

## PREPARATION AND CHARACTERIZATION OF CARBON COMPOSITE PAPER FROM COCONUT COIR FOR GAS DIFFUSION LAYER

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### ABSTRACT

The gas diffusion layer (GDL) is one of the critical components of a proton exchange membrane fuel cell (PEMFC). It is generally made of a fossil-fuel-based carbon material. In this study, carbon composite paper (CCP) for GDL was prepared by using carbon material obtained from coconut coir. To obtain the CCP, 80 wt% carbon material from the coconut coir and 20 wt% polymer binder (ethylene vinyl acetate and polyethylene glycol) were mixed in xylene solvent at 100°C, cast on molded glass, and then rolled. The carbon material consists of a mixture of carbon fibers (length: 2 mm) and powders (size: 74 μm). Subsequently, the CCP was treated with polytetrafluoroethylene solution (10 wt%). The physical properties of the CCPs, such as through-plane electrical conductivity, porosity, density, and hydrophobic properties, were investigated. Scanning electron microscopy and energy-dispersive spectroscopy mapping were used to analyze the morphology and polytetrafluoroethylene (PTFE) distribution in the CCP. The through-plane conductivity test showed that CCP with 70 wt% carbon fiber, 10 wt% carbon powder, and 20 wt% polymer was the optimum sample, and it showed the highest electrical conductivity of 2.22 S cm<sup>-1</sup>. The physical properties of PTFE-treated CCP, such as porosity, density, and contact angle, were almost similar to that of commercial carbon paper used as a GDL. Therefore, the CCP prepared from coconut coir can be applied as a GDL in a PEMFC.

**Keywords:** Carbonization; Carbon composite paper; Coconut coir; Electrical conductivity; Gas diffusion layer

### 1. INTRODUCTION

A proton electrolyte membrane fuel cell (PEMFC) is an electrochemical device that continuously converts chemical energy via a reaction to electrical energy. A PEMFC has high efficiency and power density and low emissions, operation temperature, and noise (Bhatt et al., 2012). A PEMFC stack typically consists of a membrane-electrode assembly (MEA), gaskets, and bipolar plates. The MEA, which is the main component of a PEMFC, comprises two catalyst layers: a polymer electrolyte membrane (PEM) and two gas diffusion layers (GDLs). A GDL is a porous component whose main function is to distribute the fuel gas (hydrogen and oxygen), provide an electrical pathway, remove heat, and eliminate the water produced by the cell. For this purpose, the GDL should have physical properties such as high electrical mechanical strength. A GDL is typically a carbon-based material, and it is usually in the form

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of carbon paper or carbon cloth (Bhatt et al., 2012; Han et al., 2012). Generally, they are manufactured by high-cost processes and carbon materials from fossil fuels resources such as carbon nanotubes (Poochai & Pongprayoon, 2012), synthetic graphite (Han et al., 2012), polyacrylonitrile-based carbon fiber (Hung et al., 2012; Zhi-yong et al., 2010), and Vulcan XC-72 (Chen-Yang et al., 2007). Few studies have focused on preparing carbon composite GDL using carbon materials from natural fibers (Kinumoto et al., 2015). These carbon sources are renewable and are more abundant than fossil fuel resources. Carbon materials can be obtained by a simple carbonization process of natural fibers. Natural fibers are carbon sources that consist of complex organic compounds such as cellulose, lignin, and hemicelluloses (Kinumoto et al., 2015; Lee et al., 2014). Carbonization process performed on natural fibers can produce charcoal residue that has high carbon content (50%–80%) and is porous (Indayaningsih et al., 2011).

The present study describes the preparation of carbon composite paper (CCP) with carbon material from natural fibers, i.e., coconut coir. The first objective of this study is to produce CCP as a GDL using a simple process and inexpensive raw carbon materials. The second objective of this study is to investigate the electrical properties of CCP obtained from coconut coir by applying a combination of carbon material in the form of fibers and powders.

