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Short communication

Effect of surface roughness of composite bipolar plates on the contact resistance of a proton exchange membrane fuel cell

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ABSTRACT

Polymer electrolyte membrane fuel cell performance strongly depends on properties of the fuel cell stack bipolar plates. Composite bipolar plates, though low cost and convenient in manufacturing, raise a major concern due to their high interfacial contact resistance caused by the mechanical treatment used to remove the polymer-rich layer on the surface. It is observed that most of this contact resistance is governed by electrical properties of the interface layer between the contacting surfaces. Measurements of contact resistance of mechanically polished composite bipolar plate/gas diffusion layer interface reveal a substantial influence of surface topography on the contact resistance, which varies significantly depending on the substrate surface treatment and roughness of composite bipolar plates.

1. Introduction

Fuel cells are a promising power technology with a wide variety of potential applications. With their ability to act as stationary power sources to create "micro-grids" and near-zero emissions for automotives, fuel cells are being sought to solve the energy needs and climatic problems. Particularly, proton exchange membrane fuel cells (PEMFC) have received broad attention due to their low operating temperature, low emissions and quick startup. But high material costs, low power density and lifetime of fuel cell systems remain a major barrier to their wide use. A major influence on the fuel cell cost and its power density is due to bipolar plates (BPs) that electrically connect the adjacent cells of the stack and provide the gas supply to the cells [1].

In order to become commercially viable, it is widely accepted that PEMFC bipolar plates have to be cheaper, lighter, and more compact. Bipolar plates make up 12% cost of fuel cell price [1] and currently, the most common bipolar plate materials being used are graphite, metals and composites [2]. Graphite is a good electrically conducting, corrosion resistant and light weight material but it is brittle, difficult to machine and costly. Conversely, stainless steel (SS) plates have relatively high strength, but they self passivate on exposure to air and anodic potential imposed on the plate during fuel cell operation forming an insulating passive film on the surface. This reduces electrical conductivity and results in high contact resistance leading to high ohmic overpotential [1–4].

Unlike SS or graphite bipolar plates, composite bipolar plates constitute a small fraction of the cost of machined graphite plates and less than two-thirds the cost of etched SS plates but have lower electrical conductivity [5] due to the partial polymer content. They also exhibit good electrochemical stability in the aggressive PEMFC environment and long-term durability is not a major concern [6,7]. Therefore, several kinds of composite plates for PEMFCs are currently under development to reduce cost, stack volume and weight [8,9].

2. Interfacial contact resistance

A point of concern in fuel cells is the high contact resistance in a fuel cell stack which comes from the interface between the bipolar flow channel plate and a gas diffusion layer (GDL, which is commonly made of carbon paper or carbon cloth) [10]. In a single PEMFC, a polymer membrane is inserted between two GDLs coated with electrocatalyst which are further sandwiched between two current collectors. Multiple fuel cells are stacked together, as shown in Fig. 1, to provide desired power and voltage. The cells are separated by bipolar plates, which secure electron current flow all through the stack. This set-up results in the amplification of the losses from contact resistance between contacting GDLs and bipolar plates [1].

One of the performance measures of a PEMFC is the cell potential, which decreases from its equilibrium potential during operation because of losses; ohmic resistance being the dominant

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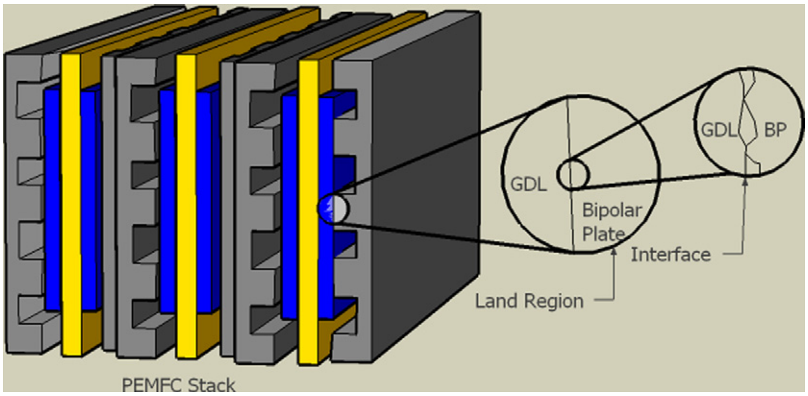

