

Evaluation of the Possibility for Using Polypropylene/Graphene Composite as Bipolar Plate Material Instead of Polypropylene/Graphite Composite

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Received: 4 January 2016; Accepted: 10 February 2016; Published online: 23 March 2016

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Abstract

To reduce the bipolar plate weight and keep the desired power density of a low temperature fuel cell, thermoplastic composite bipolar plates have been fabricated for low temperature fuel cell applications. Concerns over material weight, electrical conductivity, mechanical properties, and injection processability of the thermoplastic nanocomposites have brought forth the idea of producing a polypropylene/graphene nanocomposite for an injection moulded bipolar plate application. In this article, the properties of produced polypropylene/graphene nanocomposites were also compared with the properties of polypropylene/graphite composites to assess the feasibility for using polypropylene/graphene nanocomposites as a bipolar plate material. The effects of graphene contents and sizes on electrical conductivity and mechanical properties were reported in this work. Moreover, the obtained results were discussed in terms of morphology and state of dispersion and distribution of the graphene within the polypropylene matrix. This work is a preliminary study that offers insight into the material selection and development for low temperature fuel cells toward the ultimate goal of broad commercialization.

Keywords: Graphene nanocomposites, Bipolar plates, Polypropylene/graphene composite, Electrical conductivity, Low temperature fuel cells, PEMFC, DMFC

Please cite this article as: K. Onyu, R. Yeetsorn, M. Fowler, A. Yu, Y. S. Jun, C. Prapainainar, and W. Prissanaroon-Ouajai, "Evaluation of the possibility for using polypropylene/graphene composite as bipolar plate material instead of polypropylene/graphite composite," *KMUTNB Int J Appl Sci Technol*, vol. 9, no. 2, pp. 99–111, Apr.–June 2016.

1 Introduction

Alternative energy technologies are presently receiving substantial attention in Asia owing to their prospect to replace polluting forms of energy. The technologies also include a fuel cell technology. Nevertheless producing fuel cells to become as widely accessible as the conventional energy generators is proving difficult because there are several challenges that still require to be overcome such as hydrogen storage and material issues i.e. durability, reliability, manufacturing, and costs [1]. The portable fuel cell market has been one of the green technologies that innovators have been paid attention. Low temperature fuel cells, which are a polymer electrolyte membrane fuel cell (PEMFC) and a direct methanol fuel cell (DMFC), could be used to provide power for portable applications such as electric bicycles (Gitane), bike lights (Angstrom Power), phone charger (Brunton), batteries for electronic devices (Canon), alcohol breathalyzer (BACtrack), and etc. DMFC emits low quantity of CO_2 , while PEMFC uses direct hydrogen, with no point-of-use emissions [2].

In addition to performance of low temperature fuel cells, weight of the fuel cell must be suitable for a practical use. The technology of low temperature fuel cells is dependent on the performance of bipolar plates. The bipolar plates in a fuel cell stack require important development, because the bipolar plates account for approximately 70% of the fuel cell weight and 80% of the fuel cell volume [3]. According to the statistics, the weight of fuel cell stacks principally depends upon weight of bipolar plates. Polymer composite bipolar plates have therefore attracted the researchers due to their weight and easy fabrication [4]. Various types of bipolar plate materials have been typically explored with the overall target to accomplish the listed stringent desired properties. Using the polymeric composites results in low machining cost, brilliant corrosion resistance, and easy fabrication process for gas flow channels (via *in-situ* molding process) [4], [5]. For a proper fuel cell operation, multiple fuel cells are stacked together to give a satisfactorily high power and voltage. The method to assemble all fuel cell components together is the application of the external mechanical-force or clamping force on the components [6]. The local areas which are the interface between the lands and channels of the flow field on bipolar plates often have defects, since the areas are subjected to excessive

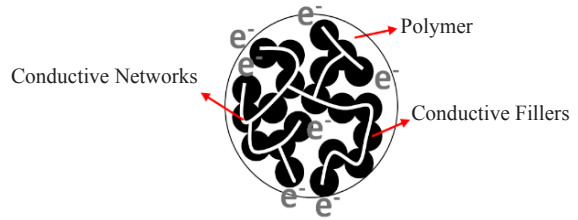


Figure 1: Schematic of the electrically conductive network in a polymer composite.

or non-uniform mechanical stresses. The stress causes mechanical degradation of membrane electrode assembly (MEA), and gas diffusion layer (GDL) [7]. Polymer composite bipolar plates are a flexible material that can have more force transfer, and then the mechanical degradation can be decreased. The weakest point of the polymer composite bipolar plates is their low electrical conductivity compared to a conventional graphite or metallic bipolar plate. To increase the electrical conductivity value of the plates, extrinsic conductive polymers have been used as bipolar plate materials [8]. An extrinsic conductive polymer is the combination of the conventional polymers with conductive loads of fillers such as carbon black or carbon fibers, graphite, metallic fibers, metallic powders [9], [10]. The electrically conductive fillers allow the creation of new polymeric composite materials with unique electrical properties. The conduction mechanism of an extrinsic conductive polymer is described by the percolation theory [9]. In Figure 1, the composite acts as an insulator if the conductive filler concentration is lower than a specific value, which is the percolation threshold. Upper this critical loading, a conductive cluster is formed and the composite becomes electrically conductive.

