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Effects of type of graphite conductive filler on the performance of a composite bipolar plate for fuel cells

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ABSTRACT

The selection of appropriate conducting fillers and resins and the optimization of their compositions are crucial for the successful design of composite bipolar plates for fuel cells. In this paper, several graphite-based conductive fillers, i.e., natural and synthetic graphites with different sizes (10, 20, and 40 μm) and shapes (flakes and lumps), are combined with phenolic resin and tested in order to investigate the effects of the filler size and shape on the conductive network and mechanical strength of the resulting composite bipolar plates. The test results show that for the same filler size/loading and resin content, the composite plates made of natural graphite (NG) exhibit higher electrical conductivities and lower flexural strengths than those made of synthetic graphite (SG). In addition, flake-like graphite powders are superior to lump-like ones for the effective formation of conductive networks. As the graphite particle size is reduced, the flexural strength of the composite bipolar plate is improved, whereas the electrical conductivity is decreased. This study clearly elucidates the electrical and mechanical behaviors of graphite/phenolic-resin-based composite bipolar plates fabricated with different graphite filler types, sizes, and shapes.

1. Introduction

Fuel cells have been recognized as alternative power devices because of their high efficiency and low emission characteristics as compared with conventional power sources such as internal combustion engines and batteries. Thus, fuel cells are a very versatile technology for portable power, stationary power generation, and transportation. As shown schematically in Fig. 1, a typical fuel-cell stack consists of repeating fuel-cell components, a membrane electrode assembly (MEA), and a bipolar plate. The bipolar plate serves many functions in a fuel-cell stack: (i) distribution of fuel and oxidant within the cell; (ii) facilitation of water management within the cell; (iii) separation of the individual cells in the stack; (iv) carrying of current away from the cell; and (v) support of the MEAs. In order to fulfill the aforementioned functions, bipolar plate materials must possess high electrical conductivity and good mechanical strength. Pure graphite is a basic bipolar plate material owing to its high electrical conductivity and chemical stability [1–3]. However, pure graphite-based bipolar plates exhibit low mechanical strength (brittleness) and incur high manufacturing costs of machining operations, rendering the fuel-cell stacks massive, voluminous, and expensive [3–5]. It has been reported that conventional bipolar plates account for approximately 80% of the total stack weight and over 40% of the total stack cost [6,7]. Alternatively, several metallic alloys have been recognized as suitable bipolar plate materials thanks to their excellent electrical conductivities, low manufacturing costs, and high mechanical strengths. However, these metallic bipolar plates suffer from corrosion problems, and thus, require surface modification and protective coating procedures [8–13].

Recently, compound materials made of graphite-based conductive fillers, carbon-based reinforcements, and polymer resins as binders have been proposed as new candidates to replace the pure graphite and metal bipolar plates, owing to the advantages of their low cost, ease of machining, low weight, and good corrosion resistance [14–20]. The electrical conductivity and mechanical strength of these composite bipolar plates depend on the graphite content [21,22], the morphology and size of the filler [23–28], and the type of resin [29–32]. Therefore, most studies regarding composite bipolar plates have focused on the improvement of the electrical conductivity and mechanical strength. Dhakate et al. [22] investigated the effects of the contents of synthetic and natural graphites on the electrical conductivity of composite bipolar plates. They reported that an electrical conductivity of 120 S/cm was achieved with a graphite content of 80 vol.%. Heo et al. [25] tested two different types of graphite fillers, i.e., spherical (25 μm) and flake-type (25 μm) fillers, for filling contents of 85 wt.%. The flake-like graphite showed superior electrical conductivity to the spherical type, owing to the high aspect ratio of the elongated flakes, which shortens the electrical path between neighboring graphite particles and consequently reduces the electrical contact resistance. Chen et al. [27] explored the influence of the particle size of flake-like graphite on the electrical conductivity and flexural strength of phenolic-resin-based composite bipolar plates. As the graphite particle size was increased from 78 to 250 μm, the electrical conductivity of the composite bipolar plates was

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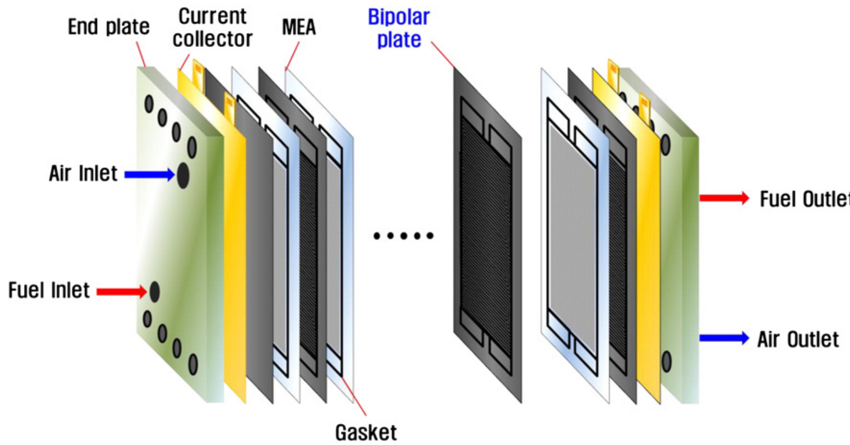

Fig. 1. Schematic diagram of a typical fuel cell stack.

enhanced from 79 to 167 S/cm, but the flexural strength was reduced from 43.6 to 35.2 MPa.

