Synergy effects of hybrid carbon system on properties of composite bipolar plates for fuel cells

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ABSTRACT

A hybrid carbon system of graphite powder (GP) and continuous carbon fibre fabric (CFF) is used for an epoxy composite to improve the electrical conductivity, mechanical properties and mouldability of a composite bipolar plate. These improvements are achieved simultaneously by inserting several layers of CFF into the GP/epoxy composite to enhance the mechanical properties and in-plane conductivity. The electrical properties, flexural strength and mouldability of the composite plates are measured as a function of conducting filler content and number of CFF layers. The composites show improved electrical conductivity, flexural properties and mouldability. Composites with 70–75 vol.% carbon fillers have the highest electrical conductivity with reasonable flexural properties. These results suggest that the poor mouldability and low through-plane electrical conductivity of the continuous fibre composite bipolar plate, as well as the weak flexural properties of GP composites, can be overcome by incorporating a GP/CFF hybrid system.

1. Introduction

The polymer electrolyte membrane fuel cell (PEMFC) has attracted considerable attention as an alternative power source in a range of portable electronic devices, as well as in stationary and transportation applications. In particular, interest in vehicular applications has focused on the PEMFC due to its high efficiency and specific power, low operating temperature, and rapid startup [1–2].

The stack of a PEMFC is composed of components, such as a polymer electrolyte, a catalyst, a gas-diffusion layer and bipolar plates. The bipolar plate plays an important role in a fuel cell stack and accounts for a large part of the stack volume, weight and cost. It performs a number of functions in a fuel cell stack [3–5], namely: (1) provides a pathway to distribute fuel and air uniformly over the entire active area; (2) connects and separates the unit cells in the stack; (3) carries an electrical current from the anode of one cell to the cathode of the next cell; (4) supports the membrane–electrode assembly and clamping forces for the stack assembly; (5) promotes

water management within the cell; (6) enables heat transfer. In order to perform these functions, bipolar plate materials must have high electrical conductivity and good mechanical properties [6–7].

Bipolar plates are commonly fabricated from a graphite plank to achieve high electrical conductivity, good corrosion resistance, and a low bulk density. Graphite plank bipolar plates, however, are brittle, expensive to fabricate, heavy and voluminous. To overcome these disadvantages, many studies have focused on alternative materials for bipolar plates in fuel cells [8–10]. Graphite/polymer composites have the potential to replace graphite bipolar plates owing to their low cost, light weight, and ease of manufacture. Unfortunately, the main disadvantages of such composites are their low electrical and mechanical properties. Therefore, it is important to develop composite materials with high electrical and mechanical properties by combining graphite powder with other types of reinforced carbon fillers [11–13]. Recently, composite bipolar plates have been developed using continuous carbon fibre prepreg (CFP) or other carbon fillers, such as synthetic graphite, carbon black, short carbon fibres and carbon nanotubes. The CFP is considered a potential material on account of its good in-plane electrical and thermal conductivities, as well as good mechanical properties [14–15]. Composites of CFP, however, have low through-plane electrical conductivity, a non-uniform distribution of carbon fillers in the channel region and poor mouldability of the composite, especially for channels. On the other hand, short carbon

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fibre and carbon nanotube reinforcing composites exhibit comparatively better mouldability and surface electrical conductivity, but at the expense of the mechanical properties [16–20]. To overcome these shortcomings, a hybrid carbon system of GP and CFF has been introduced to EP composites to enhance simultaneously their mechanical properties, in-plane conductivity, and mouldability.

2. Experimental

2.1. Materials

Synthetic GP (Timrex® KS-150) supplied by Timcal Graphite & Carbon Inc., USA was selected as the main conducting filler due to its high electrical conductivity and corrosion resistance. It has a purity of >95%, a mean particle size of 180 μm, and a density of 2.2 g cm^{-1}. The plain-weave carbon fabrics (CF1114NON) were obtained from Hankuk Carbon Co., Ltd., Korea. The fabric weaved by commercial grade 1K T-300 (Toray, Japan) carbon fibres had a thread count, weight and thickness of 6.89×6.89 cm^{-1}, 95 ± 6 g m^{-2} and 0.14 mm, respectively. This CFF can be applied to bipolar plates owing to its high electrical and thermal conductivities, good mechanical properties, light weight and high chemical resistance [14]. The carbon fibre epoxies prepreg (URN 300) (CFP) provided by SK Chemicals Co., Ltd., Korea was also used to manufacture carbon fibre prepreg bipolar plates for comparison with the bipolar plates in the hybrid system.

