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Pineapple Peel Fibre Biocomposite: Characterisation and Biodegradation Studies

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In this study, pineapple peel fibre (PAPF) based low density polyethylene (LDPE) biocomposites for green packaging was studied. The PAPF was first being treated with alkali before compounded with LDPE. The mixture was compounded using twin screw extruder and the test samples were prepared using hot press machine. The compatibility of the PAPF as biocomposites was observed through the characterisation analysis and thermal properties and also the biodegradation analysis. Melt flow index (MFI) analysis was conducted to determine the process ability of the biocomposites. As the fibre loading in the biocomposites increases, the MFI values were decreased. The amount of water absorption was increased with the increases of PAPF loading due to the higher cellulose content. Thermal stability studies of biocomposites were undergoing thermogravimetry (TGA) and differential scanning calorimetry (DSC) analysis. Melting temperature (T_m) for the biocomposite was determined from the DSC analysis while the degradation temperature was determined by using the TGA analysis. The thermal properties of PAPF biocomposites were more or less the same as the LDPE properties. The biocomposites was buried in the soil for a month and exposed to fungi environment for 28 d for biodegradation analysis and the highest PAPF/LDPE loading biocomposites degraded the most. Therefore, PAPF biocomposites was compatible for green packaging.

1. Introduction

The annual world production of polymer materials was around 150 × 10^6 t in 1996 (Ren, 2003) and the current global consumption of plastics is more than 200 × 10^6 t, with an annual grow of approximately 5 % (Siracusa et al., 2008). Packaging waste is a major contributor of municipal solid waste (MSW) and disposed of by landfill (Kale, 2007). Landfilling disposal may result in the generation of greenhouse gases and takes up. It also may contaminate land that could be used in the future. The rapid increase in production and consumption of plastics has led to the serious plastic waste problem, besides landfill depletion because of the plastic waste high volume to weight ratio and resistance to degradation (Ren, 2003). Natural fibres reinforced composites will form new class of materials which possess a significant improvement in properties without sacrificing the desirable properties. The biocomposites also contains biodegradable components from waste for biodegradability and cost effectiveness. Pineapple peel fibre (PAPF) shows significant role as cheap, exhibiting superior properties and environmental friendly biocomposite as a reinforcement fiber. Pineapple leaf fiber (PALF) exhibit high specific strength and stiffness due to the high cellulose content which is 70 - 80 % and relatively low microfibrillar angle. Due to its excellent mechanical properties, PALF have a high reinforcing efficiency for application in polyester, low density polyethylene (LDPE) and biodegradable plastic composites. There are also some limitations encounters in the PALF bio-composites due to an inadequate bonding between PALF and hydrophobic matrix. PALF biocomposites also has high susceptibility to water absorption,

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particularly at elevated temperatures (Lopattananon et al*.,* 2006). Blending PAPF with LDPE will increase the mechanical properties of the composites as they have satisfactorily high specific strength and modulus light weight. However, increasing percentage of PAPF composition in the blend will introduce to the lower mechanical properties. The usage of suitable compatibiliser can reduce the incompatibility and increase the mechanical properties of the blend. In this study, LLDPE-g-MA will be utilised as compatibiliser to improve the strength and the mechanical properties of the blend PAPF and LDPE. The cooking oil was added as a function of processing aids. In addition, the PAPF were treated with alkali and peroxide to modify the fiber surface.

