

# Water absorption and water solubility properties of sago starch biopolymer composite films filled with sugar palm particles

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**Abstract:** The influence of addition of 10, 20, 30, 40 and 50 wt % sugar palm particles (SPP) on the water absorption properties of thermoplastic sago starch biopolymer composite films was investigated. The fillers were mechanically stirred with thermoplastic sago starch mixtures for 30 minutes at 80 °C. The prepared films were then characterized for water absorption and water solubility. The SPP successfully reduce water absorption and thus increase barrier properties of thermoplastic sago starch biopolymer composite against water penetration, resulting in a more durable biocomposite films.

**Keywords:** thermoplastic sago starch, biopolymer composite films, water absorption, sugar palm particles.

## Absorpcja wody i rozpuszczalność w wodzie kompozytowych folii z biopolimeru skrobiowego sago napełnionego cząstkami włókien palmy cukrowej

**Streszczenie:** Zbadano wpływ dodatku 10, 20, 30, 40 i 50% mas. cząstek włókien palmy cukrowej (SPP) na absorpcję wody termoplastycznych folii z biopolimeru skrobiowego sago. Cząstki napełniacza rozprowadzono mechanicznie w mieszaninie termoplastycznej skrobi sago przez 30 minut w 80 °C. Wyznaczano absorpcję wody i rozpuszczalność w wodzie otrzymanych folii. Cząstki SPP skutecznie zmniejszają wchłanianie wody, zwiększając tym samym właściwości barierowe termoplastycznego kompozytu skrobi sago względem wody, co umożliwia uzyskanie trwalszych folii biokompozytowych.

**Słowa kluczowe:** termoplastyczna skrobia sago, biopolimerowe folie kompozytowe, absorpcja wody, cząstki cukru palmowego.

Sustainability has now becoming one of the key factors for developing new products. It was heard for the very first time during United Nations Conference on Sustainable Development which took place in Rio de Janeiro, Brazil and from then on, it has been increasingly taken seriously by many countries around the globe, including Malaysia. The government amended Solid Waste and Public Cleansing Management Act in 2009 where according to subsection 73 (4), the Director General may exempt collection of recyclable solid waste carried out by charity groups or any other organizations from provision of subsection (2) [1]. According to National Solid

Waste Management Department of Malaysia, this law provision aims to encourage plastic recycling behavior among the public as plastic waste ranks at number two with the second highest percentage in country's waste composition [2].

The rate of plastic waste production is far exceeding the rate of plastic degradation thus causing imbalance in the biomes. As a result, it causes water-borne diseases due to water contamination as leachate produced from the plastic waste seeps into water reserve area. Therefore, there is an absolute urgency to replace conventional petroleum based plastic with bioplastic to provide simplified end of life disposal and consequently reduced dependency on fossil fuel resources [3–11].

Bioplastic such as Mater-Bi<sup>TM</sup>, Bioska<sup>TM</sup>, Biotec<sup>TM</sup> and Potato Pak<sup>TM</sup> utilize starch as an environmentally friendly raw material, which has short term durability and can be easily and rapidly degraded [12, 13]. There are many sources of starch that can be commercialized for the production of bioplastic. However, compared to other sources of starch, sago starch is the highest contributor in terms

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**Table 1. Chemical composition of sugar palm fiber [15]**

Samples	Cellulose, %	Hemicellulose, %	Holocellulose, %	Lignin, %	Extractive, %	Ash, %
Sugar palm fibers	43.88	7.24	51.12	33.24	2.73	1.01

of productivity yield. It is estimated that around 3000 kg of sago starch can be obtained annually from one hectare of cultivation area compared to 2000 kg of potato starch and 1000 kg of maize. In Malaysia, sago starch cultivation is widely practiced in Sarawak (Eastern Malaysia) where the activity covers 12% of the land area [14].

However, sago starch has some drawback, being naturally highly hydrophilic material. Therefore, incorporation of natural fiber into sago starch matrix is a promising attempt to improve the water sensitivity of starch-based system while preserving the biodegradability of the composite [9, 15–21]. One of natural fibers available locally is sugar palm fiber which grows along the rivers and bushes in rural areas of Malaysia such as Kuala Pilah, Bruas, Raub and Jasin. It is also found in many ASEAN countries such as Indonesia, Thailand, Philippines and Vietnam, besides Taiwan and India [22]. The tree is still under-utilized and to the best of our knowledge, there is no reports on the use of sugar palm fiber as a reinforcing material in the sago starch matrix. In this study, sugar palm particles were used as a filler in thermoplastic sago starch matrix and water absorption as well as solubility of the blends were characterized. Thus, this study aims to determine the effect of addition of sugar palm particles (SPP) on water absorption properties of thermoplastic sago starch biopolymer composite films with 10, 20, 30, 40 and 50 wt % SPP.