A powder epoxy resin of the bisphenol-F type (YSLV-80XY, Tohto Kasei Co., Ltd., Japan) was chosen as the matrix binder because of its good chemical stability, heat resistance, mechanical strength, and dimensional stability [21]. It has an epoxide equivalent weight, density, molten viscosity and melting point of 190–200 g eq^{-1}, 1.23 g cm^{-3}, 0.1 Pa s at 150 °C and 78 °C, respectively. The phenol novolac type hardener (TAMANOL758, Arakawa Chemical Industries, Ltd., Japan) has a molten viscosity, melting point, density and active hydrogen equivalent weight of 0.25–0.35 Pa s at 150 °C, 80–85 °C, 1.2 g cm^{-3} and 106 g eq^{-1}, respectively. The accelerator triphenyl phosphine (TPP, Arkema Inc., Canada) has a density and melting point of 1.2 g cm^{-3} and 78.5–81.5 °C, respectively. The epoxy resin and hardener containing 3 wt.% accelerator were mixed at a 100:53 weight ratio.

2.2. Preparation of composites

The composite plates were prepared with various volume fractions of GP/CFF and EP resin to determine the optimum content of conducting fillers for a bipolar plate. The composites had total conducting fillers of 60–80 vol.% and 0–5 layers of CFF. The electrical conductivity of the composite was strongly affected by the volume of conducting fillers that make electrical path-networks.
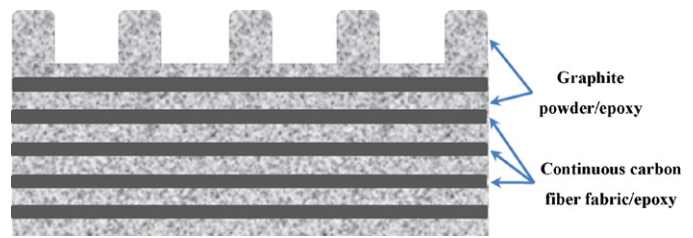

Fig. 1. Cross-sectional view of hybrid carbon-filled, composite, bipolar plate.

Therefore, the amount of conducting fillers in the composite was measured and controlled by the volume percentage. The CFF layers were inserted into the GP/EP composites, as shown in Fig. 1. The increase in the CFF layers decreases the GP content in the composites in order to maintain a constant total filler volume. The GP and EP resin were mixed homogeneously using a high speed mixer (M20, IKA, USA). The GP/EP mixture and CFF were stacked carefully in sequence into the mould with the GP/EP mixture placed in the bottom and top layer. The mould was closed and placed in a high-temperature hydraulic press that was preheated at 100 °C. Pressure was applied slowly to the mould up to 70 kg cm^{-2} and 100 kg cm^{-2} for the GP/CFF/EP and CFP/EP composites, respectively. The composite was cured at 170 °C for 35 min.

2.3. Characterization of composite plates

2.3.1. Thermo-gravimetric analysis (TGA)

The amount of carbon filler in the composite was confirmed by thermo-gravimetric analysis (50Q, TA Instruments, USA). The study was performed from room temperature to 800 °C at a heating rate of 10 °C min^{-1} under a nitrogen atmosphere.

2.3.2. Electrical conductivities

The composite plates were cut with a diamond wheel to dimensions of 127 mm × 60 mm × 3.2 mm to measure the in-plane electrical conductivity by means of the well-known four point probe method according to ASTM D4496, as shown in Fig. 2(a). A Keithley 6220 programmable current source was applied through the two outer probes and the voltage drop across the two inner probes was measured with a Kiethley 2182A nanovoltameter. Ten samples of the composite with the same composition were measured to obtain the average conductivity. The electrical resistivity (ρ) and conductivity (σ) were calculated from the value of the measured electrical resistance (R) and sample dimensions according to the following equation:

$$\rho = \frac{RWT}{L}, \quad \sigma = \frac{1}{\rho} = \frac{L}{RA} = \frac{L}{RWT} \tag{1}$$
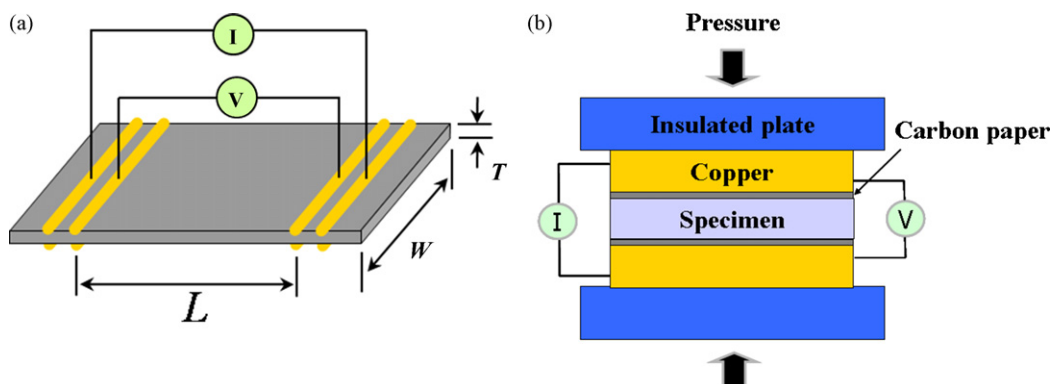

