The Effect of Kenaf Core Fibre Loading on Properties of Low Density Polyethylene/Thermoplastic Sago Starch/Kenaf Core Fiber Composites

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Abstract: *The rising concern about plastic waste disposal problems and the need for more versatile polymer-based materials has led to an increasing interest on blending synthetic polymer with degradable materials. In this present study, low density polyethylene (LDPE) and plasticised sago starch or so-called thermoplastic sago starch (TPSS) blends containing different percentage of kenaf core fibres (KCF) were prepared. The effects of different fibre loading (10–40% by weight) on the processing and mechanical properties were investigated. In order to further justify the obtained properties, the fabricated composites were characterised by Scanning Electron Microscope (SEM), Fourier Transform Infrared (FTIR), Thermogravimetric Analysis (TGA), Differential Scanning Calorimetry (DSC) and water uptake behaviour. In this work, the blend ratio of LDPE/TPSS was fixed at 90/10 (weight percentage) respectively. The results showed that the incorporation of KCF into the blend caused a considerable improvement in tensile strength and Young's modulus. Optimum strength was obtained at 6.704 MPa. FTIR analysis revealed slight changes in band position and intensities due to hydrogen bond formation occurring after the inclusion of fibre. Whereas, decomposition temperature (T_d) was improved because of higher thermal stability of fibres and DSC results illustrated the phase compatibility between components in the composite system. Moreover, water uptake tended to increase as the hydrophilic character of KCF impart to the composites.*

Keywords: Kenaf core fibres, thermoplastic sago starch, low-density polyethylene, degradability, mechanical properties

1. INTRODUCTION

Plastic is a typical organic polymer derived from petrochemicals, which are produced and used worldwide. This is due to their ease of processing, low

energy consumption during fabrication and their inertness, which makes them suitable to be used in all areas.[1] The most commonly used synthetic polymers is polyethylene since it is highly hydrophobic and has high molecular weight. Particularly, low-density polyethylene (LDPE) is the most widely used polyethylene grade, due to its relatively good mechanical properties combined with a competitive market price.

However, the ever-growing production and rapid expansion in plastic material consumption have led a tremendous problem in plastic waste disposal management. Plastic is inherently inert to the microorganisms or the chemicals in an environment.[2] Thus, the disposal of a high volume of plastic, which takes a long time to decompose posses huge environmental problems.[3] In tackling the environmental issues associated with conventional plastic, many efforts have been made in recent years to develop biodegradable materials. These materials may be synthetic, natural or combination of both.[1]

In order to impart degradability of polymers, modified plastic derived from blend with agricultural feedstock, e.g., starch offers an elegant approach to meet the requirements and ecology utilisation of resources and to guarantee at least partial degradation.[4] Starch is considered as one of the most promising available natural polymers for the development of biodegradable materials due to its attractive combination of attributes such as low cost, abundant availability and thermoplastic behaviour, in addition to being biodegradable.[3]

Starch consists of two different polysaccharides structures, which are the linear amylose and highly branched amylopectin. And, the intermolecular and intramolecular hydrogen bonds between hydroxyl groups of starch molecules lead to the crytallinity in native starch.[5] In the presence of plasticiser such as glycerol, it can behave like a thermoplastic material; at high temperature and under shear, it can readily melt and flow, allowing its use as an extruded or injected material, similar to most conventional synthetic thermoplastic polymers.[6]

In a previous work, it was found that, a higher amount of starch can be incorporated with LDPE when used in a thermoplastic (plasticised) form and the so-called thermoplastic starch (TPS).[7] The major sources of starches commonly used for production of biodegradable polymers are maize, tapioca and rice.[8] However, very few reports are available on the properties of sago starch blend. Malaysia is one of the countries with a large cultivation of sago palm. The rather excessive production with regards to the current needs of developing degradable materials make sago starch attractive and promising filler for polyethylene.

However, it was found that the addition of TPS as a minor phase into synthetic thermoplastic matrix as a major phase causes a reduction of the

mechanical properties.[9] Incompatibility arising from the hydrophilic nature of starch and the hydrophobic nature of polymers as well as the tendency of starch to absorb water results in reduced mechanical performance of the blend.[10] A previous work observed that a blend of TPS from wheat starch (0–22% w/w) with LDPE exhibited a decrease in the Young's modulus.[7] It is interesting to note that, the immiscibility between hydrophilic and hydrophobic properties of starch and LDPE respectively responsible for the reduction of mechanical properties. Apart from that, the degradation duration is still very high due to the non-biodegradable synthetic polymers.[11]

