

## EFFECTS OF GRAPHITE/POLYPROPYLENE ON THE ELECTRICAL CONDUCTIVITY OF MANUFACTURED BIPOLAR PLATE

(Kesan Komposit Polimer Pengalir Grafit/Polipropilena Komposit Polimer Pengalir Ke atas Sifat Kekonduksian Elektrik bagi Pembuatan Plat Dwikutub)

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### Abstract

Conductive polymer composite (CPC) has been used on bipolar plates by compression and injection molding methods. The CPC material comprises 25% polypropylene and 75% graphite with size variation (40, 100, 150, and 200  $\mu\text{m}$ ) according to particle size dispersion test. The composite is mixed using an internal mixer to obtain a homogeneous mixture at a process temperature of 200 °C. Electrical conductivity tests are performed on each particle size composition. The highest electrical conductivity produced by compression and injection molding methods is 17 and 12 S/cm, respectively. These values are both obtained on a 40  $\mu\text{m}$  graphite filler. Results show that the type of manufacturing process affects the value of electrical conductivity using the same material.

**Keywords:** injection molding, compression molding, bipolar plate

### Abstrak

Komposit polimer pengalir (KPP) telah digunakan pada plat dwikutub dengan kaedah pengacuan mampatan dan pengacuan suntikan. Komposisi bahan KPP adalah polipropilena (PP) 25% dan grafit (G) 75% berat dengan variasi saiz 40  $\mu\text{m}$ , 100  $\mu\text{m}$ , 150  $\mu\text{m}$  dan 200  $\mu\text{m}$  dan dijalankan ujian serakan partikel saiz. Komposit dicampur menggunakan campuran dalaman untuk mendapatkan campuran yang sekata pada suhu proses 200 °C. Pengujian kekonduksian elektrik dijalankan pada masing-masing komposisi saiz partikel. Nilai kekonduksian elektrik tertinggi ditunjukkan pada pengisi grafit saiz 40  $\mu\text{m}$ , pada masing-masing kaedah pembuatan. Kekonduksian elektrik tertinggi dihasilkan melalui kaedah pengacuan mampatan iaitu 17 S/cm dan proses pengacuan suntikan adalah 12 S/cm. Keputusan ujian menunjukkan bahawa proses pembuatan memberi kesan ke atas nilai kekonduksian elektrik menggunakan bahan yang sama.

**Kata kunci:** pengacuan suntikan, pengacuan mampatan, plat dwikutub

### Introduction

The use of conductive polymer composite (CPC) in various applications related to renewable energy has increased in recent years. The manufacture of bipolar plates for polymer electrolyte membrane fuel cell technology (PEMFC) is a material application of CPC. Bipolar plates, the main components of PEMFC, are important in continuous

system. These plates contribute to the gas and water regulation, mechanical stability, and electrical conductivity of the fuel cell [1-3]. Hence, the bipolar plate manufacture requires filler material designs and manufacturing methods that can improve the electrical conductivity. Generally, a strong relationship between the materials used and the manufacturing process of the bipolar plate can produce the most excellent CPC. One of the filler materials commonly used is a carbon-based conductive filler material, which is an alternative to metals with electrical conductivity values ranging from  $10^2$  S/cm to  $10^5$  S/cm [4].

Graphite, a carbon-based material, is proposed as a filler in the manufacture of bipolar plates. Selection of graphite as a filler material in the CPC application of bipolar plate presents advantages over metal-based materials. The carbon-based material is lighter with higher corrosion resistance and lower cost than that of metal, which is carbon malleable [5-8]. Carbon-based materials also display drawbacks, such as considerable brittleness and difficulty in machining to satisfy the specifications of the fuel cell stack. Robberg et al. [9] demonstrated that the ability filler (G) in the CPC indicates that the substantive polymer is an insulator, and graphite filler loading of more than 60% by weight in the composite can satisfy the minimum electrical conductivity.