## EXPERIMENTAL PART

### Materials

Sugar palm fiber was obtained from Kampung Kendong Lama, Kota, Negeri Sembilan, Malaysia which is located 105 km from Kuala Lumpur. The chemical compositions of the sugar palm fibers are shown in Table 1 [15]. Sorbitol (Table 2) and glycerol (Table 3) used as plasticizers were provided by Sue Evergreen Sdn Bhd (Semenyih, Malaysia).

Fibers were harvested using knives and washed to remove dirt from them. After that, fibers were cut into an inch length in a cutting mill before being ground and filtered using a vibratory screener into four different lengths namely greater than 2 mm, between 1 to 2 mm, between 0.5 to 1 mm and less than 0.5 mm. The fibers were then again filtered using a 100 mesh sieve to obtain a fine fiber fraction of less than 0.15 mm in length.

The starch used for this research was commercial sago starch, supplied by OXL Resources Sdn Bhd (Kuala Lumpur, Malaysia), which contains 85.5 g carbohydrate, 0.2 g protein and 3 mg of sodium for every 100 g of sago starch (Table 4).

### Fabrication of the biocomposite films

The blend of sago starch with sugar palm particles was prepared by dissolving 5 g sago starch into 100 cm<sup>3</sup> of distilled water together with 30% plasticizers (glycerol and sorbitol in the ratio of 1 : 1) The suspension was heated at 85 °C for 30 minutes in a water bath under constant stirring (1360 rpm) using a mechanical stirrer. The plasticized solution was then poured into a square mould (30 × 23 × 4.5 cm) and dried in an oven at 50 °C for 24 hours. This procedure was repeated by incorporating 10, 20, 30, 40 and 50 wt % sugar palm particles to the suspension. Composites were labelled as 10SPP, 20SPP,

**Table 2. Sorbitol specification**

Properties	Specification
Assay (HPLC), %	> 99.0
Heavy metals (as Pb), %	< 0.001
Mannitol (HPLC), %	< 0.2
Water, %	< 0.2

**Table 3. Glycerol specification**

Properties	Specification
Assay, %	99.8
Density 20/4°, g	1.257–1.262
Refractive index, –	1.471–1.473
pH, –	6.0–7.0
Sulphate ash, %	Max. 0.005
Chloride (Cl), %	Max 0.0001
Sulphate (SO <sub>4</sub> ), %	Max 0.0005
Ammonium (NH <sub>4</sub> ), %	Max 0.0005
Arsenic (As), %	Max 0.0004
Copper (Cu), %	Max 0.0005
Iron (Fe), %	Max 0.0005
Lead (Pb), %	Max 0.0005
Sugar (glucose), %	Max 0.0004

**Table 4. Sago starch specification**

Properties	Specification
Density, g/cm <sup>3</sup>	1.5
Ash, %	0.2
Amylose, %	24–27
Protein, g	0.2
Fat, %	0.24
Sodium, mg	3
Carbohydrates, g	85.5
Water content, %	10–20

30SPP, 40SPP and 50SPP, respectively, using one control film.

## Methods of testing

### Fourier transform infrared spectroscopy

Infrared spectra of the materials were obtained using an IR spectrometer (Nicolet 6700 AEM). The spectra were scanned 32 times for spectrum integration in the wavenumber range of  $4000\text{ cm}^{-1}$  to  $400\text{ cm}^{-1}$  using KBr for two samples, namely 10SPP and control film, to investigate the effect of sugar palm particles loading in the blends. The scanning resolution was  $4\text{ cm}^{-1}$ .

