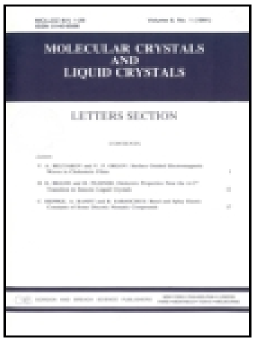

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Properties of Thermoplastic Starch/Kenaf Composite

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This paper reports on the physical and mechanical properties of the thermoplastic sago starch/kenaf fibre (TPSS/KF) composite. The composite was prepared through a compression molding technique at varying fibre contents of 0, 10, 20 and 30 wt.%, whilst the effect of the fibres incorporation was evaluated by physical and mechanical tests, as well as morphological analysis. Reduction of moisture content and denser composite were achieved with a higher fibre content. Meanwhile, the water absorption of the composite was lower than the thermoplastic with an increase in the kenaf fibre loading. Tensile testing improved strength and modulus with the increase of fibres content until an optimum was reached at 30 wt.% of fiber loading. Morphological analysis showed good wetting between the polymer matrix and fibres that provided the tensile improvement.

Keywords Sago starch; kenaf bast fibre; composite; reinforcement; plasticizer

1. Introduction

One of the major challenges in industry is related to environmental problems due to the major application of plastics. Utilization of petrochemical based plastics have been drastically increasing especially in packaging materials due to the high availabilities of materials, low cost and have good functional characteristics in terms of physical and mechanical properties [1]. This petroleum-based material however is non-biodegradable and can lead to serious ecological problems. There has been growing interest in the development of thermoplastic materials from biodegradable polymers that are derived from renewable resources. Among all of the natural plant based materials, starch has been chosen as one of the most promising candidates for future materials primarily because of the good combination of price and performance [2] Starch has polysaccharide groups of amylose and amylopectin that assist in the alteration of starch to a thermoplastic. Starch is easy to gelatinize, highly viscose, non-toxic, economical, biocompatible and broadly available throughout the year [3]. Unfortunately, thermoplastic starch alone does not show good properties under mechanical or physical tests, and needs to be modified or reinforced with

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other materials. Natural fibres were introduced and kenaf fibre was used as reinforcement to yield a lighter and cheaper yet durable composite [3]. Kenaf fibre (*Hibiscus canabinus*) has long been considered environmentally friendly and has superior stiffness and strength due to the existence of 60–80% cellulose, while hemicelluloses are responsible for biodegradation, moisture absorption and thermal degradation of the fibres, and are mainly utilized as reinforcement for Polymer Matrix Composites [4]. The aim of this research is to study the effect of the physical and mechanical properties of thermoplastic sago starch/kenaf fibre composites (TPSS/KF) with different fibre loadings.

2. Materials and Methods

2.1. Materials

Commercial sago starch powder (25% amylose) was purchased from Hup Seng Heng Sdn. Bhd, Malaysia. Kenaf bast fibres were supplied by the Institute of Tropical Forestry and Forest Products, Malaysia. Glycerol (99.5% purity) with density of about 1.261 g/cm^3 acted as the non-volatile plasticizer and was supplied by Merck Chemicals, Malaysia.

2.2. Fabrication Process

Sago starch was dried in an oven at 80°C for 8 hrs while the untreated kenaf fibres were overnight dried at 75°C under vacuum in order to reduce the humidity content. Kenaf fibres were cut into a length of approximately 400 μm. The weight ratio of glycerol/starch was 30/70 before adding the short fibres at 0, 10, 20 and 30 wt.%, which were mechanically stirred for 10 min. The mixture was then stored overnight at an ambient temperature of 20–25°C (R.H. 60 $\pm$ 5°C) in air tight polyethylene bags to avoid from moisture inclusion before melt blended using an internal mixer (Brabender). The following processing parameters have been used: temperature 130°C, rotor speed 100 rpm, residence time 10 min. The mixture was then granulated and stored under controlled conditions for 24 hrs before being compression moulded in an electrical heated hydraulic press (preheating at 165°C for 6 min and compression at 150°C for 6 min). The compression moulded sheet (150 $\times$ 150 $\times$ 3 mm) was cold pressed for 3 min. The samples were abbreviated as TPSS/0KF, TPSS/10KF, TPSS/20KF and TPSS/30KF for thermoplastic sago starch/kenaf fibre at 0, 10, 20 and 30 wt.% of kenaf fibre loadings.

