Cellulosic Pulp Fiber as Reinforcement Materials in Seaweed-Based Film

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Composite materials made from renewable resources can minimize the environmental pollution. In this work, biocomposite films were produced using seaweed as matrix and empty fruit bunch (EFB) pulp fibers as reinforcement. Based on the results, the EFB pulp-seaweed composite films exhibited better mechanical properties than the seaweed film. It was also observed that 50% EFB pulp loading gave the highest tensile strength (81.4 MPa) and elongation at break (5.4%). This phenomenon was supported by SEM analysis, in which more fiber breakage than fiber pull-out was observed on the tensile fracture surface of composite film. Additionally, no agglomeration of the pulp fibers was observed. Instead, the pulp fibers were homogenously distributed throughout the film. In contrast, the contact angle of the seaweed-based films started to decrease once the pulp fibers were added. The decrease in the contact angle was attributed to the hydrophilic nature of the pulp fibers. Nevertheless, the contact angle values of all composite films were still comparatively high and thus, this would not affect their application as a packaging film.

Keywords: Seaweed; Oil palm empty fruit bunch; Composite film; Mechanical; SEM; Contact angle

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INTRODUCTION

The world is threatened by a number of issues, such as the shortage of non-renewable petroleum, climate change, and geopolitical conflicts that are connected to minerals, metals, and incineration waste residues. To address such problems there is an interest in the development of sustainable materials and environmentally friendly processes. For example, plastic produced from renewable feedstock (biomass and waste) is a promising replacement for petroleum-based polymers, as it can reduce the global dependence on fossil fuel resources and supply simplified end-of-life disposal (Khalil *et al.* 2012). Moreover, bio-composites that mix natural fibers (*i.e.*, flax, hemp, kenaf, jute, and cotton) with polymer matrices derived from renewable resources (*i.e.*, polylactic acid, cellulose esters, polyhydroxybutyrates, starch, and lignin) have emerged as materials that can decrease environmental impact (Mohanty *et al.* 2002).

Biodegradable polymers can be derived from animals, plants, or microorganisms. Polysaccharides, proteins, and lipids are the common natural and degradable polymers used in the packaging, automobile, agricultural, medicine, and pharmaceutical sectors (Vroman and Tighzert 2009). Of these biodegradable polymers, polysaccharides are rich

in hydroxyl (–OH) functional groups, which results in good film-forming properties due to the formation of hydrogen bonding networks that stabilize the intra- and inter-polymeric chain interactions. Furthermore, the presence of hydroxyl groups allows for good solvation in aqueous mediums and possible chemical modifications, like esterification or etherification. Seaweed is an example of a biodegradable polymer that is mainly composed of polysaccharide components, but it is also rich in essential and non-essential amino acids, as well as many other molecules (Rinaudo 2008). The main application of seaweed is food. Recently, seaweed derivatives, such as agar, alginates, and carrageenan, have been applied in drug and pharmaceutical products as thickening or gelling agents (Kadam *et al.* 2015). Alginate films with high salt concentrations are used as an edible susceptor to increase crunchiness when cooking or warming breaded foods in microwave ovens (Albert *et al.* 2012). Additionally, alginates are also prepared as emulsifiable films, which have good barrier and mechanical properties and provide better protection for encapsulated active substances (Hambleton *et al.* 2009). Siah *et al.* (2015) formed film directly from raw seaweed. Nevertheless, raw seaweed as a polymer matrix is less studied, even though no chemical and energy consumption is needed for isolation, which makes material preparation easier and cheaper.

Malaysia is one of the largest oil palm producers and exporters in the world. In a typical palm oil plantation, almost 70% of the fresh fruit bunches, such as the oil palm trunks, oil palm fronds, empty fruit bunches, palm pressed fibers, palm shells, and palm oil mill effluent, are turned into waste (Abdullah and Sulaiman 2013). Due to disposal problems, oil palm wastes have created a habitat for pests and insects, which has caused a severe environmental problem. The reuse of oil palm waste boosts the value of food and agriculture plantations by taking advantage of these biomass residues. Empty fruit bunch (EFB) fibers are among the most widely used oil palm wastes for the production of plywood (Khalil *et al.* 2010), polymer bio-composites (Chai *et al.* 2009), biofuel (Shuit *et al.* 2009), and pulp and paper (Wan Daud and Law 2011).

