Mechanical and Thermal Properties of Sugar Palm Fiber Reinforced Thermoplastic Polyurethane Composites: Effect of Silane Treatment and Fiber Loading

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ABSTRACT: The aim of the present study was to develop sugar palm fiber (SPF) reinforced thermoplastic polyurethane (TPU) composites and to investigate the effects of fiber surface modification by 2% silane treatment and fiber loading (0, 10, 20, 30, 40 and 50 wt%) on the mechanical and thermal properties of the obtained composites. Surface treatment was employed to improve the fiber-matrix interface, which was expected to boost the mechanical strength of the composites, in terms of tensile, flexural and impact properties. Thermal properties were also investigated by thermal gravimetric analysis (TGA) and dynamic mechanical analysis (DMA) to assess the thermal stability of the developed composites. Furthermore, scanning electron microscopy (SEM) was used to study the tensile fracture samples of composites with a view towards evaluating the effects of fiber surface treatments on the fiber/matrix interfacial bonding. The findings of this study reveal that the silane treatment has determined good bonding and linkage of the cellulose fiber to the TPU matrix, hence contributing to enhanced mechanical and thermal properties of the composites. The composite formulation with 40 wt% sugar palm fiber loading showed optimum values such as 17.22 MPa for tensile, 13.96 MPa for flexural, and 15.47 kJ/m2 for impact strength. Moreover, the formulations with higher fiber content exhibited satisfactory values of storage modulus and thermal degradation, while their good interfacial adhesion was evidenced by SEM images.

KEYWORDS: Sugar palm fibers, silane treatment, thermoplastic polyurethane, sugar palm fiber-reinforced composites, mechanical properties, thermo-mechanical properties

1 INTRODUCTION

The current high dependence on synthetic materials in a wide range of domains may threaten the environment and human health. Among others, threats may emerge from the non-biodegradability of synthetic materials and their environmentally irresponsible disposal, causing unnecessary emissions of carbon dioxide. In due course, such excessive carbon dioxide emissions are harmful to the human respiratory system. In this context, the development of natural fibers is greatly welcomed and expected to deter the dangerous effects of synthetic fibers. However, despite the advantages of natural fibers, such as low cost of raw material, biodegradability, environmental friendliness and good mechanical properties, they also have some drawbacks, for example, high water absorbance, low thermal stability and debonding between the polymer matrix and natural fiber, which cause limitations in utilizing natural fibers for reinforcing polymer composites. Nonetheless, such drawbacks can be overcome by strengthening the properties of natural fibers through surface modification, thus improving the physical, mechanical and thermo-mechanical properties of natural fiber-reinforced polymer composites.

A great number of studies have been reported in the literature on plant fibers such as flax, kenaf, sisal, coir, flax, oil palm empty fruit bunch, etc. [1–3]. Among plant fibers, those of *Arenga pinnata*, commonly known as sugar palm, exhibit highly promising properties for use as reinforcement in composites. Sugar palm fibers (SPF) are abundantly available in Malaysia and South Asian countries. In addition to its wide availability and the ease at which sugar palm is grown and harvested, the properties of its fibers, such as high wear

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resistance, have certain advantages. Several researchers have reported work on sugar palm-based composites such as SPF/unsaturated polyester resin [3], SPF/epoxy [4], SPF/phenolic [5], SPF/polystyrene [6], SPF/sugar palm starch [7], SPF/thermoplastic polyurethane [8] composites and many more.

Bachtiar *et al.* [9] investigated the alkaline treatment with concentrations of 2% and 6% to improve the tensile properties of sugar palm fiber-reinforced thermoplastic polyurethane (TPU). The study showed that the tensile strength of the composites containing a 30% weight fraction of alkali-treated fiber was lower than that of untreated composites. Meanwhile, the composites with 2% treated fiber had a higher tensile modulus of 440 MPa compared to the 6% alkali-treated fiber.

Kenaf fiber-reinforced thermoplastic polyurethane was produced by the process of internal mixing and hot molding [10]. Various fiber loadings (of 20% 30%, 40% and 50%) led to different properties of the kenaf-reinforced thermoplastic polyurethane. The results showed that the 30% kenaf/TPU composite presented the highest values for a tensile strength of 89 N/mm^2 and flexural strength of 148 N/mm^2.

Rashid *et al.* [11] reported the effects of alkali and seawater treatments on SPF. They found the physical properties of SPF were most affected by the seawater treatment, whereas both physical and chemical characteristics were affected by the alkali treatment. In another study, Rashid *et al.* [5] investigated the mechanical performance of seawater-treated SPF-reinforced phenolic composites with different fiber loadings (0–40 vol%). The results indicated that the mechanical properties of the composites were improved upon incorporation of fibers. Thus, the composite with 30 vol% particle loading exhibited the optimum values for flexural, compressive, and impact strength. Therefore, it can be argued that the seawater treatment of SPF contributed to improving the mechanical properties of the composites, compared with those containing untreated SPF.

Zhou *et al.* [12] evaluated the effects of silane treatment on sisal fiber. The treatment resulted in the formation of a film layer, consisting of siloxane and polysiloxane, on the fiber surface by silane adsorption. FTIR spectroscopy indicated that chemical bonds were formed between the silane coupling agent and the sisal fiber. In addition, Asumani *et al.* [13] investigated the effects of combined alkaline and silane treatment on the tensile and flexural properties of kenaf/PP composites. They found that the alkali treatment followed by the 3-aminopropyltriethoxysilane treatment significantly improved the tensile and flexural properties of short-fiber nonwoven kenaf polypropylene composites. Morphological observation by scanning electron microscopy indicated that the improvements in the tensile and flexural properties resulting from the alkali-silane treatment were due to better bonding between the kenaf fibers and the PP matrix.

Some previous studies also show that silane treatments improve the surface roughness and the performance of fibers for composite applications, and will provide sugar palm fibers suitable for reinforcing thermoplastic polymer matrices [14]. Treated SPF-reinforced polymer matrix composites have been reported to exhibit an even better performance than some other natural fiber composites [9, 15, 16]. It is known that the absence of chemical treatment leads to a poor interface between fiber and polymer matrices. Tests on treated fibers revealed their better properties compared to those of untreated fibers [17–19].

The major problem that continuously occurs when combining natural fiber and polymer matrices is related to wetting. The hydrophilic nature of natural fibers is incompatible with hydrophobic polymer matrices, and hence the former have the tendency to form aggregates [3, 20, 21]. The hydrophilic polar natural fiber exhibits poor water resistance and does not attach properly to hydrophobic nonpolar polymer matrices. Poor interlocking of natural fibers with impregnated polymer matrices results in lower mechanical properties of the composites. Therefore, surface modification of natural fibers should be performed to improve the fiber/matrix interfacial bonding and to achieve better mechanical properties of the natural fiber polymer composites. Moreover, fiber loading or the amount of filler also contributes to the performance of natural fiber composites. In general, higher fiber loading is required to attain higher mechanical properties of the composites. Hence, the influence of fiber content on natural fiber-reinforced composites should be emphasized.