One interesting approach to improve the mechanical properties and impart degradability is through the use of natural fibres to reinforce the LDPE/TPS blend. Natural fibre is preferred as it can be completely biodegraded in the natural environment and is fully sustainable. Previous studies have investigated synthetic and degradable polymer blends filled with natural fibres. For instance, cotton fibres are described in the literature as a reinforcement of starch-based thermoplastic polymers and PE blends.[9] Most natural fibres are composed of cellulose, hemicellulose and lignin in different relative amounts.[12] Thus, when natural fibres are mixed with thermoplastic starch and its blends, their mechanical properties become notably enhanced. This fact has been attributed to the chemical similarity of starch and fibres, providing good compatibility between them.[13]

Kenaf-reinforced composites are one of the most researched areas. Kenaf is an herbaceous annual plant that is available in a wide range of weather conditions. It has been actively cultivated since it grows quickly and can be harvested at a very low cost. Among lignocellulosic fibres, kenaf is attracting a special attention due to its good mechanical properties and its increasing cultivation in many developing and developed countries.[14] Kenaf fibres provide high stiffness and strength values. They have a higher aspect ratio making them suitable as reinforcement in polymer composite.[15] In view of the recent global environment issues and inadequate fibre resources, researches worldwide have begun to show interest in exploiting the full potential of kenaf and its diverse uses.[16]

Therefore, the main objective of this present study is to evaluate the potential use of sago starch (plasticised) and kenaf fibers with the polyolefin (LDPE). The effects of different amount of KCF incorporated in LDPE/TPSS blend were investigated. The work focuses on the characterisation of mechanical, thermal, morphological and water uptake properties of these composites.

2.   EXPERIMENTAL

Sago starch (13% moisture) was obtained from the Land Custody Development Authority (LCDA), Sarawak, Malaysia. It had an average particle size of 20 µm and decomposition temperature of 230°C. Glycerol (plasticiser) 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^{-1} was obtained from Titan (M) Sdn. Bhd. (Malaysia). Kenaf fibre (core) with an average length of 5 mm was supplied by National Kenaf and Tobacco Board (LKTN), Pasir Puteh, Kelantan, Malaysia. The fibres were subjected to a grinding process, which yields particles approximately 70 to 250 µm in diameter. KCF were then dried for 3 h at 70°C using vacuum before being used in the subsequent composite fabrication.

2.1   Sample Preparation

Sago starch powder was vacuum-dried by heating at 80°C for 24 h before blending and processing. The dried sago starch was then pre-mixed with glycerol by using a high-speed mixer. The weight ratio of sago starch and glycerol was maintained at 65:35 (by weight percent). The blend was stored overnight to allow the diffusion of glycerol into starch granules, which would help the melt-mixing process. TPSS was melt-blended with LDPE and kenaf fibre using an internal mixer (Haake Rheomix Mixer, Model R600/610) at 150°C and speed of 50 rpm in order to obtain a homogeneous sample.

Based on our previous work, the optimum ratio between LDPE/TPSS was obtained at 90/10. Thus, for this work, the ratio of LDPE/TPSS was fixed at 90/10 and different loading range of KCF (i.e., 10–40 wt%) was used. The processed samples were then compression-molded in an electrically heated hydraulic press (Kao Tieh Go Tech Compression Machine) at 150°C into a 1-mm thickness sheet. Table 1 shows the formulation of the composites.

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

Sample	Composition (wt%)	
	LDPE/TPSS	KCF
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

2.2   Measurement of 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^{-1} was used and the test was performed at temperature 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.

2.3   FTIR Spectroscopy Analysis

The functional groups and chemical characteristics of LDPE/TPSS/KCF composites 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.

2.4   Thermogravimetric Analysis

LDPE/TPSS/KCF composite samples were cut into small pieces (5–10 mg), which were then tested using thermogravimetric instruments (Perkin Elmer, Pyris Diamond TG/DTA) under nitrogen atmosphere within a temperature range from room temperature to 550°C at a heating rate of 15°C min^{-1}. Thermal degradation temperature was reported by the onset degradation temperature where the weight loss started to occur.

2.5   Differential Scanning Calorimeter (DSC)

The Differential Scanning Calorimeter (DSC) measurements were performed in a Perkin-Elmer DSC 7. About 10 mg of the samples was first heated from room temperature to 190°C at 5°C min^{-1} and held there for 5 min to eliminate the thermal history, cooled to room temperature and then reheated to 190°C at 5°C min^{-1} to trace the melting behaviour. Each run was performed under nitrogen atmosphere. The melting temperature (T_m) and heat of fusion of (ΔH_m) samples were determined from the peak maximum and the area under peak, respectively. The degree of crystallinity of the composites was calculated using the following equation:

$$X_c (\%) = \frac{\Delta H_m}{\Delta H_f} \times 100 \qquad (1)$$

where ΔH_m is the melt entalphy of the semicrystalline LDPE blends and ΔH_f is the heat fusion for crystalline LDPE.

