



Properties and Performance of Gas Diffusion Layer PEMFC Derived from Coconut Coir

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Abstract. In this work, carbon composite papers (CCP) were successfully produced by mixing 80 wt% carbon derived from coconut coir and 20 wt% binder (ethylene vinyl acetate and polyethylene glycol). The CCPs were prepared with two different forms of carbon material, i.e. powder (particle size: $\pm 74 \mu\text{m}$) and fiber (length: $\pm 2 \text{ mm}$, diameter: $100\text{-}500 \mu\text{m}$). Two types of papers were developed based on their composition. The first type, called CCP-1, was made from carbon in powder form (80 wt%), while the second one, CCP-2, was based on a combination of fiber (70 wt%) and powder (10 wt%). The influence of the carbon form on CCP properties were investigated, including electrical conductivity, porosity, hydrophobicity, microstructure, and its performance as a gas diffusion layer (GDL) in a stack of proton exchange membrane fuel cell (PEMFC) system. Based on the results, CCP-1 showed a slightly better fuel cell performance than CCP-2, which was also confirmed by its lower porosity, electrical conductivity, and water contact angle. The effect of carbon composite paper's properties, including its morphology and performance, are discussed in this paper in detail and compared with a commercially based GDL material (TGP-H-120).

Keywords: *cell performance; coconut coir; electrical conductivity; fuel cell; gas diffusion layer.*

1 Introduction

Gas diffusion layers (GDLs) are one of the main components used in polymer electrolyte membrane fuel cells (PEMFC). As key components in the membrane electrode assembly (MEA) of PEMFC, GDLs have the main function of diffusing reactant gas into the catalyst layer, providing an electric pathway for electrons, eliminating the water product, removing heat, and providing mechanical support [1-3]. In order to perform these functions, GDLs should have appropriate porosity, good electrical conductivity, proper wetting characteristics, and enough strength [4-8]. They are typically a porous component mainly from carbon based materials and generally available in the form of carbon paper and carbon cloth [9-12]. Commercial GDLs are generally

manufactured using polyacrylonitrile-based carbon material, which is produced from non-renewable resources, i.e. fossil fuels [13-15]. This may raise concerns regarding sustainability. This issue led several researchers to develop GDLs using carbon material from biomass sources that are abundant and renewable. Biomass sources such as bamboo [15-16], oil palm empty fruit bunches [17], and coconut coir [18,19] have been proven to be utilizable as raw material for conductive carbon composite paper that is able to act as GDL component. However, based on a previous research [18], the electrical conductivity of GDLs prepared from coconut coir carbon in powder form is lower than that of commercial carbon paper, for instance Toray TGP-H-120 from Toray Group. This will decrease fuel cell performance significantly. Therefore, two carbon composite papers (CCP) were prepared by applying a combination of carbon material from coconut coir in different forms (fiber and powder) to achieve a better GDL material in view of improving its conductivity and performance. Conductivity improvement of CCP has been successfully performed using this method in a previous study [19], whereas its influence on cell performance still needs to be investigated further. The main objective of this study was to investigate the influence of different carbon material forms on the properties of the two CCPs and their performance as GDLs in a PEMFC system.

2 Material and Method

2.1 Materials

Coconut coir was the raw material utilized as carbon source in this work. Ethylene vinyl acetate (EVA) and polyethylene glycol (PEG) obtained from Aldrich Chemical Co., Inc. (St. Louis, MO, USA), were used as binder and dispersant agent, respectively. Xylene (Brataco Chemika) was used as solvent. Teflon emulsion polytetrafluoroethylene (PTFE) 30 obtained from Fuel Cell Earth LLC was used for hydrophobic treatment of CCP. For comparison purposes, commercially available carbon paper TGP-H-120 obtained from Toray Group was also investigated.

2.2 Coconut Carbon Production

In this work, carbon material was produced in two different forms: (i) carbon fiber and (ii) carbon powder. Both carbons were obtained from coconut coir by carbonization and pyrolysis. For the carbonization process, the coconut coir was heated at 500 °C for 1 h under N₂ atmosphere. The carbonization product was then pyrolyzed at 1300 °C for 1 h under N₂ atmosphere to obtain a carbon material with high carbon content. For the final product, carbon powder was obtained by grinding and then mesh sieving to a size of about 74-μm. For the

carbon fiber process, the coconut coir was first cut to a length of about 2 mm before being subjected to carbonization and pyrolysis.

