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# Incorporation of Kenaf Core Fibers into Low Density Polyethylene/Thermoplastic Sago Starch Blends Exposed to Natural Weathering

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*The landfill disposal of a high volume of plastics that take a long time to decompose has led to a tremendous environmental problem. Incorporation of natural polymers and fibers into synthetic polymers accelerates the degradation rate by exposure to atmospheric agents such as sunlight, temperature and rainfall. In this work, thermoplastic sago starch (TPSS) and kenaf core fibers (KCF) from agricultural feed stocks were blended with low density polyethylene (LDPE) for natural weathering studies. The melt-mixed and compressed composite sheets had fiber loadings ranging from 0 to 40 wt.%, and were exposed to natural weathering conditions for a period of 3 and 6 months. The deterioration in mechanical, thermal, morphological and weight properties were investigated.*

**Keywords** Thermoplastic sago starch; kenaf; degradation

## Introduction

Plastics are widely used in almost all applications of daily life and their production is a major worldwide industry. The most commonly used commodity plastics in many applications are polyethylene (PE), polypropylene (PP), polystyrene (PS), poly(ethylene terephthalate) (PET), and poly(vinyl chloride) (PVC). They have become materials of preference owing to their low cost, especially in processing, as well as due to their outstanding performance. However, the extensive global production and usage of plastics have focused attention and concern on their environmental impact. The non-degradability of plastics causes many environmental problems associated with disposal after use. For example, polyolefin plastics are widely used in packaging (i.e. carry bags) applications which are disposed of mostly

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after one time usage. If wastes are not properly disposed, this solid mass which takes a long time to decompose, in turn possess a serious menace to the environment [1].

Industrial research is underway to develop the polymers that can degrade faster after disposal. As polymer usage is inevitable, one of the promising alternatives to augment the biodegradability of the polymers by blending them with biodegradable materials [2]. The degradability might be accelerated with minor inclusion of additives or blending in the common synthetic polymer. The search for low cost as well as environmentally friendly materials has led to the development of biodegradable composites with incorporation of natural polymers and natural fibers (i.e. starch, cellulose). It is expected, when these plastic blends are placed in environments, that the natural materials in them accelerate the degradation rate by microbial attack and/or exposure to atmospheric agents such as solar radiation, wind, rain and humid conditions [3].

As mentioned previously, the rising concern about the disposal of plastics waste and the need for more versatile polymer-based materials have led to increased interest in blending synthetic polymers with degradable materials. In order to impart degradability, biopolymers such as starch are commonly incorporated into conventional plastics which will then boost the accessibility of the plastics to oxygen and microorganisms. Modified plastics have proven to meet requirements and guaranteed at least partial degradation [4]. However, it has been found that the addition of biopolymers as a minor ingredient in the synthetic polymer causes a reduction of mechanical properties due to the immiscibility caused by the different polarities of the two constituents [5]. Apart from that, Prachayawarakorn et al. (2012) has also reported that thermoplastic starch (TPS) could be blended with synthetic polymers such as high density polyethylene (HDPE) and linear low density polyethylene (LLDPE). However, the degradation duration is still very high due to those non-biodegradable synthetic polymers [6]. Thus, another interesting approach to improve mechanical properties and impart degradation is to use natural fibers (i.e. kenaf) to reinforce synthetic polymer/biopolymer blends. To preserve renewability, biodegradability as well as to improve mechanical properties of the final products, the association between natural fiber and synthetic polymer/biopolymer blends has been investigated [7, 8]. Natural fibers are prefered in the light of the fact that they can be completely biodegraded in the natural environment.

Natural weathering, also known as environmental degradation, is considered a practical way to acquire information regarding material performance resulting from exposure to natural outdoor conditions [9]. Polymeric materials are directly or indirectly subjected to a variety of environmental conditions such as sunlight, heat, oxygen, moisture and microorganisms contribute to the degradation of polymeric materials [9]. Because Malaysia experiences a hot and wet tropical climate, it is interesting to investigate the capability of the composites to withstand the environmental degradation.

PE is resistant to attacks by microorganisms or to chemical means other than photooxidation in most naturally occuring environments. The addition of biodegradable polymers and natural fibers make the polymer more susceptible to microbial attack and enhances the natural disintegration of the blend. The gradual degradation of natural polymers increases the surface area of the polymers by creating pores which will become exposed to possible microbial attack. Those pores weaken the polymer and enable it to break apart and subsequently leave the synthetic polymer which will undergo further natural elemental attacks such as thermal oxidation and ultraviolet photo-oxidation. The composite then undergoes biological breakdown by oxidative scission of the molecular chains, which cause changes to the surface appearance and the chemistry of the composite products that usually leads to discoloration and a loss in structural integrity. UV radiation leads to radical formation,

followed by the absorption of oxygen and formation of oxidized groups such as carbonyl, carboxyl and hydroxyl [2]. This formation will cause the polymer to become brittle and will decrease the properties such as melt flow/viscosity, molecular weight and mechanical strength upon exposure [10]. The photo-degradation of natural fibers is attributed to the degradation of its components (cellulose, hemicelluloses, lignin etc.) and can usually be detected by an initial color change and surface roughening [10].

