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Journal of Power Sources 162 (2006) 207–214
JOURNAL OF POWER SOURCES
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A novel Nylon-6–S316L fiber compound material for injection molded PEM fuel cell bipolar plates

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Received 11 May 2006; received in revised form 9 June 2006; accepted 9 June 2006
Available online 27 July 2006

Abstract

This paper reports on a novel composite material for the bipolar plates of proton exchange membrane fuel cells (PEMFCs). In general, the materials used to fabricate PEMFC bipolar plates must be low-cost, easily fabricated, light, strong, mechanically stable and have a low surface contact resistance. His study fabricates bipolar plates using a composite material comprising Nylon-6 and S316L stainless steel alloy fibers. The PEMFC plates were fabricated via an injection molding process, which yielded better production rates than coating or treating metal plates with a suitable surface material. It is shown that the developed composite material has a suitable combination of properties and processability for PEMFC bipolar plate applications. Specifically, the chemical and electrochemical properties of the plates were found to be stable, and the plates demonstrated good anti-corrosion characteristics, resulting in an enhanced electrical conductivity, a stable cell performance and an extended cell life.

Keywords: Bipolar plate; PEMFC; Nylon-6 polymer thermoplastic; Injection molding; Stainless steel alloy fiber

1. Introduction

Proton exchange membrane fuel cells (PEMFCs) are a highly promising power source for future residential, mobile and automobile applications. PEMFCs have the advantages of compactness, lightweight, a high power density and a low temperature of operation. The bipolar plates in conventional PEMFCs are generally fabricated from some form of carbon material. Although these materials provide an acceptable performance, further cost reductions and an increased power density are required if fuel cells are to gain widespread commercial acceptance [1]. Furthermore, despite attempts to reduce the thickness of the bipolar plates and the membrane, the manufacturing cost of fuel cell stacks remains high and this limits the commercial viability of PEMFCs.

Metal bipolar plates such as titanium and stainless steel have low gas permeation characteristics and good mechanical properties, which enable thin plates to be manufactured with high dimensional tolerances and low scrap rates. These metals also exhibit excellent corrosion resistance in a PEMFC environment [2] because a stable and protective metal oxide film (i.e. a passive film) readily forms on their surfaces. However, the metal oxide film also acts as an electrical insulator, and this leads to an unacceptable voltage drop in the fuel cell. To minimize the voltage drop, the formation of the resistive layer must be suppressed in some way. One method of achieving this is to use only materials which form highly conductive oxide layers under fuel cell conditions. The second approach is to coat the steel surface with a highly conductive layer which is both chemically and mechanically stable.

A further potential disadvantage of using stainless steel bipolar plates is their chemical instability in fuel cell environments, particularly when they make direct contact with the acidic polymer electrolyte membrane. The corrosion product of stainless steel bipolar plates tends to poison the catalysts, thereby reducing the efficiency of the fuel cell. Therefore, contamination is an important consideration when selecting an appropriate material for the bipolar plates in a PEMFC.

As stated above, the naturally occurring oxide film on stainless steel surfaces [3] is a matter of concern in PEMFC applications. Although this surface film theoretically protects the bulk material from corrosion, it also significantly increases the

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doi:10.1016/j.jpowsour.2006.06.034

Table 1
Comparison of graphite and S316L properties

Property	Graphite	S316L
Cost (US$ kg^{-1})	75	15
Density ($gm\,cm^{-3}$)	2.25	8.02
Thickness of bipolar plate (mm)	2.5–4	1–2
Modulus of elasticity (GPa)	10	193
Tensile strength (MPa)	15.85	515
Corrosion current ($MA\,cm^{-2}$)	<0.1	<0.1
Electrical resistivity ($\Omega\,cm\,10^{-6}$)	6000	73
Thermal conductivity ($W\,m^{-1}\,K^{-1}$)	23.9	16.3
Permeability ($cm\,s^{-1}$)	10^{-2} to 10^{-6}	$<10^{-12}$

contact resistance between the bipolar plate and the electrode backing. Moreover, the oxide film formed on a stainless steel surface in air may not actually protect the bulk substrate in a fuel cell environment. Furthermore, as the corrosion film thickens over time, the contact resistance increases and the voltage output from the PEMFC therefore decreases. Nonetheless, various stainless steels have been considered for use as bipolar plates and it has been determined that the corrosion rate is generally sufficiently low that the performance of the PEMFC is not significantly degraded. Various studies have reported a stable cell output for thousands of hours [4–6].

In general, stainless steels with a high Cr and Ni content exhibit a thinner oxide film and an improved cell performance. Table 1 compares the properties of S316L stainless steel alloy with graphite (the material conventionally used to fabricate the bipolar plates in PEMFCs). It can be seen that S316L has a lower cost, a higher tensile strength, and a lower electrical resistivity. Accordingly, this study fabricates bipolar plates using a composite material comprising a Nylon-6 polymer thermoplastic matrix and stainless steel S316L fibers.