Fig. 1. Schematic showing the contact regions at the interface of bipolar plate and gas diffusion layer of a PEMFC stack where contact resistance occurs.

loss. Ohmic resistance is caused due to the obstruction to the flow of electrons at various stages in their path by GDL, bipolar plates and contact interfaces. This interfacial contact resistance constitutes a significant part of the ohmic resistance, especially when stainless steel, titanium or molded graphite is chosen as the bipolar plate material [2,11] due to the insulating layer at the interface. The performance of a PEMFC system can be significantly improved if the contact losses can be minimized [1,10]. Reported values of interfacial contact resistance in PEMFC literature largely vary, from 5 $\mathrm{m}\Omega\,\mathrm{cm}^{-2}$ [10], 25 $\mathrm{m}\Omega\,\mathrm{cm}^{-2}$ [2] to 150 $\mathrm{m}\Omega\,\mathrm{cm}^{-2}$ [4] depending on the bipolar plate material, diffusion media, compression pressure, surface roughness of contacting materials and other conditions used during the measurements; thus making it difficult for comparison.

The interfacial contact resistance between a GDL and the bipolar plate in a fuel cell stack is governed by the multi-scale surface topography of the contacting pair. The roughness features at the contacting surfaces decrease the actual area in contact, leading to a voltage drop across the interface. The clamping of a stack of individual cells causes a pressure to be applied at the interface, leading to an increase in the contact area between fuel cell components, which in turn decrease the contact resistance. However, a large pressure may deform the GDLs and membrane, causing cell leakage and internal short-circuit. Thus an optimum clamping pressure exists that trades off between the competing requirements [10].

2.1. Contact resistance of composite bipolar plate

In case of polymer/graphite composite bipolar plates, different thermoplastic and thermoset polymers, e.g. phenolic and epoxy resins, and vinyl esters are used as binders and processes such as compression, injection, injection–compression, transfer and slurry molding are used for molding [12]. Typically, one of the chief difficulties with molding polymer/graphite plates is the formation of a polymer-rich, non-conductive skin over the surface of the molded part, which on a bipolar plate contact region leads to an increase in the contact resistance at the bipolar plate–GDL interface [5].

Currently, the industry uses various proprietary "surface activation" methods, such as sanding and abrasion, to remove the polymer-rich resin layer from the surface of polymer/composite bipolar plates. These surface treatments generate topography on the bipolar plate surface. Previously, studies have been done by Y. Ein-Eli's group [1] on the influence of surface roughness and texturing of SS bipolar plates on its contact resistance. The roughness was caused when the surface passivating oxide was removed to decrease the contact resistance of PEMFC. Mishra et al. [10] proposed a model on the measurement and prediction of contact resistance based on the surface topography of contacting surfaces of metallic bipolar plate and GDL and Zhou et al. [11] proposed a micro-scale model for predicting contact resistance but there is little information in the literature concerning surface topography of composite bipolar plates and its influence on contact resistance. In this paper, we discuss the influence of surface roughness of a composite bipolar plate, caused due to surface activation and its influence on the interfacial contact resistance of a PEMFC.

2.2. Theory of contact resistance

The contact resistance at an interface is a function of the surface topography and the properties of the two materials. As of now, it is commonly accepted that contact resistance is governed by the surface topography of the contacting pair: the roughness features at the contacting surface decrease the actual area in contact and current flows only through the contact asperities leading to a voltage drop across the interface [1].

Contact resistance originates from current constriction in contact asperities and depends on the topography of the surfaces of the contacting members. It is intuitively clear that if contacting members are pressed one toward another, the contact resistance drops as pressure increases. Firstly, the effect takes place because of the increase in contact area under the load: each contact spot increases the area because of material deformation and the amount of contacting spots increases because of dropping of contacting member separation [13]. Further information on the theory of contact resistance can be found in references' [1,11,13].