Filler powder characterization is carried out through the filler particle size distribution analysis. Particle size distribution is generally displayed in the form of a normal distribution graph. Further statistical particle size distribution in the histogram shows three important values: the lowest value of the distribution ( $D_{10}$ ), median ( $D_{50}$ ), and the highest distribution ( $D_{90}$ ) [10]. High  $S_w$  indicates a narrow size distribution. Conversely, low  $S_w$  suggests a wide particle size distribution. Generally, the powder is easily processed by injection molding when the  $S_w$  is smaller than 2. The difficulty in feedstock injection increases when the  $S_w$  is more than 4. Feedstock injection is considerably difficult when the  $S_w$  is more than 7.

Furthermore, the manufacturing process highly influences the easy processing of powder; it can increase the electrical conductivity and the CPC properties. Compression and injection molding processes are utilized in the production of various types of products used in the manufacture of plastic products and bipolar plates. The method used is determined based on the advantages of each manufacturing method [2, 11-12].

#### Materials and Methods

This study used SM240 polypropylene (PP) (Titan Petchem Sdn Bhd) as a binder, which presents the following properties: melting point, 160 °C–170 °C; processing temperature, 190 °C–250 °C; melt flow rate, 10 g/10 min; and density, 0.9 g/cm<sup>3</sup>. The electrically conductive material used in this study is a natural graphite from Asbury 3243 (Asbury Graphite Mills, Inc.). Graphite displays the following important properties: carbon content, 99%; normal powder size,  $\leq 60$   $\mu$ m; surface area, 3 mm<sup>2</sup>; electrical resistivity, 0.036  $\Omega$ ; and density, 1.74 g/cm<sup>3</sup>. The graphite particles are sorted into the following sizes: 40, 100, 150, and 200  $\mu$ m. Particle size dispersion is carried out using a Malvern Mastersizer E version. Samples are manufactured using a compression molding method by hot compression with a capacity of 200 tons and injection molding of the plunger DSM Xplore (semiautomatic). Critical powder loading test determined that the compositions of PP binder and graphite (the highest filler material) are 25% and 75% by weight, respectively [13]. Particle size analysis and particle size distribution are conducted on each graphite size by using a Malvern Mastersizer E version. The electrical conductivity of the composite is evaluated using four-point probe tool window (Multi Height Four-Point Probe) with test unit RM3. Scanning electron microscopy (SEM) is used to observe graphite powder.

#### Results and Discussion

Figure 1 shows the graphite powder shape, which displays advantages and disadvantages for compression and injection molding processes. Graphite flake can produce high electrical conductivity of approximately 420 S/cm in the minus 320 mesh [14]. The flake shape causes difficulties in the flow injection molding. Mixing of particles with various sizes affects the particle arrangement in composites.

