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# Effect of PTFE Content and Sintering Temperature on the Properties of a Fuel Cell Electrode Backing Layer

*The effects of the polytetrafluoroethylene (PTFE) content and sintering temperature on the properties of a fuel cell electrode backing layer are studied in this work. Characterization of the electrical conductivity, hydrophobicity, and surface structure of the backing layer is carried out for various PTFE content values (15–45 wt. %) and sintering temperatures (175–400 °C). The results showed that, generally, the electrical conductivity of the backing layer surface decreased whereas the hydrophobicity increased as the PTFE content and the sintering temperature increased. Based on the observations made via scanning electron microscope (SEM) analysis and testing of the electrical conductivity and hydrophobicity, the PTFE content should not exceed 35 wt. %, and the best sintering temperature was 350 °C. [DOI: 10.1115/1.4026932]*

*Keywords: backing layer, PTFE, conductivity, hydrophobicity*

## 1 Introduction

Proton exchange membrane fuel cells (PEMFCs) are devices that produce electricity from an electrochemical reaction between hydrogen gas (the fuel) and oxygen (the oxidant) with a proton exchange membrane serving as the electrolyte and a catalyst layer contained in the electrodes [1]. PEMFCs are widely considered to be promising energy conversion devices for vehicles, utilities, and mobile systems due to their high power density, energy efficiency, and zero emissions [1–3].

The membrane electrode assembly (MEA) is the core component of a PEMFC and the point where the conversion of hydrogen fuel and oxygen occurs to produce electricity and water. The MEA consists of two electrodes (a cathode and an anode) separated by a polymer electrolyte membrane (usually a Nafion membrane). The electrode itself consists of several layers comprising a backing layer (i.e., carbon paper or carbon cloth), a gas diffusion layer (GDL), and a catalyst layer. Some researchers mention that the backing layer was itself a GDL.

The backing layer is a layer that functions as an electrode holder and that retains moisture from the electrode. The backing layer must be able to conduct electrons well but be hydrophobic enough to reject and remove water and to easily transport gas to react with the catalyst layer [4]. Some factors that must be

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reviewed in an effort to improve the performance of the backing layer are the PTFE content, time, and sintering temperature.

An investigation of the influence of the PTFE content on the performance of the backing layer is expected to provide information about the optimal conductivity, hydrophobicity, and morphology of the backing layer (the role of the backing layer in the mass transport mechanism and in producing the reactant product has not been discussed extensively).

Wang et al. [5] analyzed the performance of carbon paper and carbon cloth in a PEMFC and showed that carbon cloth is the better option for use in high humidity conditions and that carbon paper is more suitable for dry conditions. Efforts to compare the performance of carbon paper and carbon cloth were also made by Radhakrishnan and Haridoss [6]. Radhakrishnan and Haridoss studied the effect of the structure of carbon paper and fabric design flows and found that the intrusion of carbon paper is smaller than that of the carbon cloth.

The effect of the backing layer thickness on the performance of the MEA was studied by Lin et al. [7]. Lin et al. used a nonwoven teflonized carbon with different thicknesses and carbon black mixtures of pure and 75% vapor-grown carbon fibers (VGCFs) and 25% PTFE in a 2-propanol ink solution as the backing layer. Lin only tested the performance of the GDL thickness of the backing layer and the addition of VGCF and found that a GDL thickness of 330  $\mu\text{m}$  has the highest current density of 3.0  $\text{mg}\cdot\text{cm}^{-2}$  carbon content compared with a thinner layer. Lin et al. also identified the role of VGCF in increasing the homogeneity of the GDL ink.

In addition to the influence of the type of backing layer and the thickness, research on the backing layer is also directed at understanding the influence of the hydrophobicity of the backing layer on PEMFC performance. Lim and Wang [8] conducted research to determine the effect of the hydrophobic fluorinated ethylene propylene (FEP) content in the GDL on the performance of a PEM fuel cell using Toray 090 as a backing layer. Lim and Wang found that the 10 wt. % FEP content in the GDL was best, considering the perspective of the current density. Meanwhile, Liu et al. [9] analyzed the influence of FEP content on the thickness, contact angle, gas permeability, and through-plane resistivity of the carbon cloth and determined that the thickness, contact angle, gas permeability, and through-plane resistivity increased with increases in the FEP content. In contrast, Chiu and Wang [10] used the addition of PTFE to improve the hydrophobicity of carbon cloth with a sputtering method to obtain results that were more successful than the normal spraying method. Chiu suggested the sputtering method because the typical method very often involves the blocking of macropores by large PTFE molecules.

