

# Characterisation and Biodegradation of Poly(Lactic Acid) Blended with Oil Palm Biomass and Fertiliser for Bioplastic Fertiliser Composites

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This work presents a new technique for producing a slow-release fertiliser with bioplastic polymer coating. Poly(lactic acid) (PLA) was blended with granular NPK fertiliser and empty fruit bunch (EFB) fibres using extrusion technique. The polymer coatings were characterised using thermal gravimetric analyser (TGA) and diffraction scanning calorimetry (DSC). The PLA and EFB fibres complemented each other in terms of their thermal stability in the BpF composites. A homogenous BpF blend was observed under a scanning electron microscope (SEM). In biodegradation the percentages of weight loss for PLA/EFB/NPKC1 and PLA/EFB/NPKC2 were higher due to the presence of EFB fibres, which were 64.3% and 76.3%, respectively.

*Keywords:* Bioplastic; Oil palm biomass; Fertiliser; Composites; Leaching

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## INTRODUCTION

Due to much economic and environmental concern, the agricultural sectors have been asking for more research on optimizing the use of fertilisers while achieving considerable increases in yield (Wu *et al.* 2008; Teodorescu *et al.* 2009; Ni *et al.* 2010; Jin *et al.* 2011). Depending on the soil condition and method of application, the wastage of conventional fertilizers with regard to plant nutrient uptake may reach up to 50% (Vassilev and Vassileva 2003; Entry and Sojka 2008). Most fertilizers contain water-soluble inorganic salts. The dissolution of these salts at high rate causes excessive levels of nutrient concentration that disrupt the plant nutrient uptake while bringing hazard to the surrounding environment.

Bioplastic fertilisers (BpFs) have been used in the agricultural industry for decades. They are considered an advanced approach within the controlled-release fertiliser (CRF) industry and play an important role in efficiently delivering the nutrients for plant uptake while controlling the longevity of the product. Common CRF normally only provides nitrogen, while BpF provides extra micro and macronutrients due to compared traditional water-soluble fertilisers. Various techniques of controlled-release fertiliser applications have been introduced to enhance the fertiliser efficiency and reduce the concerns and frequency of their applications (Wu *et al.* 2008; Teodorescu *et al.* 2009; Ni *et al.* 2010; Jin *et al.* 2011). Two approaches seem to have proven most effective. The

first one involves the mechanism of progressive dissolution to attain slow nutrient release, which can be achieved by synthesising novel compounds with low water solubility (Bhattacharya *et al.* 2007; Bandyopadhyay *et al.* 2008). The second option is to coat the fertilizers with organic materials that have low water solubility (Jarosiewicz and Tomaszewska 2002; Ni *et al.* 2010). The coating allows control over the rate of nutrient release by means of its diffusibility and biodegradability characteristics, depending on the varying physical and chemical properties of the coating material. In order to ensure the sustainability and environmental safety of its usage, it would be advisable to obtain the coating material from renewable sources.

As a means to achieving such goals, many bioplastics have been synthesised from renewable resources, such as poly(lactic acid) (PLA), polyamide, polycaprolactone, polyhydroxyalkanoates, *etc.* PLA can be considered a very useful compostable and biodegradable bioplastic/thermoplastic. Due to its compostable characteristic, it has a high potential to be used in various applications (Vink *et al.* 2003; Lima *et al.* 2008). However, it has one disadvantage, that is, its brittleness, which makes it unsuitable as a structural component (Cai *et al.* 1996; Pilla *et al.* 2010). Recently, scientists have discovered its significant use as matrix for polymer composites. They found it suitable for industrial production lines at large scale, such as thermoforming, extrusion, moulding, or blow moulding under its degradation temperature, which is 200 °C (Ramli *et al.* 2013; Chowdhury *et al.* 2013; Mina *et al.* 2014). It is crucial to maintain the processing temperature below PLA's degradation temperature to ensure the stability of its thermal properties and molecular weight.

Researchers are interested in reinforcing polymer-based composites with natural fibres because they have many advantages such as lower cost, lighter weight, lower density, higher strength-to-weight ratio, better biodegradability, better thermal and insulating properties, and other acceptable specific properties (Kim *et al.* 2012). Using natural fibres in polymer composites reduces the market cost, promotes global sustainability, and increases commercial use in various applications (Jawaid *et al.* 2011). However, there are limitations to using natural fibres as a reinforcing material because of their poor wettability, high moisture absorption tendency, poor adhesion with synthetic materials, and low thermal stability during processing (Demir *et al.* 2006). Oil palm empty fruit bunch (EFB) fibres are considered as agricultural wastes and renewable resources. This resource competes with other natural fibres as a promising reinforcing agent for polymer matrix (Rozman *et al.* 2000; Alam *et al.* 2011). The amount and properties of the lignocellulosic component determine the properties of the fibres and contribute to the properties of the composite materials. Regardless of their potential, EFB fibres as well as other natural fibres possess some weaknesses, such as having low thermal stability, moisture affinity, and low compatibility with polymer matrix.

