

Effect of seaweed on physical properties of thermoplastic sugar palm starch/agar composites

R. Jumaidin^{1,4}, S.M. Sapuan^{1,3*}, M. Jawaid⁴, M.R. Ishak,² and J. Sahari⁵

¹Department of Mechanical and Manufacturing Engineering, Universiti Putra Malaysia, 43400 UPM Serdang, Selangor, Malaysia

*Email: sapuan@upm.edu.my

Phone: +603-89471788; Fax: +603-86567122

²Department of Aerospace Engineering, Universiti Putra Malaysia, 43400 UPM Serdang, Selangor, Malaysia

³Laboratory of Biocomposite Technology, Institute of Tropical Forestry and Forest Products (INTROP), Universiti Putra Malaysia, 43400 UPM Serdang, Selangor, Malaysia

⁴Department of Structure and Material, Faculty of Mechanical Engineering, Universiti Teknikal Malaysia, Melaka, Hang Tuah Jaya, 76100 Durian Tunggal, Melaka

⁵Faculty of Science and Natural Resources, Universiti Malaysia Sabah, 88400, Kota Kinabalu, Sabah, Malaysia

ABSTRACT

The aim of this paper is to investigate the physical properties of thermoplastic sugar palm starch/agar (TPSA) blend when incorporated with seaweed. The ratio of starch, agar, and glycerol for TPSA was maintained at 70:30:30. Seaweed with various contents (10, 20, 30, and 40 wt.%) were mixed with TPSA matrix via melt mixing before compression were molded into 3 mm plate at 140°C for 10 minutes. The prepared laminates were characterized for moisture absorption, water absorption, thickness swelling, water solubility, and density. The results showed that increasing seaweed loading from 0 to 40 wt% has led to a drop in moisture content from 6.50 to 4.96% and 9% reduction of the density. TPSA matrix showed 52.5% water uptake and 32.3% swelling whereas TPSA/seaweed composites (40 wt% loading) showed 97% water uptake and 74.8% swelling respectively. Higher water solubility was also shown by TPSA/seaweed composites (57 wt%) compared to that of the TPSA matrix (26 wt%). After 16 days of storage, the equilibrium moisture content for TPSA and TPSA/seaweed (40 wt% loading) were 23.2 and 25.2% respectively. In conclusion, TPSA/seaweed composites show good environmental friendly characteristics as a renewable material. In future, the properties of this material can be further improved by hybridization with more hydrophobic fillers for better resistance against water.

Keywords: Seaweed; thermoplastic starch; agar; water absorption.

INTRODUCTION

Non-environmentally friendly petroleum based plastics have been widely used in all areas of human activity. The disposal of these materials has created serious environmental problems since they are not readily biodegradable. Therefore, intense research has been carried out to develop alternative materials that are easily disposable

but not environmentally harmful [1]. Nowadays, biopolymer derived from natural resource is getting more attention since it offers a practical solution to the accumulation of petroleum based plastic in the environment [2]. Starch is one of the most promising material for biopolymer development since it is widely available, low cost, biodegradable, renewable, and can possess thermoplastic behaviour in the presence of plasticizer [3]. However, biopolymer derived from starch is known to possess poor mechanical properties. This problem has been addressed by previous researches through various modifications such as reinforcing it with natural fibre i.e. coir, sugar palm and blending with other polymer i.e. agar [4–6]. In our previous work, incorporation of agar into thermoplastic sugar palm starch has successfully improved the mechanical properties of this biopolymer, which was also accompanied with the enhanced thermal stability [7]. Application of natural filler into polymer composites is a practical solution to enhance the properties of the composites while improving the environmental characteristics of the material as well [8–10]. Various natural fillers have been used in previous work such as snail shell, seashell, olive pit, oil palm shell, and coconut shell [11–15]. Though natural filler has been used in polymer composites, however, the hydrophobic nature of the polymer matrix used has led to poor filler-matrix compatibility of the composites [16].