The bipolar plate is a multi-functional component of the PEMFC. Its primary roles include delivering the reactant gases to the fuel cell electrodes, providing an electrical connection between adjacent cells in the stack, removing the water by-product from the cell, and dissipating the reaction heat. A bipolar plate must meet the following material and construction requirements [7,8]:

1. high electrical conductivity (DOE target >100 $S\,cm^{-1}$);
2. high thermal conductivity (PlugPower's target >10 $W\,m^{-1}\,K^{-1}$);
3. good mechanical properties (PlugPower's target targets: tensile strength >41 MPa, flexural strength >59 MPa, impact strength >40.5 $J\,m^{-1}$; DOE target: crush strength >4200 kPa);
4. thermal stability at fuel cell operating temperature (−40 to 120 °C for fuel cell driven vehicles);
5. chemical stability in the presence of fuel, oxidant and product water, which may be slightly acidic (DOE target: corrosion rate <16 $\mu A\,cm^{-2}$);
6. inexpensive materials and processing costs.

The high cost of the graphite-based bipolar plates used in many PEMFCs is the result of the brittleness of the graphite,

Table 2
Nylon-6 properties

Property	Test method	Value
Density at 25 °C	ISO 1183	1128 $kg\,m^{-3}$
Hardness (15 s value)	ISO 868	78 Shore D
Heat distortion temperature at 1.81 MPa	ISO 75	65.5 °C
Vicat softening point at 10 N	ISO 306	21.5 °C
Melt flow index (275 °C 10 kg^{-1})	ISO 1133	43 g 10 min^{-1}
Melt flow index (275 °C 10 kg^{-1})	ISO 1133	9 g 10 min^{-1}
Flexural modulus	ISO 178	2300 MPa
Flexural strength	ISO 178	7 MPa
Tensile strength at break	ISO 527	45 MPa
Tensile strength at yield	ISO 527	50 MPa
Elongation at break	ISO 527	15%
Notched izod impact at 25 °C	ISO 180	11 $kJ\,m^{-2}$

which increases the machining costs of the flow channels on the plates. Accordingly, extensive research has been conducted to identify low-cost, high performance alternatives to graphite. Various candidate materials have been proposed, including metal-based bipolar plates [9–11], carbon-filled polymers [5], and carbon/carbon composites [9]. As shown in Table 2, Nylon-6 has a low-cost, good gas tightness, high nobility, excellent chemical resistance, favorable mechanical properties, and good impermeability. Accordingly, Nylon-6 is chosen as the polymer matrix in the present study and is blended with S316L stainless steel fibers to create a composite material with which to fabricate the current bipolar plates.

For materials with high carbon concentrations, injection molding enables the mass production of low-cost bipolar plates [12]. Furthermore, injection molding and compression molding are also suitable for the fabrication of bipolar plates using carbon-filled polypropylene (PP) and polyphenylene sulfide (PPS). Accordingly, injection molding provides a convenient technique for fabricating the current Nylon-6/S316L stainless steel bipolar plates.

2. Experimental

2.1. Preparation of PEMFC

As discussed above, the current bipolar plates are fabricated using a composite material comprising Nylon-6 polymer thermoplastic and S316L stainless steel alloy fibers. Although Nylon-6 is a non-conductor, the addition of the S316L stainless steel fibers ensures that the bipolar plates have good electronic conductivity and excellent corrosion resistance. Furthermore, Nylon-6 is known to form homogeneous mixtures when combined with suitable resins. Generally, the electrical properties and mechanical stability of bipolar plates are improved with an increased stainless steel alloy fiber content. Therefore, the present study specifies a basic composition for the bipolar plates of 40 wt.% Nylon-6 polymeric composite with adhesive and 60 wt.% stainless steel alloy fiber (see Table 3). Using an injection molding process, bipolar plates were fabricated with a total thickness of 3.5 mm and an active area of 25 cm^2.

Table 3
Composition of bipolar plates

Component (wt.%)	
Nylon-6 polymeric thermoplastic (including stearic acid)	40
Stainless steel alloy fiber	60
Fiber length (mm)	6.4

A membrane-electrode assembly (MEA) was then fabricated by placing electrodes on either side of a pre-treated Nafion 112 (50 μm) membrane (ElectroChem), with platinum loadings of 0.2 and 0.4 mg cm^{-2} for the anode and cathode, respectively. A fuel cell was then constructed using the prepared MEA, a Teflon gasket, and two Nylon-6 flow field plates positioned on either side of the MEA (see Fig. 1).