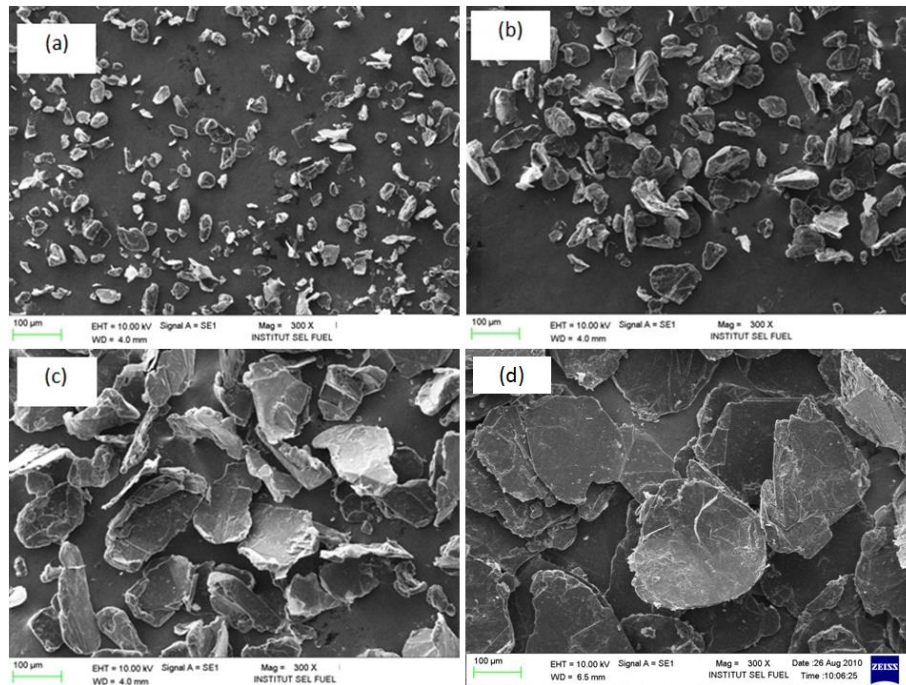


Figure 1. Scanning electron microscopy images of graphite type 3243 with the following particle sizes after sieving: (a) 40, (b) 100, (c) 150, and (d) 200 µm

Analysis results of the filler particle size distribution are shown in Table 1. The distribution percentage of particle size shows particle size distributions of 10%, 50%, and 90%, which are named as  $D_{10}$ ,  $D_{50}$ , and  $D_{90}$ , respectively. The median size of the powder is a cumulative percentage of 50%, which is often named as the  $D_{50}$  size (Figure 2(a)) of the graphite particle with a size of 40 µm. The size distribution curve ( $S_w$ ), which fills the space between large and small particles in the filler composition percentage of 75% by weight, shows large graphite particle sizes.

Table 1. Average particle size of the graphite powder filler

(µm)	Percentage Distribution of Particle Sizes			Surface Area	
	$D_{10}$	$D_{50}$	$D_{90}$	(m <sup>2</sup> /g)	$S_w$
As received	18.8	56.4	127.8	0.161	2.12
40	18.1	34.8	62.3	0.204	4.76
100	26.5	104.3	212.3	0.125	2.83
150	27	151.7	291.9	0.095	2.47
200	39.8	201.4	364.8	0.075	2.66

The median particle size ( $D_{50}$ ) and distribution curve ( $S_w$ ) are key factors in the analysis of conductive powder.  $S_w$ , the cumulative distribution curve lognormal, is equivalent to multiplied or standard deviation of the particle size distribution. The distribution curve ( $S_w$ ) affects the performance of the injection molding process. When the  $S_w$  value is high, the particle size distribution is narrow. By contrast, the low  $S_w$  value indicates a wide particle size distribution.

High  $S_w$  value indicates narrow particle size distribution. Low  $S_w$  value results in even distribution and easy injection process. The graphite filler powder presents a wide particle size distribution and low  $S_w$  value of 2, thereby

causing its easy injection. A graphite with 100  $\mu\text{m}$  particle size (Figure 2. (b)) also shows  $S_w$  in wide particle size distribution, which indicates easy injection. The same trend is observed on graphite with particle sizes of 150 (Figure 2(c)) and 200  $\mu\text{m}$  (Figure 2(d)). Generally, the CPC (G/PP, 75%/25%) exhibits a low  $S_w$  value, which suggests its mold ability.