Utilization of seaweed as natural filler in polymer composites has been explored in previous studies. Albano et al. [17] explored the potential of seaweed residue as fillers in high-density polyethylene (HDPE) matrix. The finding showed that the incorporation of seaweed into the polymer matrix resulted in high porosity and poor mechanical properties of the composites. Earlier study by Hassan et al. [18] showed that the incorporation of green seaweed from *Ulva lactuca* (sea lettuce) species onto polypropylene matrix has caused a drop in the tensile strength of the material. More recent study by Bulota et al. [19] explored the potential of various kind of seaweeds i.e. green, brown, and red seaweeds as fillers in poly (lactic acid) (PLA) matrix. Despite the environmental friendly characteristics of PLA, the hydrophobic characteristic of this biopolymer is not favourable to achieve compatibility with natural filler. The author reported a distinct separation between the filler and the polymer matrix accompanied by the drop in the tensile strength and the elongation of the composites. In general, it can be seen from previous studies that utilization of seaweed as natural filler in hydrophobic polymer matrix often led to negative results due to the incompatibility of the two materials.

Moreover, bio composites derived from the combination of natural filler and synthetic petroleum based polymer are still non-fully biodegradable, therefore, the environmental friendly characteristics of the bio composites are not entirely achieved. Biomass residues from agricultural wastes have shown great potential as natural fillers in polymer composites [20–22]. *Eucheuma cottonii* (also known as *Kappaphycus Alvarezii*) is a marine alga that belongs to the “red seaweed” family. The extraction of carrageenan (seaweed hydrocolloids) from this marine algae produces an enormous amount of solid wastes due to the low weight ratio (25 to 35%) of carrageenan in the raw seaweed [23]. Even though there are existing studies utilizing seaweed as natural fillers in polymer composites, it is clear from literature that there is no study utilizing *Eucheuma cottonii* wastes as natural fillers in thermoplastic starch/agar blend matrix. Therefore, the objective of this study is to utilize seaweed wastes as natural fillers for biopolymer matrix derived from thermoplastic sugar palm starch/agar blend in order to investigate the physical properties of this fully bio composite material.

MATERIALS AND METHODS

Materials

Sugar palm starch was extracted from sugar palm trees in 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 in order to dissolve the starch into the water. Water solution containing the starch was filtered in order 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 open air for 48 hours 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 Sciencechem. Seaweed wastes from *Eucheuma cottonii* species were obtained as waste materials from seaweed extraction. The solid wastes were obtained after hot alkaline extraction process to obtain carrageenan. These by-products were 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 process. The average particle size, moisture content, and water absorption capacity of the seaweed wastes were 120 μ m, 0.75 \pm 0.2% and 8.65 \pm 0.12 g g⁻¹ respectively. Figure 1 shows the micrograph of seaweed wastes.



Figure1. *Eucheuma cottonii* seaweed wastes.

Sample Preparation

Preparation of thermoplastic sugar palm starch/agar (TPSA) was conducted according to the previous work [7]. For the preparation of TPSA, the weight ratio of starch, agar, and glycerol was maintained at 70:30:30. All materials were pre-mixed using high speed mixer at 3000 rpm for 5 min. 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 40 tonnes. The same processes were used for the modification of TPSA with 10, 20, 30, and 40 wt. % of seaweeds. All samples were pre-conditioned at 53% RH for at least 2 days prior to testing. Figure 2 shows the flowchart of the composites preparation.