### Water absorption

Specimens with dimensions of  $10 \times 10 \times 3\text{ mm}$  were weighed and next dried in an air circulating oven at  $105 \pm 2\text{ }^\circ\text{C}$  for 24 h in order to remove existing moisture and then immersed in water at room temperature ( $23 \pm 1\text{ }^\circ\text{C}$ ) for 24 h. After that, the samples were weighed again. This test was repeated 10 times using another samples of the same formulation and the weight gain was recorded for 2, 4, 6, 8, 10 and 24 hours. The percentage of water absorption was calculated using the following equation, where final and initial weights are denoted by  $w_f$  and  $w_i$ , respectively.

$$\text{Water absorption} = \frac{w_f - w_i}{w_i} \cdot 100\% \quad (1)$$

### Water solubility

Before the test, the samples were cut and dried at  $105 \pm 2\text{ }^\circ\text{C}$  to determine their initial weights. Water solubility of thermoplastic sago starch blended with sugar palm particles was determined by immersing  $10 \times 10 \times 3\text{ mm}$  samples in  $30\text{ cm}^3$  distilled water.

The samples were stirred gently from time to time. After 24 h immersion the samples were taken from the beaker and filter paper was used to remove the remaining water from the surface. Then, the samples were dried at  $105 \pm 2\text{ }^\circ\text{C}$  for 24 h to determine their final weight. The experiment was repeated 10 times. The percentage of water solubility was calculated using the equation (1).

## RESULTS AND DISCUSSION

### Results

Figure 1 shows unfilled thermoplastic sago starch (control film), sugar palm particles with size lower than  $0.15\text{ mm}$  and thermoplastic sago starch blended with 50 wt % sugar palm particles.

Visual observation shows that sugar palm particles are dispersed homogeneously inside sago starch matrix with

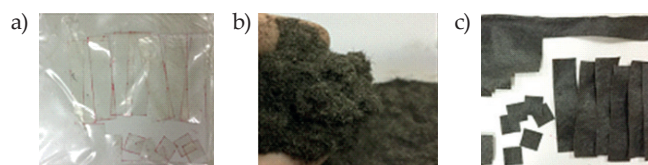


Fig. 1. a) Thermoplastic sago starch without sugar palm particles (control film), b) sugar palm particles, c) thermoplastic sago starch blend with 50 wt % sugar palm particles

no voids, cracks and apparent phase separation. This suggests that both the matrix and the reinforcement have good compatibility, thus the wettability between sugar palm particles and sago starch matrix is possibly high. This observation is supported by various experiments conducted by Sahari *et al.* [23], Sanyang *et al.* [24], and Ilyas *et al.* [3] in which good compatibility between reinforcing fibers and biopolymer matrices can be indicated by surface morphology analysis showing no phase separation between these components. Moreover, blending of sugar palm particles into sago starch matrix also decreases film flexibility defined as the ability of material to be bent. This happens due to the nature of sugar palm particles which is more rigid compared to the sago starch matrix [25].

### FT-IR

Infrared spectroscopy is used to detect the chemical changes after the addition of sugar palm particles into the thermoplastic sago starch system. Figure 2 shows the results for control film and sago starch blended with 10 wt % sugar palm particles where both spectra are dominated by the peaks at  $3441\text{--}3446\text{ cm}^{-1}$  corresponding to hydroxyl group (-OH) in sago starch for control film and sago starch and cellulose for blended film [3, 26]. An increase in hydroxyl peak intensity is detected after introducing sugar palm particles into the thermoplastic sago starch. Also, the absorption peaks characteristic for cellulose can be found at wavenumbers  $897$ ,  $1160$ ,  $1316$ ,  $1370$  and  $1424\text{ cm}^{-1}$ , whereas the band of aromatic skeletal vibration ( $\text{C}=\text{C}$ ) of lignin and lignocellulose can be found

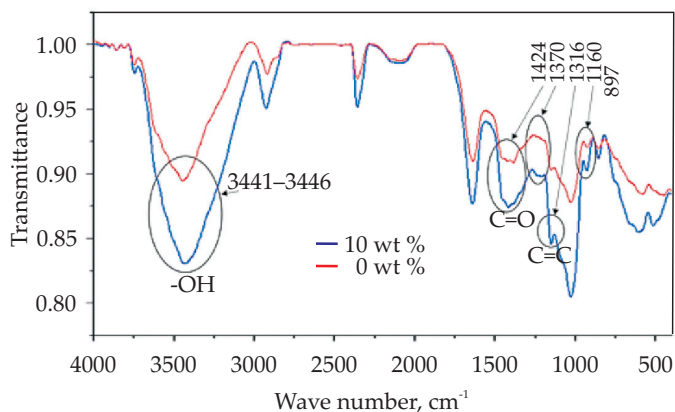


Fig. 2. FT-IR spectra for control film (above) and sago starch blend with 10 wt % sugar palm particles

at wavelength of 1510–1520  $\text{cm}^{-1}$  [16]. When FT-IR results are compared with water absorption and solubility, it can be seen that even though the amount of hydroxyl group increases in the system due to addition of sugar palm particles, this factor solely does not increase the film water absorption and solubility as both properties show reduction. This suggests that there are other factors that are more dominant in influencing these properties as discussed below.