The electrical conductivity of the backing layer was one important parameter in determining the performance of the fuel cell. Several researchers have investigated the conductivity of the backing layer. Zhou and Liu [11] investigated the effect of the conductivity of the GDL on the performance of the fuel cell. Zhou and Liu found that the performances increased if the conductivity was higher. Zhou and Liu also found that the through-plane conductivity has a direct effect on the ohmic loss and the in-plane conductivity effect on the overpotential distribution and the local current density.

Ismail et al. [12] described the effect of PTFE on the conductivity of a GDL comprising carbon paper from SGL Technologies GmbH, with and without the addition of PTFE. Ismail et al. determined that the PTFE content has an effect on the in-plane conductivity of the GDL. Ismail et al. [12] reported anisotropic in-plane conductivity. The conductivity measured along perpendicular directions varied by a factor of 2. Moreover, the addition of PTFE content does not change the conductivity significantly. The authors also found that there is not a correlation between the increase in the PTFE content in the microporous layer (MPL) and the conductivity.

Several methods for measuring conductivity were suggested by Mironov et al. [13]. They conducted research to compare the four electrode (FE) and the four point probe (FPP) methods to measure the electrical conductivity of a polymer composite based on

graphite. Mironov found that the conductivity difference between FE and FPP methods was due to inconsistencies and inhomogeneities in the composite material.

Zhang and Shen [14] analyzed the effect of temperature on the electrical resistance and the tensile strength of the backing layer, and they determined that the resistance and tensile strength decreased with increasing temperature using SEM and XRD observations.

The effect of PTFE content and sintering temperature on conductivity, hydrophobicity, and surface structure backing layer made of carbon paper Avcarb CPS P75 of Ballard will be assessed in this study. Studies on the effect of adding PTFE to the properties of the carbon paper is expected to be a comparison of what has been done by Chiu and Wang [10] and Lim and Wang [8]. While the influence of the sintering temperature is expected to be an input and proof of the best sintering temperature.

## 2 Materials and Methods

**2.1 Washing Carbon Paper.** The carbon paper used in this experiment was Ballard Avcarb CPS P75 (Canada), and the hydrophobic agent was 60 wt. % PTFE from Ion Power Inc. (USA). The carbon paper was washed with acetone for 10 min and dried in an oven at 110 °C for 30 min before use.

**2.2 Preparation of the Backing Layer.** The backing layer contained PTFE in the amounts of 15, 25, 35, and 45 wt. % (by weight of carbon paper). The carbon paper was dipped into the PTFE solution, heated at 105 °C for 30 min and then sintered for 3 h at 350 °C. The effect of the sintering temperature on the carbon paper was studied using carbon paper containing 35 wt. % PTFE at sintering temperatures of 175, 200, 250, 275, 300, 350, 375 and 400 °C for 3 h.

**2.3 Characterization of the Backing Layer.** The characterization of the backing layers coated with various amounts of PTFE and sintered at various temperatures included measurements of the surface electrical conductivity, morphological structure, and hydrophobicity of each backing layer sample.

**2.3.1 Electrical Conductivity Measurement.** The electrical conductivity of the backing layer was analyzed using the FPP method, followed by the Smith method with the Jandel RM3 model with eight observation points: four points on the top and four points on the bottom. On each side, measurements were performed for each of the two observation points in the 0 deg and 90 deg directions. Calculation of the conductivity follows the general equation for a thin layer using the FPP method, as demonstrated by Zhou and Liu [11], Mironov et al. [13], and Ismail et al. [12]. The electrical conductivity was measured in the in-plane (longitudinal) direction, which is commonly used to determine the conductivity of a homogeneous sheet [13] of carbon paper and to analyze the changes that have occurred [12].