2.3 CCPs Preparation and Characterization

The CCPs were prepared by mixing the carbon materials with EVA and PEG in xylene solvent at 100 °C for 2 h to form a slurry. The slurry was cast on molded glass, rolled, and then dried at room temperature for 24 h to evaporate the solvent to form paper. In this work, two types of CCPs were made with different shapes of carbon. The first type (CCP-1) was composed of carbon powder, while the second one (CCP-2) was composed of a combination of carbon fiber and powder. The composition details of each type of CCP are summarized in Table 1. These compositions were applied in accordance with the results from a previous research [19], where CCP-1 had the lowest conductivity and CCP-2 had the highest conductivity. Both types of CCPs were treated to improve their hydrophobic properties by dipping in PTFE suspension for 30 min and drying at room temperature for 24 h. Then, they were heat treated at 150 °C for 30 min and at 350 °C for 30 min in air atmosphere.

Table 1 Composition details of CCPs.

Substrates	Composition (wt %)		EVA (wt %)	PEG (wt%)
	Carbon powder	Carbon fiber		
CCP-1	80	-	14	6
CCP-2	10	70	14	6

The CCPs and commercial carbon paper (TGP-H-120) were characterized using the same instruments and techniques for equitable comparison. The surface and cross-section morphology of the samples were imaged using a HITACHI SU-3500 scanning electron microscope (SEM). A HIOKI 3522-50 HITESTER LCR-meter was used to measure the through-plane electrical conductivity. Meanwhile, the porosity and bulk density were determined using the Archimedes method in a kerosene medium based on the BS 1902: Part 1A testing method. Contact angle measurement was conducted using a sessile drop test. The measurements were analyzed using Bashforth and Adams tables [20] to determine the hydrophobic properties.

2.4 MEA and Cell Performance of PEMFC Test

In this work, a commercially available catalyst coated membrane (CCM) was applied for the three different tested GDLs, namely CCP-1, CCP-2, and commercial Toray paper (TGP-120). The GDLs were assembled at both sides of the membrane. The MEA performances were evaluated using a polarization curve, or J-V curve. A commercial PEMFC single cell test fixture (WonATech) with an active area of 25 cm² was used to test the cell performance using a

SMART2 test station (WonATech). Pure hydrogen and oxygen were fed into the anode and cathode compartment respectively, at a flow rate of 300 ml/min, operated in open-end conditions. The cell and reactant humidification temperature were set at 55 and 50 °C, respectively, to maintain proper hydration of the membrane. All GDL samples were activated before cell performance measurement was carried out by operating the cell at 0.6 volt for an hour. This activation process was performed under the same conditions as the cell performance measurement process.

3 Result and Discussion

3.1 Morphology of Carbon Papers

Figures 1 and 2 show the microscopic surface and cross section morphologies of CCP-1, CCP-2, and the commercial carbon paper (TGP-H-120), respectively. The surface of CCP-1 with carbon material in powder shape appears smooth with a homogenous distribution of pores with sizes of about 1-50 μm , Figure 1(a). The carbon powder seemed uniformly distributed in the matrix binder. This surface morphology is very similar to the micro-porous layer (MPL) on the carbon paper investigated by Shim, *et al.* [21]. The pore morphology can be seen more clearly in the cross section view of CCP-1 (Figure 2(a)), which had a thickness of about 370 μm . The insert of Figure 2(a) clearly shows the original shape of the coconut fiber with some parallel hollow tube-like shapes close to each other with a diameter of 2-10 μm . Figure 1(b) reveals the surface morphology of CCP-2, a composite of carbon fiber and powder. From the top view of the surface it can be seen that the morphology is completely different from that of CCP-1. It shows a discontinuous fiber-like shape with larger dimensions and a higher aspect ratio (L/D), spread randomly throughout the composite surface. Seen from the top, the pore size is about 25-250 μm , i.e. much lower than for CCP-1. The pores are formed by the internal spaces between the carbon fibers. These fibers can be seen more clearly at higher magnification in the cross section view of CCP-2 in the insert of Figure 2(b). It reveals that the fibers have a hollow tube-like morphology that is similar to that of CCP-1 (Figure 2(a)). The fibers appear to be arranged irregularly and form pores with larger sizes than those of CCP-1. Meanwhile, Figure 1(c) and 2(c) show the surface and cross section morphology, respectively, of the commercial carbon paper (TGP-H-120). It can be seen that commercial carbon paper is composed of solid and non-porous carbon fiber with a finer diameter and higher aspect ratio compared with CCP-2. Seen from the top the pore size is about 10-100 μm . The pores are formed by the internal spaces between the carbon fibers, as in CCP-2.

