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Effect of Multi-Sized Graphite Filler on the Mechanical Properties and Electrical Conductivity

(Kesan Pengisi Grafit Berbilang Saiz pada Sifat Mekanikal dan Kekonduksian Elektrik)

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ABSTRACT

This research successfully fabricated conductive polymer composites (CPCs) prepared using multiple sizes of graphite filler (40, 100, 150, and 200 µm) that provided excellent network formation within the fillers and polypropylene matrix which further improved both electrical conductivity and flexural strength. An important discussion on the fabrication technique, including compression moulding and injection moulding was conducted, to manufacture CPC materials with a thickness less than 3 mm. The findings of this study suggested that fabricating CPCs using the compression moulding technique with a graphite composition of 75 wt. % exhibited better network connectivity as the electrical conductivity increased to 15 Scm^{-1}. Also, compared to the three sizes of graphite filler (40/100/200 µm) it resulted in 13 Scm^{-1}, with two sizes (40/200 µm) reporting better electrical conductivity at 15 Scm^{-1}. This demonstrated that the addition of multiple sizes was not necessarily due to agglomeration occurring. The resultant graphite composites of 40/200 µm possessed a more stable structure having a thin composite layer (2.5 mm) which promoted better electrical conductivity suitable for bipolar plate used in proton exchange membrane fuel cells.

Keywords: Carbon; composites; electrical conductivity; fuel cells; mechanical properties

ABSTRAK

Kajian ini berjaya menghasilkan konduktif polimer komposit (KPK) dengan menggunakan beberapa jenis saiz pengisi grafit (40, 100, 150 dan 200 µm) yang berupaya menghasilkan jaringan elektrik yang cemerlang antara pengisi dan matrik polimer sekaligus meningkatkan nilai keberaliran elektrik dan kekuatan tegangan. Perbincangan penting ditekankan pada kaedah pembuatan termasuk penggunaan kaedah pembentukan mampatan dan pengacuan suntikan dalam pembikinan bahan KPK berketebalan kurang 3 mm. Penemuan kajian ini menunjukkan pembuatan bahan KPK menggunakan kaedah pembentukan mampatan pada komposisi grafit sebanyak 75 % bt. mampu menghasilkan jaringan keberaliran elektrik yang baik dengan nilai keberaliran elektrik direkodkan sebanyak 15 Scm^{-1}. Perbandingan ke atas tiga saiz pengisi grafit berbeza iaitu (40/100/200 µm) memperoleh nilai keberaliran elektrik sebanyak 13 Scm^{-1} manakala penggunaan dua saiz pengisi grafit memperoleh nilai sebanyak 15 Scm^{-1} iaitu jauh lebih baik. Keadaan ini menunjukkan bahawa pertambahan saiz berbeza tidak semestinya meningkatkan nilai keberaliran elektrik kesan daripada pergumpalan yang lebih mudah berlaku. Kajian menunjukkan komposit grafit dengan saiz 40/200 µm mempunyai struktur yang lebih stabil serta nilai keberaliran elektrik lebih tinggi dengan ketebalan 2.5 mm bersesuaian dengan aplikasinya sebagai plat dwikutub dalam sel fuel membran penukar proton.

Kata kunci: Karbon; keberaliran elektrik; komposit; sel fuel; sifat mekanikal

INTRODUCTION

Conductive composites are acknowledged as powerful material nowadays given the world has moved more towards renewable energy and green technology. However, one may ask, how do conductive composites relate to those fields? The answer is, by introducing conductive composites, this material has been applied in many diverse types of applications such as fuel cell technology, electronic sensors and even medical biosensor equipment (Folorunso et al. 2019; Leigh et al. 2012; Rozlosnik 2009; Ul-Islam et al. 2015). Although, the research in this study, focused on conductive composites as a major component in proton exchange membrane fuel cells (PEMFCs). Prior studies have discussed conductive composites used as a bipolar plate, which are the main components in PEMFCs (Liu et al. 2019; Mohd Radzuan et al. 2017a). In fact, industries have mass-produced bipolar plates for fuel cells which are available in today's market. Even though conductive composites have been produced and discussed in detail, none of the industries and researchers has been able to fabricate a conductive bipolar plate as thin as 2.5 mm in size due to limitations around brittleness and electrical conductivity inhibition (Liu et al. 2019; Wang et al. 2016). A competitive materials fabrication among industrial lead on graphite and conductive polymer composites (CPCs) thickness within the range of 0.6 to 2.5 mm. Yet, the fabrication of thinner (< 2.5 mm) graphite and CPCs materials are intensely hard to manufacture compared to the metal sheet that proven ability to be as thin as 0.1 mm as reported by another study (Martín et al. 2014). These occurred due to the fact that as the thickness of graphite or CPCs materials reduce lower than 2.5 mm, the materials either become brittle and delicate hence defeat the purpose.