3. Experimental

3.1. Materials

BMC 940-8649 flat composite plaques (Bulk Molding Compound, USA) of thickness 3 mm, 60–70% graphite content and molded using vinyl ester polymer are cut to 5 cm × 5 cm for contact resistance measurements. The in-plane and through-plane resistivity for this material are 0.005 $\mathrm{m}\Omega\,\mathrm{cm}$ and 0.01 $\mathrm{m}\Omega\,\mathrm{cm}$ respectively [14]. Plain untreated Toray® TGP-H-060 carbon papers (ElectroChem Inc., USA) are used as GDLs to ensure good electrical contact between the two copper electrodes and the measured sample. The in-plane and through-plane resistivity for Toray GDL are 5.8 $\mathrm{m}\Omega\,\mathrm{cm}$ and 80 $\mathrm{m}\Omega\,\mathrm{cm}$ respectively (with a porosity of 78% and thickness 0.19 mm). Mirror polished copper plates electroplated with gold coating of 1 μm thickness are used as electrodes.

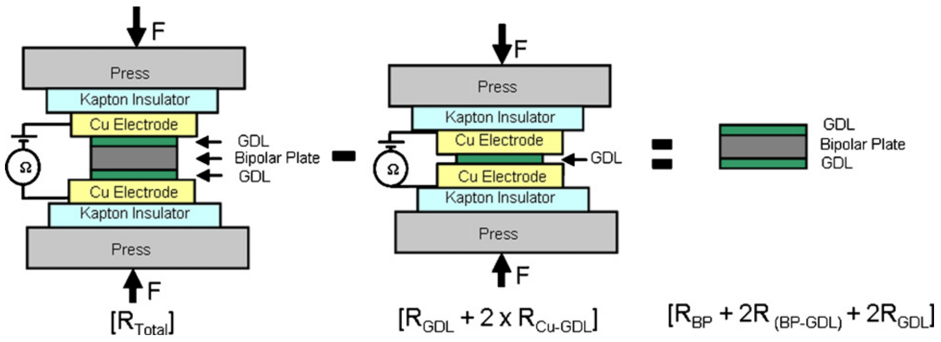

Fig. 2. Schematic of contact resistance measurement set-up.

3.2. *Contact resistance measurement set-up*

Contact resistance measurements are conducted with a set-up as shown in Fig. 2. A Keithley® 2200 sourcemeter (min resolution 1 μΩ) is used to measure the total electrical resistance of the set-up and a Carver® hydraulic press (max force 20 T) is used to provide a series of prescribed clamping pressures. Kelvin clip leads are used to connect the gold coated copper plates to the sourcemeter. To insulate the electric circuit from the press, 0.2 mm thick Kapton® polymer films are placed between the platens of the press and the electrodes. A DC current in the range of 1 μA to 1 A is applied by the sourcemeter and the potential drop is measured to calculate the total resistance (R_{Total}) to the GDL/bipolar plate assembly.

Applied pressures ranged from well below typical plate land pressures of 1.0–1.5 MPa up to around 6.0 MPa. Sandpapers of various grit sizes (United States Coated Abrasive Manufacturers Institute, CAMI, grit designation: 80, 150, 240, 400, 600 and 1000) are used to polish the 5 cm × 5 cm composite bipolar plate samples. Each plate is gently hand polished by the sandpaper until the shiny surface of polymer layer is removed. Three bipolar plate samples are used for measurement of the contact resistance and roughness for each grit size. The surfaces of polished bipolar plate and the carbon paper are cleaned with isopropanol and dried to remove any particle debris. Surface profile scans are performed to measure the average roughness using a profilometer Tencor® AlphaStep 200 at a scan rate of 50 μm s^{-1} over a 2 mm length. Each sample is scanned at 10 different regions and average roughness (R_a) calculated from the 30 values to give statistically significant number (10 values from each of the three samples for one grit size). Hitachi S-4000 scanning electron microscope (SEM) equipped with energy dispersive spectroscopy (EDS) facility is used for surface imaging and elemental analysis.

3.3. *Contact resistance measurement method*

Two calibration set-ups were used to isolate the value of the bulk and that of the contact resistance from one another. Fig. 2 presents the two set-ups used for the calibration procedure. In set-up 1, the resistance of one bipolar plate sandwiched between two carbon papers is measured between two copper electrodes while in set-up 2 carbon paper is placed between the copper electrodes. As all copper surfaces are mirror polished and electroplated with gold, it is assumed that both the copper electrodes have same surface finish.

