

Properties of carbon composite paper derived from coconut coir as a function of polytetrafluoroethylene content

Fredina Destyorini¹, Indriyati², Nanik Indayaningsih¹, Bambang Prihandoko¹ and Anne Zulfia Syahrial³

¹Research Center for Physics, Indonesian Institute of Sciences (LIPI), South Tangerang, Indonesia

²Research Unit for Clean Technology, Indonesian Institute of Sciences (LIPI), Bandung, Indonesia

³Department of Metallurgical and Materials Engineering, Faculty of Engineering, Universitas Indonesia, Depok, Indonesia

E-mail: fred007@lipi.go.id; fredinadestyorini@gmail.com

Abstract. The carbon composite papers were produced by utilizing carbon materials from coconut coir. In the present work, carbon composite papers (CCP) were prepared by mixing carbon materials in the form of powder and fibre with polymer (ethylene vinyl acetate and polyethylene glycol) in xylene at 100°C. Then, polytetrafluoroethylene (PTFE) with different content was used to treat the surface of CCP. The properties of PTFE-coated CCP were analysed by means of contact angle measurement, tensile testing, porosity, density, and electrical conductivity measurements. As expected, all CCP's surfaces treated with PTFE were found to be hydrophobic with contact angle $>120^\circ$ and relatively constant during 60 minutes measurement. Furthermore, water contact angle, density, and mechanical properties of CCP generally increase with increasing PTFE content. However, the porosity and electrical conductivity of CCP decrease slightly as the PTFE content increased from 0 wt% to 30 wt%. Based on the observation and analysis, the optimum PTFE content on CCP was 20 %, in which the mechanical properties and hydrophobicity behaviour were improved significantly, but it was only caused a very small drop in porosity and electrical conductivity

1. Introduction

Coconut coir is an abundant agricultural waste in Indonesia but not yet optimal utilizations. It contains hydrocarbon such as cellulose, hemi-cellulose and lignin as major composition [1]. The carbonization processes of hydrocarbon in inert gas to temperature of 1300°C produces charcoal with high carbon content and conductive [2]. Due to this abundance and advantages, several researchers have initiated to use the potential of coconut coir for raw material of carbon paper for gas diffusion layer (GDL) in proton exchange membrane fuel cells (PEMFC) [3,4]. Previous study in our lab revealed that coconut carbon fibre has internal porous (in the range of 1-5 μm) to enhance reactant gases reach active sites in the catalyst layer [2].

GDLs are generally available in the form of carbon paper and carbon cloth. At present, they are composed of carbon materials, which are produced from fossil fuels such as coal and petroleum,



raising concerns about sustainability. The utilization of coconut coir as carbon source for the manufacture of carbon paper for GDLs could resolve these issues.

As one of the critical components in PEMFC, GDL main functions are to supply reactant gases, remove product water, and conduct electron between adjacent components, and provide mechanical support for membrane electrode assembly (MEA) [5]. In order to perform these functions, carbon paper as GDL should be porous (50%-90% of porosity), hydrophobic (contact angle $>90^\circ$), conductive, and strong enough [6]. Hydrophobic property controlled by hydrophobic treatment is needed to remove liquid water saturation in the cathode effectively. Although carbon substrates used as GDL are usually hydrophobic, they are typically treated with a hydrophobic agent such as polytetrafluoroethylene (PTFE), which is used for the commercial carbon paper [7,8]. PTFE treatment applies to both, anode and cathode sides. This feature reduces the flooding of water in the GDL and provides the reactant access to the catalyst layer [9]. However, its impact on other parameters such as electrical conductivity and porosity should be considered for an appropriate cell design. It has been reported that usage PTFE with too high content could decrease porosity and conductivity of carbon paper [8,10]. On the other hand, too low PTFE content would make water generated in the cell cannot be removed effectively. Therefore, PTFE content in the GDL should be balanced and optimized to achieve the best performance of fuel cell.

In this study, carbon paper or GDL was prepared from coconut coir. The coconut coir carbon paper (CCP) was then treated with various concentration of PTFE to improve its hydrophobic property. The optimum PTFE content was determined based on investigation of their physical and mechanical properties. Relationship among those parameters will be revealed as an effect of PTFE content.