Nowadays, industries and researchers alike can produce a thin layer of the bipolar plate which is metal-coated based resulting in excellent electrical conductivity having a thickness of 1 mm (Li et al. 2019; Wang et al. 2012). While these sound promising, the metal-coated bipolar plate often relies on its durability and ability to withstand corrosion effects which deteriorates the plate over time and affects the overall performance of the fuel cell (Antunes et al. 2010; Pollet et al. 2019). Previous studies have showed that there are various types and compositions of conductive composites in which its percolation threshold has examined to obtain the maximum electrical conductivity (Folorunso et al. 2019; Qu et al. 2017). Additive fillers including carbon black, graphene, carbon nanotube, in numerous geometry shapes have often been added to improve the conductivity of composite materials in which the addition of filler often results in impressive conductivity output (Breuer & Sundararaj 2004; Li et al. 2015; Sharma & Pollet 2012). Noted that, the single size filler has been discussed elsewhere, which reported that the maximum electrical conductivity of 11 Scm^{-1} obtained when using graphite reinforced polypropylene materials (Adloo et al. 2016). However, the addition of filler somehow increases fabrication costs which are avoided by industries as the additional fillers used were mainly graphite-based (Alegre et al. 2019; Yang et al. 2019). Also, the addition of filler will result in producing a thick layer of the composites on the bipolar plate, increasing the total weighted of the fuel cells stacked together (Antunes et al. 2011). Therefore, this study proposes using a simple graphite material with different geometric sizes which will assist in maintaining the flexural properties as the fabricated composites had a thickness of 2.5 mm.

MATERIALS AND METHODS

MATERIALS AND FABRICATION OF CONDUCTIVE COMPOSITES

The conductive polymer composites fabricated for this study consisted of pure graphite and polypropylene as its matrix. The conductive graphite fillers, grade Asbury 3243, were supplied on multiple sizes of 40, 100, 150, and 200 µm obtained from Asbury Graphite Mills, Inc., New Jersey. Graphite fillers often exist in several geometries and shapes. However, this study used a flake-like structure given its ability to produce electrical conductivity at the upmost value of 420.3 Scm^{-1} compared to a spherical structure at 387.3 Scm^{-1} (Chen et al. 2018). The graphite fillers had a carbon content of 99%, an average surface area of 3.0 mm2, a density of 1.74 gcm^{-3}, and an electrical resistance of 0.036 Ω, as prescribed by the supplier. Meanwhile, the polymer resin which was polypropylene (PP), was supplied by Titan Petchem Sdn Bhd., grade TitanPro SM420. The polypropylene had a melting temperature between 160 and 170 °C, average electrical conductivity of 10-14 Scm^{-1}, a flow rate of 25 g 10 min^{-1}, and an average density of 0.9 gcm^{-3}. This study considered polypropylene as its polymer matrix since polypropylene can easily degrade when compared to epoxy resin which is the thermoset resin that is normally used in bipolar plate fabrication (Arutchelvi et al. 2008; Tavares et al. 2017). Although it is known that thermoset materials such as epoxy/carbon fibre able to reach the maximum electrical conductivity of 64.83 Scm^{-1}, their limited incorporation with fewer amount of filler is intolerable (Planes et al. 2012). These features are crucial as the bipolar plate itself is purposely used for renewable technology in fuel cells. Hence, the intention of developing a green product was one of the key factors in this research (Adloo et al. 2016).

