Soil Burial of Polylactic Acid/Paddy Straw Powder Biocomposite

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The objective of this work was to study the biodegradability of polylactic acid (PLA)/paddy straw powder (PSP) biocomposites. Environmental degradation was evaluated by composting the biocomposite samples into the soil. Different techniques, including mechanical tests and scanning electron microscopy (SEM), were used to obtain a view of the degradation that occurred during the soil burial of the biocomposites. Results of the mechanical tests showed that an increasing content of PSP in the biocomposites decreased the tensile strength and elongation at break (EB), while it increased the modulus of elasticity after six months of exposure. Scanning electron microscopy on the surface after soil burial showed that the filler was poorly wetted by the matrix. This explains the reduction in tensile strength and the elongation at break after soil burial. Differential scanning calorimetry results indicated that the crystallinity of the biocomposites increased with longer composting periods.

Keywords: Biodegradable; Biocomposite; Paddy straw powder; Polylactic acid; Soil burial

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INTRODUCTION

Today, much effort has been made to develop degradable materials, largely because of the global environmental problems resulting from petroleum-based polymers. Polyethylene is the largest-volume plastic used for packaging (Hernadez *et al*. 2000); it is highly resistant to environmental degradation and biodegradation. Therefore, a renewable resource-based biopolymer has been investigated to produce a polymer that can undergo environmental degradation (Kumar *et al*. 2010; Tabi and Kovacs 2011).

Polylactic acid (PLA) is one biopolymer that is comparable with polyethylene in terms of the properties. PLA has the advantage of not only being biodegradable, but also being renewable since its raw material, lactic acid, can be generated by microbial fermentation. This polymer can be synthesized from renewable resources, through the polymerization of monomers derived from agricultural resources, such as corn, sugarcane, and sugar beet (Masud *et al*. 2005). Previous studies have shown that the PLA matrix demonstrates good mechanical behavior; however, PLA alone is not sufficient to produce a favorable product (Lee and Wang 2006). Formerly, PLA composites improved the properties of composites through blends with low cost, renewable, and fully biodegradable natural fillers, such as wood flour, starch, lignocellulosic fibers/fillers, and natural cellulose fibers (Oksman *et al*. 2003; Masud *et al*. 2005; Anuar 2007; Salmah *et al*. 2012). PLA has the advantage of being both renewable and biodegradable, since lactic acid is produced

from biological processes. Paddy straw is a common lignocellulosic fiber. It is an agricultural co-product found abundantly in Malaysia, especially in the northern region.

Generally, biodegradation studies are carried out in soil and/or compost to enhance the biodegradation of these materials that occur in the presence of compost. This complex biological environment allows for a high microbial diversity, and therefore, an increase in the degradation potential for polymeric compounds (Itävaara *et al.* 2002; Shogren *et al*. 2003; Wu 2005; Martucci and Ruseckaite 2009). In the near future, discarded biocomposite byproducts will become biocomposite wastes, and these products will break down naturally by the air, moisture, climate, and soil, and disintegrate into the surrounding land. However, as more and more biodegradable materials pile up, there is an increased threat to the environment. To remedy this, during the processing of materials, additives can be included to enhance the rate of biodegradability of biocomposites, depending on the end-uses of the products. Currently, there are a few studies that have reported positives finding when conducting tests on the biodegradability of biocomposites, especially on natural soil burial and weathering (Alvarez *et al.* 2006; Ismail *et al*. 2008; Sam *et al*. 2011).

In view of this, it is of interest to conduct biodegradation studies of biocomposites of natural fiber reinforced PLA. The changes in the biocomposites after soil burial were studied using a tensile test machine and scanning electron microscopy (SEM). The weight losses of the biocomposites during the soil burial test were investigated to study the biodegradation effect from microorganisms and a distinct leaching effect from environmental moisture.

EXPERIMENTAL

Materials

Pelleted PLA (type 4032D) was supplied by Biomer® (Krailling, Germany). The PLA has a specific gravity of 1.24 g/cm^3 20,000 g/mole of molecular weight.. Paddy straw was obtained from paddy fields in Perlis, Malaysia. The paddy straw was ground using a grinder to an average size of 68 µm. The paddy straw powder was dried in the oven at 80 °C (24 h) to eliminate the moisture content before being used for biocomposite preparation.

Methods

Blending of PLA/Paddy straw powder

The formulation of the PLA/PSP biocomposite is shown in Table 1.

Table 1. Formulation of PLA/PSP Biocomposites

Sample	PLA matrix (%wt)	PSP (%wt)
PLA	100	0
PLA-5PSP	95	5
PLA-10PSP	90	10
PLA-15PSP	85	15

PLA: polylactic acid; PSP: paddy straw powder

Biocomposites were prepared in a Haake Polydrive with a Rheomix R600/610 (Thermo Fisher Scientific,USA). Mixing was done at 180 °C at a rotor speed of 50 rpm for

10 min. The PLA was first added into the mixer to start melt mixing. After 2 min, PSP was added, and the mixing was continued for another 8 min. The PLA/PSP biocomposites were compression moulded using a hot press. The compression-molded procedures involved preheating for 3 min, followed by compression (pressure :10 MPa) for 3 min at 180 °C, and subsequent cooling under pressure for 1 min. Moulded samples were cut into dumb-bell shapes according to ASTM D638 (2014), before being exposed to the environment.

Natural soil burial testing

The biodegradability of the PLA/PSP biocomposites was measured during a natural soil burial test which lasted for 6 months. The test was conducted at the Universiti of Malaysia, Perlis, (latitude 6°26'N, longitude 100°14'E) from June 2014 to November 2014. Meteorology data, such as average temperature, rainfall, and relative humidity were recorded from the nearest meteorology station in Mata Ayer (latitude 6 °28 'N, longitude 100° 14 'E). Figures 1 and 2 show the data obtained from the meteorology station, and data from each month was averaged, compiled, and utilized.

