Properties of Slow Release Fertilizer Composites Made from Electron Beam-irradiated Poly(Butylene Succinate) Compounded with Oil Palm Biomass and Fertilizer

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Electron beam irradiation at certain absorption doses can affect the chain scission and crosslinking of poly(butylene succinate) (PBS) molecules, as well as their thermal properties. In this study, slow release fertilizer composites were produced by compounding neat PBS with NPK fertilizer and oil palm empty fruit bunch using a twin-screw extrusion method. It was found that granular PBS irradiated with up to 50 kGy remarkably improved the bonding and dispersion of the PBS matrix. The subsequent experiment also showed that the biodegradation of slow release fertilizer composites in soil could be improved *via* electron beam irradiation.

Keywords: Biodegradable polymer; Electron beam irradiation; Oil palm biomass; Composites; Degradation

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INTRODUCTION

The world has seen an increased awareness regarding public health, natural resource sustainability, and environmental protection. Every industry is working towards prioritizing these aspects. In agriculture, scientists are trying to produce more environmentally friendly fertilizers that can reduce nutrient loss in leaching or run-off, while at the same time increasing the yield. Even though soil fertilization and the production of slow release fertilizers (SRFs) has been increasing, the nutrient use efficiency (NUE) is still relatively low. The assimilation of nitrogen and potassium by crops in the first year of using of a SRF is estimated to be approximately 50% to 60%, and the percentage of phosphorus assimilation is in the range 10% to 25%. The coming years will probably see a reduction of 1% to 2% in the assimilation of these nutrients per year (Trenkel 2010). Such a low efficiency in nutrient assimilation affects the environment and may jeopardize the well-being of humans and animals (Newbould 1989). Additionally, a low NUE adversely affects the economic aspect of agriculture through material losses, wasted labor, and energy losses (Cruse *et al.* 2010). To counter the problem, application of polymer-coated controlled-release-fertilizers (PC-CRFs) is on the rise nowadays. According to Majeed *et al.* (2015), polymer-coated controlled-release-fertilizers (PC-CRFs) are the type of organic and inorganic encapsulated fertilizers coated with various types of polymeric materials that provide a better means of targeted delivery of controlled-fertilizer supply for the agricultural crops.

Natural fibers are renewable, as well as environmentally friendly, and therefore the demand for them has increased in recent years. They also have a promising use as reinforcements in polymer composites (Jawaid *et al.* 2011). Oil palm empty fruit bunch is one of these natural fibers. These fibers are considered waste from the palm oil industry (Li and Sun 2010). Some studies have used them as reinforcement materials for polymer matrices (Bledzki and Gassan 1999; Rozman *et al.* 2000; Alam *et al.* 2011). However, because of their limitations, such as a low thermal stability, poor polymer–matrix compatibility, and high moisture affinity, the process must be optimized to improve the adhesion of the fiber matrix and increase the quality of the composites.

Poly(butylene succinate) (PBS) is a type of aliphatic thermoplastic polyester. It is a well-known biodegradable polymer that having superior materials properties compared to that of the prevalently used polyethylene (PE) which produced large amount of greenhouse gases during decomposing (Henke *et al.* 2017; Hong *et al.* 2017). It has good mechanical properties, a low processing temperature, and most importantly, is biodegradable (Liu *et al.* 2009). The synthesis of PBS involves a reaction between aliphatic succinic acid and glycol 1,4-butanediol (Kim *et al.* 2005). Its degradation may occur via hydrolysis because of the breaking down of polymer structures metabolized by microorganisms or biological enzymes, which forms water and carbon monoxide (Wang *et al.* 2009; Ahmad Thirmizir *et al.* 2011a). This degradation process can happen in various environments, including in seawater, moist soil, or even compost (Kim *et al.* 2006). Because of this property, PBS can potentially be used to manufacture disposable plates and cups, packaging films, and landfill covers, as well as in other similar applications. Regardless of its useful properties, its high cost makes it less favorable in the industry. Therefore, by incorporating relatively low-cost natural fibers into PBS composites, such as oil palm empty fruit bunch (EFB), it is hoped that the price can be reduced (Ahmad Thirmizir *et al.* 2011b).

Although PBS possess many advantageous characteristics, a main drawback that limits the application and processing of PBS is its low thermal resistance. In addition, weak adhesion between EFB fiber and NPK fertilizer is also a main problem in the production of bioplastic composites. Saffian *et al.* (2017) produced bioplastic fertilizer (BpF) composites made of poly(butylene succinate) blended with oil palm biomass and fertilizer and revealed that complete debonding from the PBS matrix happened as a result of weak adhesion between EFB fiber and NPK fertilizer. However, it can be improved by using ionizing radiation such as electron radiation beam to crosslinking new composition (Kim *et al.* 2015). Electron beam irradiation (EBI) is an environmental friendly treatment that aims to enhance the properties of polymer materials such as fibres, films, and composites (Then *et al.* 2014). Gamma rays can affect the chain scission and crosslinking of poly(butylene succinate) (PBS) (Suhartini *et al.* 2003). Electron beam irradiation of a polymer can cause competitive occurrences of chain scission and crosslinking of the polymer molecules. The degree of such an effect depends on the polymer type and electron beam energy (Loo *et al*. 2005; Quynh *et al.* 2007). Then *et al.* (2015) produced biocomposite with alkali treated oil palm mesocarp fiber and PBS and reported that the mechanical and physical properties of the resultant biocomposite is superior that of the untreated oil palm mesocarp fiber.