Fig. 2. Schematic diagram of experimental setup for measuring (a) in-plane and (b) through-plane conductivities.

where R is the electrical resistance, and L, W and T are the length, width and thickness of the composite specimen, respectively.

To measure the through-plane conductivity, a composite specimen with dimensions of 50 mm × 50 mm × 3.2 mm was placed between two pieces of carbon paper that made the same contact situation with the fuel cell stack and better electrical contact of the plate with a gold-coated copper electrode, as shown in Fig. 2(b). The measurement was performed using a hydraulic press with an increasing applied pressure to determine the proper clamping pressure of the composite bipolar plate in a fuel cell. The resistance was calculated based on Ohm's law.

The through-plane electrical conductivity of the specimen can be calculated by measuring the resistances of the specimen with carbon paper and the carbon paper separately under the same conditions. The resistance of the specimen was calculated using the following equation [22]:

$$R = R_T - 2R_C \qquad (2)$$

where R, R_T and R_C are specimen resistance, measured system total resistance and carbon paper resistance, respectively. The through-plane electrical conductivity was calculated from the measured R value and sample dimensions using Eq. (1).

2.3.3. Flexural strength

The flexural strength of the composite plate was measured using a three-point bending test on a Universal Testing Machine (STM-10, United, USA) based on the procedure of ASTM D790. Five samples were tested in each case and the mean flexural strength was obtained. The rectangular plate with dimensions of 127 mm × 60 mm × 3.2 mm was used to measure the flexural strength. The crosshead speed and span length for the test were 1.3 mm min^{-1} and 48 mm, respectively. The flexural strength was calculated using the expression:

$$\text{Flexural strength}\,(\sigma^f) = \frac{3PL}{2WT^2} \qquad (3)$$

where P, L, W and T are the load at the yield, span length, width and thickness of the specimen, respectively.

2.3.4. Mouldability

An optical microscope (E600, Nikon, Japan) was used to measure the mouldability of the composite bipolar plate. The mouldability was defined as the ratio of the moulded cross-sectional area to the designed area of the channels in a half-cell bipolar plate. The moulded bipolar plate was cut using a sophisticated diamond wheel to obtain an undamaged cross-section. The cross-sectional area was measured by means of an image analyzer program. An average value for 20 channels on each sample was determined and compared with the designed area of the channel.

3. Results and discussion

3.1. Thermal stability of composites

The TGA curves of prepared GP/EP and CFP composites are given in Fig. 3. These curves suggest that the prepared composite has the desired graphite content and a good thermal stability up to 300 °C that will be sufficient for application in PEMFCs. There is little improvement in thermal stability with increasing graphite content. The TGA curve of the CFP also shows a similar pattern to that of the composite filled with 70 vol.% GP.

3.2. Electrical properties of composites

Bipolar plates collect and transport electrons generated by the electrochemical reaction in fuel cell stacks. Therefore, the materials
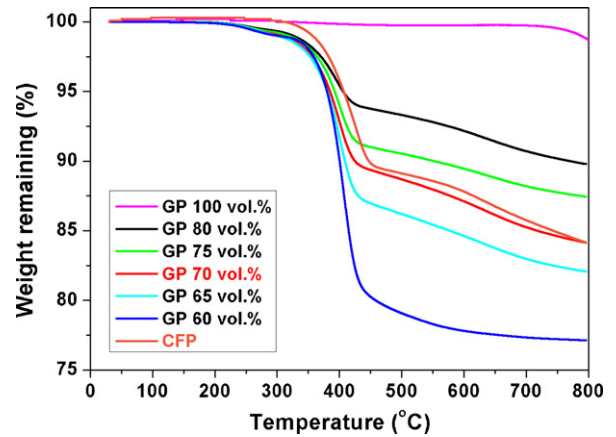

Fig. 3. TGA curve of GP/EP and CFP composites with various GP contents.

for bipolar plates require high electrical conductivity to minimize voltage loss. The electrical properties are one of the most important characteristics among the requirements for a bipolar plate. The property is largely influenced by the total filler loading and filler type. The composite filled with graphite and carbon fibre has anisotropic electrical conductivity depending on the distribution direction of the fillers. For example, a stacked CFP composite has a comparatively high in-plane conductivity (plane direction) and low through-plane conductivity (thickness direction) because it has a resin-rich region between carbon fabrics [17,22–24]. Hence, the in-plane and through-plane electrical conductivity of a composite were measured separately.