One of the pathways to tailor the electrically conductive materials is to incorporate carbon based conductive fillers into a polymer matrix. The fillers typically are carbon black (CB), multi walled and single walled carbon nanotubes (MWCNT and SWCNT), carbon fibers (CF), expanded graphite and their combinations [11], [12]. Carbon nanotubes with high aspect ratio exhibit either semiconducting or metallic behavior, so they have induced a very large scientific interest for the electrically conductive composites. Carbon nanotubes can ease the formation

of the electrically conductive network, thus decreasing percolation threshold more than other fillers [13]. Academic and industrial researchers are recently interested in a graphene (GnP) filler. Graphene has been utilized for fuel cells in three main aspects; as the fuel cell electrodes, as an additive to a polymer electrolyte membrane, and as a filler to bipolar plates [14]. Graphene considerably enhanced the fuel cell performance, resulting in better durability and increased lifetime of fuel cells. Graphene is a flake carbon-nanofiller (2-dimensional carbon nanofiller). Its nanostructure contains carbon atoms which are bonded in hybridized sp^2 [9]. That means a carbon atom in graphene is surrounded by three neighbour atoms. Electrons in sp^2 -bonded carbon atoms can be nearly free delocalize, and the delocalized electrons take place in an orbital perpendicular to the carbon plan [15], [16]. In other words, the graphene structure forms flat sheets with π -type electrons above the sheet. In terms of a lattice type, the carbon atoms in this nanostructure are self-assembled in a honeycomb 2D lattice [15]. The particular structure of graphene confers to good mobility of charge carriers (10 to $100\text{ cm}^2\text{V}^{-1}\text{s}^{-1}$), high electrical conductivity ($2 \times 10^5\text{ S/m}$), low resistivity ($10^{-6}\ \Omega\cdot\text{cm}$), and high thermal conductivity ($5000\text{ Wm}^{-1}\text{K}^{-1}$) [15]. The composite materials of the bipolar plate must have specific properties because of its multiple responsibilities and the challenging environment in which the fuel cell operates. The ideal material for bipolar plates should combine proper electrical and thermal conductivity and also good mechanical properties according to target values that are referenced from the Department of Energy (DOE) [5]. Graphene polymer nano-composites can be prepared by simple approach, such as solution mixing, melt blending, and in-situ polymerization [15]. If the polymer can be dissolved in simple polar solvents, the solution mixing will be an interesting strategy to prepare polymer/GnP composites [17], [18]. The composites produced from the solution mixing will contain the uniform dispersion of GnP in a polymer matrix. Poly(vinyl alcohol), poly(methyl methacrylate), epoxy resin, poly(acrylic acid), phenol formaldehyde resins and polyurethane are typically used for the solution mixing [19]–[21]. To create polymer composite bipolar plates which can be manufactured by an inexpensive process, polypropylene (PP) or polyethylene is widely used as a polymer matrix [10]. The solution mixing technique becomes difficult when

using commodity plastics such as polypropylene or polyethylene. polypropylene or polyethylene cannot dissolve in organic solvent at room temperature, such that they are soluble in xylene and trichlorobenzene at $> 120^\circ\text{C}$ [17]. Melt mixing technique is, therefore, more suitable for the commodity plastics than the solution mixing technique. Altered bipolar plate production techniques as high performance machining and plastic processing (compression molding, injection molding, and thermoforming) have been used to fabricate the required shape. Compression molding which has been proposed molds mixtures containing conductive fillers with polymer binders. Thermosetting composites are compression molded and cured at low temperature [22]. The plates are capable to be produced speedily and economically. Further studies of the bipolar plate fabrication are required to speed up the process of fuel cell commercialization [23]. An injection molding process is cost competitive even for comparatively small series in the range of numerous thousand plates [5], [24]. PP was selected to be polymer matrix for this research work because it has the combination of the reasonably low cost, good processability for an injection molding process, well balanced physical and mechanical properties. Consequently, PP has been a polymer base for fabricating composite bipolar plates in the past few years. Incorporating graphene 1 wt% with PP composites (fillers: natural graphite 64 wt%, carbon black 5 wt%, and carbon fibers 5 wt%) can improve electrical conductivity (through plane: from 99.70 to 130.17 Scm^{-1}), mechanical strength (flexural strength: from 54.23 up to 57.28 MPa) [25]. It is also corrosion current mitigation. Those advantages are reasons for the use of graphenes (graphene sheet and graphene oxide) as a conductive filler of bipolar plates, even the high production costs has limited their use [26]. A novel approach to scale down the weight of bipolar plates is another intention of the graphene application. Many layers of graphene stacked together with 1 m^2 sheet have around 0.77 mg of weight [2]. The addition of graphene sheet into polymer such as PP or polyphenylene sulphide has dramatically positive effect on the rheological properties of the nanocomposites [15], [26]. At low frequency range, liquid-like behavior of composite turns to be solid-like behavior for loading which is higher than 0.2 – $0.4\text{ vol}\%$ of percolation threshold [15]. To enhance the electrical conductivity of polymer composites, a synergistic effect between graphene and

carbon fillers has been focused. The synergistic effect has offered a useful comprehension into the production of polymeric bipolar plates with tailored properties to meet the stringent requirements. Researchers studied the synergetic effect of two fillers together by processing double-fillers; carbon fiber/graphene [25] and carbon black/graphene [27], and triple-filler; carbon fiber/carbon black/graphene [25]. This article presents a melt-mixing technique for obtaining PP/graphene nanocomposites that offers the feasibility of mass production, though using classical processing methods such as extrusion. Experimental work focused on shaping composite plates via injection molding, since injection molding is suitable for producing large scale quantities. Nevertheless, the process is still difficult when a high concentration of a filler is used. The high conductive filler content increases the viscosity of the blends.