From set-up 1, at a given pressure, the components which contribute to observed resistance is described by the following equation:

$$R_{\text{Total}} = [2R_{\text{Cu}} + 2R_{\text{GDL}} + R_{\text{Bp}} + 2R_{(\text{Cu}-\text{GDL})} + 2R_{(\text{Bp}-\text{GDL})} + R_{\text{i}}] \quad (1)$$

$$R_{(\text{Bp}-\text{GDL})} = 0.5\ [R_{\text{Total}} - 2R_{\text{Cu}} - 2R_{\text{GDL}} - R_{\text{Bp}} - 2R_{(\text{Cu}-\text{GDL})} - R_{\text{i}}] \quad (2)$$

where, R_{Total} = total resistance of the set-up, R_{Cu} = bulk resistance of copper electrode, R_{Bp} = bulk resistance of bipolar plate, R_{GDL} = bulk resistance of carbon paper, R_{i} = internal resistance of the system (sourcemeter, wiring etc). R_{i} is measured between the electrodes using a fixed resistor and is found to be negligible. $R_{(\text{Cu}-\text{GDL})}$ = contact resistance between electrode and GDL, $R_{(\text{Bp}-\text{GDL})}$ = contact resistance between bipolar plate and GDL.

All the components of Eq. (2) are known except for the contact resistance between copper electrode and the carbon paper, $R_{\text{Cu}-\text{GDL}}$. From set-up 2, the components which contribute to observed resistance is described by the equation:

$$R'_{\text{Total}} = [2R_{\text{Cu}} + R_{\text{GDL}} + 2R_{(\text{Cu}-\text{GDL})} + R_{\text{i}}] \quad (3)$$

$$R_{(\text{Cu}-\text{GDL})} = 0.5\ [R'_{\text{Total}} - 2R_{\text{Cu}} - R_{\text{GDL}} - R_{\text{i}}] \quad (4)$$

Substituting the $R_{\text{Cu}-\text{GDL}}$ from Eq. (4) in Eq. (2) will give the interfacial contact resistance between the composite bipolar plate and gas diffusion layer.

4. Results and discussion

4.1. *"Polymer skin" on bipolar plate surface*

The SEM images of unpolished surface of the bipolar plate show the presence of a non-uniform "foggy" layer on the unpolished plate surface (Fig. 3(a)). The layer shows "very high charging effects" when the electron beam is focused on it, indicating its insulating properties. No such layer is present (Fig. 3(b)) on the polished sample and minimal charging effects were observed when the electron beam is focused on it's surface. The visual and charging effects indicate the presence of a non-conducting layer on the unpolished surface.

EDS is performed on the surfaces of the two BPs to identify the elements of the layer and as shown in the plot in Fig. 4(a), the "polymer-rich" layer on the surface of the unpolished bipolar plate shows a higher oxygen peak compared to the "almost negligible" oxygen peak in Fig. 4(b). The presence of oxygen on the unpolished BP's surface could be due to the vinyl ester from the composite, which has a polymer structure with two ester groups on either side of the polymer chain. The C=O, C—O and C—OH groups of the vinyl ester polymer could be essentially contributing to the oxygen peak in EDS analysis.

Ideally, the polished bipolar plate surface should also indicate the presence of oxygen peak in EDS plot as it contains the vinyl ester polymer as well. But since the maximum penetration volume for a 5 kV electron beam is ~1 μm, the electron beam is directly on the graphite particles which are more in number on the surface of a polished sample. Thus the SEM image, charging effects and the ED spectra indicate the presence of polymer-rich layer on the surface and in this case, vinyl ester polymer.