Natural fiber composites have been an issue for a long time, as seen by the many research articles, but very few if any attempts focusing on fiber modification of SPF with 2% silane treatment are available. To date, few studies have considered the mechanical and thermal properties of sugar palm fiber-reinforced thermoplastic polyurethane composites with variations in fiber loading (0–50 wt%). The present study focuses on the effect of fiber surface modification of sugar palm fiber by using 2% of silane treatment and the effect of fiber loading on the properties of sugar palm fiber-reinforced thermoplastic polyurethane composites. Various fiber loadings were used: 0, 10, 20, 30, 40 and 50 wt% of untreated and silane-treated sugar palm fiber, to reinforce thermoplastic polyurethane. The mechanical and thermal properties of 2% silane-treated and untreated SPF-reinforced TPU composites were studied in this work. The morphology of tensile fracture samples of composites was investigated by scanning electron microscopy. Figure 1 shows the

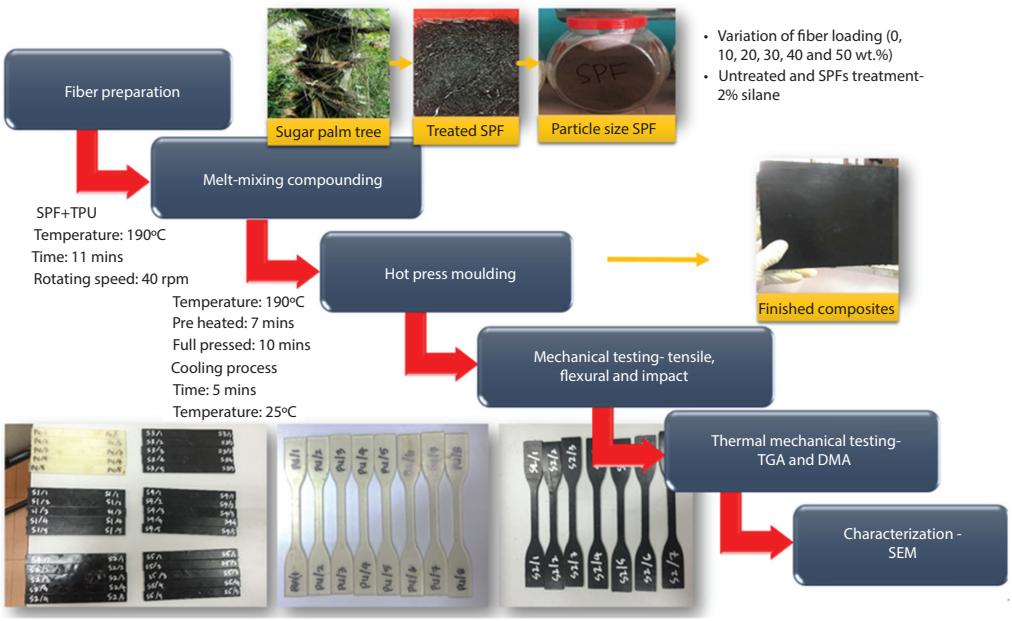

Figure 1 Schematic illustration of research methodology flow diagram.

research methodology flow diagram for the development, mechanical and thermo-mechanical properties of silane-treated sugar palm reinforced TPU composites.

2 EXPERIMENTAL

2.1 Materials

Estane® 58311 TPU was supplied in pellet form, with the density of 1.13 g/cm^3, by Innovative Pultrusion Sdn. Bhd. and was used as polymer matrix. Sugar palm fiber (SPF) was obtained from a sugar palm in Jempol, Negeri Sembilan, Malaysia.

2.2 Preparation of Sugar Palm Fiber

Sugar palm fibers were cut to a length of about < 8–10 cm. The fibers were then cleaned with tap water several times to get rid of any impurities attached to them. The SPF was kept in open air for a moment and dried in an air circulating oven at 60 °C for 48 h. The dry SPF was then ground to achieve the size of 10–15 mm using a plastic crusher machine and then a pulverizer machine. After this, the SPF particles were sieved to obtain the size of 125–250 µm.

2.3 Silane Treatment of Sugar Palm Fiber

The obtained SPFs of particulate size (125–250 µm) were immersed in a 2% silane solution for 3 h. For the silane treatment, 3-aminopropyl-triethoxysilane was mixed with a mixture of methanol-water (90/10 w/w) from the hydrolysis process. In order to get a solution pH of 3.5, the mixture was stirred with acetic acid for 10 min. Then the fibers were soaked in this solution for 3 h under agitation. After this time, SPF were washed with distilled water and decanted. Then, the fibers were oven-dried at 60 °C for 72 h to remove any moisture. This procedure is based on the modified method described by Atiqah *et al.* [22]. The reaction schemes are given as follows [23]:

$$CH_2CHSi\left(OC_2H_5\right) \xrightarrow{H_2O} CH_2CHSi(OH)3 + 3C_2H_5OH \quad (1)$$

$$CH_2CHSi(OH)_3 + \text{Fiber-OH} \rightarrow CH_2CHSi(OH)_2 \text{O-Fiber} + H_2O \quad (2)$$

2.4 Fabrication of SPF-Reinforced TPU Composites

The SPF/TPU composites were prepared using the melt-mixing compounding method, followed by hot-press molding. The drying process was carried out for sugar palm particles (of 125–250 µm size) and for thermoplastic polyurethanes in pellet form in an electric oven at 80 °C for 48 h. Six sets of SPF/TPU composites, with different loadings of sugar palm fiber (of 0, 10, 20, 30, 40 and 50 wt%) were prepared, as shown in Table 1. The process of homogenization was carried out at

Table 1 Relative amount of reinforcing materials in composites.

Sugar Palm Fiber (%)		Thermoplastic Polyurethane (%)	Composites
Untreated	0	100	0/100 USP/TPU
	10	90	10/90 USP/TPU
	20	80	20/80 USP/TPU
	30	70	30/70 USP/TPU
	40	60	40/60 USP/TPU
	50	50	50/50 USP/TPU
Treated	10	90	10/90 TSP/TPU
	20	80	20/80 TSP/TPU
	30	70	30/70 TSP/TPU
	40	60	40/60 TSP/TPU
	50	50	50/50 TSP/TPU

optimum processing parameters: 40 rpm, for 11 min at 190 °C, using a Haake Polydrive R600 mixer. Hot-press molding was performed using a Vechno Vation 40 ton compression molding machine, with the following regime: the samples were preheated for 7 min, full pressed for 10 min at 190 °C and, finally, cold-pressed for 5 min at 25 °C [24].

3 CHARACTERIZATIONS

3.1 Tensile Test

The tensile properties were determined using a universal testing machine from Lloyd Instruments. The tensile tests were carried out on flat dog-bone shaped samples, as per ASTM D618 test standards. The specimens were tested by a calibrated universal testing machine with a crosshead speed of 50 mm/min at 25 ± 3 °C. The tests were performed on six samples and the average of the six values was taken as a final result.

3.2 Flexural Test

The flexural properties of the developed SPF/TPU composites were determined according to ASTM D790-03 (3-point bending) standard. The testing was carried out using a Lloyd (Ametec) universal testing machine, with 60 mm span length and 12 mm/min crosshead speed, which was determined by using Equation 3:

$$R = \frac{ZL^2}{6D} \tag{3}$$

where R is the rate of crosshead speed (mm/min), L is the support span (mm), D is depth of beam (mm) and Z is the rate of straining of the outer fiber (mm/mm/min [Z = 0.01]).

Finally, flexural strength was calculated using Equation 4 and flexural modulus was obtained automatically by the software controlled machine:

$$FS = \frac{3FL}{2BD^2} \tag{4}$$

where F is the maximum load (MPa), L is the support span (mm), B is the width (mm) and D is the thickness of the tested sample (mm).