Basically, the combination of different environmental factors such as oxygen, temperature, sunlight, water, living organisms and pollutants, which are responsible for the degradation of polymers, may result in synergistic effects on the polymer degradation rate [9]. Although the previous literatures have shown that extensive studies on weathering effects on the polymer matrix and natural fibers have been carried out, the existence of any work on the effect of natural weathering on LDPE/TPSS blend reinforced with KCF is hardly identified. Associated to this, efforts were made in this present work to investigate the biodegradation behaviour and functionality of kenaf as biodegradants under outdoor weathering. Mechanical changes on composite, as well as structural and morphological were characterized.



## Experimental

### *Materials*

Sago starch (13% moisture) was obtained from the Land Custody Development Authority (LCDA), Sarawak, Malaysia. It had an average particle size of 20 $\mu$m and decomposition temperature of 230°C. Glycerol (plasticizer) was an analytical grade reagent, purchased from Merck Chemicals (Malaysia) and used as received. Low density polyethylene (LDPE, LDF 260GG) with melt flow index of 5 g/10 min was obtained from Titan (M) Sdn. Bhd. (Malaysia). Kenaf fiber (core) with average length of 5 mm was supplied by National Kenaf and Tobacco Board (LKTN), Malaysia. The fibers were subjected to grinding process which yields particles approximately 70 to 250 $\mu$m in diameter. KCF were then dried for 3 hours at 70°C using vacuum before being used in the subsequent composite fabrication.

### *Sample Fabrication*

Sago starch powder was vacuum dried by heating at 80°C for 24 hours before blending and processing. The dried sago starch was then pre-mixed with glycerol by using a high speed mixer. The ratio of sago starch to glycerol was maintained at 65:35 (by weight percent). The blend was stored overnight to allow the diffusion of glycerol into starch granules and would help the melt-mixing process. TPSS was melt-blended with LDPE and kenaf fiber using an internal mixer (Haake Rheomix Mixer, Model R600/610) at the temperature of 150°C at speed of 50 rpm in order to obtain a homogeneous sample. The ratio between LDPE/TPSS was fixed at 90/10 and different loading of KCF (i.e. 10-40 wt.%) were used. The processed samples were then compression molded in an electrically heated hydraulic press (Kao Tieh Go Tech Compression Machine) at a temperature of 150°C into a 1 mm thickness sheet. Table 1 shows the formulation of the composites.

### *Outdoor Weathering Test*

The tests were performed by exposing dumbbell samples of LDPE/TPSS blends reinforced with KCF composites to outdoor weathering. The tests were conducted at an open area in Universiti Sains Malaysia (USM), Penang, Malaysia (latitude 5°28′N, longitude 100°29′E)

**Table 1.** Formulation of LDPE/TPSS/KCF composites

| | Composition | |
|---|---|---|
| Sample | LDPE/TPSS (wt.%) | KCF (phr) |
| LDPE/TPSS | 100 | 0 |
| LDPE/TPSS + 10% KCF | 90 | 10 |
| LDPE/TPSS + 20% KCF | 80 | 20 |
| LDPE/TPSS + 30% KCF | 70 | 30 |
| LDPE/TPSS + 40% KCF | 60 | 40 |

*Note: wt.% - weight proportion, phr – parts per hundred of resin.



for a period of 3 months and 6 months, from March 2012 to August 2012. The dumbbell samples were arranged on an exposure rack facing to the south and at an inclination angle of 45°. After the exposure period, the samples were subjected to further mechanical and analytical test. The samples were washed with distilled water, dried and weighed after drying to a constant weight in an air-drying oven, maintained at 70°C. Weather reports for the 6 months exposure period were obtained from the Meteorological Department of Penang State, Malaysia and are summarized in Table 2. Table 2 depicts the climate of the weathering site in Northern part of Peninsular Malaysia, which featured uniform temperature and relatively variation of rainfall.