The effect of total filler content on the in-plane conductivity of the composites filled with GP and CFF is shown in Fig. 4. CFF1 and CFF2 indicate 1 and 2 layers of CFF respectively, and so on. The electrical conductivity of GP/EP composites increases initially with increasing GP content up to 70 vol.%. The in-plane conductivity of the composite then decreases with further increase in GP content for all types of composite. Compared with GP/CFF/EP composites, the CFP composite has higher in-plane conductivity due to the high conductivity of carbon fabric in the plane direction.

As shown in Fig. 4, the electrical conductivity of the composites increases with increasing number of CFF layers up to a total filler content of ≤65 vol.%. The trend in electrical conductivity is reversed, however, when the total filler content is >70 vol.%. This suggests that CFF layers effectively do not affect the additional conducting networks in the composites at higher filler loading. The in-plane conductivity of the GP/CFF/EP is lower than that of the CFP compos-
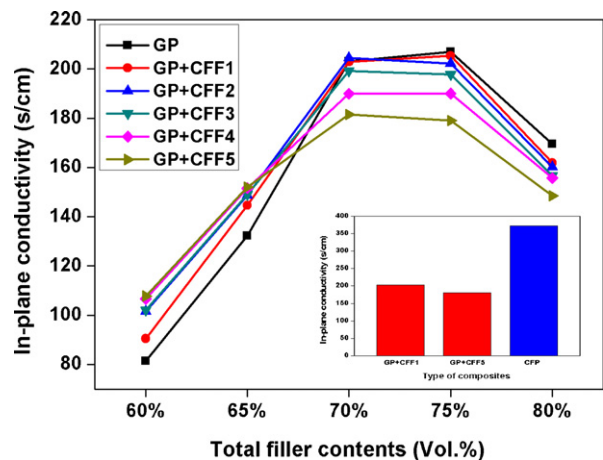

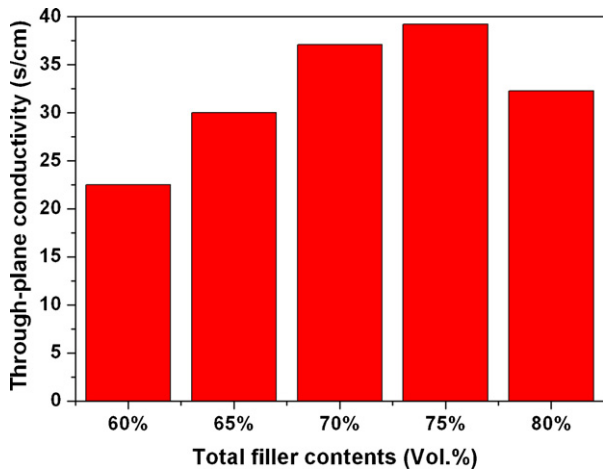**Fig. 4.** In-plane conductivity of GP/CFF/EP composites with increasing filler content.

Fig. 5. Through-plane conductivity of GP/EP composites with increasing graphite content measured at 4 MPa.

Fig. 7. Area specific resistance of GP/EP composites with increasing compaction pressure.

ite containing a 70 vol.% filler contents. Although the conductivity of the GP/CFF/EP composites decreases with increasing number of CFF layers, it is >180 S cm^{-1}, which is much higher than the U.S. Department of Energy (DOE) target value of 100 S cm^{-1}.

The though-plane conductivity of a bipolar plate is also very important because electrons are collected from the surface of the bipolar plate and migrate through the plates in a fuel cell [23]. The through-plane conductivity calculated from the area specific resistance indicates the electrical properties of the plate in the thickness direction. The through-plane conductivity of the composites with various GP contents shown in Fig. 5, has a similar pattern to the in-plane conductivity but is considerably lower. This might be due to the fact that graphite particles of the plank type might orient with in-plane direction when making the plate by compression.