The graphite fillers were initially prepared at two different compositions; the combination of two fillers and three fillers, as shown in Table 1. Theoretically, the combination of different types of filler geometry will assist in compactness and improve conductivity network formation since most of the graphite fillers hold one another, which results in tunnelling effects (Ardanuy et al. 2011; Zare & Rhee 2019). Also, the 75 wt. % of graphite filler loading was selected since it is the maximum percolation threshold for graphite materials as prescribed in other studies ((Dhakate et al. 2008; Radzuan et al. 2019). These different sizes of graphite then were pre-mixed using a ball milling machine (model Fritsch Pulverisette 6) with 20 stainless steel balls at a rotational speed of 300 rpm for 90 min (Suherman et al. 2013; Zakaria et al. 2015). Next, the pre-mixed fillers were mixed with the polypropylene resin using an internal mixer (model Haake Rheomix Roller rotor 600R) at a mixing temperature of 200 °C and a rotational speed of 50 rpm for 17 min (Mohd Radzuan et al. 2017a; Zakaria et al. 2015). The composites compound mixed were then crushed using the crusher at room temperature until the compound was of an average size of 2 mm (Radzuan et al. 2019). The prepared composites compound was later used for bipolar plate fabrication employing two different processing techniques, namely injection moulding, and compression moulding.

The compound underwent the injection moulding process to undergo a rheology study using a rheometer (model Shimadzu CFT-500) having a length of 10 mm and a diameter hole of 1 mm based on the JIS K7210 (Ismail et al. 2008). The prepared composite compound was later injected in the form of square shape plate using an injection moulding machine, (model DSM Xplore Semi-automatic) at a maximum pressure of 16 bar and nozzle temperature of 200 °C, while, the heater temperature was maintained at 350 °C. Regarding the compression moulding technique, 200-tonne compression moulding was used to fabricate the composite compound with a sample size of 140 × 60 × 2.5 mm. During the fabrication process, the temperature applied was 185 °C for 25 min pre-heating beforehand and compressed at a pressure of 750 kN for 15 min.

TABLE 1. Combination of different graphite geometry and compositions

Compositions	Polypropylene (wt. %)	Graphite (wt. %)			
		40 µm	100 µm	150 µm	200 µm
40 µm / 100 µm	25	37.5	37.5	-	-
40 µm / 150 µm	25	37.5	-	37.5	-
40 µm / 200 µm	25	37.5	-	-	37.5
40 µm / 100 µm / 150 µm	25	30	35	10	-
40 µm / 100 µm / 200 µm	25	30	35	-	10
40 µm / 150 µm / 200 µm	25	30	-	35	10

CHARACTERISATION

The characterisation performed onto the polypropylene reinforced graphite mainly focused on the particle size and morphological analysis as the fabricated composite compound consisted of different sizes. The micrograph images were analysed at the sample's cross-section using a Scanning Electron Microscope (SEM), (model Zeis EVO MA10, Malaysia). For the particle analysis of two and three particle sizes, the analysis was performed using a Malvern Master Sizer E-version 5.54 in a wet condition of graphite fillers. Aside from conducting the morphological analysis, the fabricated composite compound was also tested for in-plane electrical conductivity to identify its electrical performance. This is crucial as composite materials used as bipolar plate materials acquire good electrical conductivity in very thin sizes less than 3 mm (Middelman et al. 2003). Therefore, the in-plane conductivity was measured using a Jandel four-point probe and recorded using an RM3 test unit. The prepared polypropylene reinforced graphite samples then underwent a flexural test using a Universal Testing Machine (UTM), (model Instron 5567) with dimensions of 100 × 12.7 × 2.5 mm. The gauge length was fixed at 50 mm based on the ASTM D760-03 standard with the crosshead speed of 5 mmmin^{-1}.