EFB fiber is well known for its toughness and high tensile strength (John *et al.* 2008; Ahmed *et al.* 2010). Several studies have been carried out that incorporate EFB fibers into a polymer matrix, and the resulting composite material has improved tensile strength, Young's modulus, flexural strength, and flexural modulus (Prasad *et al.* 2009; Ahmed *et al.* 2010; Tshai *et al.* 2016). However, the compatibility between the polymer matrix and the lignocellulosic fiber as the reinforcement material has always been an issue. Hydrophobic polymer matrices and hydrophilic lignocellulose biomass matter are not compatible. Only weak bonds are formed between them, such as Van de Waals forces or mechanical anchoring induced by compressive residual stress. Thus, agglomeration of fibers often occurs in the polymer matrix, which eventually deteriorates the strength of the composite (Bax and Müssig 2008; Širvaitiene *et al.* 2013).

Alkaline pulping is the most common pulping approach that chemically separates cellulose fibers from wood and non-wood biomasses. Alkaline pulping has been utilized to modify fiber surfaces to improve their interfacial adhesion with a polymer matrix. Alkaline pulp improves the mechanical properties of polymer matrix composite due to its higher crystallinity index and rougher surface compared with fibers that had not been subjected to the alkaline conditions. Alkaline pulping tends to remove the cementing materials (*i.e.*, lignin and hemicellulose) and makes the cellulose fibrils align along the direction of tensile deformation (Bledzki and Gassan 1999; Mwaikambo and Ansell 2002). According to Tay *et al.* (2010), soda pulp from EFB fiber shows a low probability to agglomerate, a high aspect ratio, and better distribution when incorporated into

polypropylene matrices. Khan *et al*. (2016) also verified the enhancement of thin film mechanical properties when fractionated bleached pulp fibers are reinforced into a biodegradable polymer (alginate) matrix.

To date, the incorporation of pulp fibers into a seaweed matrix has not yet been explored. Moreover, the use of seaweed directly as the raw material for the matrix instead of using a seaweed-derived polymer has not been studied either. The purpose of this research was to develop bio-composite films from seaweed and EFB pulp and to determine the characteristics of the developed composite films.

EXPERIMENTAL

Materials

Oil palm empty fruit bunch (EFB) fiber and dry seaweed were obtained from the Malaysian Palm Oil Board (MPOB), Selangor and Sabah, Malaysia, respectively.

Methods

Preparation of EFB pulps

The EFB pulp was produced by the soda-anthraquinone pulping process in a 4 L stationary stainless steel digester fitted with a computer-controlled thermocouple. The pulping condition was as follows (Rodríguez *et al*. 2010): solid to liquid ratio (S:L) of 1:6 (w/w), pulping reaction time (*t*) of 70 min, pulping reaction temperature (*T*) of 170 °C, 15% (w/w) alkali charge based on the oven-dried weight of EFB fibers, and 1% (w/w) anthraquinone dosage based on the oven-dried weight of fibers. Upon completion of the pulping, the pulp was washed with running water in a 200-mesh size screen box. The cleaned pulp was mechanically disintegrated in a three-bladed mixer before it was screened on a flat-plate screen with 0.15 mm slits (Somerville Screen, Testing Machines Inc., DE, USA). The screened pulp was collected with the 200-mesh size screen box. After dewatering and homogenizing, the accepted pulp was stored at 4 °C for further use.

Preparation of seaweed particles

The raw seaweed was washed with tap water several times to drain away contaminants such as salts and sand particles. The cleaned seaweed was cut into small pieces and oven-dried at 40 °C for 2 to 3 days until a constant weight was obtained. The seaweed particles were stored in an oven at 40 °C before use. Drying and storing the seaweed at 40 °C prevented thermal degradation of the seaweed structure.

Preparation of seaweed/EFB pulp composite film

A seaweed solution with a concentration of 2% (w/v) was prepared. Solutions of EFB pulp fiber filled seaweed composites were prepared by loading 0%, 10%, 20%, 30%, 40%, and 50% of the oven-dried weight pulp fiber, which was calculated based on wt.% of the seaweed, into the seaweed matrix. The seaweed particles were first softened by soaking in distilled water overnight. After soaking, the gelatinized seaweed was formed. The EFB pulp was dissolved with distilled water for 1 min before being added and mixed together with the seaweed solution. The mixture was occasionally stirred to prevent hardening of the solution, and it was also kept below 100 ºC to prevent thermal degradation of the pulp. After the mixture was completely dissolved, it was evenly

poured into a container (32 cm × 25 cm × 9 cm) and dried at room temperature for 24 to 48 h. All films were stored in a desiccator before testing.