**2.3.2 Morphological Investigations of the Backing Layer.** The morphological investigations of the backing layer include an analysis of the surface structure using SEM (model EVO MA10, Carl Zeiss, Germany).

**2.3.3 Hydrophobicity Measurement.** The hydrophobicity measurement was made using the EasyDrop contact angle meter DSA30S from Kruss, Germany, and additional visualization was performed using a Canon EOS 500D camera with a macro lens.

## 3 Results and Discussion

### 3.1 Backing Layer Characterization for Various Amounts of PTFE

**3.1.1 The Electrical Conductivity and Surface Hydrophobicity of the Backing Layer.** The electrical conductivity and hydrophobicity are two important parameters of the backing layer and of

the electrode in general. The electrical conductivity can illustrate the ability of the backing layer and the electrode to conduct electrons to and from the catalyst layer. Meanwhile, hydrophobicity is the ability to refuse and drain water. A good backing layer should have a high electrical conductivity and hydrophobicity. To increase the hydrophobicity of the backing layer, we must add a hydrophobic agent, PTFE, for example. However, adding PTFE can cause a decrease in the conductivity.

The relation between the electrical conductivity and the surface hydrophobicity of the backing layer is shown in Fig. 1.

Figure 1 indicates that the electrical conductivity of the backing layer generally decreases with increasing PTFE content. The decrease in conductivity occurred because PTFE is a hydrophobic agent and is not conductive. The decrease in conductivity is not too large (the conductivity decreases by  $5.8 \text{ S}\cdot\text{cm}^{-1}$  for each percentage increment of PTFE). According to Ismail et al. [12], the addition of PTFE does not significantly lower the backing layer conductivity.

Meanwhile, the hydrophobicity of the backing layer increased significantly with the addition of PTFE, starting with a content of 15 wt. % and continuing to increase with the increasing content of PTFE, although further increases were not too large. Hydrophobicity is the characteristic of water-repellent substances, defined as the tendency to repel and not absorb water. In this case, the hydrophobicity of the backing layer is the capability to reduce water-contact and eliminate water from the structure of the backing layer. A hydrophobic backing layer is needed to assist elimination of water and avoid water-clogging within the pores, which is caused by adhesion force between water and carbon substrate. On the other hand, the increment of hydrophobicity by addition of PTFE would reduce electrical conductivity. Hydrophobicity is measured by measuring the contact angle between the surface of the backing layer and a drop of water. A more hydrophobic surface will have a larger contact angle. In this experiment, the contact angle was measured using the Kruss EasyDrop equipment and a picture taken using the camera with a macro lens is shown for comparison.

To determine the level of homogeneity of the PTFE, conductivity measurements were taken on both sides of the backing layer in a direction perpendicular to the surface (0 deg and 90 deg), and the results are shown in Fig. 2.

Figure 2 shows that the electrical conductivities are not significantly different in the 0 deg and 90 deg directions. This lack of difference indicates that the distribution of PTFE onto the surface of the backing layer is homogeneous. The small differences indicate that the carbon paper coating process is homogeneous on both sides (top and bottom) and in two directions (0 deg and 90 deg). The homogeneous in-plane conductivity was very helpful

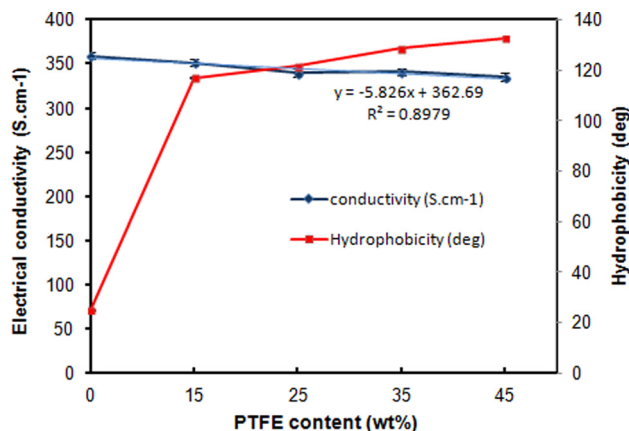


Fig. 1 Electrical conductivity of the surface of the backing layer at various PTFE content (wt. %)