***Tensile Properties***

Tensile tests were carried out with a Universal Testing Machine (Instron 3366) according to ASTM D638. Dumbbell specimens of 1 mm thickness were cut from the compression molded sheets with a Wallace die cutter. A crosshead speed of 5 mm/min was used and the test was performed at temperature of 25 ± 3°C and relative humidity of 60 ± 5%. Five specimens were used to obtain average values for tensile strength, elongation at break, and Young's modulus. The retention of these properties was calculated as follows:

$$\text{Retention}\,(\%) = \frac{\text{value after degradation}}{\text{value before degradation}} \times 100\% \tag{1}$$

**Table 2.** Climatic conditions during the weathering test

| Exposure (month) | Mean temperature (°C) | Mean rainfall (mm) |
|---|---|---|
| March 2012 | 27.5 | 176.4 |
| April 2012 | 27.9 | 287.0 |
| May 2012 | 28.3 | 264.2 |
| June 2012 | 28.6 | 97.0 |
| July 2012 | 27.6 | 178.0 |
| August 2012 | 28.1 | 105.4 |

### *Fourier Transform Infrared Analysis*

The functional groups and chemical characteristics of LDPE/TPSS/KCF composites after natural weathering test as a function of KCF content were obtained by Fourier Transform Infrared Spectroscopy (FTIR, Perkin Elmer System 2000) with a resolution of 4 $cm^{-1}$ in a spectral range of 4000–600 $cm^{-1}$ using 32 scans per sample. The carbonyl index (CI) was used as a parameter to observe the degree of degradation of LDPE/TPSS blends reinforced with KCF composites. CI was calculated as the ratio of the intensity of the peak at 1720 $cm^{-1}$ to the reference peak intensity of 1460 $cm^{-1}$. The peak at 1720 $cm^{-1}$ corresponded to the absorption from the presence of a carbonyl groups which was the by product of polymer degradation.

### *Weight Loss*

The biodegradability was assessed and evaluated by measuring weight loss before and after testing. The weight loss of weathered samples was calculated and evaluated using the following Eq. 2:

$$\text{Weight loss}\,(\%) = \frac{W_0 - W_1}{W_0} \times 100 \tag{2}$$

where $W_0$ and $W_1$ are sample weights before and after the composting test respectively.

### *Morphology Evaluation*

Scanning electron micrographs of surfaces of LDPE/TPSS/KCF composites before and after outdoor weathering were obtained by using a Scanning Electron Microscope (SEM, model ZEISS Supra 35 VP). The samples were sputter coated with a thin layer of carbon to avoid electrostatic charging during the examination.

## Results

### *Tensile Properties*

Figures 1–3 depict the tensile properties of LDPE/TPSS/KCF composites with variation of KCF loadings and its ability to withstand degradation after 3 to 6 months of outdoor weathering exposure. Overall the specimens experienced a progressive decay in tensile properties after natural weathering. The deterioration was more severe with an increase in exposure time. Generally, increasing the KCF loading up to 10 phr resulted in an increase in the tensile strength of the un-weathered composites, while a further increase of fiber loading decreased the tensile strength. However, for the weathered composites, the tensile strength decreased with increasing fiber loading. Table 2 indicates that the retention of tensile strength decreased as the KCF loading increased. This result suggested that the reinforcing effect of the KCF has deteriorated upon natural weathering. In all cases, the strength of the composites showed a decline with increasing weathering time, most probably due to degradation of the composites during exposure due to thermo-oxidation, photo-oxidation and chain scission reactions [10]. Upon weathering, photo-oxidation degrades the polymer matrix by breaking down polymer chains. Exposure of polymers to UV during natural weathering results in chain scission due to oxidation of the polymers. As a result, the melt flow/viscosity, molecular weight and mechanical properties of the polymer have weakened.

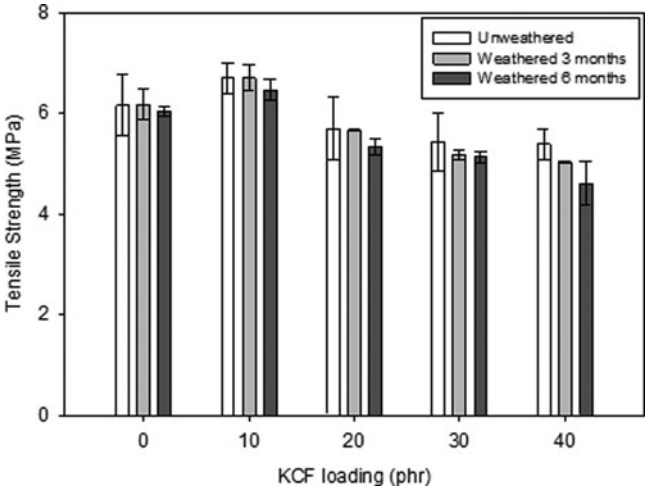


**Figure 1.** Tensile strength of LDPE/TPSS blends reinforced KCF before and after exposure to natural weathering.

It is worth noting that with the addition of starch to the synthetic polymer, the blend was prone to degradation. Exposure of starch to UV radiation leads to degradation by photo-oxidation of amylose (mainly in amorphous region of starch) [11]. Concurrently, when fiber reinforced polymer composites are exposed to outdoor weathering, fiber components such as lignin, hemicelluloses and cellulose also underwent rapid degradation. The absorption of UV by lignocellulosic components during natural weathering results in photo-chemical degradation of the lignocellulosic components (especially lignin) [10]. Photo-chemical degradation will act in combination with moisture, temperature and agents such as oxygen and/or ozone to depolymerise lignin and cellulose components, thus leading to a reduction in some physical, chemical and biological properties of the lignocellulosic materials [10].