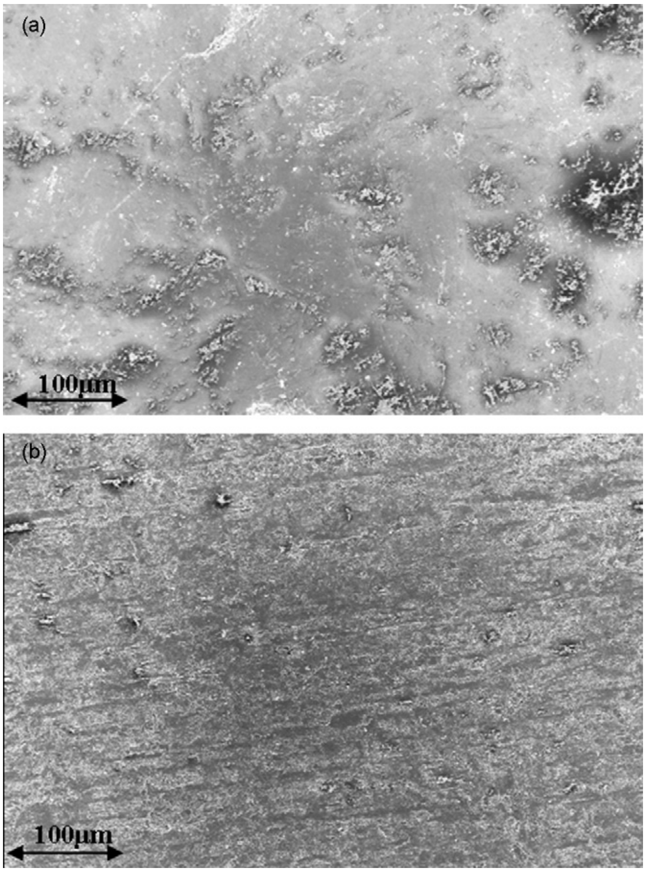

Fig. 3. (a) SEM image of BMC bipolar plate – unpolished surface. (b) SEM image of BMC bipolar plate – polished surface.

4.2. Sand paper polishing

Since mechanical polishing is the common method used to remove the polymer layer from the surface, sand papers of various grit sizes, as mentioned in Section 3.1, were used to polish the composite bipolar plates for removing the polymer layer. Table 1 shows the different grit-sized sand papers used for polishing the surface of the bipolar plate and the resulting bipolar plate roughness measured using a profilometer. The graph in Fig. 5 indicates that there is a linear trend between particle size of sand paper and the resulting roughness of the bipolar plate surface. Bigger particle sized sand papers result in increased surface roughness. Since different surface roughness values are obtained for the bipolar plate based on the grit size of sand paper used, it may be hypothesized that there will be a specific or a range of roughness values, corresponding to the grit size of sand paper, which can result in optimal lower contact resistance between the polished bipolar plate and the carbon paper.

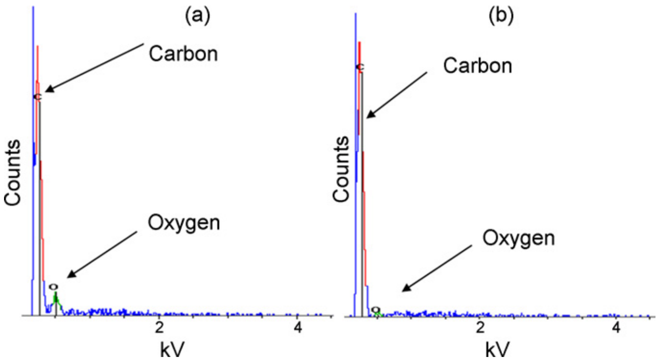

Fig. 4. (a) Energy dispersive spectra of BMC composite bipolar plate – unpolished surface. (b) Energy dispersive spectra of BMC composite bipolar plate – polished surface.

Table 1
Average roughness (R_a) of bipolar plates polished with various grit size* paper of given particle size.

Polishing sand paper		Bipolar plate roughness (μm)
Grit size	Particle size (μm)	
80	190	7.63
150	92	2.69
240	53	2.15
400	23	0.98
600	16	0.76
1000	10.3	0.44

* US CAMI grit designation, as mentioned in Section 3.1.

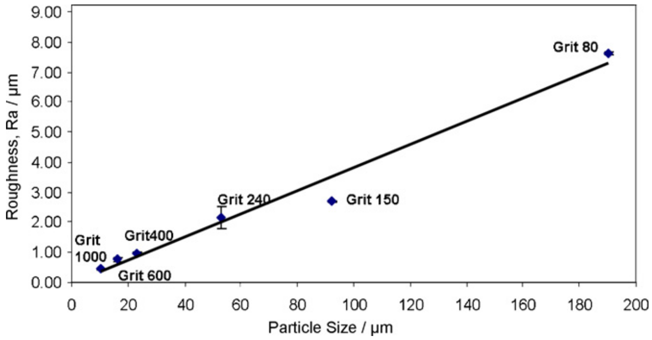

Fig. 5. Surface roughness of BMC composite bipolar plate vs. particle size of sand papers used for polishing.