The through-plane electrical conductivity of GP/CFF/EP composites with different numbers of CFF layers and CFP composites is presented in Fig. 6. In the case of GP/CFF/EP composites, the through-plane conductivity decreases gradually with increasing number of CFF layers because of the increase in contact resistance between the CFF and GP in the thickness direction. The GP/CFF/EP composites have approximately 4–7 times higher through-plane conductivity than the CFP composites. The comparatively low through-plane conductivity of the CFP composite may be due to the resin-rich region between the CFP layers, which acts as an insulating layer.
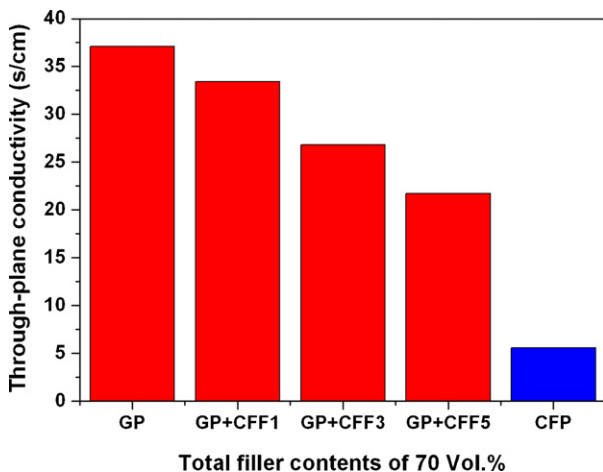
More pathways to transfer electrons were created in the composites by increasing the graphite content up to 75 vol.% which results in a corresponding increase in in-plane conductivity. At a GP content >75 vol.%, however, the amount of polymer resin is not sufficient to bind the graphite particles tightly. Therefore, increasing the filler content leads to a decrease in the compactness of the composites. As a result, the contact resistance between graphite particles increases and the electrical conductivity decreases.

It should be noted that whereas the CFP composite has the highest in-plane conductivity because the continuous carbon fibres have good electrical conductivity, it has the lowest through-plane conductivity. A hybrid system of the CFF and GP would help compensate for the weakness of each component in the composite bipolar plate.

The area specific resistance includes the bulk and contact resistance, and strongly affects fuel cell performance. The contact resistance is quite important because the electrons generated at the electrode pass through the gas-diffusion layer (GDL) to the bipolar plate [25–26]. The measured area specific resistance of the GP/EP and GP/CFF/EP composites is given in Figs. 7 and 8, respectively. The resistance of the composite is very high at low pressure and decreases dramatically with increasing compaction pressure. By contrast, the resistance of the composite is almost constant at high pressure. This suggests that the compaction pressure in a fuel cell stack assembly is an important parameter [27]. Although
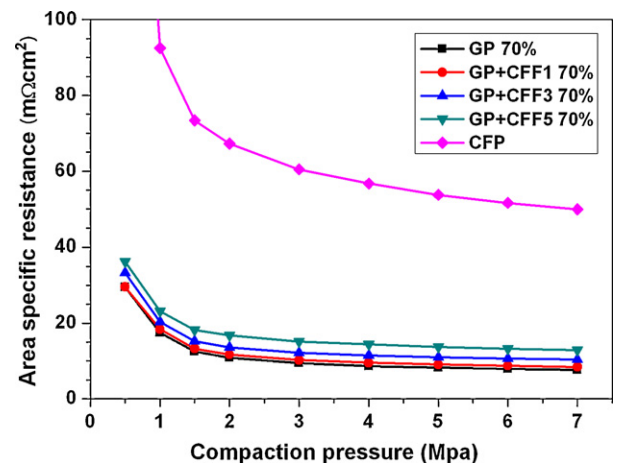

Fig. 6. Through-plane conductivity of GP/CFF/EP and CFP composites measured at 4 MPa.

Fig. 8. Area specific resistance of GP/CFF/EP and CFP composites with increasing compaction pressure.
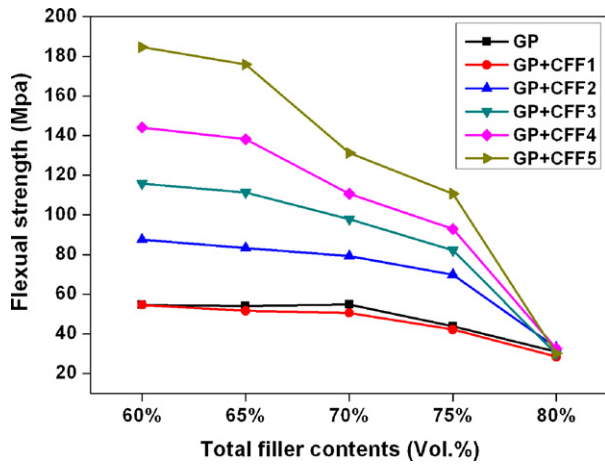
Fig. 9. Flexural strength of GP/CFF/EP composites with various filler contents and layers of CFF.