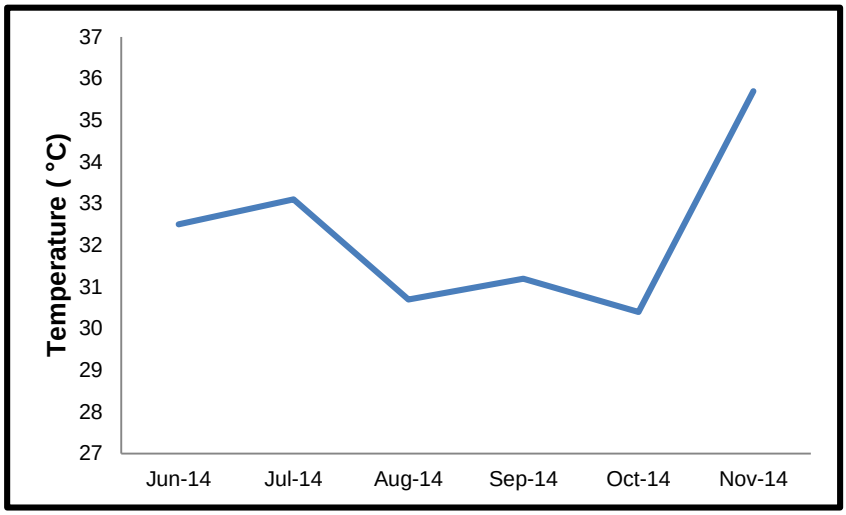
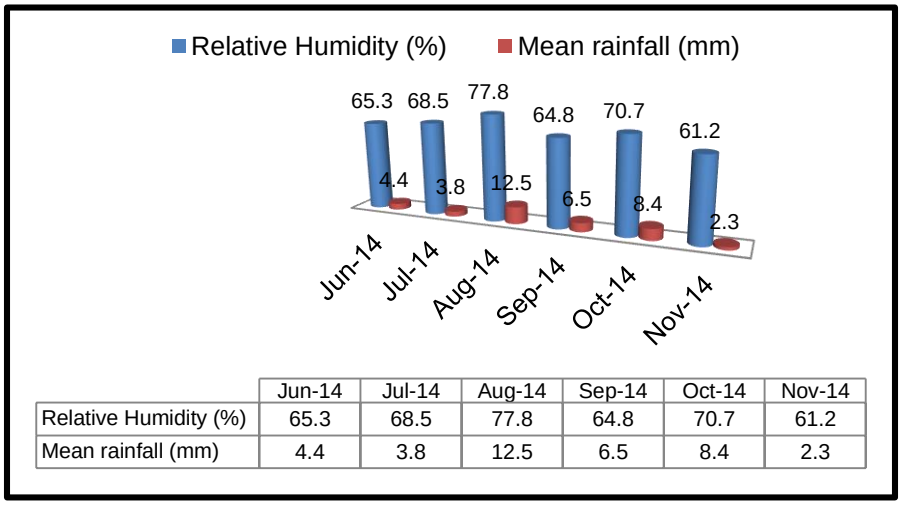


Fig. 1. Variation in the temperature during the natural soil burial test



	Jun-14	Jul-14	Aug-14	Sep-14	Oct-14	Nov-14
Relative Humidity (%)	65.3	68.5	77.8	64.8	70.7	61.2
Mean rainfall (mm)	4.4	3.8	12.5	6.5	8.4	2.3

Fig. 2. Variation in the relative humidity and mean rainfall during the soil burial test

The test was carried out based on a modified method from Kim *et al*. (2005). Samples were prepared, buried, and removed monthly for 6 months. Upon removal, these samples were washed in distilled water and dried in an air drying oven at 70 °C for 24 h before undergoing weight loss and tensile property tests.

Tensile test

Tensile tests were performed using an Instron Universal Testing Machine (Illinois Tool Works Inc., Norwood, MA, USA) at a crosshead speed of 5.0 mm/min. Five measurements were obtained and the final values were determined from the average. The retention of the tensile properties was computed using Eq. 1:

$$\left(\textbf{retention}\ (\%) = \frac{\textbf{Value after degradation}}{\textbf{Value before degradation}}\right) \quad (1)$$

Morphological study

The surface aspects of the exposed areas of the samples were analyzed using a variable pressure field emission scanning electron microscopy (VPFE-SEM; Zeiss SUPRA 35VP). The surfaces of the samples were mounted on aluminum stubs and sputter coated with a thin layer of gold to avoid electrostatic charging and poor resolution during examination.

Weight loss

After exposure to the natural soil burial test, the samples were rinsed thoroughly using distilled water and dried to a constant weight in the oven. The weight loss percentage was calculated using Eq. 2,

$$\textbf{Weight loss}\ (\%) = \frac{Wi - Wf}{Wi}\textbf{X 100} \quad (2)$$

where W_i and W_f are the weight before soil burial and the weight after soil burial, respectively.

Fourier transform infrared (FTIR) analysis

Structural changes after soil burial were investigated using FTIR spectrometer (Perkin-Elmer Model Series 2).The equipment was operated at a 4 cm^{-1} resolution level, in the 4000-650 cm^{-1} scanning range.

Differential scanning calorimetry

Crystallinity studies were conducted using a Perkin Elmer Differential Scanning Calorimetry (DSC) thermal analyzer under a nitrogen atmosphere. Five to ten mg of sample was capsulated on aluminum pans and subjected to heating and cooling cycles. The samples were subjected to an initial heating process to remove the patterns of their previous heating history. Then, the samples were cooled to room temperature at a constant rate of 10 °C/min to favor crystallization. Then, the second heating process was set at 10 °C/min in the temperature range of 25 to 200 °C. The heat of fusion was calculated by integrating the area under the endothermic curve. The percentage of crystallinity of the biocomposites was calculated using Eq. 3,