Promising progress has been achieved in recent years in understanding the mechanisms of polymer-coated SRFs and quantifying the nutrients that are released (Shaviv 2001; Shaviv *et al.* 2003a). Most of the slow release agents that are currently sold in the market are made of non-biodegradable polymer emulsions or spraying, but

compounding techniques are seldom applied (Shaviv *et al*. 2003b). In this work, tests were done to assess the effects of electron beam irradiation on the properties of slow release fertilizer composites made from EFB, irradiated PBS, EFB, and NPK fertilizer compounded *via* an extrusion method to produce SRF composites. The degradation data of the SRF composites after a soil burial test, including the melting point index, thermal properties, and degradation, was analyzed using an analysis of variance (ANOVA) performed by SAS software 9.4 (Kuala Lumpur, Malaysia).

EXPERIMENTAL

Materials

The PBS polymer used in this study was acquired from Innovative Pultrusion Sdn. Bhd. (Senawang, Malaysia). The EFB was obtained from Poly Composite Sdn. Bhd. (Teluk Intan, Malaysia), while the commercial slow-release NPK fertilizers were acquired from Diversatech Fertilizer Sdn. Bhd (Bangi, Malaysia).

Preparation of Electron Beam Irradiation

Prior to the irradiation procedure, all of the granulated PBS samples were put in polyethylene bags, pressed, and vacuum-sealed to remove the air. The bags were put on a moving conveyor belt at a speed of 0.3 m/min to 1.2 m/min. The PBS samples were irradiated at dosages of 0 kGy, 10 kGy, 20 kGy, 30 kGy, 40 kGy, and 50 kGy *via* electron beam (EB) irradiation using an EB machine (EPS Model-3000kV, Nissin High Voltage Co. Ltd., Kyoto, Japan). The dose rate, beam current, and acceleration energy were 10 kGy/pass, 2 mA, and 2 MeV, respectively.

Sample Preparation of the SRF Composites

Compounding of the materials was done using a twin-screw extruder. Irradiated and non-irradiated PBS were used in the extrusion process for the preparation of SRF composites. In this study, irradiated PBS (50 kGy dose), EFB fibers, and NPK fertilizer were used to produce SRF composites.

Table 1. Formulation of the Irradiated Bioplastic Fertilizer Composites

Composite	PBS Matrix (%)	EFB Fiber (%)	NPK Fertiliser (%)
NPK	0	0	100
PBS/NPK	40	0	60
PBS-EB/NPK	40	0	60
PBS/EFB/NPK	30	10	60
PBS-EB/EFB/NPK	30	10	60

Note: EB - electron beam-irradiated

Table 1 shows the formulations of the irradiated SRF composites. A drying oven at a temperature of 103 °C ± 2 °C was used to dry all of the materials overnight. The extruder screw was set at a constant speed of 50 rpm, while the screw rotating speed was maintained at 80 rpm. The temperature of the reverse barrel was 130 °C, while the temperatures used in the hopper for molding were 110 °C and 130 °C. The process was completed in

approximately 15 min and the PBS, fertilizer granules, and EFB fibers were compounded homogeneously.

Melt Flow Index

A melt flow index (MFI) instrument (Toyoseiki Co., Tokyo, Japan) was used to measure the melt index of the irradiated and non-irradiated PBS composites. At 190 °C, the MFI of the samples was measured for a mass of 2.16 kg. Six replicates of each sample were obtained, and the unit of measurement was grams per 10 min. The extrusion and cutting began after the polymers were kept in the barrel for 7 min. An applied weight of 2.16 kg was used for extrusion from the MFI die.

Thermogravimetric Analysis

The thermal properties analysis was done using a thermogravimetric analyzer (TGA; Q500 TA Instruments, New Castle, DE, USA). The amount of sample used in each analysis was 10 mg. The temperature range was 35 °C to 600 °C with a heating rate of 10 °C/min. The nitrogen flow rate was 20 mL/min. The weight reduction was plotted against the temperature.

Scanning Electron Microscopy

The sample surface analysis was conducted using scanning electron microscopy (SEM) (SEM-3400N, Hitachi, Japan) with a cathode acceleration voltage of 15 kV. The gold coating of the samples was done using an Emitech K550X coater (Kent, UK) (600 s and 35 mA) at a pressure of 0.2 bar.

Biodegradation of the SRF Composites in the Soil Burial Test

A simple soil burial test was used to test the biodegradability of the SRF composites. To simulate a natural soil environment, natural soil that did not contain any enzyme activity or composting materials was obtained. Samples were buried in polybags containing natural soil for a total of 24 weeks. The samples were recovered and analyzed every 4 weeks. To obtain a constant dry weight, the moisture was removed from the samples using a vacuum oven at 80 °C. The biodegradability of each sample was measured based on its weight loss after the soil burial test using Eq. 1,

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

where W_0 is the weight before the test (g) and W_1 is the weight after the test (g).

Leaching of Nitrogen Using Soil Column Method

Topsoil from surface to 15 cm depth was collected from University Agriculture Park, Universiti Putra Malaysia. The collected topsoil was tested for physical and chemical properties in Soil Department of Faculty of Agriculture, UPM. The results indicated that the topsoil had a density of 1.031 g/cm^3, with the chemical composition content of N = 0.14%, P = 6.15%, K = 46.17%. Mg = 43.20%, C = 1.63%, and S = 0.02%. The soil columns made of acrylic material with a 30 mm internal diameter were prepared. Each column was filled with 3 kg air-dried soil, occupying a height of about 400 mm. The SRF composites were inserted to a depth of 100 mm of the soil columns. After the soil in columns was saturated with water, they were kept in a controlled temperature room at 30 ± 1°C. The soil in columns was rinsed with 100 mL of distilled water in 4, 8, 12, 16, 20, and 24 weeks, and the leachates from column were collected. 10 mL of collected leachate

was analyzed for nitrogen at Soil Department of Faculty of Agriculture using TruMac CNS, Version 1.1x, LECO, USA.