2.3. Physical Testing

Five samples of dimensions 10 $\times$ 10 $\times$ 3 mm from each composition were tested for average density, water absorption and moisture content. The density test was conducted using a high precision electronic balance, densimeter (model-MD300s). The water absorption test was performed through immersion in water at room temperature for a period of two weeks, and the mass of the specimen before and after immersion was recorded. For the moisture content, each specimen were weighed, dried in a circulating oven at 80°C for 24 hrs and reweighed.

2.4. Tensile Test

Tensile test was carried out in accordance with ASTM D 412 using a Universal Testing Machine (Instron 3366). A crosshead speed of 5 mm/min was used and the test was performed at ambient temperature.

2.5. Morphology

The morphology of the TPSS/KF composites was observed under Scanning Electron Microscope (SEM) (Model Quanta 200 MK2) at 7 kV and 50/60 Hz. The surface of the samples was sputter-coated with gold-palladium since the specimens were non conductive. The specimens were placed on the sample holder by mounting with double-sided carbon tape and the observation was made on the morphology of the fracture surface of the specimens after the tensile test.

3. Results and Discussion

Figure 1 shows physical properties of the TPSS/KF composites in terms of moisture content and density. Figure 1(a) shows the density of the TPSS/KF composites at different loadings of kenaf fibres. Additions of kenaf fibres greatly influenced the density of the composites. The density of the TPSS/KF composites increased with the increment of fibre loading from 1.372 to 1.428, and to 1.519 g/cm^3 for fibre contents of 10, 20 and 30 wt.%, respectively. The increment of composite density was due to less air inclusion occurring. Hence, as fewer voids were formed, the material was denser [3].

Figure 1(b) illustrates the decrease in moisture content with the increase of fibre loading. The TPSS/0KF yielded a 9.5% moisture content, while for TPSS/30KF, the moisture content was the lowest at 5.5%. This was similar to that reported by [3] where the greater of

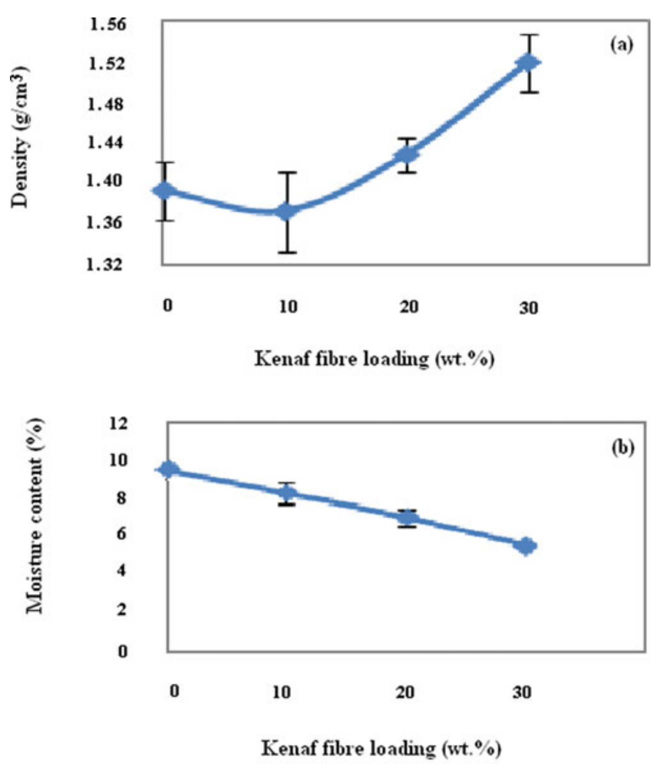

Figure 1. (a) Density, and (b) moisture content of TPSS/KF composites.

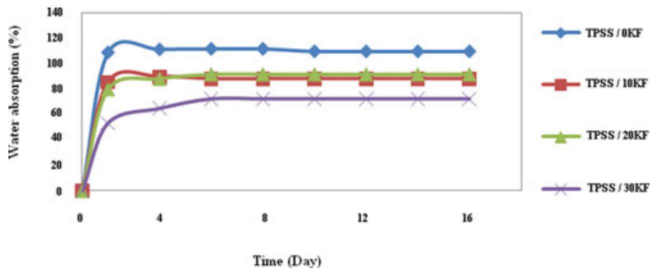

Figure 2. Effect of kenaf fibre on water absorption of TPSS/KF composites at different fibre loadings.

fibre loading, the lower the moisture content. Hence, the reinforced TPSS/KF composites absorbed less moisture compared to unreinforced TPSS/0KF. Lower moisture content with the TPSS/KF composites is due to a less hydrophilic behavior with the kenaf fibres than with starch [5]. In addition, good interfacial adhesion between the reinforcement and matrix leads to a minimum chance of water molecules from environment being attracted to the hydroxyl groups of the matrix [6].