2.6   Morphology Evaluation

Scanning electron micrographs of tensile fracture surfaces of LDPE/TPSS/KCF composites 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. The image results were analysed to investigate the distribution of natural fibres in polymer matrix and their interaction.

2.7   Water Absorption

Water uptake measurement was carried out as per ASTM D570. Newly prepared samples were first dried in an oven at 70°C for 24 h until a constant weight was attained and then dipped in the distilled water at ambient temperature. After immersion for a specific interval, the samples were then removed from the water, gently dried by wiping with a clean cloth, and immediately weighed to the nearest 0.001 g. The percentage of water absorption was calculated as followed:

$$WA\,(\%) = [(M_1 - M_0)/M_0] \times 100 \qquad (2)$$

where M_0 and M_1 were the dried weight and final weight of the sample, respectively.

## 3.   RESULTS AND DISCUSSIONS

3.1   Processing Properties

The melt processing characteristics of the LDPE/KCF/TPSS composites have been studied with processing torque-time curves. Figure 1(a) shows the processing torque of the melt mixed blends with different KCF loadings. A similar pattern of processing torque curves was observed for all composites. It can be observed that, an initial torque revealed corresponded to the initiation of rotor and charging of solid material into the mixing chamber, which in this case is the LPDE. It is assumed that, the increase in torque is due to high shear force to rotate the rotors in the presence of LDPE prior to melting. The loading peak is highly dependant on the charged amount of LDPE.

For LDPE/TPSS blend, the torque is higher compared to its composites. This is originated from the greater amount of LDPE dosage subjected into the mixing chamber. Torque decreased with the increase in KCF loading due to the reduction of charged weight of LDPE significantly. After loading, the blend started to melt under shear and temperature. After 3 min, the torque began to

decrease gradually due to reduction in viscosity, which indicates completion of melting.[17]
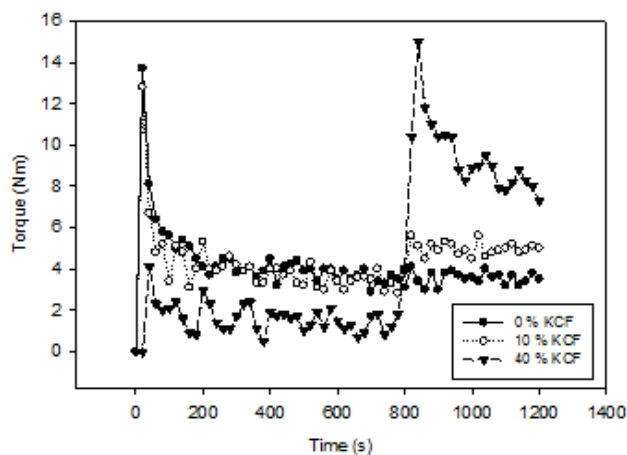
(a)
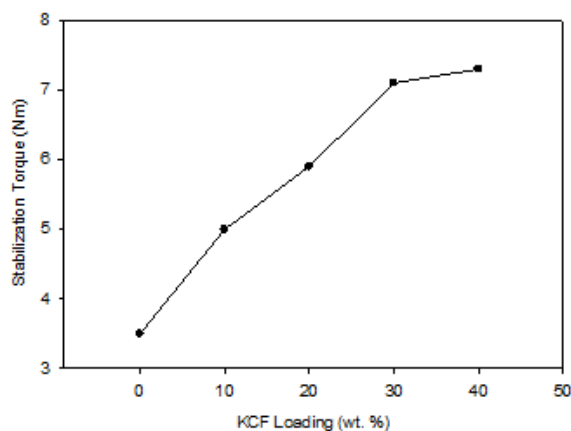
(b)

Figure 1: The plots of (a) processing torque and (b) stabilisation torque of LDPE/TPSS/KCF composites at different KCF loading.

The addition of minor content of TPSS at min-3 did not show any prominent loading peak. It is believed that the melt viscosity is almost stable due to the plasticisation of the blends with the presence of glycerol in TPSS.[18] However, an increase of torque can be observed as KCF were loaded into the molten LDPE/TPSS blend at min-13. The inclusion of KCF caused a reduction of the mobility of polymer chain and thus increased the melt viscosity.[17] Therefore, greater KCF loading in the composite exhibited higher torque values.