RESULTS AND DISCUSSION

Melt Flow Index

The MFI values of the PBS after being irradiated from 0 kGy to 50 kGy are shown in Fig. 1. The results show that the MFI of the PBS irradiated samples increased from 8.40 MVR at 0 kGy to 16.0 MVR at 50 kGy. The results agreed with Kim *et al.* (2015) who reported that irradiation at 50 kGy resulted in the most enhancement in thermal and mechanical properties of PBS. Therefore, the PBS in this study was irradiated PBS with 50 kGy dose.

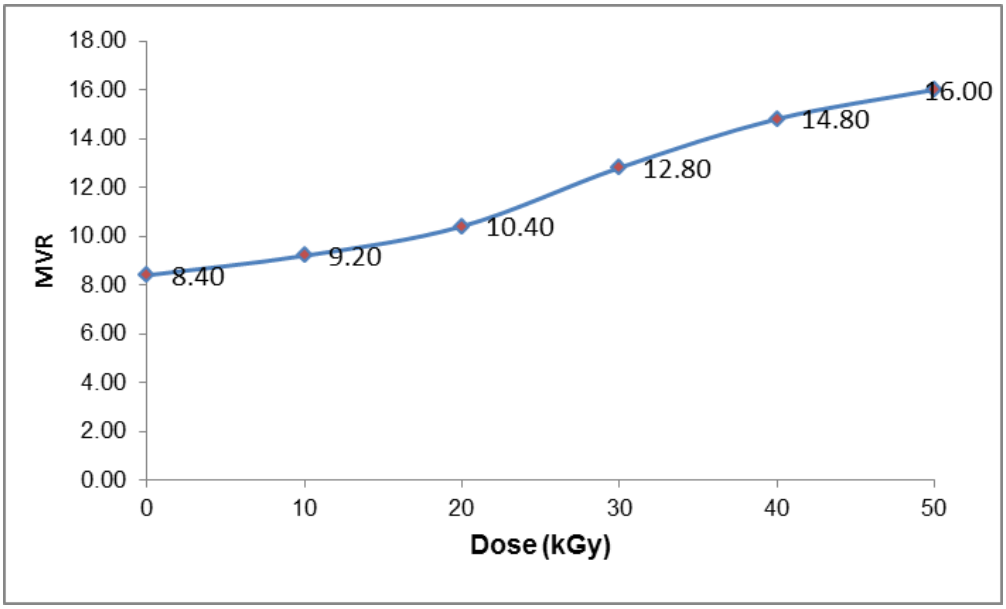

Fig. 1. MFI of the irradiated PBS

Although EB irradiation can result in cross-linking (*i.e.* an increase in the molecular weight) and scission (*i.e.* a reduction in the molecular weight) at the same time, the dominant process determines the end result of the irradiation process. It was observed that the MFI of the PBS increased after the irradiation treatment because of an increase in the occurrence of molecular scission as the dosage increased. A lower molecular weight caused easier material flow and produced an increase in the melting rate of the PBS biopolymer. The melt flow properties of materials can be influenced by increasing the radiation dose. Degradation can be achieved by several means, such as chain scission, oxidative degradation, depolymerisation, and trans-esterification. Thermal degradation can be further influenced by residual catalysts, reactive end groups, and unreacted starting monomers, as well as impurities (Carrasco *et al.* 2010).

Thermogravimetric Analysis

Table 2 shows the initial and final weights of the pure PBS, irradiated PBS, EFB, and SRF composites. The weight loss versus temperature curve is shown in Fig. 2. The

degradation temperature range of the composites was 244.0 °C to 412.6 °C. The lower degradation temperature range of the composites (EFB, PBS/NPK, PBS-EB/NPK, PBS/EFB/NPK, PBS-EB/EFB/NPK) compared with that of the PBS and PBS-EB was attributable to the low degradation temperatures for the other components in the composites. The EFB fiber degraded within the temperature range of 250 °C to 350°C and the fertilizer degraded within the temperature range of 50 °C to 375°C (Saffian *et al.* 2016).

Table 2. Peak Temperature and Percentage Weight Loss of the Pure PBS, Irradiated PBS, PBS/NPK, and PBS/EFB/NPK Composites

Composite	T_{IDT} (°C)	T_{FDT} (°C)	Residual (%) (at 550 °C)
PBS	338.56	411.02	1.68
PBS-EB	342.76	398.10	-
EFB	251.18	342.82	23.11
PBS/NPK	279.95	412.62	26.37
PBS-EB/NPK	271.48	366.24	30.32
PBS/EFB/NPK	282.62	407.83	29.92
PBS-EB/EFB/NPK	243.96	361.71	34.11

T_{IDT}: initial decomposition temperature; T_{FDT}: final decomposition temperature