Fig. 10. Flexural strength of GP/CFF/EP and CFP composites at 70 vol.% filler.

the contact resistance of the composites decreases with high compact pressure during stack assembly, the application of excessive pressure damages the carbon paper and causes separation of the electrolyte and electrode. These results suggest that a compaction pressure >4 MPa will be sufficient for these bipolar composite systems in terms of resistance.

3.3. Flexural strength of composite plates

Bipolar plates require good mechanical strength to withstand the high clamping forces of the stacking and vibrations during vehicular applications [14,16]. The flexural strengths of GP/CFF/EP composites with various GP and CFF contents are given in Fig. 9. The flexural strengths of the GP/CFF/EP composite systems tend
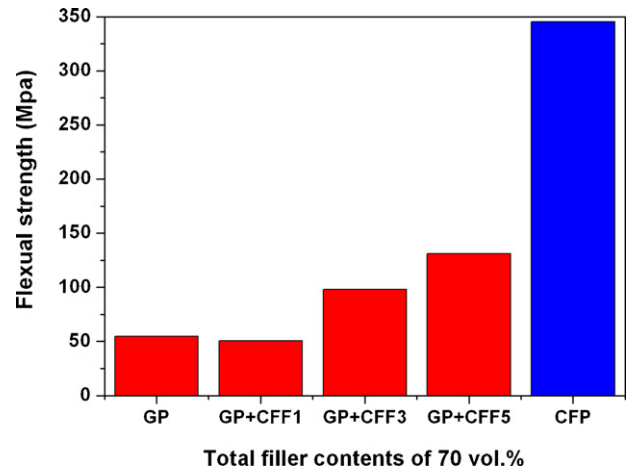
to decrease gradually with increasing carbon filler content over the entire range. In addition, the flexural strength decreases dramatically when the filler content is >75 vol.%. This suggests an increase in imperfection binding between the fillers and epoxy with increasing filler content. The flexural strength of the hybrid composite increases significantly with an increasing number of CFF layers when more than two layers are stacked in the range of filler contents of 60–75 vol.%. When the filler content reaches 80 vol.%, however, the flexural strength has a low value of approximately 30 MPa, regardless of the number of CFF layers because the EP resin is not sufficient to bind carbon materials.

The flexural strength of the composite plates with a 70 vol.% filler content and with different CFF layers is compared with that of a CFP composite plate Fig. 10. It should be noted that the flexural strength of the GP/CFF/EP composite with 5 CFF layers is highest (131.2 MPa,
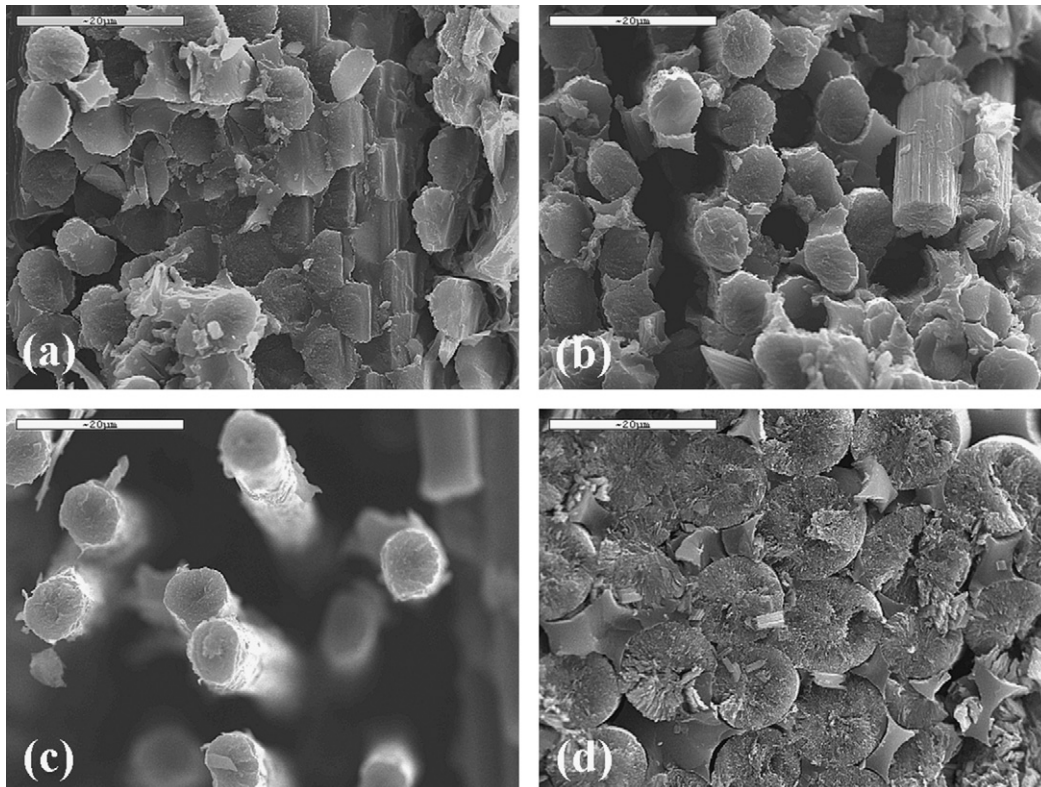