3.3 Impact Test

Standard Notched Izod impact test specimens were cut out from the SPF/TPU composite plates using an abrasive waterjet machine (Excel WJ 4080), according to ASTM D256. Averages of the five samples were taken as the final impact strength for selected treated and untreated SPF/TPU composites. The Izod impact strength was calculated using Equation 5:

$$\text{Impact Strength (Izod)} = \frac{\text{Energy absorption (kJ)}}{\text{Area (m2)}} \tag{5}$$

3.4 Scanning Electron Microscopy (SEM)

Morphological investigations were performed on untreated and treated SPF with a Hitachi S-3400N SEM operated at an emission current of 58 μA and an acceleration voltage of 5.0 kV; the working distance was set to 6.2 mm. Before the SEM analysis, the samples were coated with gold.

3.5 Dynamic Mechanical Analysis (DMA)

The DMA properties, including storage modulus, loss modulus and damping factor (Tan delta), were measured as a function of temperature (–100–140 °C), using a TA 2980 DMA equipped with a dual-cantilever bending fixture at a frequency of 1 Hz and a heating constant rate of 10 °C/min.

3.6 Thermogravimetric Analysis (TGA)

Thermal analysis was performed using a PerkinElmer Pyris 1 TGA. The samples (5–10 mg) underwent thermal scanning from 25 °C to 800 °C at a heating rate of 10 °C/min under nitrogen atmosphere. The nitrogen flow rate was 20 ml/min.

4 RESULTS AND DISCUSSION

4.1 Mechanical Properties

4.1.1 Tensile Properties

Figure 2 illustrates the tensile strength of untreated and silane-treated SPF reinforced thermoplastic polyurethane composites. Tensile strength provides a good indication of the interfacial bonding between the untreated or silane-treated SPF and the TPU matrix. It was found that the amount of untreated and silane-treated SPF (0–50 wt%) had a great influence on the tensile strength of the composites. Thus, in the untreated SPF-reinforced TPU composites with 0 to 20 wt% fiber content, an initial decrease of tensile strength was observed from 11.83 MPa to 7.80 MPa, while as the fiber content increased to 30 and 40 wt% the tensile strength increased to 8.85 and 14.52 MPa, respectively. However, the tensile strength of the composites decreased with further increasing of the fiber content to 50 wt%, as illustrated in Figure 2. The same behavior was exhibited by the silane-treated SPF-reinforced TPU composites. The optimum addition of silane-treated fiber (40 wt%) led to a 13% increase in tensile strength, from 15.22 MPa to 17.22 MPa. Our results reveal an improvement compared to the previous findings of Mohammed *et al*., who used 10% w/w untreated SPF to reinforce thermoplastic polyurethane using the extrusion method [8]. Moreover, SEM analysis revealed more fiber pull-out during the tensile tests; thus, a good interlocking surface between fiber and matrix is likely to contribute to higher tensile strength because of the tendency to avoid fiber pull-out.

The tensile modulus of the natural fiber-reinforced polymer composites increases with increasing fiber loading [1]. From Figure 2, it can be observed that the tensile strength of the composites reinforced with untreated and silane-treated sugar palm fiber increased with increasing fiber loading. Compared with untreated SPF/TPU, the tensile strength of 10, 20, 30, 40 and 50 wt% silane-treated SPF/TPU increased by 19%, 21%, 24%, 19% and 20%, respectively. The tensile modulus of the composite materials in general increases with an increase in SPF content up to 40 wt% of SPF content. The tensile modulus was almost two and a half times higher at 40 wt% SPF content than that of neat TPU (0/100 USP/TPU) composites, as depicted in Figure 3. Beyond this optimum fiber loading, the tensile modulus decreased; thus, at 50 wt% treated SPF it dropped to 354.69 MPa. The improvement in the mechanical properties of silane-treated (2%) fiber-reinforced polymer composites is due to the enhanced interfacial bonding, as has been also reported for silane-treated coir-reinforced polyethylene matrix [25].

4.1.2 Scanning Electron Microscopy

The SEM micrographs taken at 500x magnification of the untreated and 2% silane-treated SPF, after tensile testing, are shown in Figures 4 and 5. The images exhibit the morphology of the fracture surfaces for the various fiber loadings investigated in this study (0%, 10%, 20%, 30%, 40% and 50% by weight).

Thus, it may be observed that 0/100 USP/TPU (100% TPU) had a smooth surface compared to those of the SPF/TPU composites. Figure 4b,c indicates that the surfaces of untreated SPF/TPU composites were uneven, with some porous spots, which resulted in more fiber pull-out compared to the silane-treated SPF/TPU composites shown in Figure 4d,e. The fiber pull-out phenomenon contributed to poor mechanical properties in terms of tensile strength and modulus, as may be seen in Figures 2 and 3. Also, it should be noted that the morphology of untreated SPF composites (Figure 4b,c

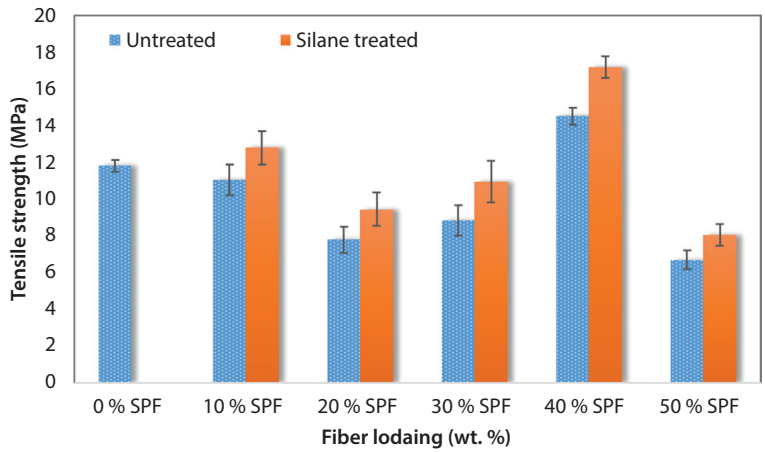

Figure 2 Tensile strength of untreated and silane-treated SPF/TPU composites.

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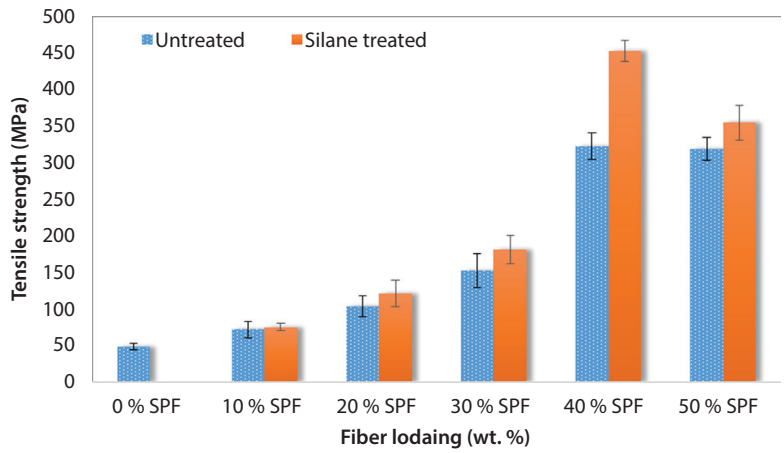

Figure 3 Tensile modulus of untreated and silane-treated SPF/TPU composites.