Properties of Seaweed/EFB Pulp Composite Film

Film thickness

The thickness of the composite films was measured with a precision digital micrometer to the nearest 0.0001 mm at 20 random locations on each film. The mean thickness value from all samples was used in the tensile strength calculations.

Mechanical test

The tensile tests were run using a single arm texture analyzer TA.XT plus (Stable Micro System Ltd., Godalming, UK) under 58% relative humidity and room temperature. It was controlled by Texture Exponent software (Godalming, UK) using a 30 kg load cell based on ASTM D882-02. In the tensile testing, at least five probes were cut into rectangular pieces of 100 mm × 20 mm. Initially, the leaving initial grips separation (L_o) was 60 mm with a cross-head speed of 100 mm/min. The tensile strength (TS) and elongation at break (EAB) were calculated from the tensile force and length of specimen after fracture was obtained.

Scanning electron microscopy (SEM)

The surface morphology and tensile fracture surface morphology of the composite films were examined by a scanning electron microscope (EVO MA10, Carl-ZEISS SMT, Oberkochen, Germany). The samples were placed onto the SEM holder with double-sided electrically conducting carbon adhesive tape to prevent a surface charge from forming on the specimens when they were exposed to the electron beam. The specimens were then coated with a thin gold-palladium layer using a Polaron SC515 sputter coater (Fisons Instruments, UK). The SEM applied a focused beam of high energy electrons to produce a variety of signals at the surface of the solid specimens.

Contact angle measurement

The static contact angle of the films was measured using a KSV CAM 101 (KSV Instruments Ltd., Finland), where the sessile drop technique was applied. A drop of test liquid was uniformly placed on the film surface, and the image was recorded for 5 s at a speed of 5 frames per s. Each film test was repeated 5 times, and the mean value of each film was calculated.

Statistical analysis

DSAASTAT ver. 1.101 by Andrea Onofri was used for statistical analysis of all data from each testing. Data was analyzed by analysis of variance (ANOVA) and multiple comparisons of means were carried out using Turkey's test.

RESULT AND DISCUSSION

Thickness of Film

Table 1 shows the thickness of the seaweed and seaweed/EFB pulp composite films. The thickness of the blank seaweed film (0% EFB) was 0.74 ± 0.03 mm. When the EFB pulp was reinforced into the seaweed matrix, the thickness of the films increased with increasing pulp fiber loadings. This was due to the increase in stacking layers of

pulp fibers in the composite film. A similar observation was also reported by Singh *et al.* (2011), in which the thickness of the bagasse fiber/polypropylene composite increased when more bagasse fibers were incorporated. Nevertheless, it was found that the thicknesses of some composite films were not significantly different when 10 to 30%, 20 to 40%, and 40 to 50% of EFB pulp loadings, respectively. In addition, the resulting composite films were more opaque with the increase in pulp fiber (Fig. 1). Atef *et al.* (2015) stated that the thickness of the film affected the transparency of the film. Thus, higher pulp loading in the seaweed matrix led to more opaque film, as shown in Fig. 1. Furthermore, black spots were observed on the films after the pulp fibers were added (Fig. 1). The spots on the composite films were silica bodies embedded in the EFB pulp fibers (Gunawan *et al.* 2009; Harsono *et al.* 2016). Hence, more EFB pulp added into the seaweed matrix resulted in more black spots on the surface of the film.

Table 1. Thickness of Seaweed Films Incorporated with EFB Pulp

Pulp Loading (%)	Thickness (mm^2)*
0	0.74 ± 0.03^{a}
10	1.20 ± 0.07^{b}
20	$1.39 \pm 0.10^{b,c}$
30	$1.40 \pm 0.10^{b,c}$
40	$1.58 \pm 0.13^{c,d}$
50	1.71 ± 0.15^{d}

* Thicknesses are the mean ± standard deviation.
[a, b, c, d] Values along each row with the same capital letter are not significantly ($p>0.05$) different as analyzed by Tukey's Test