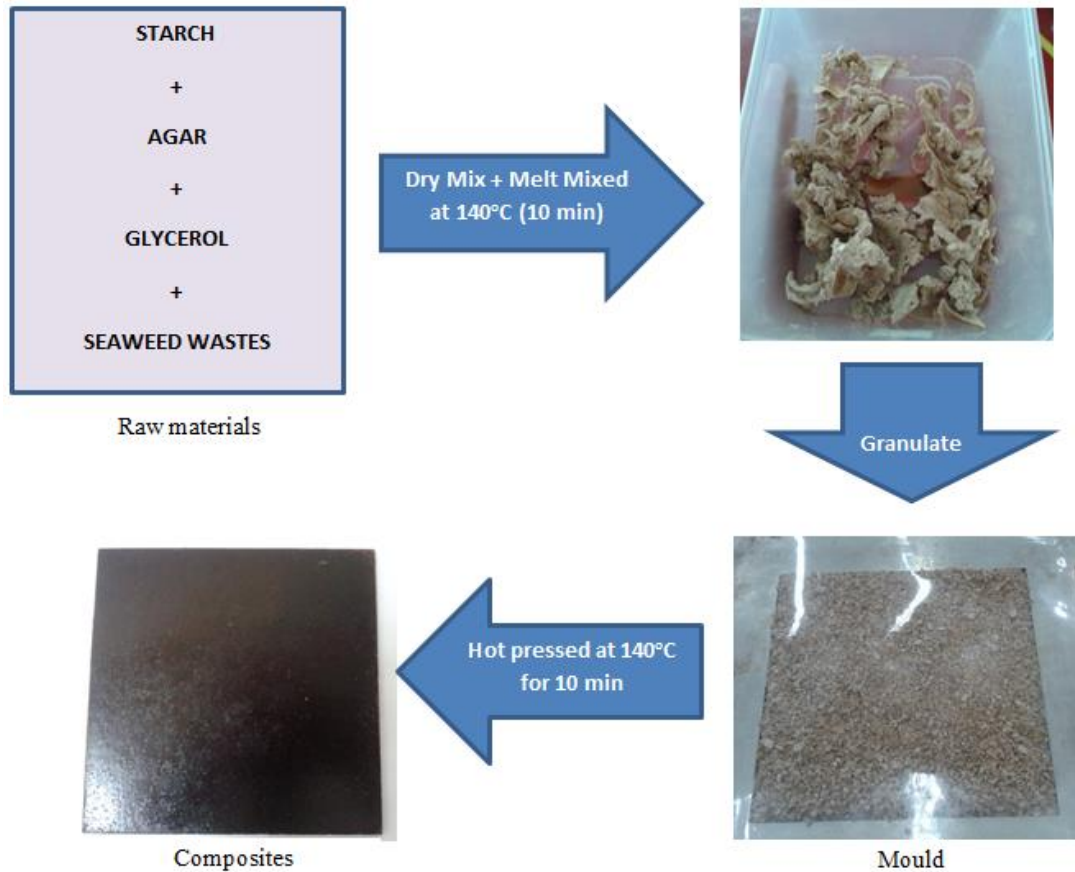


Figure 2. Flowchart for composites preparation.

Moisture Content

Moisture content of samples was determined following the previous study [24]. Samples (10 × 10 × 3 mm) were prepared for the moisture content investigation. All samples were heated in an oven for 24 h at 105 °C. Weights of samples before, M_i and after, M_f the heating were measured in order to calculate the moisture content. Moisture content was determined using the following equation:

$$\text{Moisture content (\%)} = \frac{M_i - M_f}{M_i} \times 100 \quad (1)$$

The tests were conducted in five replications and the average value was calculated.

Density

Density determination balance (XS205 Mettler Toledo) was used to measure the density of the composites. Five measurements were conducted at 27 °C and the average value was calculated.

Water Absorption

Specimens with dimensions of 10 × 10 × 3 mm were dried in an air circulating oven at 105°C±2 for 24 h in order to remove existing water and then immersed in water at room temperature (23±1 °C) for 0.5 and 2 h as proposed by previous studies [1,24]. The

samples were weighed before, W_i and after immersion, W_f and the water absorption of the laminates was calculated using Eq. (2):

$$\text{Water absorption (\%)} = \frac{W_i - W_f}{W_i} \times 100 \quad (2)$$

Thickness Swelling

To determine the percentage of thickness swelling, similar testing parameters were used as mentioned in Section 2.5. The samples were measured before, T_i and after, T_f immersion using a digital vernier (Model: Mitutoyo) and have 0.01 accuracy. The thickness swelling ratio of the laminates was calculated using Eq. (3):

$$\text{Thickness swelling (\%)} = \frac{T_i - T_f}{T_i} \times 100 \quad (3)$$

Water Solubility

Water solubility (WS) of the samples was determined according to the method by Kanmani and Rhim [25] with slight modification. For this, a piece of sample ($10 \times 10 \times 3$ mm) was cut and dried at $105^\circ\text{C} \pm 2$ for 24 h. Initial weight of samples (W_o) was measured before being immersed into 30 mL of distilled water with gentle stirring. After 24 h of immersion, the remaining piece of sample was taken from the beaker and filter paper was used to remove the remaining water on the surface. Then, the samples were dried again at $105^\circ\text{C} \pm 2$ for 24 h to determine the final weight (W_f). The WS of the sample was calculated as follows:

$$\text{Water solubility (\%)} = \frac{W_o - W_f}{W_o} \times 100 \quad (4)$$

Moisture Absorption

Samples were stored at $75 \pm 2\%$ relative humidity (RH) at a temperature of $25 \pm 2^\circ\text{C}$ in order to analyze the moisture absorption behaviour of the samples. The 75% RH was obtained by using a saturated solution of sodium chloride (NaCl) in a closed desiccator. Prior to the moisture absorption measurements, samples with the dimension of $10 \text{ mm} \times 10 \text{ mm} \times 3 \text{ mm}$ were dried at $105^\circ\text{C} \pm 2$ for 24 h. The samples were weighed before, M_i and after absorption, M_f for certain period until constant weight was obtained. The moisture absorption of the samples was calculated using the following equation:

$$\text{Moisture absorption (\%)} = \frac{M_f - M_i}{M_i} \times 100 \quad (5)$$

RESULTS AND DISCUSSION

Moisture Content

Figure 3 shows the moisture content of TPSA composites with various seaweed loadings. Increased seaweed loading from 0 to 40 wt% has led to a slight drop of moisture content from 6.50 to 4.96%. Despite the hydrophilic behaviour of seaweed, the moisture content of the composites showed the opposite trend. This effect can be

attributed to a reduction in the mobility of polysaccharide matrix following the addition of fillers which resulted in lower moisture content of the composites [26]. Moreover, this might as well be attributed to the low moisture content of seaweed ($0.75 \pm 0.2\%$) which was used in the preparation of these composites. The moisture content reported for the seaweed/TPSA composites was relatively lower than the previous work of biocomposite based on thermoplastic starch (more than 10%) [26]. On the other hand, lower moisture contents (0.75 to 1.35%) of thermoplastic starch based composites were also reported in previous work [27].

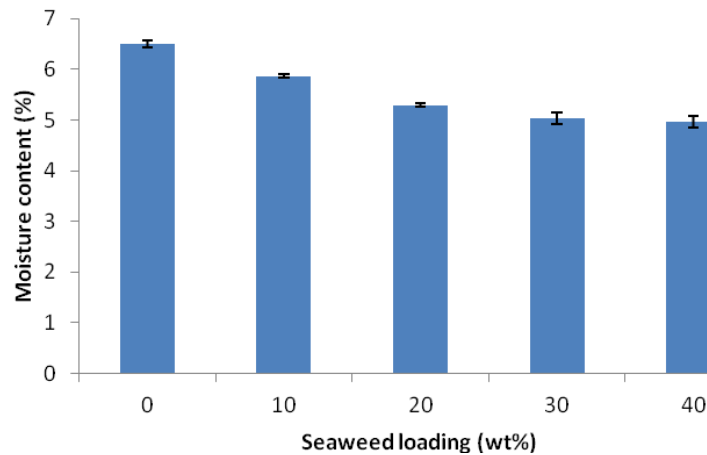


Figure 3. Moisture content of TPSA/seaweed composite.

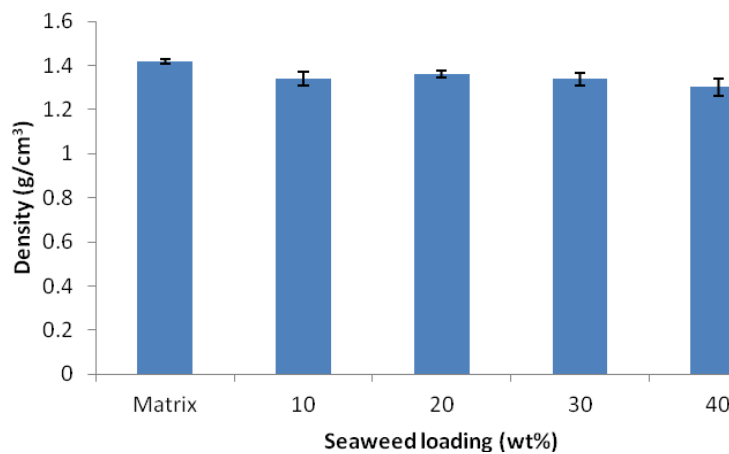


Figure 4. Density of TPSA/seaweed composite.

Density

Reducing the weight of material is one of the primary reasons for composite fabrication. Lightweight material is often desirable due to its easy handling which might aid in improving the performance of the end product as well as reducing the transportation costs. Density of seaweed composite was shown in Figure 4. In general, it can be seen that incorporation of seaweed into TPSA matrix led to a decrease in the density of the composite. At 10 wt% of seaweed loading, the density of composite was reduced by 2.8%. Further incorporation of seaweed at 40 wt% led to 9% reduction of the density. This might be attributed to the formation of voids following the incorporation of fillers into the matrix. Ibrahim et al. [28] reported a decline in the density of composites

following the addition of date palm fibre into thermoplastic starch (TPS) matrix, which was attributed to the formation of voids in the composites.