Distinctive water absorption curves with different kenaf fibre loadings immersed in water at room temperature for TPSS/KF composite are shown in Fig. 2.

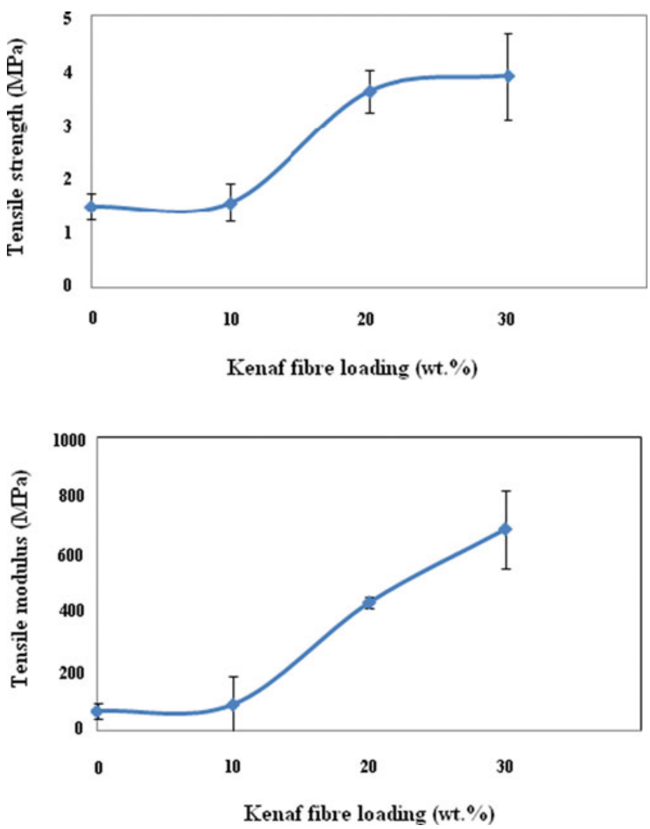

Figure 3. Tensile strength and tensile modulus of TPSS/KF composites at different fibre loadings.

In all cases, the percentage of water absorption was initially sharp and leveled off for a length of time as it approached equilibrium. In general, for the TPSS/KF composite, the equilibrium point of water absorption reached a maximum after 3 days of conditioning. However, it can be noted that unreinforced TPSS/0KF had the highest water absorption compared to the reinforced TPSS/KF composite due to the hygroscopic nature of glycerol and the hydrophilicity of the starch, which enhanced the water absorption. The existence of pores in the network of amylose within the starch entraps more water that is absorbed [3]. Meanwhile, the incorporation of kenaf fibres in the TPSS/KF composite decreases the water absorption capacity. The water absorption of the composites decreased with an increase in kenaf fibre loading due to the lower hydrophilic behavior of the kenaf fibre compared to starch. At the same time, good interfacial bonding at the fibre/matrix caused fewer cracks and voids. This result led to greater penetration of water through the voids [7].

Figure 3 represents a comparison of the tensile strength and modulus of TPSS/KF composites with various kenaf fibre loadings.

It is interesting to note that there was improvement in the tensile strength when the kenaf fibre was incorporated into the TPSS/0KF, where the maximum tensile strength was reached at 3.89 MPa at a fibre loading of 30 wt% (Fig. 3). The high strength of the composite was basically due to the fibre reinforcement and good interfacial adhesion between the matrix and the fibre, since both of these are hydrophilic materials [8]. Other than that, well dispersed of fibre in the polymer matrix contributed to high strength of the composite, as shown in the fracture surface in Fig. 4(b)–4(d) [9]. This data contradicts the report by [3], where the maximum tensile properties were achieved at 20 wt.% of fibre content. The enhancement of tensile properties also was due to hydrogen bonding between