To address the problem this study was directed at applying graphene with low loading as an electrically conductive filler. This is due to graphene can produce a dramatic improvement in properties at very low filler content. In addition to electrical conductivity and mechanical property, obtained nanocomposites are reported and discussed in terms of morphology and the state of dispersion and distribution of the graphene as assessed by microscopic techniques.

2 Experimental Methodologies

2.1 Materials

The main materials used in this study were injection-grade PP (DOW 560.01) purchased from Dow Chemical Co. and natural graphite flake (10 mesh of agglomerate size, 99.9%) purchased from Alfa Aesar. Two grades of graphene supplied from XG Sciences were as follows: GnP-M-5 grade (99.5% carbon) with 5 microns of average diameter and <10 nm of average thickness, GnP-H-100 grade (> 99.5% carbon) with 200 microns of average diameter and <15 nm of average thickness.

2.2 PP grinding process

To investigate the effect of PP sizes on mixing ability and final properties of composite plates, PP pellets were ground. Prior to the filling PP pellets into a

grinder, they were immersed in liquid nitrogen to make easier grinding. The different particle sizes of ground PP powder were separated via a woven screen sieve (mesh number 18 and 35). Selected PP particle sizes for this work are shown in Table 1.

Table 1: Different sizes of PP particles

Samples	Particle size (μm)
PP(A)	<500
PP(B)	500–1,000
PP(C)	pellet

2.3 Preparation of PP/natural graphite and PP/graphene nanocomposites

PP and natural graphite (G) or graphene (GnP) were premixed using a solution blending technique, and ethanol was used as a solvent for the process. Initially, PP and G or GnP was dispersed in ethanol using a homogenizer (mechanical stirring) to obtain G or GnP slurry. After the solution blending step, the ethanol was removed from the PP mixture by purging with nitrogen gas. Before mixing, PP mixture was kept in an oven at 60°C overnight. PP/G and PP/GnP nanocomposites with 5.0 and 7.5 wt% fillers were prepared by a melt-mixing technique. PP and the electrically conductive fillers were mechanically mixed in counter-rotating twin screw extruder (SJ2S-07, 11.00 millimeters of screw diameter, Wuhan Ruiming Machinery, China). Note that 5.0 wt% and 7.5 wt% of fillers concentration used for composites were formulated based on a percolation threshold [28]. Specific operating conditions of composite mixing are 195°C of molten temperature, screw speed at 18 round per minute, 5.00 minutes time for mixing circulation.

2.4 Nanocomposite plate molding

PP nanocomposite plaques for ex-situ testing were injected via mini injection molding (SZS-20, Wuhan Ruiming Machinery, China). The molten composites from the extrusion were transfer to the mini injection by specific hot holder. The injecting condition consisted of 230°C of injecting temperature, 60°C of mold temperature, 0.7 MPa of injection pressure, and 0.3 MPa of mold compression pressure.

2.5 Electrical conductivity measurement

Surface electrical conductivity and volume electrical conductivity values were evaluated according to ASTM D257-07 using electrometers (Keithley 6517 and Keithley 8009 resistivity test fixture). The specimen size was 8.00 cm × 1.00 cm × 0.40 cm. Three specimens of each composition were used for the measurements.

2.6 Morphological investigation

The morphology of the fracture surfaces was observed with a field emission scanning electron microscopy (SEM: Zeiss Leo 1550). The broken surface cryofractured samples obtained after compounding and after injection molding were used for SEM analysis. Small pieces of each sample were freeze fractured using liquid nitrogen in the cross-sectional direction, and affixed to an aluminium stub using double-sided conductive tape. Then, the samples were sputter coated with gold at a thickness of approximately 10 nm prior to loading into the SEM machine.

2.7 Mechanical property test

Mechanical properties including flexural, and hardness tests were performed as following details. Flexural test was carried out in accordance with ASTM D790 through three-point bending technique using Universal Testing (Tensile Hounsfield HSK-S, 0006). Tests were conditioned at room temperature, and 1 mm/min of compression speed. Specimen size used for all tests was 8.00 cm × 1.00 cm × 0.40 cm, and five specimens were tested for each sample.

Surface hardness or shore hardness of PP nanocomposite plaques measured by Super EX durometers (shore D from e-Asker). According to ASTM D2240, the durometers allow for a measurement of the initial hardness, or the indentation hardness after a given period of time. Five specimens were tested for each sample.

3 Results and Discussion

3.1 A comparison between PP/GnP nano-composites and PP/G composites

Figure 2 illustrates the changes of surface electrical conductivity (in-plane conductivity) and volume

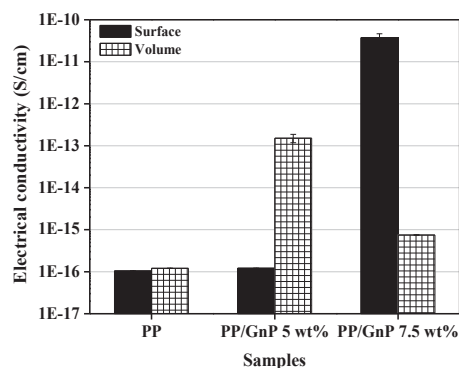


Figure 2: Electrical conductivity of neat PP and PP/GnP (GnP-M-5) nanocomposites.

