Mechanical Performance of Roselle/Sugar Palm Fiber Hybrid Reinforced Polyurethane Composites

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The effect of sugar palm fiber (SPF) loading was studied relative to the mechanical properties of roselle (RF)/SPF/thermoplastic polyurethane (TPU) hybrid composites. RF/SPF/TPU hybrid composites were fabricated at different weight ratios (100:0, 75:25, 50:50, 25:75, and 0:100) by melt mixing and hot compression. The mechanical (tensile, flexural, and impact test) and morphological properties of tensile fractured samples were examined using a universal testing machine, impact machine, and scanning electron microscope. It was found that the hybridization of RF/SPF increased its impact strength corresponding to the increases in the SPF content of the composites. The tensile and flexural properties of the hybrid composites decreased due to poor interfacial bonding between the fiber and matrix. Scanning electron micrographs of the tensile fractured surface of the RF/SPF hybrid composites revealed fiber pullouts and poor adhesion bonding. In conclusion, the hybridization of SPF with RF/TPU composites enhanced its impact strength while decreasing the tensile and flexural strength.

Keywords: Roselle fibers; Sugar palm fibers; Thermoplastic Polyurethane; Mechanical properties; Hybrid composites

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INTRODUCTION

The use of petroleum-based plastic in human activities is increasing rapidly with the increase human population. Moreover, the disposal of these materials is a major issue of environmental pollution (Jumaidin *et al.* 2016b, 2017). Increasing awareness in society has led to numerous efforts to reduce the impact of this material on the environment (Ilyas *et al*. 2017; Jumaidin *et al*. 2017b). Environmentally friendly materials, such as natural fibers derived from renewable resources, are a promising solution for this problem (Jumaidin *et al.* 2016; 2017a). The application of this naturally derived material improves the properties of polymer composites (Kasim *et al*. 2015). Natural fiber products are widely applied in various industries such as furniture, packaging, automotive, and construction (Alamri and Low 2013; Kumar *et al.* 2016; Edhirej *et al*. 2017; Radzi *et al*. 2017b). The demand from industries that use natural fiber (kenaf, hemp, banana, and oil palm) sources is increasing. This material source is more friendly to the environment, low cost, easy to recycle, and biodegradable, making it a good alternative to replace the use of commercial fibers (Razali *et al.* 2015).

Roselle and sugar palm fibers are two natural fibers that are abundantly available in Malaysia, along with oil palm and kenaf fiber. The roselle plant grows in South Asia (Nadlene *et al.* 2015, 2016a), and the fibers from this plant have been used in polymer composites (Chauhan and Singh 2012; Nadlene *et al.* 2015a,b, 2016). The roselle tree (*Hibiscus sabdariffa* L.) is easy to grow and is found in tropical areas (Razali *et al*. 2015; Nadlene *et al.* 2016a). Normally, these trees are used to produce medicine, food, ropes, jute, and textiles.

The sugar palm tree (*Arenga pinnata* Merr.), also known as Ijuk, belongs to the 'Palmae' family. It is the most widely used sugar palm for the harvest of sugar and fibers. The sugar palm tree can produce traditional sugar blocks more commonly known as Enau or Gula Kabung, which are often used as sweeteners in food and sugar palm fiber. Ijuk is also utilized in products for daily use, such as for brooms, brushes, and ropes, mats, and paint brushes (Huzaifah *et al.* 2017).

Hybridization is a combination of more than one fiber used to reinforce a matrix to increase its mechanical properties and reduce the cost of producing the hybrid fiber reinforced matrix composites (Pandya *et al.* 2011; Nunna *et al.* 2012; Saw *et al.* 2012). The hybridization of natural fibers in polymer composites is another promising approach in improving the physical properties of the material. Additionally, the hybridization of natural fibers with natural or synthetic fibers can improve the thermal properties, stiffness, ductility, damping, and strength of the matrix composite (Nunna *et al.* 2012; Sathishkumar *et al.* 2014; Saba *et al*. 2016; Aggarwal *et al.* 2017). The hybridization of sugar palm fiber with seaweed waste in a thermoplastic sugar palm starch/agar matrix increases the thermal, mechanical, and biodegradation characteristics of the material (Ridhwan *et al.* 2017c). This finding is attributed to the similar hydrophilic nature of the natural reinforcement, which resulted in good compatibility and adhesion between them.

