

Conductive Polymer Composites Made from Polypropylene and Recycled Graphite Scrap

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ABSTRACT: *Electric Discharge Machining (EDM) process uses electrodes made from graphite that wear out over time and are turned into scrap. In this research, EDM electrode scraps were recycled and turned into graphite powder (rGP). This rGP was used as a conductive filler to produce conductive polymer composite (CPC) material by combining it with polypropylene (PP) resin via melt compounding and compression moulding processes. The percolation threshold of this composite material changed when 30 wt% of rGP was added, whereby the insulative material changed became antistatic. The composite was able to achieve surface resistivity as low as 10^3 ohm/sq. However, the addition of higher rGP content deteriorated the tensile properties of composite, whereby the tensile strength of composite significantly decreased as compared to neat PP. The results also showed that the tensile modulus of this composite became higher, and the material became more brittle as compared to neat PP. However, the PP/rGP composite with 50 wt% filler content reduced the tensile modulus due to plasticising effect caused by the agglomeration of rGP. The addition of high filler content on PP/rGP composite also caused an increase in processing torque. This was due to the restriction of rGP particles to the melt flow of molten PP. The morphological analysis found that the PP/rGP composites with higher amounts of filler content were highly agglomerated and formed conductivity*

paths within the PP matrix. The increase of rGP content highly improved the thermal stability of composite. The findings of this study show that the rGP has the potential to be used as a conductive filler for producing conductive composite material.

Keywords: recycled graphite, polypropylene, conductive polymer composite

1. INTRODUCTION

Polymers are good electrical insulators, but they can be tailor-made into conductive materials by combining a polymer with conductive filler. This material is called conductive polymer composites (CPC) as it contains conductive fillers dispersed in polymer matrix. The conductive characteristic of CPC is due to the presence of conductive fillers, and this results in CPC having a broad range of conductivity which can be controlled by the filler content and type of conductive filler.¹ There is a wide range of applications of CPC such as semiconducting material for dissipation of static electricity, electromagnetic interference shielding, electronic equipment, and floor heating elements.^{1,2} In the past, CPC materials were mainly produced by adding carbon-based conductive filler, such as carbon black, carbon nanotube, and graphite.^{3,4} However, studies related to the production of CPC material using recycled waste material are relatively rare. Therefore, this research focuses on utilising recycled graphite for producing CPC material.

Graphite is a type of crystalline carbon that is obtained from nature and synthetically prepared from inorganic material. Graphite is a good electrically and thermally conductive material.⁵ For EDM machines, the majority of industries use graphite electrode material. The graphite electrode of EDM machine wear out after certain cycles of usage and requires replacement. Then, the scrap of electrodes is disposed, and it becomes waste. In this context, this research was carried out to recycle graphite electrode scraps that are obtained from EDM machining industry and utilise them as conductive filler in producing CPC.

The present research is focused on the effects of filler content on processing torque, electrical, tensile, thermal, and morphological properties of CPC made from recycled graphite powder (rGP) and polypropylene (PP). Therefore, this research explores the potential of recycling graphite electrode scraps from EDM machines by turning them into conductive fillers to produce conductive composites.

2. EXPERIMENTAL

2.1 Raw Material

In this experiment, EDM graphite electrode scraps were collected from Pre-Tech Solution Sdn. Bhd. (Penang, Malaysia). PP-type co-polymer (grade: SM340) was chosen as the matrix to produce CPC. Therefore, the PP resin was purchased from Lotte Chemical Titan (M) Sdn. Bhd. (Johor, Malaysia).

2.2 Preparation of Conductive Filler

The collected graphite electrode scrap was recycled by breaking it into small pieces using a hammer. Then, the graphite pieces were ground into fine powder using a miniature powder grinder (Mill Powder Tech, Tainan, Taiwan). The rGP was sieved to obtain particles of homogenous size, which was then ready to be used in the experiment. The size of metal sieve used in this experiment was 200 mesh. The average particle size of obtained rGP was 130 microns as measured by Melvin Particle Analyser (Cambridge, United Kingdom).