Stabilisation torque is measured as the average value around 18–20 min of mixing. The stabilisation torques of composites at different KCF loadings are shown in Figure 1(b). Evidently, the stabilisation torque gradually increased as higher KCF amount was loaded in the composites. It is worth noting that, the torque became more stable when the KCF were homogenised and dispersed within the polymer matrix. Thus, it can be postulated that, higher KCF loading required high shear force, resulting high melt viscosity and yielding high stabilisation torque.

3.2   Tensile Properties

Figure 2 shows the plots of tensile strength, Young's modulus and elongation at break of LDPE/TPSS blend reinforced with different amount of KCF. It can be seen from the plot that the addition of 10% (by weight) KCF into the LDPE/TPSS blend, the initial strength trebled up to 6.704 MPa. The enhancement in the strength is indicative of a good degree of adhesion between fibres and matrix, and a relatively high aspect ratio of fibres, exceeding critical level.[19] Thus, it is believed that, the addition of 10% (by weight) KCF is sufficient to provide the reinforcing effect to the composite system. Stress is transferred efficiently from matrix to the fibre.

Another contributing factor could be the chemical similarity of both TPSS and KCF, by virtue of having hydroxyl groups. The existence of such interaction increases the hydrogen bond formation as revealed by IR spectra in Figure 3 and the fibre surface wetting presented by SEM micrograph in Figure 6. The results were in accordance to the findings reported by Prachayawarakon et al.[9] and Ma et al.[20]

However, the incorporation of 20%–40% (by weight) KCF into the composite system caused a reduction in tensile strength. Tensile strength tended to decrease due to excessive content of KCF that caused the discontinuity of the matrix.[8] Weight fraction is perhaps the most important factor, since most of mechanical properties increase with increase amount of fibre, until a maximum amount, because at higher reinforcement loading, low dispersion of fibres in the matrix can occur.[3] Apart from that, the reduction of strength might be due to incompatibility of highly hydrophilic KCF and non-polar hydrophobic LDPE.

Because of different polarities, a weak interfacial adhesion between KCF and LDPE appeared, providing a site for failure to initiate and propagate. Reduction in the effective surface area of fibre phase yielded an inefficient stress transfer from matrix.[21] As a result, the tensile strength was not improved. The drop in tensile strength became more drastic at higher KCF loading (40 wt%). This is because fibre-fibre interaction became more prominent than fibre-matrix

interaction with increasing KCF content. The applied force could not be transferred efficiently from polymer matrix to the fibre. This is justified by the SEM micrograph in Figure 6.
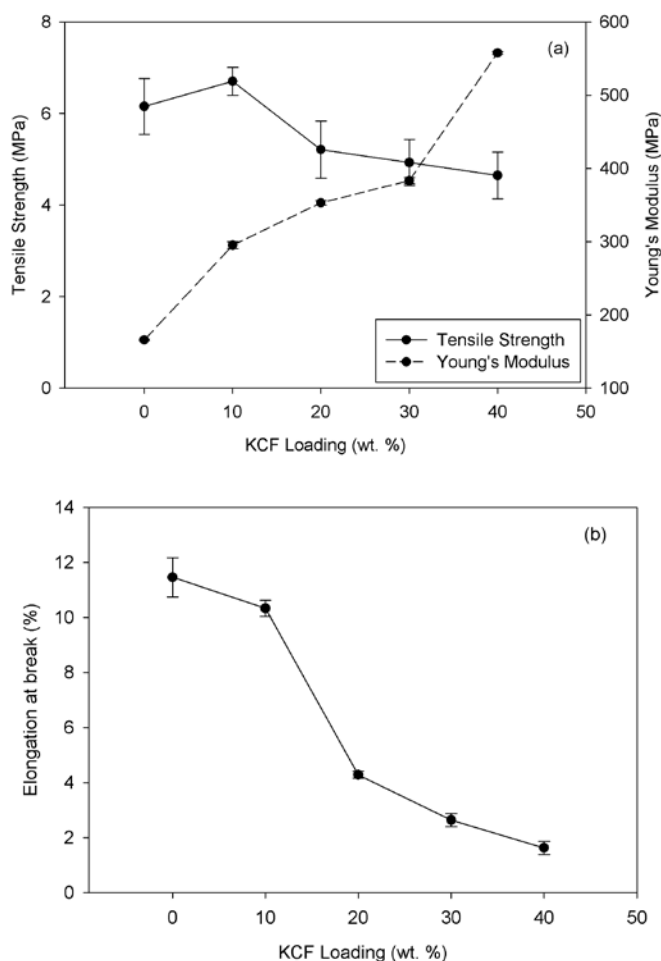

Figure 2:   The plots of (a) tensile strength and Young's Modulus and (b) elongation at break of LPDE/TPSS/KCF composite at different KCF loading.