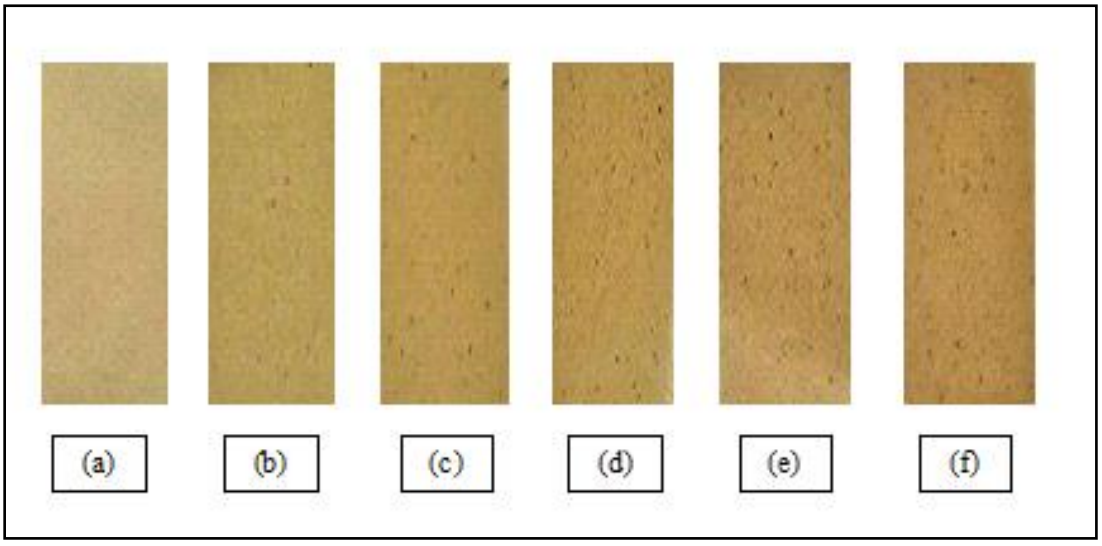


Fig. 1. Samples of the composite films with varying EFB pulp fiber loadings: (a) 0%, (b) 10%, (c) 20%, (d) 30%, (e) 40%, and (f) 50%

Mechanical Properties of Film

When composite films are utilized for packaging applications, it is expected that they are capable of withstanding different kinds of stress encountered during use (Cagri *et al.* 2001). The common mechanical tests used to identify the mechanical properties of the films are of the TS and EAB. TS is the maximum tensile stress that can be sustained by the sample before failure, whereas EAB indicates the film extensibility and flexibility, which is determined at the point where the film breaks during the tensile testing (Thongsane 2009; Siah *et al.* 2015).

According to Table 2, the TS of the blank seaweed film was the lowest at 45 MPa. The TS of the composite films increased with increased EFB pulp content (Table 2). The enhancement of the TS in the composite films indicated that there was good compatibility between the EFB pulp and seaweed. This phenomenon was due to the good bonding between the hydrophilic EFB pulp and seaweed matrix (Yang *et al.* 2006). Because the treated EFB pulp surfaces and seaweed matrix were mostly covered with hydroxyl groups (-OH groups), 3D continuous hydrogen bonding networks formed between these components (Xu *et al.* 2003). Thus, any stress applied on the composite film was efficiently transferred from the matrix to the pulp. Similar findings were reported by Eng *et al.* (2014), where the silane treated hydrophobic oil palm mesocarp fiber (OPMF) showed better compatibility compared with unmodified hydrophilic OPMF in the hydrophobic PLA/PCL/nanoclay/OPMF hybrid composites.

Table 2. Tensile Strength (TS) and Elongation at Break (EAB) of Seaweed Films Incorporated with EFB Pulp

Pulp Loading (%)	TS (MPa)*	EAB (%)*
0	45 ± 2^{a}	2.5 ± 0.3^{a}
10	53 ± 4^{b}	3.3 ± 0.4^{b}
20	61 ± 2^{c}	4.1 ± 0.3^{c}
30	$64 \pm 3^{c,d}$	$4.6 \pm 0.5^{c,d}$
40	68 ± 2^{d}	5.0 ± 0.1^{d}
50	81 ± 1^{e}	5.4 ± 0.2^{d}

* Results are the mean ± standard deviation.
[a, b, c, d, e] Values along each row with the same capital letter are not significantly ($p>0.05$) different as analyzed by Tukey's Test