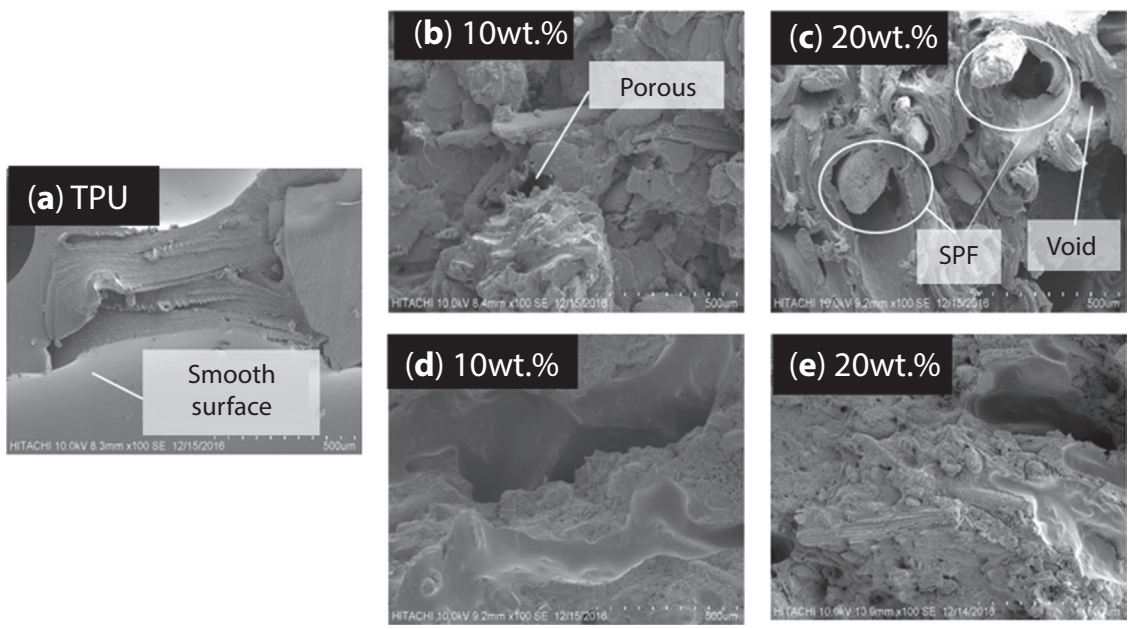

Figure 4 SEM of tensile testing of untreated (a) 0/100 USP/TPU, (b) 10/90 USP/TPU, (c) 10/90 USP/TPU and silane-treated (d) 10/90 TSP/TPU, (e) 20/80 TSP/TPU composites.

and Figure 5f–h) reveals some gap around the fibers, which could easily lead to fibers being pulled out, thus indicating weak interfacial bonding between the fibers and the polymer matrices. In contrast, in Figure 4d,e and Figure 5i–k, an improved TPU-SPF adhesion upon the silane treatment can be noticed, which contributes to higher interface bonding between the fibers and polymers. Besides, in the case of untreated SPF fiber composites, the fibers detached easily from the TPU matrix and were damaged, but no scattered fibrils were detected, which means that the adhesion between the matrix-fiber phases is not strong enough.

Furthermore, during the addition of SPF into the TPU matrix, air or other volatile substances may be trapped inside the composites, determining the formation of voids in the TPU matrix surface, as can be seen in Figure 4c.

The SEM images in Figure 5i–k reveal a smooth surface of the SPF due to the appropriate surface modification employed. Besides, sufficient adhesion between the fiber and the matrix, as well as a homogeneous matrix and few voids, have been observed for the composites with incorporated silane-treated fiber, compared to those with untreated fiber. Moreover, it was

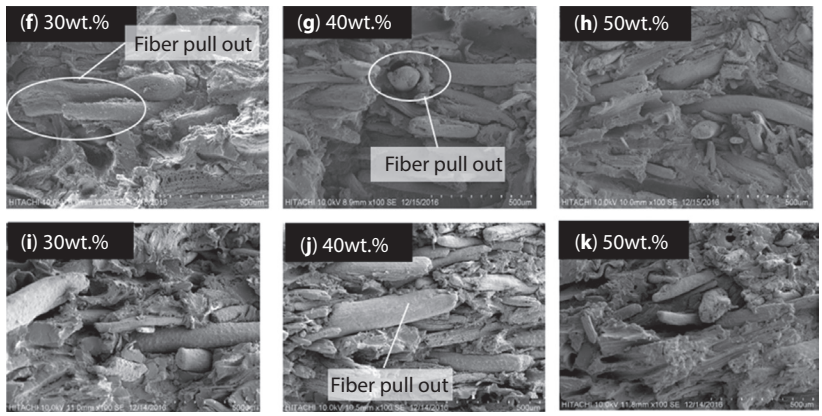

Figure 5 SEM of tensile testing of untreated (f) 30/70 USP/TPU, (g) 40/60 USP/TPU, (h) 50/50 USP/TPU and silane-treated (i) 30/70 TSP/TPU, (j) 40/60 TSP/TPU, (k) 50/50 TSP/TPU composites.

found that the addition of 40 wt% of SPF treated with 2% silane determined an increase in the tensile properties of the resulting composites (Figures 2 and 3), proving that the SPF fiber attached well to the TPU matrix, as evidenced in Figure 5j. After the silane treatment, the sugar palm fibers exhibited a cleaner and rougher surface. This enabled both mechanical interlocking and fiber/matrix interaction due to the exposure of the silanol groups during compounding [12]. In addition, morphological studies revealed the presence of spherical lumps on the surface of 10 and 20 wt% untreated SPF/TPU composites at regular intervals [26]. It has also been observed that the presence of voids on the surface of sugar palm fiber from the removal of these globular protrusions may cause improvement in the mechanical interlocking of the fiber/matrix bonding when treated with 2% silane. Conversely, in the case of untreated sugar palm fiber, the cell wall of the fibers is thicker and they cannot attach well enough to the TPU matrix, which leads to lower mechanical properties of the composites [27].

4.1.3 Flexural Properties

Figure 6 illustrates that the composites reinforced with 40 wt% untreated and silane-treated sugar palm fiber exhibited higher flexural strength compared to the other formulations of SPF/TPU composites. Results show that the flexural strength of the 40 wt% SPF composite was slightly increased by around 11% from 12.47 MPa (before the treatment) when composed with the untreated fiber to ~13.96 MPa (after 3 h) when composed with the silane-treated fiber. Generally, as the fiber loading was increased, for both untreated and treated SPF, the flexural strength of the composites was also boosted until the threshold of 40 wt% SPF, and beyond this optimum fiber loading, the value of flexural strength dropped to 7.59 MPa. This situation is in agreement with the findings of another study on kenaf fiber-reinforced TPU composites [10]. The highest flexural strength was measured for the 40 wt% silane-treated SPF/TPU composites with 13.96 MPa. This enables both mechanical interconnecting and the attachment reaction, due to the exposure of the hydroxyl groups, during melt compounding of SPF/TPU [28].

It can also be observed that the flexural strength of the 100 wt% TPU is almost similar (3.92 MPa) to those of 10 and 20 wt% untreated SPF/TPU composites (3.90 MPa and 3.65 MPa). Flexural modulus results (Figure 7) showed an increasing trend as the loading of the SPF was increased for all the composites except composite with 50% SPF/TPU. Moreover, the same trend was observed for the stiffness of the SPF/TPU composites, compared to that of untreated SPF. These results are related to the tensile behavior of the composites depicted in Figures 1 and 2.

