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PERFORMANCE OF GAS DIFFUSION LAYER DERIVED FROM CARBON POWDER OF COCONUT COIR FOR PEMFC APPLICATION

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ABSTRACT

Gas diffusion layer (GDL) plays an important role in polymer electrolyte membrane fuel cell (PEMFC). It provides diffusion media that transport reactant gas into catalyst layer, mechanical support for electrolyte membrane, electrical pathways for electron, and serves channel product water away from the electrode. In this study, the main physical properties (hydrophobicity, electrical conductivity, and porosity) and the performance of GDL made from activated carbon powder from coconut coir were investigated. Its properties were also compared to commercially available GDL, Toray TGP-H-120 (TGP-120). Gas diffusion layer coconut carbon paper (CCP) was fabricated by mixing the coconut carbon powder, ethylene vinyl acetate, polyethylene glycol, and xylene using hot plate stirrer at 100°C. The carbon slurry was casted to make carbon paper and then dried at room temperature. The carbon paper was also treated with PTFE solution to improve its hydrophobicity. SEM images showed that CCP had smoother surface morphology and denser compared to TGP-120, however, its porosity was quite similar, 69% and 74%, respectively. This was estimated because CCP had internal porous on the surface of coconut fibres. Although low conductivity of CCP still needed a further improvement, its relatively high hydrophobicity would be benefited the water management inside the cell. By measuring the cell performance in a single cell proton exchange membrane fuel cell (PEMFC) with an active area of 25 cm^2 using H_2/O_2 reactants, CCP showed a good fuel cell performance with current density of about 232 mA/cm^2 at 0.6 volt, compared to 284 mA/cm^2 for TGP-120. Power peaks were revealed at 168 and 208 mW/cm^2 for both CCP and TGP-120, respectively.

Keywords: gas diffusion layer, activated carbon, coconut coir, PEMFC.

INTRODUCTION

Polymer electrolyte membrane fuel cell (PEMFC) has high potential to reduce the needs of fossil fuel energy and pollutant emissions. It converts a fuel (hydrogen) and oxidant (oxygen/air) into electricity with water as the by-product by electrochemical reaction. Moreover, its low operating temperature that assuring instant start-up makes a PEMFC applicable for power sources of portable electronics, electric cars, and backing-up telecommunication systems [1].

Gas diffusion layer (GDL) are one of the key components in membrane electrode assembly (MEA) used in PEMFC. It plays essential roles to diffuse reactants into catalyst layer, to ensure sufficient humidification by allowing water vapour to diffuse into MEA, and to facilitate the removal of liquid water to prevent flooding inside PEMFC. Gas diffusion layer also serves as supporting structure for elastic membrane and plays as electrical connection that transfer electrons between catalyst layer and flow field [2]. Gas diffusion layer is made of porous electrically conductive materials that mainly from carbon based. Commercially available, there are two types of GDL, carbon paper and carbon cloth [3-4]. In carbon paper, the carbon fibres are held together in a carburized resin matrix, while in carbon cloth no such binder is necessary as the fibres are woven. Not only differ in their structure, those GDLs also have different properties, such as mass transport, mechanical strength, porosity, hydrophobicity, and conductivity.

Many studies have been carried out to investigate the performance of GDL based on carbon cloth or carbon paper. For example, researchers have found that performance of carbon cloth was higher than carbon paper at high humidity and high current density, while carbon paper was more superior in dry condition [5-7]. However, only a few researchers that studies on the preparation of GDL without carbon paper or carbon cloth have been reported [8]. Moreover, as commercial carbon materials for GDL are mainly produced from fossil fuel sources, it raises concerns about sustainability. To overcome this issue, some researchers have started to develop GDL derived from carbon of biomass sources. Taro et.al used carbon from bamboo [9]. Based on previous research, carbon coconut coir could also be a promising source for GDL because it has unique structure [10]. Carbon coconut coir has many parallel hollow like a tube and along the tube surface there is micro porous that could be beneficial in distributing reactant gases.