2. Materials and Methods

The PAPF was supplied by Lee Pineapple Co. (Pte.) Ltd. LDPE was supplied by Titan Polyethylene (M) Sdn. Bhd. The PAPF was treated with alkali treatment, NaOH to improve the adhesion between fiber and matrix and mixed with the Linear low-density polyethylene-grafted maleic anhydride (LLDPE-g-MA) and cooking oil. The fiber was immersed in 0.5 % NaOH solution for half an hour before being washed several times with cold water and finally with acidified water, HCl 0.1 N. Then, the fiber was dried in an air oven at 60 °C for 24 h. PAPF based biocomposites were dried in the oven at 80 °C for 24 h to eliminate moisture content. LDPE, PAPF and compatibiliser were mixed manually. First, the LDPE was well mixed with the plasticiser. Then, PAPF and compatibiliser were added into the mixture and were mixed again. The compounding of PAPF biocomposites were done using SINO PSM 30 co-rotating twin screw extruder in the feed, compression, metering and die zone with suitable temperature (150 °C, 155 °C, 165 °C, 165 °C, 150 °C, and 140 °C) and formulations were listed in Table 1. The pre-mix PAPF biocomposite were fed to feed hopper into the feed section of the barrel. The biocomposites were melted and mixed in various shearing by shearing action of the screw at speed 80 rpm. Then, continuous strands were formed through the die when the composites passed through the die zone. The biocomposites were pelletised using pelletiser machine.

Water absorption test were carried out to determine the resistance ability of the samples in the water. The samples were first being dried at 50 °C in an oven for 24 h and immediately weighed. According to ASTM D570, the samples of thickness 3.00 ± 0.05 mm were entirely immersed in a container of distilled water. At specified interval, each sample was removed from the water container, wiped with a clean cloth, and consequently weighed. The weight gains of the samples were recorded and the percentages of weight gain, M_t were determined by Eq(1);

$$M_t\ (\%) = \frac{(W_w - W_d)}{W_d} \times 100 \quad (1)$$

where, W_d = initial weight samples and W_w = Weight of samples after exposure to water absorption.

The samples were buried in the soil for about a month in a preparation box and the initial weight of the samples were recorded. On the testing day, the samples were wiped cleanly, and then will be dried in the oven at 60°C for 24 h. The final weights of the samples were recorded and the percentages of weight loss were calculated using the following Eq(2);

$$\text{Weight loss}\ (\%) = \frac{(W_o - W_s)}{W_o} \times 100 \quad (2)$$

where, W_o = initial weight of the samples and W_s = final weight of the samples

Table 1: Formulations of PAPF based biocomposites

Samples	LDPE (%)	PAPF (%)	Plasticiser (%)	LLDPE-g-MA (%)
LDPE	100	-	-	-
LDPE/PAPF:50/50	35	50	5	10
LDPE/PAPF:60/40	45	40	5	10
LDPE/PAPF:70/30	55	30	5	10
LDPE/PAPF:80/20	65	20	5	10
LDPE/PAPF:90/10	75	10	5	10

3. Results and Discussion

3.1. Water Absorption Test

Water absorption test is important to determine the water absorptivity of the biocomposites during certain period. The abundant of cellulose content when used to reinforced hydrophobic matrices; the result is a very

poor interface and poor resistance to moisture absorption. Cellulosics fibre is difficult to dissolve because of their high crystallinity but they tend to retain liquids in the inter-fibrillar space. Based on Figure 1, the amount of water absorption increased with the increases of PAPF loading due to the higher cellulose content. The LDPE/PAPF with 50 % PAPF composition shows the highest increment in the weight due to the higher water absorption.

The percentage of weight gained for 50 % PAPF loading was 7.6 %. The 10 % of fibre loading shows no significant changes in their weight compared to the others. The treated PAPF reduced the water absorption because of better interfacial bonding. To promote the adhesion between the fibre and the matrix, chemical treatment or modifications are considered. Chemicals activate the hydroxyl groups or introduce new moieties that can effectively interlock with the matrix (George et al., 1998). Alkali treatment has two effects on the fibre which are increasing the surface roughness resulting in better mechanical interlocking; and increasing the amount of cellulose exposed on the fibre surface, thus increasing the number of possible reaction sites. Alkali treatment also reduces the polarity of the PAPF which increased the crystallinity and reduced the sorption capacity of the fibre. The chemically modified LDPE/PAPF exhibited a reduction in water uptake.