2.2. *Decision-making and experimental processes involved in fabrication of current bipolar plates*

Fig. 2 presents a flowchart of the decision-making processes and experimental procedures conducted in the current study. In the first step, the suitability of S316L stainless steel alloy as the main component of the bipolar plate composite material was appraised. Through mechanical and electrical testing, it was established that the optimal fiber length was 6.4 mm (0.25 in.). Having chosen S316L and Nylon-6 as the materials for the bipolar plates, electrical conductivity tests and scanning electron microscopy (SEM) analysis were performed to establish a suitable mix of the two components. Subsequently, the bipolar plate flow fields and the injection molds were designed and fabricated. Using the Nylon-6/S316L composite material, bipolar plates with an interdigitated type flow field (see Fig. 3) were fabricated via injection molding under the processing conditions shown in Table 4. The bulk resistance of the plates was measured using a four-point probe (4PP) method with a 280CI probe purchased from SE Technologies Co. The quality of the injection molded plates (in terms of the distribution and direction of the stainless steel alloy fibers within the composite material) was analyzed using a scanning electron microscope. The resistance, Voltage–current (*V*–*I*), power density–current (*P*–*I*), and open circuit voltage (OCV) characteristics of the PEMFC incorporating the composite bipolar plates were then evaluated and compared to the results obtained from a PEMFC constructed using conventional graphite bipolar plates.

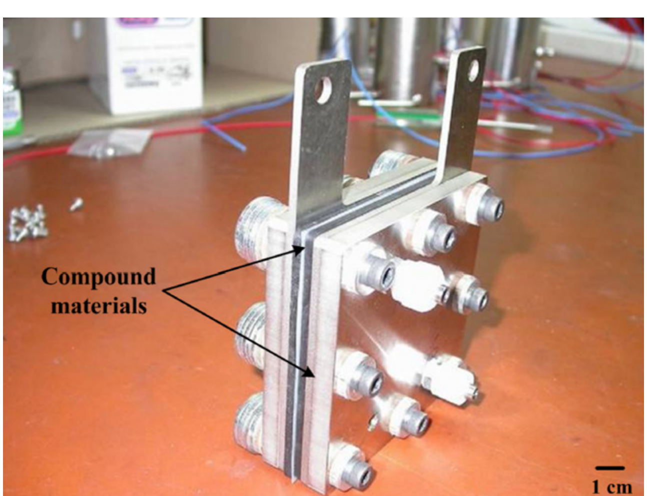

Fig. 1. Photograph of single PEM fuel cell containing Nylon-6/S316L stainless steel bipolar plates.

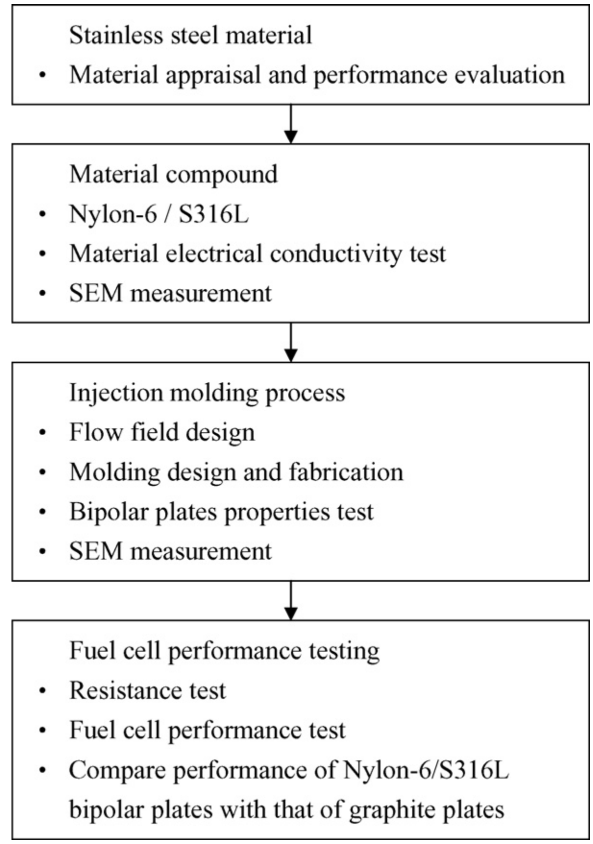

Fig. 2. Flowchart of current decision-making and experimental procedures.

