Effect of Graphene Nano-fillers on Mechanical and Thermal Properties of Polypropylene (PP)/Pineapple Leaf Fibre (PALF) Composites

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ABSTRACT

GRAPHICAL ABSTRACT



impact strength and Young modulus of PP/PALF

Polypropylene (PP) is a popular commodity plastic due to having relatively good mechanical properties and processability. However, further modification of PP is needed in order to compete with engineering polymers. In this study, pineapple leaf fibre (PALF) and graphene are being incorporated to enhance properties without increasing the cost significantly. The PP composites were prepared with different graphene content (0.5, 1.0 and 2.0 phr) using melt blending method followed by injection moulding and the neat PP and PP/PALF act as control. The main focus of this study was to investigate the effect of adding both fillers on mechanical strength and thermal properties of PP. Tensile test, impact test and thermal gravimetric analysis (TGA) were carried out to study the properties of PP composites. Results showed that tensile strength (MPa) and Young's Modulus of PP increased by addition of PALF but decreased of elongation at break. The addition of graphene (0.5, 1.0 and 2.0 phr) on PP/PALF composite increased the Young's Modulus and tensile strength but it decreased the percentage of elongation at break. From Impact test, the use of PALF had increased the impact strength of PP. The use of graphene on PP/PALF composite had also increased the impact strength. In general, higher content of graphene (2.0 phr) decreased the mechanical properties of PP/PALF composite. From TGA test, the maximum decomposition of PP occurred within temperature 475 to 480°C. The addition of PALF and graphene gave no significant changes to thermal properties of PP.

Keywords: Polypropylene (PP), pineapple leaf fibre (PALF), graphene, mechanical strength, thermal properties

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INTRODUCTION 1.

Nowadays, many societies are interested in applying the principle of green chemistry in the production of polymers that are more environmentally friendly and are biodegradable. The polymers should be designed so that at the end of their usage, they will break down into innocuous degradation products and do not persist in the environment [1]. Many researchers are contributing in producing polypropylene (PP) composite using biofillers due to its low cost of production, good thermal and mechanical properties. The use of bio-filler and inorganic filler in PP is significant in this study to investigate if it can further improve properties of PP.

Pineapple leaf fibre (PALF), which is rich in cellulose, relatively inexpensive and is abundantly available, has the potential to be used in polymer-reinforced composites. Arib et al. (2006) investigates the tensile and flexural behaviours of PP/PALF composites as a function of volume fraction [2]. The tensile modulus and tensile strength of the composites were found to be increasing with increasing fibre content until it reaches the optimum amount of fibre content. Thus, PALF can act as bio-filler in PP to improve the mechanical strength of biopolymer. Besides, the usage of graphene has also been found to be a great potential as nanofiller in improving the yield strength, Young's Modulus and thermal oxidative stability as graphene has high mechanical strength [3]. Thus, the present study aims to investigate the effect of graphene on the mechanical and thermal properties of PP/PALF composites using melt blending method.

2. **EXPERIMENTAL**

The graphene powder was purchased from Sigma Aldrich and a commercial grade of PP (PP5032E1) purchased from ExxonMobil Chemical. Maleated PP was purchased from Dupont Fusabond P353. Pineapple leaf fibre (PALF) was produced from leaves of pineapple. It was purchased from agrowaste management lab at school of chemical engineering, UTM, Skudai, Johor Bharu. All the materials were used as received.

The melt blending process was used in this study to ensure the dispersibility of PALF and graphene in PP matrix and the PP/PALF composites were prepared according to formulation in Table 1. The percentage of PP and PALF used were 85% and 10% respectively. The malleated polypropylene (5%) was added into each formulation to ensure the interaction occur between PP with PALF and PP with graphene.

Graphene contents used were 0.5, 1.0 and 2.0 phr. The mechanical properties of composites were tested using tensile test and impact test while the thermal property was characterized using thermal gravimetric analysis (TGA) under nitrogen atmosphere at heating rate of 10°C min⁻¹. From tensile test, the tensile strength, elongation at break and Young's Modulus were obtained. From impact test and TGA, the impact strength and degradation temperature of PP were obtained respectively.

Samples	Coding	PP (wt%)	Maleated PP (wt%)	PALF (wt%)	Graphene (phr)
(Control 1)	PP	100	-	-	-
(Control 2)	PP/PALF	85	5	10	-
1	PP/PALF/0.5 Graphene	85	5	10	0.5
2	PP/PALF/1.0 Graphene	85	5	10	1.0
3	PP/PALF/2.0 Graphene	85	5	10	2.0

Fable 1. Preparation of PP/PALF/Graphene compose

3. RESULTS AND DISCUSSION

3.1. Characterization by thermogravimetric analysis

TGA results are shown in Table 2 which higlighted the $T_{20\%}$ (°C), T_{max} (°C) and residue (%). $T_{20\%}$ is the temperature at which 20% of PP weight was loss. T_{max} is the temperature at which the rate of decomposition of polypropylene is at maximum. The term residue means the weight of polypropylene composites that were left at the end of analysis. The analyses were carried out within temperature 25 to 600°C.