The incorporation of KCF was found to increase the tensile modulus of LDPE/TPSS blends [as shown in Figure 2(a)]. The moduli were increased from 172.5 MPa until 557.75 MPa when 10%–40% (by weight) of KCF was incorporated in the composite system. The addition of KCF was expected to increase the modulus resulting from the inclusion of rigid filler particles in the soft matrix.[17] Additionally, this results suggest that, the stiffening effect had occurred because of the existence of fibre inside the composites.[22] Tensile

modulus indicates the stiffness of a composite, i.e., the higher the tensile modulus, the stiffer the composite is.[23] This finding is in agreement with the work reported by Abdul Khalil et al.[23] and Ismail et al.[24] whereby, the incorporation of starch or cellulosic fibre had improved the stiffness of the composite materials.
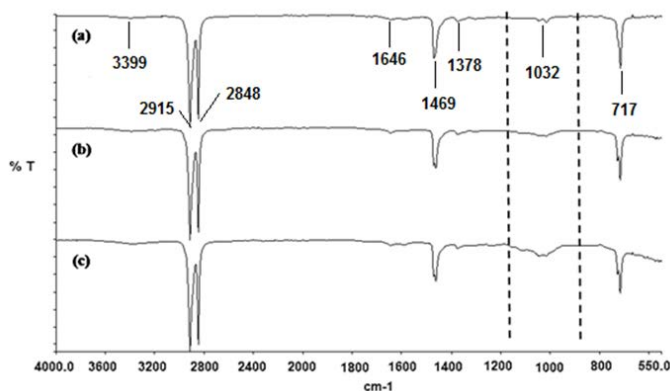

Figure 3:   IR spectra of LDPE/TPSS/KCF composites at different KCF loading (a) 0%, (b) 10% and (c) 40% (by weight).

On the contrary, the addition of KCF brings about a drop of elongation at break values. This is in agreement with another report.[25] This effect is expected considering that the high crystallinity of fibres decreases the polymer chain mobility and deformability of a rigid interface between fibre and matrix, which leads to an inevitable decrease in the degree of ductility of material. Besides, physical incorporation of starch in LDPE weakens the forces between LDPE layers and also, starch (a low molecular weight polymers) has lower elongation than LDPE. Since there is no chemical interaction between both constituents, starch inclusion forms discontinuity, resulting in lower elongation.[13] In synthetic polymer blends, the addition of second immiscible phase to a ductile matrix material usually significantly decreases the elongation at break.[6]

3.3   IR Spectroscopic Study

IR spectra of LDPE/TPSS blend reinforced with different KCF loading were shown in Figure 3. The observed peaks are the functional groups of LPDE, TPSS and KCF. However, there are some slight changes in band positions and intensities. It is known that, if two polymers are compatible, a distinct interaction (hydrogen bonding or dipolar interaction) exists between the chains of one polymer and those of the other, causing the IR spectra of composites to change.
It can be seen in Figure 3(a) that the LDPE/TPSS composite exhibits IR main peaks positions at 2915 cm^{-1}, 1469 cm^{-1} and 717 cm^{-1} representing C–H stretching, –CH$_3$ bending and –CH$_2$– vibration, respectively. However, in the

case of LDPE/TPSS composite filled with TPSS, IR peaks related to KCF become prominent. It is evident in Figure 3(b) that when KCF is added to the composite, it is characterised by a broad absorption peak occurring at 1260–900 cm^{-1} attributed to the stretching of C–O group.[26] In fact, a slight increase in the IR peak could be observed as more KCF was loaded to the composite (40 wt%) as shown in Figure 3(c). These changes could be interpreted in terms of the rearrangement of hydrogen bonds between starch and fibre. The change in IR spectra indicates a distinct interaction between the chains of polymers.

Apart from that, starch is characterised by two strong and broad absorption peaks occurring at 3000–3900 cm^{-1} (O–H) and at 1000–1250 cm^{-1} (C–O–C) stretching. The presence of OH group peak at 3360.46 cm^{-1} is related to the presence of amylose and amylopectin in sago starch. The characteristic peaks appeared due to the intermolecular hydrogen bond formation occurring when TPSS were incorporated into the composite.

Whereas, the peak position at approximately 1646 cm^{-1} is due to the bound water present in the starch.[6] All the characteristic peaks of LDPE and TPSS have appeared in the blends. These results indicate that the KCF is distributed within LDPE/TPSS blend. And, it is believed that the hydrogen bond formation is correlated with the tensile properties discussed earlier.