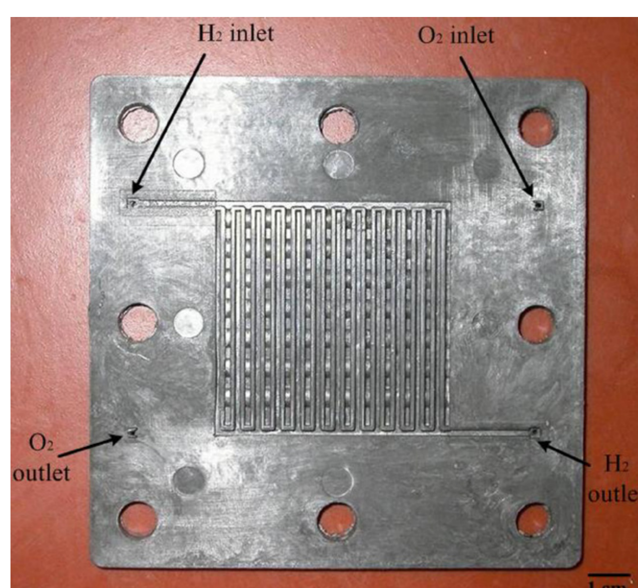

Fig. 3. Injection molded bipolar plate with interdigitated type gas flow channels.

Table 4
Nylon-6 and stainless steel alloy fiber injection molding conditions

Temperature (°C)	
Rear zone	249–271
Center zone	232–266
Front zone	221–260
Melt	243–279
Mold	54–93
Pressure (MPa)	
Injection	69–103
Hold	34–69
Back	0.34–0.69
Speeds	
Fill	13–51 mm/s
Screw	30–60 rpm
Drying	4 h at 79 °C
Dew point	−18 °C
Moisture content	0.20%

2.3. Characterization of PEMFC

The voltage–current, power density–current and open circuit voltage characteristics of the fabricated PEMFC were obtained using a Bean 300M digital multimeter (Bean Co., Taiwan) linked to an IBM PC. Meanwhile, the quality of the injection molded composite bipolar plates was evaluated from sectional SEM photographs of the electrolyte-membrane acquired using a JSM-5610 SEM (JEOL).

It has been reported that thermoplastic with low additive loadings of 50 wt.% achieve specific bulk conductivities of approximately 50 S cm^{-1} [13]. Standard composite materials consist of a thermoplastic polymer matrix and a carbon compound mixture with additional additives to increase the electrical conductivity of the compound material. This study improves the electrical conductivity of the composite mixture by using stainless steel fibers in place of carbon compound additives.