RESULTS AND DISCUSSION

ELECTRICAL CONDUCTIVITY ANALYSIS

Since the development of a thin layer (2 to 3 mm) of composites is now viewed as the main strategy in bipolar plate applications, its behaviour towards electrical conductivity due to its compactness and lesser conductive pathway performed by the composite materials itself is often discussed (Middelman et al. 2003). However, as indicated in Figure 1(a), through the compression moulding process, the electrical conductivity was able to be maintained between 10 and 15 Scm^{-1} compared to the injection moulding process for both the two sizes and three sizes of graphite fillers. This shows that regardless of the sizes and morphological aspects used during the fabrication of conductive composites, the compression moulding process results were on par between the two sizes and three sizes of graphite fillers. This can be observed as shown in Figure 1(a), indicating 15 Scm^{-1} for 40/200 µm, which results in higher electrical conductivity compared to 40/150/200 µm at 6 Scm^{-1} (Figure 1(b)). These findings were also supported by other research in which the addition of other filler sizes caused an agglomeration or even an unsmooth networking pathway during the process (Wang et al. 2011). Theoretically, the excellent conductivity formation depends on filler contact (end-to-end contact) compared to redundant (body-to-body) or (end-to-body) contact (Lux 1993; Mohd Radzuan et al. 2017b). Hence, this study proved that the optimum filler addition only involved one type of geometric shape of the same filler rather than multiple sizes applied. In contrast, the use of 40/100 µm and 40/150 µm did not seem to show much difference regarding the electrical conductivity of 12 and 10 Scm^{-1}, respectively, compared to 40 /200 µm with the value of 15 Scm^{-1}. Prior studies showed that this was due to the smaller particle sizes, which may have filled the gap within the primary filler of 40 µm, thus resulting in tunnelling effects (Hamimah et al. 2010; Wang et al. 2017). Although, under some circumstances, the fillers with a higher contact area will further result in leap tunnelling conductive occurrence (Ardanuy et al. 2011). These key findings are most important given researchers have only tended to focus in fabricating conductive composites of a thick size (> 3 mm). Industries have started to concentrate on using a thinner layer of the conductive composite while maintaining its electrical conductivity and brittleness at the utmost level. Noted that, the through-plane conductivity of conductive composite materials has not been measured and reported in Figure 1 due to the fact that theoretically, through-plane conductivity is half of the in-plane conductivity of the materials (Radzuan et al. 2017).

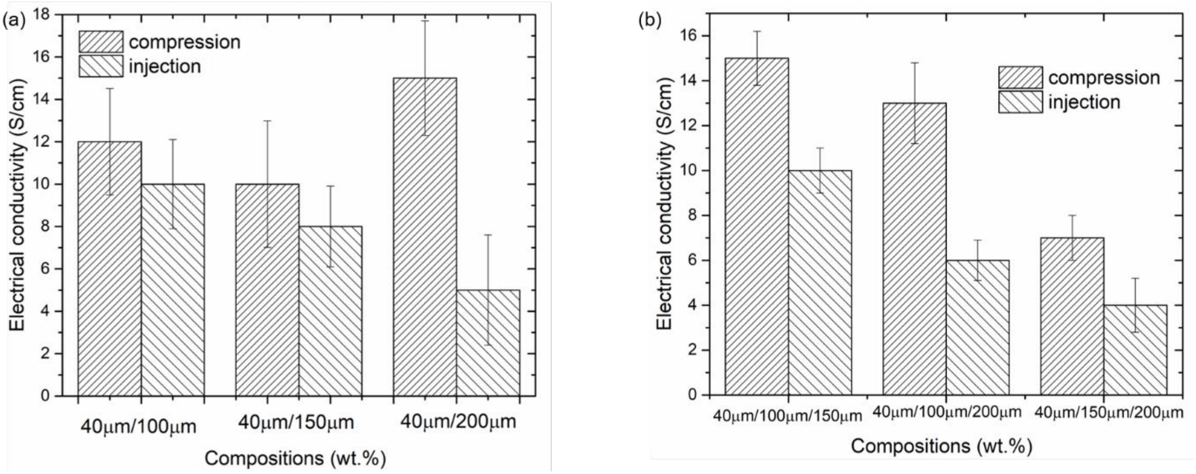

FIGURE 1. In-plane electrical conductivity of (a) two sizes (40/100 µm, 40/150 µm, 40/200 µm) and (b) three sizes (40/100/150 µm, 40/100/200 µm, 40/150/200 µm) of graphite filler developed by the injection moulding and compression moulding technique