In the present study, NPK fertiliser was compounded in a biodegradable plastic together with empty fruit bunch (EFB) for extrusion. It is clear that slow-release agents on the market are made of non-biodegradable polymer/plastic through spraying, emulsion, or dipping; however, a compounding technique was applied in this study as a novel approach to produce a slow-release fertiliser using biodegradable plastics like PLA. The extrusion method-developed bioplastic fertiliser composites can degrade in soil over a longer period of time as compared to conventional fertilisers. The main objective of the present study is to determine the biodegradation rate of bioplastic fertiliser (BpF) composites under soil burial test.

## EXPERIMENTAL

### Materials

Poly(lactic acid) (PLA) was obtained from the Innovative Pultrusion Sdn. Bhd., Senawang, Negeri Sembilan, Malaysia. The PLA (Nature Works™) used had the following properties: density of 1.25 g/cm<sup>3</sup>; 58 °C glass transition temperature ( $T_g$ ); 160 °C melting point ( $T_m$ ); 195,000 to 205,000 g/mol molecular weight ( $M_w$ ); and a melt flow index (MFI) of 2 to 5 g / 10 min (190 °C / 2.16 kg). Oil palm empty fruit bunch (EFB) fibres were used as the natural reinforcement for the polymer composites and were obtained from Poly Composite Sdn. Bhd., Teluk Intan, Perak, Malaysia. NPKC1 (granular) and NPKC2 (formulation commercial CRF) were obtained from Diversatech Fertilizer Sdn. Bhd, Bangi, Selangor, Malaysia.

### Methods

#### *Bioplastic fertiliser (BpF) formulation and compounding process*

The compounding of the materials was carried out by using the twin-screw extruder (MICROMAC, Malaysia). PLA, EFB fibres, and NPK fertiliser were dried at 103±2 °C overnight in an oven drying. The product was called bioplastic fertiliser (BpF) composites, containing biodegradable plastic, EFB fibres, and NPK fertiliser. The bioplastic fertilisers were dried to remove any moisture until processing. The reverse barrel temperature profile was 160 °C, and the extruder screw speed was maintained at 50 rpm. The temperature ranges were as follows: Section I, 145 to 150 °C; section II, 150 to 155 °C; and section III, 155 to 160 °C. All the components were premixed until the mixture became homogeneous. The homogeneity of the compound mixture was determined through the speed and temperature fluctuation panel. Table 1 shows the formulation of the bioplastic fertiliser (BpF) composites.

**Table 1.** Formulation of Bioplastic Fertiliser (BpF) Composites

Composites	PLA (%)	EFB Fibre (%)	NPK Fertiliser (%)
PLA/NPKC1	40	0	60
PLA/NPKC2	40	0	60
PLA/EFB/NPKC1	30	10	60
PLA/EFB/NPKC2	30	10	60

#### *Thermogravimetric analysis (TGA)*

Thermal properties were analysed by a thermogravimetric analyser (TGA Q500 TA Instruments, USA). Ten milligrams of the samples were heated in the temperature range of 35 to 600 °C at a rate of 10 °C/min. The analysis required a nitrogen atmosphere with a nitrogen flow rate of 20 mL/min. The weight reduction of the samples was recorded and plotted against temperature.

#### *Differential scanning calorimetry (DSC)*

The equipment used for DSC analysis was a differential scanning calorimeter (Q20 TA Instruments, USA). The analysis required an argon atmosphere with an argon flow rate of 50 mL/min. The heating rate was set at 5 °C/min and an aluminium sample pan was used to hold the samples. Ten milligrams of each sample was observed between

25 and 200 °C. The melting temperature ( $T_m$ ), glass transition temperature ( $T_g$ ), and cold crystallisation temperature ( $T_c$ ) of each sample were recorded.

#### *Scanning electron microscopy (SEM)*

The surfaces of the fractured test bars were analysed using a scanning electron microscope (Hitachi S-3400N, Japan). The acceleration voltage at the cathode was 15 kV. The samples were first sputter-coated with gold using an Emitech K550X (UK) coater (600 s, 35 mA) at a pressure of  $2 \times 10^{-1}$  bar.

#### *Biodegradability test*

Biodegradation testing of the BpF composites involved the simulation of a natural soil environment in a simple soil burial test. The natural soil was collected previously and did not contain any composting materials or enzyme activity. In this test, samples were simply buried in the natural soil inside polybags and were kept at a temperature between 25 and 30 °C, with a relative humidity of approximately 65%. The test spanned a total of 24 weeks and the samples were recovered every four weeks for analysis. Thus, there were seven degradation stages, starting from Week 0, Week 4, and so on, until Week 24. For each stage, the samples recovered were rinsed under running water to remove the soil residues from the surface. A vacuum oven with a temperature of 80 °C was used to completely remove the moisture until a constant dry weight was obtained for each sample. The weight loss after the soil burial test indicated the level of biodegradability of the samples and was calculated using Eq. 1,

$$\text{Weight loss (\%)} = (W_0 - W_1) / W_0 \times 100\% \quad (1)$$

where  $W_0$  is the weight before the test and  $W_1$  is the weight after the test.