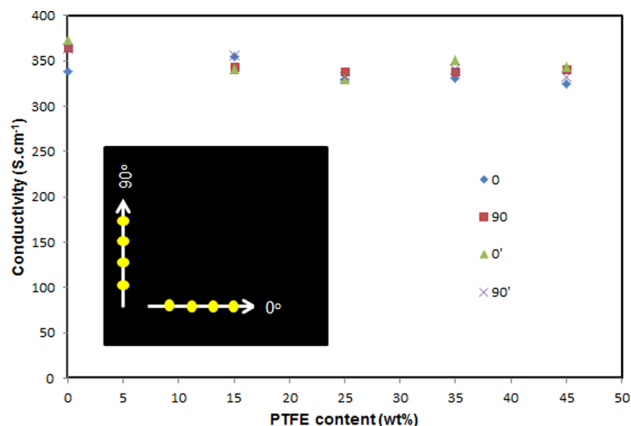


Fig. 2 Electrical conductivity of the surface of backing layer in the direction of 0 deg and 90 deg, respectively, to the top and bottom of the backing layer at various PTFE content

for facilitating electron transport at various points on the surface of the backing layer.

This finding is contrary to the results obtained by Ismail et al. [12] who found that the conductivity difference between 0 deg and 90 deg was approximately twofold. The reasons for differences between our results and those reported by Ismail et al. could be due to different values of PTFE concentration and the method of measurement (we measured the in-plane conductivity of the top and bottom side of the backing layer while Ismail et al. took only single-surface measurements). Moreover, in our research, we fabricate our own backing layer, but Ismail et al. used a commercialized backing layer.

Based on the result in Fig. 3, it can be observed that the higher the PTFE content, the higher the refuse of water. The water droplets form a more rounded shape and have a larger contact angle, which explains the increase in the hydrophobicity of the backing layer.

3.1.2 Structural Properties and Morphology. The structural properties and surface morphology of the backing layer were

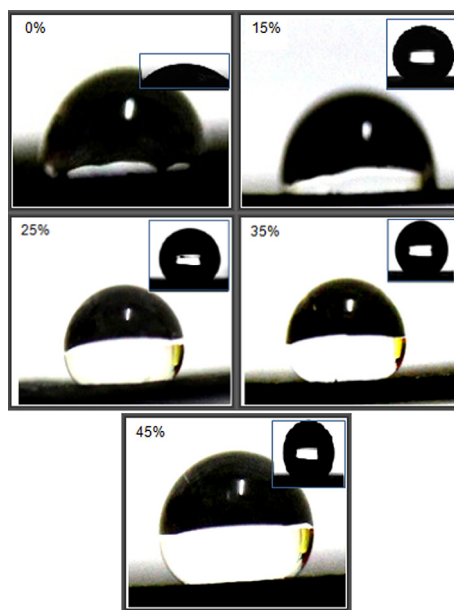


Fig. 3 Water droplets on carbon paper with the various contents of PTFE based on observations of ordinary cameras (inset picture from contact angle meter)