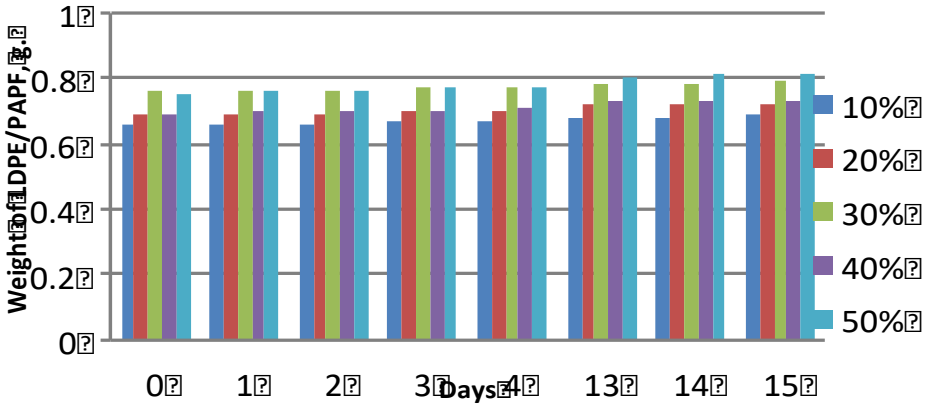

Figure 1: Weight gained of PAPF based biocomposites with various fibre contents

3.2 Melt Flow Analysis

Table 2 shows that the MFI values for LDPE/PAPF decreased as the fibre loading increased. The decreasing of MFI values indicates the decreasing in the flow ability of the biocomposites. The decreasing in the MFI values also indicates the increasing of the viscosity of the biocomposites. The viscosity of the system increased with fibre loading due to an increased hindrance to the flow. Addition of bonding agents increases the viscosity of PAPF biocomposites due to the increased fibre-matrix interaction. The mechanical interlocking between fibre and matrix was increased due to the removal of the waxy material present on the surface of the fibre (George et al., 1996).

Table 2: Melt flow index of PAPF based biocomposites

Samples	Composition	MFI value (g/10 min)
LDPE	100	4.78
LDPE/PAPF	90/10	6.92
LDPE/PAPF	80/20	5.10
LDPE/PAPF	70/30	2.89
LDPE/PAPF	60/40	0.90

3.3 Thermogravimetry Analysis

The thermal degradation behaviour of biocomposite passes through two stages upon heating from 34 °C to 960 °C and was shown in Figure 2. Within the first stage up to 250 °C, there was minor significant difference in thermal stability between PAPF and LDPE. However, the difference of thermal stability can be easily observed within the major degradation region from 250 °C to 490 °C, in which all the PAPF biocomposites display lower thermal stability due to the degradation of cellulose, hemicellulose and lignin in samples compared to polymer LDPE. This indicates that the thermal stability of sample decrease when PAPF was added into the polymer LDPE (Zarate et al., 2008). Hydrogen bonding is the major source of stability in cellulose which allow the thermal energy to be distributed over different type others bonding. Thus, interaction

between PAPF and polymer LDPE may decrease with the increase of the PAPF content. In others word, the addition of PAPF enhancement contributes to the reduction at 140 °C of the biocomposite stability when compared with polymer LDPE.