## RESULTS AND DISCUSSION

### **Fabrication of Bioplastic Fertiliser (BpF) Composites**

The extrusion process from the feeder to the mould took approximately 5 min. The product that extruded over the first 5 min of each run was discarded, and then the strands of the BpF fertiliser composites that extruded through the moulded cylinder-shaped mould were cut at approximately 150 mm in length before being divided further into 30-mm cylinders for the biodegradation soil tests. Next, the BpF composites were then placed in an airtight plastic bag. Samples of the BpF fertiliser composites are shown in Fig. 1.

The processing conditions, which determine the dispersion and the adhesion/interaction between the fillers and the matrix, influence the enhancement of the resulting composite material (William *et al.* 2000; Oksman *et al.* 2004). One of the best methods to obtain good dispersion of fillers in the biopolymer matrix is by using the twin-screw extrusion technique (Mishra *et al.* 2002; Oksman *et al.* 2003); however, according to Lee *et al.* (2005), the sensitivity of the temperature during the compounding processes involving extrusion or injection moulding is limited. The interaction between the matrix and the fillers (reinforcement), which provides a large surface area, improves the mechanical properties as well as the dimensional and thermal stability of the composite (Felix and Gatenholm 1991; Paul and Thomas 1997). From the aspect of processing, it is

also important to understand the thermal behaviour of mixtures involving polymers and cellulosic materials. There are many scientific reports on the effects of co-pyrolysis of synthetic polymer and cellulose derivative mixtures (Jakab *et al.* 2000, 2001).



Fig. 1. Bioplastic fertiliser (BpF) composites

The dispersion of fertiliser and filler material in the PLA matrix determines the degree of reinforcement in bioplastic fertiliser composites. It is also influenced by the interaction between the surfaces of the PLA phase and the filler. To have good dispersive mixing, the use of a typical single screw extruder would not be enough to properly disintegrate the additive agglomerates. This limitation can be overcome by having a twin-screw extruder for a better dispersive mixing of materials. Based on the results, when extruding PLA, it is important to minimise the residence time and processing temperature. In terms of resin formulation, it should be noted that the reverse depolymerisation and hydrolysis reactions may be catalysed by the presence of residual polymerising catalysts (Gruber *et al.* 1996; Witzke 1997), which explains the varying molecular weight drop for the melt-processed PLA.

### Thermogravimetric Analysis (TGA)

Thermogravimetric analysis (TGA) is based on the weight change *versus* temperature. In this study, TGA was used to quantitatively determine the composition and the degradation rate of the matrix as well as the filler fibres in the composite. The derivative thermogravimetric (DTG) curve further illustrated the behaviours of the components of the composites and their reactions at different temperatures, based on the location and magnitude of the peaks. As shown by Table 2 and Fig. 2, the thermal degradation of pure PLA started at 274.63 °C and the fastest weight loss occurred at 357.21 °C. Approximately 98.8% of PLA weight loss occurred at 550 °C. Additionally, the thermal stability of the PLA/NPKC2 composites was the highest, with an initial decomposition temperature of 294.34 °C.

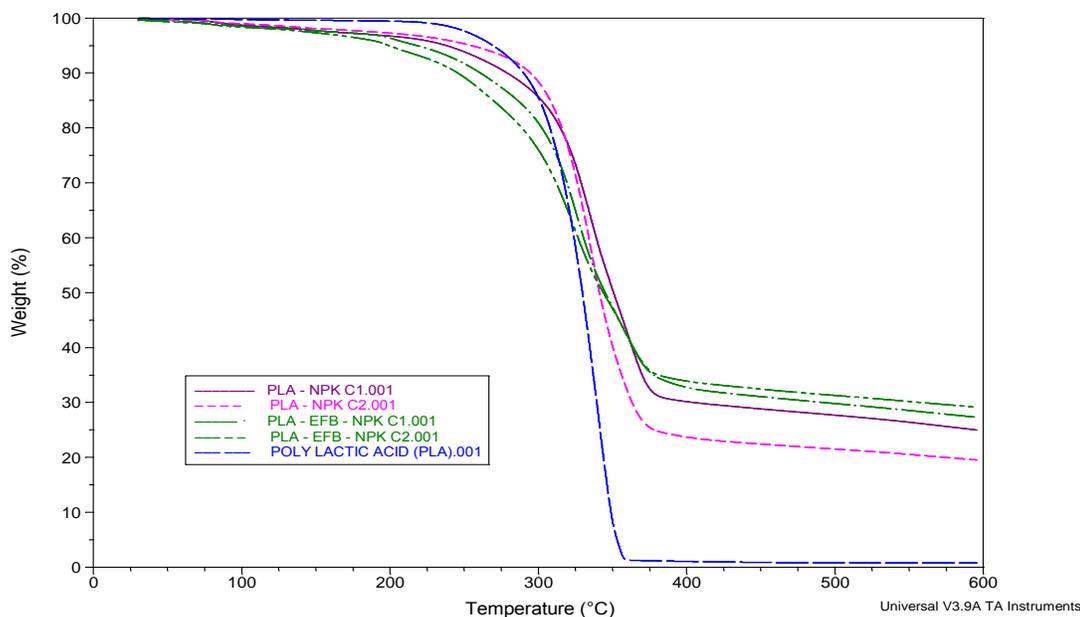
The PLA/NPKC2 composites displayed higher weight loss as thermal degradation increased but its weight loss was lower than those of the PLA/NPKC1 composites. The percentage weight loss for the PLA/EFB/NPKC2 composites with the uncoated fertiliser decreased at 550 °C. In the low-temperature region, the presence of PLA in the composites compensated for the low thermal stability of the EFB fibres. Similarly, in the high-temperature region, the presence of EFB fibres compensated for the low thermal stability of the PLA. As can be seen in Table 2, decreasing the percentage weight loss and enhancing the percent residue resulted in an increase in the thermal stability of the