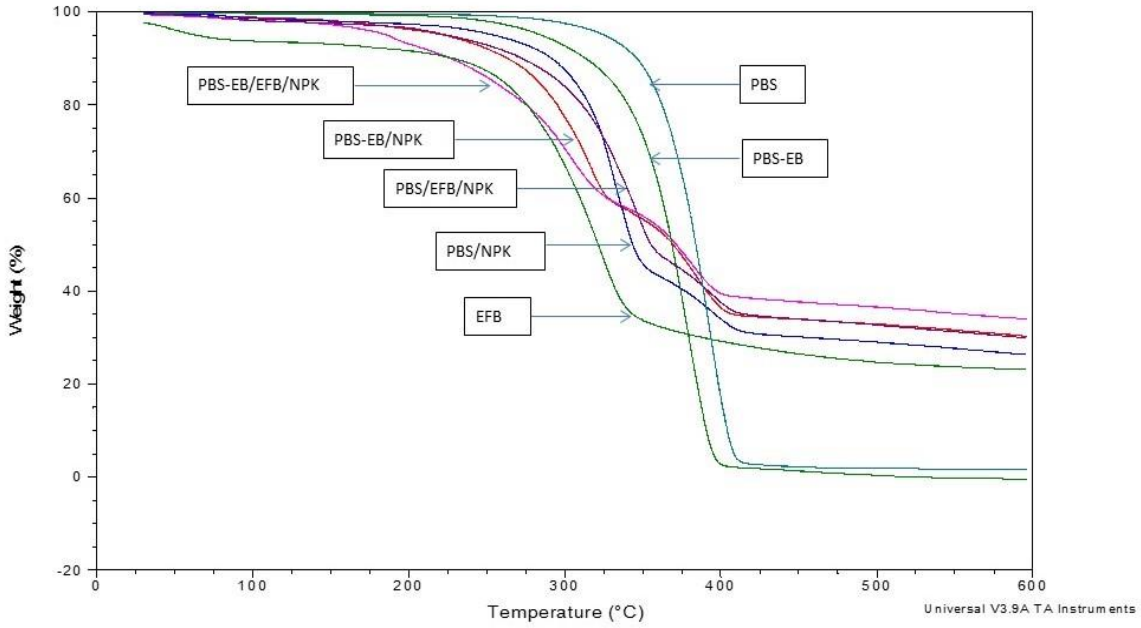

Fig. 2. TGA thermograms showing the thermal properties of pure PBS, EFB, PBS-EB, PBS/NPK, PBS/EFB/NPK, PBS-EB/NPK and PBS-EB/EFB/NPK composites

The thermal stabilities of the irradiated and non-irradiated PBS were comparable, and were 342.8 and 338.6 °C, respectively. However, the irradiated PBS had a lower capability to protect the NPK fertilizer compared with the non-irradiated PBS. The initial decomposition temperature of the PBS-EB/NPK composite was lower compared with the PBS/NPK composite, and were 271.5 °C and 278.0 °C, respectively. Similarly, the final decomposition temperature of the PBS-EB/NPK composite was lower compared with the PBS/NPK composite, and were 366.2 and 412.6 °C, respectively. Likewise, the thermal stability of the PBS-EB/EFB/NPK composite was reduced by 38.7 °C for the initial

degradation temperature and by 46.1 °C for the final degradation temperature, when compared with those of the PBS/EFB/NPK composite. Therefore, it was concluded that the irradiation of PBS at 50 kGy reduced the thermal stability of the SRF composites.

Scanning Electron Microscopy

A SEM analysis was performed on the surfaces of the PBS irradiated composites for direct observation of the surface structure of the SRF composite. The SEM micrographs of the surface of the non-irradiated and irradiated PBS composites are shown in Figs. 3a and 3b, respectively. The smooth surface of the irradiated PBS composites indicated that the components were well dispersed within the composites. Additionally, the SEM micrographs of the fractured PBS presented in Figs. 3c and 3d show visible gaps at the interface between the PBS, EFB fibers, and NPK fertilizers. Poor interfacial adhesion resulted from the incompatibility between the hydrophilic EFB fiber and hydrophobic PBS.

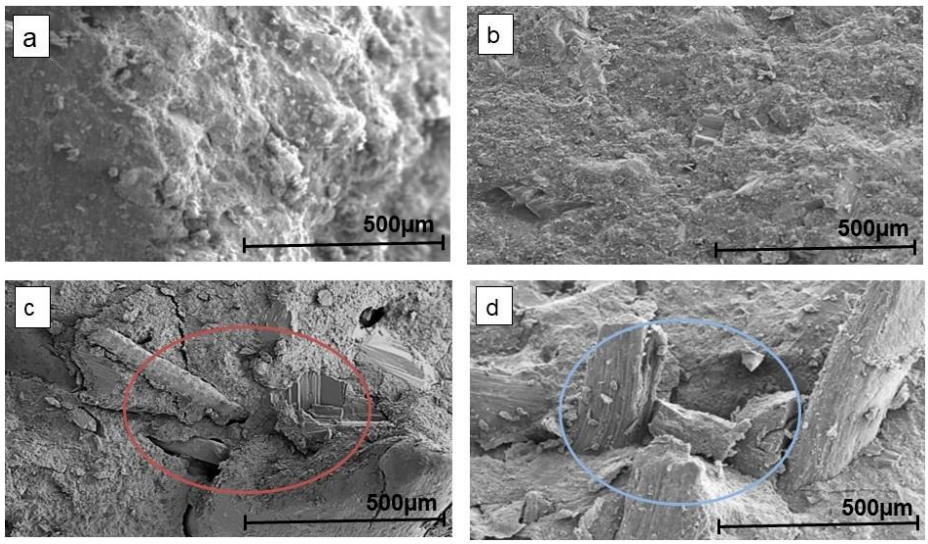

Fig. 3. SEM micrographs of the irradiated PBS, EFB, and NPK composites: a) PBS/NPK, (b) PBS-EB/NPK, (c) PBS/EFB/NPK, and (d) PBS-EB/EFB/NPK

