



Original Article

Physical and thermal properties of treated sugar palm/glass fibre reinforced thermoplastic polyurethane hybrid composites



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ABSTRACT

Lignocellulosic/natural fibre and glass fibre-based hybrid composites are considered as high-performance composites but very limited numbers of researchers worked on substituting sugar palm fibre with glass fibre in hybrid composites. The main goal of this research to investigate the effect of various treatment such as 6% alkaline (TNSP), 2% silane (TSSP) and combined 6% alkaline–2% silane (TNSSP) on physical and thermal properties of sugar palm/glass/thermoplastic polyurethane hybrid composites were carried out. Taking into account of physical properties, the combined alkaline–silane treated hybrid composites (TNSSP) showed the lowest density, thickness swelling, and water absorption as compared to other composites. The good thermal stability was discovered for treated as compared to untreated sugar palm fibre based composites (UTSP). Overall, the treated sugar palm/glass/thermoplastic polyurethane hybrid composites suitable for the fabrication of automotive components.

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1. Introduction

All across the countries, natural fibres have become well-known as an important material in many industries application. There are several types of established natural

fibres such as sugar palm, kenaf, coconut husk, jute, sisal, curaua, flax and so on. Talking about sugar palm, its trees are widely grown and its fibres are largely abandoned in many Asian countries mainly in Malaysia. This opportunity has gathered many researchers' attention to study further these sugar palm fibres as a potential material to be used as reinforcement in polymer composites [1,2].

As the research growth, sugar palm fibre has shown to have many significant advantages to be considered. As in environment, sugar palm fibre is famous in low priced, biodegradable,

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plentiful in nature and so on. While in term of properties, sugar palm fibre has shown low density, good mechanical strength, and also good in thermal properties [3]. Apart from those mentioned above, sugar palm fibre also has some disadvantages such as non-adhesive with polymer matrices, high moisture, low modulus and low strength in properties. Therefore, certain methods like chemical treatment were employed in order to improve the attraction between both constituents in the composite [2,4].

After witnessing the disadvantages of the sugar palm fibre being improved by the chemical treatment to enhance fibre's properties, hybridization is next to be used in this research study. In this study, glass fibre reinforced thermoplastic polyurethane is hybridizing in the same matrix with sugar palm fibre in order to improve its properties. This current method of reinforcement in hybrid form is used in composites for the purpose of achieving superior physical properties as well as dimensional stability. There are several factors that influenced the performance of the hybrid composites such as the matrix itself, the interfacial bonding (between fibre and matrix), composite's shape and length, and the fraction volume of both fibres [5]. There are some works reported that done on fibre treatment and hybridize with glass fibre to enhance the properties of the hybrid composites such as kenaf [6,7], flax [8], basalt [8], sisal [9], Palmyra palm leaf [10], coir [11], etc.

Furthermore, the previous works that investigated with combination treatment of alkaline and silane as shown in Table 1 such as kenaf [12,13], sisal [14], bamboo [15], hemp [16] also showed the good properties of composites. In this research, fibre modification with NaOH or mercerization process can defibrillate the surface of the sugar palm fibre, so that TPU can hold further into the fibre to enhance both physical adhesion. Other than that, sugar palm fibre is treated with silane mixture in order to change the structural and chemical composition in the fibre and eventually can improve the fibre's properties. Lastly, alkaline and silane mixture are mixed together as for the fibres treatment. There are few or

no attempt on sugar palm fibre surface modification with hybridization of glass fibre. So, the main objective of this research was to investigate the effect of untreated and treated sugar palm fibre incorporated in glass fibre reinforced TPU composites on physical and thermal properties.

2. Experimental

2.1. Materials

The hybrid composites consisted sugar palm fibre, glass fibre and thermoplastic polyurethane. Sugar palm fibres were collected from local plantation at Jempol, Negeri Sembilan. Glass fibres and TPU were supplied in the form of mill seized of 6 mm and pellet form by Innovative Pultrusion Sdn. Bhd.