## 2. EXPERIMENTAL SETUP

### 2.1. Materials

Carbon materials were prepared from the pyrolysis products of coconut coir. Ethylene vinyl acetate (EVA) and polyethylene glycol (PEG) obtained from Aldrich Chemical Co., Inc. (St. Louis, MO, USA), were used as the binder and dispersant, respectively. Xylene obtained from Brataco Chemika was used as a solvent for making the CCP. Teflon emulsion polytetrafluoroethylene (PTFE) 30 obtained from Fuel Cell Earth LLC was used as a hydrophobic agent for CCP. Teflon emulsion PTFE 30 should be diluted to 10% concentration before being coated on the CCP surface. For the purpose of comparison, commercial carbon paper TGP-H-090 with 10% PTFE obtained from Toray was also studied.

### 2.2. Production Process of Carbon Materials

In this study, carbon material was used in two forms: carbon fiber and carbon powder. Both carbon materials were prepared from coconut coir by a two-stage process: carbonization and pyrolysis. In the carbonization process, coconut coir was heated at 500°C for 1 h under N<sub>2</sub> atmosphere and cooled down to room temperature to produce charcoal having high carbon content. Subsequently, the charcoal was pyrolyzed at 1300°C for 1 h under N<sub>2</sub> atmosphere to eliminate impurities and improve its electrical conductivity and other properties. The coconut coir was cut to a length of  $\pm 2$  mm before being subjected to carbonization and pyrolysis to produce the carbon fiber. The carbon powder was obtained by grinding the carbon material into a powder of 74- $\mu$ m size.

### 2.3. Preparation of CCP

To prepare a CCP, the carbon materials (carbon fiber and carbon powder) obtained from the previous processes were mixed with a polymer binder (EVA and PEG) in xylene solvent at 100°C for 2 h to form a slurry. The carbon materials:EVA:PEG mass ratio was 80:14:6 wt%. To form paper, the slurry was cast on molded glass, rolled, and then dried at room temperature for 24 h to evaporate the solvent. In this study, nine groups of samples were made with various carbon fiber and carbon powder compositions, as shown in Table 1.

The CCP with highest electrical conductivity was treated with a hydrophobic agent to improve its hydrophobic properties. The hydrophobic treatment was performed by dipping the CCP in 10 wt% PTFE suspension for 30 min and dried at room temperature for 24 h. Next, it was

heated at 150° in an oven for 30 min to evaporate the surfactant and sintered at 350°C for 30 min to melt PTFE.

Table 1 Variation of material composition of CCP

Sample	Carbon materials	
	Carbon fiber (wt%)	Carbon powder (wt%)
CCP-1	0	80
CCP-2	10	70
CCP-3	20	60
CCP-4	30	50
CCP-5	40	40
CCP-6	50	30
CCP-7	60	20
CCP-8	70	10
CCP-9	80	0

#### 2.4. Characterization of Carbon Paper

Characterization and analysis were carried out on CCP samples and commercial carbon paper as a comparison. The through-plane electrical conductivity was measured using a HIOKI 3522-50 HITESTER LCR-meter. The porosity and density were determined by the kerosene density method using the Archimedes principle in accordance with BS 1902: Part 1A standard. The hydrophobic properties were determined from the contact angle using a sessile drop test. For each measurement, a 50- $\mu$ L water droplet was placed on the carbon paper surfaces by placing the tip of the syringe close to the sample surface, and images were captured every 20 min for 1 h after the droplet was attached to the sample surface. Furthermore, the droplet shape was analyzed using Bashforth and Adams tables (Bashforth & Adams, 1883) to determine the contact angle of the samples. A HITACHI SU-3500 scanning electron microscope (SEM) was used to observe the surface and cross-section morphology of the samples. Energy-dispersive spectroscopy (EDS) mapping (X-maX, Horiba, Japan) was performed to analyze the PTFE distribution in the carbon paper.