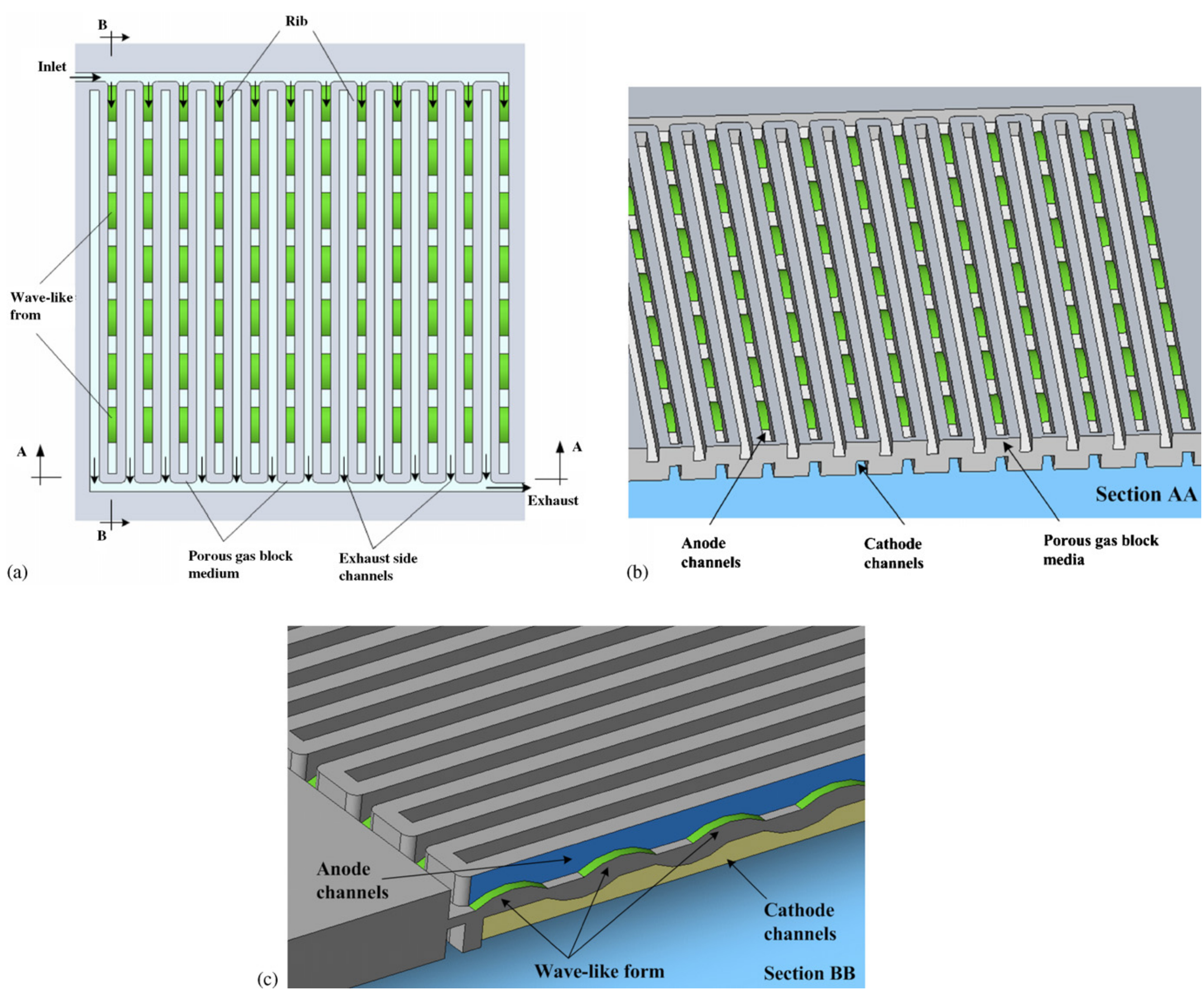

Fig. 4. (a–c) Interdigitated flow field channel in Nylon-6/S316L stainless steel bipolar plate.

2.4. Operation and performance evaluation of PEMFC

To evaluate the performance of the fabricated PEMFC, humidified hydrogen and oxygen were supplied to the anode and cathode, respectively. The PEMFC was tested under room temperature conditions (25–30 °C). The humidity of the pure hydrogen (99.999% H_2; 200 sccm) supplied to the anode was varied from 10 to 100% during the open circuit voltage measurement test, and maintained at 100% for all other tests. Meanwhile, humidified oxygen gas (200 sccm) was supplied to the cathode side throughout all of the measurement operations.

2.5. Resistance measurement

To evaluate the performance of the current PEMFC, the *V–I* and *P–I* characteristics of the cell were measured and compared to those of the conventional graphite-based bipolar plate fuel cell. The measured resistance includes contributions from the bulk material and from the contact resistance between the diffusion media and the plates. Note that the influence of lead resistances and other parasitic resistances can be neglected because the four-wire-system used for measurement purposes in this study eliminates these resistances by comparing the voltage drop across the sample with that measured across a defined reference element.

2.6. Flow field design

The interdigitated flow field, which consists of dead-ended inlet and outlet channels, forces the reactant gas to flow through the surface of the electrode. Fig. 4 presents various schematics of the interdigitated flow field on the current bipolar plates. In general, the interdigitated flow field design converts the transport mechanism of the reactant and product gases to and from the catalyst layer from a diffusion dominant mechanism to a forced convection dominant mechanism. As a result, the thickness of the diffusion layer in the backing layer of the electrode is reduced from the whole backing layer thickness to a much thinner stagnant layer above the catalyst. More importantly, the shear force exerted by the gas flow provides an effective mechanism for removing the liquid water which becomes entrapped in the inner layers of the electrode during operation of the fuel cell, and hence reduces the electrode-flooding problem. Experimental results have shown that an interdigitated flow field design leads to a PEMFC performance improvement ranging from 50% to more than 100% [14]. To enhance the removal of the water by-product, Issacci and Rehg [15] proposed the use of a gas block mechanism at the cathode and anode sides of the fuel cell. In their design, the authors used one or more porous gas block media at points adjacent to the flow field, as illustrated in Fig. 4(a and b). The pore size of the gas block medium was such that the water was sipped off to the outside of the flow field by capillary flow while the cathode gas was prevented from flowing through the medium. In the current bipolar plates, the flow channels are designed with a novel wave-like form (see Fig. 4(c) in order to enhance the gas diffusion flow in the channels, and hence improve the fuel cell performance.

3. Results and discussion

The interdigitated channels in the current bipolar plates differ from serpentine and grid channels in that they are discontinuous, i.e. they do not directly connect the inlet and outlet manifolds. In this way, the reactant gases are forced to pass through the porous gas diffusion layer as they travel from the inlet channels to the outlet channels. Convection through the porous layer shortens the diffusion path and helps remove the water which would otherwise accumulate in the gas diffusion layer, thereby enhancing the gas reaction transport and hence the fuel cell performance, particularly at higher current densities. In the current gas flow channels, an increased amplitude of the wave-like form (see Fig. 4(c)) enhances the gas reaction and hence improves the fuel cell performance. However, depending on the properties of the gas diffusion layer, the interdigitated flow field may also result in a higher pressure drop, which is clearly undesirable. In general, many factors affect the cell performance and the cell life, including the MEA characteristics, the bipolar plate performance, and the stacking and operating parameters, and in practice it is necessary to achieve a trade-off between these factors to optimize the PEMFC performance.