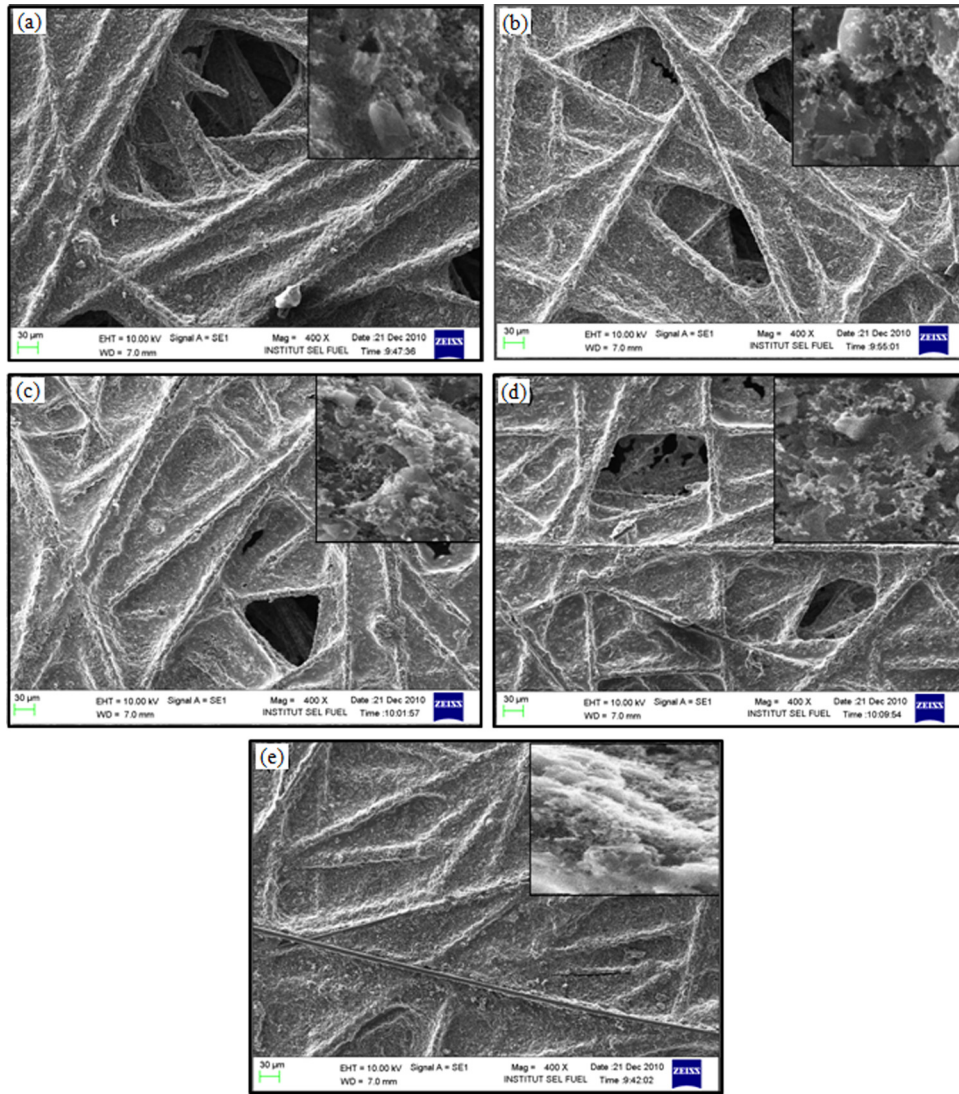


Fig. 4 SEM images of backing layer at various PTFE content: (a) 0 wt. % (carbon paper Avcarb P75 PCS (Ballard)), (b) 15 wt. %, (c) 25 wt. %, (d) 35 wt. %, (e) 45 wt. % (inset, magnification 20,000 $\times$ )

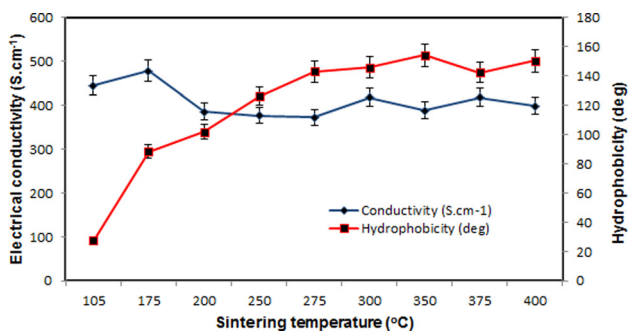


Fig. 5 The level of conductivity and hydrophobicity of the backing layer at various sintering temperatures

characterized using SEM. The results of the observation can be seen in Fig. 4.