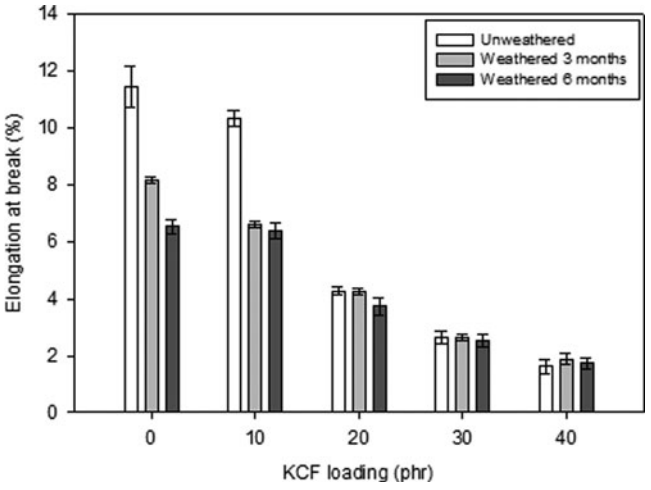


**Figure 2.** Elongation at break of LDPE/TPSS blends reinforced KCF before and after exposure to natural weathering.

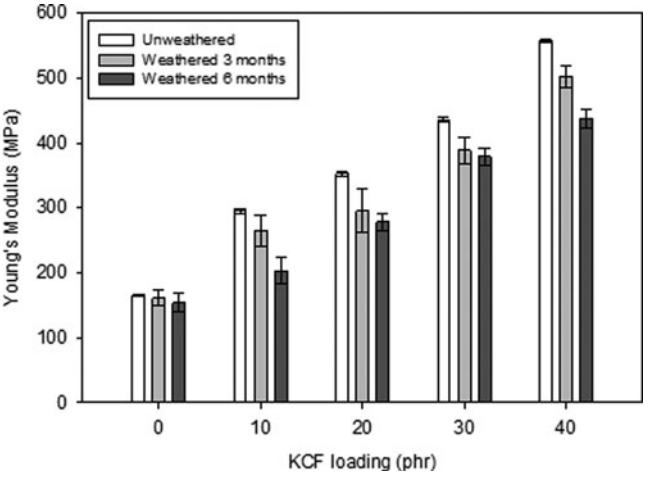


**Figure 3.** Young's modulus of LDPE/TPSS blends reinforced KCF before and after exposure to natural weathering.

As shown in Fig. 1, after 6 months of weathering testing, all specimens showed a severe decline in tensile strength. It is apparent that the retention of tensile strength decreased as the KCF loading increased. Most blends showed diminutions of tensile strength after the exposure as a result of the combined effects of atmospheric factors such as solar radiation, temperature and humidity as well as seasonal variations and environmental pollutants [12]. When a sample was subjected to natural weathering for a longer period, severe surface cracks developed, and this permitting chemical degradation, water absorption, microorganism attack to take place and eventually deteriorate the properties of the composite [11]. In fact, the surface cracks of the composite structure also resulted from the cyclic expansion and contraction of the sample resulting from variable climatic conditions. Thus moisture could absorb into the composites through fiber-matrix interfacial gaps, fiber absorption and diffuse through polymer chains [10]. These will then lead to diminution of fiber-matrix interfacial adhesion.

Similar trends were observed for the Young's modulus values for all samples. This finding is common for lignocellulosic polymer composites where exposure to photo-oxidation and thermo-oxidation reactions during natural weathering results in more brittle composites [10]. Photo-oxidation concentrated near the surface of the composites because of higher permeability to oxygen. Upon exposure, the degradation of the samples started at the outer surface and penetrated gradually into the matrix. As mentioned previously, the development of more surfaces cracks allow chemical degradation and water absorption to occur. The chemical degradation and water absorption can be caused by exposure of the blends to rain water and moisture which can seep through cavities of the surface cracks, causing chemical attack to the polymer bonds [10]. These results are evident in the SEM micrographs in Figs. 6 and 7. The susceptibility of the starch and fibres to absorb rain water and moisture subsequently promote larger surface for degradation due to leaching out of starch and detachment of kenaf. The voids being occupied by either microbes or water make the main chain of polymers more accessible for further microbial assimilation. The reduction of chain length tended to reduce elongation at break as the polymer flexibility and stiffness decreased. Apart from that, the crack on the surface exposes the inner layer to atmospheric

