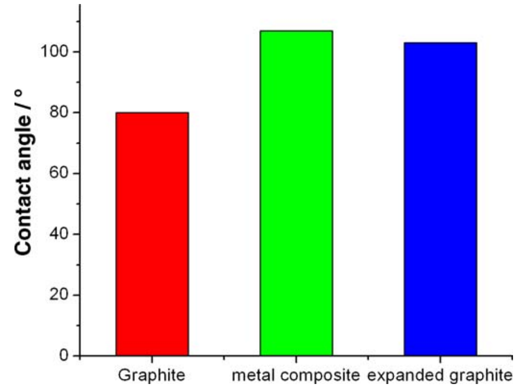

Fig. 2. Static water contact angle of graphite plate, metal composite plate, and expanded graphite plate.

To evaluate performance of expanded graphite bipolar plates, single cells have been fabricated using graphite, metal composite, and expanded graphite bipolar plates. Fig. 3 shows initial performance of the single cells with three different bipolar plates. The performance of the fuel cell using expanded graphite bipolar plate is slightly lower than the performance of fuel cell using pure graphite bipolar plate. This phenomenon can be explained by the higher contact resistance of the expanded graphite bipolar plate. The performance of metal composite bipolar plate fuel cell is the same as that of expanded bipolar plate fuel cell, which is owing to the surface oxidation of metal composite bipolar plate which results in the contact resistance increasing during the cell operating.

Fig. 4 shows the short-term performance of the fuel cell, the fuel cell is operated over 200 h at a constant current density 500 $\mathrm{mA\,cm^{-2}}$. The performance does not degrade obviously, but it is not enough for fuel cell vehicles. The life-time of fuel

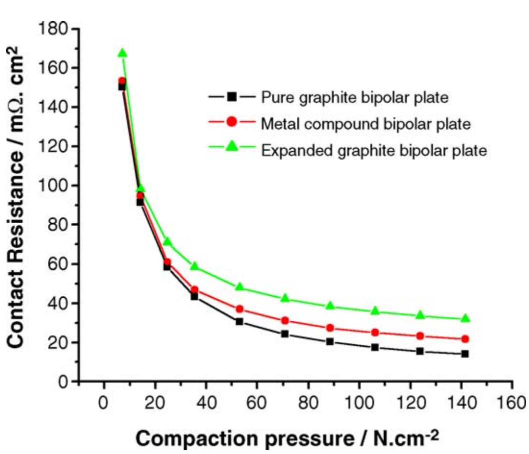

Fig. 1. Contact resistance of three kinds of bipolar plates measured at various compaction pressures.

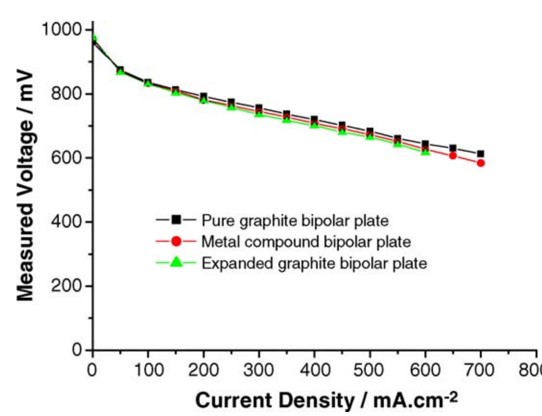

Fig. 3. *I–V* curves for the single cells using graphite, metal composite, and expanded graphite bipolar plates; operating temperature = 60 °C; operating pressure = ambient; $\lambda_{H_2} = 1.5$ and $\lambda_{air} = 2.5$; $RH_{H_2} = 60\%$; $RH_{air} = 80\%$.

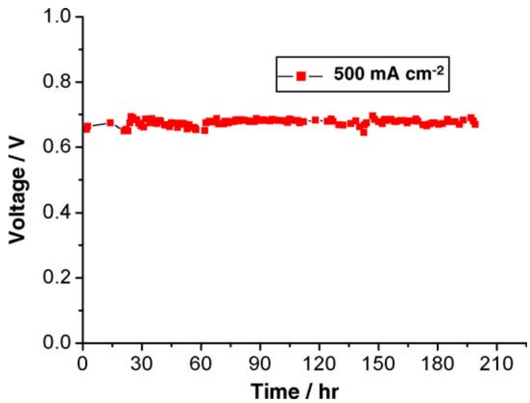

Fig. 4. Life-time test of the single fuel cell using the expanded graphite bipolar plate at a constant current density of 500 mA cm^{-2}.

cell should be over 5000 h if fuel cells are applied in vehicles. Therefore, the durability examination is needed.