Fig. 11. SEM images of fractured surfaces of GP/CFF3/EP composites; (a) 60 vol.%, (b) 70 vol.%, (c) 80 vol.% GP/CFF, and (d) CFP composites.
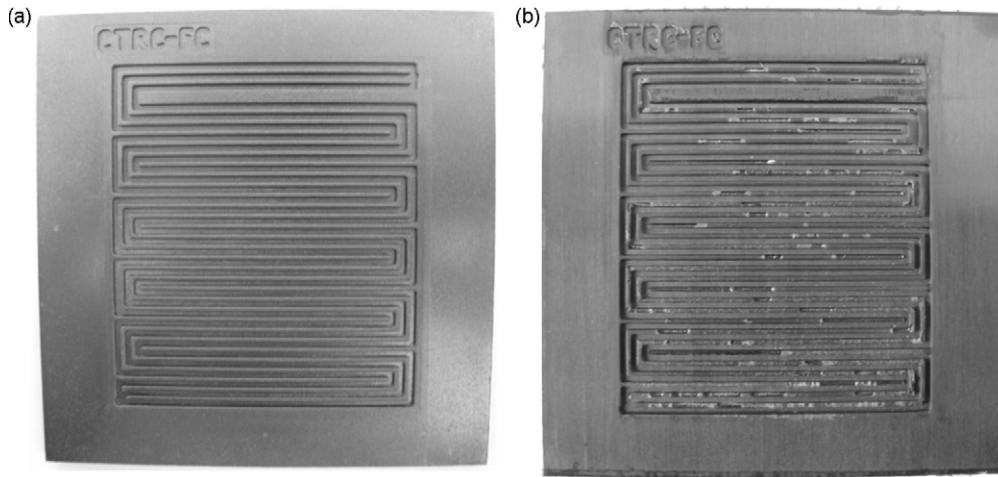
Fig. 12. Moulded bipolar plates; (a) GP/CFF3/EP composite and (b) CFP composite.

238% higher than that of the GP/EP composite) but much lower than that of the CFP composite, 345.2 MPa.

The flexural strengths and electrical properties of the composites can be explained by observation of a fractured surface of the composites. Scanning electron microscopy (SEM) images of the fracture surfaces of the CFP composite and the CFF/GP/EP composite with various graphite contents are presented in Fig. 11. Fibre breakage, pull-out marks and pulled-out fibres are observed in the composites. With increasing filler content, more pulled-out fibres and fewer broken fibres are observed because of the imperfect bonding between the fibres and the resin. Short broken fibres and a compact structure, as shown in Fig. 11(a), suggest that the interface between the carbon fillers and the epoxy resin is strong. The pull-out marks in Fig. 11(b) and pulled-out carbon fibres in Fig. 11(c)

show a relatively weak interface and incomplete bonding between the fillers and the epoxy resin. These results can explain why the flexural property of the composites decreases with increasing filler loading. In the case of the CFP composite, the fracture surface forms a very tight structure with short broken fibres (Fig. 11(d)). This might be due to the very high strength of the CFP composite.