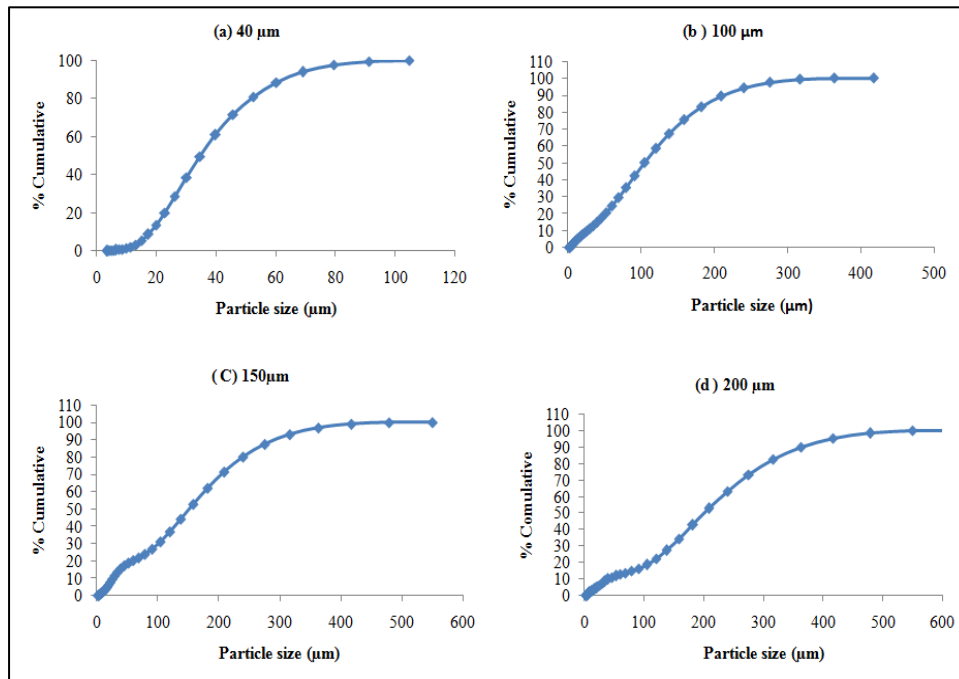


Figure 2. Cumulative percentage of the volume of graphite particle size distribution: (a) 40, (b) 100, (c) 150, and (d) 200 $\mu\text{m}$

The electrical conductivity test results (Figure 3) show that the electric conductivity value decreases with increased particle size. This trend is observed in both CPC plate manufacturing methods. The electrical conductivity produced by compression molding method displays a higher value (17 S/cm) than that of the injection molding method (12 S/cm) in the 40  $\mu\text{m}$  particle size. Consequently, the filler particle dispersion produced by compression molding is higher than that by injection molding method. Particle shape influences the particle dispersion of fillers. The flake particle shape is more suitable for compression molding method in producing high electrical conductivity value than those of other forms, such as sphere [14]. This condition is attributed to the flake shape, which can easily form the dispersion of compression molding due to the large pressure area on the compression. The electrical conductivity devaluation and subsequent increase in particle size are also shown in Figure 3.

This phenomenon is due to the formation of empty spaces between large particles. The free space formation for electrical conductivity hinders the establishment of electricity networks because PP, as binder, will fill the empty space formed between particles. This observation is supported by the SEM image in Figures 4 and 5 for the injection and compression molding processes, respectively. Large particle size will increase the pore size and electrical resistance of the composite material. Figure 3 displays that the electrical conductivity produced by injection molding approaches the values of compression molding method. This trend is due to the particle form, which increases the electrical conductivity value produced by the former method. In general, the particle shape also affects the electrical conductivity value. The graphite flake used as filler in this research can improve the electrical conductivity value. However, the flake shape of the filler material for injection molding can constrain the feedstock flow due to the rotation of particles at the injection proses of feedstock, which results in irregular formation of a mutable block in the flake structure [15]. Increasing the injection pressure can reduce the effects of constraints

caused by agglomeration. This finding is supported by the microstructure observation using SEM, as shown in Figures 4 and 5 for injection and compression molding methods, respectively.