**Table 3.** Retention of tensile properties for LDPE/TPSS/KCF composites after 3 and 6 months weathering

| | Retention (%) | | | | | |
|---|---|---|---|---|---|---|
| | Tensile strength | | Elongation at break | | Young's modulus | |
| Sample | 3 months | 6 months | 3 months | 6 months | 3 months | 6 months |
| 0 | 100.34 | 98.21 | 71.29 | 57.01 | 97.10 | 93.12 |
| 10 | 99.96 | 96.44 | 63.73 | 61.64 | 89.98 | 68.84 |
| 20 | 99.61 | 93.66 | 99.35 | 87.27 | 83.58 | 78.89 |
| 30 | 95.29 | 94.37 | 100.20 | 95.36 | 89.25 | 86.88 |
| 40 | 93.59 | 85.62 | 116.23 | 107.34 | 90.04 | 78.46 |



factors (i.e. heat, light, and rain), which accelerates the loss of plasticizer through migration and evaporation. Once the plasticizer content was reduced or lost, the specimen loses its flexibility and cannot elongate to the extent it previously could [11].

### *Fourier Transform Infrared (FTIR) Analysis*

Structural changes of LDPE/TPSS/KCF composites due to natural weathering can be accurately detected by FTIR. Figure 4 shows FTIR spectra obtained for LDPE/TPSS/10 phr KCF before and after weathering. The representative FTIR results demonstrate the changes that occurred in the weathered specimens especially the presence of degradation products such as carboxylic and carbonyl groups. As shown in Fig. 4, the assignments of the main absorption bands are 3000–3600 $cm^{-1}$ (O—H stretching), 2915 $cm^{-1}$ (C—H stretching), 2848 $cm^{-1}$ (stretching vibration of $CH_2$ group), 1463 $cm^{-1}$ (scissoring vibration

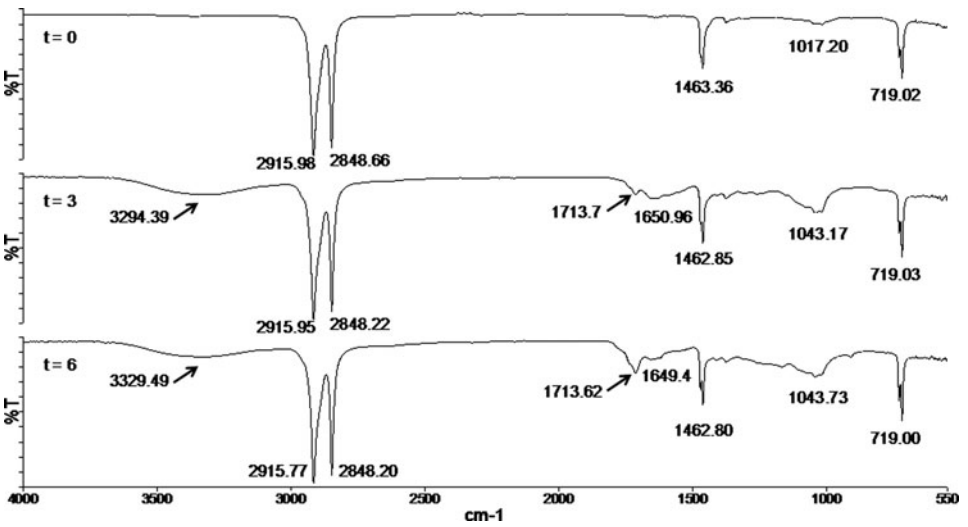


**Figure 4.** Representative FTIR spectra for LDPE/TPSS/10 phr KCF composites before and after weathering exposures.

of $CH_2$ group), 1017 $cm^{-1}$ (C—O stretching) and 719 $cm^{-1}$ (C—C stretching vibration of $CH_2$ groups).

Upon exposure to natural weathering, there are peaks that are very well defined and their intensities increased with an increase in weathering time. The FTIR analysis suggested that the degradation of the composites became prominent after prolonged weathering. Referring to Fig. 4, the appearance of a weak shoulder at 3200–3400 $cm^{-1}$, which represents O-H stretching, can be seen in both composite samples after weathering for 3 and 6 months periods. This is probably due to the presence of hydroxyl groups from the generation of hydroperoxide and hydroxyl species [10]. It is reported that UV radiation causes reduction of the chain size and forms groups such as carbonyl, carboxyl and hydroxyl. The intermediate products such as hydroperoxides, peroxides, ketones and aldehydes resulting from the partial oxidation of LDPE are present in small amounts [13]. The appearance of hydroxyl region might be due to the O-H stretching of KCF exposed on the weathered composite surface.