2.3 Preparation of Conductive Composites

For compounding, rGP was mixed with PP resin in various filler contents, which were 10 wt%, 20 wt%, 30 wt%, 40 wt%, 45 wt%, and 50 wt%, using Brabender® Plastograph Internal Mixer (Duisburg, Germany). The compounding temperature was fixed at 180°C with a rotor speed of 50 rpm. For the compounding procedures, PP resin was first transferred into a mixing chamber and was left to melt for 2 min. Then, the rGP was introduced into the melted PP and was allowed to mix for 6 min until the torque stabilised. After that, the rotor was stopped, and the compound was removed from the mixing chamber. During compounding, the data of processing torque against time was recorded by the machine. The PP/rGP composite compound was further moulded into a sheet of 1 mm thickness using the Gotech (Taichung, Taiwan) compressing moulding machine (model GT-7014-H30C). The operating temperature was set at 180°C and compression pressure at 5 tonnes. For moulding sequence, the compound was first pre-heated in the mould for 6 min, and then, the compound was softened and compressed for 1 min. After 1 min, the mould was left to cool down for 10 min. Lastly, the composite sheet was removed from the mould and further cut into tensile specimens using a dumbbell cutter. The dimensions of tensile specimens were according to ASTM D638, type IV.

2.4 Testing and Analysis

The PP/rGP composites with different filler contents were tested for their tensile properties with the aid of Instron (Norwood, United States) universal testing machine (model 5569). The test was carried out by referring to the standard ASTM D638. The crosshead speed of the machine was set to 30 mm/min and load cell to 15 kN.

PROSTAT® surface resistivity apparatus (Bensenville, United States) was used to measure the surface resistivity of CPCs and the test was done following the standard ASTM D257. The test was conducted using two miniature point probes placed at five different locations on a 100 mm × 100 mm sheet to obtain surface resistivity. The average values of surface resistivity were recorded.

Thermal properties of the PP/rGP composites were analysed using Pyris Diamond Thermogravimetric Analyser (TGA) (Perkin Elmer, Waltham, United States). The specimen was cut into small sizes with specimen weight of 5 mg to 9 mg. Then, the specimen underwent thermal scan from temperatures of 30°C to 700°C with a heating rate of 10°C/min. The analysis was run under nitrogen atmosphere with a nitrogen gas flow rate of 20 ml/min.

Field Emission Scanning Electron Microscope (FESEM) (model FEI Quanta 400, Hillsboro, United States) was used to examine the fracture surface of tensile specimens. Prior to examination, all specimens were coated with an ultrathin layer of gold to prevent electrons from charging during the scanning. The electron acceleration was fixed at 20 kV.

3. RESULTS AND DISCUSSION

3.1 Compounding Behaviour

Figure 1 illustrates the processing torque-time curves of the PP/rGP composites with different filler contents. The results showed a drastic increment of torque in the first min, and then a gradual decrease. This was because the rotor required more torque in the initial stage to shear and melt the solid PP resin, thus, the processing torque increased. When the PP resin had melted due to continuous shearing at high temperature, the torque slowly decreased. When rGP powder was slowly added to the melted PP resin over a period of 2 min, the torque slowly increased. The increase in torque was due to the restriction of rGP particles towards the melt flow. After the compound was homogeneously mixed, the torque became stable, and the mixing process was completed. The first peak lowered as the amount of rGP was

increased. Then, the second torque peak occurred after 2 min, and it increased as the filler content increased. This phenomenon was caused by the reduction of PP resin that decreased the amount of torque required to shear the PP resin. The change of second peak was correlated with the amount of rGP filler. As the filler content was increased, the restriction from rGP particles also became higher. For this reason, the second observed torque became higher when more rGP was added to the melt. The compounding behaviour of PP/rGP composite was like other composites filled with different particle fillers.^{6,7} Table 1 shows that the processing torque of PP/rGP compound increased with the increase in filler content. As mentioned earlier, the presence of rGP particles restricted the melt flow of molten PP and increased the viscosity of compound. This finding agreed with the findings reported by Chun et al. They also agreed that the viscosity of composite compound was directly proportional to the increase of filler content.⁸