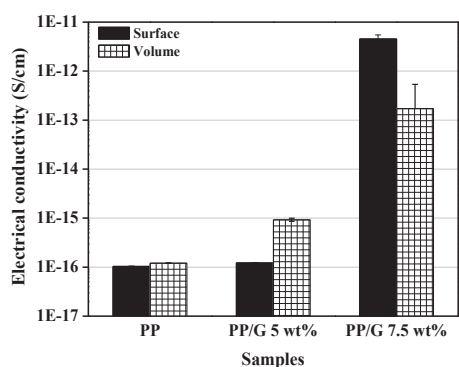


Figure 3: Electrical conductivity of neat PP and PP/G composites.

electrical conductivity (through-plane conductivity) of PP/GnP nanocomposites compared with those conductivity values of PP/G composites in Figure 3. Please note that GnP used in this experimental part was GnP-M-5. As known, it is important to find a composite formulation that offers a good combination in terms of conductivity and processability. This work is the preliminary work that was designed to use 5 and 7.5 wt% of filler content based on a percolation threshold [28]. Figure 2 and 3 show the changes of the electrical conductivity of PP/G and PP/GnP nanocomposite plates as a function of the graphene or graphite content. Electrical conductivity of neat PP was measured to be 10^{-16} S/cm, indicating that the PP plates were electrically insulating. At 5 wt% of conductive filler loading, the volume conductivity increased more than three orders of magnitude for the PP/GnP composite, but it rose just 10 times in case of the PP/G

composite. It is reasonable to describe that uniform dispersion of the nanofiller constructs an efficient electrically conductive pathway, and the path helps to obtain easier electron mobility throughout the insulating polymer and thus enhance electrical conductivity. Graphene delivers a high electrical conductivity, high aspect ratio (in the range of 3,200 to 5,200 [26]), and high surface area, hence graphene could homogeneously disperse in PP with a three-dimension random orientation. It implies that created three-dimension electrically conductive path of graphene throughout the PP matrix results in a higher volume electrical conductivity enhancement than that of graphite. The volume electrical conductivity was diminished, when PP/GnP composites containing 7.5 wt% of graphene content. The polarity difference between PP and GnP leads to the agglomeration of graphene sheets. The PP/GnP composites containing 7.5 wt% of graphene content cannot attain uniform dispersion because of the agglomeration of graphene sheets. When graphene particles align themselves perpendicularly to the composite plate, they create less conductive networks. On the contrary, a growth in the volume conductivity of PP/G composites can be seen at the same loading of the filler.

In terms of surface conductivity, both composites exhibited an electrically static dissipative surface or low conductivity surface. The probable reason is that the fillers tend to align parallel to the surface of the composite plate during compression molding but the connection between fillers particle were not enough to construct a conductive pathway at low graphene or graphite content. If filler concentration is higher (7.5 wt%), nanofiller particles incline to align themselves along the flow direction of the plate during lateral material flow in the injection molding process. The result demonstrates that the number of physical contacts between graphene or graphite particles increases with the increment of the nanofillers loading. Thus more electric charges can flow through the nanocomposite plate. The introduction of graphene at 7.5 wt% to PP increased the surface conductivity of the resulting composites by more than 5 orders of magnitude (from 1.21×10^{-16} to 3.76×10^{-11} S/cm.), while adding graphite into PP increased the surface conductivity around 4 orders (from 1.21×10^{-16} to 4.54×10^{-12} S/cm.). It is a surprise that the surface or in-plane electrical conductivity was not dropped as the volume conductivity even the nanofiller concentration was increased.

The orientation on the skin may be another reason that surface electrical conductivity is higher than volume electrical conductivity. This also contribute a higher in-plane electrical conductivity in parallel to the injection flow direction comparing to the value measured perpendicular to the flow direction.

The generated heat from a low temperature fuel cell has to be dissipated from the fuel cell stack through the bipolar plate to a cooling channel in order to maintain constant operating temperature in the fuel cell system. Therefore, consideration of the thermal conductivity of bipolar plates also requires for management of the heat in fuel cell stacks. Song *et al.* reported that PP/GnP composite with 2.1 vol% (5 wt%) of GnP loading gave a thermal conductivity of 0.396 W/mK, while PP/G composite with 3.0 vol% of exfoliated graphite loading provided 0.4 W/m K [17]. The reported values have not achieved the requirement of DOE (20 W/m K) [29]. Higher loading of GnP and G than 5 wt% is required in order to accomplished DOE requirement. Furthermore, the improvement of filler dispersion and orientation are necessary for thermal conductivity enhancement [30].

The electrical conductivity results were further supported by SEM micrographs. Figure 4 and 5 show the SEM micrographs of the fracture surfaces of PP/G and PP/GnP composites. The morphological characteristics of graphite-filled or graphene-filled PP composites at various nanofiller loading were observed. It can be seen in the micrographs that the existence of small flaky graphite particles dispersed in the PP matrix between the larger graphite particles.