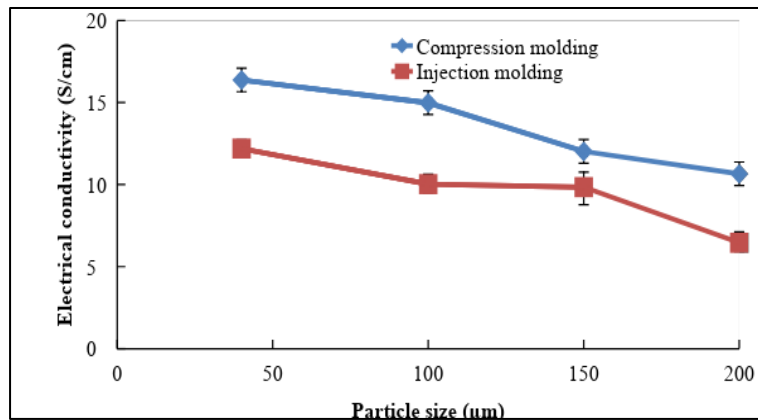


Figure 3. Electrical conductivity by compression and injection molding methods

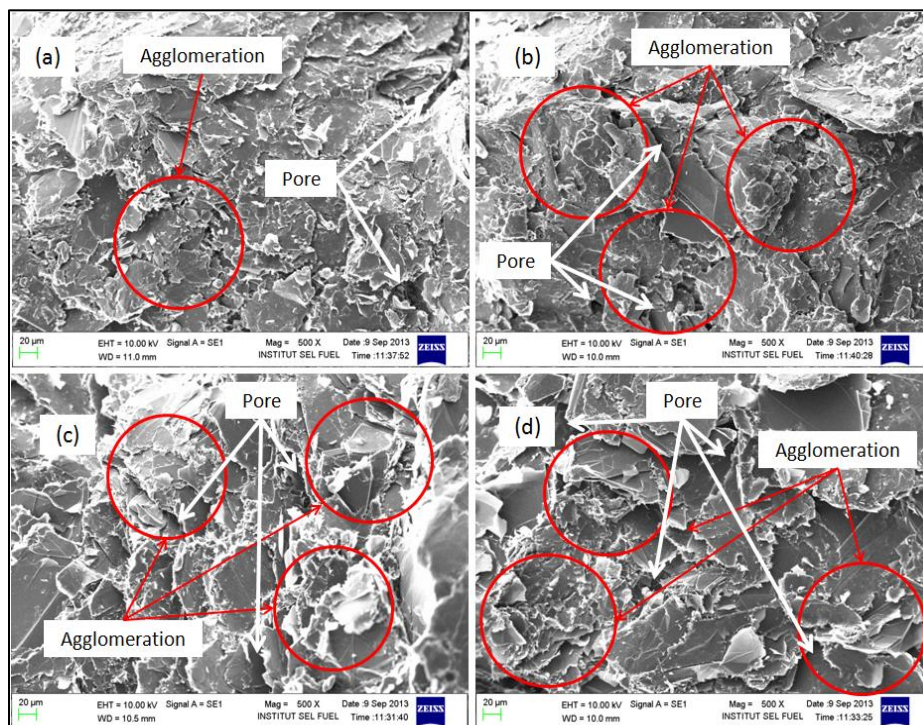


Figure 4. SEM images for the injection molding method:(a) 40, (b) 100, (c) 150, and (d) 200 µm

The SEM image in Figure 4. demonstrates the uniform particle dispersion and the particle agglomeration, which can also result in networking between particles. Consequently, the electrical conductivity increases, and the graphite filler content is higher than 60% by weight [3]. Figure 4(a) shows the particle size agglomeration of 40 µm on several fronts, and the solid bond between particles is shown in red circle. Thus, the electrical conductivity increases, which can affect the flow ability of the feedstock in the injection molding process. The small pores of the composite, which are indicated by an arrow at a composite size of 40 µm, increase the electrical conductivity. By

contrast, Figures 5 (a), 5(b), 5(c), and 5(d) present an increase in agglomeration, as indicated by the red circle. This increase also influences the increase in pore volume in composites indicated by the arrow. Such result is similar to Mahyoedin [16] who found that particle size affects the flow ability of feedstock during the injection molding process. Increased pore size will reduce the networking between particles, as shown in the compositions of particles with sizes of 100, 150, and 200  $\mu\text{m}$  (indicated by the arrow), and decrease the electrical conductivity.