In this study, carbon composite GDL was fabricated by mixing carbon powder made from coconut coir with a polymer binder, namely ethylene vinyl acetate. The main properties of coconut carbon paper (CCP) that affect fuel cell performance, such as hydrophobicity, electrical conductivity, and porosity were investigated. Surface and cross section morphology of the GDL have also been examined using scanning electron microscopy. In additional, a single cell operating test of CCP for GDL material was demonstrated for the first time. Comparing to the commercial one, Toray carbon paper of TGP-H-120 has been used as reference.

EXPERIMENTAL

Coconut carbon powder production

Coconut carbon powder was produced by two main steps, namely carbonized and pyrolysis process. In the carbonization process, coconut coir was carbonized at temperature of 500°C for an hour in furnace with nitrogen atmosphere. This step was used to degrade the chemical compound of the coconut coir and produce charcoal. After that the charcoal was moved to further process of pyrolysis at temperature of 1300°C for an hour to improve its carbon quality. Before used to make GDL, the carbon fibres were ground into carbon powder in size of 200 meshes (75 μm).

Gas diffusion layer fabrication and characterization

Carbon coconut paper was fabricated by mixing the coconut carbon powder (80 wt. %) and polymer (20 wt. %). The adhesive polymer used in this study was ethylene vinyl acetate (70 wt. %) and a plasticizer of poly ethylene glycol (30 wt. %). The mixture was stirred in xylene at temperature of 100°C for 90 min. Next, the mixture was cast using tape casting technique and dried at room temperature. To improve its hydrophobicity, the CCP was treated with polytetrafluorine ethylene (PTFE) solution 10 wt. % (30 min) and then hot – treated at temperature of 350°C (30 min). As comparison, Toray carbon paper (TGP-H-120) was used as a reference.

A scanning electron microscopy (Hitachi SU3500) was used to observe the surface morphology and cross – section of the GDL CCP and TGP-120. The cross-sectional surface of the samples was mechanically fractured after immersion in liquid nitrogen. Electrical conductivity of the samples was characterized using HIOKI 3522-50 LCR-meter Hitester. To estimate the hydrophobicity, contact angle was measured using sessile drop method, and it was calculated by Bashforth and Adam Table [11]. Moreover, the GDL porosity was determined using Archimedes principle in accordance with BS 1902: Part 1A:1966 standard.

MEA and fuel cell performance

A commercial PEMFC single cell test fixture (WonATech) with active area of 25 cm^2 was used to test the performance of CCP and TGP-120. The same commercially available catalyst coated membrane (CCM) was used for both GDLs. The GDLs were assembled at both sides of the membrane. The cell was tested using SMART2 test station (WonATech) with reactants of pure hydrogen and oxygen. Flow of the reactants was set at 300 ml/min and operated at open – end condition. The cell and reactant humidification temperature were set at 55°C and 50°C, respectively. Before measuring the cell performance, both GDL samples were activated by operating the cell at 0.6 volt for an hour.

RESULTS AND DISCUSSION

Physical properties analysis

Tabel-1 lists the physical characteristics of gas diffusion layer based on CCP and commercial TGP-120. As summarized in Tabel-1, porosity of TGP-120, which represents the ability of the porous media to diffuse and transport reactants/products in the electrode, is slightly larger than porosity of CCP. Different porosity is typically attributed to different porous structures [5, 8]. In this research, it came from different morphology of non-woven carbon paper TGP-120 that consisted of carbon fibres and CCP that was made from carbon powder.

To study further about the porous structures, surface morphology and cross-section images of both GDLs can be seen in Figure-1 and Figure-2. The surface morphology of CCP seems very smooth and the cross-sectional images reveal that CCP is denser compared to TGP-120. It is also clearly seen that the TGP-120 is characterized by a nonwoven structure with many open – pores. From the SEM images, pores of TGP-120 formed by internal space between carbon fibres are in the range of 10 – 100 μm, while those for CCP are smaller, about 5 – 20 μm.

Table-1. Physical characteristics for CCP and TGP-120.