The dispersion of graphite particles allows electrons transfer throughout the PP matrix. In case of graphene composites, graphene particles are not seen clearly, but rough surfaces illustrate graphene dispersion in the PP matrix. As seen in the figures, the graphene particles were well dispersed in the PP matrix. That implies the melt-mixing process provides good flow-ability and wetting of the graphene nanofiller. The figures also presented the graphene particle which were encapsulated by the matrix, indicating considerable interfacial interaction between the graphene particles and PP. The result was confirmed by no separation of graphene fillers from the PP on fracture surface as that can be found in the micrographs of graphite composites.

Bipolar plates have to provide mechanical strength and rigidity to support the thin membrane and

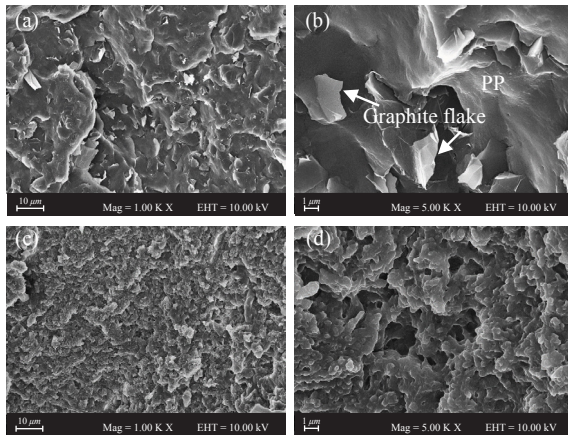


Figure 4: SEM micrographs of PP composites: (a) PP/G composite with 5 wt% of G content at 1.00 KX of magnification (b) PP/G composite with 5 wt% of G content at 5.00 KX of magnification (c) PP/GnP-M-5 composite with 5 wt% of GnP-M-5 content at 1.00 KX of magnification (d) PP/GnP-M-5 composite with 5 wt% of GnP-M-5 content at 5.00 KX of magnification.

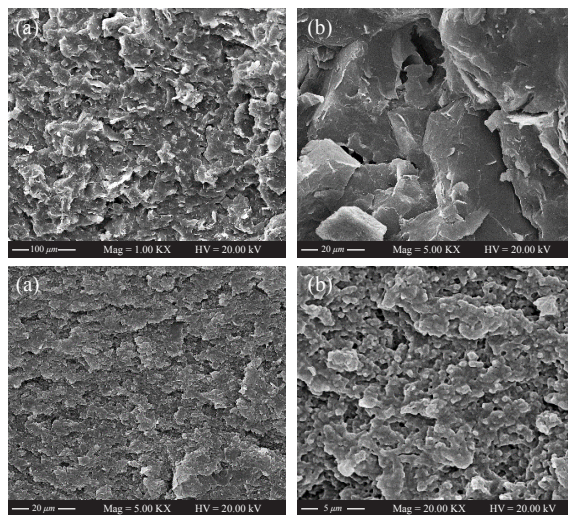


Figure 5: SEM micrographs of PP composites: (a) PP/G composite with 7.5 wt% of G content at 1.00 KX of magnification (b) PP/G composite with 7.5 wt% of G content at 5.00 KX of magnification (c) PP/GnP-M-5 composite with 7.5 wt% of GnP-M-5 content at 5.00 KX of magnification (d) PP/GnP-M-5 composite with 7.5 wt% of GnP-M-5 content at 20.00 KX of magnification.

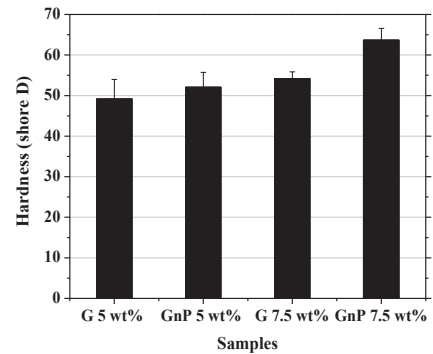


Figure 6: Shore D hardness of PP nanocomposite plates.

electrodes and clamping forces for the stack assembly. Their mechanical properties should be considered for fuel cell applications. The surface hardness of composite plate was measured by scleroscope shore hardness, type D. The surface hardness of the composite plates increases with increasing graphite or graphene content as shown in Figure 6.

The hardness value increases from 52.1 to 63.7 as the graphene (GnP-M-5) content increases from 5 to 7.5 wt%. The rigid graphite or graphene particles inserted among the PP chains along the composite plate's surface, resulting in hardness reinforcement. Hardness values of PP/GnP composites are higher than the values of PP/G composites (from 49.2 to 54.2). Graphene particles had better dispersion in PP than the dispersion of graphite particles, and then PP/GnP provided higher hardness reinforcement than the reinforcement of PP/G. The increase in hardness may not be significant when the filler content is further increased. The hardness value is the surface phenomena, and then the high filler concentration in polymer bulk does not contribute to the material hardness. Bipolar plate hardness requirement from DOE should be >50. The example of hardness values for existing commercial bipolar plates are 60 for Schunk 138 bipolar plates and 52 for the bipolar plate of fuel cell store [31]. The results reveal that PP/GnP and PP/G composites are sufficiently hard for bipolar plate application. A fuel cell stack in a portable device has to withstand the vibration occurred from the device mobility. As displayed in Table 2, flexural strength and flexural modulus of PP/GnP and PP/G composites were systematically observed. The flexural strength values of PP/GnP nanocomposites at 5 and 7.5 wt% of graphene