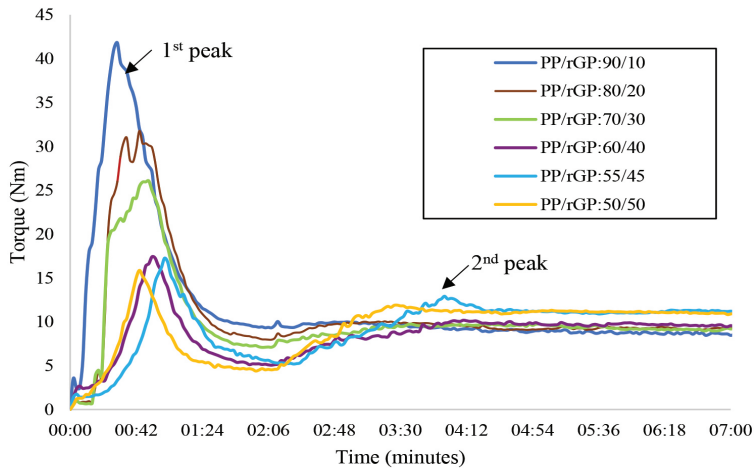


Figure 1: Processing torque verses mixing time of PP/rGP with various contents of filler.

Table 1: Stabilised torque of PP/rGP composites with various filler contents.

Sample	Stabilised torque (Nm)
Neat PP	6.8
PP/rGP:90/10	7.2
PP/rGP:80/20	8.4
PP/rGP:70/30	9.0
PP/rGP:60/40	9.5
PP/rGP:55/45	9.8
PP/rGP:50/50	10.5

3.2 Electrical Surface Resistivity

Figure 2 shows the electrical surface resistivity of PP/rGP composites with various filler contents. The electrical surface resistivity of PP/rGP composites, with filler contents ranging from 10 wt% to 20 wt%, fluctuated within an insulative range. This occurred because the amount of rGP was insufficient to form a conductive network. However, when the filler content was increased to 30 wt%, the surface resistivity of composite initially decreased, and it significantly reduced from 10^{12} ohm/sq to 10^{11} ohm/sq once the filler content reached 30 wt%. This indicated that the conductivity of composite changed from insulative to antistatic. The percolation threshold of PP/rGP composite was found at 30 wt%. When the amount of rGP was above 40 wt%, the surface resistivity of composite dramatically decreased from antistatic to static dissipative region. This indicated that when the filler content exceeded 40 wt%, the rGP particles formed a conductive network within the PP matrix. Therefore, the electrons could move freely around the conductors. The morphological analysis also confirmed that the formation of conductive network occurred in the composite when filler content reached 50 wt%. In the research by Gulrez et al. the conductivity of PP-based conductive composite showed similar percolation trends but the amount of conductive filler to cause percolation threshold was different.⁹ In this study, the amount of filler content that caused percolation threshold was higher because the particle size of the rGP prepared in the experiment was considered large as compared to the sizes used by other researchers. However, the findings proved that the graphite electrode scrap can be recycled and used as a conductive filler.

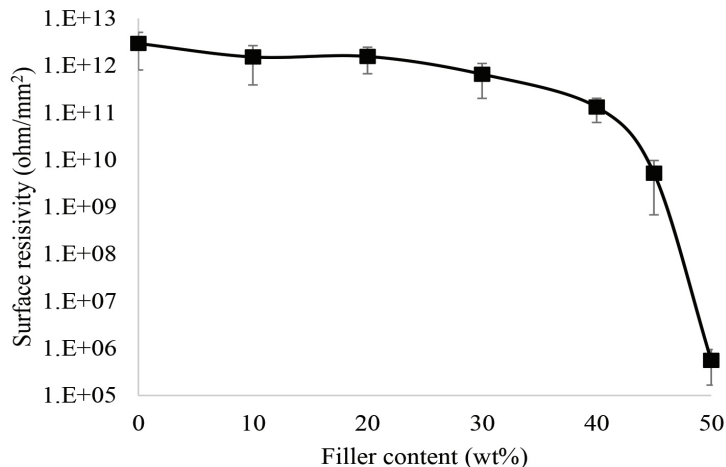


Figure 2: Trendline of electrical surface resistivity over various amounts of filler in PP/rGP composites.