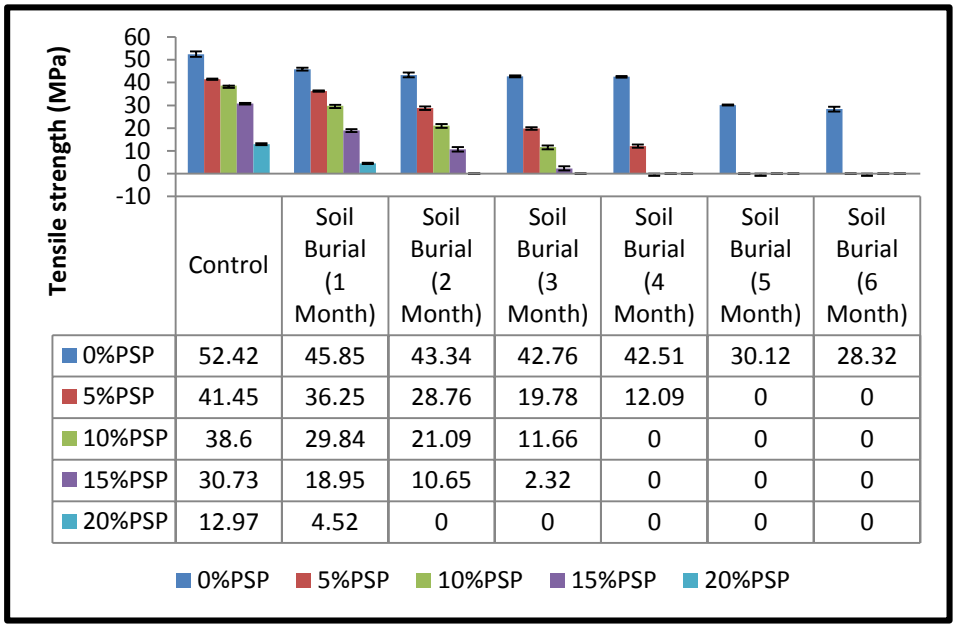
$$\textbf{Crystallinity}\ (\%) = \frac{\Delta H^*_f}{\Delta H^\circ_f}\ \textbf{X}\ 100 \quad (3)$$

where ΔH^*_f is the heat of fusion for the semi-crystallinity of biocomposite and ΔH°_f is the heat of fusion for 100% crystalline PLA.

RESULTS AND DISCUSSION

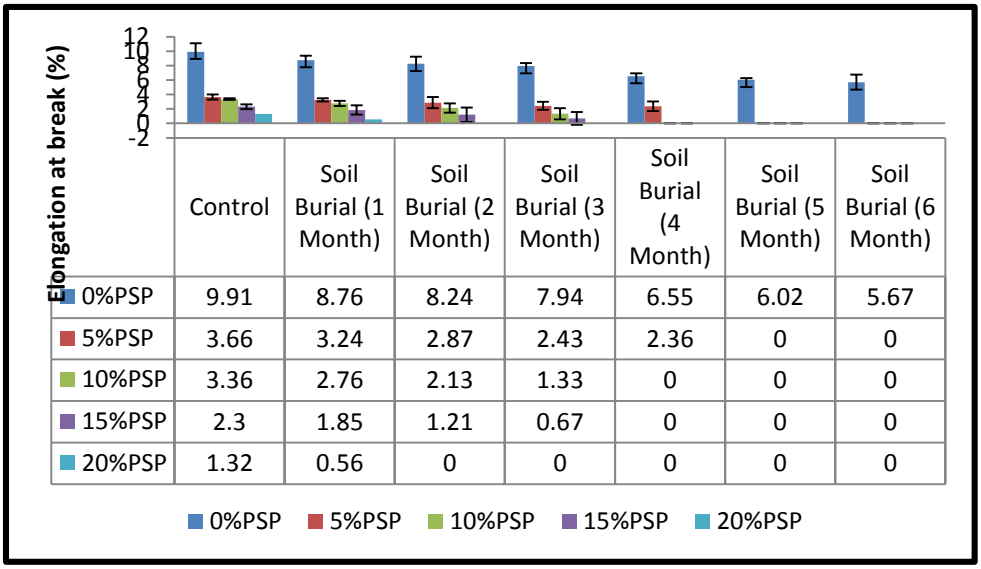
Tensile Properties

Figures 3 through 5 illustrate the effect of natural soil burial on the tensile properties via tensile strength, elongation at break (EB), and modulus of elasticity (MOE) of PLA/PSP biocomposites before and after natural soil burial testing from1 to 6 month periods. It can be seen that the tensile strength (Fig. 3) and EB (Fig. 4) decreased after composting.



	Control	Soil Burial (1 Month)	Soil Burial (2 Month)	Soil Burial (3 Month)	Soil Burial (4 Month)	Soil Burial (5 Month)	Soil Burial (6 Month)
0%PSP	52.42	45.85	43.34	42.76	42.51	30.12	28.32
5%PSP	41.45	36.25	28.76	19.78	12.09	0	0
10%PSP	38.6	29.84	21.09	11.66	0	0	0
15%PSP	30.73	18.95	10.65	2.32	0	0	0
20%PSP	12.97	4.52	0	0	0	0	0