Increased pulp fiber loading reduces the amount of matrix used during the preparation of composite film, and the TS of this composite film declines as the pulp content increases (Tay *et al.* 2010). In the present study, increased pulp fibers incorporated into a constant concentration of seaweed matrix increased the TS of these composite films, as shown in Table 2. The highest TS (81 MPa) was attained at the highest EFB pulp concentration (50%). This finding verified that the amount of seaweed matrix applied in this study was sufficient to hold the incorporated EFB pulp fibers. Moreover, the pulp fiber was effective as a reinforcement to the matrix, where the pulp fiber attributed resistance to the stress that was applied (Thongsane 2009). In addition, the homogenous distribution of pulp fibers in the matrix might contribute to the improvement of the TS. This was further examined by SEM. A previous study reported that the highest TS was attained when up to 65% of alkaline treated baggase fibers were incorporated into a biodegradable aliphatic polyester (Satyanarayana *et al.* 2009).

In comparison to the blank seaweed film, the EAB increased with the addition of EFB pulp in the seaweed films (Table 2). However, the results of the EAB testing was in

contrast to the results obtained by Tay *et al.* (2010), where the incorporation of the EFB pulp decreased the EAB values of the EFB pulp/propylene composite. This phenomenon might be due to the hydrophilic properties of the EFB pulp-seaweed composite film compared with the Tay *et al.* (2010) study. Because EFB pulp and seaweed are both hygroscopic materials, they tend to absorb moisture from the surrounding environment. Therefore, the uptake of water by these materials indirectly imparted a plasticization effect in the composite films, as water is a common plasticizer in most hydrophilic films (Zhang and Han 2008). Hence, it was believed that the increase of pulp fiber filling increased the EAB values of the composite films (Table 2). This finding was further verified through the contact angle measurement. Additionally, it was observed that the EAB value of composite film with 30%, 40% and 50% of EFB pulp loadings were insignificantly different.

Morphology Properties of Film

Surface morphology

The morphology of the composite films incorporated with different EFB pulp concentrations are shown in Fig. 2.

At 100 times magnification (Fig. 2), the blank seaweed film exhibited a smooth surface. The surface of composite films became rough when the EFB pulp was added. An even distribution of EFB pulp fibers across the seaweed matrix was observed, even for the highest EFB pulp content (50%). This indicated that the pulp fibers and seaweed matrix were homogenously well-blended with each other. Moreover, the flattened fibers were overlapping and entangling with each other in the composite films with an average diameter of 15.60 ± 2.8 µm (as indicated in Fig. 2. (f) and (h)). No agglomerations of the pulp fibers were observed. Hence, a good mechanical strength (*i.e.*, TS) was achieved (Table 2).

Fracture surface morphology

The TS behavior of the composites was further studied by examining the fracture surface of these films under SEM. Without the addition of the EFB pulp, breakage manner and plate-like areas on the fracture surface of the seaweed films were observed (Fig. 3. (a) and (b)). This phenomenon was due to the brittleness of the seaweed matrix. A similar observation was seen for the breaking pattern of polylactide film in the study carried out by Plackett *et al.* (2003).

At 10% pulp fiber loading, the pulp fiber matrix with a layer of the matrix covering it was pulled out from the seaweed, as indicated by a circle in Fig. 3(c). This result indicated that the EFB pulp fibers had an effective interaction with the seaweed matrix. As both materials were hydrophilic, they were able to form hydrogen bonds between each other (Bax and Müssig 2008). Subsequently, strong adhesion interaction formed between both components with better wetting of the pulp fiber by the seaweed matrix. According to Coutinho *et al.* (1997), good wetting is essential for the establishment of strong interfacial adhesion. Thus, the pulp fibers supported stress transfer from the seaweed matrix to obtain optimum strength properties of the composite.