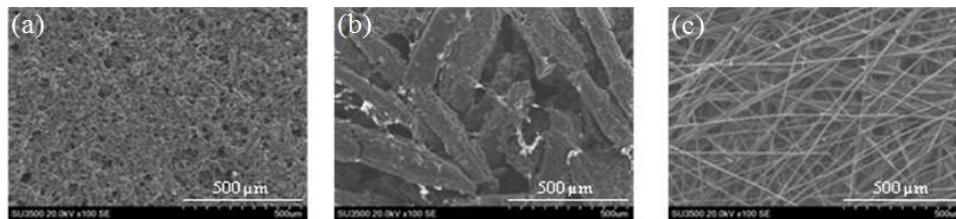


Figure 1 Surface morphologies of (a) CCP-1; (b) CCP-2; (c) commercial carbon paper.

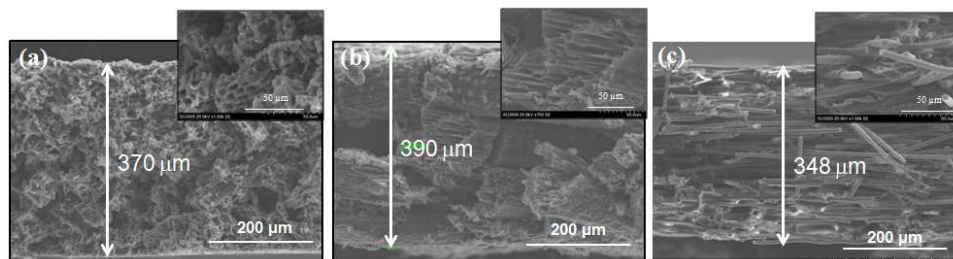


Figure 2 Cross section morphologies of (a) CCP-1; (b) CCP-2; (c) commercial carbon paper.

3.2 Physical Properties of Carbon Papers

The physical properties of CCP-1, CCP-2 and the commercial carbon paper are summarized in Table 2. It shows that CCP-1 and CCP-2 have significantly different values of electrical conductivity and porosity. The combined use of carbon in fiber and powder shape in CCP-2 clearly enhanced the electrical conductivity compared with the CCP-1 (pure carbon powder shape). This finding is consistent with the work of Wen, *et al.* [22] and Shen, *et al.* [23]. In CCP-2, carbon fibers with high aspect ratios were interconnected with each other (Figure 1(b)), thus establishing a continuous electrically conductive path. Hence, the electrons can be transported over a longer continuous path with fewer barriers than in CCP-1. This can also be explained by the carbon powder filling the gaps between adjacent carbon fibers, which creates more interconnected fibers. Thus, the increasing number of conductive paths leads to higher electrical conductivity of the composite CCP-2 than that of CCP-1. However, the conductivity of the commercial carbon paper was still much higher than that of our developed samples. As described above, CCP-1 had pores with smaller sizes and in greater numbers compared with CCP-2 (Figure 1(a-b) and 2(a-b)), spread homogeneously throughout the composite. This probably causes a higher porosity of CCP-1 than that of CCP-2, and a lower bulk density. Furthermore, all the tested materials exhibited relatively similar hydrophobic properties, while their contact angle value was not significantly

different. The ideal hydrophobicity for GDL has a contact angle between 90° and 180° based on water as wetting agent [24].