Fig. 3. Tensile strength of the PLA/PSP biocomposites after soil burial testing



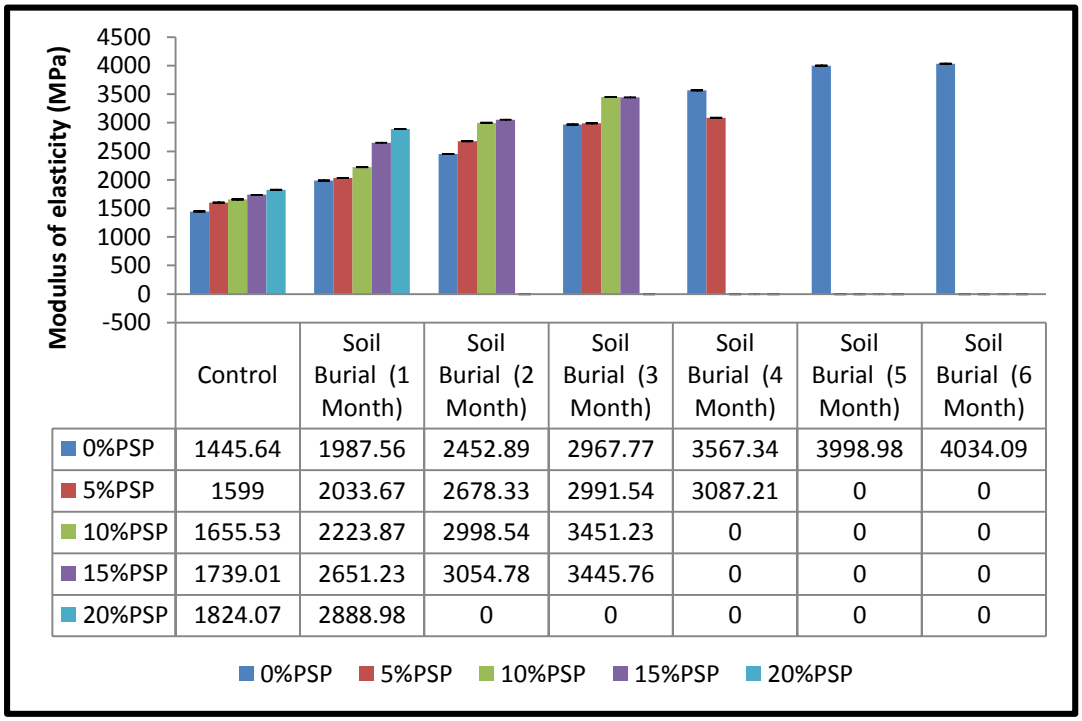
	Control	Soil Burial (1 Month)	Soil Burial (2 Month)	Soil Burial (3 Month)	Soil Burial (4 Month)	Soil Burial (5 Month)	Soil Burial (6 Month)
0%PSP	9.91	8.76	8.24	7.94	6.55	6.02	5.67
5%PSP	3.66	3.24	2.87	2.43	2.36	0	0
10%PSP	3.36	2.76	2.13	1.33	0	0	0
15%PSP	2.3	1.85	1.21	0.67	0	0	0
20%PSP	1.32	0.56	0	0	0	0	0

Fig. 4. Elongation at break of the PLA/PSP biocomposites after soil burial testing

After 5 months of soil burial exposure, the PLA/5PSP biocomposites underwent fragmentation. The biocomposites of 10 wt.% and 15 wt.% of PSP fragmented after 4 months of soil burial, while the biocomposites of 20 wt.% of PSP fragmented after 3 months of soil burial. Therefore, all of the samples could not be subjected to tensile testing.

Figure 4 shows the elongation at break (EB) of biocomposites before and after soil burial testing. The EB trend was similar to the tensile strength trend (Fig. 3). The decrease of the EB over 6 months of exposure was attributed to the consumption of biodegradable (lignocellulotic) materials by indigenous microorganisms, *i.e.*, the paddy straw powder present in the biocomposites. Several fungi that contributed to the lignocellulolytic activity were *Fusarium sp*. (Phutela and Sahni 2011) and *Aspergillus oryzae* (Viji and Neelanayanan 2015).

The modulus of elasticity of the biocomposites after the soil burial testing is shown in Fig. 5. The modulus of elasticity increased with increased exposure period. The increase in the modulus of the biocomposites after composting can be attributed to the enhanced brittleness and stiffness caused by microbial attack (Ismail *et al*. 2008).



	Control	Soil Burial (1 Month)	Soil Burial (2 Month)	Soil Burial (3 Month)	Soil Burial (4 Month)	Soil Burial (5 Month)	Soil Burial (6 Month)
0%PSP	1445.64	1987.56	2452.89	2967.77	3567.34	3998.98	4034.09
5%PSP	1599	2033.67	2678.33	2991.54	3087.21	0	0
10%PSP	1655.53	2223.87	2998.54	3451.23	0	0	0
15%PSP	1739.01	2651.23	3054.78	3445.76	0	0	0
20%PSP	1824.07	2888.98	0	0	0	0	0

Fig. 5. Modulus of elasticity of the PLA/PSP biocomposites after soil burial testing

Figure 6 shows the two fungi, known as *Aspergillus fumigatus* and *Aspergillus terreus*, were obtained from the buried soil after the isolation technique was conducted. Both fungi were identified based on their morphological structure (Table 3).

The tensile strength of all of the buried biocomposites decreased, especially after longer periods of composting. As expected, the tensile strength of the biocomposites after burring was lower than the biocomposites before burring because of the polymer degradation rate. Sam *et al*. (2011) agreed that decreasing the tensile properties during soil burial was attributed to the occurrence of chain scission from polymer degradation.

Aspergillus fumigatus

Aspergillus terreus

Fig. 6. The morphology of the identified fungal strains

Table 3. Morphological Characteristics and Identification of Fungi Isolated from Soil

Color of aerial shape	Color of septate hyphae	Nature of hyphae	Shape	Presence of special structure	Sporangiophore or conidiophore	Characteristics of spore head	Identified fungal strains
Bluish-green	Brown	Septate	Oval greenish	Presence of foot cell	Long erect non septate	Multinucleate green vesicles	*Aspergillus fumigatus*
Brown	Brown	Septate	Oval	Presence of foot cell	Conidiophores	Conidial and hyaline	*Aspergillus terreus*

Table 4 shows the retention of tensile properties after 6 months of composting. After 6 months, all of the samples showed fragmentation. Only the pure PLA sample yielded tensile strength, elongation at break, and modulus of elasticity values of 54.02, 57.21, and 279.05, respectively.

Table 4. Retention of Tensile Properties for PLA/PSP Biocomposites after 6 Months of Soil Burial

	Retention of Biocomposites (%)		
Sample	Tensile Strength	Elongation at Break	Modulus of Elasticity
PLA	54.02	57.21	279.05
PLA/5 PSP	Fragmentation	Fragmentation	Fragmentation
PLA/10PSP	Fragmentation	Fragmentation	Fragmentation
PLA/15PSP	Fragmentation	Fragmentation	Fragmentation
PLA/20PSP	Fragmentation	Fragmentation	Fragmentation

Morphological Study

The surface morphology of the PLA/PSP biocomposites before and after soil burial testing every 1, 3, and 6 months is illustrated in SEM micrographs (Figs. 6 to 8). Figures

6(a-e) show the surface of the PLA/PSP biocomposites after 1 month of soil burial testing. Figure 6(a) shows the surface of the PLA without PSP. The surface was free of cracks and fungi colonization. However, cracks and fungi were found on the surface of PLA/PSP biocomposites (Fig. 6b-e). The size of the cracks or pores was larger in the biocomposites with a higher PSP content. The pores on the surface may have been formed because of the leaching of PSP from the biocomposites. Consequently, the samples decreased in strength. This is in agreement with the reduction in tensile strength for the biocomposites with higher PSP content after the soil burial testing.