Based on Fig. 4, increasing the PTFE content reduced the porosity of the backing layer. The optimum porosity of the backing layer was observed for 35 wt. % PTFE content. The samples with a PTFE content above 35 wt. % were covered with macropores. Based on the enlarged image (20,000 $\times$ , inset), increasing

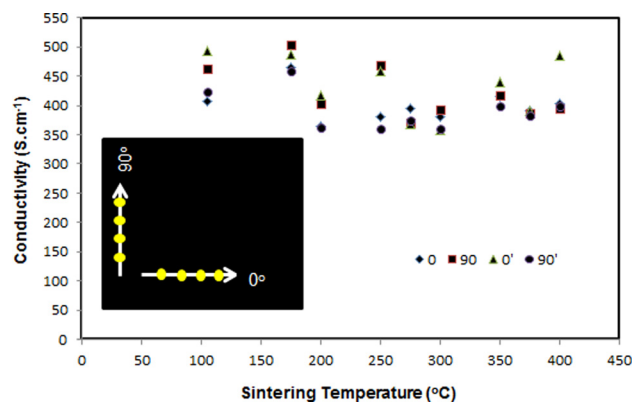


Fig. 6 Electrical conductivity of backing layer in the direction of 0 deg and 90 deg, respectively, to the top and bottom of the backing layer at various sintering temperatures

the concentration of PTFE covered the exterior of the backing layer surface. At 45 wt. % PTFE, the existing pores were covered. Thus, based on pore size, 35 wt. % PTFE is the highest content of PTFE that can be used.

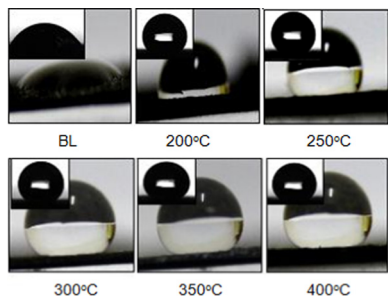


Fig. 7 Water droplets on the backing layer at various temperatures sintering ( $^{\circ}\text{C}$ ) based on observations of ordinary cameras (inset picture from contact angle meter)

### 3.2 Backing Layer Characterization at Various Sintering Temperatures

*3.2.1 Electrical Conductivity and Hydrophobicity Properties of the Backing Layer Surface.* Sintering is a method involving fusing together small particles by applying heat below the melting point. Sintering of the backing layer was performed to absorb and adhere the PTFE to the carbon rod from the backing layer through the pores. The impact of the addition of PTFE through the sintering process would lower the conductivity and increase the hydrophobicity. The electrical conductivity and hydrophobicity properties of the backing layer surface at various sintering temperatures are illustrated below in Fig. 5.

The surface electrical conductivity of the backing layer decreased dramatically after  $175^{\circ}\text{C}$  and tend to continue to decline with increasing temperatures up to  $275^{\circ}\text{C}$ . The surface