composites. Fine fibre–matrix adhesion caused the PLA/EFB/NPK composites to be more thermally stable than pure PLA.

**Table 2.** Peak Temperature and Percent Weight Loss of Pure PLA, PLA/NPK, and PLA/EFB/NPK Composites

Composites	Degradation Temperature (°C)		Residual (%) (at 550 °C)
	$T_{IDT}$	$T_{FDT}$	
NPKC1	76.12	285.45	41.80
NPKC2	76.01	372.63	40.82
EFB	251.18	342.82	23.11
PLA	274.63	357.21	0.77
PLA/NPKC1	271.96	376.39	25.00
PLA/NPKC2	294.34	370.53	19.54
PLA/EFB/NPKC1	266.63	380.12	27.30
PLA/EFB/NPKC2	244.79	375.86	19.54

Notes:  $T_{IDT}$ : initial decomposition temperature,  $T_{FDT}$ : final decomposition temperature, PLA (polylactic acid), NPKC1 (uncoated), NPKC2 (coated) fertiliser and EFB (empty fruit bunch)



**Fig. 2.** TGA thermograms showing the thermal properties of pure PLA, PLA/NPK, and PLA/EFB/NPK composites

In a previous study, NPK was observed to have multiple degradations at 78, 172, 247, 286, and near 339 °C. In preliminary work, NPK was approximately 57.5% at 550 °C. The degradation temperature of NPK when combined with PLA was lower than the neat PLA temperature (Harmaen *et al.* 2015). Complex mineral fertilisers with

ammonium nitrate are able to decompose on their own by means of exothermic heat, provided that some conditions are fulfilled. These conditions include the following: local heating, a closed storage room with an insufficient heat exchange, and a high environmental temperature. A closed system catalyses the decomposition reaction because of the overheating experienced by the whole mass of the substance. The overheating leads to thermal explosion and releases gaseous products such as  $N_2O$ ,  $N_2$ ,  $NO_2$ ,  $H_2O$ ,  $Cl$ , and  $NH_4Cl$ . In other words, the fertiliser will start to decompose only if the temperature is high enough to initiate the process. Degradation of the fibres changes the fibre structure and has negative effects on the mechanical properties of the composites. In addition, the degradation also produces volatile residues that form microvoids on the surfaces of the composite components, which leads to debonding and failure of the material. It is also important to note the thermal degradation of the natural fibre composites because it influences the optimum temperature that should be used during processing. Degraded natural fibres deteriorate the mechanical as well as the organoleptic properties, such as colour and odour (Rennekar *et al.* 2004).

### Differential Scanning Calorimetry (DSC)

Table 3 summarises the thermal properties that resulted from the DSC studies: melting temperature ( $T_m$ ), crystallisation temperature ( $T_c$ ), and glass transition temperature ( $T_g$ ). The  $T_g$  of the PLA was 58.80 °C, while its  $T_m$  was 117.50 °C. The  $T_m$  and  $T_g$  remained almost the same after the fertiliser and EFB fibres were added to the PLA matrix (Table 3). The  $T_c$  of both fertilisers NPKC1 and NPKC2 decreased by approximately 6 °C, indicating a hindrance of the diffusion and migration of PLA molecular chains within the composites (Kokta *et al.* 1985; Albano *et al.* 2003). The melting temperature of the PLA was 155.50 °C, and it decreased by up to 5% when mixed with EFB and NPK. The results of the DSC show a small reduction in  $T_m$  after the EFB fibres were added to the PLA/NPK composites (Fig. 3).