3.3 Tensile Properties

The tensile strengths of PP/rGP composite with different filler contents are displayed in Figure 3. The tensile strength of PP/rGP composite decreased when the amount of filler was increased. As compared to neat PP, the tensile strength of composite decreased by approximately 60% when 50 wt% of rGP was added. The rGP was particulate and irregular in shape as observed from the scanning electron microscope (SEM) micrograph. Generally, this type of filler has a poor load-bearing capacity.⁷ Furthermore, the rGP was highly agglomerated at high filler content, whereby the agglomeration acted as a stress concentrator. The presence of stress concentrator and a low load-bearing capacity resulted in a lower composite strength. The presence of filler agglomeration was proven by the morphological study as well. The geometrical and stress concentrator factors of the filler were key factors that caused the decrement of strength in the conductive composite filling, as reported by other researchers.^{10,11} In contrast, the tensile modulus of PP/rGP composites increased when the filler content was increased from 10 wt% to 45 wt%. The addition of rGP particles highly restricted the chain mobility of PP matrix. Therefore, it was concluded that the addition of more rGP content increased the stiffness of composite. However, the tensile modulus of PP/rGP slightly increased when the filler content was increased to 50 wt%, as displayed in Figure 3.

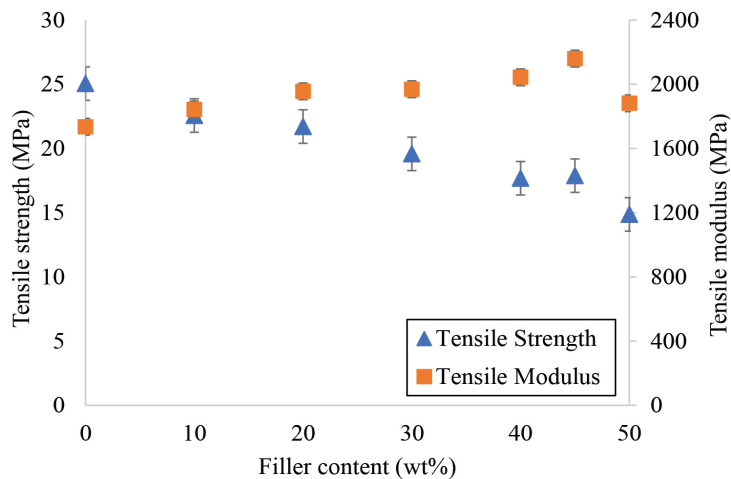


Figure 3: Tensile strength and tensile modulus of PP/rGP with various contents of filler.

Figure 3 illustrates that the tensile modulus of PP/rGP composites increased when the filler content increased from 10 wt% to 45 wt%. When the filler content reached 50 wt%, the tensile modulus of composite decreased. The rGP filler had stiff and rigid particles. It could be concluded that the addition of rGP generally increased

the stiffness of composites. Furthermore, the addition of rGP restrained the chain mobility of PP matrix due to the friction that occurred at interfacial region. As a result, the composite became more rigid and stiff. According to Bhagat and Verma the addition of more graphite flakes increased the tensile modulus of composites.¹² However, when the amount of filler reached 50 wt%, a decrement in the modulus was observed. As shown in Figure 4, the current high number of agglomerates may cause a lubricating effect on the composite. When a tension load was applied to the composite, the shear stress caused sliding in agglomerated rGP particles, resulting in a lubricating effect on the composite.

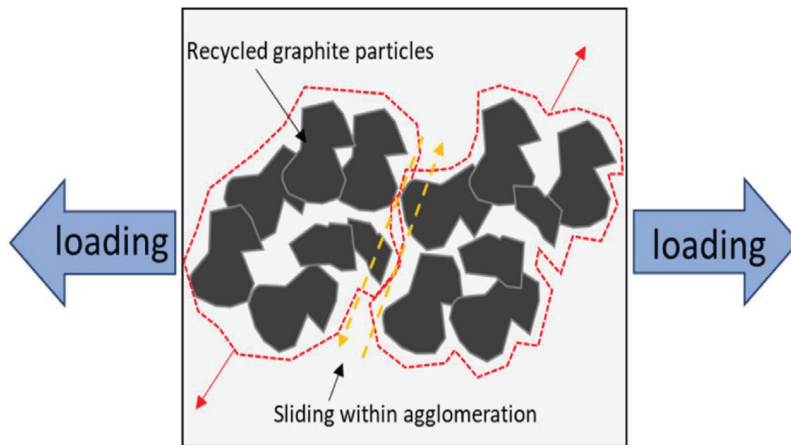


Figure 4: Proposed mechanism of lubricating effect due to filler agglomeration.