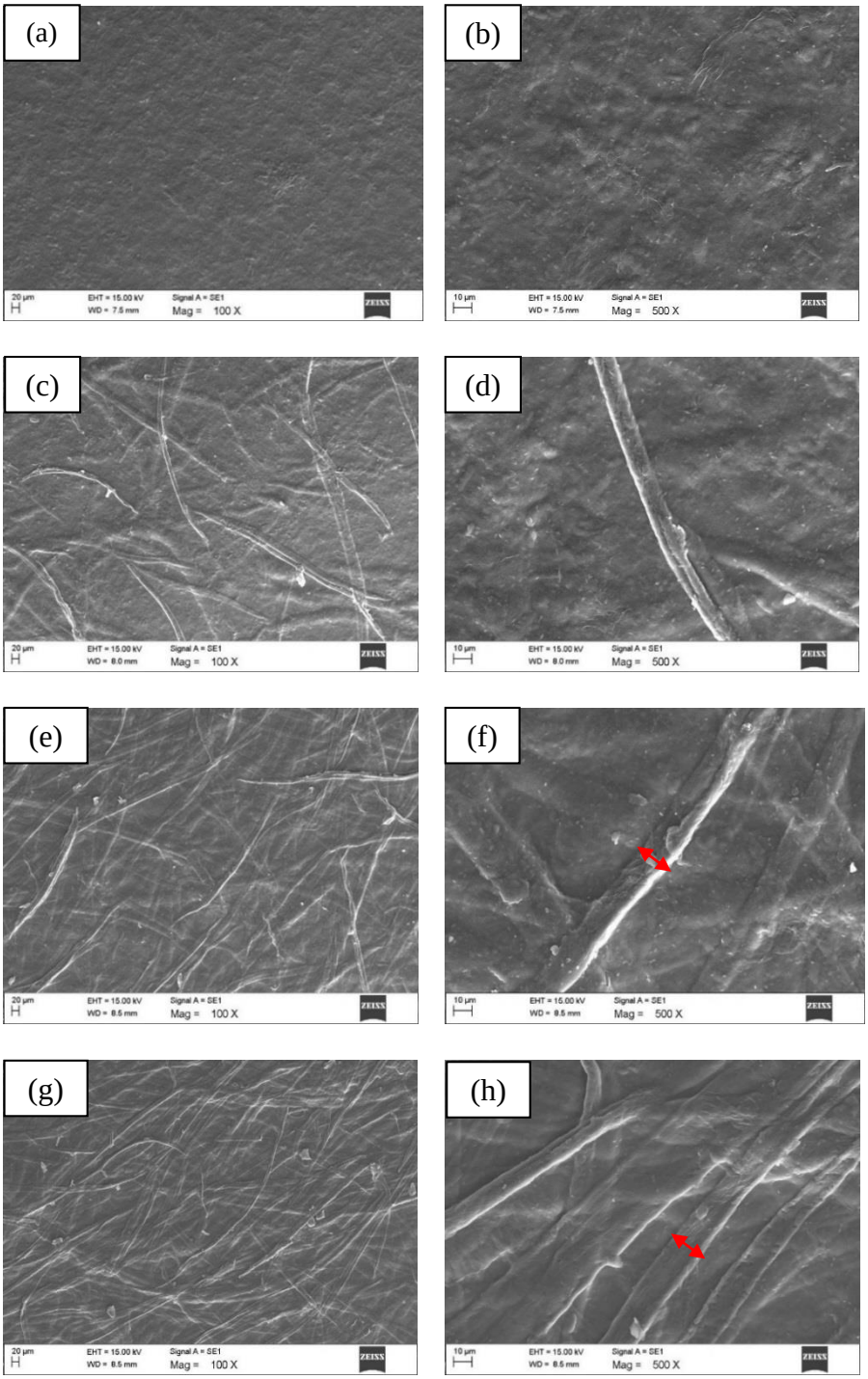
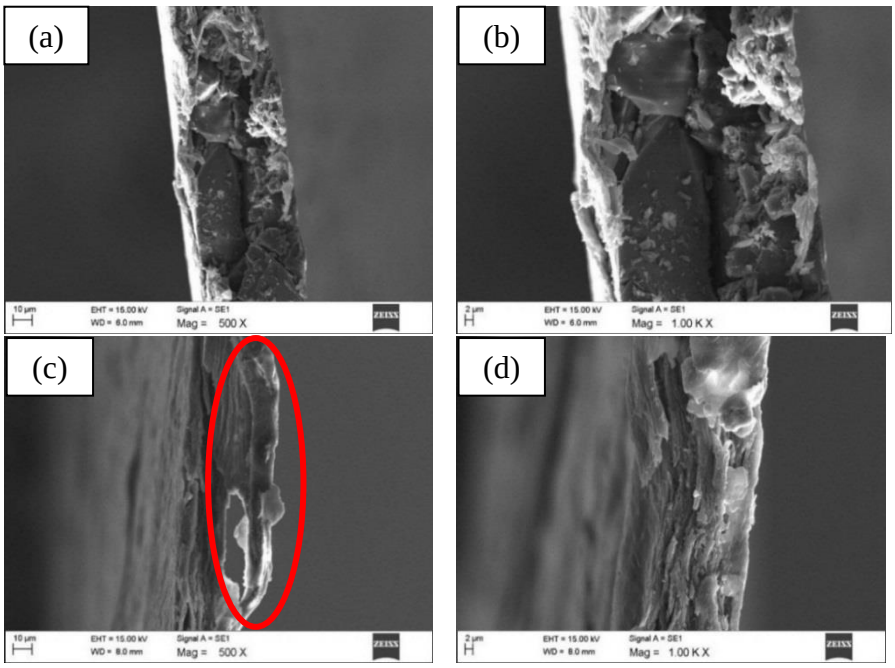