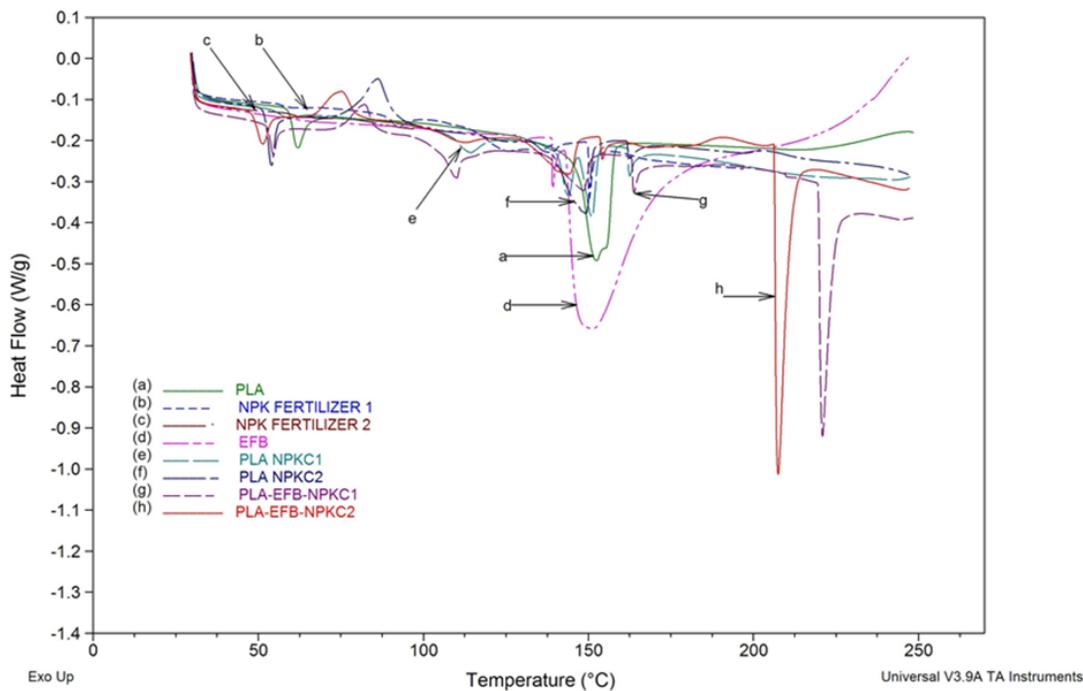
**Table 3.** DSC Characterisation of Pure PLA, PLA/NPK, and PLA/EFB/NPK Composites

Sample	$T_g$ (°C)	$T_c$ (°C)	$T_m$ (°C)
NPKC1	62.75	92.48	150.34
NPKC2	ND	107.80	197.20
EFB	ND	138.80	177.60
PLA	58.80	117.50	155.50
PLA/NPKC1	63.10	117.02	152.10
PLA/NPKC2	60.25	115.10	150.00
PLA/EFB/NPKC1	60.20	110.50	150.00
PLA/EFB/NPKC2	60.10	105.55	150.10

Notes:  $T_g$ : glass transition temperature,  $T_c$ : cold crystallisation temperature,  $T_m$ : melting temperature, PLA (poly lactic acid), EFB (empty fruit bunch), NPKC1 (uncoated), NPKC2 (coated) fertiliser and ND (not determined)

This reduction was probably caused by the restricted chain mobility of the PLA molecules or simply because of the weight reduction as a result of fibre addition. No significant effect was observed after fibre addition, and the same observation has been obtained by other researchers (Mishra *et al.* 2002; Shibata *et al.* 2002; Lee *et al.* 2008). They reported that the addition of a small amount of cellulose fibres to the PLA matrix did not lead to a change in  $T_g$ . It also showed that the PLA exhibited a single endothermic

melting peak at 151 °C and no exothermic peak, whereas the pure PLA and most of the PLA composites showed two distinct endothermic melting peaks, at 147 to 151 °C and 153 to 156 °C, respectively, in addition to one exothermic crystallisation peak at 99 to 110 °C.