3.1. Interfacial contact resistance

In order to characterize the cell assembly, tests were conducted to establish the relationship between the pressure and the interfacial contact resistance. Due to the sandwich-layered structure of the PEMFC, the surface characteristics and the surface passive (oxide) film may affect the interfacial contact resistance between the bipolar plate and the MEA. Fig. 5 presents a schematic illustration of the experimental setup used to measure the interfacial contact resistance. Note that this setup was originally proposed by Wang et al. [11]. The method basically involves measuring the potential difference across two gold plates, which sandwich the bipolar plates and the MEA, as a fixed electrical current is passed through them. The variation in the potential difference is recorded as the compaction force applied to the assembly is gradually increased. By measuring the voltage drop, it is possible to calculate the total resistance, i.e. $R = V/I$.

The interfacial contact resistance was investigated under different compaction forces. The corresponding results are shown in Fig. 6 for compaction forces in the range 80–150 N cm^{-2}. In

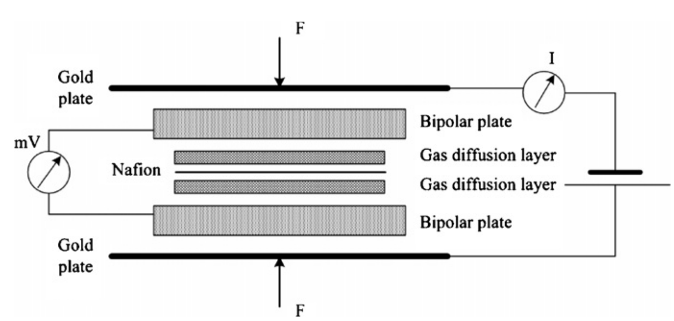

Fig. 5. Schematic diagram of experimental setup for measuring interfacial contact resistance of current bipolar plates.

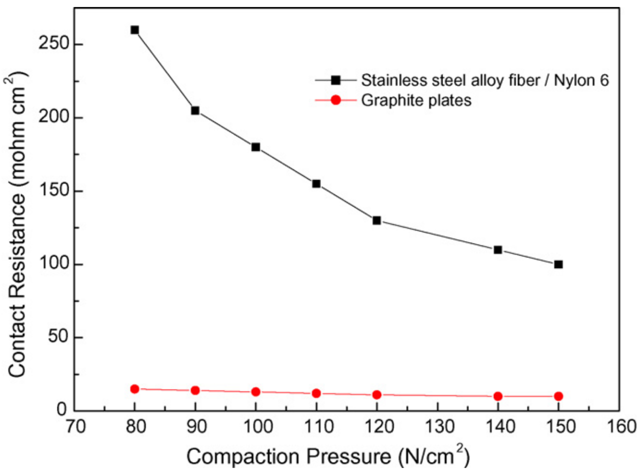

Fig. 6. Variation of interfacial contact resistance with compaction pressure.

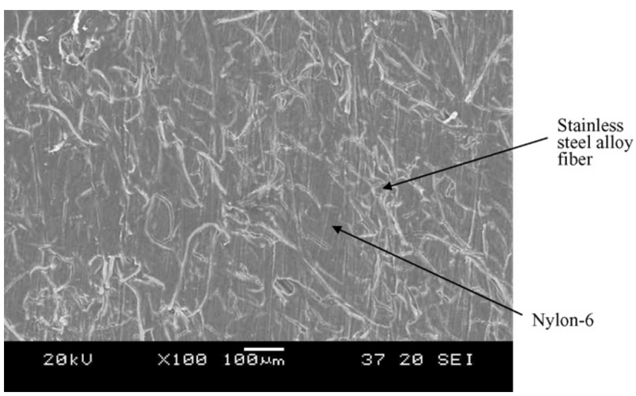

Fig. 7. Scanning electron microscopy (SEM) image of Nylon-6/stainless steel alloy fiber composite material.

general, the results show that the contact resistance decreases with an increasing compaction force. For the range of compaction force considered in the figure, the interfacial contact resistance varies from 100 to 260 mΩ cm^2. Although a reduced contact resistance is desirable, an excessive compaction force may damage the membrane-electrode assembly.

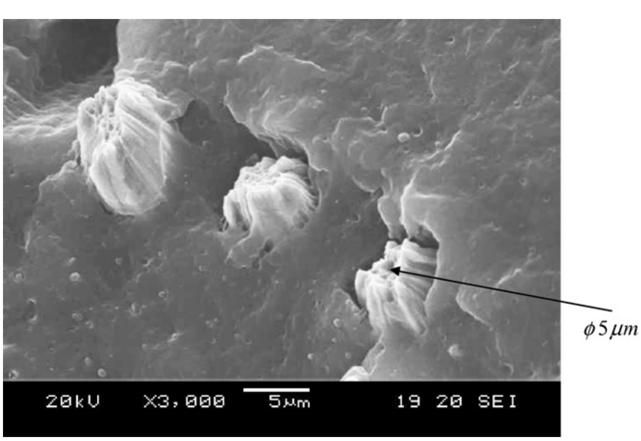

Fig. 8. High magnification scanning electron microscopy (SEM) image showing dimensions of micro-scale stainless steel alloy fiber (Ø5 μm).