Roselle fiber (RF) has the advantages of good tensile strength and durability in water. RF also has a high impact on the mechanical properties when it is used as fiber reinforcement in polymer composites (Nadlene *et al.* 2015a,b). Therefore, the hybridization of RF with natural fiber such as SPF aims to get the best combination hybrid fibers on TPU composites. The use of SPF for hybridization is due to its good fiber strength, resistance sea water and durability to developing unique hybrid composites (Saw *et al.* 2012; Kasim *et al*. 2015; Huzaifah *et al*. 2017). To date, there are no studies on RF/SPF hybrid reinforced thermoplastic polyurethane (TPU) composites. The aim of this paper is to study the properties of RF/SPS hybrid reinforced TPU composites with different percentages of RF /SPF hybrid composites (100:0, 75:25, 50:50, 25:75, and 0:100). The tensile strength, flexural strength, impact strength, and morphology of RF/SPF/TPU composites were examined.

EXPERIMENTAL

Materials

RF was obtained from a roselle plantation area in Mersing, Johor, and SPF was obtained from Jempul, Negeri Sembilan, Malaysia. To produce fibers from roselle plant, it was necessary to start with a water retting process that takes 14 days after soaking with water. Tap water was used to wash the retted stem, and RF was pulled out manually to separate the fiber and stem roselle. The SPF was washed with tap water to remove impurities and dried under sunlight. Both fibers were crushed and sieved using a sieve shaker machine. The size range used, 300 to 425 µm fiber length, was fixed throughout the study. The polymer material used as a matrix in this study was TPU with a density of 1.10

g/cm^3, tensile strength 9.18 Mpa, flexural strength 5.24 MPa, and impact strength 9.88 (Radzi *et al*. 2017b). TPU was supplied by Pultrusion Sdn. Bhd, Seremban, Malaysia.

Sample Preparation

A Brabender Plastograph internal mixer (Duisburg, Germany) was used for the pre-mixing process of the RF/SPF/TPU hybrid composites at 170 °C. A hot press machine was used to pre-heat and full press the sample into sheet form at 170 °C under the load 40 tons. The fiber was arranged randomly. Lastly, the sample in sheet form was cool-pressed. The weight percentage of the reinforcement is shown in Table 1. The TPU as a matrix was maintained at 60 wt.% according to previous work (Radzi *et al*. 2017a) with designation for single roselle fiber with 40 wt% (RF-T) and sugar palm fiber with 40wt% (SPF-T).

Table 1. Composition of the Materials

RF (%)	SPF (%)	Designation
75	25	RS-1
50	50	RS-2
25	75	RS-3

Tensile Testing

Tensile properties were determined using a universal testing machine (UTM; Instron model 5556, (Norwood, MA), according to the ASTM D-638 (2002). The temperature used was 23 ± 1 °C. Humidity was 50 ± 5%, and the maintained speed was 5 mm/min.

Flexural Testing

Flexural test was determined using the UTM according to ASTM D-790 (2003). There were five samples prepared for this test, and they were cut into 130 mm x 13 mm x 3 mm. The temperature was 23 ± 1 °C, the humidity was 50 ± 5% and speed was maintained at 2 mm/min.

Impact Test

The impact test was determined by using an Instron CEAST 9050 pendulum impact tester (Norwood, MA) according to ASTM D-256 (2001). 5 samples notched were cut into 60 mm x 13 mm x 3 mm.

Scanning Electron Microscopy (SEM)

Morphological studies were performed on the fractured surface of the samples strength using scanning electron microscopy (Hitachi, model S-4300N, Japan). The sample composites were gold coated to provide electrical conductivity in order to obtain good quality morphological results before their surface was examined

RESULTS AND DISCUSSION.

Tensile Properties

The tensile strength of the RF/SPF/TPU hybrid composite is shown in Fig. 1. The tensile strength and modulus of the RF/SPF/TPU hybrid composites decreased with increasing in SPF content. RS-1 showed the highest value in tensile strength compared to the other hybrid composites. The highest tensile strength value recorded was 13.4 MPa, and the lowest was 11.1 MPa. Furthermore, adding the SPF into the RF composites caused a decrease in their tensile strength and modulus. This may be due to poor adhesion, high void, fiber pullouts, incompatibility, or agglomeration, which may have led to poor load transfer between the fiber and the matrix (Rozman *et al.* 2001; Saw *et al.* 2012; Yahaya *et al*. 2014).