Water Absorption

Figure 5 shows the water absorption percentage of TPSA incorporated with different amount of seaweeds. It can be seen that after 0.5h of immersion, TPSA showed 26.9% water uptake while TPSA composite with 40 wt. % seaweed showed an increment of water uptake at 54.1%. It was apparent that water uptake of all materials increased with longer immersion time. TPSA and TPSA/seaweed composites continued to show gradual increment of water uptake with addition of seaweed after 2h of soaking in distilled water. TPSA showed 52.5% of water uptake while TPSA composite with 40 wt. % seaweed showed 97% of water uptake.

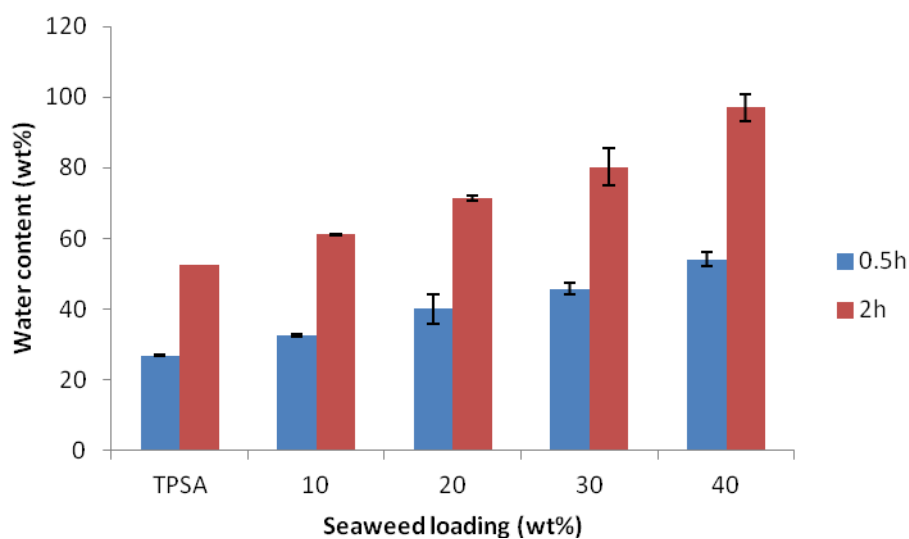


Figure 5. Water absorption of TPSA/seaweed composites.

In general, it can be seen that the incorporation of seaweed has increased the water absorption capacity of the composites. This effect can be assigned to the hydrophilic character of seaweed that facilitates the diffusion of the water molecules within the material. Hassan et al., also reported an increment of water uptake when introducing seaweed as fillers in polypropylene matrix [18]. Moreover, the presence of residual carrageenan inside the seaweed might also contribute to this phenomenon since the seaweed hydrocolloids are known to possess high water absorption capacity [29]. Similar findings were also reported for incorporation of other natural fibres i.e. kenaf, jute, and oil palm fibre into the polymer matrix [30,31]. After 3h of soaking, the composites with higher filler loading (30 and 40 wt. %) began to disintegrate, which prevented accurate measurement of the water uptake. This phenomenon might be attributed to higher amount of seaweeds inside the matrix that led to excessive swelling and eventually weakened the filler-matrix bonding of the composites.

Thickness Swelling

The swelling characteristics of TPSA/seaweed composites were investigated using the swelling ratio in order to investigate the effect of seaweed on the dimensional stability of the composites. Figure 6 shows the swelling percentage of TPSA and the composites with various seaweed loadings. It was obvious that the thickness of TPSA and the

composites was affected by both immersion time and the filler loadings. TPSA showed the lowest swelling, while this increased with increasing amount of seaweed in the composites. The difference in swelling percentage between the composites was more evident after 2h of immersion, where incorporation of seaweed from 0 to 40 wt. % showed an increase in swelling from 32.3 to 74.8% respectively. This effect can be attributed to the nature of seaweed that preserves water in order to maintain the structure of the branch. The preserved water was removed from the structure of seaweed during the process that involved various drying stages from sun drying to oven drying. Therefore, seaweed has higher tendency to regain the water loss in the structure during the immersion which eventually led to swelling of the composites. Yahaya et al., [31] also reported an increase in the thickness swelling of the composites when kenaf fibre was introduced to the polymer matrix.