3.4   Thermogravimetric Analysis (TGA)

Thermogravimetric analysis (TGA) of the composite was carried out to assess its thermal stability and degradation temperature. The onset degradation temperature and thermogravimetric curves for different LDPE/KCF/TPSS composite samples are shown in Figure 4. It can be observed that thermal degradation of the samples occurred in three steps: 1st region (250°C–280°C), 2nd region (335°C–400°C) and 3rd region (above 400°C). The thermal degradation of LDPE/KCF composite occurred in a two-step degradation process. The LDPE/TPSS exhibited initial mass loss from approximately 302°C to 370°C, mainly due to the decomposition of TPSS. After that, second thermal degradation step of LDPE was observed.[27] The thermal degradation appeared around 488°C is associated with the greatest weight loss, and was due to thermal degradation of the LDPE backbone.[19]
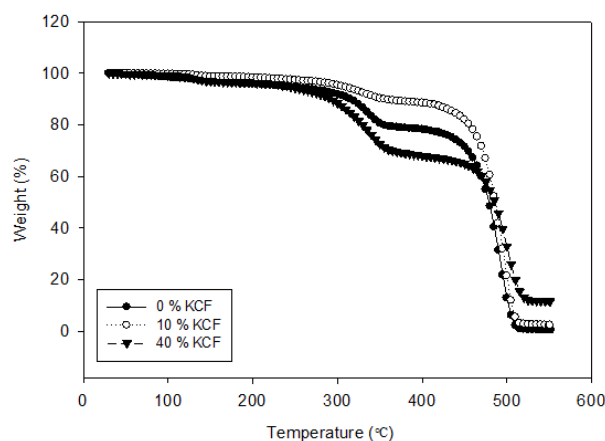
Figure 4: TGA thermograms of LDPE/TPSS/KCF composites at different KCF loading.

With the incorporation of KCF in the LDPE/TPSS blend, it was observed that the decomposition temperature (T_d) found at 176.2°C and 295.8°C were due to the decomposition of glycerol and starch, respectively. The process continued progressively up to 300°C, where thermal degradation of starch occurred.[16] T_d value of the TPSS was 332°C when 10% (by weight) of KCF was incorporated into the composite system. This is attributed to higher thermal stability of fibre and phase compatibility between TPSS and KCF. Due to their similar chemical structures, the two structures was linked through hydrogen bonds.

It is noted that the T_d of LDPE component was obtained at the temperature of 491°C. Similar pattern was observed for LDPE/TPSS blend with 40% (by weight) KCF. It was also found that the maximum weight losses were 99.38%, 97.51% and 88.28% for the KCF loading of 0, 10 and 40% (by weight), respectively. According to the KCF content, it is believed that the reduction in weight loss was the result of the improvement in thermal stability.[28]

3.5   Differential Scanning Calorimetry (DSC)

Figure 5 illustrates the second heating cycles of DSC thermograms for LDPE and its blends, whereas their melting temperature (T_m), enthalpy of melt (ΔH_m) and degree of crystallinity (X_c) are summarised in Table 2. DSC thermograms of LDPE and its composites show one endothermic transition peak, indicating that the material demonstrates compatibility between components in blends. It can be observed that, the endothermic peak at 108.3°C assigned to the melting temperature of pure LDPE. In comparison with the LDPE and its blends, the DSC results show that there is a slight decrease in T_m of LDPE blend with the addition of fibre. It is believed that, the fibres create hindrance in the melting of

LDPE. This decrease can be explained based on the decrease in the crystalline portion in the system.
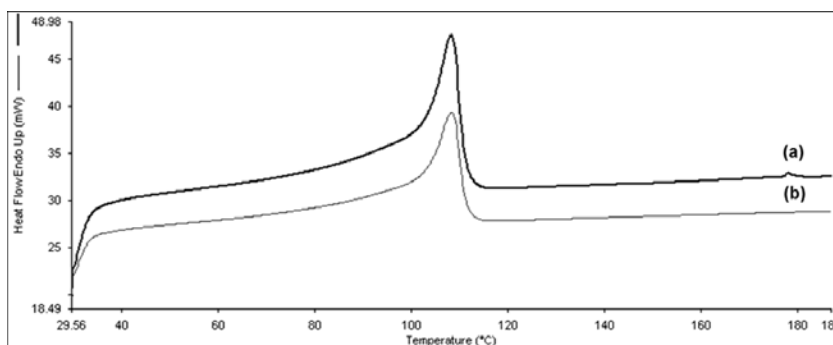

Figure 5: DSC thermograms of (a) LDPE and (b) LDPE/TPSS/KCF composites.