2. Experiment

2.1. Materials

Coconut coir obtained from local market, was cut into a length of ± 2 mm. Ethylene vinyl acetate (EVA) and poly ethylene glycol (PEG) purchased from Aldrich Chemical Co., Inc. (St. Lois, MO, USA) were used as binder and dispersant, respectively. Xylene (Brataco Chemica) was used as solvent for making the CCP. Polytetrafluoroethylene (PTFE) emulsion obtained from MTI Corporation was used as hydrophobic agent of CCP.

2.2. Production of carbon materials

Coconut coir was processed by two stages, namely: carbonization and pyrolysis. In the carbonization process, coconut coir was heated at 500°C for an hour under N_2 atmosphere and cooled down to room temperature to produce charcoal which had high carbon content. Subsequently, the charcoal was processed through pyrolysis at 1300°C for an hour under N_2 atmosphere to improve the electrical conductivity, eliminate impurities, and improve other properties of carbon. The resulting carbon material was named as carbon fibre. Some of carbon fibre was grinded into a powder of ± 74 μm sizes (200 mesh).

2.3. Preparation of CCP

Carbon fibre (70 wt%) and carbon powder (10 wt%) were mixed with EVA and PEG in xylene at 100°C for 2 hours to make slurry. To form a paper, the slurry was cast on molded glass, rolled, and then dried at room temperature for 24 hours to evaporate the solvent. To provide the hydrophobic property, the CCPs were treated with different quantities of PTFE for 30 minutes, varied from 10 to 30 wt%, and dried at room temperature for 24 hours, and finally sintered at 350°C .

2.4. Characterization of CCP

Characterization and analysis were carried out on untreated and PTFE-treated CCP samples. For evaluating the hydrophobic properties of a CCP, surface contact angles between water and the carbon

paper were measured using sessile drop method. For each measurement, a 50 μL water droplet was placed onto the surface of CCP and images were captured every 20 minutes for 1 hour after the droplet attached to the sample surface. Furthermore, the droplet shape was analyzed using Bashforth and Adams tables [11] to determine the contact angle of the samples. Large contact angles imply high hydrophobia. Porosity and density were examined by the kerosene density method using Archimedes principle in accordance with BS 1902: Part 1A standard. Through-plane electrical conductivity was measured using HIOKI 3522-50 HITESTER LCR-meter. The mechanical properties of the carbon paper samples were measured by using a universal testing machine (Orientec Co. Ltd, Model UCT-5T) according to ASTM D828-97. For each sample, 7 rectangular specimens (70 $\times$ 10 mm) were tested and an average tensile strength and Modulus Young were determined.

3. Results and discussion

3.1. Hydrophobicity (water contact angle) of CCP

The hydrophobic properties of a solid surface could be determined by the surface contact angle. When liquid water was used as a wetting agent, hydrophilic carbon papers will have contact angle smaller than 90° , while hydrophobic carbon papers have contact angle between 90 and 180° [12]. Fig. 1 shows the contact angle of CCP with different content of PTFE. The contact angle of untreated CCP (0 wt% PTFE) changes during 60 minutes of measurement process. At the first minute when the water droplet attached to the surface of untreated CCP, it showed the hydrophobicity with contact angle greater than 90° , indicating that the substrate is resistant to wetting. However, water droplet started to penetrate into the porosity of CCP, then untreated CCP became wet and the contact angle dropped to 0° (40 to 60 min). This indicates that the untreated CCP is not able to resist the water penetration for a long period. When the paper is impregnated with PTFE, contact angles increase from approximately 115° up to 135 – 145° . All the PTFE-treated CCPs exhibit high contact angles and are relatively constant during the measurement process as can be seen in Fig. 1. It is noted that the droplets show relatively similar contact angle on CCPs with different PTFE content (10 to 30 wt%). This elucidates that PTFE treatment significantly affects the hydrophobicity of carbon paper, but the contact angle does not change too much for different PTFE content.

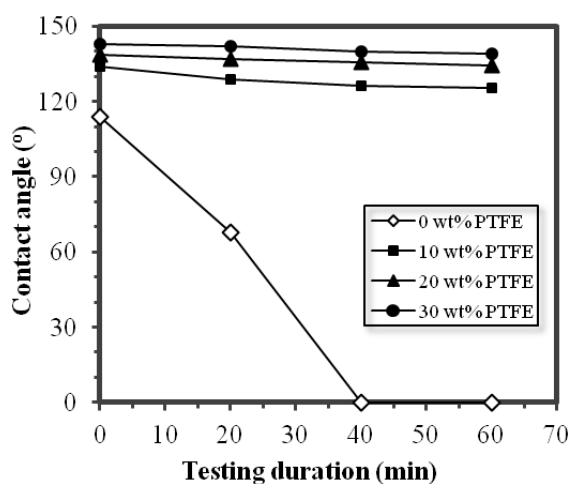


Figure 1. Contact angle of CCP with different PTFE content

3.2. Porosity and density of CCP

Porosity is needed to diffuse and transport reactant/product in the electrode. The porosity and density measurements using kerosene density method were only performed on PTFE-treated CCPs. These