In general, similar increasing trend was observed for water absorption in the previous section which indicates that swelling characteristics of the composites are highly dependent on the amount of water absorbed. This finding is in good agreement with previous study on thermoplastic starch/coir fibre composites which reported similar situation [1]. According to Jawaid et al. [30] the hydrophilic properties of materials and the capillary action will cause water absorption during immersion, and thus increase the dimension of the composites.

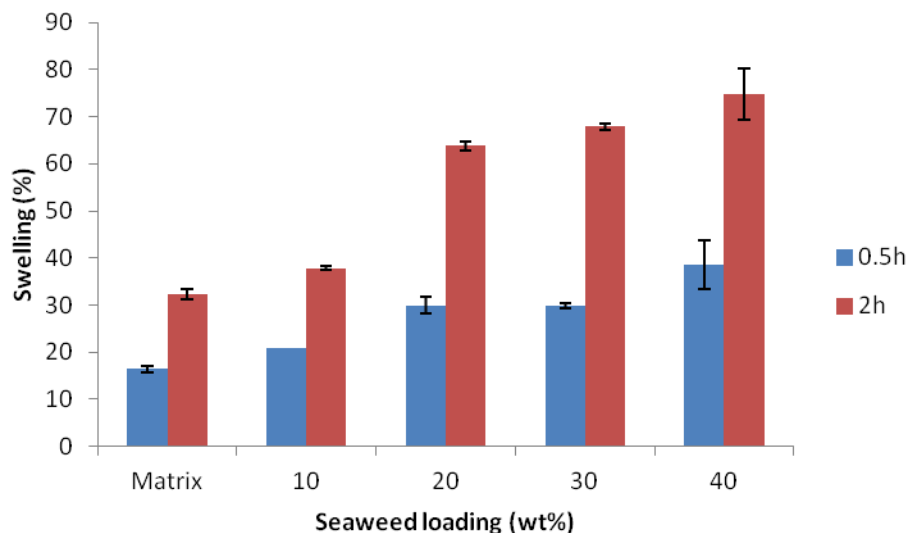


Figure 6. Thickness swelling of TPSA/seaweed composite.

Water Solubility

Disposal of waste material on water often creates serious problem to the ecosystem due to the non-biodegradable characteristics of the material. One leading advantage of bio – based material is the readiness to decompose when disposed in water. Water solubility shows the percentage of weight loss of a material when disposed in the water. Figure 7 shows the water solubility of TPSA and the composites with various seaweed loadings. It can be seen that the incorporation of seaweed into TPSA has increased the solubility of the composites. TPSA/seaweed with 40 wt. % seaweed shows 57% solubility whereas TPSA matrix shows only 26% of solubility. Again, this effect can be attributed to the hydrophilic nature of seaweed that tends to absorb more water, which leads to swelling and disintegration to take place. The residual carrageenan in the seaweed might as well contribute to this behaviour. According to Flores et al. [27] carrageenan is more soluble in water than neutral hydrocolloids i.e. starch because the negatively charged

sulphate groups are more hydrophilic. Since bio-based material is mainly designed for short life products, therefore, improvement in water solubility of TPSA when incorporated with seaweed gives more positive attributes to this biomaterial in terms of the environmental friendly characteristics. Similar findings were reported by Flores et al. [27] on the increase in water solubility of thermoplastic cassava starch following the incorporation of carrageenan in the matrix. Nevertheless, it should be noted that higher water solubility also indicates weak resistance of material when exposed to water, therefore, increased amount of seaweed might as well be associated in weakening the matrix structure upon contact with water.