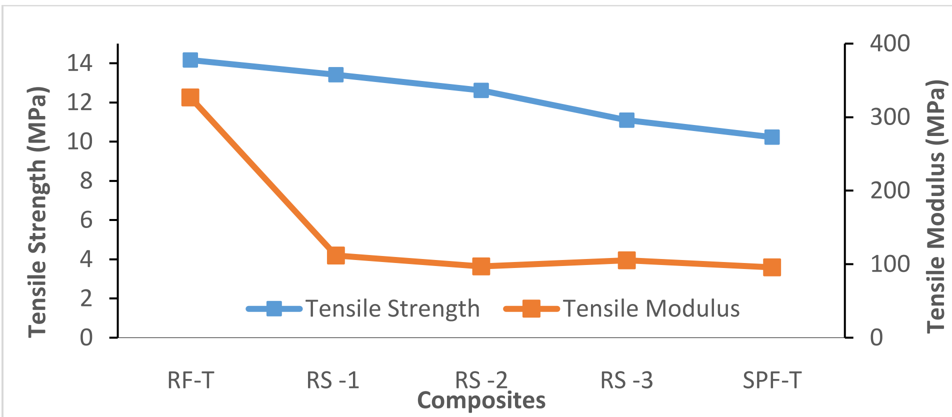

Fig. 1. Tensile strength and modulus of RF/SPF/TPU hybrid composites

Good interfacial bonding between fibers, compatibility, and matrix are an important factor for the transferring stress from the matrix to the fibers (Threepopnatkul *et al.* 2009; Kasim *et al*. 2015). Good adhesion can enhance a sample's mechanical properties. Also, the addition of SPF was one of the factors that weakened interfacial bonding of the hybrid composites. The SPF-T showed the lowest tensile strength compared to the RF-T and other hybrid composites. It is possible that when the SPF was added into the RF composites, it decreased the properties as shown in Fig. 1, as the SPF contents may have created more stress points and an increased incapability area on the polymer composite between the fibers and the matrix. Similar results have been reported by several authors (Rozman *et al.* 2001; Datta 2015; Essabir *et al.* 2016; Radzi *et al.* 2017b), whose studies have shown that the creation of high-stress points and an incapability area between reinforcements and matrix caused a weakening in tensile strength.

Figure 2 shows, at 50x magnification, the fractured surface with different RF/SPF/TPU hybrid percentages of composites. The image of RF and SPF can be observed in the SEM of the fiber in Figs. 2A and 2B. Figure 2 shows the distribution of RF/SPF hybrid composites with different percentage content of hybrid fibers, and it is clear that there is poor adhesion between the hybrid fibers and the matrix when the incorporated in the composites. Figures 2A and 2B show good interfacial bonding with and without the incorporation of SPF content in the composites.