### 3. RESULTS AND DISCUSSION

#### 3.1. Effect of Carbon Fiber Content Variation on Electrical Conductivity of CCP

Figure 1 shows the through-plane electrical conductivities of CCPs with different carbon fiber content (0%–80 wt%) before PTFE treatment. The electrical conductivity of CCP increases alongside number of carbon fiber content. The CCP-8 sample with 70 wt% carbon fiber has highest electrical conductivity. The carbon fiber has higher aspect ratio (L/D) than the carbon powder. The aspect ratio plays an important role in enhancing the electrical conductivity of the composites. Carbon fiber with high aspect ratio formed a more electrically conductive path than carbon powder. The conductivity of the composite increased with the number of electrically conductive paths formed on it. However, the CCP-9 sample with higher carbon fiber content (80 wt%) had lower electrical conductivity than the CCP-8 sample because the carbon powder (10 wt%) filled the gap between adjacent carbon fibers and formed a more electrically conductive path. Powders and fibers used in combination have greater influence on the electrical conductivity of composites compared to fibers alone (Wen & Chung, 2007).

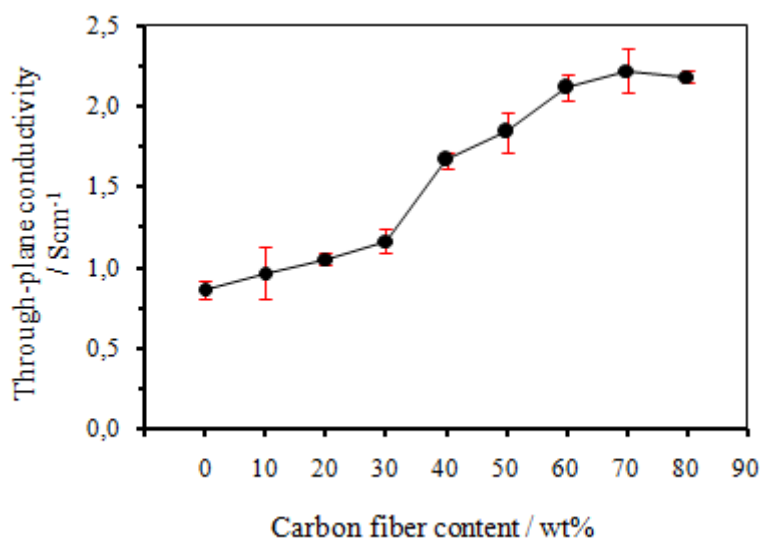


Figure 1 Electrical conductivity of CCP with various carbon fiber contents

### 3.2. Effect of PTFE Treatment on Physical Properties of CCP

PTFE treatment was performed on CCP-8, which had the highest electrical conductivity. This treatment is aimed at improving the hydrophobic property of carbon paper used as a GDL by avoiding water-flooding in the GDL. Table 2 shows the physical properties of CCP and commercial carbon paper. It is clearly seen that 10 wt% PTFE treatment of CCP improved the hydrophobic properties; the contact angle of CCP after PTFE treatment increased by 20.3° compared to that before the treatment. This value is comparable to that of commercial carbon paper.

Table 2 Physical properties of CCP and commercial carbon paper

Properties	Untreated CCP	PTFE-treated CCP	TGP-H-090
PTFE content (%)	0.00	10.00	10.00
Contact angle (°)	113.70	134.00	137.50
Bulk Density (gram/cm <sup>3</sup> )	...	0.42	0.48
Porosity (%)	...	73.63	75.74
Through-plane conductivity (S/cm)	2.22	2.09	5.12