Table 2 Electrical and physical properties of CCPs and commercial product.

Parameter	CCP-1	CCP-2	TGP 120
Electrical conductivity (S/cm)	0.50	1.53	4.50
Porosity (%)	74.24	69.45	74.02
Bulk density (gram/cm ³)	0.43	0.47	0.50
Contact angle (°)	139	137	131
BET surface area (m ² /gram)	15.72	14.86	213.80
Average pore diameter (nm)	4.02	6.49	5.56

3.3 Cell Performance from PEMFC Test

Figure 3 shows the performance test of the GDLs based on CCP-1, CCP-2, and the commercial product (TGP-H-120). The curve indicating the MEA performance based on CCP-1 and CCP-2 is almost the same for both, especially for low current density (with voltage higher than 0.7 V). The slight difference between these two types of as-developed GDLs is noticeable only in the medium and high current density region (about 100-500 mA/cm²). As is already known, the polarization curve of the J-V curve is the most important characteristic and is a convenient tool for the design and optimization of PEMFC systems [25]. There are three main parameters that can be obtained directly from the J-V curve to evaluate cell performance, namely: (i) open circuit voltage (OCV), (ii) current density at operation voltage of 0.6 V ($J_{0.6}$), and (iii) maximum power density (P_{max}), as can be seen in Table 3.

From the J-V curve, voltage losses or overpotential of the cell can be estimated. During the operational test, the cell potential is decreased from its ideal performance due to three main overpotentials, namely: (i) activation, (ii) ohmic, and (iii) diffusion overpotential [26,27]. Activation overpotential dominates at low current density and is related to the slowness of the reaction that is taking place on the electrode surface. This overpotential highly depends on the activity of the catalyst sites. In Figure 3 it can be seen that the J-V curves of the cell based on the three different GDLs (CCP-1, CCP-2, and TGP-120) are almost identical at low current density. This shows that the catalytic activity of the catalyst layer of the GDLs was almost the same, because the three GDLs used the same CCM catalyst. Starting from a current density of 50 mA/cm² and clearly noticeable in the middle current density region (about 100-400 mA/cm²), the three GDLs revealed different performances.

At the middle current density known as the ohmic overpotential region, the performance losses are dominated by the electrical resistance of the electrode

and the resistance of the ion (proton) flow in the electrolyte. Because the three GDLs used the same CCM catalyst and test apparatus, the ohmic overpotential can be attributed to the electrical conductivity of the GDLs, as shown in Table 2, where the conductivity of TGP-H-120 is much higher than that of CCP-1 and CCP-2. On the other hand, the J-V curves of CCP-1 are slightly higher than those of CCP-2 in this current density region, whereas the conductivity of CCP-2 is higher than that of CCP-1.

This indicates that high electrical conductivity only cannot directly lead to high performance of the cell. This can also be contributed to the surface morphology and pore size of the GDL itself. CCP-1 has a smoother surface and smaller pore sizes than CCP-2 (Figure 1(a) and 1(b)). These two factors probably prevent the catalyst ink from penetrating into the pores of the carbon paper (during the preparation process) [28] and this will affect the reduction of the contact resistance of the catalyst layer/GDL [10], as illustrated in Figure 4.

The third contribution of the overpotential is diffusion or mass transport. This overpotential is clearly shown at high current density and it appears since there is a concentration gradient of reactant due to the limited supply of reactants. At cell voltage of 0.3 V, the cell based on TGP-H-120 had a current density of 580 mA/cm², whereas for CCP-1 and CCP-2 it was 476 and 488 mA/cm², respectively. From previous results, diffusion overpotential is closely related to porous electrodes, since the pore size distribution and hydrophobicity of the GDL influences the possibility of water flooding inside the cell [29]. However, in this study, apart from the three GDLs having a similar porosity and contact angle (Table 2), differences in mass transport overpotential were still strongly detected.