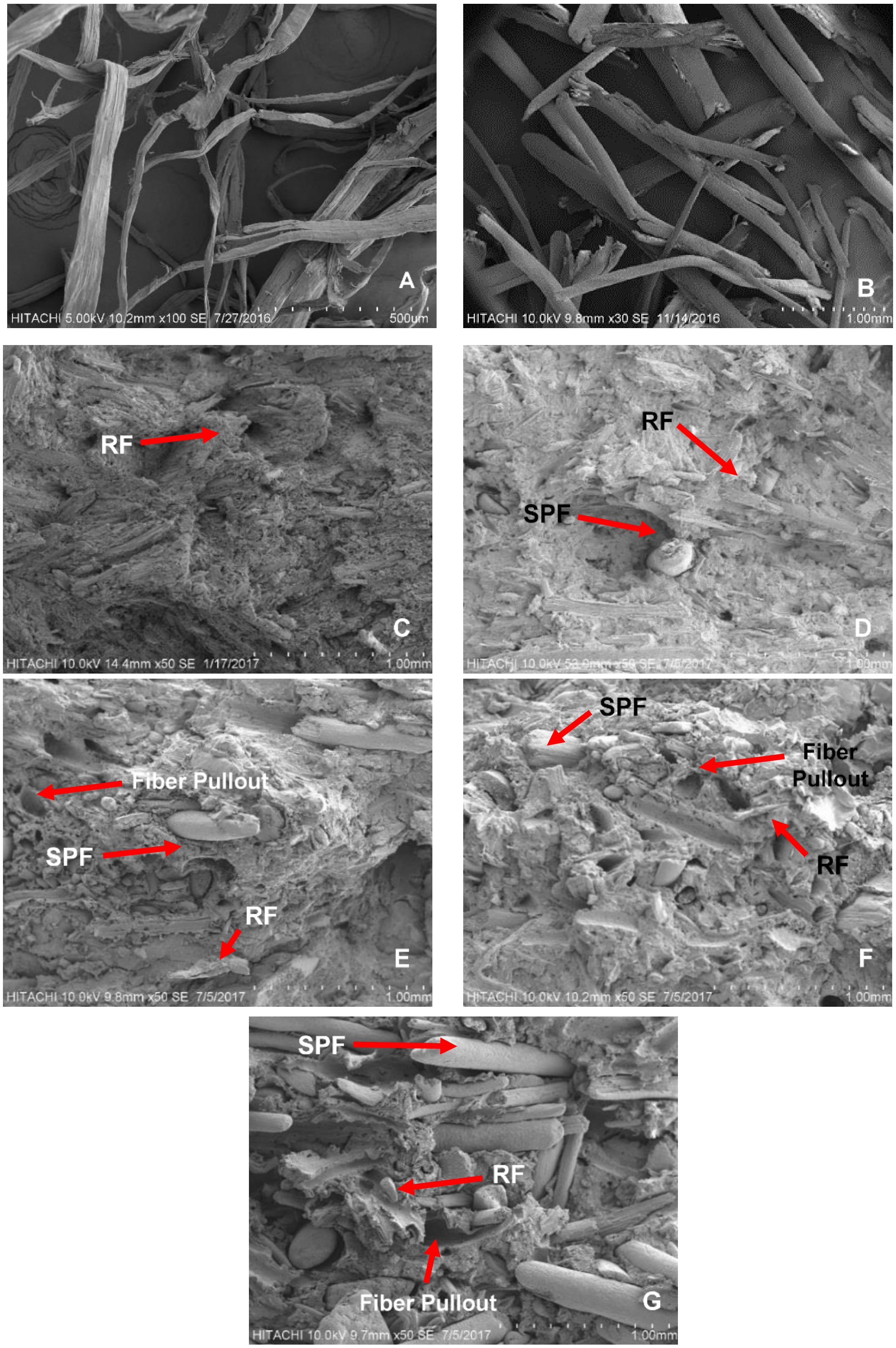

Fig. 2. Morphology of fractured surface of RF/SPF/TPU hybrid composites; (A) Roselle fiber (Radzi *et al.*, 2017b), (B) Sugar palm fiber, (A) RF-T composites, (B) RS-1 composites, (C) RS-2 composites, (D) RS-3 composites, and (E) SPF-T composites

In was observed that the low effectiveness of energy transfer from the matrix to the fiber has affected the sample's tensile strength as proved by the low tensile strength value (Fig. 1) (Lomelí Ramírez *et al.* 2011; Ridhwan *et al.* 2017d). Additionally, the tensile strength depends on the effect of adhesion between matrix and fiber. Such relationships also have been reported for hybrid composites such as kenaf/thermoplastics with cassava starch (Zainuddin *et al.* 2013) and seaweed / sugar palm fiber reinforced thermoplastic sugar palm starch/agar hybrid composites (Ridhwan Jumaidin *et al*. 2017d). Therefore, the inclusion of SPF in the composite leads towards the formation of voids, which can be clearly seen in the individual composite SPF morphology (Fig. 2G). From Figs. 2C to 2E, the voids, fiber pullouts, poor interfacial bonding, and less wettability fiber of the matrix can clearly be seen. These results contribute significantly to the failure in terms of decreased tensile strength with increased SPF percentage in the hybrid composite. Singh *et al.* (2015) reported that the presence of poor adhesion, voids, and fiber pullouts contributed to the decreased tensile strength of composites. In addition, the figures also show the SPF/RF in the form of fiber bundles, which prevented the stress transfer efficiency between fibers and the matrix. Besides, the size, agglomeration, and distribution of fibers also lead to the low ability of stress transfer between fibers and the matrix. Thus, these factors can cause low tensile strength (Rozman *et al.* 2001; Idicula *et al.* 2005; Busu *et al.* 2010).