The effects of the silane treatment can be assessed by the evolution of the flexural modulus of the composites. Thus, it should be noted that the fiber treatment with silane achieved increased surface roughness, removed surface impurities and reduced the diameter of sugar palm fiber [27], which consequently improved the flexural properties of the composites, as depicted in Figures 6 and 7. The results also show that the composites with silane-treated SPF display higher flexural modulus than those with untreated SPF. The flexural modulus of the 10, 20, 30, 40 and 50 wt% silane-treated SPF/TPU increased by 8%, 25%, 65%, 23% and 96%, respectively, compared with that of untreated SPF/TPU. This demonstrates that the introduction of silanols enhanced the interaction between the SPF and the TPU matrix. This was possible through the reaction of the silane coupling agent attached to the SPF with

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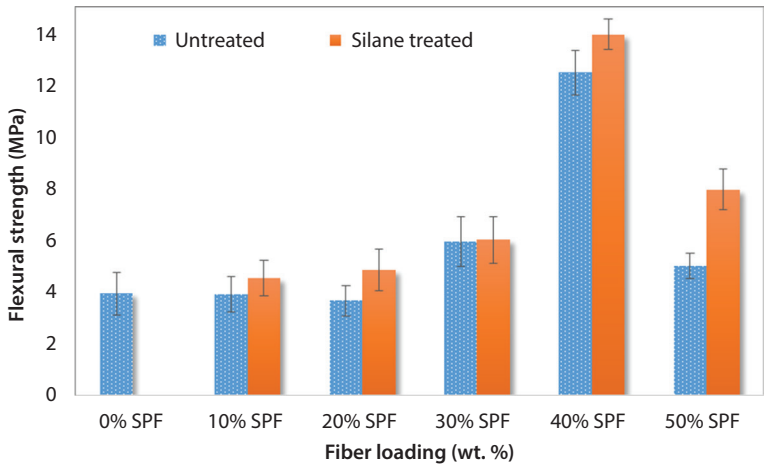

Figure 6 Flexural strength of untreated and silane-treated SPF/TPU composites.

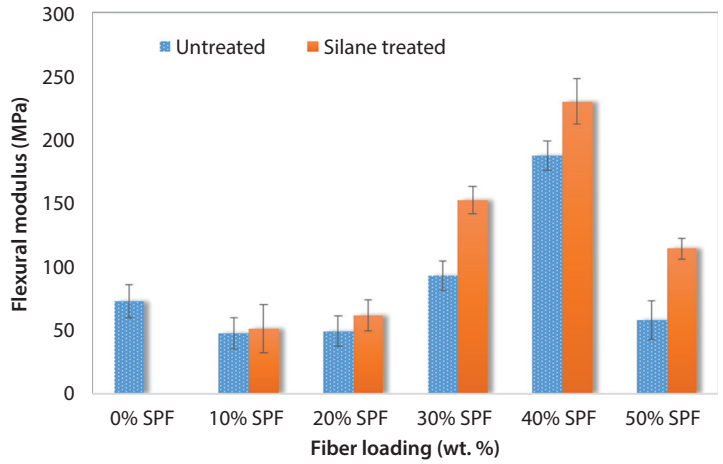

Figure 7 Flexural modulus of untreated and silane-treated SPF/TPU composites.

the OH from the thermoplastic polyurethane, indicating that, besides the linkages formed between the OH from SPF, additional bonds were formed in the silane-treated composites [26]. The results agree with the findings reported for silane-treated ijuk fiber-reinforced polypropylene composites [14].

4.1.4 Impact Properties

Figure 8 illustrates that the impact strength of the SPF-reinforced TPU composites increased when the fiber loading was increased from 0% to 50 wt%. It has been noticed that neat TPU (0/100 USP/TPU) has a lower impact strength value, i.e., 7.4 kJ/m^2. When introducing different loadings of sugar palm fiber into the TPU, the impact strength of the resulting composites was boosted. The highest impact strength (15.47 kJ/m^2) was observed for the silane-treated SPF/TPU composite at 40 wt% fiber loading, whereas the corresponding untreated SPF/TPU exhibited a value of 14.03 kJ/m^2 at the optimum observation energy, which is attributed to the good bonding between the SPF and the TPU matrix. The Izod impact test also demonstrated that improved bonding resulted in higher resistance to crack propagation [26]. A lower impact strength value was observed under similar conditions, when the polyester was combined with short natural fiber [29].

4.2 Thermal Properties

4.2.1 Storage Modulus (MPa)

Figure 9 presents the variation of storage modulus (E′) of untreated and silane-treated SPF/TPU composites as a function of temperature. A similar trend of the DMA curves was found for both untreated and treated SPF composites. It demonstrates that the storage modulus of both untreated and treated SPF composites is higher than that of the TPU matrix, which is due to the effect of the reinforcement of the polymer matrices by

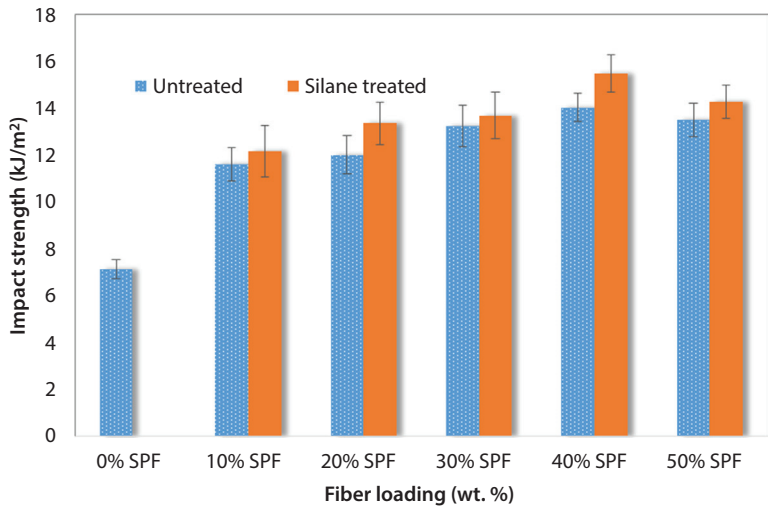

Figure 8 Impact strength of untreated and silane-treated SPF/TPU composites.

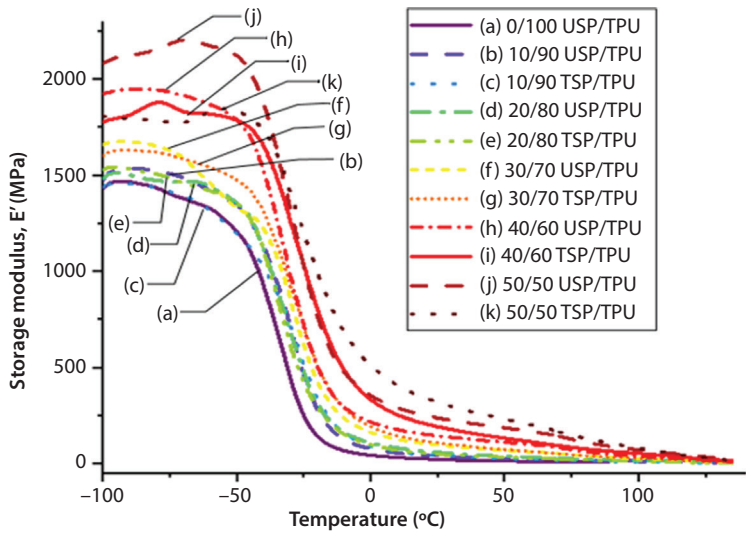

Figure 9 Storage modulus of untreated and silane-treated SPF/TPU composite in different proportions.

the sugar palm fiber [30, 31]. As the temperature was increased from –100 °C to 140 °C, the storage modulus of the 100% TPU continuously decreased. However, the storage modulus curves showed an increasing trend with an increment in fiber loading in the following order: 0/100USP/TPU < 10/90USP/TPU < 20/80USP/TPU < 30/70USP/TPU < 40/60USP/TPU < 50/50USP/TPU.