The SEM images in Figs. 7 and 8 show the microstructure of the Nylon-6/S316L stainless steel fiber composite material. It is apparent that the injection molding process results in an orderly end-to-end linking of the stainless steel fibers within the Nylon-6 polymer matrix, which in turn ensures an improved electrical conductivity of the bipolar plates.

3.2. Long-term OCV test

To examine the output of the PEMFC without load, a long-term OCV test was carried out for 48 h. The test was performed at an operating temperature of 65 °C and oxygen gas was supplied to the cathode side at a rate of 200 sccm. The corresponding results are presented in Fig. 9. As shown, an initial open circuit voltage of 0.92 V was obtained. This voltage fell to a value of 0.87 V after approximately 5 h and remained stable thereafter. The theoretical output was calculated to be 1.23 V. The discrepancy between the experimental results and the theoretical results is a consequence of the contact resistance and the obstruction of the water by-product. The contact resistance of the stainless steel fiber resin increases when an oxide layer is formed on the end plates. This effect can be controlled by optimizing the compaction pressure in the PEMFC. Meanwhile, the volume of entrapped water can be reduced by improving the fluent flow field channel of the cathode fuel and reducing the compaction pressure on the MEA.

The nature of the reaction between the two reactants, and hence the overall efficiency of the PEMFC, is determined by the shape of the gas flow channel. During performance testing, forced convection of the reactants (H_2 and O_2) into the catalyst layer takes place. Testing was performed at room temperature (25 °C) and ambient pressure (1 atm) with H_2 and O_2 reactant mass flow rates of 200 sccm, as shown in Table 5. Note that during operation, the temperature of the cell did not exceed 65 °C.

Fig. 10 compares the *V–I* curves of the current PEMFC with composite bipolar plates and a conventional PEMFC with graphite bipolar plate. In both cases, the voltage drops with

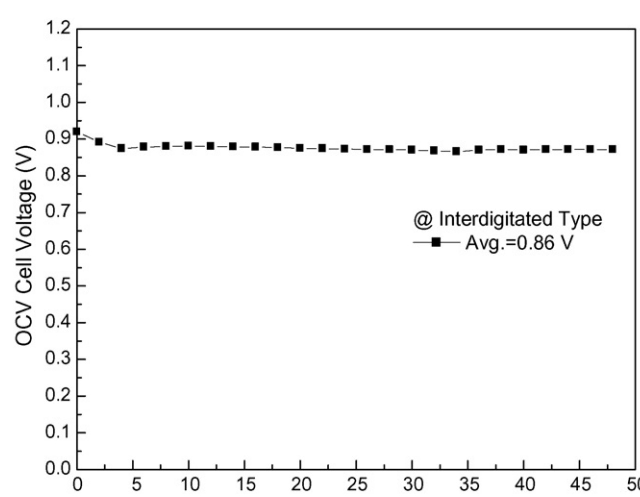

Fig. 9. Variation of open circuit voltage (OCV) over 48 h voltage test of bipolar plate with interdigitated type gas flow channel configuration.

Table 5
Bipolar plate test conditions

Condition	Value
Reaction area (cm^2)	25
Temperature (°C)	25
Anode and Cathode side pressure (atm)	1
Channel depth (mm)	1.5
Channel width (mm)	1.2
Rib width (mm)	1.5
Bipolar plate thickness (mm)	3.5
Mass flow rate (anode) (sccm)	200
Mass flow rate (cathode) (sccm)	200
Catalyst layer thickness (anode) (mm)	0.018
Catalyst layer thickness (cathode) (mm)	0.026
Diffuser layer thickness (mm)	0.2

increasing current. However, the voltage drops more rapidly in the current PEMFC because the compound surface contains micro holes, which increase the resistance and therefore reduce the voltage.

Table 6 summarizes the basic mechanical properties of the current PEMFC, i.e. the tensile strength, flexural strength, impact strength, elongation at fracture, and Rockwell hardness test. The properties are consistent with the targets specified for conventional PEMFCs.