Property	CCP	TGP-120
Thickness (μm)	380	370
Porosity (%)	69	74
Average contact angle θ_c (°)	139	131
Conductivity σ (S/cm)	0.5	4.5
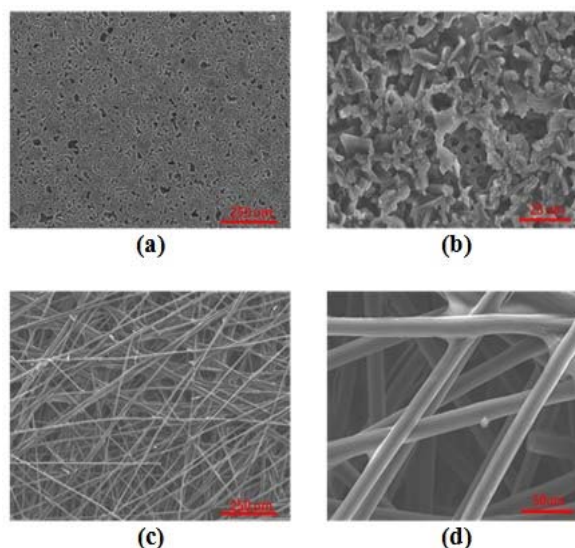
Figure-1. SEM micrographs of surface of CCP (a and b) and TGP-120 (c and d).

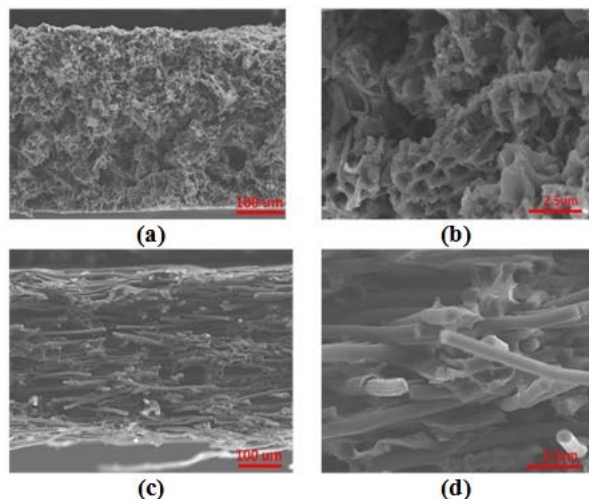ARPN Journal of Engineering and Applied Sciences

Figure-2. SEM micrographs of cross-sectional of CCP (a and b) and TGP-120 (c and d).

If only comparing the SEM images (surface and cross – section), it seems that porosity of TGP-120 is much higher than CCP. However, from calculation (Table-1), their porosity is almost similar, 69% and 74% for CCP and TGP-120, respectively. This could be the result of existence of internal porous (in the range of $1-5$ μm) on the surface of coconut fibres that clearly seen in Figure-3 [10]. Those complex porous structures of GDL CCP might produce additional transport channels that could enhance reactants to reach active site in the catalyst layer, thus improve the cell performance.
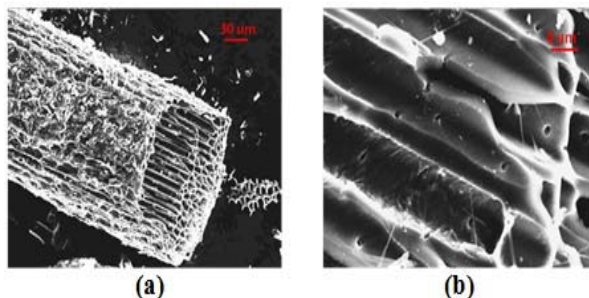

Figure-3. SEM micrographs of coconut carbon fibre [10]