3.2. Performance of 1 kW PEMFC stack

The new kind of bipolar plate for fuel cell must be validated by the performance of fuel cell. Therefore, a kW class PEMFC stack with 16 single cells is assembled using the expanded graphite bipolar plates to validate the feasibility of this kind of bipolar plate. The PEMFC stack is activated for 24 h at ambient pressure, then, the *I–V* curves of the stack is measured to estimate performance of the stack. The result indicates that the PEMFC stack assembled using the expanded graphite bipolar plate can be operated steadily. It can be seen in Fig. 5 that the output power can reach to 1.6 kW and the power density can exceed 0.37 W cm^{-2}. In addition, Fig. 6 gives the performance curves of the stack under different operating pressures, it can see that the power of the stack can reach to 2.4 kW and the power density can exceed 0.54 W cm^{-2} when the PEMFC stack runs under higher working pressure. These data indicate that the performance of this kind of PEMFC stack is very good and the expanded graphite bipolar plate is a nice choice for PEMFC.

The histogram in Fig. 7 shows the voltage distribution of every single cell in the 16-cell stacks when the stack is operated

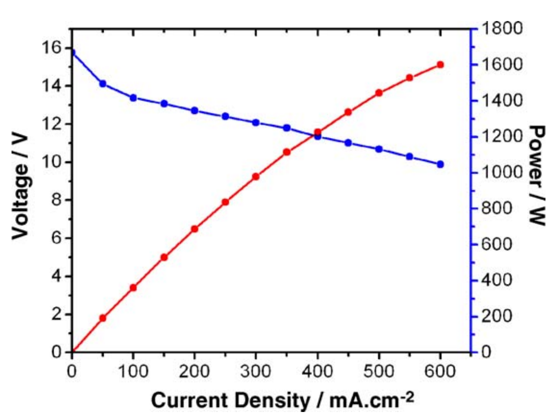

Fig. 5. The PEMFC performance curves of the 16-cell short stack with expanded graphite bipolar plates; operating temperature = 60 °C; operating pressure = ambient; λ_{H_2} = 1.5 and λ_{air} = 2.5; RH_{H_2} = 60%; RH_{air} = 80%.

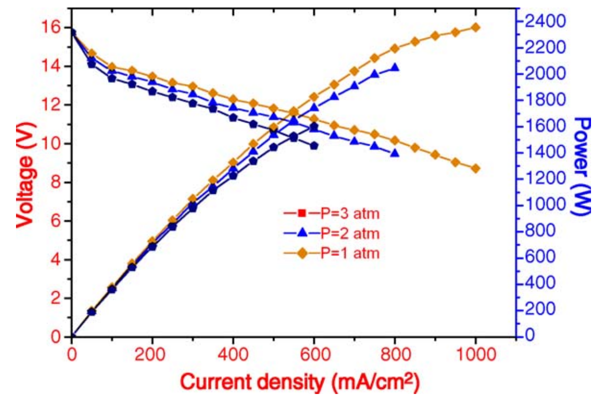

Fig. 6. The performance curves of the short expanded graphite bipolar plates PEMFC stack with 16 single cells at various operate pressures; operating temperature = 60 °C; λ_{H_2} = 1.5 and λ_{air} = 2.5; RH_{H_2} = 60%; RH_{air} = 80%.

at the current density of 500 mA cm^{-2}, the histogram indicates the performance of every single cell in this short stack is considerable uniform and the difference between the maximum and the minimum voltages of the single cells among the stack is less than 60 mV.

3.3. Performance of 10 kW class PEMFC stack

The feasibility of expanded graphite bipolar plates in fuel cell is validated by assembling a 10 kW class PEMFC stack with these plates, the stack has 130 single cells and the appearance of the stack is shown in Fig. 8. The performance of the stack and the uniformity of gases distributing on the bipolar plate among the stack is reflected by the following operating record of the stack.