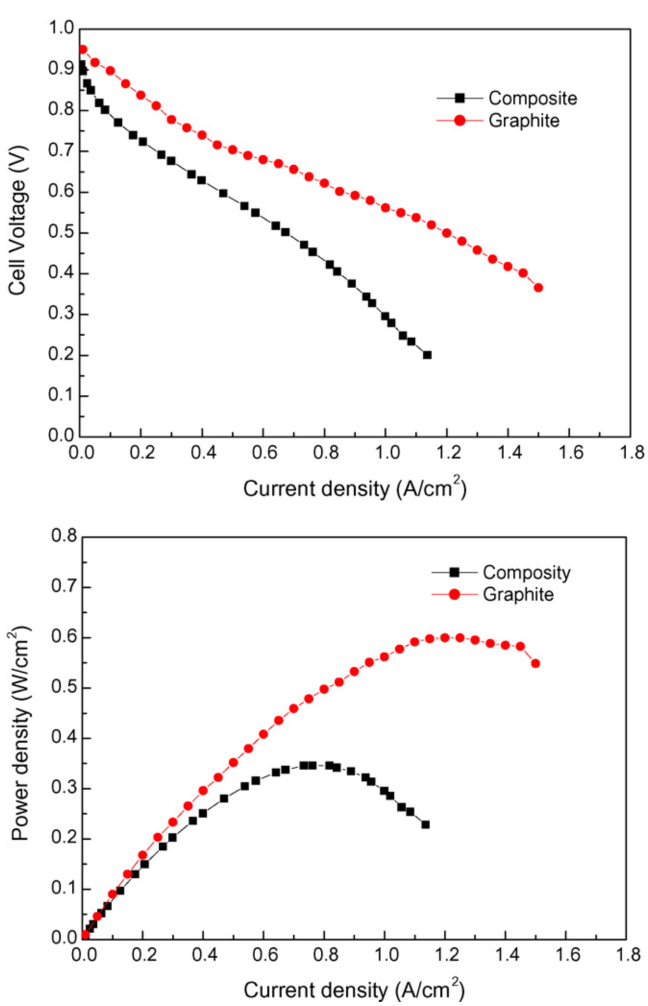

Fig. 10. PEMFC performance test results for composite and graphite single fuel cell.

Table 6
Mechanical properties of composite bipolar plate

Condition	Value
Tensile strength (MPa)	52.3
Elongation at fracture (%)	2.39
Flexural strength (MPa)	68.5
Impact strength (J m^{-1})	102.8
Rockwell hardness test (HRM)	54.4

Table 7
Bipolar plate costs

Condition	Value (US$ plate^{-1})
Material	5
Injection molding process	0.5
Equipment	0.5
Energy	0.5
Labor	0.5
Other	1
Total price	8

The various costs associated with fabricating the current composite bipolar plate are estimated in Table 7. As shown, the total estimated cost of each plate is US$ 8. However, the use of injection molding proposed in this study for fabricating the bipolar plate provides the potential for mass production and therefore this cost will be significantly reduced.

The Nylon-6/stainless steel alloy fiber composite compound has a high electrical conductivity (60 S cm^{-1}). However, increasing the stainless steel alloy fiber content causes the composite to become stiff. Hence, fabricating the bipolar plates using an injection molding process is difficult. Improving the electrical conductivity of the bipolar plates, while simultaneously developing the means for their precision mass fabrication is an important area of future research. Furthermore, with an operating voltage of 0.6 V (the general operating voltage for a PEMFC), the current PEMFC provides a current density of 350 mA cm^{-2} (see Fig. 10) and a low power density of 210 mW cm^{-2}. Therefore, improving the current density is also an important topic for further research.

4. Conclusion

This study has used a Nylon-6/S316L stainless steel alloy fiber composite material to mold conductive plastic bipolar plates for a PEM fuel cell. The resulting bipolar plates are inexpensive, have favorable corrosive, hardness, and gas tightness properties, are lightweight and small, and are easy to manufacture. Although the performance of the current PEMFC fails to match that of conventional PEMFCs with graphite bipolar plates, the present results have suggested that, with further improvement, the current Nylon-6/S316L stainless steel composite material may provide a feasible alternative to graphite for the bipolar plates in PEMFCs. Despite the relatively poorer performance of the current PEMFC in its present state of development compared to the conventional graphite-based PEMFC, the Nylon-6/S316L stainless steel composite material has a lower cost than graphite, while the use of injection molding provides the potential for rapid, low-cost mass production. Therefore, the current bipolar plates have commercial potential and merit further investigation and improvement.

Accordingly, in future studies, the current authors intend to enhance the performance of the developed PEMFC by improving its water draining characteristics. Specifically, the authors intend to investigate the effect of the fuel cell orientation (e.g. 0°, 45°, and 90° orientations) on the water draining performance, and to enhance the molding process such that the smoothness of the flow channels is improved. Furthermore, attempts will be made to reduce the contact resistance by increasing the S316L fiber length and coating Ni and Ti alloys on the fiber surface. Finally, polymer research will be performed to reduce the holes in the matrix following injection molding and to improve the surface hydrophobic effect of the composite material.

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