It was noted that, for the non-irradiated PBS, the surface of the EFB fiber was relatively clean without PBS adhered to the fracture of the composite, which indicated that their interphase bonding was relatively poor. Reduced gaps between the irradiated PBS, EFB fibers, and NPK fertilizers can be seen in Fig. 3d, which shows that the incorporation of irradiated PBS resulted in better adhesion with the EFB fibers and NPK fertilizer. Based on the cracks on the fibrous end, the interphase bonding was stronger. This was evident from the observation that the fiber was not pulled out during fracture, but instead underwent breakage. The irradiated PBS can be seen adhering to the surface of the EFB fibers with a layer of the matrix materials covering the fibers. These observations showed that the interfacial strength of the PBS composites had been improved through the chain scission and crosslinking processes caused by irradiation.

Biodegradation of the SRF Composites in the Soil Burial Test

A simple soil burial test was used to test the biodegradability of the SRF composites. Samples were buried in natural soil for a total of 24 weeks. Five replicate samples were recovered and analyzed every 4 weeks. The data are presented in Fig. 4 and

Table 3. Figure 4 shows that within a period of 12 weeks, the degradation of the PBS-EB/NPK and PBS-EB/EFB/NPK composites was faster than that of the other SRF composite samples. The PBS-EB/NPK composite experienced a weight loss of 65.5%, while the PBS-EB/EFB/NPK composite had a weight loss of 75.5%.

Table 3. Weight Loss of the Bioplastic Fertilizer Composites during the Soil Burial Test

Composite	Week						
	0	4	8	12	16	20	24
PBS/NPK	0	28.95%^{c} (3.99%)	39.2%^{b} (12.82%)	44.20%^{b} (6.33%)	57.83%^{a} (3.51%)	56.99%^{a} (3.79%)	54.01%^{a} (8.40%)
PBS-EB/NPK	0	45.77%^{e} (6.10%)	52.74%^{d} (4.59%)	55.69%^{d} (2.25%)	58.92%^{bc} (5.22%)	63.22%^{ab} (2.14%)	65.52%^{a} (2.72%)
PBS/EFB/NPK	0	38.39%^{e} (3.71%)	58.66%^{d} (1.09%)	50.31%^{c} (2.21%)	59.59%^{c} (0.48%)	64.37%^{b} (1.88%)	72.68%^{a} (3.93%)
PBS-EB/EFB/NPK	0	55.24%^{e} (1.37%)	56.27%^{e} (2.08%)	60.57%^{d} (2.68%)	65.01%^{c} (2.77%)	68.43%^{b} (0.63%)	75.54%^{a} (2.62%)

Mean of the weight loss of the composites with the same letter is not significantly different at $P < 0.05$; Standard deviation is given in parentheses; Five replicates were made for each sample

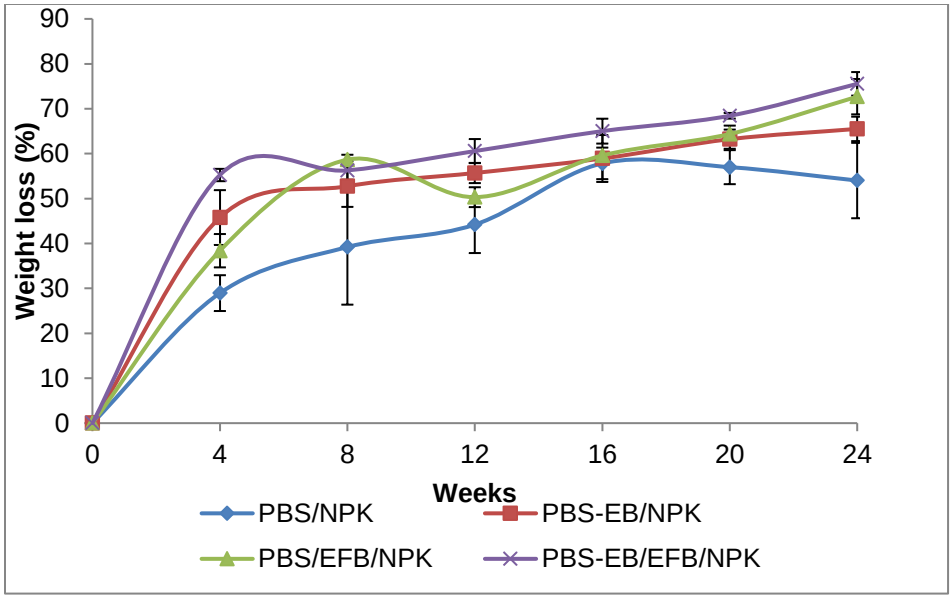

Fig. 4. Mean of the standard deviation and weight loss of the PBS/NPK, PBS-EB/NPK, PBS/NPK/EFB, and PBS-EB/EFB/NPK composites from the soil burial test

The findings were in agreement with Benyathiar *et al.* (2016), who reported that the poly(lactic) acid (PLA) irradiated with electron beam had an increased biodegradation rate compared to that the non-radiated commercial PLA film. Nugroho *et al.* (2001) suggested that chain-scission reaction is the dominant reaction when the polymers are exposed to radiation in the presence of air. Excitement followed by scission of weaker bonds in the polymer's macromolecules occurred as the energy absorbed by the exposed

plastic materials. As a result, the molecular weight of the samples decreased and led to higher biodegradation rate (Hamilton *et al.* 1996). Although some crosslinking reaction might occur due to the irradiation, subsequently leading to a lower degradation rate. The reaction will eventually dominate by chain scission after some period of time, eventually increasing the biodegradation rate.