Fig. 9 shows the performance curves of the 130-cell stack. It can be seen that the output power of the PEMFC stack under ambient pressure can reach to 14.3 kW, this result indicates that the electrochemical performance of large stack assembled with expanded graphite bipolar plate is favorable and can satisfy the qualification of the PEMFC. Fig. 10 gives the voltage distribution curves of every single cells in the 130-cell stack at different current levels, the cell voltages are more uniform which indicates that the stack can run steadily and the flow field structure and

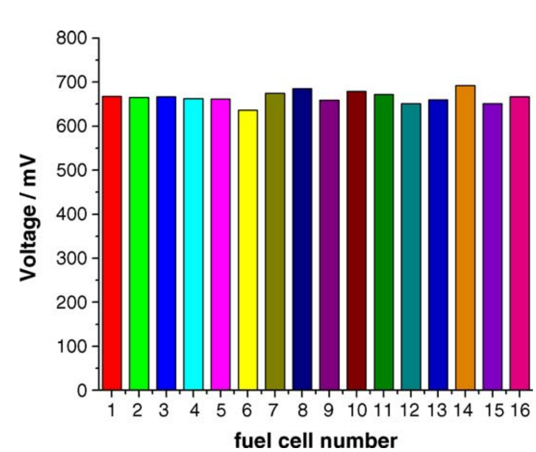

Fig. 7. Voltage distribution in the 16-cell stack at current density 500 mA cm^{-2}.

Fig. 8. The 10 kW (130-cell) stack using the expanded graphite bipolar plates. Active area was 270 cm^2.

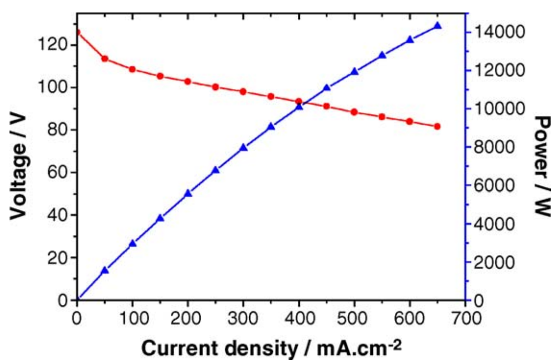

Fig. 9. The ambient pressure performance curves of the 130-cell stack assembling using expanded graphite bipolar plates; operating temperature = 60 °C; operating pressure = ambient pressure; λ_{H_2} = 1.5 and λ_{air} = 2.5; RH_{H_2} = 60%; RH_{air} = 80%.

manufacture technology of expanded graphite bipolar plate have already satisfied the requirement of gas distribution among every single cell in the large stack. Above all, the weight of this 130-cell stack is 30 kg and the weight power density is 475 W kg^{-1} at ambient pressure, therefore, expanded graphite can sever as

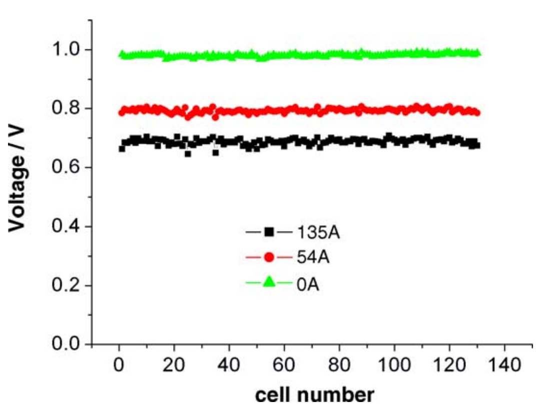

Fig. 10. The voltage distribution of every single cell among the 130-cell stack at the current 135, 54 and 0 A; operating temperature = 60 °C; operating pressure = ambient pressure; λ_{H_2} = 1.5 and λ_{air} = 2.5; RH_{H_2} = 60%; RH_{air} = 80%.

bipolar plate material in fuel cell and can be applied widely in the future.

3.4. *Cost of the expanded graphite bipolar plate*

Because the expanded graphite plate need not been pretreated at high temperature the material cost is very low, its price is US$ 4–5 kg^{-1} in China market. The gas flow channels and cooling water channels can be stamped directly, eliminating the step for costly machining. To evaluate the cost of material and technology a typical plate design was used. The size of the bipolar plate was 410 mm × 100 mm. The thickness of the bipolar plate was 3 mm. The weight of the bipolar plate was 130 g. The bipolar plate included gas channels, cooling channels and integrated seals. The cost of expanded graphite plate material and polymer resin is less than US$ 1 per piece; the cost of molds, equipment, and energy consumption is about US$ 1.3 per piece for a capacity of 2000 plates per month. Currently, the cost of labor is about US$ 0.7 per piece, thus the total cost price of per expanded graphite bipolar plate was about US$ 3 per piece for the area of 410 cm^2. The cost of bipolar plates can be more reduced if the bipolar plates are mass-produced.