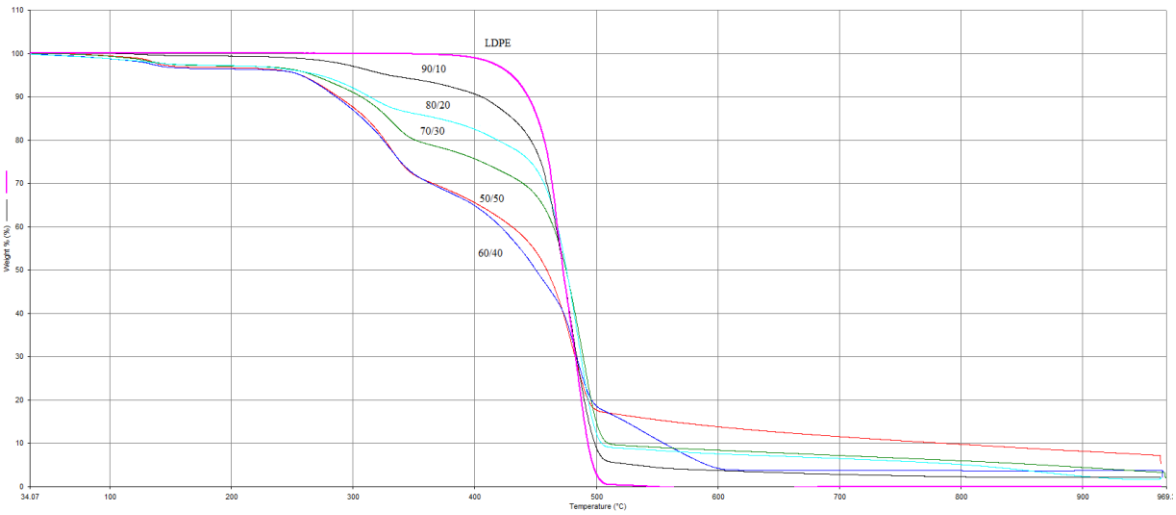

Figure 2: Thermogravimetry analysis of PAPF composites

3.4 Differential Scanning Calorimetry Analysis

Differential scanning calometry analysis for PAPF composites were shown in From Figure 3 and it can be observed that the introduction of PAPF significantly changes the thermal behaviour of investigated sample in glass temperature. The lowest melting temperature was observed with the smallest content of PAPF, which is 101 °C with 10 % PAPF content while higher melting temperature is 120 °C with 50 % PAPF content. The reason of this change can be related to the increasing of PAPF content in the composites. Thus, adding of PAPF into LDPE enhances the thermal transition of biocomposite.

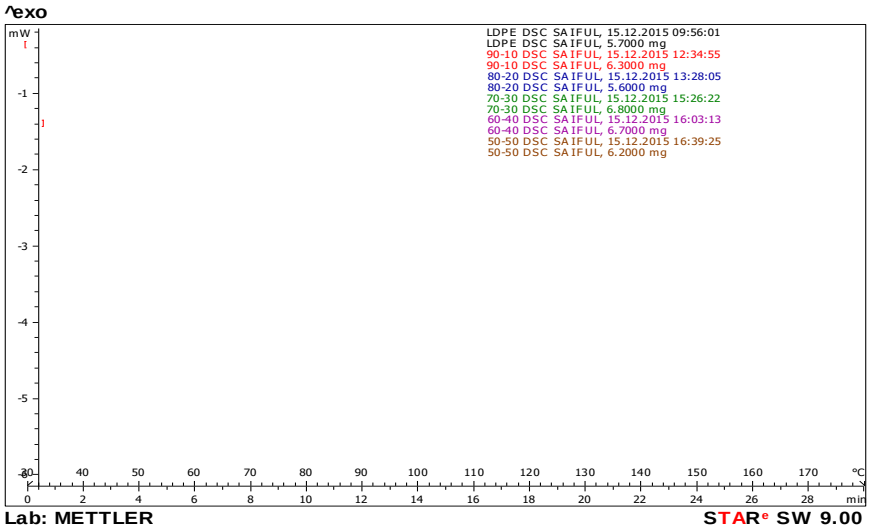

Figure 3: DSC thermogram of PAPF biocomposites

3.5 Soil Burial Test

Different sample of pineapple peel based biocomposite was buried under soil surface that consisted compost, and garden soil. Soil environment contains different kind of microorganism and macroorganism. From Figure 4, it can be observed that the mass losses of pineapple peel based biocomposite were measured due to the degradation time for 6 month. Notable trend here is the rate of biodegradation was significantly increased from

30 - 50 wt % of pineapple peel content. This result was expected due to the higher amount of pineapple peel fibre content that increased the rate of biodegradation (Kijchavengkul et al., 2006). Microorganisms were attracted to the pineapple peel content of blends. Microorganisms consume pineapple peel in the polymer and caused a fracture in the LDPE chain. Because of the existence of maleic anhydride as compatibiliser that made a chemical bond between PAPF and LDPE, degradation of pineapple peel caused a fracture in the polymer matrix (Wang and Yu, 2005; Ali et al., 2013). However, pineapple peel content with show a slightly decrease in weight loss from 10 - 20 wt %. This problem was occurred due of pineapple peel content might be contaminated. Thus, the contaminated sample was interrupt the microorganism to attack and metabolised the polymer matrix.