In comparison, composites that contain EFB, which are PBS-EB/EFB/NPK and PBS-EB/EFB/NPK, exhibited higher biodegradation rate. These composites absorbed more water from the surroundings because of the hydrophilicity of the fiber, as well as the holes and cracked lines in the SRF composites, which are shown in Fig. 3. The EFB fibers swelled and degraded further after 16 weeks in the soil, which increased the weight loss of the PBS-EB/NPK and PBS-EB/NPK/EFB composites, and slowly released the fertilizer. Microorganisms are said to be the main cause for the degradation of cellulose and lignocellulosic components (Ishiaku *et al.* 2002). The fiber itself possessed a high degree of degradability because of its cellulosic and lignocellulosic components. Therefore, the fiber content determined the overall biodegradability of the SRF composites in the soil environment. The presence of water assisted hydrolysis at the ester linkages, and allowed bacteria and fungi in the soil to degrade the PBS (Kim and Rhee 2003). The hydrolytic depolymerization of the cellulose materials *via* microorganisms was also involved in the degradation of the EFB fibers to monomeric glucose units.

Leaching of Nitrogen

Figure 5 shows the mean cumulative of N releases from SRF composites. The curves show that the release pattern was slow and steady but gradually increased with the passage of time. Release of N in all types of SRF composites approached equilibrium after 12 weeks, and it continued to be released in small amounts. A different release pattern was observed for all types of SRF composite, with the irradiated samples showing higher release rates compared to that of the non-irradiated samples. The pattern of release was correlated well to the biodegradability of the SRF composites, as shown in Fig. 4, where the irradiated samples degrade and release N faster than that of the non-irradiated samples.

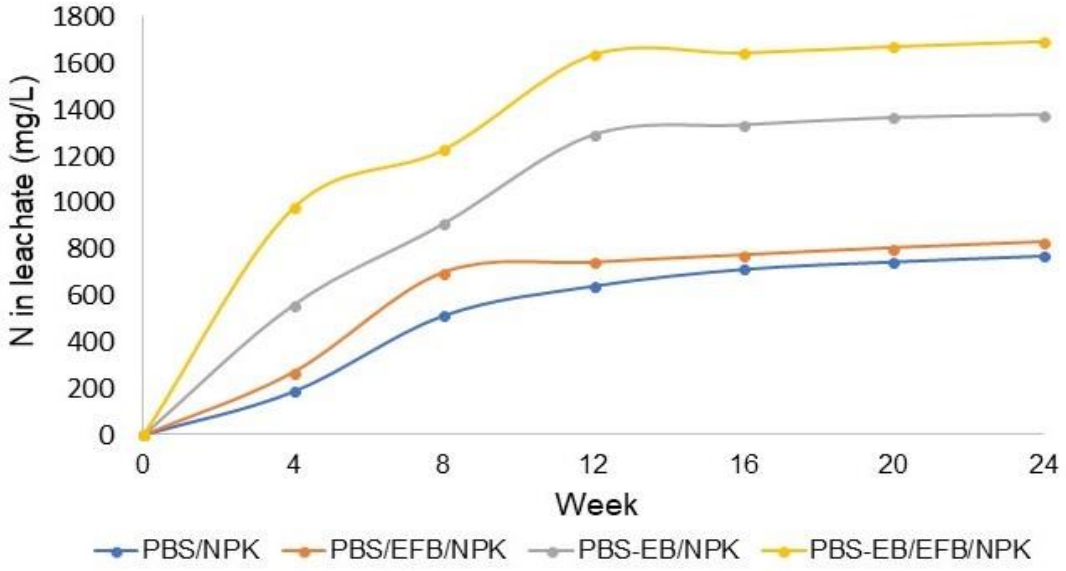

Fig. 5. The leaching of nitrogen from PBS/NPK, PBS-EB/NPK, PBS/NPK/EFB, and PBS-EB/EFB/NPK composites from the soil burial test

Different nutrient release patterns can be applied for different applications depending on the nutrient demands of the cultivated crops. Crops that require a persistent level of nutrients over a long period of time can be fertilized with fertilizer with slower release pattern that can reside a relatively longer time in soil. Meanwhile, short duration plants can benefit from a fertilizer that shows high N release at an early stage (Abdul Khalid *et al.* 2015).

CONCLUSIONS

1. A new approach to produce slow-release fertilizers (SRFs) was conducted using irradiated poly(butylene succinate) (PBS) *via* an extrusion method. The SRF composites with non-irradiated PBS were observed to have weak adhesion and poor stress transfer, which led to the complete debonding of the empty fruit bunch (EFB) fibers and nitrogen-phosphorus-potassium (NPK) fertilizers from the PBS matrix.
2. After an irradiation treatment of 50 kGy on the PBS, the bonding improved. After 12 weeks, the PBS-EB/NPK and PBS-EB/EFB/NPK composites degraded more, which was determined based on the weight loss, as well as the observed cracked lines and holes on their surfaces. The highest total weight loss in the biodegradation test was reached at the end of the test period (24 weeks).
3. Generally, EFB fibers are hydrophilic in nature, while the matrix polymer is hydrophobic. The mixture of these two materials causes weak adhesion and poor fiber wetting, which leads to faster biodegradation. The irradiation of PBS prior to mixing improved the adhesion and increased the biodegradation rate compared with the non-irradiated SRF composites.