4. Conclusions

The expanded graphite is a low-cost bipolar plate material; it has low contact resistance and high surface energy. A 1 and 10 kW class PEMFC stacks using expanded graphite bipolar plates are successfully assembled and the performance of the stacks is excellent.

The power of the 16-cell short stack can reach to 1.6 kW and the power density of the MEA in the stack can surpass 0.37 W cm^{-2} when the stack is running under ambient pressure. Furthermore, when the stack is operating under higher pressure (2 bar) and the maximum working current density exceeds 1000 mA cm^{-2}, the performance of the stack can reach to 2.4 kW and the power density of the MEA in the stack can exceed 0.54 W cm^{-2}.

In addition, the performance of the 130-cell stack is also satisfied, the performance of every single cell in the stack is very uniform and the performance is steadily.

The expanded graphite is a promising bipolar plate material for PEMFC commercialization development.

References

[1] L.G. Marianowski et al., Institute of Gas Technology, Technical Quarterly Progress for July to September 30, 1998.
[2] O.J. Murohy, A. Cisar, E. Clarke, Electrochim. Acta 43 (1998) 3829.
[3] T.M. Besmann, J.W. Klett, J.J. Henry Jr., E. Lara-Curzio, J. Electrochem. Soc. 147 (2000) 4083.
[4] H. Tsuchiya, O. Kobayashi, J. Hydrogen Energy 29 (2004) 985.
[5] S. Joseph, J.C. McClure, R. Chianelli, P. Pich, P.J. Sebastian, J. Hydrogen Energy 30 (2005) 1339.
[6] P.L. Hentall, J.B. Lakeman, G.O. Mepsted, P.L. Adcock, J.M. Moore, J. Power Sources 80 (1999) 235.
[7] D.R. Hodgson, B. May, P.L. Adcock, D.P. Davies, J. Power Sources 96 (2001) 233.

[8] M.C. Li, C.L. Zeng, S.Z. Luo, J.N. Shen, H.C. Lin, C.N. Cao, Electrochim. Acta 48 (2003) 1735.
[9] A. Hermann, T. Chaudhuri, P. Spagno, J. Hydrogen Energy 30 (2005) 1297.
[10] D.N. Busick, M.S. Wilson, Fuel Cell Bull. 2 (1999) 6.
[11] J. Scholta, B. Rohland, V. Trapp, U. Focken, J. Power Sources 84 (1999) 231.
[12] H. Maeda, A. Yoshimura, H. Fukumoto, T. Hayashi, Abstracts of Fuel Cell Seminar, 2002, p. 58.
[13] A. Heinzel, H. Kraus, C. Kreuz, F. Mahlendorf, O. Niemzig, Abstracts of Fuel Cell Seminar, 2002, p. 153.
[14] J. Stewart, C. Robert, US Patent 4,670,300 (1987).
[15] T. Uemura, S. Murakami, US Patent 4,737,421 (1988).
[16] D. Busick, M. Wilson, US Patent 6,248,467 (2001).
[17] T.M. Besmann, J.W. Klett, J.J. Henry Jr., E. Lara-Curzio, J. Electrochem. Soc. 147 (2000) 4083.
[18] J. Huang, D.G. Baird, J.E. McGrath, J. Power Sources 150 (2005) 110.
[19] E.A. Cho, U.-S. Jeon, H.Y. Ha, S.-A. Hong, I.-H. Oh, J. Power Sources 125 (2004) 178.
[20] D.N. Busick, M.S. Wilson, Fuel Cell Bull. 2 (1999) 6.
[21] E.A. Cho, U.-S. Jeon, S.-A. Hong, I.-H. Oh, S.-G. Kang, J. Power Sources 142 (2005) 177.
[22] M.R. Angelo, C.J. Paul, W.M. Lee, W.T. William, Europe Patent WO0064808 (2000).
[23] H. Zhang, M. Hou, China Patent 200410020906 (2004).
[24] P.R. Gibb, Europe Patent WO0041260 (1999).
[25] E.A. Cho, U.-S. Jeson, H. Ha, S.-A. Hong, I.-H. Oh, J. Power Sources 125 (2004) 178.
[26] A.J. Cisar, O.J. Murphy, K.-I. Jeng, S. Simpson, D. Weng, US Patent 6, 426,161 (2002).