The most prominent peak to indicate the oxidation and degradation of the system during the period of outdoor exposure corresponds to the carbonyl groups [14]. Formation of carbonyl groups after weathering confirms that photo-degradation took place and that the chemical structure of the polymer was changed [15]. As seen in Fig. 4, upon exposure to natural weathering, the band at 1713 $cm^{-1}$ which is attributed to C=O vibration starts to develop at $t = 3$ (3 months exposure). After the exposure period of 6 months, the intensities of the functional groups seem to be slightly higher indicating more deterioration of the system. The results demonstrated that the degradation of the composites became severe at prolonged weathering times. This is probably due to the presence of KCF in the composites which increase the carbonyl functionality and makes the composites more susceptible to undergo degradation. It is also worth noting that the band associated with C=O stretching vibrations resulted from carboxyl groups in the xylan components of hemicelluloses and also the chemical groups of lignin. The increase of the intensity of this peak signified the oxidation during the period of outdoor exposure, as well as the presence of oxidation products such as a mixture of carboxylic acid and ketone species [10].

Many researchers have used the carbonyl index as a parameter to observe surface oxidation and degradation of blend systems [11]. In this work, the index showed a gradual increase (i.e. more degradation products were generated) with an increase in weathering time. For this work, it was found that, the CI for $t = 3$ and $t = 6$ are 1.2662 and 1.2707, respectively. Generally, the increment of carbonyl index concluded that the exposure of polymer to UV light during weathering will result in carbonyl degradation via Norrish Type II. This is due to presence of KCF that led to an increase in porosity of the blend, which then easily allowed the permeation of light and oxygen throughout the inner part of the matrix.

As shown in Fig. 4, weathered specimens also showed a strong O—H band in a region of 1640–1655 $cm^{-1}$. The broad 1650 $cm^{-1}$ O—H band can be attributed to water molecules absorbed by starch molecules. This intense peak is caused by the high water absorption of the weathered specimen from the environment [11]. Another significant peak appearing in weathered samples was at around 1043 $cm^{-1}$. By comparison, it can be seen that the intensity of the peak appearing at 1043 $cm^{-1}$ assigned to C—O stretching (combination of cellulose, hemicelluloses and lignin) becomes broader and stronger upon exposure to natural weathering [13].

Accordingly, significant changes in hydroxyl (O—H) and carbonyl (C=O) regions were observed after weathering. The presence of both carbonyl and hydroxyl functional

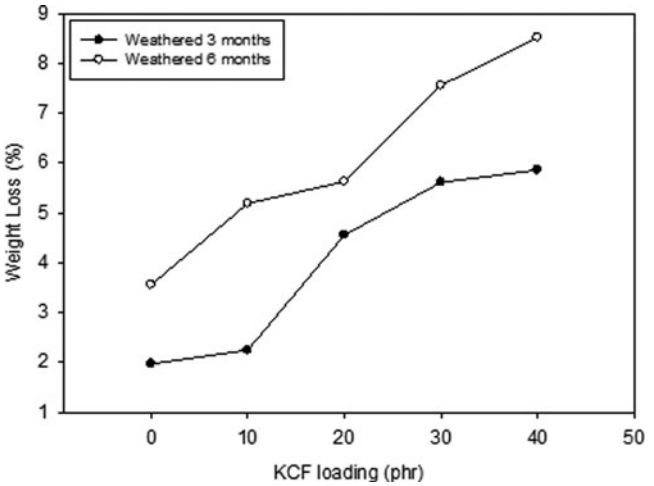


**Figure 5.** Weight loss of LDPE/TPSS/KCF composites after exposure to natural weathering for 3 and 6 months.

groups signified the photo-oxidation of both blends during the period of outdoor exposure [12].

### *Weight Loss*

The effect of deterioration by outdoor exposure on LDPE/TPSS/KCF composites was evaluated qualitatively by weight loss data. Weight losses of polymer strips upon weathering could be assumed as an indicator of biodegradation in the natural environment. Weight loss data retrieved from the samples loaded with KCF ranged from 10–40 wt.% after exposure to natural weathering for a period of 3 and 6 months. After recovery, the samples were washed, dried and weighed. Figure 5 summarizes the weight loss obtained. It was found that the weight loss observed over 3 months weathering was larger with increasing KCF content in the composite system. As expected, a prolonged exposure time resulted in a higher weight loss percentage.