The contact angle value shown in Table 2 was measured at the first observation after the droplet attached to the sample surface. Figure 2 shows the results of the contact angle measurements performed every 20 min for a further 1 h, and Figure 3 shows photographs of the water drops on the sample surface after 20 min. The contact angle of untreated CCP after 20 min was below 90°, and it decreased to 0° after 40–60 min. Untreated CCP shows hydrophilic characteristics owing to the absence of a hydrophobic agent on its surface. The contact angle of PTFE-treated CCP remained relatively constant from 0 to 60 min, as in the case of commercial carbon paper. This indicated that the hydrophobic properties of CCP obtained from coconut coir by PTFE treatment were comparable to those of commercial carbon paper. The porosity and bulk density of the PTFE-treated CCP are also almost similar to those of commercial carbon paper. Carbon paper used as a GDL allows the reactant gases to reach the reaction zones and the product water move out. To perform these functions effectively, carbon paper should have high porosity of 50%–90% (Yoon & Park, 2013); the porosity of PTFE-treated CCP satisfied this requirement. The effect of PTFE treatment on the electrical properties of carbon paper can be seen from the through-plane conductivity values listed in Table 2. PTFE treatment caused the electrical

conductivity of CCP to decrease slightly from 2.22 to 2.09 S cm<sup>-1</sup>. This was because the presence of nonconductive PTFE particles in the composite resulted in several conductive carbon materials failing to form electrically conductive paths. The electrical conductivity of PTFE-treated CCP was around 0.4 times that of commercial carbon paper.

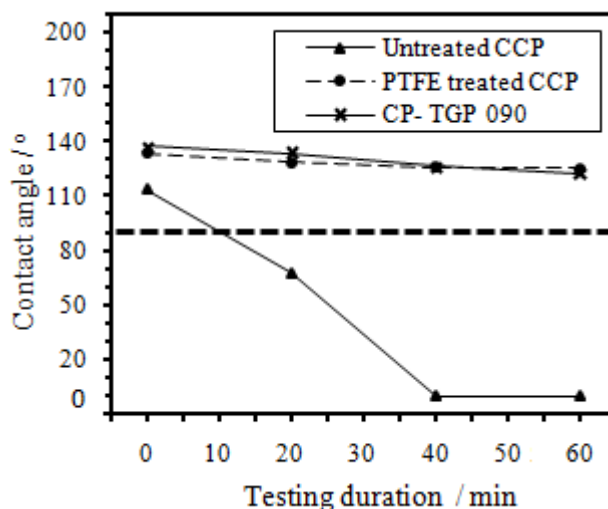


Figure 2 Contact angle of CCP obtained from coconut coir and commercial carbon paper

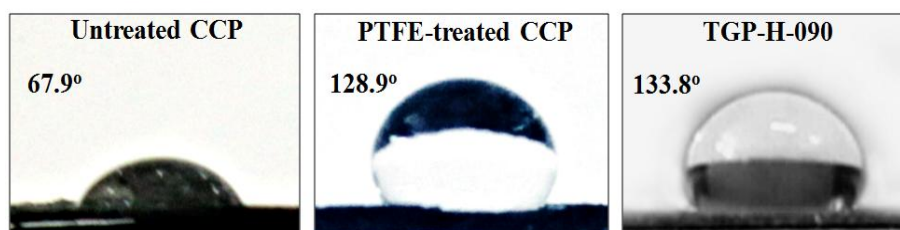


Figure 3 Photographs of water drops on untreated CCP, PTFE-treated CCP obtained from coconut coir, and commercial carbon paper at 20 min

### 3.3. Morphology and PTFE Distribution of CCP

Figure 4 shows SEM images of the surfaces of untreated CCP, PTFE-treated CCP, and commercial carbon paper. As shown in Figures 4a–4d, untreated CCP and PTFE-treated CCP show almost similar surface morphologies, namely, carbon fibers with length of 1–2 mm obtained from coconut coir. Observations with greater magnification, as shown in Figure 4b and Figure 4d, revealed that each carbon fiber comprises several hollow tubes. Furthermore, pores with ~1- $\mu$ m diameter were spread across almost the entire wall of the hollow tube. The main pores of the CCP were in the range of 25–250  $\mu$ m; they were formed by the internal spaces between carbon fibers. In contrast, commercial carbon paper comprises longer and smaller-diameter carbon fibers. This carbon paper has main pores only in the range of 10–100  $\mu$ m; they were formed by the internal spaces between carbon fibers and were spread homogeneously across the surface of the carbon paper. Figure 4f shows that the carbon fibers were solid and nonporous.