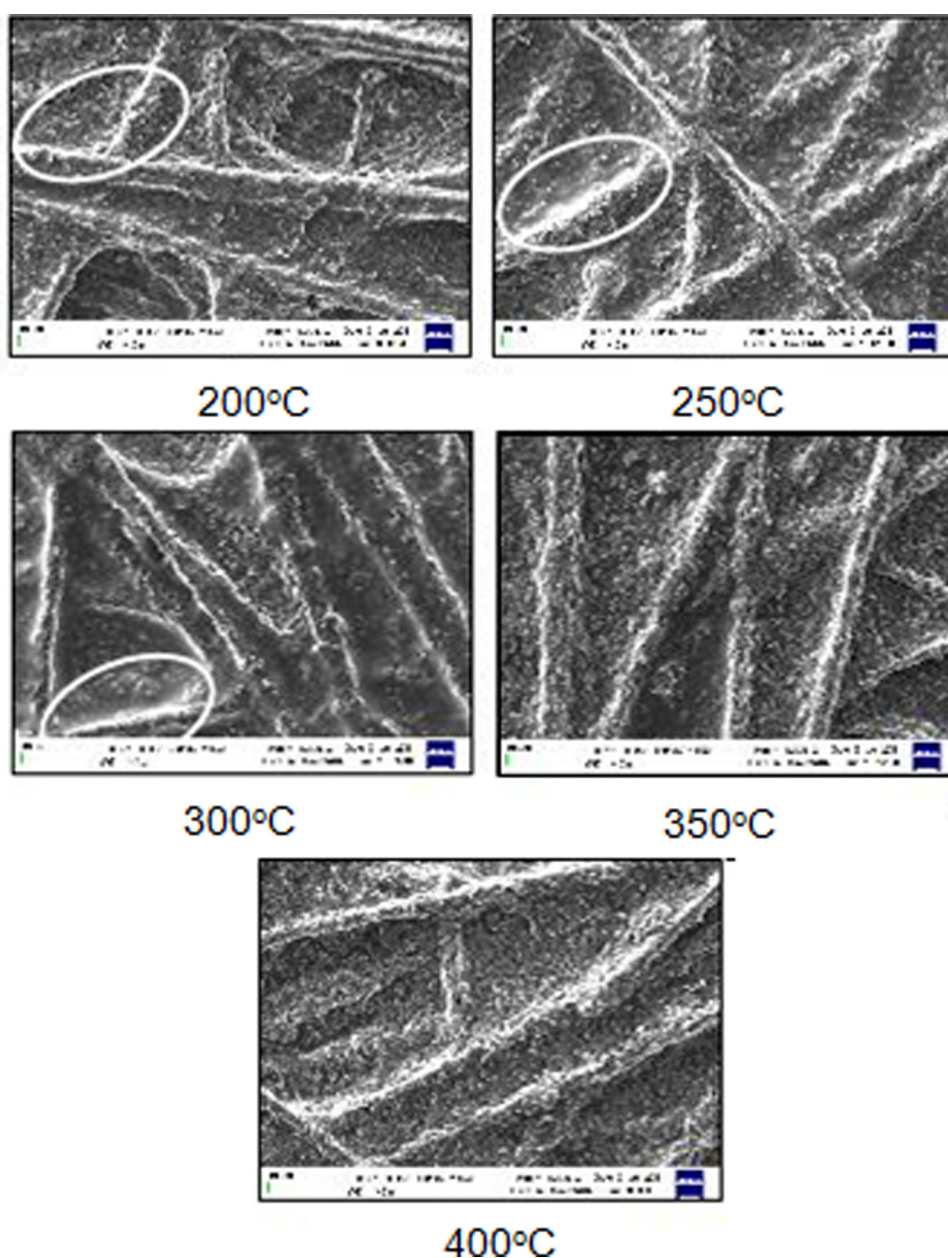


Fig. 8 SEM images of backing layer at various sintering temperatures

conductivity then increased again after 300 °C because a certain amount of PTFE is melted and absorbed into the structure of the backing layer. This phenomenon can be observed from the SEM images in Fig. 8. At the same conditions, the hydrophobicity of the backing layer increased as the sintering temperature increased and reaches a maximum at 350 °C.

The effect of the sintering temperature on the conductivity of the backing layer on both sides and in both directions is shown in Fig. 6.

Figure 6 shows that there were differences between the backing layer conductivity at various temperatures and in different directions. The value of the conductivities on the top and bottom of the backing layer were different. This difference indicates a nonhomogeneous PTFE coating, especially under 300 °C. This is understandable because under a temperature of 300 °C, PTFE did not melt, and in some areas, it still visibly accumulated (Fig. 8).

The effect of the sintering temperature on the hydrophobicity of the backing layer can be observed in Fig. 7.

Based on Fig. 7, it can be observed that at the higher sintering temperatures, water rejection from the backing layer is higher, which means an increased hydrophobicity. As mentioned before, there are correlations between increases in the electrical conductivity and decreases in the hydrophobicity.

**3.2.2 The Structure and Morphology of the Backing Layer.** The effect of the sintering temperature on the morphological structure of the backing layer can be observed in Fig. 8.

Based on Fig. 8, it can be observed that up to 300 °C, liquid PTFE was still visible on the backing layer. This liquid began to disappear above 300 °C. Thus, the best sintering temperature was above 300 °C, especially at 350 °C with maximum hydrophobicity value.

## 4 Conclusion

Important properties of a backing layer include high electrical conductivity and hydrophobicity. In order to achieve this, we have added a PTFE as hydrophobic substance to the backing layer. Based on our findings, hydrophobicity of the backing layer increases significantly as 15% PTFE is added and tends to increase further with the addition of the PTFE content. Meanwhile, the conductivity decreases by around 5.8 S·cm<sup>-2</sup> for each percentage increment of PTFE. On the other hand, the hydrophobicity of the backing layer increases with increasing sintering temperature, while under the melting point of PTFE, the surface electrical conductivity decreases with increasing sintering temperature.