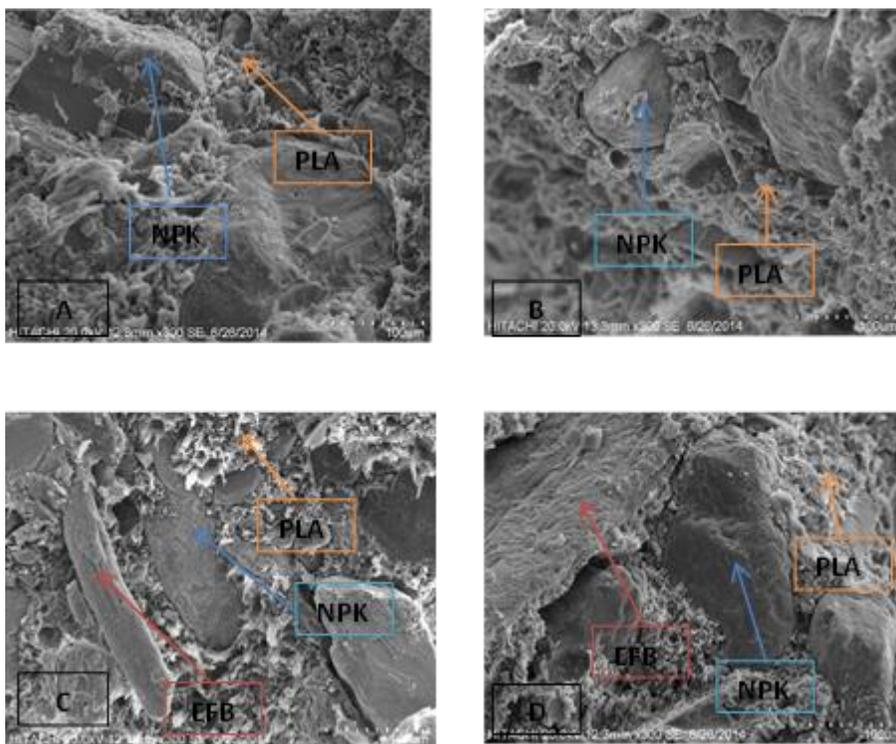
**Fig. 3.** DSC characterisation of pure PLA, PLA/NPK, and PLA/NPK/EFB composites

There are several factors that affect the crystalline and thermal behaviours of PLA-based composites, such as thermo-mechanical processing, the nature of the fillers, the composition, the time-temperature relationship of the formed composite, and the molecular weight of the PLA matrix (Tuominen *et al.* 2002). In this study, the dual melting peaks for the PLA sample were initially present; therefore, the two melting peaks observed were considered to have no association with the addition of EFB fibres. They were more likely related to the multi-step processing conditions of the PLA and the PLA-based composites. Tuominen *et al.* (2002) also reported an observation of dual melting peaks. Theoretically, they may be caused by the rearrangement of the lamellar structure when the PLA crystallised. This rearrangement formed less perfect crystals on the melting endotherms, in addition to the original crystallites. The  $T_g$ ,  $T_c$ , and  $T_m$  values for the PLA/EFB/NPK experienced some changes because of the presence of the EFB fibres, which promoted progressive hydrolysis of the PLA during thermal processing. The normalised PLA crystallinity decreased as a result of the thermal processing of the PLA in the composites as well as the pure PLA.

### Scanning Electron Microscopy (SEM)

In Fig. 4, parts A (PLA/NPKC1), B (PLA/NPKC2), C (PLA/EFB/NPKC1), and D (PLA/EFB/NPKC2) show the SEM micrographs of the fractured surface of the PLA/NPK, PLA/EFB/NPK, and the composites after extrusion. A smoother fracture surface of the PLA/NPK indicated the brittleness of the polymer matrix. On the other hand, the fractured PLA/EFB/NPK indicated ductile behaviour. This was attributed to the

combination of low interfacial adhesion between the PLA and its composites, and to the much higher viscosity of the PLA-dispersed phases. In Fig. 4, the EFB fibre and NPK fertiliser in the PLA matrix were observed. The crack propagation groove was directly generated from the agglomeration. On the other hand, the cryo-fractured surface of the PLA after fibre and fertiliser addition was rougher, implying that local ductile surfaces were generated during fracture. Adding EFB fibres did not change the continuous morphology, though some spherical dispersed phases were observed, which indicates a slight change in the melt rheology of the blends. For the PLA, it was found that the PLA-dispersed phases were higher than those of the fibre blend. This was due to higher shearing stress introduced by the relatively higher viscosity of the PLA matrix mixed with NPK.



**Fig. 4.** SEM of bioplastic fertiliser (BpF) composites. A (PLA/NPKC1), B (PLA/NPKC2), C (PLA/EFB/NPKC1), and D (PLA/EFB/NPKC2)

In the PLA with added fibres, crack propagation lines were observed on the fracture surface, and the crack grooves were more emphasised than those on the PLA. Under SEM, the PLA-based composites showed debonding of fibres and fertiliser from the matrix. Compared to the fertiliser, the PLA with its polar nature was more compatible with the fibre. To transfer and disperse stress more effectively, the composites require stronger interaction between the components (Herrera and Valadez 2005).

