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Manufacture of a polymer-based carbon nanocomposite as bipolar plate of proton exchange membrane fuel cells

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ABSTRACT

The aim of this paper is to prepare a polymer-based carbon nanocomposite reinforced by carbon fiber cloth (CF) to be utilized as bipolar plate of proton exchange membrane (PEM) fuel cell. For this purpose, some single, double, and triple-filler composites were manufactured by using phenolic resin as polymer (P) and graphite (G), carbon fiber (CF) and expanded graphite (EG) as fillers. The production method was comp ression-molding technique. The electrical conductivity, flexural strength, toughness, hardness, porosity, and hydrogen permeability tests were then measured to determine the mechanical and physical properties. A triple-filler composite containing 45 wt.% G, 10 wt.% CF, 5 wt.% EG, reinforced by a layer of CF cloth, was selected as composite bipolar plate. The electrical conductivity, thermal conductivity, and flexural strength of this composite were 74 S/cm, 9.6 W/m K, and 74 MPa, respectively, which are higher than the specified value by department of energy in USA (DOE). The composite bipolar plate used in the single fuel cell assembly showed a maximum power density 810 mW/cm². In this paper, a material selection was performed on the different materials of bipolar plates. It can be concluded that the composite bipolar plates are more suitable for high life time stationary applications.

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1. Introduction

Bipolar plate is a multi-functional component in proton exchange membrane (PEM) fuel cells. It provides the electrical connection from cell to cell and separates the reactive gases [\[1\]](#page--1-0). 80% of the PEM fuel cell stack weight and 38% of the cost are ascribed to the bipolar plate [\[2–4\]](#page--1-0). Based on department of energy (DOE) criteria [\[2,5\],](#page--1-0) composite bipolar plates should achieve some properties such as high through-plane electrical conductivity (>100 S/ cm), low area specific resistance (<30 m Ω cm²) [\[6\],](#page--1-0) chemical stability in the slightly acidic water $(pH < 4)$, corrosion resistance (\leq 16 μ A/cm) [\[6\],](#page--1-0) high thermal conductivity ($>$ 10 W/m K) [\[7\],](#page--1-0) low permeability to hydrogen and oxygen (<2 \times 10^{–6} cm³/(cm² s) [\[6\],](#page--1-0) flexural strength > 59 MPa, and impact strength >40.5 J/m [\[6,7\].](#page--1-0) There are many studies concerning the composite bipolar plates, however in most of them the properties of the composite have not attained DOE criteria. Carter [\[8\]](#page--1-0) attempted to manufacture single and double fillers with graphite (G) and carbon fiber (CF). However, the electrical conductivity of these composites is lower than the DOE criteria. Phenolic resin/CF (composite of phenolic resin and CF) possesses higher mechanical strength and toughness than the DOE criteria, but the electrical conductivity does not attain DOE criteria, even with 60 wt.% CF. Moreover, increasing the CF loading in the composite substantially increases hydrogen permeability of bipolar plates, that is resulted from the tendency to agglomeration $[1]$. Dhakate et al. $[9]$ manufactured a composite by using polymer and expanded graphite, but the flexural strength value was lower than that of DOE criteria. Kang et al. [\[10\]](#page--1-0) manufactured a P/G composite by hot compression-molding technique. Through-plane electrical conductivity and flexural strength reached 80 S/cm and 55 MPa, respectively, which are lower than DOE criteria.

It has been attempted in the current work to increase the composite electrical and mechanical properties, simultaneously. Increasing the strength of composite bipolar plate leads to decreasing composite thickness, thereby decreasing fuel cell stack size. Therefore, the present research is an attempt to obtain a high-quality composite bipolar plate with emphasizing on the mechanical properties. The CF and carbon cloth are two effective fillers applied for increasing the mechanical properties. Other carbon-based fillers such as G and EG (expanded graphite) are mainly utilized to increase composite electrical conductivity. The type and value of fillers should be optimized in the composite. Therefore, some single and double-filler composites were manufactured from G, EG and CF to clarify the fillers properties and the synergetic effect

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among fillers. To the best of our knowledge, there are few reports on the synergetic effect between fillers [\[4\]](#page--1-0). The single-cell assembly of the composite bipolar plate was also prepared to specify the power and performance curves of the cell. In this research, a material selection was also performed among common graphitic, metallic, and composite bipolar plates.