The effect of the 2% silane treatment on the E′ curves of the composites can be observed by comparing the sample series USP/TPU with TSP/TPU. Thus, the storage modulus of treated SPF samples 10/90 TSP/TPU, 20/80 TSP/TPU, 30/70 TSP/TPU, 40/60 TSP/TPU and 50/50 TSP/TPU increased by 1%, 5%, 11%, 33% and 34%, respectively, compared to their untreated counterparts 10/90 USP/TPU, 20/80 USP/TPU, 30/70 USP/TPU, 40/60 USP/TPU and 50/50 USP/TPU at 25 °C. In the case of the 50/50 TSP/TPU composite, a significant increase in storage modulus was attained with the silane treatment as compared to the 100% TPU. It is clear that this situation is due to the enhanced bonding between the silane-treated sugar palm fibers and the TPU matrix. It has been observed that the 100% TPU is an amorphous polymer, representing a glassy-to-rubber transition phase. The result for the storage modulus of the 100% TPU obtained in this study agrees with the findings of another study focusing on the storage modulus of kenaf-reinforced TPU [10].

As may be noted in Figure 9, the TSP/TPU composites presented higher E′, as compared to the USP/TSP composites. This is attributed to the improved bonding between the sugar palm fiber and the TPU matrix after fiber modification by the silane treatment. Thus, the removal of lignin from the fiber is one of the methods to achieve high modulus composites [26]. From the DMA results, it was found that the stiffness and damping properties of the composites decreased, whereas the storage modulus increased with an increment in the SPF content. Specifically, the storage modulus of the SPF/TPU composites increased remarkably with the addition of up to 50 wt% SPF from the minimum of the 0/100 USP/TPU to the maximum of the 50/50 TSP/TPU. Otherwise said, the high storage modulus of the 50/50 TSP/TPU (319.21 MPa) is the optimum value of storage modulus among the SPF/TPU composites. Additionally, it has also been noted that an increase in the SPF content of SPF/TPU composites indicates an increase in their stiffness and thermal stability, as compared to 0/100 USP/TPU. Thus, it can be concluded that, in comparison with the untreated sugar palm fiber, the silane-treated one possesses higher thermal stability. These findings are similar with those of Goriparthi *et al.* [32], who discovered a similar trend of storage modulus for silane-treated jute fiber PLA composites.

4.2.2 *Loss Modulus (MPa)*

In the present study, the relaxation of 100% TPU, as well as that of untreated and 2% silane-treated SPF/TPU composites, has been investigated from the loss modulus curves depicted in Figure 10. In Table 2, the glass transition temperature (T_g) is derived from the loss modulus curves of the composites. Generally, the T_g is understood as the maximum of the loss modulus curves acquired during dynamic mechanical analysis. As shown in Table 2, the T_g of the silane-treated SPF-reinforced composites moved to a higher temperature due to the fiber modification that SPF underwent before compounding. The shift of T_g towards higher temperatures is correlated with the reduced movement of the TPU matrix chains, which indicates enhanced interfacial bonding between the fibers and the matrix due to the coupling effect of the silane. It has been shown that the T_g for the 0/100 USP/TPU (100% TPU) is around –34.14 °C, increasing to –30.13 °C for the 50/50 USP/TPU and to –24.88 °C for the 50/50 TSP/TPU, which is based on the reinforced effect promoted by the silane-treated fibers physical interactions with the TPU that results in the decrease of the mobility chains. Similar results have been reported in previous works investigating silane-treated banana fiber composites, namely, a higher loss modulus was obtained for silane-treated banana fiber/PLA composites, compared with those including untreated banana fiber [33].

4.2.3 *Damping Factor (Tan Delta)*

Figure 11 presents the evolution of the damping factor (Tan delta) for the 100% TPU, as well as for the untreated and treated SPF/TPU composites, as a function of temperature. The measurement of energy

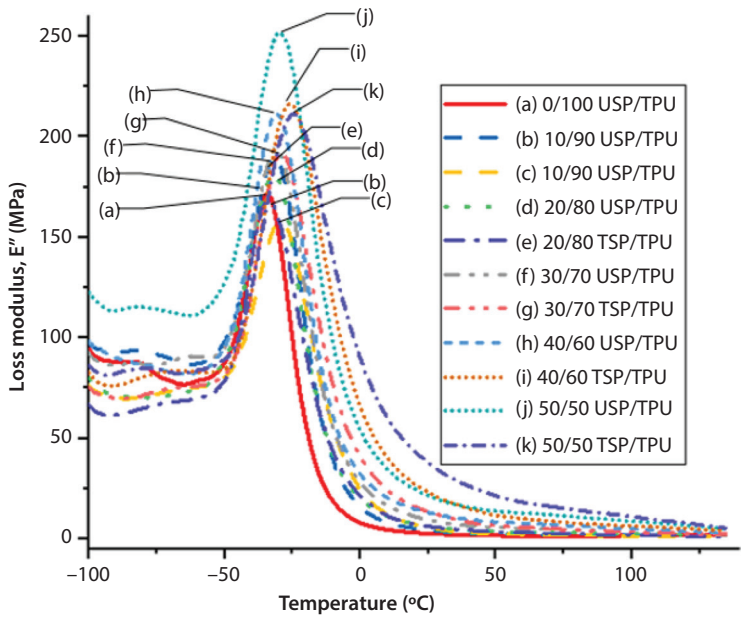

Figure 10 Loss modulus of untreated and silane-treated SPF/TPU composite in different proportions.

dissipation is the ratio of E′ and E″, which gives the tangent of the phase angle delta.

An increment in Tan δ is observed in both bending and shear modes with the reduction of sugar palm content in the TPU composites. The higher the fiber content (50 wt%), the lower the value of the damping peak (Tan δ), as may be noted comparing values for the 0/100 USP/TPU (100% TPU) and the 50/50 TSP/TPU (0.40550→0.20890). Damping factor in the transition region determines the deficiencies in the elasticity of a polymer, with the possibility of additional losses occurring at the matrix-fiber interface [34, 35]. As shown in Figure 11, the increasing fiber content from 10 to 50 wt% determined lower damping values with regard to the neat TPU matrix (0/100 USP/TPU). Therefore, it is recommended that, for incorporating sugar palm fiber into TPU, initially the molecular motion of TPU should be ensured, which would lead to good adhesion between the fiber and the matrix. Furthermore, the removal of lignin from silane-treated SPF involves modifications in the extent of hydrogen bonding, which affects the tan delta of the composites. In addition, when the fiber is treated with silane, the presence of unreactive organic groups from organosiloxanes will reduce the crosslinking of the interfacial network that caused increased damping properties of the composites compared to the untreated SPF composites. Thus, it can be deduced that lower Tan δ suggests good bonding due to improved adhesion between the fiber and the matrix, as compared to untreated composites. Hong *et al.* [34] reported similar observations regarding surface treated jute-reinforced polypropylene composites.

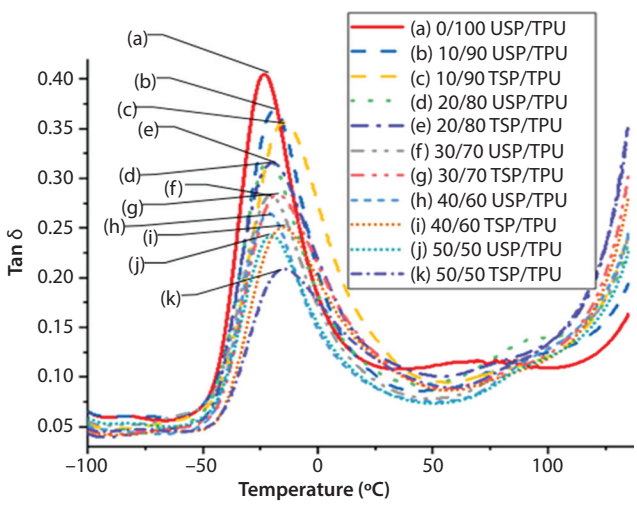

Figure 11 Tan delta of untreated and silane-treated SPF/TPU composite in different proportions.