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REFERENCES CITED

Abdul Khalid, N. N., Ashaari, Z., Mohd Haniff, A. H., Mohamed, A., and Lee, S. H. (2015). "Treatability of oil palm frond and rubber wood chips with urea for the development of slow release fertiliser," *J. Oil Plam Res.* 27(3), 220-228.

Ahmad Thirmizir, M., Mohd. Ishak, Z. A., Mat Taib, R., Rahim, S., and Mohamad Jani, S. (2011a). "Kenaf-bast-fiber-filled biodegradable poly(butylene succinate) composites: Effects of fiber loading, fiber length, and maleated poly(butylene succinate) on the flexural and impact properties," *J. Appl. Polym. Sci.* 122(5), 3055-3063. DOI: 10.1002/app.34046

Ahmad Thirmizir, M., Mohd. Ishak, Z. A., Mat Taib, R., Rahim, S., and Mohamad Jani, S. (2011b). "Natural weathering of kenaf bast fibre-filled poly(butylene succinate)

composites: Effect of fibre loading and compatibiliser addition," *J. Polym. Environ.* 19(1), 263-273. DOI: 10.1007/s10924-010-0272-2

Alam, A. K. M. M., Shubhra, Q. T. H., Barai, S., Islam, M. R., and Rahman, M. M. (2011). "Preparation and characterization of natural silk fiber-reinforced polypropylene and synthetic E-glass fiber reinforced polypropylene composites: A comparative study," *J. Compos. Mater.* 45(22), 2301-2308. DOI: 10.1177/0021998311401082

Benyathiar, P., Selke, S., and Auras, R. (2016). "The effect of gamma and electron beam irradiation on the biodegradability of PLA films," *J. Poly Environ.* 24, 230-240. DOI: 10.1007/s10924-016-07660-7

Bledzki, A. K., and Gassan, J. (1999). "Composites reinforced with cellulose based fibres," *Prog. Polym. Sci.* 24(2), 221-274. DOI: 10.1016/S0079-6700(98)00018-5

Carrasco, F., Pagès, P., Gámez-Pérez, J., Santana, O. O., and Maspoch, M. L. (2010). "Processing of poly(lactic acid): Characterization of chemical structure, thermal stability and mechanical properties," *Polym. Degrad. Stabil.* 95(2), 116-125. DOI: 10.1016/j.polymdegradstab.2009.11.045

Cruse, I., Liebman, M., Raman, D. R., and Wiedenhoeft, M. H. (2010). "Fossil energy use in conventional and low-external-input cropping systems," *Agron. J.* 102(3), 934-941. DOI: 10.2134/agronj2009.0457

Hamilton, J. V., Greer, K. W., Ostiguy, P., and Pai, P. N. (1996). "Anisotropic properties in ultrahigh-molecular-weight polyethylene after Cpbalt-60 irradiation," in: R. L. Clough, and S. W. Shalaby (eds.), *Irradiation of Polymer*, American Chemical Society, Washington.

Henke, L., Zarrinbakhsh, N., Endres, H., Misra, M., and Mohanty, A. K. (2017). "Biodegradable and bio-based green blends from carbon dioxide-derived bioplastic and poly(butylene succinate)," *J. Polym. Environ.* 25, 499-509. DOI: 10.1007/s10924-016-0828-x

Hong, G., Meng, Y., Yang, Z., Cheng, H., Zhang, S., and Song, W. (2017). "Mussel-inspired polydopamine modification of bamboo fiber and its effect on the properties of bamboo fiber/polybutylene succinate composites," *BioResources* 12(4), 8419-8442

Ishiaku, U. S., Pang, K. W., Lee, W.S., and Ishak, Z. A. M. (2002). "Mechanical properties and enzymic degradation of thermoplastic and granular sago starch filled polycaprolactone," *Eur. Polym. J.* 38(2), 393-401. DOI: 10.1016/S0014-3057(01)00125-2

Jawaid, M., Abdul Khalil, H. P. S., and Abu Bakar, A. (2011). "Woven hybrid composites: Tensile and flexural properties of oil palm-woven jute fibres based epoxy composites," *Mat. Sci. Eng. A-Struct.* 528(15), 5190-5195. DOI: 10.1016/j.msea.2011.03.047

Kim, B. K., Cho, D., Kwon, O. H., Park, W. H., and Lee, J. (2015). "Effects of electron beam irradiation on the gel fraction, thermal and mechanical properties of poly(butylene succinate) crosslinked by multi-functional monomer," *Mater. Design* 87, 427-435. DOI: 10.1016/j.matdes.2015.08.046

Kim, D. Y., and Rhee, Y. H. (2003). "Biodegradation of microbial and synthetic polyesters by fungi," *Appl. Microbiol. Biot.* 61(4), 300-308. DOI: 10.1007/s00253-002-1205-3

Kim, H. S., Kim, H. J., Lee, W. J., and Choi, I. G. (2006). "Biodegradability of bio-flour filled biodegradable poly(butylene succinate) bio-composites in natural and compost soil," *Polym. Degrad. Stabil.* 91(5), 1117-1127.