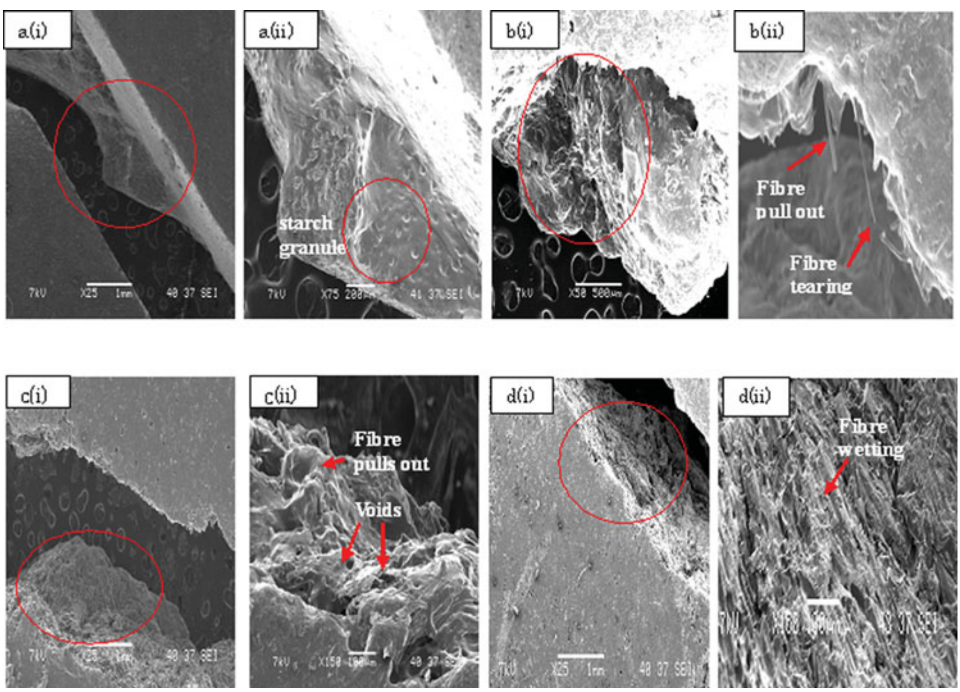

Figure 4. SEM images of TPSS/KF composite at (a) 0 wt.%, (b) 10 wt.%, (c) 20 wt.%, and (d) 30 wt.% of kenaf fibres.

the hydroxyl and carboxyl groups in the starch and kenaf fibre through the processing of the composites [10]. Hence, this improvement showed the good dispersion of the kenaf fibre in the composite and the efficiency of stress transfer from the matrix to the fibre [11].

The results obtained in Fig. 3 are validated with the fracture morphology between the unreinforced TPSS/0KF and reinforced TPSS/KF composites. From the surface conditions, TPSS/0KF had a clear and smooth surface as seen in Fig. 4 (ai), with no starch granules, showing that the starch was completely gelatinized [3]. At the same time, some of starch granules, and no cavities, were noticed on the fracture surface. This shows a high interfacial adhesion between the hydrophilic sago starch and glycerol. Thus, glycerol acts as an effective plasticizer in the thermoplastic in order to increase the flexibility of the material [12]. Meanwhile, Fig. 4(b)–4(d) show the rough surfaces of the TPSS/10KF, TPSS/20KF and TPSS/30KF composites and the wetting of kenaf fibre with the starch matrix. However, the fibres were not fully covered by the matrix and more fibres can be seen coming out in Figure (bii) and (cii). There are also gaps between the fibre and matrix, which contributed to a poor interfacial adhesion [8]. Figure 4 (dii) for TPSS/30KF illustrates good interfacial adhesion between the matrix and the fibres, where fewer fibres came out from the matrix, which can be traced to the fracture surface. More energy was required prior failure due to a strong interfacial bonding and good wettability [13]. Hence, the kenaf fibre acted as an effective reinforcement and may act as load carrier when the composite are subjected to load.

4. Conclusions

The physical, mechanical and morphological properties of starch-based biopolymer composite reinforced with kenaf fibre have been studied. The incorporation of kenaf bast fibre in the thermoplastic starch has potential, based on the enhancement obtained with regards to tensile strength and Young's modulus. Furthermore, SEM analysis showed good interfacial bonding between starch and the fibres, leading to enhanced strength. The reinforcing effect also showed that kenaf fibres have a good distribution within the matrix, resulting in efficient stress transfer. TPSS/0KF exhibited more moisture content compared to the reinforced TPSS/KF composites, as the addition of more fibres reduced the water absorption due to the inherent hydrophilicity of the fibre and the matrix, along with fewer voids appearing at the fibre-matrix interface.

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