Contact resistivity of the bipolar plate/GDL interface of various polished plates is measured using the model mentioned in Section 3.3 and is plotted against surface roughness of the bipolar plate. As can be seen in Fig. 6, the contact resistivity is nearly independent of the surface down to the samples with roughness of ∼1 μm. The plates polished using sand paper of grit sizes 80, 150, 240 and 400 did not vary drastically in contact resistance and the measured values are within the error bars. For the given conditions of graphite content in the composite bipolar plate and the type of carbon paper used, Grit 600 sand paper appears to be giving the optimal contact resistivity with roughness of 0.76 μm.

As the roughness decreases further, it may be assumed that the surface becomes smoother and the actual area of contact at the interface increases. Since pressure = force/area and the actual contact area at the interface increases, the actual pressure diminishes.

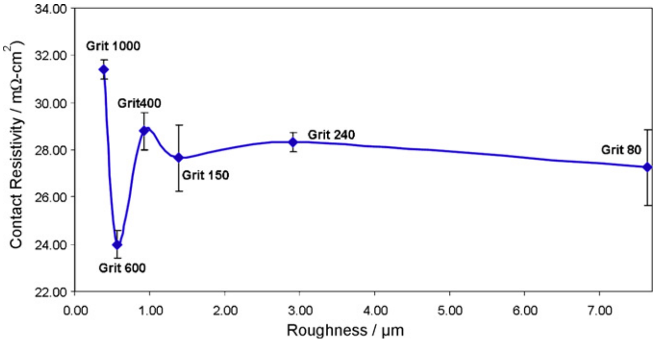

Fig. 6. Surface roughness vs. contact resistivity obtained from BMC composite bipolar plate surfaces (treated with different grit size sand papers) in contact with Toray paper at 0.5 MPa.

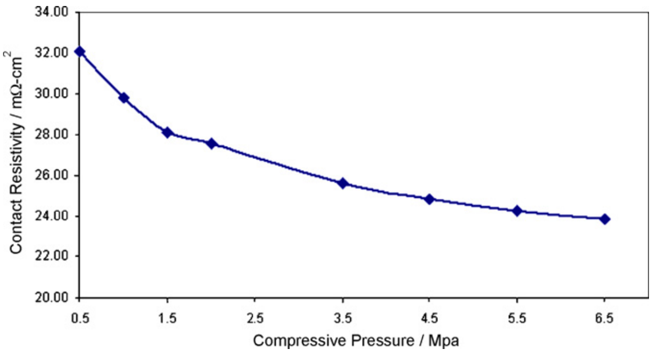

Fig. 7. Contact resistivity vs. compressive pressure for an unpolished BMC composite bipolar plate surfaces in contact with Toray paper.

This is because the load which is being applied in consideration to maintain the same force per unit of the sample apparent area is unchanged. The decrease of the pressure is proportional to the increase of the interface contact area, whereas, the dependence of contact resistance vs. pressure has non-linear and very steep character as shown in Fig. 7; as a result, the increase of the specific contact resistivity cannot be compensated by increasing the contact area and the overall resistance is expected to grow up as can be seen exactly in Fig. 6 for the 1000 grit sand paper polished plate whose surface roughness is ~0.46 μm. The plot in Fig. 6 shows high similarity to the contact resistivity behavior vs. surface roughness plot for the polished SS bipolar plates reported by Y. Ein-Eli's group [1].

5. Conclusions

The results and data show the significant impact of the bipolar plate's surface topography on the contact resistance of composite bipolar plate/GDL interface. Counter to the intuitive argument of smoothening the surface roughness to attain lower contact resistance, the above results show that a smoother surface may result in a steep increase of the contact resistivity at a given pressure. It is crucial to choose a suitable mechanical treatment and a surface topography for the bipolar plates to reduce the composite bipolar plate/gas diffusion layer interfacial contact resistivity. The optimal parameters of the bipolar plate surface roughness are to be chosen in consistency with the compressive pressure of the stack, porosity and mechanical properties of the chosen gas diffusion media, graphite content and design of the bipolar plate. It should be emphasized that no general composite bipolar plates/gas diffusion layer contact resistance interface model concerning the most appropriate bipolar plate surface roughness can be drawn from this study, since different gas diffusion layers may be applied in different fuel cell designs and the composite bipolar plates may also vary in graphite content, design or graphite particle size. It may be suggested that the choice of polishing method used for removing surface polymer layer of composite plates cannot be justified without a detail consideration of the bipolar plate cannot be adequately made without proper monitoring of the bipolar plate surface topography.

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