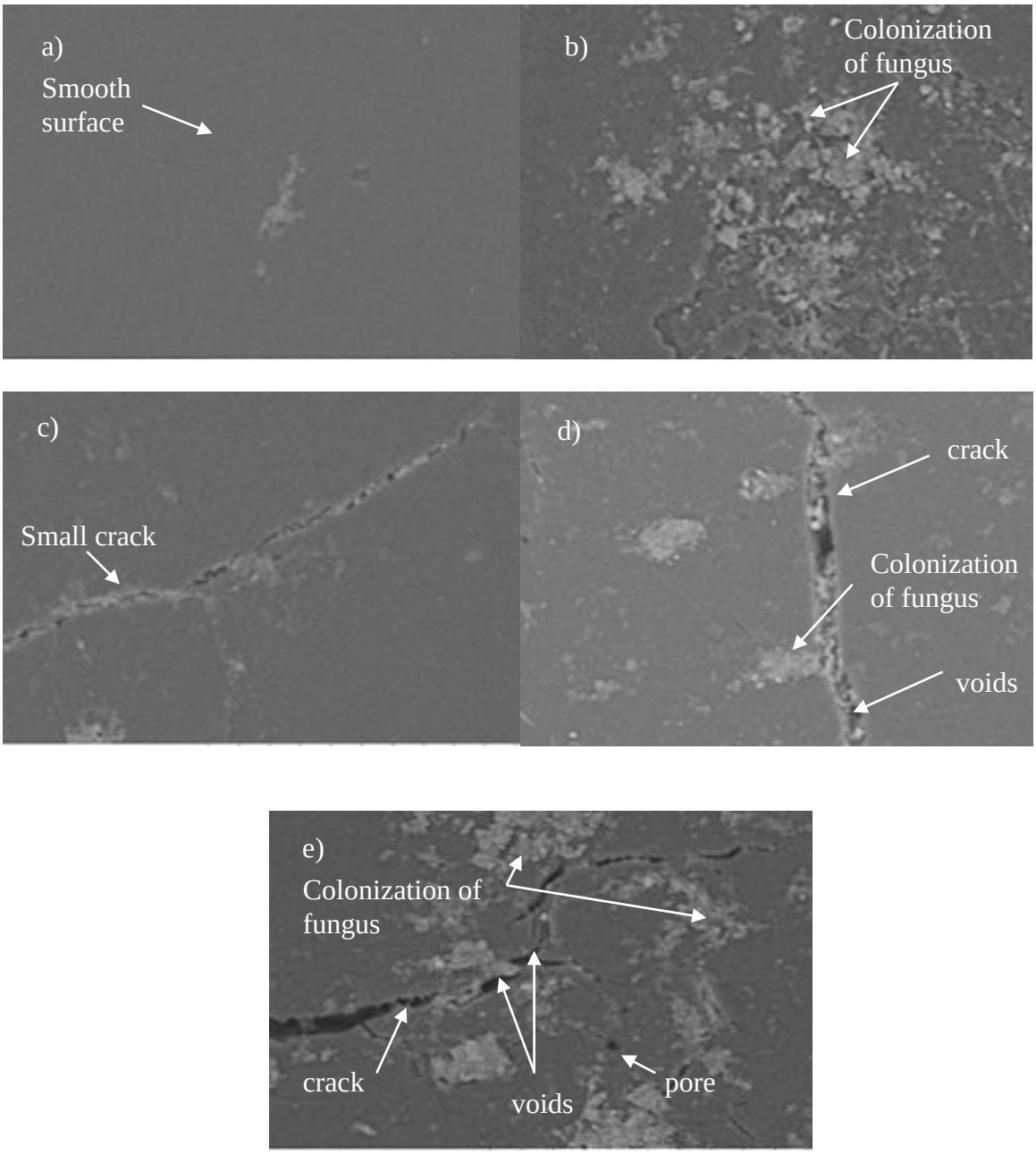


Fig. 6. SEM micrograph (1000x) of PLA/PSP biocomposites with a PSP content of a) 0 wt%; b) 5.0 wt%; c) 10 wt%; d) 15 wt%; e) 20 wt%, after 1 month of composting

Figure 7(a-e) demonstrates the surface of the PLA/PSP biocomposites after 3 months of soil burial testing. The surface of the 100% PLA (Fig. 7a) exhibited no obvious differences compared to the surface of the 1 month soil buried test specimen. Nevertheless, the number of fungi colonies and the size of the pores were larger (Fig. 7b-e).