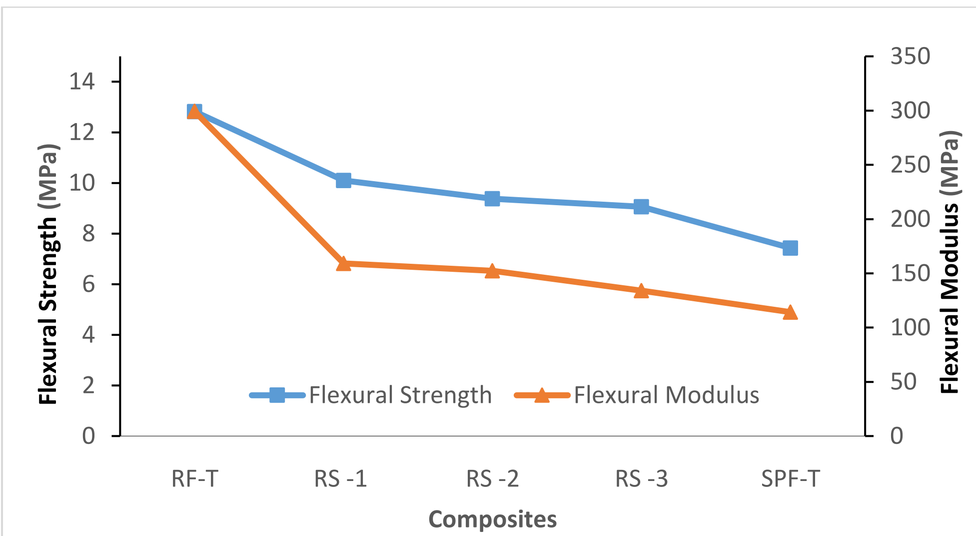

Fig. 3. Flexural strength and modulus of RF/SPF/TPU hybrid composites

Flexural Properties

Figure 3 shows the flexural strength and modulus of the RF/SPF/TPU hybrid composites. The results showed a similar trend with the tensile strength, in which the strength and modulus decreased with the increases in the incorporation of SPF. The RS-1 hybrid composites showed the highest strength; the strength value was 10.19 MPa and the modulus value was 168.23 MPa. This improvement in flexural strength can be attributed to the similar reasons with the increase in tensile strength. However, the decreased in strength and modulus might be due to non-uniform interfacial bonding and poor distribution of fibers between RF and the SPF and matrix (Kasim *et al.* 2015; Singh *et al.* 2015). The existence of voids also contributes to decrease the flexural strength (Busu *et al.* 2010). Same results have been reported by several authors (Shibata *et al*. 2005; Ozturk

2010; Kaewkuk *et al*. 2013; Salit *et al*. 2015), where the poor wettability between fibers and the matrix had contributed to the decrease in flexural strength. Additionally, Fig. 3 shows that the strength and modulus of single fiber SPF/TPU composites decreased more compared to those of the RF/TPU composites. This was possibly due to the SPF/TPU composites weak flexural strength which did not help improve their mechanical properties when the SPF was incorporated into the hybrid system. The decrease of the flexural strength may be due to the fibers agglomeration, which causes weaknesses in the interface between fibers and the matrix. Overall, all hybrid composites showed no effectiveness in the interaction between the RF/SPF hybrid and TPU composites. Several characteristics of hybrid composites such as good adhesion bonding, good wettability, fewer fiber pullouts, less void/pore, and agglomeration prevention are the main focus areas to be improved in terms of mechanical properties