parameters cannot be measured by this method for the untreated CCP because EVA as a binder will dissolve in kerosene [13]. The porosity and density value of PTFE-treated CCPs are shown in Fig. 2 and Fig. 3, respectively. It is proved that increasing PTFE content from 10 to 30% decrease the porosity of CCPs for about 8 %.

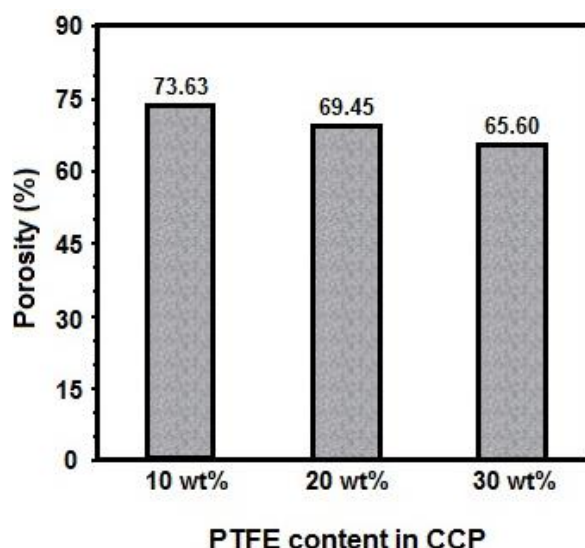


Figure 2. Porosity of CCP with different content of PTFE

This could be explained that PTFE particles penetrate through the pores of CCP and deposit onto the carbon fibres or accumulate at the fibre crossing. As a consequence, it reduces the overall porosity of CCPs. Yoon and Park [6] reported that carbon paper should have high porosity of 50-90% to effectively function as GDL, and the porosity of all treated CCPs is in this range. It should be noted that higher GDL porosity improves mass transport, leading to higher current density produced by PEMFC, but it will decrease the electrical conductivity as well as the mechanical properties [5] that will be shown in the next section. Fig. 3 shows the density of CCP as a function of PTFE content. The density of CCP gradually increased as PTFE content increased. This is a reasonable result since PTFE penetrate into the CCP's pores and accumulate at the fibre crossing as mentioned above. The densities increase from 0.42 to 0.55 gram/cm³ for PTFE 10 to 30%, respectively.

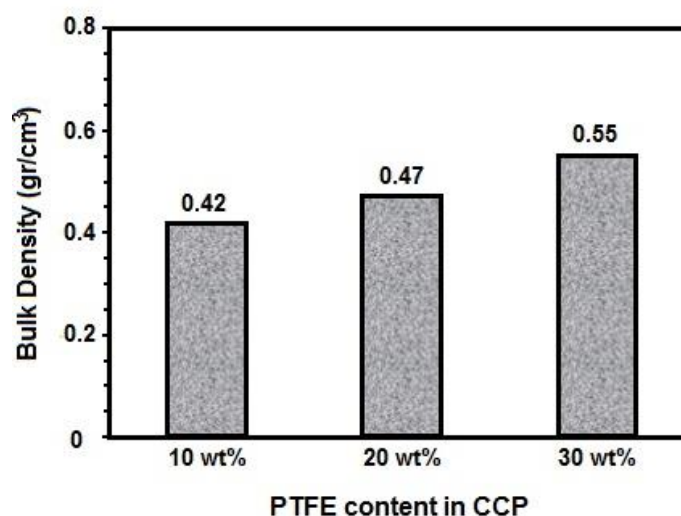


Figure 3. Density of CCP with different content of PTFE

3.3. Electrical conductivity of CCP

The values of through-plane electrical conductivity of all samples with different PTFE content are represented in Figure 4. As predicted that an increase of PTFE content up to 30% leads to decrease in the electrical conductivity for almost 50% compared to untreated CCP. It occurs because of the increasing numbers of non-conductive PTFE particles that isolated the conductive carbon materials and disconnected the conduction path in the CCP [14].

