Effects of Soil Burial on Properties of Linear Density Polyethylene (LDPE)/Thermoplastic Sago Starch (TPSS) Blends

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ABSTRACT
Linear density polyethylene (LDPE)/thermoplastic sago starch (TPSS), blended with and without the addition of compatibilizer [Polyethylene-grafted-Maleic Anhydride, (PE-g-MA)] were prepared for soil burial test. The test was conducted in the natural soil environment for 3 and 6 months. Different loading of TPSS (10, 20, 30, 40, and 50 wt. %) were used in this study. After soil burial, the blends were evaluated for their tensile properties and scanning electron microscopy (SEM) to observe the surface morphology properties after the test. For LDPE/TPSS, it was observed that the tensile strength decreased with the increase of soil burial time, as well as Young modulus and elongation at break (EB). The LDPE/TPSS/PE-g-MA also showed the same trend for the tensile properties, but with higher properties as compared to uncompatibilized blends. The tensile properties also decreased with the increase in the TPSS loading for both the LDPE/TPSS and LDPE/TPSS/PE-g-MA. Meanwhile, the scanning electron microscopy (SEM) on the blend surfaces after the soil burial test showed that degradability increased with the increase in the exposure time as well as the TPSS loading.

Keywords: Soil burial, mechanical properties, biodegradation, TPSS

INTRODUCTION
The increase in the plastic production through the whole nation had introduced the world to environmental problems because most of the plastic materials have remained in the garbage deposits and landfills for decades. These situations may contribute to serious environmental problems (Bikiaris et al., 1998; Chandra and Renu, 1997; Dash et al., 2002; Iionna, 2007; Ismail and Awang, 2008; Krishna Sastry et al., 1998). However, the use of hydrocarbon plastic materials that are not readily biodegradable has increased. Meanwhile, some alternatives have been considered to increase the use of biopolymer in reducing environmental problems.

Biopolymer, such as starch, is a good example of degradable polymer that can be used to replace the hydrocarbon plastic material. The addition of starch and other plastic additives like plasticizers and fillers is usually susceptible to microbial attack. This leads to physical embrittlement of the polymer, leaving a porous and mechanically weakened polymer. The microbes, in turn, release non-specific oxidative enzymes that could attack the synthetic polymer. In addition, the gradual degradation of the natural polymer leads to increased surface area by erosion and pitting. This will accelerate the degradation of the synthetic polymer by diffusion of oxygen, moisture, and enzymes into the porous polymer matrix (Torabi et al., 2004; Usarat and Duangdao, 2006).
In this article, LDPE/TPSS blends were subjected to soil burial. The objective of the paper is to study the behaviour of the samples after the soil burial environment and the effects of the addition of PE-g-MA. The tensile test and surface morphology were applied to characterize the change of tensile properties after the soil burial test. The difference between the uncompatibilized and compatibilized blends was also discussed in this article.

THE EXPERIMENT

Materials
Sago starch was obtained from the Land Custody Development Authority (LCDA), Sarawak. Its moisture content was determined at 13 wt. %. The granules sizes ranged from 9 to 35 μm, with an average granule size of 20 μm. It is crucial to note that sago starch decomposes at 230°C. Pellets of low density polyethylene (LDPE), with the melting temperature of 138°C, were obtained from Titan Polyethylene (M) Sdn. Bhd. Meanwhile, polyethylene-grafted-Maleic Anhydride (PE-g-MA) was obtained from Sigma-Aldrich (M) Sdn. Bhd. Glycerol was obtained from Merck Chemicals grade ACS Reag. Ph Eur.

Preparation of the LDPE/TPSS Blends
TPSS was melt-blended with LDPE in a Haake Rheomix Model R600/610. Mixing was performed at 150°C and 50 rpm for 10 minutes. LDPE was first added into the mixing chamber, followed by the TPSS after 3 minutes of mixing. The total mixing time was 10 minutes. The TPSS loadings varied from 10, 20, 30, 40, and 50 wt. %. PE-g-MA was used at an amount of 10 wt. % based on the TPSS weight. Both the melting temperature and torque were recorded during the mixing periods.