3.4 Morphological Properties

For the micrograph, the region highlighted with yellow colour square was captured at a magnification of 5000 \times . Figure 5 shows the SEM micrographs of the fracture specimen of neat PP. From Figure 5(a), the neat PP exhibited a uniform matrix tearing surface. In addition, the microfibrils on the fracture surface of neat PP were clearly observed at higher magnification, as shown in Figure 5(b). The presence of microfibrils was due to the ductile fracture of material after crazing. This finding indicated that the neat PP was a ductile material. The SEM micrographs of fracture surface of PP/rGP composites at selected filler contents are displayed in Figures 6 to 8. It can be observed that the rGP was in particulate form and irregular in shape, and the composites exhibited a brittle fracture surface with the addition of rGP. This was due to the addition of more rGP filler, which made the composite more rigid. The SEM result clearly showed that the rGP was highly agglomerated when the filler content was increased. As the filler content increased, the conductive network within PP matrix formed. The proposed mechanism of

formation of conductive network is displayed in Figure 9. From Figure 6, there was no agglomeration that could be observed and the rGP particles were separated by the PP matrix. Therefore, the PP/rGP composite with 10 wt% of filler content was insulative. Figure 8 displays that the rGP was agglomerated and the rGP particles were contacted by each particle. This observation indicated that when the rGP particles agglomerated, they initially formed conductive network within the PP matrix. As the filler content increased to 50 wt%, the agglomeration of filler became more visible. The micrograph in Figure 8(b) clearly shows that the rGP particles are highly agglomerated, indicating that PP/rGP with 50 wt% of rGP is the most conductive. In addition, a matrix tear found in Figure 8(b) indicated a lubricating effect on the PP/rGP composite with 50 wt% filler content. Therefore, this finding supported the reason as to why the tensile module of composite reduced at 50 wt% of filler contents.

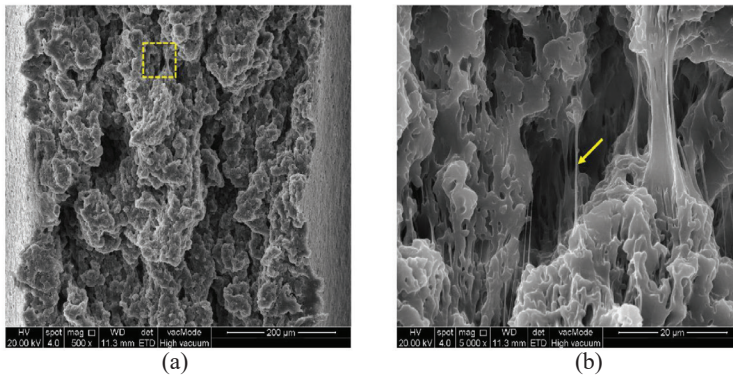


Figure 5: SEM micrographs of tensile fracture surface for neat PP at (a) 500× and (b) 5000× magnification.

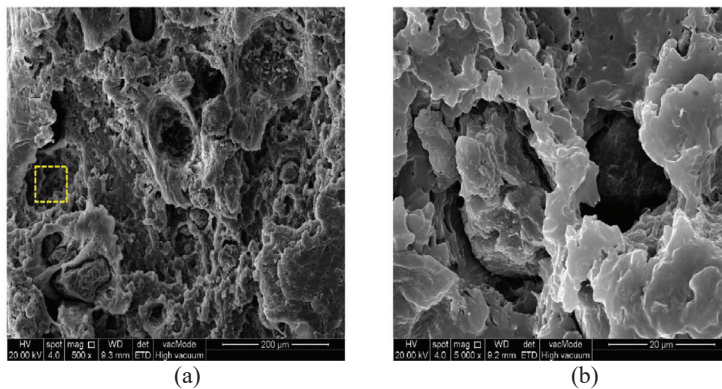


Figure 6: SEM micrographs of tensile fracture surface of PP/rGP composite with 10 wt% of filler content at (a) 500× and (b) 5000× magnification.