Although considerable efforts have been made to design and optimize composite bipolar plates, a more detailed study is required to clarify the complex behavior of the graphite fillers and polymer resin. The purpose of this study is to investigate the effects of the graphite particle nature, size, morphology, and interaction with the polymer resin on the electrical and mechanical behaviors of composite bipolar plates. This work can provide further insights for the design and optimization of composite bipolar plates.

2. Experimental details

2.1. Preparation of composite materials

In this study, we prepared the two different grades of graphite fillers, i.e., natural graphite (NG) and synthetic graphite (SG) powders, with different filler sizes and shapes. The samples used were flake-like NG (Graftech Co., Ltd), lump-like NG (BTR Co. Ltd), flake-like SG (Timcal Co., Ltd), and lump-like SG (Timcal Co., Ltd) with three different sizes (10, 20, and 40 μm). Detailed information on the graphite powders used in this work is given in Table 1 [33]. The morphologies of the graphite samples were observed by scanning electron microscopy (SEM), as shown in Fig. 2. The graphite particles and phenolic resin were mixed in a kneader mixer to obtain uniform mixtures of graphite–phenol compounds. A range of phenolic resin content employed during the experiment was from 10 to 25 wt.%, i.e. corresponding to the contents of graphite fillers from 90 to 75 wt.%, respectively. Then, the compounds were dried and shattered before the molding process. Finally, the mold was placed into a hot press (Carver Inc. 30 tons) and heated to 150 °C for 5 min under a pressure of 25 MPa. The hot-press temperature of 150 °C was set slightly higher than the glass transition temperature of phenolic resin, i.e. 135 °C. On the other hand, the curing time of 5 min was chosen based on the previous work of Heo et al. [34]. The pressure condition was optimized from carbon composite sample test results in Fig. 3 wherein 25 MPa leads to superior electronic conductivity and flexural strength. The dimensions of the specimens were 6 mm (width) × 6 mm (height) × 3.5 mm (thickness). A schematic diagram of the detailed procedure for specimen fabrication is shown in Fig. 4.

Table 1
Properties of the graphite materials [33].

Properties	Natural graphite (NG)		Synthetic graphite (SG)	
Shapes	Lump	Flake	Lump	Flake
Surface area, [m^2/g]	4.3	7.56	10.25	13.08
Density, [g/cm^3]	1.82–1.89		2.25–2.28	
Electrical conductivity, [S/cm]	648		523	

2.2. Characterization of composite bipolar plates

Several sample tests were conducted to understand the electrical and mechanical behaviors of the composite bipolar plates. The electrical conductivities of the bipolar plates were measured with an FPP-5000 four-point-probe instrument. The three-point bending test was performed to measure the flexural strengths with a universal testing machine (Instron, 5569), using a procedure based on ASTM D790 [35–37]. All the specimens were 35 mm in length, 4 mm in width, and 3 mm in thickness.

3. Results and discussion

Fig. 5 shows the electrical and mechanical behaviors of the composite plates made of lump-like SG as a function of the phenolic resin content (10 to 25 wt.%). These bipolar plates, based on SG particle sizes of 10, 20, and 40 μm, were fabricated under a constant molding pressure of 25 MPa. It is observed clearly in Fig. 5 that the electrical conductivity of the composite bipolar plates decreases with increasing resin content, whereas the flexural strength exhibits the opposite trend, decreasing with increasing resin content. The trend observed experimentally can be explained as follows: as the resin content is increased in the composite compound, the resin fills more gaps between the graphite particles, reducing the number of conductive networks. This hinders the movement of electrons and lowers the overall electrical conductivity of the composite plate. On the other hand, the flexural strength of the composite plate must be enhanced with increasing resin content because the greater amount of resin improves the wetting of the graphite particles and reduces the number of voids in the composite plate.

It can also be observed from Fig. 5 that the electrical conductivity increases as the size of the graphite particles increases from 10 to 40 μm. A smaller particle size leads to an increase in the number of graphite particles per individual composite compound, which results in lower conductivity owing to the higher inter-particulate contact resistance between particles. On the other hand, the flexural strength of the composite bipolar plate is enhanced with decreasing particle size from 40 to 10 μm. This is because the smaller graphite particles have a larger external surface to volume (A/V) ratio, and hence, contact between neighboring graphite particles becomes stronger, enhancing the compaction of the composite plates. Regarding the target values presented by the US Department of Energy (DOE), i.e. above 100 S/cm electrical conductivity and 25 MPa flexural strength, the optimum composition of composite bipolar plates is 15 wt.% phenol resin and 85 wt.% 40-μm NG.

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