In order to study surface wettability of both GDLs, contact angel θ_c on the GDL surface was measured. Static contact angle formed between water droplet and GDL surface was measured every 10 min for 50 min. When liquid water was used as a wetting agent, θ_c was smaller than 90° on hydrophilic GDLs, while for hydrophobic GDLs, θ_c was between 90 and 180° [12]. From Figure-4, it describes that during the measuring time of 50 min, θ_c for both GDLs decreases in which the surface layer of TGP-120, with average θ_c of 131° (Tabel-1), is less hydrophobic than CCP (θ_c is 139°). Different

hydrophobicity of those GDLs could come from their surface roughness (Figure-1). Higher the fineness of the surface is greater in its contact angle, since higher surface roughness results in higher wet ability [13, 14]. Less hydrophobic GDL has advantage when PEMFC cell is operated at dry condition, as it could retain more water vapour to hydrate the membrane. In other hand, higher hydrophobicity would benefit when the cell is operated at high humidity to prevent the over swelling of electrolyte membrane and flooding inside the cell.
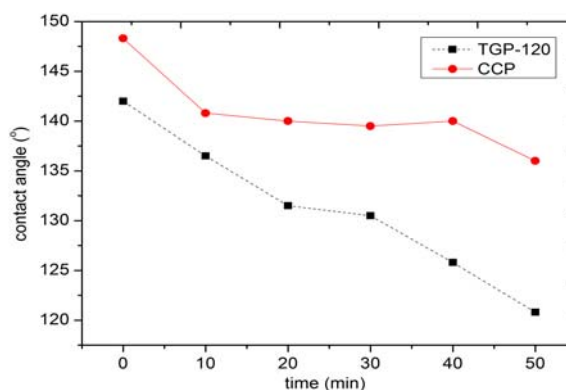

Figure-4. Contact angles of MEA based on CCP and TGP-120 at different time holding.

Based on results in Table-1, conductivity of TGP-120 is almost nine times higher than CCP, which is 4.5 S/cm and 0.5 S/cm, respectively. Significant different in the electrical conductivity highly affected the electron transfer, and it was observed during cell performance test using the polarization curve.

The performance of single cell

Figure-5 shows the change of current density at cell voltage of 0.6 V during activation process for MEA based on CCP and TGP-120. This process, which also called pre-conditioning or break-in period, is required since PEMFC cell does not reach the best and stable performance immediately after first starting-up. Gradual change of the cell performance during this process was related to hydration of the electrolyte membrane and availability of active site in catalyst layer [15, 16].

In Figure-5, the curve of MEA using TGP-120 stabilizes just after being operated for 10 min, while CCP needs more than 30 min to reach its stability. As both GDLs used the same CCM, this indicated that TGP-120, which was less hydrophobic, could retain more water vapour to hydrate the membrane, thus it was shorten the activation time. This trend was similar to the result obtained by Wang et al [5] in which GDL carbon paper could prevent the loss of water product to dry gas streams thus increasing the membrane hydration level due to its high tortuous structure. From Figure-5, it also presents that during activation process, current density for CCP is only 25%, which is lower than those for TGP-120.
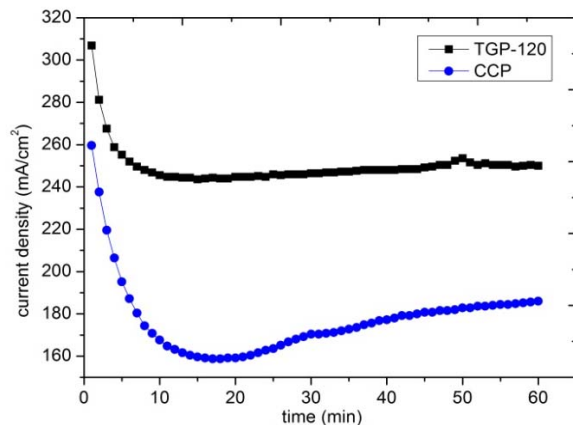
Figure-5. Activation process form MEA based on GDL
CCP and TGP 120.