FLEXURAL STRENGTH ANALYSIS

As discussed earlier regarding graphite composites, using only two different sizes will allow good electrical conductivity formation, and thus more filler sizes are unnecessary. The flexural strength analysis further supports these findings, as shown in Figure 2, as both graphite sizes were on par with 26 and 30 MPa for 40/200 μm and 40/150/200 μm, respectively. As such, the results demonstrate (as shown in Figure 2(a)) that by only using two types of graphite sizes, it was able to acquire the standard strength (> 25 MPa). Figure 2(a) shows the graphite composite fabricated through the compression moulding process encountering lower flexural strength compared to the injection moulding process. Even though these phenomena were rarely discussed, prior studies have indicated that compression moulding can only produce flexural strength of around 40 MPa, while the injection moulding process can reach up to ~70 MPa, respectively (Adloo et al. 2016; Kuo & Chen 2006; Lee et al. 2009). This was mainly because injected composite was able to align and adjust the filler directions and arrangement (Qu et al. 2017; Shou et al. 2013). Notwithstanding, studies have shown that due to the geometric shape (i.e. flake-like structure) of the graphite filler, it was able to withstand excessive load applied, illustrated in Figure 4. As shown in Figure 2(b), the graphite filler of 40/150/200 μm exhibited a slightly higher strength of 30 MPa compared to the injection moulding process with 29 MPa. Interestingly, this finding contradicted with other graphite filler sizes combinations, as well as the findings in previous studies. This was also supported as shown in Figure 4(f), where during the injection moulding process, the conductive composite experienced the occurrence of voids, consequently deteriorating its flexural strength. Meanwhile Figure 3(f) also shows evidence of smaller void formation compared to Figure 4(f), resulting in higher flexural strength of 1 MPa.

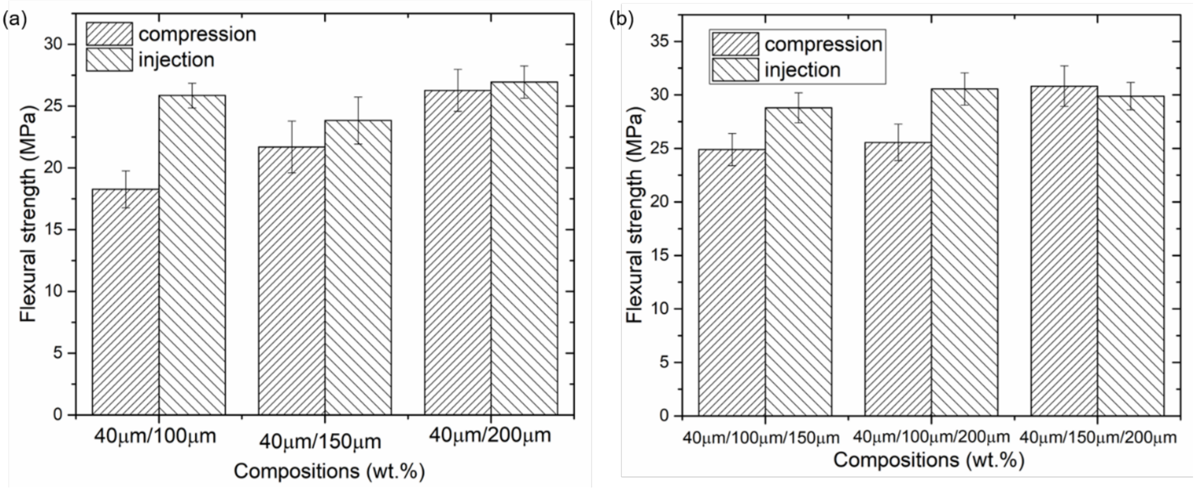

FIGURE 2. Flexural strength of (a) two sizes (40/100 μm, 40/150 μm, 40/200 μm), and (b) three sizes (40/100/150 μm, 40/100/200 μm, 40/150/200 μm) of graphite filler developed by injection moulding and compression moulding techniques