4.3 Thermogravimetric (TGA) and Derivative Thermogravimetric (DTG) Analyses

4.3.1 *Effect of Fiber Loading on Thermal Properties of SPF/TPU Composites*

The TGA and derivative (DTG) curves for neat TPU (0/100 USP/TPU), untreated and treated SPF/TPU composites are presented in Figures 12, 13 and 14. Thermal decomposition of the TPU matrix takes place in three stages between 327 °C and 490 °C, when the scission of the polyol-isocyanate bond formed during polymerization occurs, then the isocyanate vaporizes and recondenses as a smoke, while polyol remains to further decompose later [36]. Almost similar results have also been reported by other researchers [10, 37]. The peak showing the temperature of the maximum decomposition rate appears at around 435 °C for TPU. El-Shekeil *et al.* investigated the degradation of kenaf-reinforced TPU composites. The onset temperature of thermal decomposition was found to have similar values for all the TPU composites, at around 363 °C [10].

The degradation temperature for natural fiber-based composites regularly occurs between the decomposition temperature of the reinforcement and that of the matrix [1]. The same pattern is also depicted by the untreated sugar palm reinforced thermoplastic polyurethane. According to Yakubu *et al.* [38], the onset degradation is measured at the 5% weight loss of composites. As seen in Figure 12, the initial decomposition temperature is around 270–350 °C and the final decomposition at 800 °C, which allows concluding that

Table 2 The thermo-mechanical properties of the TPU and its composites.

Designation	Peak height of Tan δ curve	Tg from Tan δmass (°C)	Tg from E″max (°C)
0/100 USP/TPU	0.4055	–23.59 °C	–34.14 °C
10/90 USP/TPU	0.3688	–20.00 °C	–32.39 °C
10/90 TSP/TPU	0.3568	–14.52 °C	–29.11 °C
20/80 USP/TPU	0.3185	–21.78 °C	–31.99 °C
20/80 TSP/TPU	0.3161	–19.15 °C	–32.88 °C
30/70 USP/TPU	0.2835	–21.16 °C	–31.55 °C
30/70 TSP/TPU	0.2840	–17.97 °C	–29.06 °C
40/60 USP/TPU	0.2653	–22.04 °C	–30.89 °C
40/60 TSP/TPU	0.2509	–15.81 °C	–26.29 °C
50/50 USP/TPU	0.2429	–20.80 °C	–30.13 °C
50/50 TSP/TPU	0.2089	–15.12 °C	–24.88 °C

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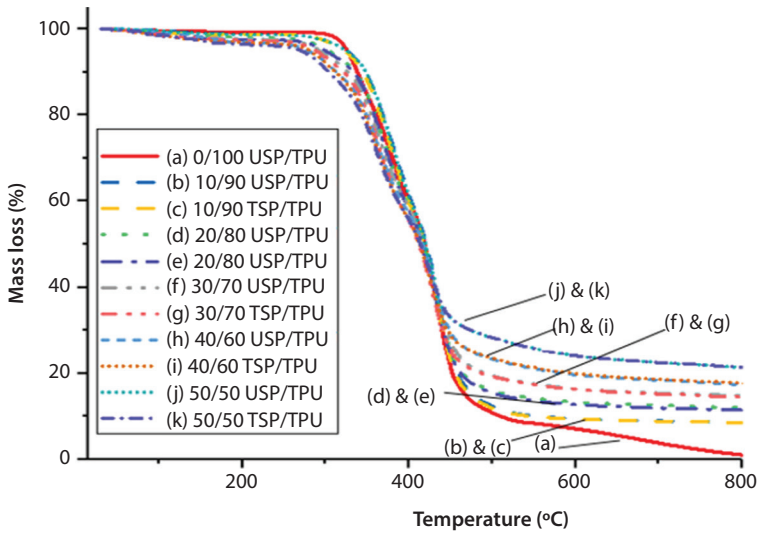

Figure 12 Analysis of TGA curves showing the effect of increasing fiber loading (0, 10%, 20%, 30%, 40% and 50% of SPF) in untreated and silane-treated SPF/TPU composites.

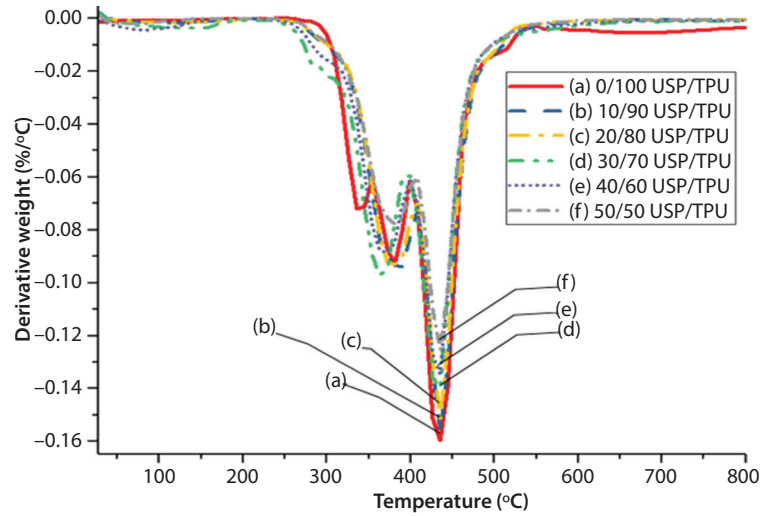

Figure 13 DTG curves of neat TPU and untreated SPF/TPU composites with fiber content of 0, 10, 20, 30, 40 and 50 wt% of SPF.

the existence of the cellulose component in the sugar palm fiber influences the decomposition process. All the formulations of untreated SPF/TPU composites degrade in three stages. In the first degradation stage of the 10/90 USP/TPU, moisture evaporation starts at 255 °C and ends at 309 °C, with a peak at the temperature of 291 °C. The second step reached the maximum temperature of 390 °C, whereas the third occurred between 426 °C and 498 °C, with a maximum peak at 436 °C. Overall, these peaks refer to a weight loss of about 91.44%. For the 20/80 USP/TPU, the first degradation stage took place between 274 °C and 399 °C, with a maximum at 390 °C; the second, in the range of 417–480 °C, with a maximum temperature of 435 °C; and the third between 498 °C and 552 °C, with a maximum at 437 °C. The degradation for the 20 wt% fiber loading represented a rapid and complex process, entailing a weight loss of about 87.98%.