This phenomenon could be related to the different structure of the GDLs, especially for CCP-1 and CCP-2. At high current density, GDL has two main functions. Firstly, it can effectively transport sufficient reactants to the electrodes, and secondly, it provides a good water management system that facilitates water vapor to flow out from active sites to flow field plates in order to avoid flooding inside the cell. It has been reported that roughness features presented on the surface of the GDL contribute highly to water pinning and evaporation. A lower surface roughness or smoother surface of the GDL indicates higher pinning and lower water evaporation [30]. This means that CCP-1, which has a smoother surface, as shown by the SEM image in Figure 1, would need more time to remove water droplets into flow channels compared to CCP-2. This could increase the possibility of water clogging and hinder the reactant distribution, as indicated by the lower J-V curve in the high-current region.

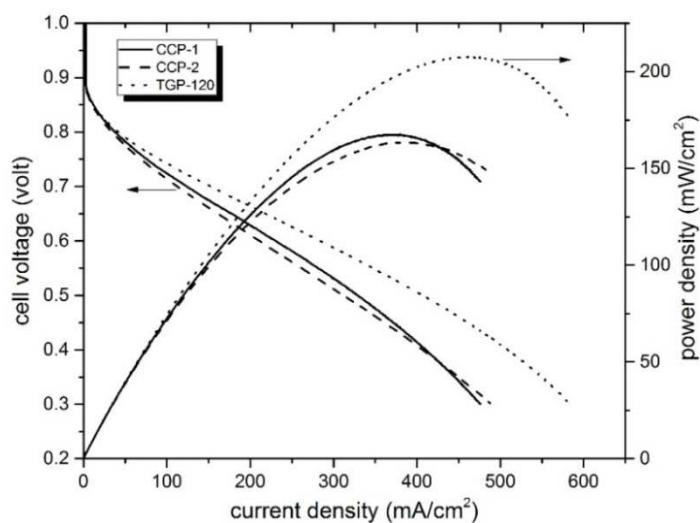


Figure 3 Cell MEA performance based on GDL CCP-1, CCP-2, and commercial carbon paper.

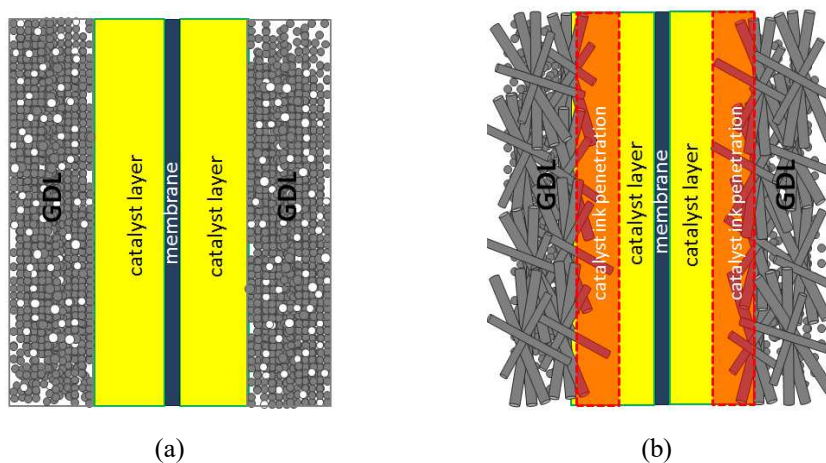


Figure 4 Schematic representation of MEA with (a) CCP-1 and (b) CCP-2 as GDL.

Table 3 Parameters of cell performance for the three kinds of MEAs.

Substrates	OCV (V)	$J_{0.6}$ (mA/cm ²)	P_{max} (mW/cm ²)
CCP-1	0.997	232	168
CCP-2	0.979	212	164
Commercial carbon paper	1.000	284	208

4 Conclusion

In this study, two carbon papers for use as GDLs were successfully prepared with different forms of carbon material, i.e. powder and fiber. SEM micrographs revealed that the carbon paper prepared with carbon powder (CCP-1) had a smoother morphology and smaller pores compared to the carbon paper composed of fiber and powder (CCP-2). This prevented the catalyst ink from diffusing into the pores of the carbon paper and reduced the contact resistance between the catalyst layer/GDL. This also resulted in a higher current and power density of CCP-1, whereas, its electrical conductivity was lower than that of the other carbon papers that were tested.

Acknowledgments

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