2.2. Treatment of sugar palm fibre

Sugar palm fibre was initially immersed in huge tap water for several days then rinsed using water and dried in the ambient temperature for 7 days. The SPFs were crushed into small size of 5–10 mm using plastic crusher machine this due to the physical attributes of the SPFs is very hard to cut using the usual machine cutter. Subsequently then were transferred to pulverize machine followed by siever machine in order to get the size of 150–250 μm . Then the SPFs were undergoing the next process of fibre modification as follows:

Alkaline treatment: The SPFs were treated for 3 h using alkali (NaOH) solutions with a concentration of 6%. The alkaline solutions were used to remove the surface impurities and hemicelluloses within the fibre. Next, the SPFs were repeatedly cleaned using distilled water then dried at 25 °C for 72 h and oven dried at 60 °C for 24 h [6].

Silane treatment: The 2 wt.% APS (weight percentage compared to the fibre) was dissolved for hydrolysis in a mixture of water-ethanol were employed on sugar palm fibre for 3 h then washed and kept in air for 72 h. Lastly, the sugar palm

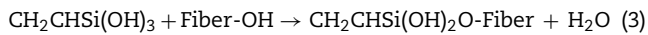
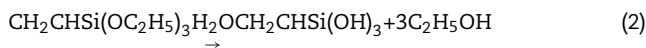
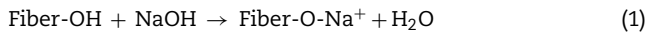
Table 1 – Previous worked working on combined alkaline-silane for natural fiber treatment.

Matrix	Reinforcement	Fibre treatment	References
Polypropylene	Kenaf fibre	Alkaline-silane	[12]
Polylactic acid	Kenaf	Alkaline-silane	[13]
Polylactic acid	Wheat husk	Alkaline-silane	[17]
Polylactic acid	Sisal	Alkaline-silane	[14]
Polylactic acid	Hemp	Alkaline-silane	[16]
Polylactic acid	Empty fruit bunch	Alkaline-silane	[18]
Polylactic acid	Ramie	Alkaline-silane	[19]
Epoxy	Bamboo	Alkaline-silane	[15]
Phenolics	Kenaf and pineapple leaf	Alkaline-silane	[20]
Polylactic acid	Phormium tenax	Alkaline-silane	[21]
Polyvinyl alcohol	Oil palm empty fruit bunches fibre	Alkaline-silane	[22]
–	Hemp	Alkaline-silane	[23]
Epoxy	Bamboo	Alkaline-silane	[24]
Thermoplastic polyurethane	Flax	Alkaline-silane	[25]
Polylactic acid	Jute fibre	Alkaline-silane	[26]
Polylactic Acid	Corn fibre	Alkaline-silane	[27]
Polypropylene	Kenaf	Alkaline-silane	[28]
Nanoclay	Banana	Alkaline-silane	[29]
–	Phormium tenax	Alkaline-silane	[30]

Table 2 – Formulation of untreated and treated SP/G/TPU hybrid composites.

Type of composites	TPU (wt. %)	SP (wt. %)	G (wt. %)	Fiber treatment
UTSPP	60	30	10	Untreated
TNSP	60	30	10	6% alkaline
TSSP	60	30	10	2% silane
TNSSP	60	30	10	6% alkaline–2% silane

fibre was oven dried at 60 °C for 24 h [31]. The chemical reaction of alkaline (1) [32] and silane treatment (2–3) [33] are given as follows.



Combined alkaline–silane treatment: First, sugar palm fibres were treated with alkaline treatment as described in the alkaline treatment step. Then, the sugar palm fibres were treated by silane treatment as described in the silane treatment step.

The untreated and treated sugar palm fibre/glass reinforced thermoplastic polyurethane was fabricated using melt-mixing compounding followed by hot pressing moulding. The weight fraction of the sugar palm fibre, glass fibre and TPU is shown in Table 2. The hybrid composites specimens including untreated, alkaline treated, silane treated, alkaline–silane treated sugar palm fibre. The fibres reinforcement with TPU was mixed together using an internal mixer with temperature 190 °C, 11 min, 40 rpm before the batch were hot pressed with the preheat 7 min and full pressed 10 min, temperature 190 °C and cooling press for 5 min with temperature 25 °C [34].

3. Characterization

3.1. Density

The developed untreated and treated hybrid composites were weighed in the air using digital weighing scale and in water using the density determination balance (XS205 Mettler Toledo). Ten samples were cut into a square shape with the dimension 10 × 10 × 3 mm. The test was performed according to ASTM D4018 standards at 27 °C. The density was then recorded for all samples and presented in Fig. 1.