Figure-6 shows the polarisation (J-V) curve for
the two GDLs. In general, cell performance of CCP is
lower than those for TGP-120. From Tabel-2, maximum
power density of TGP-120 is 208 mW/cm2 (0.45 V; 462
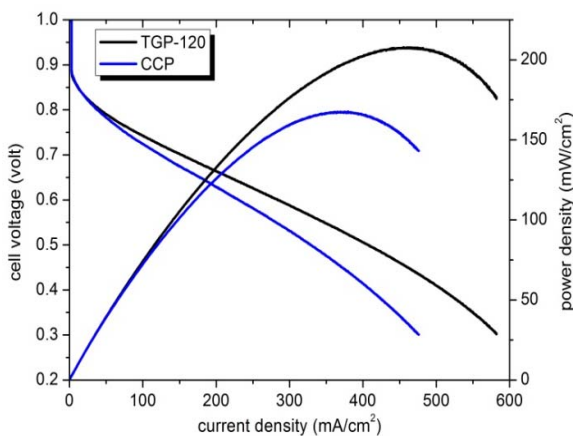mA/cm2), while those for CCP is 168 mW/cm2 (0,44 V;
381 mA/cm2).

Figure-6. Cell performance of MEA based on GDL TGP-
120 and CCP.

During operational, the cell potential was
decreased from its ideal performance due to three main
over-potentials, namely activation, ohmic, and diffusion
over-potential [17]. Activation over-potential that was
dominated at low current density was related to the
slowness of reaction taking place on the surface of the
electrode. This over-potential highly depended on the
activity of the catalyst sites. From Figure-6, the J-V curves
for both GDLs at low current density (< 100 mA/cm2) are
almost identical. This revealed the catalytic activity of
catalyst layer for both GDLs were almost similar. The
difference was mainly showed at middle current density.
This linear region as called ohmic over-potential was
dominated by electrical resistance of the electrode and the

resistance to the flow of ion in the electrolyte. Since there
was used the same commercial CCM, MEA preparation
procedure, and test apparatus, ohmic over-potential is
mainly related to the conductivity of the GDLs. Although,
the conductivity of CCP was much lower than that for
TGP-120, area specific resistance (Ra) of CCP, obtained
from the slope of linear portion of the polarisation curves,
was only 20% higher than that for TGP-120 (Table-2). In
addition, current density at operating condition, 0.6 volt
(J0.6) for that TGP-120 and CCP were 284 and 232
mA/cm2 respectively. This trend needed further
investigation.

Tabel-2. Parameters of the cell performance for MEA
based on GDL TGP-120 and CCP.

Parameter	CCP	TGP-120
OCV (V)	0.997	1.00
$J_{0.6}$ (mA/cm^2)	232	284
P_{max} (mW/cm^2)	168	208
R_a (Ωcm^2)	0.95	0.76

The third over-potential was diffusion over-
potential or mass transport over-potential. This over-
potential was clearly showed in high current density, and it
appeared since there was a concentration gradient of
reactant due to limited supply of reactants. At cell voltage
of 0.3 V, MEA based on TGP-120 had current density of
580 mA/cm2, whereas it was 476 mA/cm2 for CCP. Han
et al. [18] found that diffusion over-potential had closely
related to porous electrode, since pore size distribution and
hydrophobicity of GDL had influenced the possibility
water flooding inside the cell.

By comparing the three regions of over-potential,
it was known that in spite of significantly lower of CCP
conductivity compared to TGP-120, there was only
slightly difference in the cell performances. This might
indicate that the unique structure of CCP had advantage to
enhance cell performance, and it still required further
investigation.

CONCLUSIONS

The main physical properties (hydrophobicity,
electrical conductivity, and porosity) and the performance
of GDL made from activated carbon powder from coconut
coir were investigated. Comparing to the commercial one,
Toray Paper TGP-120, CCP morphology and electrical
conductivity were significantly different. However, from
the J-V curves, by comparing three over-potential regions,
there was insignificantly difference in the cell
performances. This result might be confirmed that
existence of microstructures on the surface of coconut
fibres and relatively high hydrophobicity of CCP had
advantage to enhance cell performance. Nevertheless,
further studies are required to evaluate the CCP durability
and improve its electrical conductivity.

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