As mentioned in the previous section, natural weathering involves the degradation of the composite sample by UV radiation, whereby the matrix (LDPE) undergo photo-oxidation reactions that involve the abstraction of a hydrogen atom by free radicals generated from hydroperoxides. These radicals then propagate, forming further radicals and result in chain scission of the polymer, thereby leading to a decrease in molecular weight [11]. Consequently, UV radiation of starch granules induces oxidative depolymerization and yield smaller molecules. The weight loss occurred after microorganisms consumed these fragments of the polymer as their food source. Starch consumption by microorganism results in presence of microorganisms, pores and crack on the specimen's surface as been viewed in SEM micrographs (Fig. 6 and 7). These porous structures enhance the accessibility of water, oxygen and microorganisms into the polymer matrix and caused fractures in the LDPE chains. In fact, cellulosic materials were also degraded into a lower molecular weight compound [15]. By some means, the exposed surfaces permit moisture penetration that promotes microbial attack on the starch/fiber and hydrolysis of the matrix component. These might contribute to the leaching out of the components to the outer surface and

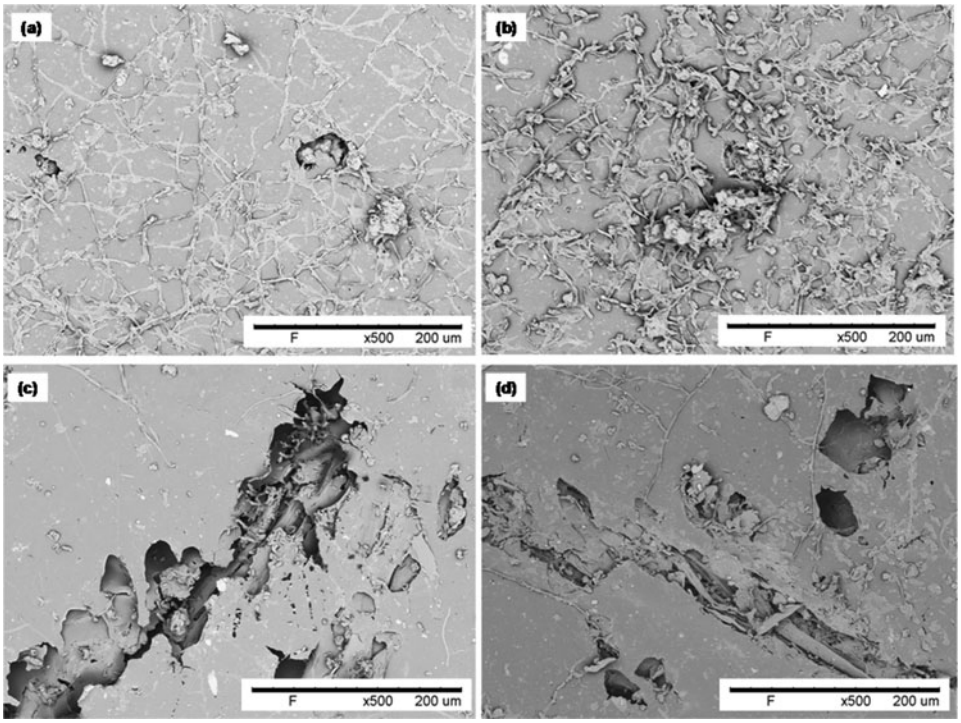


**Figure 6.** SEM micrographs of LDPE/TPSS blends reinforced with (a) 0 phr KCF (b) 10 phr KCF (c) 30 phr KCF and (d) 40 phr KCF after 3 months exposure to natural weathering.

resulted in overall weight loss. Thus, composites with lower KCF content degrade slowly and result in a low percentage of weight loss, but composites with higher KCF amounts increase the weight loss percentage.

It is suggested that the attack of outdoor agents takes place mainly on the exposed surface. The weight loss might be one of the factors for the decrease in tensile properties besides the cracks that occurred on the surface of the blends.

### *Morphological Properties*

Figures 6 and 7 show the micrographs of exposed surfaces of LDPE/TPSS/KCF composites after a 3-month and 6-month weathering. Referring to Fig. 6 (a), SEM micrographs of LDPE/TPSS blends signify minor changes on the surface microscopy. It seems that small amounts of deposits appeared on the surface of composites, believed to be the additives present in TPSS that leached out onto the surface [13]. The additives are likely to leach out from the LDPE matrix on prolonged weathering time (Fig. 7 (a)). Most of the additives had leached out due to the environment effect such as UV sunlight, rain and wind [16].