MICROGRAPH ANALYSIS

The micrograph analysis *via* SEM analysis is shown in Figures 3 and 4 for both the compression and injection moulding fabrication technique, respectively. Figure 3(a) - 3(c) depicts the two sizes of graphite filler in which Figure 3(c) is the most compact with a smooth cross-section structure as compared to Figure 3(a) and Figure 3(b) given the existence of voids and an uneven, through the nonhomogeneous surface. Accordingly, this proves that the flake-like graphite filler geometry shape was able to ensure excellent conductivity network formation and thus, its electrical conductivity was able to acquire 15 Scm^{-1} compared to the other two filler sizes. Moreover, the low electrical conductivity between 5 and 10 Scm^{-1} of the injected two sizes of graphite composites was due to the occurrence of voids, as shown in Figure 4(a) - 4(c). The formation of voids within the conductive composite will eventually drop the electrical conductivity given the graphite filler will be unable to form a conductive pathway (Li et al. 2015; Zare & Rhee 2019). On the other hand, as shown in Figure 4(d) - 4(f), the addition of the three sizes of graphite filler induced an extreme void formation even though the graphite was in a homogenous structure. These findings was also discussed in previous studies as the compactness of the composite matrix was reduced, causing the huge void, as illustrated in Figure 5(b) (Antunes et al. 2010). However, from the flexural analysis performed, the studies showed that the formation of homogenous graphite has aided in well distributing the load applied during the testing. It was further resulting in greater flexural strength (~30 MPa) when compared to compression moulded graphite composites. These studies, therefore suggesting that this occurrence was due to the high mechanical strength possessed by the homogenous graphite fillers, thus leading to an increase in flexural strength (Zakaria et al. 2015).

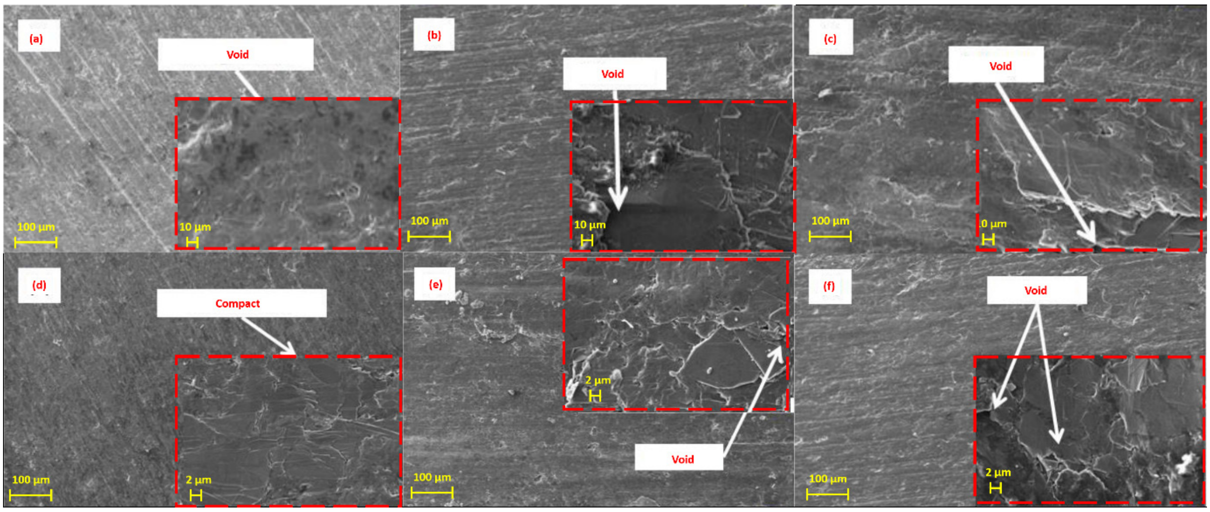

FIGURE 3. Micrograph images of (a) two sizes (40/100 µm, 40/150 µm, 40/200 µm), and (b) three sizes (40/100/150 µm, 40/100/200 µm, 40/150/200 µm) of graphite filler developed by the compression moulding technique