2. Experimental details

2.1. Materials

The polymer resin used in this research was the novolac phenolic resin in powder form with 60 μ m size and was purchased from Resitan Co. Ltd. The G powder was purchased from Merck Co. Ltd., containing size <50 μ m and the bulk density of 20–30 g/(100 ml). The expandable graphite and CF were purchased from Qingdao Yanxin Graphite Products Co., Ltd. and Highborn International Co., Ltd., respectively. Expandable graphite was inserted into the furnace with 1000 \degree C to be converted to EG containing nanosheets. CF cloth that is the textile of CF was purchased from Toray Co.

2.2. Preparation of composites

All composites were produced by the hot compression-molding technique. The die temperature, pressing time and pressing pressure were 200 \degree C, 50 min and 230 bar, respectively. The composites contain phenolic resin as binder and G, CF, and EG as fillers. Three categories of composites were manufactured: single-filler, doublefiller and triple-filler composites . The codes of all categories have been shown in Table 1. The double-filler composites have been designed so that the synergetic effect can be derived by comparing single and double-filler composites . In double-filler composites by codes P/G/EG (the composite of polymer with G and EG), P/G/ CF, and P/EG/CF, the total filler loadings were considered 20, 40, and 60 wt.%, so that the values of two fillers were equal. The composite bipolar plate which is a triple-filler nanocomposite containing 40 wt.% polymer, 45 wt.% G, 10 wt.% CF, and 5 wt.% EG sandwiched by CF cloth was labeled P/45G/10CF/5EG/CC (CC is abbreviation of carbon fiber cloth). The CF cloth was embedded within the middle of the composite during the molding (Fig. 1). The composition of composite bipolar plate was selected based on the attained properties of single and double-filler composites.

2.3. Characterization

The bulk densities of the samples were determined by using highly precise digital balance (model ME 40290) based on the Archimedes principle. The determined porosity (closed porosity) values were calculated by using the density values. The throughplane electrical conductivity was measured by a laboratory-made setup [\[11\]](#page--1-0). The sample dimension was 10 \times 10 \times 2.5 mm³. Firstly, the conductive silver adhesive paste covered the two parallel surfaces of the sample. The ampere and voltage were determined by

Table 1

The codes of single, double and triple filler composites.

Fig. 1. The schematic of components in sandwiched composite (P/45G/10CF/5EG/ C_C).

digital mili-voltmeter and microampermeter placed in circuit. The resistance of sample, thereby electrical conductivity can be derived from the $I-V$ curve slope. Thermal property measurement was performed using a thermal conductivity test apparatus, with its design based on the guarded heat flexmeter device recommended by ASTM: D5470-12 with a number of modifications [\[12\]](#page--1-0). The mechanical properties of the composites were investigated by flexural, impact and hardness tests. The three-point flexural strength was measured according to ASTM: D790-10 using the Instron Universal testing machine (model 4411). The unnotched impact tests were conducted according to ASTM: 4812-11 by the Zwick model apparatus. The universal digital hardness tester was KOOPA (Model UV1). The method was Vickers and the set was adjusted in the simulation mode. In order to evaluate the gas permeability of composites containing different fillers, N_2 gas permeability measurements were made based on ASTM: F316-03 using a laboratory-made apparatus [\[13,14\].](#page--1-0) This test was conducted on P/50G, P/50EG, P/50CF composites. The device precision was up to 2 \times 10⁻⁷ cm³/(cm² s). In order to evaluate the composite bipolar plate in the cell, this bipolar plate was inserted into a PEM fuel cell. The membrane was selected nafion 112 and Pt loading was 0.4 $\rm g/cm^2$ and surface area was 25 cm². Channel type was serpentine with 1 mm width and 1 mm height. The stoichiometry of $H₂$ (purity 99.999%) and O₂ (purity 99.9%) was kept 1, while the back pressure was preserved 3 bar.

3. Results and discussion

3.1. Fracture surface and microstructures

[Fig. 2](#page--1-0)a shows the composite contains 10 wt.% CF. It can be seen that some fibers have been pulled out from their sites but some polymer has adhered to the fibers. This shows that the bridging

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