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## References

- [1] Wang, Z.-B., Zuo, P.-J., Chu, Y.-Y., Shao, Y.-Y., and Yin, G.-P., 2009, "Durability Studies on Performance Degradation of Pt/C Catalysts of Proton Exchange Membrane Fuel Cell," *Int. J. Hydrogen Energy*, **34**(10), pp. 4387–4394.
- [2] Taylor, A. D., Kim, E. Y., Humes, V. P., Kizuka, J., and Thompson, L. T., 2007, "Inkjet Printing of Carbon Supported Platinum 3-D Catalyst Layers for Use in Fuel Cells," *J. Power Sources*, **171**(1), pp. 101–106.
- [3] Daud, W. R. W., Mohamad, A. B., Kadhum, A. A. H., Chebbi, R., and Iyuke, S. E., 2004 "Performance Optimisation of PEM Fuel Cell During MEA Fabrication," *Energy Convers. Manage.*, **45**(20), pp. 3239–3249.
- [4] Du, C., Wang, B., and Cheng, X., 2009, "Hierarchy Carbon Paper for the Gas Diffusion Layer of Proton Exchange Membrane Fuel Cells," *J. Power Sources*, **187**(2), pp. 505–508.
- [5] Wang, Y., Wang, C.-Y., and Chen, K. S., 2007, "Elucidating Differences Between Carbon Paper and Carbon Cloth in Polymer Electrolyte Fuel Cells," *Electrochim. Acta*, **52**(12), pp. 3965–3975.
- [6] Radhakrishnan, V., and Haridoss, P., 2011, "Differences in Structure and Property of Carbon Paper and Carbon Cloth Diffusion Media and Their Impact on Proton Exchange Membrane Fuel Cell Flow Field Design," *Mater. Des.*, **32**(2), pp. 861–868.
- [7] Lin, J. F., Wertz, J., Ahmad, R., Thommes, M., and Kannan, A. M., 2010, "Effect of Carbon Paper Substrate of the Gas Diffusion Layer on the Performance of Proton Exchange Membrane Fuel Cell," *Electrochim. Acta*, **55**(8), pp. 2746–2751.
- [8] Lim, C., and Wang, C. Y., 2004, "Effects of Hydrophobic Polymer Content in GDL on Power Performance of a PEM Fuel Cell," *Electrochim. Acta*, **49**(24), pp. 4149–4156.
- [9] Liu, C.-H., Ko, T.-H., Shen, J.-W., Chang, S.-I., Chang, S.-I., and Liao, Y.-K., 2009, "Effect of Hydrophobic Gas Diffusion Layers on the Performance of the Polymer Exchange Membrane Fuel Cell," *J. Power Sources*, **191**(2), pp. 489–494.
- [10] Chiu, K. F., and Wang, K. W., 2007, "Hydrophobic Coatings on Carbon Electrodes for Proton Exchange Membrane Fuel Cells," *Surface Coatings Technol.*, **202**(4–7), pp. 1231–1235.
- [11] Zhou, T., and Liu, H., 2006, "Effects of the Electrical Resistances of the GDL in a PEM Fuel Cell," *J. Power Sources*, **161**(1), pp. 444–453.
- [12] Ismail, M. S., Damjanovic, T., Ingham, D. B., Pourkashanian, M., and Westwood, A., 2010, "Effect of Polytetrafluoroethylene-Treatment and Microporous Layer-Coating on the Electrical Conductivity of Gas Diffusion Layers Used in Proton Exchange Membrane Fuel Cells," *J. Power Sources*, **195**(9), pp. 2700–2708.
- [13] Mironov, V. S., Kim, J. K., Park, M., Lim, S., and Cho, W. K., 2007, "Comparison of Electrical Conductivity Data Obtained by Four-Electrode and Four-Point Probe Methods for Graphite-Based Polymer Composites," *Polymer Test.*, **26**(4), pp. 547–555.
- [14] Zhang, X., and Shen, Z., 2002, "Carbon Fiber Paper for Fuel Cell Electrode," *Fuel*, **81**(17), pp. 2199–2201.