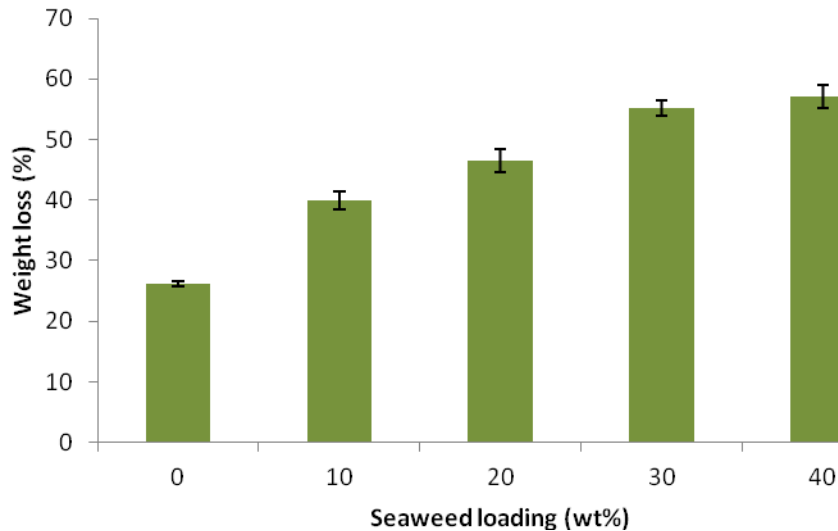


Figure 7. Solubility of seaweed/TPSA composites.

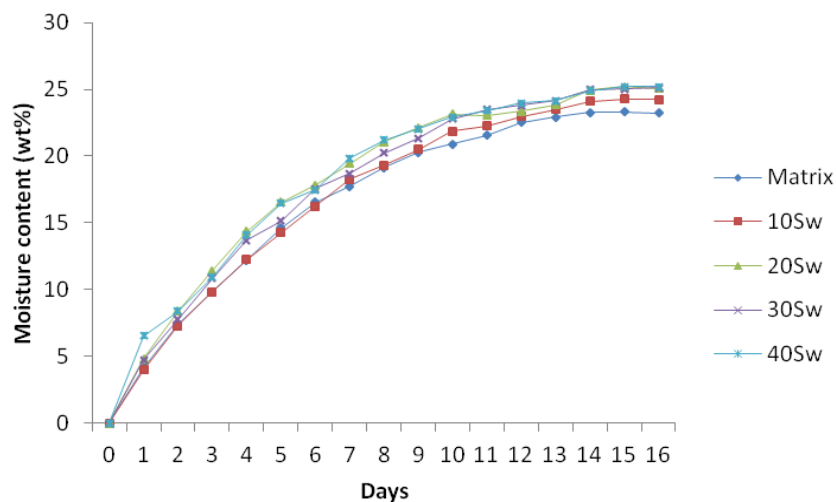


Figure 8. Moisture absorption curves of seaweed/TPSA composites.

Moisture Absorption

Figure 8 shows the moisture absorption of seaweed composites during 16 days of storage at $75 \pm 2\%$ RH at a temperature of 25 ± 2 °C. In general, all composites showed similar increasing trend for moisture content with increased storage time. It can be noted that the moisture sorption of the composites was more rapid at the initial stages and became slower as the storage time increased. More stable moisture sorption curve of the

composites can be seen after 14 days of storage. This is because after 14 days, the moisture content of the composites began to achieve equilibrium with the surrounding. Similar finding was reported for coir fibre reinforced thermoplastic starch composites where the moisture absorption became stable after 14 days of storage [1]. The effect of seaweed incorporation into TPSA matrix can be noted by a higher moisture content shown by the composites when compared to the matrix. After 16 days of storage, the incorporation of fillers from 0 to 40wt% has led to the increase in the equilibrium of moisture content from 23.2 to 25.2%. This finding is in agreement with the water absorption behaviour shown by the composites. Again, this effect can be ascribed to the more hydrophilic nature of seaweed than the matrix.

CONCLUSIONS

Bio composites derived from seaweed and TPSA blend have been successfully produced in this study. The combination of this material has led to variations in their physical properties. Increasing the addition of seaweed from 0 to 40 wt. % resulted in (i) a decrease in moisture content from 6.50 to 4.96% (ii) a decrease in density from 1.42 to 1.30 g/cm³ (iii) an increase in water absorption from 52.5 to 97% (iv) an increase in thickness swelling from 32.3 to 74.8%, (v) an increase in water solubility from 26.2 to 57%, and (vi) an increase in moisture absorption from 23.2 to 25.2%. In conclusion, the bio composites prepared in this work shows great potential as a renewable material that possesses good environmental friendly characteristics. However, the composites prepared also show weak water resistance which could affect the performance of the final product. Therefore, hybridization of seaweed with more hydrophobic filler is a highly potential research to be explored in the near future.

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