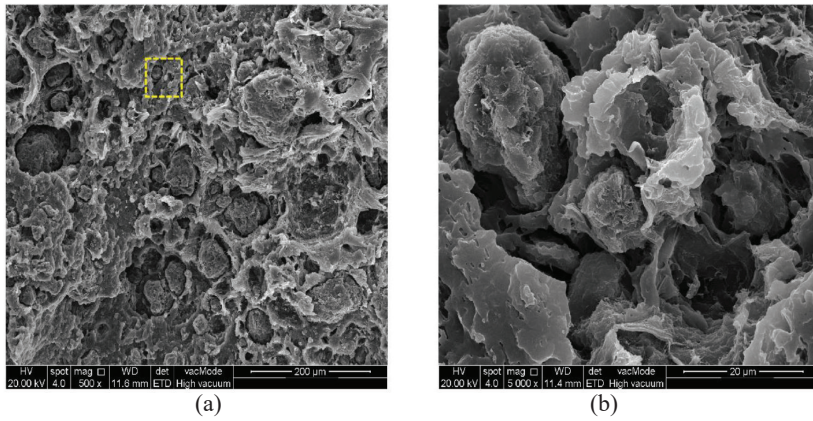


Figure 7: SEM micrographs of tensile fracture surface of PP/rGP composite with 30 wt% of filler content at (a) 500× and (b) 5000× magnification.

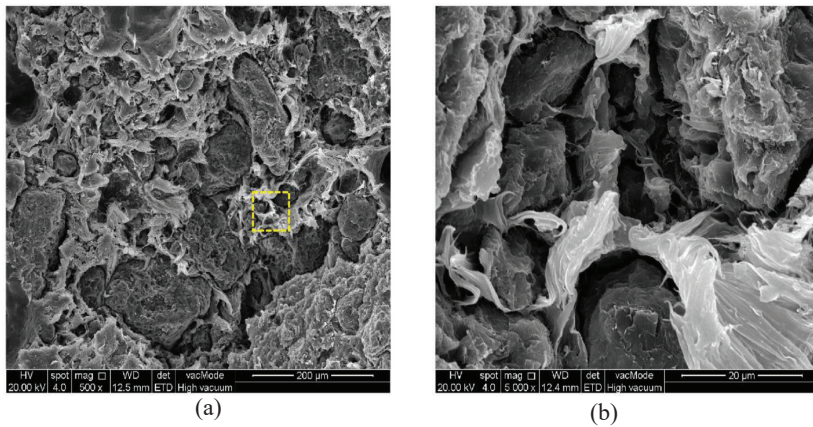


Figure 8: SEM micrographs of tensile fracture surface of PP/rGP composite with 50 wt% of filler content at (a) 500× and (b) 5000× magnification.

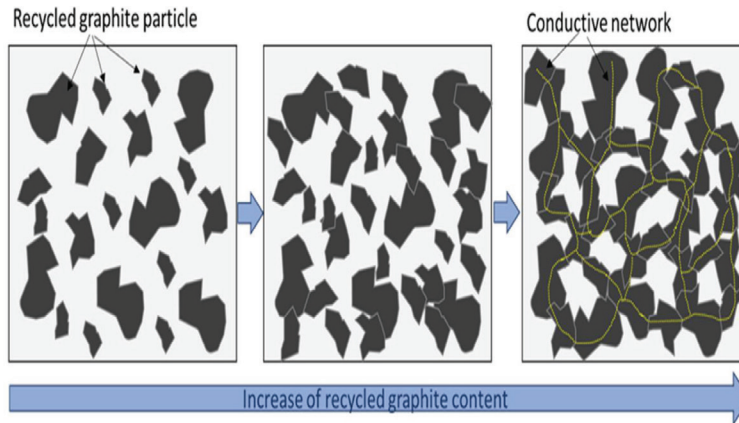


Figure 9: Proposed mechanism for formation of conductive network occur within PP/rGP composite.