A similar pattern was observed for the 40/60 and 50/50 USP/TPU, as shown in Figure 13. The 40/60 USP/TPU degraded quickly with three steps of decomposition, in the range 255–516 °C, with maximum at 291 °C, 363 °C and 438 °C, and a weight loss of about 82.68%. Meanwhile, for the 50/50 USP/TPU, the maximum temperatures were 291 °C, 381 °C and 435 °C, and the weight loss was about 78.63%, the

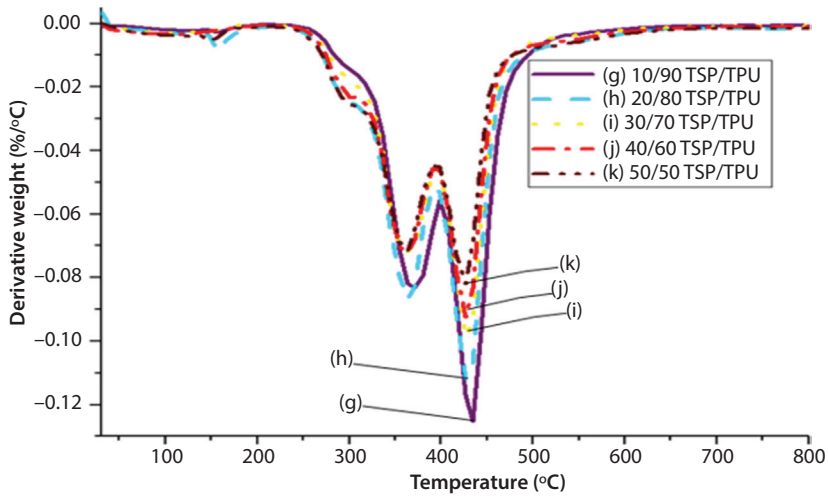

Figure 14 DTG curves of silane-treated SPF/TPU composites with fiber content of 10, 20, 30, 40 and 50 wt% of SPF.

values reached being higher than those of the peaks corresponding to the neat TPU (0/100 USP/TPU) matrix. Thus, it can be concluded that the thermal stability of USP/TPU composites is better than that of the neat TPU.

As may be noted in Figure 12, higher fiber loading contributes to higher thermal stability for both untreated and treated SPF/TPU composites, with weight losses of 21.37% and 21.19%, respectively.

4.3.2 *Effect of Silane Treatment on Thermal Properties of SPF/TPU Composites*

Figures 12 and 14 show TGA and derivative (DTG) curves of untreated and treated SPF/TPU composites. It can be observed that after fiber surface modification by the silane treatment the thermal stability of the TSP/TPU composites improves slightly. The thermal decomposition of the untreated SPF shows similar stages with those of treated SPF/TPU composites, indicating the decomposition temperatures of the different components. The SPF decomposed in four phases in the temperature range of 45–400 °C as follows: (i) moisture evaporation ranging between 45–123 °C, (ii) decomposition of hemicelluloses at 220–315 °C, (iii) decomposition of cellulose at 300–370 °C, and (iv) decomposition of lignin at 160–900 °C [39]. The neat TPU (0/100 USP/TPU), as well as the untreated and treated SPF/TPU composites, demonstrated the first stage of decomposition above the temperature of 300 °C. The data acquired from the TGA curves for untreated and treated SPF composites are presented in Table 3. Untreated SPF/TPU composites undergo early thermal decomposition at 300 °C, which is due to the thermal decomposition of the hemicelluloses in sugar palm fiber generally ranging between 250–300 °C. The addition of SPF improved the thermal stability of TPU, as may be concluded from the T_{dmax} values, which were higher than that of neat TPU (0/100 USP/TPU). Besides, the total weight loss at 800 °C exhibited a decrease with a rise in the sugar palm fiber loading.

This behavior demonstrates that the presence of SPF improves the thermal stability of the TPU matrix. The improvement of thermal stability leads to char formation during the pyrolysis of SPF, whereas the char acts as a preventive wall that defeats the thermal degradation of the TPU matrix [36]. The T_{dmax} values of the silane-treated SPF/TPU composites increased and their weight losses decreased at temperatures between 300 °C and 800 °C with regard to the untreated composites.

This can be clarified by the fact that the silane treatment improves the fiber-matrix interfacial bonding and leads to better dispersion of sugar palm fiber in the TPU matrix, which causes the establishment of a more homogenous char layer that gives better thermal protection [26]. Therefore, the thermal stability of the silane-treated SPF/TPU composites is slightly higher than that of the untreated composites.

Moreover, the results revealed that the thermal stability of both untreated and silane-treated SPF/TPU composites was higher than that of neat TPU (0/100 USP/TPU). The thermal degradation of TPU took place through random chain scission and a radical chain mechanism. The thermal decomposition of neat TPU (0/100 USP/TPU) undergoes several steps, where the initial weight loss happens at around 320 °C and finishes at 460 °C. Oppositely, this degradation in composites initiates at a relatively lower temperature. However, the major source of thermal degradation

Table 3 Weight loss percentage of designation of SP/TPU composites.

Designation	Tdmax(°C)	Weight loss (%)	
		300 °C	800 °C
0/100 USP/TPU	435	98.83	0.95
10/90 USP/TPU	436	97.29	8.56
10/90 TSP/TPU	437	97.23	8.49
20/80 USP/TPU	437	95.27	12.02
20/80 TSP/TPU	439	94.87	11.45
30/70 USP/TPU	438	94.07	14.77
30/70 TSP/TPU	440	93.35	14.42
40/60 USP/TPU	438	92.14	17.32
40/60 TSP/TPU	440	92.07	17.23
50/50 USP/TPU	439	97.24	21.37
50/50 TSP/TPU	444	90.76	21.19

in SPF was the degradation of the chains of the TPU polymer, which included C=C (double bond chains) [40]. The maximum mass loss temperatures of the thermoplastic polyurethane filled with different loadings of SPF are depicted in Table 3. The weight loss of neat TPU (0/100 USP/TPU) took place very slowly at temperatures below 400 °C, while as the temperature exceeded 400 °C, this process occurred very rapidly. These results demonstrate that the thermal stability of the composites increased as the content of SPF was increased. This is a logical consequence of the high thermal stability of the sugar palm fiber, compared with that of the TPU matrix [10]. The thermal decomposition of the composites was retarded above 500 °C because of the increase in the ash content. The treated fiber composites demonstrated an higher decomposition temperature compared to untreated fiber composites with similar fiber contents, which is in agreement with other previously reported findings [41]. Moreover, it can be seen that the weight loss of the silane-treated composites has higher values than neat TPU and untreated composites.

5 CONCLUSION

The present study has demonstrated that the fiber loading and silane treatment are influential factors, leading to different mechanical properties of the developed SPF/TPU composites. It can be concluded that the tensile, flexural and impact properties of the SPF/TPU composites improve with increasing sugar palm fiber content. A fiber loading of 40 wt% has been found as the optimum for achieving good tensile and flexural properties. In addition, the silane treatment has been shown to contribute to an enhancement in mechanical properties, including tensile, flexural and impact properties of the SPF-reinforced thermoplastic polyurethane composites. This is explained by the fact that the silane treatment of SPF improves the fiber/matrix interfacial bonding in SPF/TPU composites. SEM analyses have verified that the silane-treated fiber attached well to the thermoplastic polyurethane at the interface. Based on the results, it can be concluded that 40 wt% silane-treated fiber loading is the optimum reinforcement in SPF/TPU composites, yielding the best tensile, flexural and impact strength, compared to untreated fiber composites. The values of storage modulus and loss modulus were found to be the maximum for the formulation containing 50 wt% SPF. The highest Tan δ value was exhibited by the 50 wt% silane-treated SPF/TPU composite and lowest value by the neat TPU (0/100 USP/TPU). The value of T_g obtained from the loss modulus curve is lower than that derived from the Tan δ curve. The highest T_g value was observed for the 50/50 TSP/TPU composite among all other composites. Moreover, the addition of sugar palm to the TPU matrix reduced the damping factor, due to a good fiber-matrix interface. The silane treatment slightly affected the TGA results. Bearing in mind the advantages that sugar palm fiber presents, it should be considered as an alternative natural fiber for reinforcing thermoplastics.

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