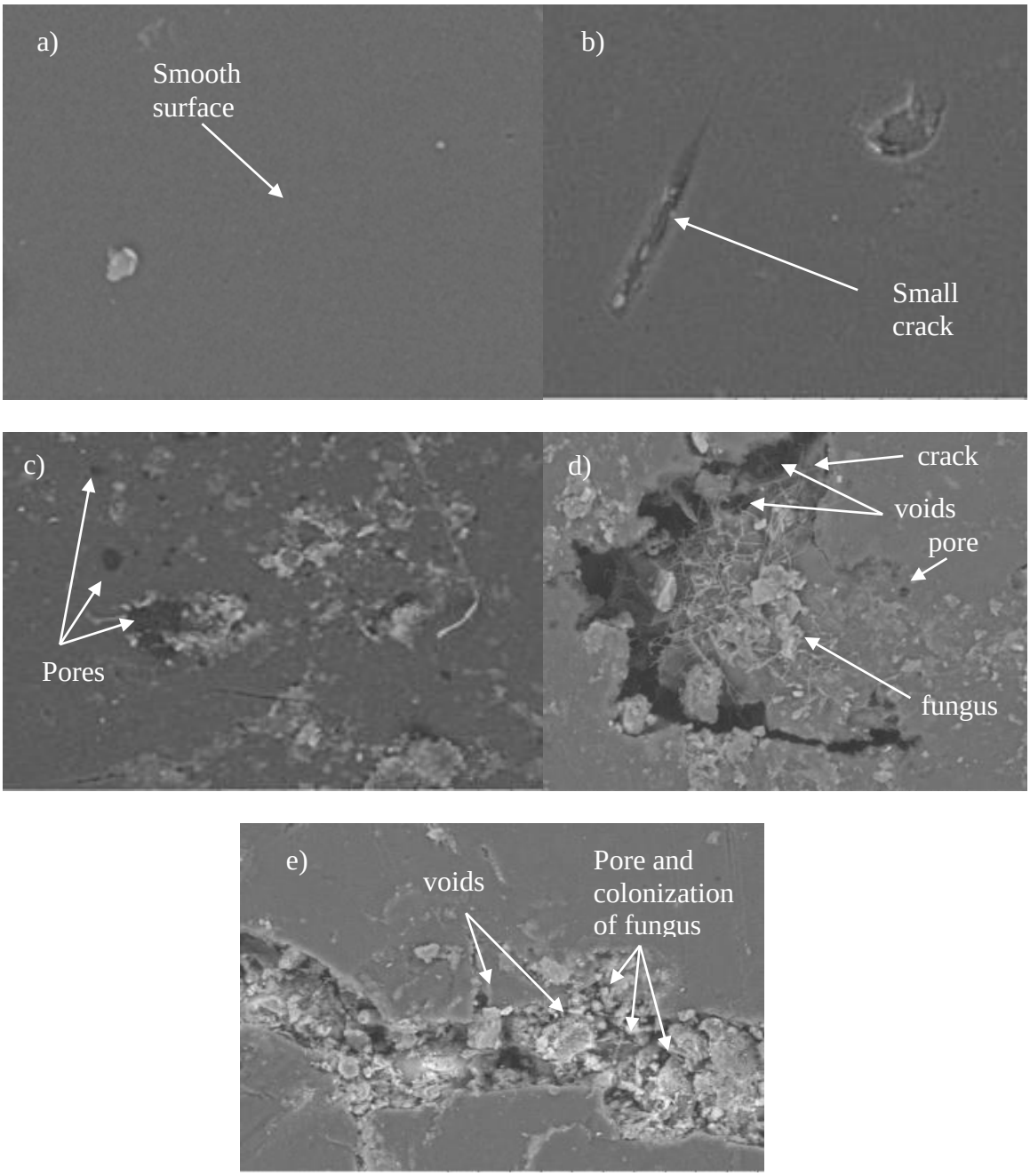


Fig. 7. SEM micrograph (1000x) of PLA/PSP biocomposites with a PSP content of a) 0 wt%; b) 5.0 wt%; c) 10 wt%; d) 15 wt%; e) 20 wt%, after 3 months of composting

Figure 8 (a-e) illustrates the surface of the PLA/PSP biocomposites after 6 months of soil burial. The surface of the 100% PLA (Fig. 8a) shows small crack compared to the surface of the 1 and 3 month specimens. The number of fungi colonies and the size of the pores were larger than the surface specimen for the 1 and 3 month specimens (Fig. 8b-e).