loading were found to be slightly higher than that of the neat PP. However, no significant change was observed for the comparison of flexural performance of PP/GnP nanocomposites at 5 and 7.5 wt% of graphene loading. 7.5 wt% of graphene loading may cause the stacking together of graphene sheets owing to the strong van der Waals force between the graphene sheets. That results in low effective reinforcement. In terms of the PP/GnP nanocomposite at 7.5 wt% of graphene concentration, the flexural strength of the composite increased and was illustrated to be 29.97 MPa comparing with the value of 27.45 MPa of the PP/G composite at 7.5 wt% of graphite loading. The values of flexural modulus for PP/GnP composites are distinctly increased when compared to the values of the neat PP. Nonetheless, PP/G composites with 7.5 wt% of graphite loading did not give the improvement of flexural strength due to the loading has not achieved mechanical percolation threshold. Among all the composite samples, PP/G composites with 5 wt% of graphite loading showed the lowest value of flexural modulus.

Table 2: Flexural strength and flexural modulus of PP, PP/G and PP/GnP at 5 and 7.5 wt% of loading

Samples	Flexural Strength (MPa)	Flexural Modulus (MPa)
PP	27.29	647.24
PP/G 5 wt%	20.73	564.30
PP/G 7.5 wt%	27.45	799.31
PP/GnP 5 wt%	29.76	922.35
PP/GnP 7.5 wt%	29.97	954.16

The resulting stress-strain curves were presented in Figure 7. The flexural modulus of the PP/GnP and PP/G composites were observed to be scientifically escalated with increasing graphene or graphite contents, following an additive rule. However, a clear decrease in the values of flexural modulus can be observed from Figure 7 for the PP/G composites with 5 wt% of graphite loading. The enhancement in flexural performance of PP/GnP composites relative to PP/G composites was believed to be due to a uniform dispersion and strong interfacial bonding between the nanofiller and the PP matrix [32], [33] as can be seen in Figure 4 and 5. All results exhibited the dependencies of the graphene and graphite (flake or platelet fillers) on reinforcing effect

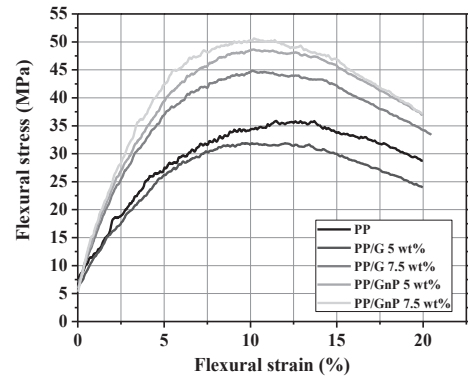


Figure 7: Flexural stress-strain curve of neat PP, PP/G and PP/GnP composites.

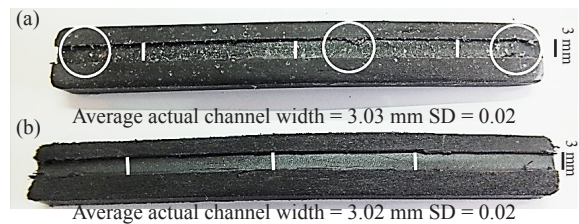


Figure 8: (a) PP/G and (b) PP/GnP composite specimens with a gas flow channel made by CNC milling machine.

from the filler volume fraction. E.V. Kuvardina et.al. reported that platelet-particle fillers especially graphene showed the highest reinforcement, while spherical-shape fillers displayed the lowest reinforced effectiveness [28]. Furthermore the mechanical performance of flake graphite was lower than the flake graphene.

The bipolar plates perform a number of functions within low temperature fuel cells providing a flow path for gas transport and distribute the gases over the entire electrode area uniformly is one important function. Flow channels of bipolar plates should be therefore machined smoothly. Introducing solid fillers in to polymer matrix typically makes a polymeric material brittle, and the composite plates may be cracked during the machining. Figure 8 displays cutting surface of PP/GnP and PP/G composite plates. The figure shows that both composite plates can be used for creating gas flow channels by CNC milling machine without cracking defects from machining process. The machined flow channels on both composite samples indicated good accuracy. Figure 8 shows setting channel width is 3.00 mm and the average actual channel width of both samples is in the range of 3.02–3.03 mm.

3.2 Graphene particle size effects

It is well known that filler characters such as aspect ratio, diameter, and surface area suggest significant improvement in physical properties of composites. Therefore, GnP-M-5 and GnP-H-100 were used in experiments to study the influences of different diameters and thicknesses of graphene on electrical conductivity and mechanical performance. GnP-M-5 grade had 5 microns of average diameter and <10 nm of average thickness, whereas GnP-H-100 grade had 200 microns of average diameter and <15 nm of average thickness. Figure 9 illustrated that both of graphene types improved surface and volume electrical conductivity of the composites, comparing to the neat PP sample prepared by the same methods. Furthermore, the highest increase in electrical conductivity of graphene filled PP was evidently observed at the PP/GnP-M-5 composites containing 7.5 wt% of graphene. The introduction of 7.5 wt% GnP-M-5 to PP increased the surface electrical conductivity of the resulting composite by more than five orders of magnitude, while volume electrical conductivity of the composite increased more than one order of magnitude. GnP-M-5 is the smallest particle, has lowest thickness, and has highest surface area. Those characteristics would promote well dispersion and conductive path according to the percolation theory. However, the volume conductivity of composites containing both graphene types was much lower than the PP/G composite. The agglomeration of graphene sheets and poor dispersion of graphene in PP matrix affected on conductive network formation.