### Biodegradation of Bioplastic Fertiliser (BpF) Composites

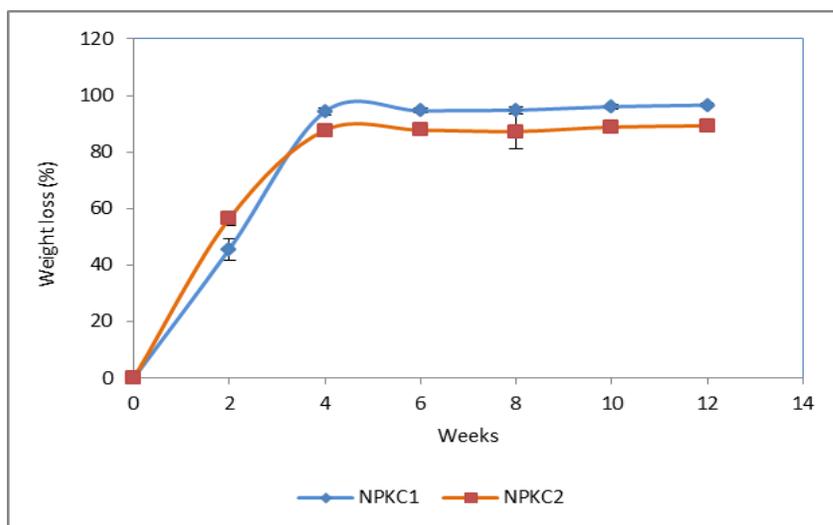
Table 4 and Fig. 5 show that the mean of weight loss values of conditions sharing the same letter were not significantly different at  $P < 0.05$ , NPKC1 (uncoated) and NPKC2 (coated). The weight loss was 45% and 90% after 2 and 4 weeks, respectively. NPKC1 was degraded more than NPKC2.

**Table 4.** Mean and Percentage of Weight Loss of Bioplastic Fertiliser (BpF) Composites from Soil Buried Test

Treatment	Weeks						
	0	2	4	6	8	10	12
NPKC1	0	45.60b (3.77)	94.36a (1.27)	94.60a (0.79)	94.81a (1.27)	96.05a (0.71)	96.62a (0.48)
NPKC2	0	56.45a (2.47)	87.70b (0.75)	87.76b (0.81)	87.27b (6.02)	88.85b (0.81)	89.29b (0.57)

Note: Mean of weight loss in same letter is not significantly different at  $P < 0.05$ . Standard deviation are given in parentheses. NPKC1 (uncoated) and NPKC2 (coated).

The degradation of the fertilisers after 10 weeks was 90 to 96%. After eight weeks and until 10 weeks, the degradation of NPKC1 was higher, with 94.81% to 96.62% weight loss as compared with 87.81% to 89.29% weight loss for the NPKC2. During the soil burial test, the fertiliser swelled and leached through the tea bag. Temperature, moisture content, and water in the soil influenced the degradation and solubility of the fertiliser.

**Fig. 5.** Mean of standard deviation and weight loss NPKC1 (uncoated) and NPKC2 (coated) in soil buried test

The degradation of PLA happened in stages. Initially, water diffused into the material, causing the hydrolysis of ester bonds and the reduction of molecular weight. Then, intracellular uptake of lactic acid oligomers and catabolism occurred. The increase in temperature and water content increased the rates of hydrolysis (Ho *et al.* 1999). This process was also catalysed by the free carboxyl group of the hydrolysed PLA ends. The higher concentration of carboxylic acid inside a thick sample further enhanced the hydrolysis reaction. The weight losses of the bioplastic fertiliser (BpF) composites are shown in Table 5 and Fig. 6. The bioplastic fertiliser composites deteriorated as a function of degradation time. Different composition ratios directly affected the degradation behaviour. The PLA products initially showed the highest weight losses during the soil burial experiment. Then, the weight loss rate was high until week 8 of soil burial time, after which it slowed down.

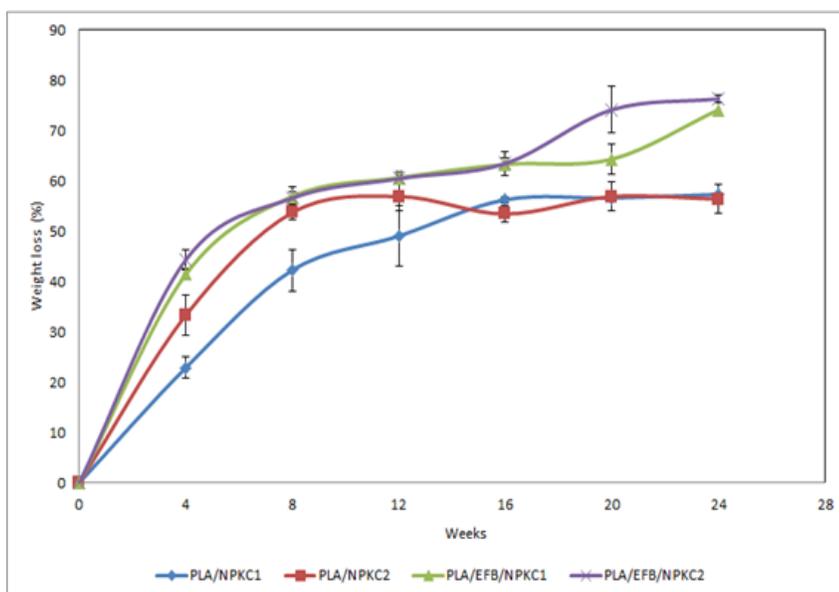
A constant rate of water removal from the structure of the PLA/NPK was observed. After 16 weeks, further degradation of the structure of the BpF composites was observed. Interestingly, it was greater than that of commercial PLA without fibre. Perhaps the higher molecular weight of commercial PLA affected its structure, making it stronger than the PLA composites.