3.2. Water absorption and thickness swelling

Ten replicates of untreated and treated SP/G reinforced TPU composites were oven dried at 60 °C for 24 h. The specimens with the dimension of 20 × 20 × 3 mm were then immersed in distilled water at ambient temperature. Then, each sample was removed from the bath and carefully dried with an

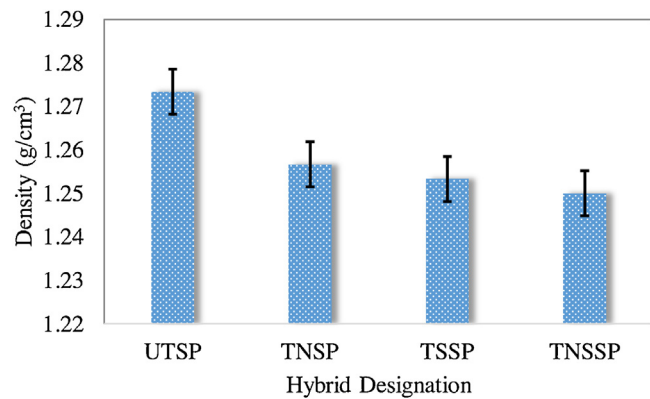


Fig. 1 – Density of untreated and treated hybrid SP/G reinforced TPU composites.

absorbent paper before weighing. The weight gain, W_g due to water absorption was calculated using Eq. (1):

$$W_g = \frac{W_t - W_0}{W_0} \times 100\% \quad (1)$$

W_t is the weight of the specimens after a certain time of immersion.

W_0 is the initial weight dried specimens.

Furthermore, the thickness swelling, T_g was calculated using Eq. (2):

$$T_g = \frac{T_t - T_0}{T_0} \times 100\% \quad (2)$$

3.3. Thermogravimetric analysis (TGA)

The thermal stability of the TPU and sugar palm fibre/glass reinforced hybrid composites was determined by thermogravimetric analysis (TGA), using a PerkinElmer Pyris 1. TGA analysis was performed under a nitrogen atmosphere at a flow rate of 20 ml/min to avoid oxidation. The untreated and treated SP/G samples weighed 10–15 mg. The heating rate was maintained 20 °C/min as the samples were heated to the temperature of –45 to 800 °C.

4. Results and discussion

4.1. Physical properties

4.1.1. Density

The density of untreated and treated SP/G hybrid composites is shown in Fig. 1. From Fig. 1, the density of combined alkaline–silane treated showed 1.25 g/cm³ followed by silane treated, alkaline treated and untreated. The order

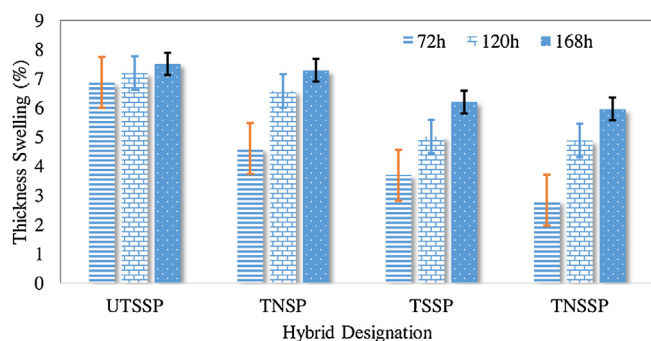


Fig. 2 – Thickness swelling of untreated and treated hybrid SP/G reinforced TPU composites.

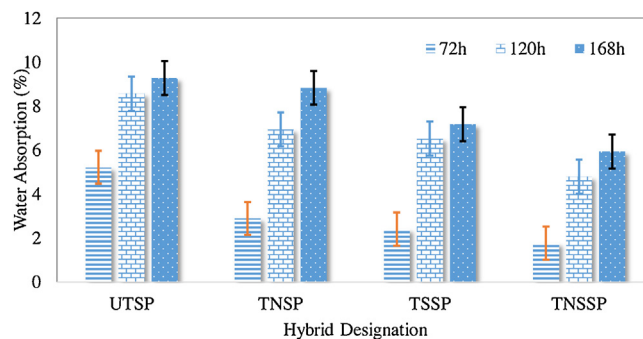


Fig. 3 – Water absorption of untreated and treated hybrid SP/G reinforced TPU composites.

of decreasing value of density SP/G hybrid composites are as follows: UTSP < TNTP < TSSP < TNSSP. Reduction in density of treated fibres was also reported in previous work [35]. In the study, they investigated a study on the effect of fibre treatment using alkaline and silane showed the density of treated fibre led to the lower density of composites as compared untreated [35].