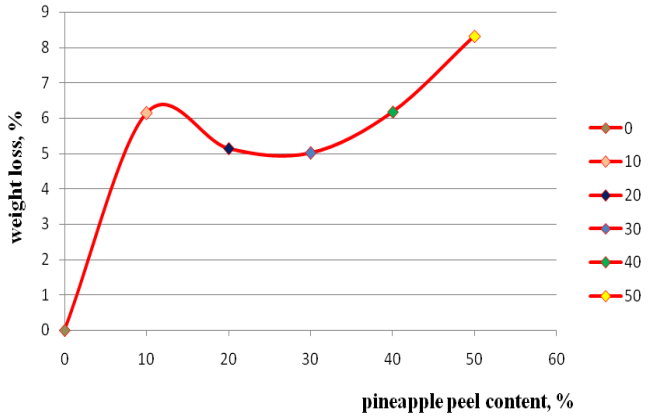

Figure 4: Percentage of weight loss of PAPF biocomposites for soil burial test

3.5 Exposure to Fungi Analysis

The fungi behaviour of biocomposite was determined by using Aspergillus Niger as a microorganism. The sample has been done in laboratory for 28 d. A weight loss of the sample as a function of biodegradation time obtained from fungi test was shown in Figure 5. From Figure 5, it can be seen clearly that trend of fungi degradation exhibit the positive results which has been describe with increasing the curve in percentage of weight loss. The pineapple peel content ranging from 10 - 50 wt % created variation in mass losses. The 50 wt % of PAPF biocomposites show the highest percentage in weight loss compared to others. This result indicates there has activity of fungi been occurred in polymer matrix. The higher fibre content enhances the degradation of bio composite (Liu et al., 2003). Microorganism recognises the pineapple peel link as a nutrient source. Consumption of polar hydrophilic pineapple peel caused fracture in the polymer chain. Maleic anhydride created a link between two incompatible particles, so with removal of pineapple peel, a gap appeared in the polymer chain. Through the gap, microorganism had access to the link of low density polyethylene (Labuzek et al., 2004).

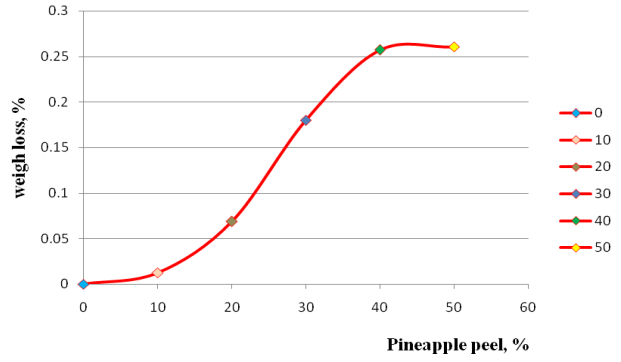

Figure 5: Percentage of weight loss of PAPF biocomposites for fungi analysis

4. Conclusions

It can be conclude that the antimicrobial PAPF based biocomposite was developed in this research. The PAPF up to 50 wt % are still able to be processed by using conventional plastic machinery which is injection moulding. The analysis of PAPF based biocomposite show the presence of O-H group in the fibre that would promote to absorb the water to its surface. The degradation of biodegradable plastic can be enhances with addition of PAPF as a natural fibre. These also suggest that the hydrolysis ability of the polymer matrix can be controlled by altering the amount of the added PAPF.

Acknowledgments

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