Fig. 2. SEM micrographs of the seaweed films with varying EFB pulp concentrations. (a) 0%, (c) 10%, (e) 30%, and (g) 50% at 100 times magnification. (b) 0%, (d) 10%, (f) 30%, and (h) 50% at 500 times magnification. The red arrows in (f) and (h) indicate the average diameter of fiber.

More pulp fiber pull-outs and holes were observed for 30% pulp loading in the seaweed film, as indicated by a circle in Fig. 3(f). The gaps between the pulp fiber and seaweed matrix might have been due to the fiber-matrix debonding during mechanical testing, poor approximation during composite production, or poor interphase adhesion between the reinforcing agent and the matrix element (Bax and Müssig 2008). However, the addition of 50% pulp fiber in the seaweed matrix showed more fiber breakage than fiber pull-out at the facture surface of the composite film, as indicated by the circles in Fig. 3(h). Fiber breakage at the fracture point indicated strong bonding between the EFB pulp and the seaweed matrix (Sangthong *et al.* 2009). This observation correlated with the TS results, in which the composite films with the highest fiber loading, 50%, had the highest TS.

Contact Angle Measurement

The contact angle measurement was conducted to study the surface hydrophobicity and wettability properties of the composite films (Rane *et al.* 2014). When the contact angle is less than 90°, fluid spreads over a large area on the surface, which means that the wettability of the surface is favorable. Thus, the surface is less hydrophobic. In contrast, if the contact angle is more than 90°, fluid minimizes its contact with the surface and forms a compact liquid droplet. Thus, the wettability of the surface is unfavorable, and the surface is more hydrophobic (Yuan and Lee 2013).



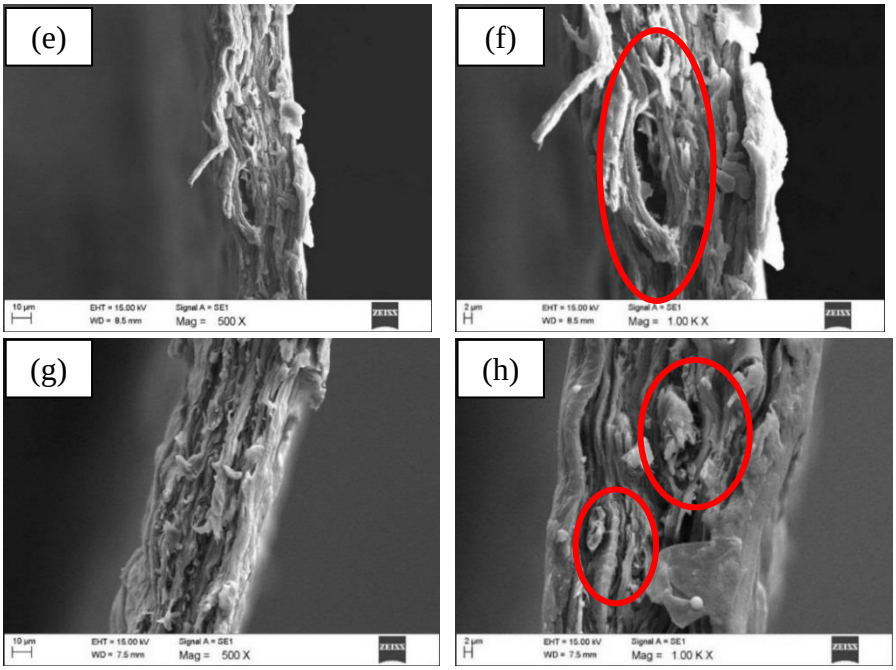


Fig. 3. SEM micrograph of fracture surfaces of the seaweed films with varying EFB pulp content. (a) 0%, (c) 10%, (e) 30%, and (g) 50% at 500 times magnification. (b) 0%, (d) 10%, (f) 30%, and (h) 50% at 1000 times magnification. The red circles in (c), (f), and (h) indicate fiber pull-out and breakage.