Impact Properties

Figure 4 shows the impact strength of the RF/SPF/TPU hybrid composite. The results show that in contrast with the tensile and flexural strength, increases in the SPF content of the hybrid had increased its impact strength. Factors that may affect the composite impact properties include adhesion of fiber/matrix, fiber properties, and stiffness and rigidity properties (Rozman *et al.* 2001; Shibata *et al.* 2005; Ozturk 2010). Besides that, the increment of impact strength might be attributable to several factors such as incorporation and the presence of void lead to crack propagation (Nadlene *et al.* 2016b; Ridhwan *et al.* 2017d). Also, the fiber pull-out mechanism absorbs energy during friction impact (Nadlene *et al.* 2016b; Ridhwan *et al*. 2017d). Therefore, a good interlocking between the fibres and matrix surface may produce low resistance and a tendency for pull fiber-pull out. In this phenomenon, fiber breakage is more likely to occur with slight changes in the cracking of the plane rather than fiber pull-out (Nadlene *et al.* 2016b; Ridhwan *et al.* 2017d). This finding is in agreement with Nadlene *et al.* (2016b), who studied the effect of decreasing impact strength on a composite roselle / vinyl ester, and also Ridhwan *et al.* (2001), who studied the effect seaweed/SPF reinforced thermoplastic SPF /Agar hybrid composites.

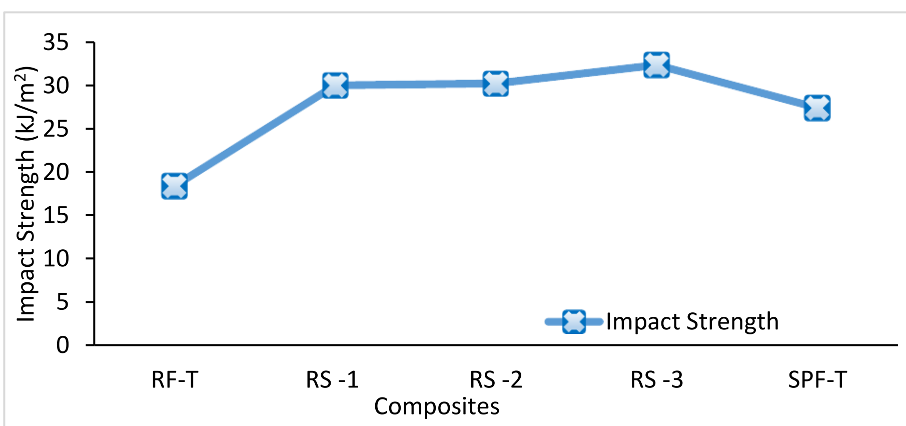

Fig. 4. Impact strength of RF/SPF/TPU hybrid composites

The increased impact strength may be due to the fact that the RF/SPF hybrid composites can absorb impact energy efficiently and have small localized deformations of fiber. In addition, the increase in stiffness and rigidity of the hybrid fibers has been a major factor influencing the impact properties of composite (Kalia *et al.* 2009). Also, the single fiber SPF/TPU composites showed the highest value compared RF/TPU composites. It is possible that the impact property was improved when the SPF was incorporated into the hybrid system. The incorporation of fiber/matrix and the low number of void that can cause microcracking may have contributed to the increased impact strength. Additionally, fibers and matrix with good flexibility can absorb impact energy (Jumaidin *et al.* 2017a,c). The RS-3 shows the highest value of 32.4 kJ/m^2 followed by 18.4 kJ/m^2 (RF-T), 30 kJ/m^2 (RS-1), 30.22 kJ/m^2 (RS-2), and 27.4 kJ/m^2 (SPF-T).

CONCLUSIONS

1. Hybrid composites generated from the roselle fiber/sugar palm fiber/thermoplastic polyurethane (RF/SPF/TPU) matrix were successfully produced through pre-mix and hot compression processes. Mechanical testing showed weak strength values when SPF was added into the hybrid composites.
2. The results showed a decrement in the SPF/TPU composites compared to the RF/TPU composites which contributed to the deterioration of its tensile and flexural strength when SPF was incorporated in the hybrid composites. The impact properties had increased when the SPF content is increased.
3. A morphological study (SEM) on tensile fracture showed weak hybrid-matrix interface adhesion and fiber pullouts from the matrix.
4. The hybridization of RF with SPF in the TPU matrix increased the impact strength of the hybrid composite. However, this was accompanied by reduction of the tensile and flexural strength of the hybrid composites.

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