4.1.2. Thickness swelling

The thickness swelling of untreated and treated hybrid SP/G composites was performed as in Fig. 2. From the thickness swelling curve, it is observed that the hybrid samples absorbed water content increased with increasing immersion time. The order of decreasing the value of thickness swelling SP/G hybrid composites are as follows: UTSP < TNTP < TSSP < TNSSP. The most significant improvement of 21% thickness swelling is combined alkaline–silane treatment as compared to other treated hybrid composites. This is due to cleaning by using alkaline treatment which removed the waxy like substances and the impurities on the fibre surface rougher [16,36]. This lead to enhance fibre–matrix interaction resulting from the fewer pores and void space that prevent adsorption of moisture from the sugar palm fibre. Similar findings have been informed in the literature, where it is reported that the thickness swelling of alkaline and silane treated of wood fibre reinforced recycle polymer composites was minimum [37].

4.1.3. Water absorption

Fig. 3 shows the combined alkaline–silane is beneficial for the moisture resistance of hybrid SP/G composites. The order of decreasing value of water uptake SP/G hybrid composites are as follows: UTSP < TNTP < TSSP < TNSSP. The moisture absorption decreases considerably by employing fibre treatments on sugar palm fibre. In the case of untreated hybrid composites (UTSP), higher water absorption, 9.27% may be due to poor wettability of untreated sugar palm fibre to TPU matrix. At the first stage of alkaline treatment, the impurities that adhere to and hemicelluloses part were removed from sugar palm fibre. Then, the silane treatment prevents the hydroxyl groups on the sugar palm fibre surface by inducing hydrophobic silanes groups [16,18]. The existence of the hydroxyl group origins from the moisture absorption properties. This combined treatment yielding the lowest water uptake value of as compared to TNTP and TSSP. Those fibre treatments lower

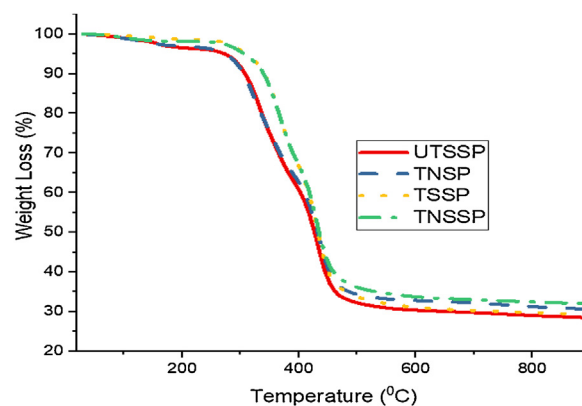


Fig. 4 – Thermogravimetric (TGA) curves for UTSP, TNTP, TSSP and TNSSP hybrid composites.

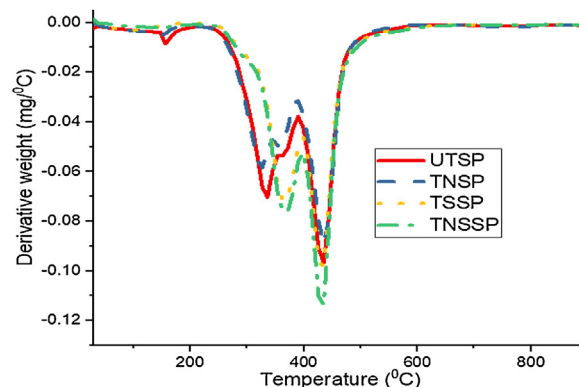


Fig. 5 – Derivative thermogravimetric (TGA) curves for UTSP, TNTP, TSSP and TNSSP hybrid composites.

the water absorption than those of untreated sugar palm fibre. Similarly, Abdullah and Ahmad [38] reported that water absorption of coir reinforced unsaturated polyester resin composites decreased with treated silane and alkaline treatment. The decrease of water absorption of natural fibre and its composites after alkaline and silane treatment was also reported by other researchers [15,17,37]. The concentration and time of treatment of fibres also influenced the water absorption of the hybrid composites.