Table 2: DSC data for LDPE/TPSS/KCF composites.

Samples	T_m (°C)	ΔH_m (J g^{-1})	X_c (%)
LDPE	108.3	74.57	27.02
LDPE + TPSS + 10% KCF	108.2	71.25	25.82

It also can be observed that there was a reduction in enthalpy of melt (ΔH_m) for the blends compared to pure LDPE. ΔH_m is obtained from the total area of the melting peak. The reduction of ΔH_m is correlated with the decrease in LDPE content in blend specimen.[29] In addition, there is a slight decrease in crystallinity of the LDPE and its blends. This suggests that the greater the compatibility between components in blends, the weaker their ability to crystallise will be.

3.6   Morphology

Polymer properties can be determined from the morphological structure. The SEM micrographs of tensile fracture surface of LDPE/TPSS blend and morphological effects of KCF incorporation are depicted in Figure 5. Rough surface of TPSS could be observed in Figure 6(a). Without KCF loading, the tensile fracture surface of LDPE/TPSS blend indicates the spots of starch granules within the LDPE continuous phase. Such particles seem to be embedded in LDPE matrix, although there were a few gaps and separate starch particles were visible. Surface roughness in the blend appeared due to the resulting incompatible blend of TPSS and LPDE. The immiscibility of the two components caused the weak interfacial adhesion and starch granules tend to agglomerate,

resulting in low strength. The tensile properties obtained in previous section are evidenced by this morphological structure.

Figure 6(b) illustrates the tensile fracture surface of LDPE/TPSS blend reinforced by 10% (by weight) KCF. It can be observed that the fibre is embedded in the matrix and its surface wetted by the matrix. There were no obvious starch granular figures. This infers that, with the increase in fibre content, the starch granular fusion was affected by the fibre during processing. The fibre seemed to be well dispersed in the polymer blend and the fracture surface showed intimate mixing of fibre within the matrix. The good dispersion corroborates the presence of chemical bonding between KCF and TPSS probably due to the formation of hydrogen bonding,[26] good stress transfer from matrix to fibre and the reinforcing effect of fiber.[27] It is an evidence of phase compatibility and yielded tensile strength.
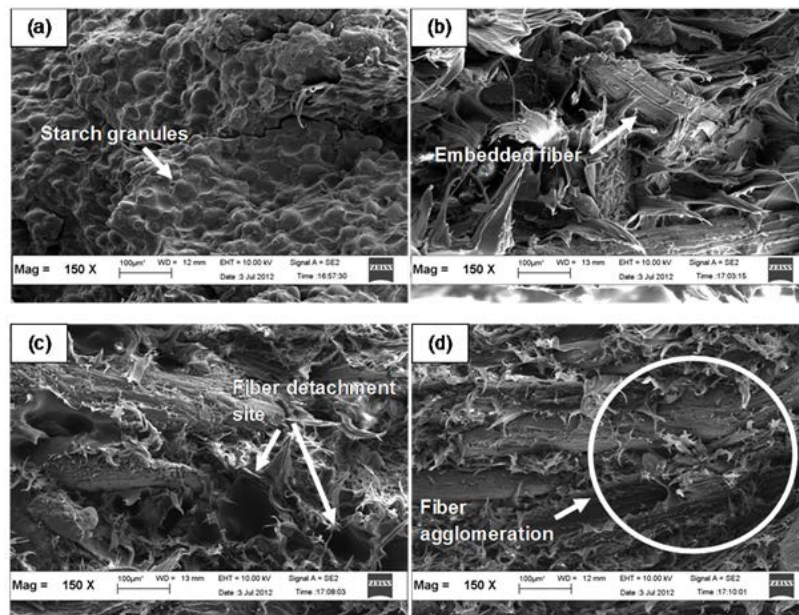

Figure 6: SEM micrographs of LDPE/TPSS/KCF composites at (a) 0% (b) 10%, (c) 20% and (d) 40% (by weight) of KCF loading.

On the other hand, Figure 6(c) illustrates the tensile fracture surface of LDPE/TPSS/KCF composite when KCF were loaded at 30% (by weight). In this case, the presence of fibres was more visible. KCF appeared not wholly embedded into matrix. Moreover, the fibres were strongly damaged and the occurrences of some debonding phenomena indicated poor adhesion between fibre and matrix.[14] And, there was even formation of holes as a result of fibre

detachment in matrix. The fracture surface micrographs in Figure 6(d) show poor wettability between fibre and matrix in which fibre agglomeration become more prominent in the composites. The non-uniform distribution of fibre gave rise to the formation of stress concentration points.[21] This situation is reflected in the mechanical properties. It is assumed that, increasing the fibre loading decreases interfacial adhesion and homogeneity. The mechanical properties already discussed also support these findings.