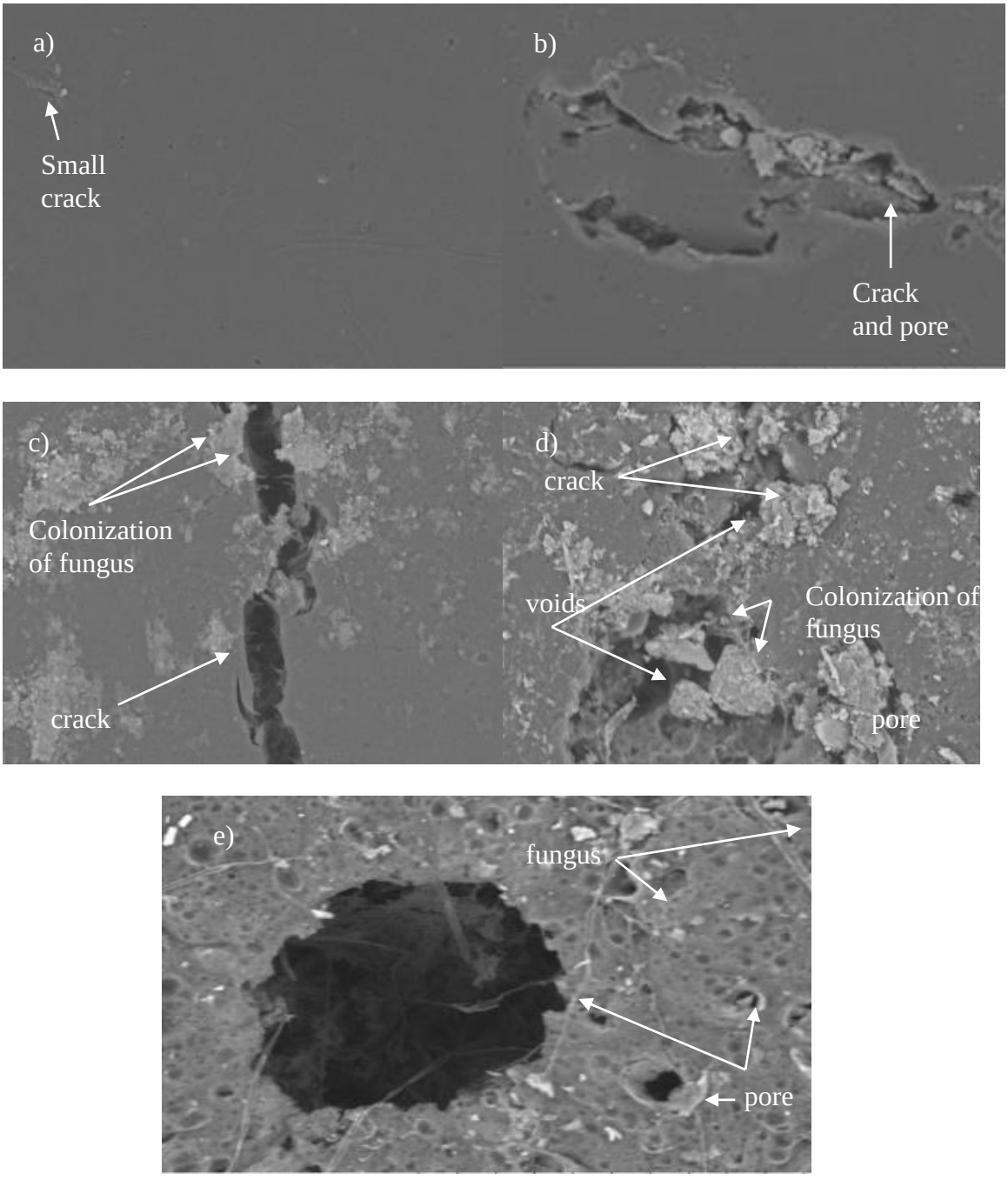


Fig. 8. SEM micrograph (1000x) of PLA/PSP biocomposites with a PSP content of a) 0 wt%; b) 5.0 wt%; c)10 wt%; d) 15 wt%; e) 20 wt%, after 6 months of composting

The degradation that took place in PLA-PSP biocomposites due to their exposure to the natural microbial consortium during soil burial testing is clearly shown. The diffusion of water from the soil caused swelling in the polymer which boosted the microorganism attack (Falk *et al*. 2000) and enhanced the microbial activity. Microbial growth is dependent on the presence of water and oxygen. Previous study by Stark *et al*. (2004) on soil burial testing of sisal fibre/Mater Bi-Y biocomposites found that the soil microflora constituted a mixture of microbial populations, including bacteria,

actinomycetes, and fungi. These microorganisms can act synergistically during the degradation process, and reproduce under naturally occurring conditions (Stark *et al*. 2004). It was highlighted by Karlsson and Albertsson (1998) that biodegradable polymers, whether natural or synthetic, may be more susceptible to microbial and/or enzymatic degradation. However, the rate of this biodegradation is correlated with the type of repetitive unit, morphology (*e.g*., crystallinity and size of spherulites), hydrophilicity, surface area, and the presence of additives.

Weight Loss

Outdoor soil burial is a bio-geophysical test that can provide a realistic environment because soil humidity, temperature, and microorganisms types vary depending on the season. Change in weight is a direct way to measure the biodegradability of polymers. The weight loss percentages of the PLA/PSP biocomposites are shown in Table 5.

Table 5. Weight Loss (wt.%) of PLA/PSP Biocomposites over Different Periods of Soil Burial Testing

	Periods in Months					
Paddy Straw Powder Content (wt.%)	1	2	3	4	5	6
0	0.13	0.52	0.67	0.92	2.05	2.22
5	1.26	1.88	2.95	3.31	10.23	22.24
10	2.56	4.51	7.15	7.91	29.69	38.05
15	4.04	6.74	8.37	10.65	32.91	43.45
20	5.2	9.41	14.14	15.87	35.65	49.86

With increasing PSP content, the percentage of weight loss increased more rapidly. This result indicates that the degradation of PLA/PSP biocomposites is faster than the PLA because lignocellulosic materials are more easily attacked by microorganisms. Other than that, this phenomenon happened due to the increase in water uptake of the biocomposites, which causes major deterioration. The high water uptake is attributed to the hydrophilic nature of the PSP. Swelling effect by the water uptake also led to the microcracking of the biocomposites, resulting in a weight loss. The activities of microorganisms in the soil reduced the samples weights. The void caused by the presence of moisture also permitted the colonization by microorganisms (Figs. 6 to 8).

Fourier Transform Infrared (FTIR) Analysis

The structural changes of the PLA and PLA/PSP biocomposites after soil burial were further analyzed using FTIR (Figs. 9 to 10). Figure 9 presents the FTIR spectra of pure PLA before and after soil burial. The PLA spectrum shows C=O stretching overtones at 3660 to 3500 cm^{-1} and –CH- stretching at 3000 to 2940 cm^{-1}. The -C=O ester carbonyl stretching at 1752 cm^{-1} and the O-C=O stretching at 1190 to 1087 cm^{-1} are characteristic of ester bonds.

Biodegradation will result in increase in carboxyl and alcohol end groups. Buried sample spectra show band in the range 1046 to 1183 cm^{-1} corresponding to –C-O- stretching and 1754 cm^{-1} corresponding to C=O stretching and 1046 cm^{-1} corresponding to

–OH bending increased. Suganti (2008) also observed similar trends and suggested increase of band in the range 1070 to 1180 cm^{-1} is due to increasing in –C-O- stretching of alcohol and increase of band 1745 cm^{-1} is due to increasing in carboxylic stretching while increase in band 1041 cm^{-1} due to increasing of –OH end chain.