Compression Moulding
The LDPE/TPSS blends were compression moulded in an electrical heated hydraulic press. Hot press procedures involved preheating at 150°C for 6 minutes, followed by compression with a pressure of 1000kg/m² for 3 minutes at the same temperature. All the compression moulded sheets (150 x 150 x 1mm) were cold pressed for 2 minutes.

Soil Burial Test
The soil burial test was conducted for 3 and 6 months. The reason for choosing 3 and 6 months of burying time was primarily to study the effects of biodegradation after the short period of time, as the material is partially biodegradable. In this method, the samples in a dumbbell shape size were buried in alluvial deposit soil environment, whereby all the samples were put in a box covered by soil and thus subjected to the action of micro-organisms, i.e. both fungi and bacteria which are normally present in the soil. After the test, the samples were removed, washed in distilled water and dried at 105°C in an oven for 24 h and then kept in a desiccator.

Tensile Properties
The tensile tests for the unexposed and exposed samples were carried out with a universal testing machine, Instron 3366, according to ASTM D638. The dumbbell specimens of 1mm thickness were cut from the compression moulded sheets with a Wallace die cutter. A crosshead speed of 5 mm/min was used and the test was performed at 25 ± 3°C. Five specimens were used to obtain the average values for tensile strength, elongation at break and Young’s modulus.
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*Morphology Study*  
Studies on the surface morphology of the exposed samples of the LDPE/TPSS blends were carried out using a field emission scanning electron microscope, FESEM SUPRA36VP-24-58. The surfaces of the sample were mounted on aluminium stubs and sputter coated with a thin layer of gold.

**RESULTS AND DISCUSSION**

*Tensile Properties*  
Figs. 1 to 3 show the tensile properties of LDPE/TPSS and LDPE/TPSS/PE-g-MA blends that were buried in the soil. From the figures, it can be seen that the tensile strength, Young’s modulus and elongation at break decreased with the increase in the TPSS loading, as well as the burying time. Meanwhile, the decrease in the tensile strength, Young’s modulus and elongation at break was due to the pit and voids which occurred after the assimilation of the TPSS particles on the surface of LDPE/TPSS blends. The pit and voids act as stress concentrator and lead to a decrease in tensile strength and Young’s modulus. In addition, micro-organism attacks starting with starch. As the micro-organisms consumed the surrounding starch, the blends lost their structural integrity. This process could lead to the deterioration of the tensile properties. The degradation of the blends normally occurs due to the vacation of starch sites, which are occupied by either microbes or water and thus, leads to extensive degradation of the blends. During the experiment, the water inside the soil diffuses into the polymer sample, causing swelling and enhancing biodegradation. In addition, extra cellular enzymes made by the microbes also attack the resin and may be responsible for the fine cracking and tearing that lower the elongation at break, and can lead to further degradation and lower the tensile properties. The blends containing a higher percentage of TPSS (40 and 50% TPSS content) degraded faster as compared to the blends with lower TPSS loading in first 3 months, during which the maximum starch content was accessed. Over the next 6 months, a gradual decrease of TPSS was observed.

The tensile properties of the LDPE/TPSS/PE-g-MA blends with the addition of compatibilizer buried in the soil showed a higher tensile strength as well as the Young’s modulus and elongation at break. This result might be due to the compatibilizing effect of the PE-g-MA in the blends that could prevent early degradation to the blends. Meanwhile, the effect of the PE-g-MA reduced the volumes of voids which created the stress concentration area in the blends. As a result, the mechanism of degradation was reduced.
Fig. 1: Tensile strength of control and buried samples (a) LDPE/TPSS blends; (b) LDPE/TPSS/PE-g-MA blends
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**Fig. 1:** Tensile strength of control and buried samples (a) LDPE/TPSS blends; (b) LDPE/TPSS/PE-g-MA blends