The micrographs for every degradation period indicated that the presence of KCF resulted in higher degradation rate in the blends. As can be seen, the surfaces of LDPE/TPSS/10KCF composites exposed to outdoor weathering for 3 months were occupied by fungus and micro pores (Fig. 6 (b)). The tiny white deposits observed on the surface probably represents impurities that either come from the surrounding materials or by the KCF. However, a small crack was found in LDPE/TPSS samples after 6 months of

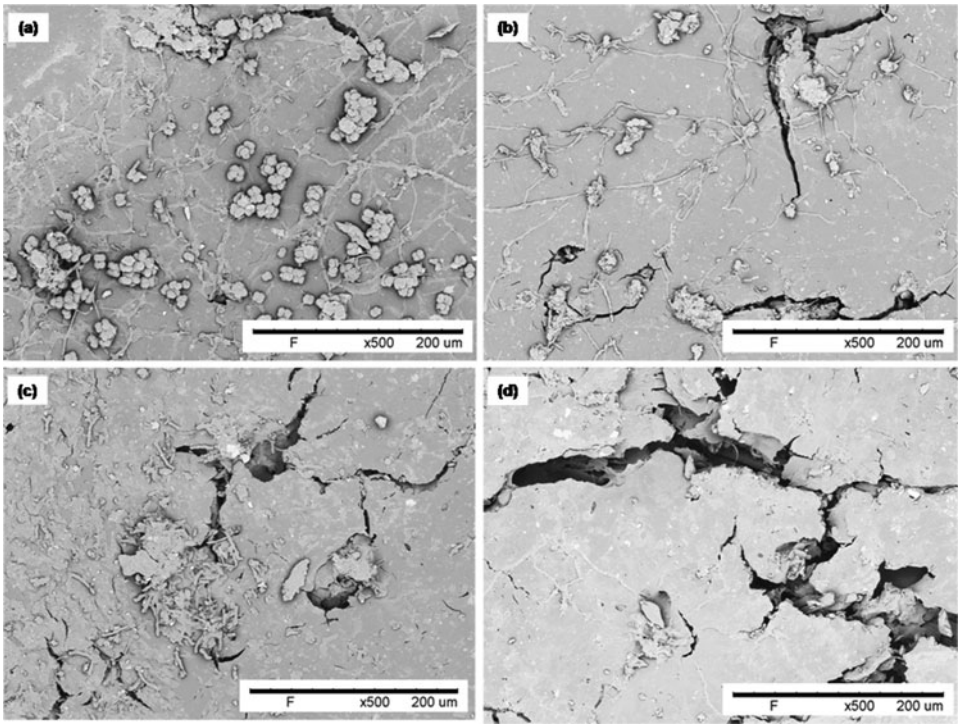


**Figure 7.** SEM micrographs of LDPE/TPSS blends reinforced with (a) 0 phr KCF (b) 10 phr KCF (c) 30 phr KCF and (d) 40 phr KCF after 6 months exposure to natural weathering.

weathering (Fig. 7 (b)). The matrix was probably attacked by thermal and UV degradation. It is worth noting that, as the content of the KCF increased, more fungus colonized on the surface of the samples and larger pores were observed. In addition, the weathered surface in Fig. 6 (c) revealed that the surface cracks exposed the embedded fibers to the surface. It is possible that upon weathering the hygroscopic nature of KCF increased the moisture absorption and thus facilitates the formation of severe cracks on the composites [10]. Thus, the fibers were easily seen and the detachment of KCF from the matrix proved that they had undergone degradation leading to poor fiber-matrix interactions and consequently deterioration of mechanical properties [9]. However, biodegradation by fungus was obviously shown in Fig. 7 (c). Higher KCF content caused more fungus colonies to form on the surface of the blends and the localized consumption by the microorganism resulted in the appearance of pores and cracks. The surface of LDPE/TPSS/KCF composites after 6 months of weathering was remarkably changed to a rougher surface with large continuous crack formation and detachment of starch and fibers on the surface. It appears that the exposed surface showed deeper crack formation compared to 3 months of weathering.

As the KCF content increased, the severity of the cracks was evident in the micrograph (Fig. 6 (d)). After 6 months of natural weathering exposure of some large cracks appeared on the surface (Fig. 7 (d)). Crack formation probably occurred due to thermal contraction and material loss during prolonged weathering time. Surface cracks had propagated deeply into the inner part of the samples [10]. The extreme daily changes in temperature and humidity cause surface cracking on the composites [3]. As can be seen, cracks can be found throughout the blend surface, indicating the occurrence of photo and thermal degradation.

As discussed above, the increase in carbonyl index signifies that his has occurred. Generally, biodegradation of the composites are believed to occur resulting from microbial attack to polymer chains in amorphous regions, weight loss then begins followed by a deeper microbial invasion along with humidity leading to the extensive degradation of the material [17].

## Conclusions

The tensile strength, Young's modulus and elongation at break showed a progressive decay as KCF loading increase after exposure to natural weathering conditions for a period of 3 and 6 months. This is evident in SEM micrographs that at low KCF loading, the morphology exhibited a relatively rough surface with some pores occupied by fungus. However, composites with greater KCF amount viewed fungal colonization and large crack formation. The presence of both carbonyl and hydroxyl groups signified the deterioration of components during the period of weathering. With increasing KCF content and weathering period, the observed weight loss of the composite increased assuming microbial attack on the starch/fiber and hydrolysis of the matrix component.

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