The distribution of the hydrophobic agent could be seen using SEM with elemental mapping on the cross-section of the CCP and commercial carbon paper, as shown in Figures 5 and 6, respectively. Based on the results of carbon mapping shown in Figure 5b, the CCP comprised carbon material (cyan-colored region) and pores (black-colored region). These pores were also seen on the surface, indicating that CCP has pores that are spread not only on the surface but

also in the deeper part of the surface. The same was observed in commercial carbon paper (Figure 6b).

Figures 5c and 6c showed fluorine mapping of the CCP and commercial carbon paper, respectively. These images indicate how far the PTFE (represented by elements of fluorine) dispersed in the carbon paper. As shown in Figure 5c, the PTFE particles (red-colored region) are spread on the surface and penetrate the CCP, as in the case of commercial carbon paper (Figure 6c). This indicates that hydrophobic material was deposited successfully in the CCP using the dipping technique, resulting in the improved hydrophobic properties of CCP, as shown in Figures 2 and 3.

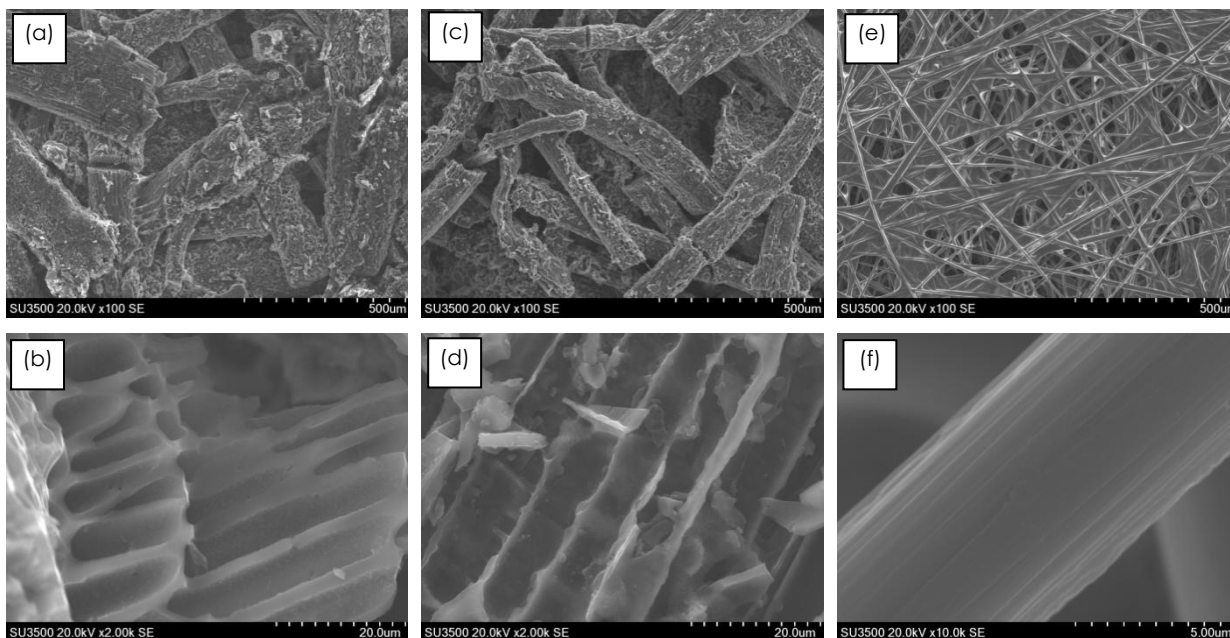


Figure 4 Surface morphologies of all samples under different magnifications: (a) untreated CCP-100 $\times$ ; (b) untreated CCP-2000 $\times$ ; (c) PTFE treated CCP-100 $\times$ ; (d) PTFE treated CCP-2000 $\times$ ; (e) commercial carbon paper-100 $\times$ ; and (f) commercial carbon paper-10000 $\times$