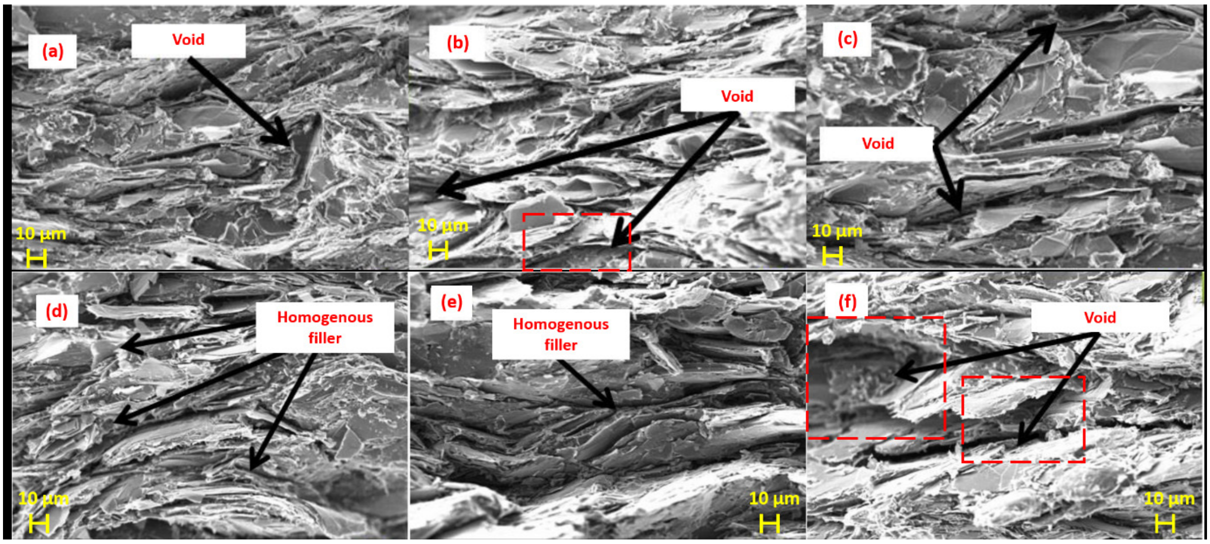

FIGURE 4. Micrograph images of (a) two sizes (40/100 µm, 40/150 µm, 40/200 µm) and (b) three sizes (40/100/150 µm, 40/100/200 µm, 40/150/200 µm) of graphite filler developed by the injection moulding technique

PARTICLE SIZE EFFECT

Prior studies have widely discussed that the distribution of particle sizes of conductive fillers reflected the percolation threshold since it affected the conductivity pathways (Heo et al. 2006). Hence, it is important to identify the suitable combinations of graphite fillers, as too many (three sizes) of different geometrical fillers will cause a serious porosity which will ultimately lead to the lack of electron tunnelling (Fulmali et al. 2020; Zare & Rhee 2020). As such, the overall electrical performance will deteriorate (Heo et al. 2006; Hui et al. 2009). Figure 5(a) displays the distribution of particle sizes of both graphite composites. The results demonstrate that the two sizes of graphite filler, 40/200 µm, encounter sharp filler distribution compared to 40/100/150 µm with larger particle size distribution ranges. Based on the image, as shown in Figure 5(b), it demonstrates that the three sizes of filler easily experience voids or larger particle size distribution, as seen in Figure 5(a). This phenomenon will eventually affect the electrical performance and the conductivity pathway developed especially for the thin composite structure. Prior studies have proposed that excess filler will not significantly reduce the resistivity that occurs, but rather disturbs the conductivity pathway formed (Balogun & Buchanan 2010). This is supported by the findings on the graphite fillers in which the addition of two instead of three graphite, sizes, was able to enhance the chemical composition differences since it related to the percolation concentration of the graphite fillers (Balogun & Buchanan 2010; Heo et al. 2006).