Coding	T20%	T _{max}	Residue
	(°C)	(°C)	(%)
PP	438.1	475.1	0.2
PP/PALF	438.2	478.2	0.5
PP/PALF/0.5 Graphene	439.5	478.2	1.3
PP/PALF/1.0 Graphene	438.1	480.2	0.5
PP/PALF/2.0 Graphene	431.8	476.5	0.3

Table 2. TGA Analyses of PP/PALF/Graphene composites

The results indicated that the use of PALF in neat PP had increased the degradation temperature from 475.1 to 478.2°C. In addition, the use of graphene (0.5 and 1.0 phr) on PP/PALF composite showed slight increase in the degradation temperature up to 1.0 phr graphene but decreased afterwards at 2.0 phr graphene. It was noted that for all composites, T_{max} (°C) were higher compared to PP. However, in general, the thermal properties of PP composites showed no significant changes after addition of PALF (10%) and graphene (0.5, 1.0 and 2.0 phr). In addition, the graphs of percentage of weight of PP composites versus temperature were plotted for all formulations. Figures 1 and 2 shows the graphs obtained from thermogravimetric analyzer. It can be seen that the degradation temperature of PP composites occurred within temperature 475 to 480°C.



Figure 1. TGA of neat PP and PP/PALF composite



Figure 2. TGA of neat PP and PP/PALF/graphene composites

From Figures 1 and 2, the first transition from 40 to 130°C due to the release of absorbed moisture in the fibres was missing since the PALF used is already in a dried form before the melt blending process. The second transition (the temperature range of the decomposition from 195 to 360°C) indicated the degradation of cellulosic substances such as hemicellulose and cellulose in PALF as stated in previous study [4]. The third stage (360 to 470°C) of the decomposition was due to the degradation of non-cellulosic materials in PALF [5]. The addition of graphene into PP/PALF composite did not show the degradation of any other substances as shown in Figure 2.

3.2. Mechanical properties

3.2.1. Tensile Properties

The effect of PALF on neat PP had increased the tensile strength as shown in Figure 3(a). The enhancement might be due to PALF being perfectly aligned with matrix of PP. Figure 3(b) showed the effect of adding graphene on tensile strength of PP/PALF composite. Tensile strength increased with increasing graphene content. These enhancements are attributed to the homogeneous dispersion of graphene and effective load transfer from the matrix of PP to graphene due to their strong interfacial adhesion [6].



Figure 3. Effect of (a) PALF content on tensile strength of PP and (b) graphene content on tensile strength of PP/PALF

The addition of PALF and graphene had given a negative impact to the elongation at break for the composites. Figure 4(a) and 4(b) show the effect of adding PALF on PP and the effect of adding graphene on PP/PALF respectively. Generally, it can be seen that the elongation at break (%) of PP/PALF and PP/PALF/graphene composites is lower as compared to neat PP. The addition of PALF had decreased the elongation at break of PP and the addition of graphene had decreased the elongation at break of PP/PALF. The negative results showed that the polymer composites became more brittle than neat PP. This is due to fibre (PALF) breakages that are wholly pull-out of the matrix [4].



Figure 4. Effect of (a) PALF content on elongation at break of PP and (b) graphene content on elongation at break of PP/PALF

The addition of PALF and graphene show an increased in the Young's Modulus of PP composites. The results can be seen in Figures 5(a) and 5(b). The addition of pineapple leaf fibre had increased the Young's Modulus of PP and may be caused by adhesion forces between the PALF and the PP matrix [5]. An adhesion forces between PALF and PP makes it feasible for stress transfer to take place from the PP matrix to the PALF, thereby improving the strength of the composites. The addition of graphene also had increased the Young's Modulus of PP/PALF. A non-polar characteristic of graphene makes it more compatible with hydrophobic PP and results in better interaction between them.



Figure 5. Effect of (a) PALF content on Young's Modulus of PP and (b) graphene content on Young's Modulus of PP/PALF

3.2.2. Impact Properties

The impact strength of PP had increased after addition of pineapple leaf fibre (PALF). Figures 6(a) and 6(b) show the results of impact strength after adding both fillers. The addition of graphene (0.5 and 1.0 phr) also increased the impact strength of PP/PALF. These enhancements were due to stress distribution was more uniform and allow both fillers to be easily incorporated into the matrix of PP structure [7]. In contrast, the use of 2.0 phr of graphene content had decreased the impact strength due to abundant amount of it in matrix.



Figure 6. Effect of (a) PALF content on impact strength of PP and (b) graphene content on impact strength of PP/PALF

4. CONCLUSION

PP/PALF/graphene composites prepared using melt blending method show an enhancement in most of their mechanical properties. It can be seen that the tensile strength, impact strength and Young's modulus of PP composite had increased by addition of PALF and graphene. However, the elongation at break had generally decreased by addition of filler and thermal properties gave no significant changes after addition of PALF and graphene. The optimum graphene content that showed improvements to PP/PALF composites properties was at 1.0 phr. The TGA results showed that the thermal stability of PP did not show any significant changes compared to all PP/PALF/graphene composites.

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