Table 3 – Thermogravimetric (TGA) results obtained for UTSP, TNSP, TSSP and TNSSP hybrid composites.

Hybrid designation	Initial degradation temperature, IDT (°C)	Final degradation temperature, FDT (°C)	Final residue (%)
UTSP	156	435	28.48
TNSP	146	480	30.69
TSSP	132	481	29.35
TNSSP	138	507	32.02

4.2. Thermal properties

4.2.1. Thermogravimetric analysis (TGA)

Fig. 4 shows the results of the TGA obtained on the untreated and treated SP/G reinforced TPU composites. From Fig. 4, it can be seen that at a lower temperature, untreated hybrid composites showed the lowest weight loss followed by alkaline treated. This can be correlated to the higher moisture absorption of the hemicelluloses of the untreated sugar palm fibre as in Fig. 3. There are several works that reported that lignocellulosic fibres were not good bonding with polymers [30]. This is due to the existence of moisture in the multicellular parts or void spaces at fibre–matrix adhesion could be possibly weakening the bonding fibre–polymer [29]. The initial weight loss where the moisture starts for evaporate for UTSP, TNSP, TSSP and TNSSP are 156, 146, 132 and 138 °C, respectively. Bachtiar et al. [39] reported that initial mass loss took place around 56 °C and 165 °C for sugar palm fibre and alkaline treated sugar palm fibre reinforced HIPS composites.

The final degradation of UTSP, TNSP, TSSP and TNSSP of 534, 546, 678, and 440 °C indicated the weight loss reflects the quantity of residue. The UTSP has the lowest percentage of the final residue followed by TNSP, TSSP and TNSSP with the values of 13%, 15% and 16%. This indicates that combining alkaline and silane increases the percentage of final residue which can be suggested good thermal stability properties. Some researchers consider this is due to the removal of lignin after fibre treatment of natural fibre [13,30]. Though, based on TGA analysis that done by others, alkali treated sugar palm fibre were evaluated to leave about 5% more residue than untreated [39]. Nevertheless, it was found that when bamboo fibre pre-treated with alkaline, more weight loss as compared to single fibre treatment [40]. The TGA results of the neat, untreated and treated is summarized in Table 3.

The results of derivative thermogravimetric (DTG) analysis evaluated on the untreated and treated SP/G reinforced TPU composites are demonstrated in Fig. 5. The peaks of the DTG curves is owed to the degradation temperature of each component of the hybrid composites. It had been observed that, three peaks resulted on the untreated SP/G composites. The first peak was higher than other treated SP/G composites due to the presence of water molecules in the hemicelluloses part of SP fibre [36]. Moreover, this could be possibly due to the void that existed on the SP/G specimens during fabrication process contribute the greater peak of DTG curve. Sugar palm fibre treated with combined alkaline–silane (TNSSP) decomposed at lower temperature than TSSP, TNSP and untreated hybrid composites. In engineering application, the initial peak of decomposition is important than following peaks at higher temperature [41].

5. Conclusion

Sugar palm fibre was modified by alkaline, silane and combined alkaline–silane to enhance the adhesion bonding between sugar palm fibre with glass-reinforced TPU hybrid composites. The effects of various treatments on sugar palm fibre were characterized by physical and thermal properties. When tested at RT, the lowest density, thickness swelling and water absorption recorded for hybrid SP/G composites showed that TNSSP has the lowest density value followed by TSSP, TNSP and UTSP, respectively. The TGA analysis showed that the TNSSP had the lowest residue as the combined alkaline–silane treatment. The untreated, TNSP and TSSP had residue percentage of 29%, 30% and 32%. The hybrid composite with combined alkaline–silane degrades at a slightly lower temperature as compared with alkaline treated and silane treated. This combination of silane and alkaline treatment at last showed a significantly good result of lower density, thickness swelling, water uptake and good thermal stability as compared to other mentioned treatment and can be proposed as potential fibre treatment mainly for sugar palm fibre in fabrication of automotive component.

Conflicts of interest

The authors declare no conflicts of interest.

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