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# Thermal, mechanical, and physical properties of seaweed/sugar palm fibre reinforced thermoplastic sugar palm Starch/Agar hybrid composites

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# ABSTRACT

The aim of this research is to investigate the effect of sugar palm fibre (SPF) on the mechanical, thermal and physical properties of seaweed/thermoplastic sugar palm starch agar (TPSA) composites. Hybridized seaweed/SPF filler at weight ratio of 25:75, 50:50 and 75:25 were prepared using TPSA as a matrix. Mechanical, thermal and physical properties of hybrid composites were carried out. Obtained results indicated that hybrid composites display improved tensile and flexural properties accompanied with lower impact resistance. The highest tensile (17.74 MPa) and flexural strength (31.24 MPa) was obtained from hybrid composite with 50:50 ratio of seaweed/SPF. Good fibre-matrix bonding was evident in the scanning electron microscopy (SEM) micrograph of the hybrid composites' tensile fracture. Fourier transform infrared spectroscopy (FT-IR) analysis showed increase in intermolecular hydrogen bonding following the addition of SPF. Thermal stability of hybrid composites was enhanced, indicated by a higher onset degradation temperature (259 °C) for 25:75 seaweed/SPF composites than the individual seaweed composites (253 °C). Water absorption, thickness swelling, water solubility, and soil burial tests showed higher water and biodegradation resistance of the hybrid composites. Overall, the hybridization of SPF with seaweed/TPSA composites enhances the properties of the biocomposites for short-life application; that is, disposable tray, plate, etc.

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#### 1. Introduction

Plastic products developed from petroleum-based polymer have caused negative environmental impacts due to the accumulation of non-biodegradable waste. Therefore, interest in utilizing the available natural resources for development of a more environmental friendly polymer has been growing continuously in order to tackle the issue.

Starch has been considered as the most promising resource for development of biopolymer due to several advantages; that is, they are renewable, biodegradable, widely available and low in cost [1]. Apart from the development of thin film, the potential of starch in development of rigid material, namely thermoplastic starch (TPS), was also discovered in previous works [2–5]. In general, TPSs possess similar properties to synthetic thermoplastic which enables the uses of various fabrication machines for its production; that is, extrusion, compression moulding, injection moulding, etc. However, pure TPS also possesses several disadvantages such as poor mechanical strength and water resistance, which limits its potential application. Therefore, modification of TPS is often necessary in order to prepare this material for real application. Blending of TPS with other natural polymer appears to be a promising approach in order to preserve the biodegradable aspect of this biopolymer. Recently, the tensile, thermal and physical properties of modified TPS derived from sugar palm starch and agar has been reported [6].

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The results show significant improvement in the tensile properties of thermoplastic sugar palm starch/agar (TPSA) blend following the incorporation of agar.

Moreover, reinforcing TPS with natural fibre is another interesting approach to resolve the drawbacks of TPS. Several results have been reported on the addition of natural fibres such as kenaf [7–9], cotton [10,11], coir [12,13], sugar palm [14], kapok [15] and jute [15] into TPS matrix. Most of these studies were focused on improving the mechanical and thermal properties of TPS. In addition, enhanced water resistance was also obtained with incorporation of natural fibre into TPS material [8].

*Eucheuma cottonii* seaweed (also known as *Kappaphycus alvarezii*) belongs to the "red seaweed" family and is massively cultivated for the production of its hydrocolloids, namely kappacarrageenan (k-carrageenan). However, due to the relatively low carrageenan content in the raw seaweed (25–35%), a huge amount of solid waste was produced during processing which is yet to be utilized [16]. Meanwhile, sugar palm (also known as *Arenga pin-nata*) is a tropical tree that belongs to the *Palmae* family. Apart from the production of its neera sugar and starch [17], this tree was also known for producing natural fibre from its trunk, namely sugar palm fibre (SPF). Traditionally, SPF was used for various outdoor applications such as broom, ropes and roof materials [18]. This is due to the excellent characteristics of SPF as a natural fibre such as good tensile strength and resistance to water [19].