DOI: 10.1016/j.polymdegradstab.2005.07.002
Kim, H. S., Yang, H.-S., and Kim, H.-J. (2005). "Biodegradability and mechanical properties of agro-flour-filled polybutylene succinate biocomposites," *J. Appl. Polym. Sci*. 97(4), 1513-1521. DOI: 10.1002/app.21905
Li, Y., and Sun, X. S. (2010). "Preparation and characterization of polymer-inorganic nanocomposites by *in situ* melt polycondensation of L-lactic acid and surface-hydroxylated MgO," *Biomacromolecules* 11(7), 1847-1855. DOI: 10.1021/bm100320q
Liu, L., Yu, J., Cheng, L., and Yang, X. (2009). "Biodegradability of poly(butylene succinate) (PBS) composite reinforced with jute fibre," *Polym. Degrad. Stabil.* 94(1), 90-94. DOI: 10.1016/j.polymdegradstab.2008.10.013
Loo, J. S. C., Ooi, C. P., and Boey, F. Y. C. (2005). "Degradation of poly(lactide-*co*-glycolide) (PLGA) and poly(L-lactide) (PLLA) by electron beam radiation," *Biomaterials* 26(12), 1359-1367. DOI: 10.1016/j.biomaterials.2004.05.001
Majeed, Z., Ramli, N.K., Mansor, N., and Man, Z. (2015). "A comprehensive review on biodegradable polymers and their blends used in controlled-release fertilizer processes," *Rev Chem Eng* 31, 69-95. DOI: 10.1515/revce-2014-0021
Newbould, P. (1989). "The use of nitrogen fertiliser in agriculture. Where do we go practically and ecologically?," *Plant Soil* 115(2), 297-311. DOI: 10.1007/978-94-009-1021-8_27
Nugroho, P., Mitomo, H., Yoshii, F., and Kume, T. (2001). "Degradation of poly(l-lactic acid) by γ-irradiation," *Polym. Degrad. Stab* 72, 337-343. DOI: 10.1016/S0141-3910(01)00030-1
Quynh, T. M., Mitomo, H., Nagasawa, N., Wada, Y., Yoshii, F., and Tamada, M. (2007). "Properties of crosslinked polylactides (PLLA & PDLA) by radiation and its biodegradability," *Eur. Polym. J.* 43(5), 1779-1785. DOI: 10.1016/j.eurpolymj.2007.03.007
Rozman, H. D., Lai, C. Y., Ismail, H., and Ishak, Z. A. M. (2000). "The effect of coupling agents on the mechanical and physical properties of oil palm empty fruit bunch-polypropylene composites," *Polym. Int.* 49, 1273-1278. DOI: 10.1002/1097-0126(200011)49:11<1273::AID-PI469>3.0.CO;2-U
Saffian, H. A., Abdan, K., Hassan, M. A., and Ibrahim, N. A. (2017). "Characterization, morphology, and biodegradation of bioplastic fertilizer (BpF) composites made of poly(butylene succinate) blended with oil palm biomass and fertilizer," *Polym. Compos.* 38, 2577-2583. DOI: 10.1002/pc.23849
Saffian, H. A., Abdan, K., Hassan, M. A., Ibrahim, N. A., and Jawaid, M. (2016). "Characterisation and biodegradation of poly(lactic acid) blended with oil palm biomass and fertiliser for bioplastic fertiliser composites," *BioResources* 11(1), 2055-2070. DOI: 10.15376/biores.11.1.2055-2070
Shaviv, A. (2001). "Advances in controlled-release fertilizers," *Adv. Agron.* 71, 1-49. DOI: 10.1016/S0065-2113(01)71011-5
Shaviv, A., Raban, S., and Zaidel, E. (2003a). "Modeling controlled nutrient release from polymer coated fertilizers: Diffusion release from single granules," *Environ. Sci. Technol.* 37(10), 2251-2256. DOI: 10.1021/es011462v
Shaviv, A., Raban, S., and Zaidel, E. (2003b). "Modeling controlled nutrient release from a population of polymer coated fertilizers: Statistically based model for diffusion release," *Environ. Sci. Technol.* 37(10), 2257-2261. DOI: 10.1021/es0205277

Suhartini, M., Mitomo, H., Nagasawa, N., Yoshii, F., and Kume, T. (2003). "Radiation crosslinking of poly(butylene succinate) in the presence of low concentrations of trimethallyl isocyanurate and its properties," *J. Appl. Polym. Sci.* 88(9), 2238-2246. DOI: 10.1002/app.11944

Then, Y. Y., Ibrahim, N. A., Zainuddin, N., Arrifin, H., Wan Yunus, W. M. Z., and Abd Rahman, M. F. (2014). "Effect of electron beam irradiation on the tensile properties of oil palm mesocarp fibre/poly(butylene succinate) biocomposites," *IJAME* 10, 2070-2080. DOI: 10.15282/ijame.10.2014.23.0174

Then, Y. Y., Ibrahim, N. A., Zainuddin, N., Ariffin, H., Wan Yunus, W. M. Z., and Chieng, B. W. (2015). "Static mechanical, interfacial, and water absorption behaviors of alkali treated oil palm mesocarp fiber reinforced poly(butylene succinate) biocomposites," *BioResources* 10(1), 123-136.

Trenkel, M. E. (2010). *Slow- and Controlled-Release and Stabilized Fertilizers: An Option for Enhancing Nutrient Use Efficiency in Agriculture*, International Fertilizer Industry Association (IFA), Paris, France.

Wang, H., Ji, J., Zhang, W., Zhang, Y., Jiang, J., Wu, Z., Pu, S., and Chu, P. K. (2009). "Biocompatibility and bioactivity of plasma-treated biodegradable poly(butylene succinate)," *Acta Biomater.* 5(1), 279-287. DOI: 10.1016/j.actbio.2008.07.017

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