3.5 Thermal Properties

TGA curves of rGP, neat PP, and PP/rGP with different filler contents are displayed in Figure 10. The results showed that the neat PP remained thermally stable until 340°C, but it thermally degraded at a faster rate above this temperature. Meanwhile, the raw rGP exhibited only a 2% weight loss after being heated at a temperature of above 700°C, which indicated that the rGP has very good natural thermal stability. Table 2 shows the degradation temperature at 10% of weight loss ($T_{d10\%}$) for neat PP and composites. When the amount of rGP was increased, $T_{d10\%}$ of composites significantly shifted to higher temperatures. This means that the addition of rGP significantly improved the composites' thermal stability. This was observed because the presence of rGP provided a thermal shielding effect on PP matrix. As shown in Figure 11, the PP chains exploded and thermally degraded from direct heat. The presence of rGP particles, on the other hand, prevented a portion of the PP chain from exploding due to direct heat, resulting in a thermal shielding effect. Therefore, the thermal degradation process of PP matrix was delayed due to the thermal shielding effect, and the degradation temperature of composites shifted to higher temperature. The addition of graphite generally improved the thermal stability of composites and this finding was similar to studies by other researchers.^{13,14}

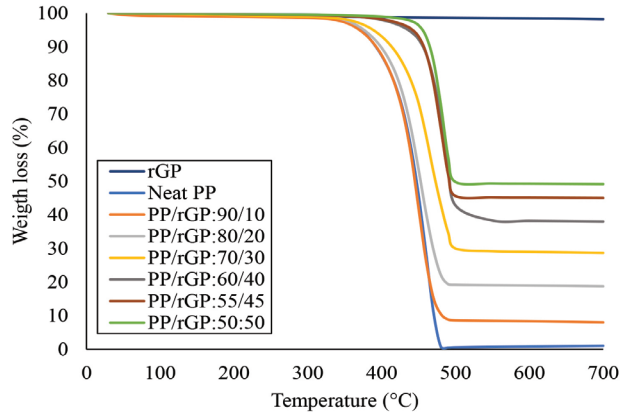


Figure 10: TGA curves of PP/rGP composites with different filler contents, neat PP and raw recycled graphite.

Table 2: $T_{d10\%}$ of PP/rGP composites with various filler contents.

Sample	$T_{d10\%}$ (°C)	Residue (%)
Neat PP	392	1.1
PP/rGP:90/10	395	8.0
PP/rGP:80/20	403	18.8
PP/rGP:70/30	414	28.7
PP/rGP:60/40	456	38.1
PP/rGP:55/45	459	45.1
PP/rGP:50/50	465	49.2

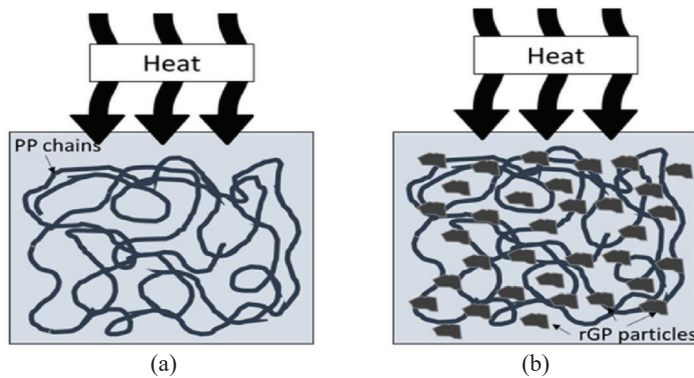


Figure 11: Different idealised thermal effects on specimen (a) without rGP and (b) with thermal shielding effect from rGP.

4. CONCLUSION

In conclusion, the EDM electrode scrap obtained from the machining industry was able to be recycled as a conductive filler for the production of conductive composite. In terms of compounding behaviour, the processing torque of PP/rGP compounding increased with rGP content. The PP/rGP composite exhibited a percolation content at 30 wt% of filler content, and the insulative composite initially changed to antistatic material. Moreover, the PP/rGP composite filled with 50 wt% of rGP achieved a surface resistivity of 10^5 ohm/sq, which shows that the composite can be used as static dissipative material. On the other hand, the tensile strength of PP/rGP composites decreased by approximately 60% as the filler content increased to 50 wt%. The tensile modulus of this composite increased and showed an optimum tensile modulus at a filler content of 45 wt%. The tensile modulus of composite decreased as the filler content was increased above 45 wt% due to the plasticising effect of rGP. The thermal stability of composite greatly increased with the addition of rGP. The morphological analysis confirmed that the increased rGP content was able to form a conductive network within the PP matrix in the composite. Therefore, this study found that rGP made from EDM electrode scrap has the potential to be used as a conductive filler for producing conductive composite material.

5. ACKNOWLEDGEMENT

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