**Table 5.** Mean and Percentage of Weight Loss of Bioplastic Fertiliser (BpF) Composites from Soil Buried Test

Treatment	Weeks						
	0	4	8	12	16	20	24
PLA/NPKC1	0	22.87c (2.08)	42.25b (4.12)	49.04c (5.98)	56.27a (0.63)	56.61b (0.28)	57.31b (0.34)
PLA/NPKC2	0	33.29b (4.02)	53.72a (1.35)	56.91b (2.93)	53.43c (1.63)	56.9b (2.90)	56.38b (2.88)
PLA/EFB/NPKC1	0	41.42a (1.17)	56.91a (1.78)	60.64a (1.24)	63.29a (1.20)	64.32a (3.09)	74.15a (1.32)
PLA/EFB/NPKC2	0	44.31a (1.98)	56.58a (1.34)	60.44a (0.89)	63.46a (2.33)	74.15a (4.66)	76.33a (0.84)

Note: Mean of weight loss BpF composites in same letter is not significantly different at  $P < 0.05$ . Standard deviation are given in parentheses. Poly Lactic acid (PLA), EFB (empty fruit bunch), NPKC1 (uncoated) and NPKC2 (coated)

Figure 6 shows the samples of different degradation stages that were retrieved from the soil. Composting is the simplest test in assessing the biodegradability of composites. The hydrolytic degradation rate of PLA depends highly on the humidity and the temperature of its surrounding environment.



**Fig. 6.** Mean of standard deviation and weight loss of PLA/NPK and PLA/EFB/NPK composites in soil burial test. Note: PLA (Poly Lactic acid), EFB (empty fruit bunch), NPKC1 (uncoated) and NPKC2 (coated)

The PLA polymers were found to degrade rapidly at 30 °C and a high level of humidity; therefore, the temperature was kept at 30 °C and the moisture content of the soil was maintained at 25%. The embrittlement of the samples after only four weeks marked the onset of PLA degradation.

Compared to pure PLA, the degradation of the composites occurred at a slower rate. It was assumed that the water uptake and diffusion in the composites experienced some resistance compared to pure PLA, hence the slower rate of degradation for the composites. After 24 weeks in the soil, both the PLA and the composites experienced significant weight loss. Figure 6 shows the residual weight percentage against time (in weeks) in the soil. The composites showed significant degradation after 16 weeks, especially for PLA/EFB/NPKC2, followed by PLA/NPKC2 and PLA/EFB/NPKC1. PLA/NPKC1 had the lowest rate of degradation compared to the other three composites.

Water that diffused into the polymer samples caused the samples to swell, enhancing the biodegradation process. It can be concluded that the PLA/EFB/NPKC1 and PLA/EFB/NPKC2 were more easily penetrated by water and thus more biodegradable than the PLA/NPKC1 fertiliser composites. The percentage of weight loss increased for the composites with added EFB fibres, *i.e.*, PLA/EFB/NPKC1 and PLA/EFB/NPKC2. Composites with the added 10% EFB fibres showed rapid degradation in the initial eight weeks of biodegradation, with a weight loss roughly equivalent to the weight of the EFB content. The PLA/EFB/NPK experienced approximately a 3 to 9 wt. % weight loss. The hydrophilic characteristic of the fibre made the composite absorb more water from the surrounding surface through cracked lines and holes of the BpF fertiliser composites (Fig. 6). After 16 weeks in the soil, the PLA/EFB/NPKC1 and PLA/EFB/NPKC2 showed an increase in weight loss because the EFB fibres swelled and were degraded while the fertiliser was being slowly released. After 24 weeks of the soil burial test, the weight losses of PLA/EFB/NPKC1 and PLA/EFB/NPKC2 were recorded to be 64.32% and 76.33%, respectively.

## CONCLUSIONS

1. In this study, a new technique for processing PLA using extrusion to produce bioplastic fertiliser (BpF) composites was applied.
2. The quality of the PLA composites was found to depend on the quality of dispersion of the fertiliser and filler material in the PLA matrix, as well as the interaction between the surfaces of the composite components. The compounded fractured surface observed between the EFB fibre and the NPK fertiliser indicated weak adhesion between the surfaces as well as poor stress transfer, which then led to complete debonding from the PLA matrix.
3. Composites composed of 10 wt.% oil palm EFB fibre achieved the greatest total weight loss after 24 weeks. In general, the hydrophilic nature of EFB fibres discouraged adhesion to the matrix polymers, which are hydrophobic in nature, leading to poor fibre wetting during processing. This problem then caused void formation in the matrix.

- The fibres in the bioplastic fertiliser composites caused an increase in biodegradation rate. Bioplastic fertiliser (BpF) composites can be applied in the agriculture and horticulture sectors, and they are also environmentally friendly.

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