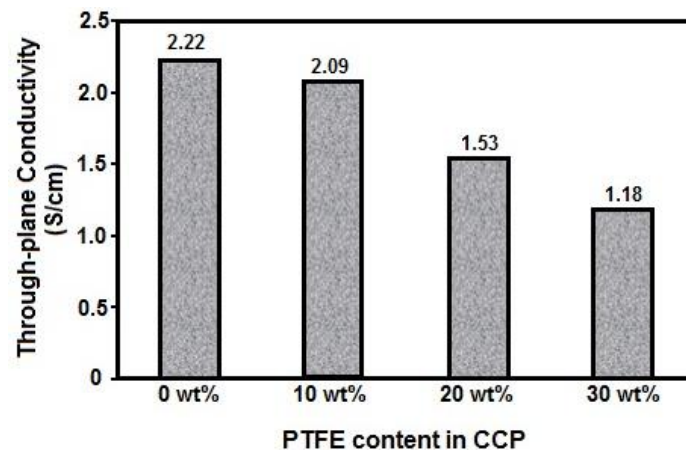


Figure 4. Through-plane electrical conductivity of CCP with different content of PTFE

3.4. Mechanical properties of CCP

Fig. 5 and Fig. 6 show the mechanical properties of untreated CCP and PTFE-treated CCPs with varying PTFE content. It is observed that PTFE treatment could improve the tensile strength and Modulus Young of CCP. In this case, PTFE also acts as the matrix in the composites that bonds the carbon fibres together and transfers loads between them. A decrease in porosity of CCP because of the addition of PTFE content may also cause an improvement of tensile properties [15] of CCP. It is noticed that increasing PTFE content from 10 to 20% gives significant improvement to the mechanical properties of CCP compared to increasing PTFE content from 20 to 30%.

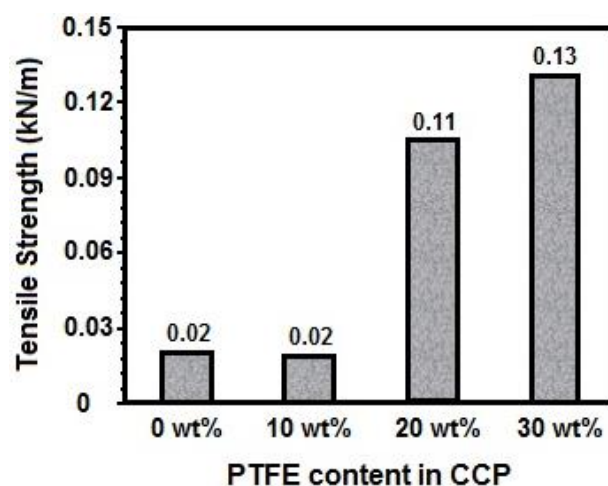


Figure 5. Tensile strength of CCP with different content of PTFE

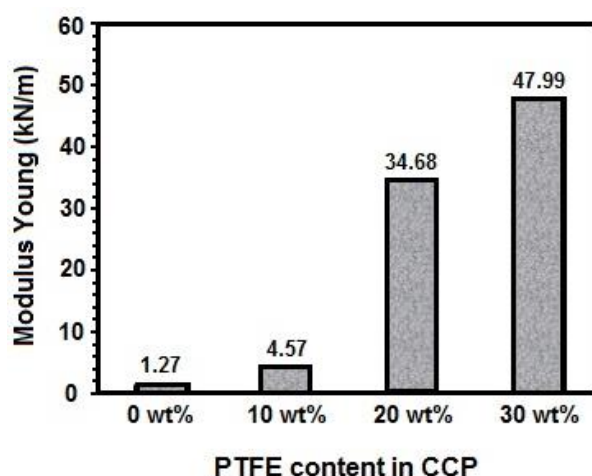


Figure 6. Modulus Young of CCP with different content of PTFE

4. Conclusion

The effects of PTFE hydrophobic polymer content on the physical and mechanical properties of carbon paper from coconut coir have been investigated. The porosity and electrical conductivity are found decreasing slightly, while the hydrophobicity and mechanical properties of carbon paper increase with the increasing of PTFE content in carbon paper. The use of 20 wt% PTFE in carbon paper from coconut coir is the optimum content because this content could improve CCP's tensile properties significantly and maintain the hydrophobicity of CCP, but it is only caused a small drop in porosity and electrical conductivity.

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