In this study, the contact angle of the blank seaweed film was 75.6°, which indicated that it was not very hydrophobic (Fig. 4). A previous study reported that the hydrophobic properties of the film increased when the reinforcing agents were added because there was better interaction between the fiber and matrix (Rane *et al.* 2014). However, that was not the case for this study. The water contact angle decreased when there were increased amounts of EFB pulp incorporated. Therefore, the hydrophobic properties of the films decreased. This phenomenon was due to the hydrophilic nature of the EFB pulp fiber. The EFB fiber is made up of cellulose, which contains more polar hydroxyl groups (Peltola 2005). Nevertheless, the wetting of all composite films was considered acceptable because the contact angles were not the lowest (Fig. 4) when compared to a previous study, in which the maximum contact angle (from the highest content of reinforcement material) was about 37° (Pu *et al.* 2007). Furthermore, this analysis also correlated with the EAB results, in which the composite films with the lowest contact angle exhibited the highest percentage of EAB, as discussed earlier.

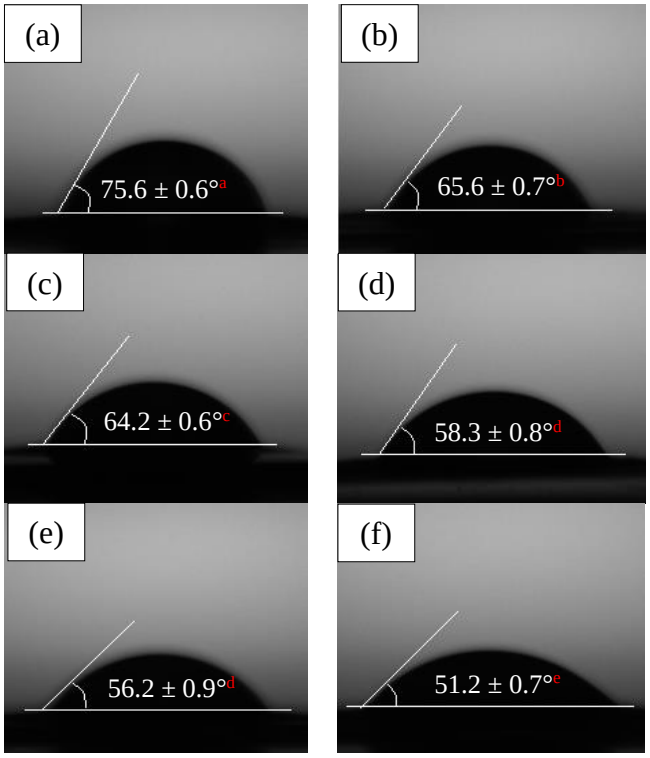


Fig. 4. Sessile drops for static water contact angle of the seaweed films with varying EFB pulp content: (a) 0%, (b) 10%, (c) 20%, (d) 30%, (e) 40%, and (f) 50% ($^{a, b, c, d, e}$ Values along each row with the same capital letter are not significantly ($p>0.05$) different as analyzed by Tukey's Test)

CONCLUSIONS

1. The incorporation of EFB pulp into the seaweed matrix had a noticeable impact on the physical, mechanical, and morphological properties of the seaweed based films.
2. The thickness and mechanical properties of the EFB pulp-seaweed composite films increased as the EFB pulp content in the films increased.
3. The contact angle of the EFB pulp-seaweed composite films decreased as the EFB pulp content in the films increased.
4. According to SEM analysis, the EFB pulp-seaweed composite films became rougher and less fiber pulled out when increased the amount of EFB pulp.
5. With the good mechanical properties and acceptable hydrophilicity, EFB pulp-seaweed composite film can be considered as a potential packaging material.

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