3.4. Mouldability of composite bipolar plates

The gas flow channels in a bipolar plate of a fuel cell are narrow and have complicated shapes in order to supply fuel and oxygen adequately to the required region and remove the water produced by the reaction. The output power and fuel cell efficiency will be reduced if the channels are not formed well. Therefore, the channel
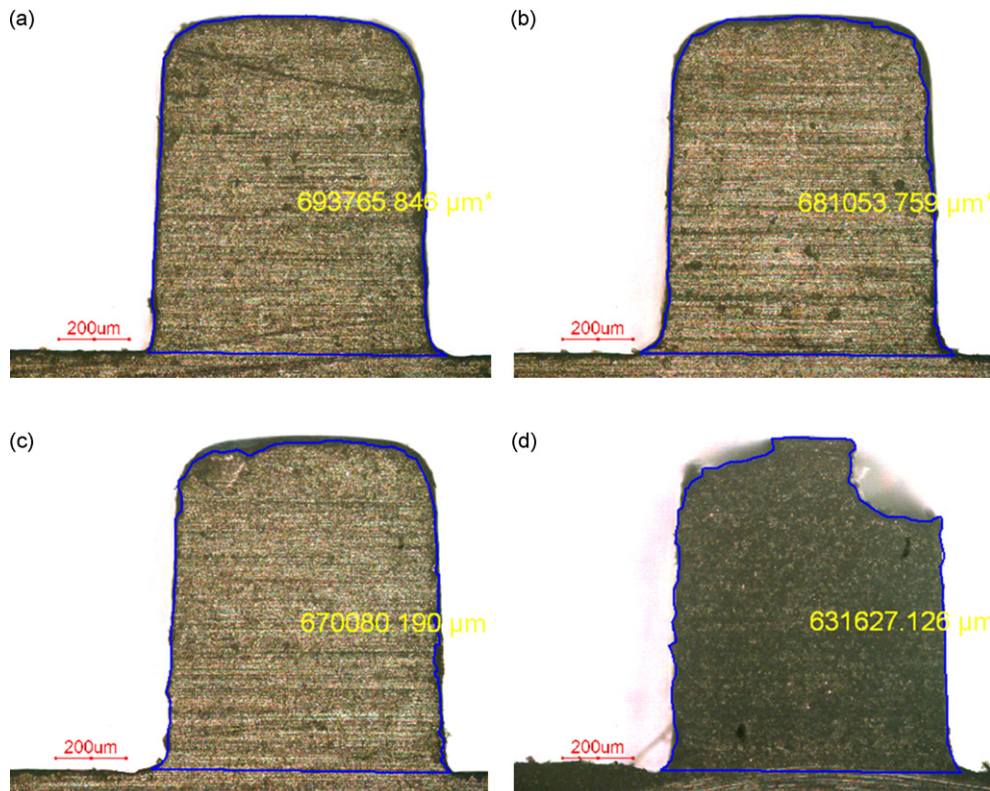

Fig. 13. Cross-sectional view of moulded bipolar plates of various composites: (a) 65 vol.%, (b) 70 vol.%, (c) 75 vol.% GP/CFF3 composites, and (d) CFP composite.

Table 1
Measured mouldability for various carbon filler contents.

Filler content	65 vol.%	70 vol.%	75 vol.%	CFP
Mouldability (area%)	98.1%	96.7%	94.9%	92.8%

mouldability of the composite materials is an important factor for a compression moulded bipolar plate. There are many parameters affecting the mouldability in compression moulding of composite bipolar plates, such as channel depth and width, radius of the corner, and moulding conditions [20,28].

A standard bipolar plate with a parallel serpentine style channel design was moulded to evaluate the mouldability of the GP/CFF/EP composite with various graphite contents and the CFP composites. Two types of compression moulded bipolar plates of GP/CFF/EP and CFP composites are presented in Fig. 12. The bipolar plate of GP/CFF/EP composites had a better formed shape and surface than the CFP composite. In order to measure the mouldability of the composites, the cross-sectional view of the channels was observed through optical microscopy, as shown in Fig. 13, and the area of the formed teeth was calculated using an image analyzer. As the graphite content in the GP/CFF/EP composites increase, channel surfaces are rough and not precisely formed. Channel formation in a CFP composite is quite poor due to the lack of movability of dense carbon fibre with impregnated epoxy resin, as shown in Fig. 13(d). The mouldability, which is calculated as the area ratio of the measured formed cross-section to the designed area of the mould channel, is listed in Table 1. The mouldability of the composites decreases with increasing filler content. Among the composites tested, the CFP composite has the lowest value of 92.8%. The mouldability of the hybrid composites with CFF is sufficient to produce dense channels with a good shape and definition. The comparatively poorer mouldability of the CFP composite might result in a low power output.

4. Conclusions

Composite materials with a hybrid filler system of GP and CFF have been developed to improve the properties of conventional composites for use in bipolar plates. The flexural properties of the GP/EP composite plates are increased significantly by adding the CFF. The lower through-plane conductivity and mouldability of CFF/EP composites could also be improved by adding GP to the composite. The CFF/EP composite shows enhanced through-plane conductivity and mouldability, and the GP/EP composite exhibits improved in-plane conductivity. This study suggests that a plain-weave carbon fabric inserted into graphite/epoxy composites can overcome the weak points of conventional composites, such as GP/EP or CFF/EP composites, in bipolar plates of fuel cells.

Acknowledgements

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