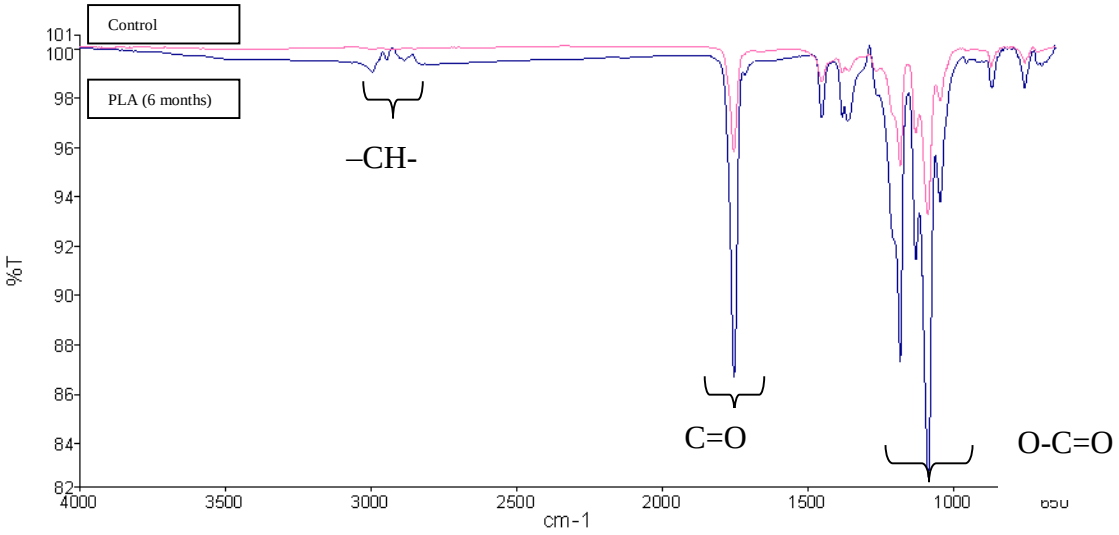


Fig. 9. IR of PLA before and after 6 months soil burial

Figure 10 shows the structural changes of biocomposites (PLA/20PSP) before and after 6 month soil burial. The peak located at 290 cm $^{1-}$corresponds to $–CH_2$ stretching. The FTIR spectra showed the same profile. However, the intensities of the absorption bands were different. The absorption band at 1640 cm^{-1} is assigned to the functional group that is present in the lignin. The increase in the intensities of carbonyl stretching (1750 cm^{-1}) shows that the biodegradation occurred.The hydroxyl group (peaking at (3400 cm^{-1}) was also generated during biodegradation. The formation of carbonyl group after the soil burial test confirms that the biodegradation occurred, leading to a change in the biopolymer chemical structure.

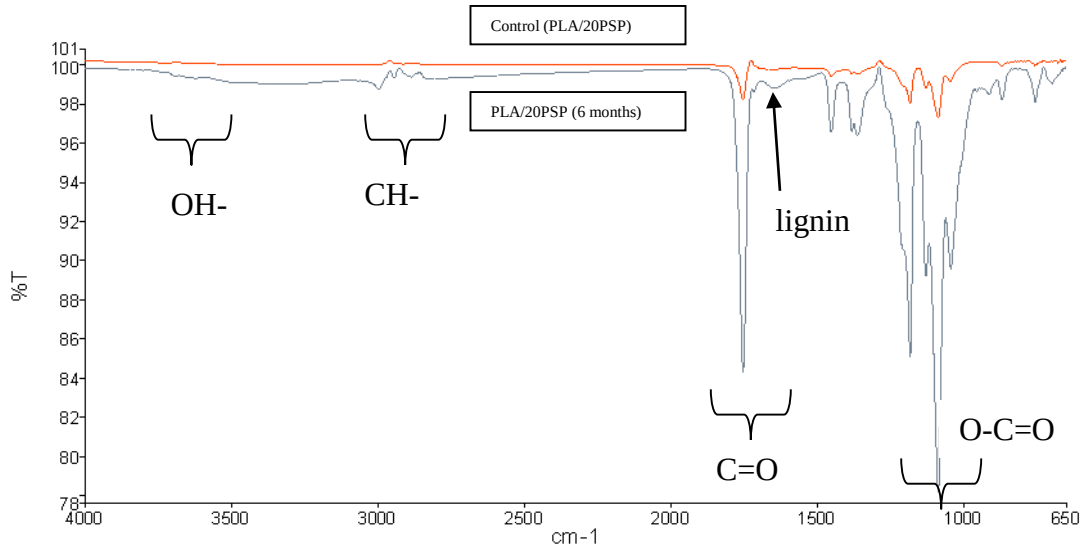


Fig. 10. IR of PLA/20PSP before and after 6 months soil burial

Differential Scanning Calorimetry

The DSC data accumulated after the soil burial testing is summarized in Table 6. The ΔHf for the pure, crystalline PLA was 93 J/g (according to Manshor *et al*. 2014), and the ΔH^*f was obtained from the total area of the second heating procedure. Both values were used to calculate the degree of crystallinity. The melting temperature (T_m) did not exhibit any significant change after the natural soil burial exposure. This indicated that there were no new crystalline domains formed after composting. However, the crystalline temperature (T_c) and heat of fusion (ΔH^*f) increased with longer composting periods, corresponding to a degradation of the amorphous phase in the biocomposites. The crystallinity of the biocomposites increased over the composting period because of the consumption of biodegradable material (paddy straw) by the microorganisms. Paddy straw in the biocomposites easily leached out and was consumed by the microorganism, which consequently left some pores in the sample's surface. Consequently, the remaining crystalline domain in the biocomposite becomes relatively higher. This is similar to the results obtained for most filled thermo-plastics, and was in agreement with previous research. Several studies have found that the crystallinity of biocomposites increased after biodegradation (Phutela *et al*. 2011; Roy *et al*. 2015; Viji and Neelanayanan 2015).

Table 6. DSC Results of PLA and PLA/PSP Biocomposites after Different Periods of Soil Burial Testing

	T_c(°C)			T_m(°C)			ΔH^*_f			Crystallinity (%)		
Samples	Control	3 month	6 month	Control	3 month	6 month	Control	3 month	6 month	Control	3 month	6 month
PLA	106.05	111.05	111.01	172.03	173.04	174.11	17.13	18.67	21.34	18.42	20.07	22.94
PLA/5PSP	90.59	93.21	92.08	170.54	171.11	172.34	35.64	39.43	41.98	38.32	42.39	45.14
PLA/10PSP	92.14	93.15	92.65	169.84	170.31	171.67	32.91	37.66	39.45	35.38	40.49	42.41
PLA/15PSP	90.26	91.66	90.89	162.43	163.03	165.12	29.34	32.45	34.67	31.54	34.89	37.27
PLA/20PSP	87.82	88.52	88.12	160.27	161.07	163.78	29.27	32.29	34.22	31.47	34.65	36.79

CONCLUSION

1. Results from this work show that the degradation of PLA in the soil can be accelerated by the addition of the lignocellulosic material, paddy straw powder.
2. The tensile strength and the elongation at break of the PLA/PSP biocomposites decreased after composting in the soil, whereas the modulus of elasticity increased. These results were support by observations from the SEM images.
3. The weight loss of the biocomposites increased with increasing PSP content. The heat of fusion and crystallinity of the biocomposites increased after composting.
4. During the soil burial process, the amorphous fraction of the materials was exposed to microorganism attack. Therefore, the microbial degradation resulted in an increase in the overall degree of crystallinity of the sample.

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