Even though there are previous studies reported on hybridization of SPF with other fibres, that is, kenaf [20] and glass fibre [21], these studies were using synthetic polymer as a matrix which is not biodegradable. It is clear from the literature review that no study has been carried out on using biodegradable TPSA as a polymer matrix for hybridized SPF/seaweed filler. SPF is known to possess both mechanical strength and water resistance; therefore, the hybridization of SPF with seaweed (Sw) seems to be a good combination to improve the properties of the material. In the present study, the effects of SPF hybridization with Sw/TPSA composites were investigated in terms of the mechanical, thermal and physical properties. Various experimental approaches were used to characterize the properties of the composites including Fourier transform infrared spectroscopy (FT-IR), Scanning electron microscopy (SEM), thermogravimetric analysis (TGA), mechanical testing, water absorption, thickness swelling, solubility and soil burial.

#### 2. Materials and methodology

# 2.1. Materials

Sugar palm starch (SPS) was extracted from the sugar palm tree population location at Jempol, Negeri Sembilan, Malaysia. The interior part of the trunk was crushed in order to obtain the woody fibres which contain the starch. These woody fibres were soaked in fresh water followed by squeezing to dissolve the starch into the water. Water solution that contained starch was filtered to separate the fibres from the solution. This solution was then left for sedimentation of the starch. The supernatant was discarded and the wet starch was kept in the open air for 48 h followed by drying in an air circulating oven at 105 °C for 24 h. Agar powder was procured from R&M Chemicals and glycerol was purchased from Science chem.

Seaweed waste from *Eucheuma cottonii* species was obtained as waste material from seaweed extraction. The solid wastes were obtained after hot alkaline extraction process to obtain carrageenan. This by-product was cleaned with water and dried at 80 °C for 24 h in a drying oven. The dried seaweed wastes were ground and sieved then kept in zip-locked bags until further pro-

#### Table 1

Relative amount of reinforcing materials in composites.

Seaweed (%)	Sugar Palm Fibre (%)	Composites
100	-	Sw
75	25	HC1
50	50	HC2
25	75	HC3
-	100	SPF

cess. The moisture content and average particle size of the ground seaweed are  $0.75 \pm 0.2\%$  and  $120 \,\mu\text{m}$  respectively. Sugar palm fibre (SPF) was obtained from sugar palm trees at Jempol, Negeri Sembilan, Malaysia. The obtained fibres were ground and screened to obtained 2 mm fibre size. The average moisture content of the fibre is  $6.55 \pm 0.1\%$ .

#### 2.2. Sample preparation

Thermoplastic SPS/agar (TPSA) matrix was prepared according to our previous work [6]. The ratio of starch:agar:gycerol was maintained at 70:30:30 (wt%). After this preliminary step, the resulting blend was melt-mixed using Brabender Plastograph at 140 °C and rotor speed of 20 rpm for 10 min. This mixture was granulated by means of a blade mill equipped with a nominal 2 mm mesh and thermo-pressed in order to obtain laminate plate with 3 mm thickness. For this purpose a Carver hydraulic thermo-press was operated for 10 min at 140 °C under the load of 10 t. The same processes were used for the modification of TPSA with hybridized seaweed/SPF. The weight fraction of the reinforcing materials was shown in Table 1 where the matrix was maintained at 80 wt%. All samples were pre-conditioned at 53% relative humidity (RH) for 48 h prior to testing.

#### 2.3. FT-IR analysis

Fourier transform infrared (FT-IR) spectroscopy was used to detect the presence of functional groups existing in thermoplastic SPS blends. Spectra of the material were obtained using an IR spectrometer (Nicolet 6700 AEM). FT-IR spectra of the sample  $(10 \times 10 \times 3 \text{ mm})$  was collected in the range of  $4000-400 \text{ cm}^{-1}$ .

#### 2.4. Scanning electron microscope (SEM)

The morphology of tensile fractured surfaces was observed under a scanning electron microscope (SEM), model Hitachi S-3400N, with acceleration voltage of 10 kV.

#### 2.5. Tensile testing

Tensile tests were conducted according to ASTM D-638 at the temperature of  $23 \pm 1$  °C and relative humidity of  $50 \pm 5$ %. The tests were carried out on five replications using a Universal Testing Machine (INSTRON 5556) with a 5 kN load cell; the crosshead speed was maintained at 5 mm/min.

### 2.6. Flexural testing

Flexural tests were conducted according to ASTM D-790 at a temperature of  $23 \pm 1$  °C and relative humidity of  $50 \pm 5\%$ . The samples were prepared with dimensions of 130 mm (L) x 13 mm (W) x 3 mm (T). The tests were carried out on five replications using a Universal Testing Machine (INSTRON 5556) with a 5 kN load cell; the crosshead speed was maintained at 2 mm/min. The support span length was set at a ratio of 16:1 to the thickness of samples.

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