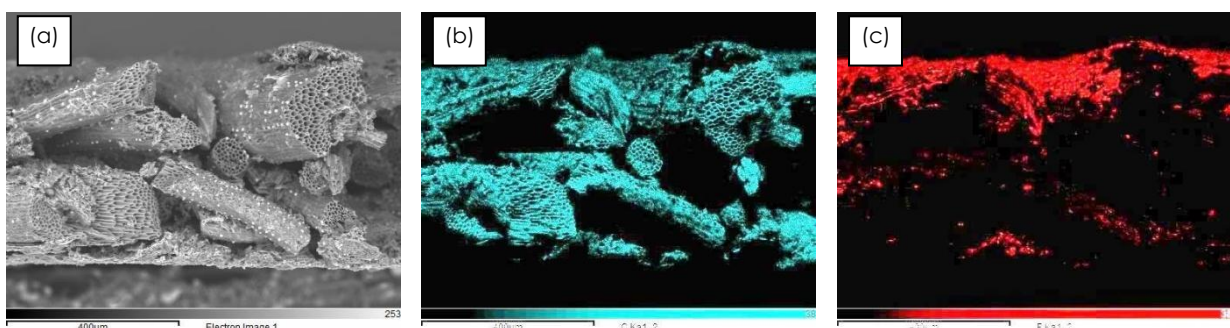


Figure 5 Cross-section morphologies and elemental mapping of PTFE treated CCP: (a) SEM image-160 $\times$ ; (b) carbon mapping; and (c) fluorine mapping



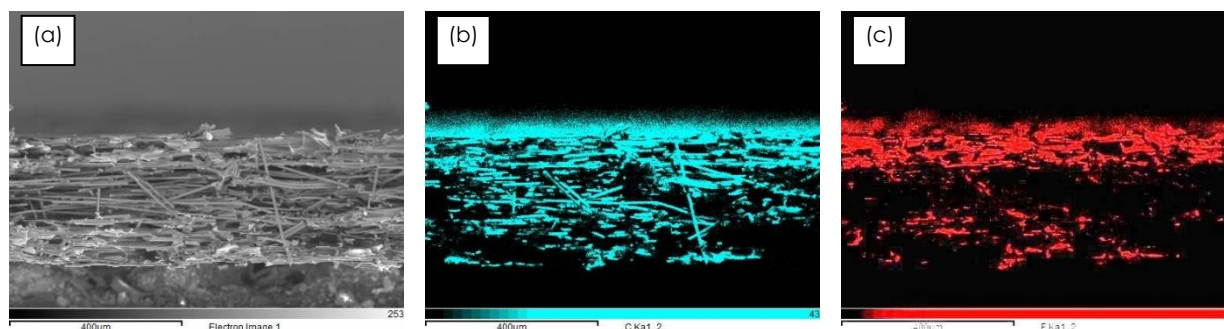


Figure 6 Cross-section morphologies and elemental mapping of commercial carbon paper: (a) SEM image-160 $\times$ ; (b) carbon mapping; and (c) fluorine mapping

#### 4. CONCLUSION

In this study, CCP was successfully prepared by using carbon materials obtained from natural fibers. The sample with 70 wt% carbon fiber, 10 wt% carbon powder, and 20 wt% polymer was the optimum one and showed the highest electrical conductivity (2.22 S cm<sup>-1</sup>). The combined use of carbon fiber and carbon powder may enhance the electrical connectivity of the composite, resulting in improved electrical conductivity. PTFE treatment of CCP reduced the electrical conductivity slightly but improved the hydrophobic properties. PTFE was successfully deposited in CCP using a dipping technique, resulting in improved hydrophobic properties of the CCP. Therefore, the CCP prepared from coconut coir can be applied as a GDL in a PEMFC.

#### 5. ACKNOWLEDGEMENT

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