In terms of mechanical performance, hardness values of PP composites did not change in the presence of either type of graphene fillers. As opposed to the hardness, flexural performance was improved by combining graphene with PP matrix.

Figure 10 showed that the flexural reinforcing effect of GnP-H-100 with larger diameter was slightly higher than that of GnP-M-5 with larger diameter. This result disagrees with previous publication [28] which stated that graphene particle with lesser length gave better mechanical performance.

3.3 PP particle size effects

The operating condition for extrusion process significantly affects on final properties of composite

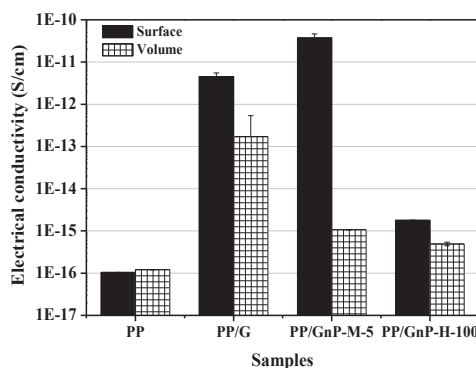


Figure 9: Electrical conductivity of neat PP, PP/G, PP/GnP-M-5 and PP/GnP-H-100 composites.

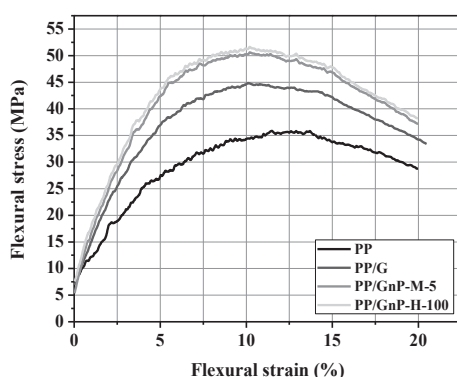


Figure 10: Flexural stress-strain of neat PP, PP/G composite, PP/GnP-M-5 and PP/GnP-H-100 nanocomposites.

products and the dispersion of nanofillers in matrix. Particle sizes of polymer and fillers are also factors that should be considered for the dispersion improvement. If particle sizes of polymer and fillers are quite different, distribution and dispersion of filler may be undesirable. Thus, the effects of particle sizes of PP on electrical conductivity and mechanical performance were investigated. There were three sizes of PP used in the experiments. PP(A) was used in a powder form with <500 μm of the particle size, while powdered PP(B) had particle sizes in the range of 500–1,000 micron. The last PP sample was PP(C) which was in the form of a commercial PP pellet. The results of the surface and volume electrical conductivity of the samples are shown in Figure 11 and 12. In general, PP composites which were melt-mixed from both PP powder delivered higher electrical conductivity than composites containing the PP pellet.

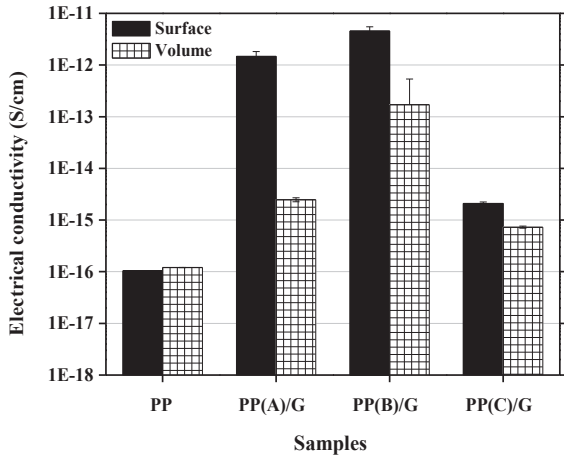


Figure 11: Electrical conductivity of neat PP and PP/G composites prepared by different PP particle sizes.

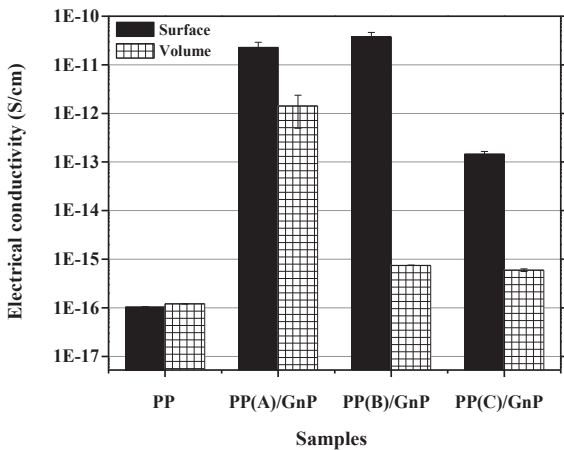


Figure 12: Electrical conductivity of neat PP and PP/GnP nanocomposites prepared by different PP particle sizes.

PP(B)/GnP composites provided the highest surface electrical conductivity which was 5.28×10^{-11} S/cm, but the highest volume electrical conductivity, which was 1.43×10^{-12} S/cm, belonged to PP(A)/GnP composites. Please note that all PP composites contained 7.5 wt% of filler concentration. PP with very small particles was easily melted causing filler particles to distribute well in PP matrix. Under optimum operating condition, the distributed filler particles were well dispersed in the matrix. The desired distribution and dispersion could support electrically conductive networks, therefore the electrical conductivity was pointedly enhanced.