**Fig. 2:** Young modulus of the control and buried samples; (a) LDPE/TPSS blends; (b) LDPE/TPSS/PE-g-MA blends

- **Fig. 2a:**

<table>
<thead>
<tr>
<th>TPSS loading (wt. %)</th>
<th>LDPE/TPSS control</th>
<th>LDPE/TPSS Soil Burial 3months</th>
<th>LDPE/TPSS Soil Burial 6months</th>
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</thead>
<tbody>
<tr>
<td>0</td>
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<td>264.7</td>
<td>225</td>
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<td>10</td>
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<td>245.733</td>
<td>172.6</td>
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<td>30</td>
<td>205.84</td>
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<td>40</td>
<td>210.5</td>
<td>141.55</td>
<td>129.65</td>
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<td>50</td>
<td>197.3</td>
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</table>

- **Fig. 2b:**

<table>
<thead>
<tr>
<th>TPSS loading (wt. %)</th>
<th>LDPE/TPSS/PE-g-MA control</th>
<th>LDPE/TPSS/PE-g-MA Soil burial 3months</th>
<th>LDPE/TPSS/PE-g-MA Soil burial 6months</th>
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<tbody>
<tr>
<td>0</td>
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<td>10</td>
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<td>50</td>
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<td>208.4</td>
<td>169.233</td>
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</tbody>
</table>
Fig. 3: Elongation at break of the control and buried samples;
(a) LDPE/TPSS blends; (b) LDPE/TPSS/PE-g-MA blends
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Fig. 4: The SEM Micrographs of LDPE/TPSS: (a) 10% TPSS loading, (c) 50% TPSS loading and LDPE/TPSS/PE-g-MA after 6 months of soil burial; (b) 10% TPSS loading, (d) 50% TPSS loading

Morphological Properties

Fig. 4 shows the SEM micrographs of LDPE/TPSS and LDPE/TPSS/PE-g-MA blends after 6 months of soil burial. From the figures, it can be seen that the sample with higher TPSS loading yields higher degradation effects. At 50% TPSS loading, continuous holes and voids can be seen due to the starch that has leached out. In addition, the samples containing a higher percentage of starch supported fungal growth. TPSS, being a natural polymer, could be readily used as a carbon source by the fungus. For the LDPE/TPSS/PE-g-MA blends, it can be seen that the samples with the addition of compatibilizer show lower voids, especially for the sample with 50% TPSS loading. The voids can still be seen but the trend is different with the blend without compatibilizer. Similarly, the voids and holes can be seen but with better distribution. The samples without the addition of compatibilizer show continuous holes and voids that prove the agglomeration has occurred inside the blends. However, the SEM micrographs for the blends with the addition of compatibilizer show a lower agglomeration but a better distribution of voids and holes.

As an evident in the SEM surface micrograph, Fig. 5 shows the percentage of weight loss for the LDPE/TPSS and LDPE/TPSS/PE-g-MA blends after soil burial for 3 months and 6 months. It is clear that the weight loss for the blends without the addition of compatibilizer is higher than the blends with the addition of compatibilizer. Meanwhile, the addition of PE-g-MA, as the compatibilizer reduced the water diffused inside the samples, has generated the degradation mechanism inside the blends.
CONCLUSIONS
After both exposure periods to soil burial test, the results from the tensile test indicated that the LDPE/TPSS/PE-g-MA blend exhibited higher tensile strength and Young modulus than LDPE/TPSS blends due to a good interfacial adhesion between the LDPE and TPSS, although both blends have shown a reduction in the tensile properties with the increase in the soil burial time. The tensile properties have also been found to decrease with the increase in the TPSS loading for both the LDPE/TPSS and LDPE/TPSS/PE-g-MA blends. Although holes were evident for both the blends, the surface morphology of LDPE/TPSS/PE-g-MA showed lower continuous holes and TPSS leached out due to the lower agglomeration and good interaction between the TPSS and LDPE.

ACKNOWLEDGEMENT
The authors gratefully acknowledge the financial support from the Research University Grant (Grant No. 1001/PBAHAN/814008).
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REFERENCES