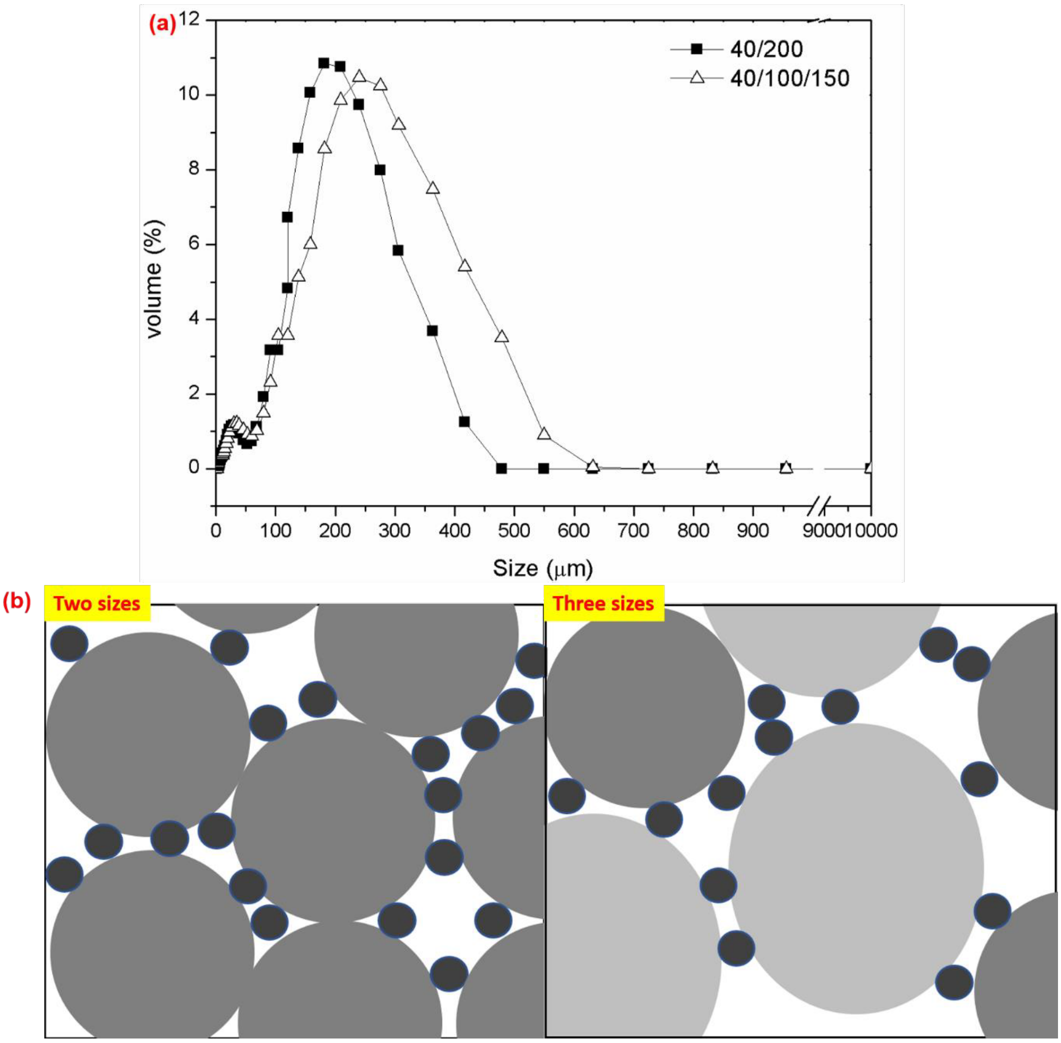

FIGURE 5. (a) Graphite composites particle size distribution with the composition of 40/100 µm and 40/100/150 µm, and (b) illustration of graphite composite

CONCLUSION

By adopting a conventional compression moulding process, a conductive composite consisting of two sizes of graphite filler was fabricated with a thickness of 2.5 mm. Indeed, this is recognized as the latest evolution in bipolar plate industry. Graphite composite of 40/200 µm filler sizes was able to withstand the flexural strength of 26 MPa. Accordingly, succeeding in fabricating conductive composite in a thin layer (2.5 mm) compared to previous research and in other industries that struggle in fabricating such a thin layer of composite. The findings of this research also indicated that by using two sizes of similar filler, both were on par and eventually acquiring better performance regarding electrical conductivity compared to three filler sizes due to the lack of network formation. Therefore, these findings can assist the needs of various industries regarding the fabrication of thinner conductive composite (<3 mm), having the flexural strength of 25 MPa. Given these findings, it is proposed that improvements be made using different types of filler to improve the electrical conductivity and hence maintaining its mechanical properties.

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