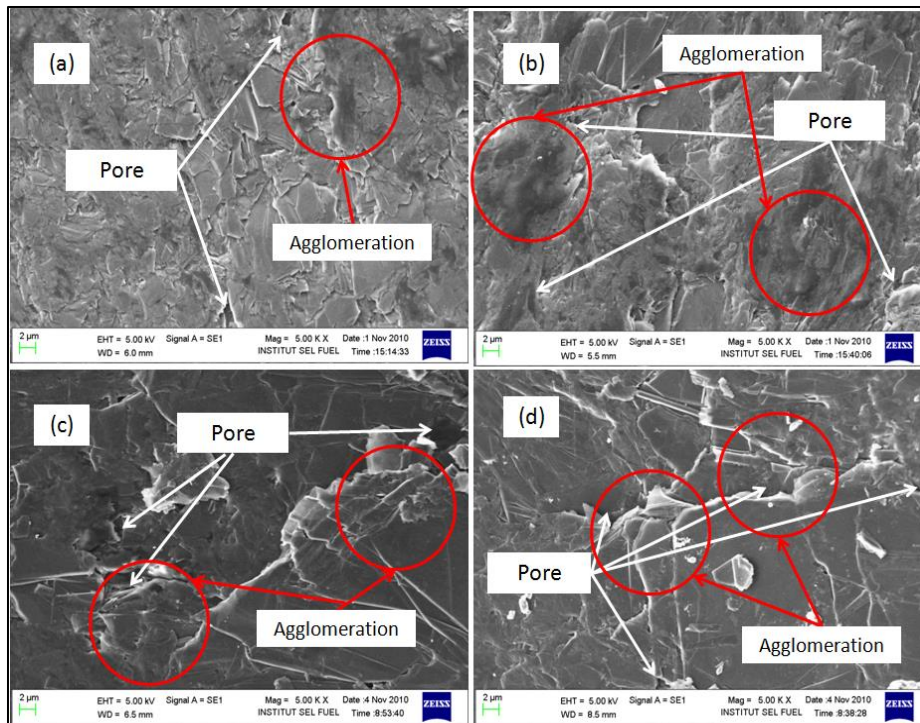


Figure 5. SEM images for the compression molding method: (a) 40, (b) 100, (c) 150, and (d) 200  $\mu\text{m}$

SEM images in Figure 5 show a compact binding between particles by compression molding. Nonetheless, the pores that can affect electrical conductivity still exist. The composite microstructure displays compact particle arrangement. Thus, the electrical network increases the electrical conductivity. Figures 5(a-d) generally indicate a more compact surface in the composite formation by compression molding than that with the injection molding method. The composite microstructure analysis results for the compression and injection molding processes are in accordance with the electrical conductivity test results presented in Figure 3.

The electrical conductivity produced by compression molding is higher than that by injection molding. The filler particles comprising a close composite by the compression process can develop better electrical networks than those by injection molding. The SEM image analysis on compression molding (Figure 5) demonstrates that the number of pores marked with arrows and the agglomeration indicated with red circle are lower than those of the injection molding process.

### Conclusion

Injection and compression molding methods are often used in the manufacture of bipolar plates with PP and G. The particle size variation in the composite (25% PP and 75% G by weight) with different manufacturing processes affects the electrical conductivity. The highest electrical conductivity produced by both manufacturing methods is observed in 40  $\mu\text{m}$  particle size; the highest value of 17 S/cm is produced by compression molding. The use of small particle sizes in the compression molding process affects the particle compaction by reducing the number of pores and agglomeration and increasing the electrical network between particles.

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