3.7   Water Absorption

Water sensitivity is a crucial measure for many products based on natural resources. The effects of KCF loading on the moisture absorption properties of LDPE/TPSS blend are shown in Figure 7. The absorption behaviour is related to the immersion of composites in water at ambient temperature and the measurement of the rate of water diffuse into the composite. With the incorporation of KCF, water absorption capacity of LDPE/TPSS blend increased progressively.

It can be seen that within the initial stage of absorption process, the kinetic of absorption is fast, and then the increasing rates slow down and finally reach a plateau. This plateau corresponds to the water absorption at equilibrium. All the composites (10–40 wt% KCF loading) have water absorption at equilibrium of the same order. It took about 5 days immersion for samples to reach absorption equilibrium. Theoretically, the existence of LDPE in the composite could dramatically restrain the water absorption and reduce water sensitivity due to the hydrophobic characteristic of non-polar LDPE. However, it is interesting to note that, the observed rise in water absorption capacity is highly dependant on KCF and TPSS content.
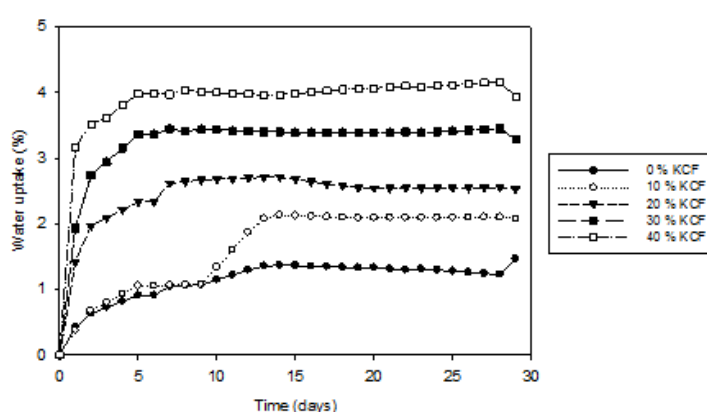

Figure 7: Water uptake of LDPE/TPSS/KCF composites at different KCF loadings.

It was found that, LDPE/TPSS blend had the lowest water uptake compared to its composites. The equilibrium water uptake for the blend was approximately 1%. While LDPE is hydrophobic, only TPSS is responsible for taking up water in the blend. TPSS is known to take up a large amount of water because it is hydrophilic due to the chemical composition of starch (amylopection and amylose), where hydroxyl group and oxygen bond with water are formed.[3]

The comparison of water uptake percentage by composites containing different amounts of KCF is also illustrated in Figure 6. It appears that the addition of KCF significantly increased the water absorption rate. The composite showed 1.09 to 3.98% water uptake with the inclusion of 10–40% (by weight) KCF, respectively. It is known that KCF is hydrophilic in nature and permeable to water. Incorporation of lignocellulosic fibre into the polymer composites generally augments the rate of water absorption ability by forming hydrogen bonding between water and hydroxyl groups of cellulose, hemicellulose in the cell wall.[17] Thus, the rise of water uptake is attributed to the hydrophilic nature of starch and fibre by virtue of the presence of abundant hydroxyl groups, which are available for interaction with water molecules.

It is also worth noting that, higher KCF content resulted in higher voids entrapped, causing higher water accumulating at the interface between fibre and matrix.[17] It was due to low degree of adhesion between hydrophobic LDPE and hydrophilic TPSS and KCF yield in cracks and voids between them, allowing easy penetration of the moisture and storage of water in the voids.[16] Thus, the percentage of water uptake arise significantly.

4.   CONCLUSION

The LDPE/TPSS/KCF composites can be prepared via melt-blending and compression moulding method. Tensile strength and modulus of the LDPE/TPSS blend could be effectively improved with the incorporation of KCF. However, elongation at break showed the opposite effect. The optimum strength was obtained at 6.704 MPa when 10% (by weight) of KCF loaded into the composite system. FTIR analysis showed the occurrences of bonding between functional groups of TPSS and KCF, which confirmed that the reactions between all components have been taking place in the composite system. Thermal degradation temperatures and thermal stability of LDPE/TPSS blends were also improved by the incorporation of KCF, whereas, DSC results demonstrated the compatibility of components in the composite system. Besides, the presences of fibres boost up the percentage of water uptake at equilibrium for the composites. The observed results of tensile properties, thermal and water uptake are explained

by interaction between fibre and matrix as evident from the fractographic studies conducted by SEM.

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