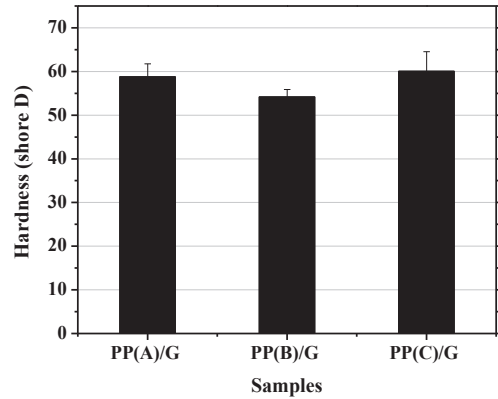


Figure 13: Shore D hardness of PP(A)/G, PP(B)/G and PP(C)/G composites.

Hardness performance of all nanocomposites prepared from different size of PP particles did not present any significant variation. The hardness values of PP(A)/GnP, PP(B)/GnP, and PP(C)/GnP nanocomposites were 64.4, 63.7, and 63.6 shore D, respectively.

In case of PP/G composites (Figure 13), the value of hardness for PP(B)/G composites was 54.2, being the lowest values. The hardness value of PP(C)/G was indistinctly increased when compared to the value of the PP(B)/G sample. There were considerable deviations from the average hardness values for testing the hardness in correlated to the inhomogeneity of the composite plates.

The results of flexural performance were shown in Figures 14 and 15. The flexural strength of the neat PP specimen was found to be about 27.29 MPa and its flexural modulus was about 647.24 MPa. A clear growth in the values of flexural strength and modulus can be observed from Figure 14 and 15 for all the types of composites.

Among all the samples, composites comprising of PP(C) showed the highest values of flexural strength. This means that using PP pellets as a matrix gives the flexural strength in a better way than using PP powder. Even though the stress-strain curves of PP/G composites present more different of flexural performance affected by PP particle sizes than the performance of PP/GnP nanocomposites, the flexural strength values were in the narrow range (27 to 31 MPa). With regard to PP/GnP composites (Figure 15), no enhancement in flexural modulus was found by varying particle sizes of PP.

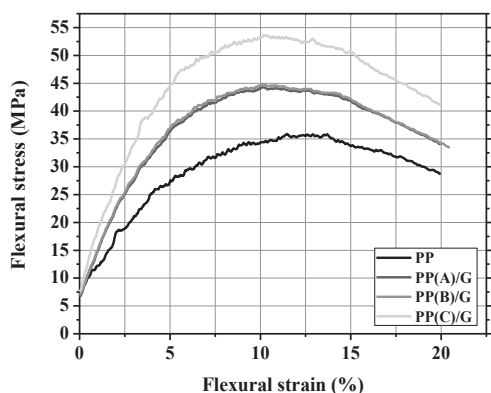


Figure 14: Flexural stress-strain of neat PP and PP/G composites with different sizes of PP particles.

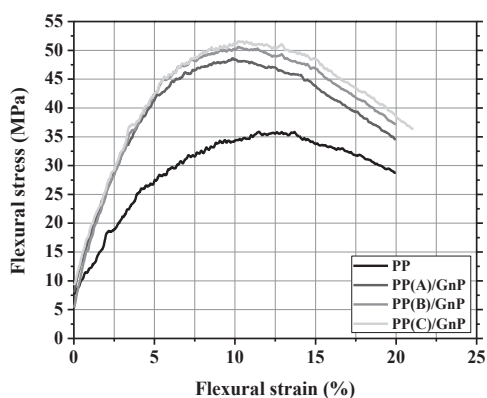


Figure 15: Flexural stress-strain of neat PP and PP/GnP nanocomposites with different sizes of PP particles.

4 Conclusions

In general, the experimental results demonstrated that the PP nanocomposites using graphene as an electrically conductive filler have enhanced electrical conductivity and mechanical performance. Those properties of PP/GnP composites for commercial application in bipolar plates, have not yet achieved. This may due to the graphene concentration should be higher or synergistic effect of mixed fillers should be concerned. Nevertheless this preliminary project can validate that graphene can be used instead of graphite. As known, graphite composite has been used for commercial bipolar plate applications. All results also show a promise for using low graphene loading to accomplish the same properties of using graphite. Some important conclusions can be drawn as follows:

- According to the electrical conductivity of the composites containing graphene, it can be said that their surface electrical conductivity values are one order of magnitude higher than graphite composites at the same filler concentration. A problem of graphene agglomeration when using 7.5 wt% of graphene loading should be concerned issue to improve the volume electrical conductivity.

- Regarding the type of composites, the flexural performance changes sequentially as follows: PP/GnP 7.5 wt% > PP/GnP 5 wt% > PP/G 7.5 wt% > PP > PP/G 5 wt% with maximum values of 29.97 MPa (Flexural strength) and 954.16 MPa (Flexural modulus) for the PP/GnP 7.5 wt% composite. The PP/GnP 7.5 wt% composite is also the hardest plate with 64 shore D.

- Graphene and PP particle size influences on distribution and dispersion of graphene in PP, and they are associated with electrical conductivity and mechanical performance of the composite products.

Acknowledgments

The authors gratefully acknowledge the financial support given for this research by National Science and Technology Development Agency (Sanction number: SCH-NR2012-213 and SCH-NR2012-214) and Department of Chemical Engineering, University of Waterloo.

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