### Water absorption

Figure 3 shows water absorption readings for control film, 10SPP, 20SPP, 30SPP, 40SPP and 50SPP after being immersed for 24 hours.

The downward trend is observed for water absorption with control film showing the highest water uptake over 120% and the lowest value about 40% for 50SPP film.

Table 5 shows the correlation result as sugar palm particles loading is varied.

Correlation analysis was applied to the data and a strong relationship can be seen with  $r = -0.966$ . This negative correlation suggests that higher sugar palm particles loading resulted in a lower water uptake. Coefficient of determination,  $R^2 = 0.932$ , means that 93.2% of the water absorption data can be explained by the amount of sugar palm particles loadings. In statistics, linear regression is a linear approach to modelling the relationship between a scalar response (or dependent variable which is 24 hours water solubility period) and one or more explanatory variables (or independent variable which is sugar palm particles loading varied from 0 wt % to 50 wt %). In particular, the purpose of linear regression is to “predict” the value of the dependent variable based upon the values of one or more independent variables. Results show a negative correlation between water absorption and sugar palm particles loading where the higher the loading, the lower the water uptake. During the test, sago starch is probably more soluble in water compared to the sugar palm as the hydrophilicity of the former is higher than that of the later. Moreover, as sago starch is in amorphous form, it is easier for water molecules to pen-

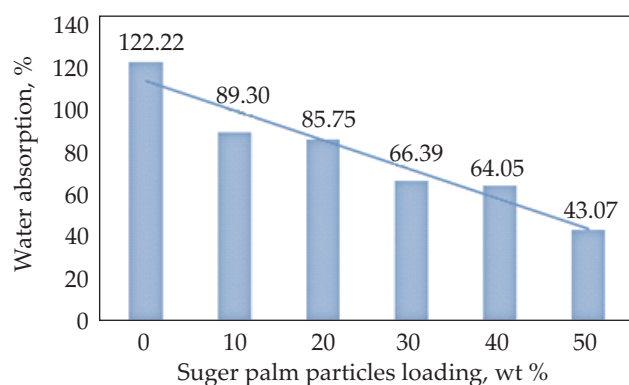


Fig. 3. Water absorption of composites as a function of sugar palm particles loading (0–50 wt %)

Table 5. Correlation between water absorption and sugar palm particles loading

Correlation matrix (Pearson)	
Variables	% sugar palm ash loadings
Water absorption (24 hours), %	$r = -0.966$ ( $p = 0.002$ )

Table 6. Analysis of variance (ANOVA) for water absorption as a function of sugar palm particles loading

Source	Mean squares	F	Pr > F
Samples of 2 hours	2324.107	4.096	0.113
Samples of 4 hours	1387.057	10.983	0.030
Samples of 6 hours	2480.609	15.036	0.018
Samples of 8 hours	7303.885	12.239	0.025
Samples of 10 hours	3672.235	21.435	0.010
Samples of 24 hours	2637.666	16.605	0.015

Computed against model  $Y = \text{mean}(Y)$ ,  $Pr > F$  – the significance probability value associated with the  $F$  value,  $F$  value – the ratio produced by dividing the mean square for the model by the mean square for error.

etrate and dissolve out the polymer. Fortunately, the introduction of sugar palm particles in the thermoplastic sago starch films restricts the movement of sago starch molecules, thus limiting the swelling of starch chains in the system [27]. This helps to reduce the amount of sago starch polymer dissolved into the water. Table 6 shows results of the analysis of variance (ANOVA) for each water absorption reading as sugar palm particles loading is varied. ANOVA analysis showed that SPP loadings of 10, 20, 30, 40 and 50 wt % are significantly different from each other, with  $p = 0.02$ .

Water absorption readings at 2, 4, 6, 8, 10 and 24 hours provide information on the trends for each of these time intervals (Fig. 4).

In the first 8 hours, all samples show a decrease in water uptake, then suddenly increase in water absorption occurs for 10 and 24 hours' readings. Coefficient of determination for each sample is  $R^2 = 0.506, 0.733, 0.790, 0.754, 0.843$  and  $0.806$  for control film, 10SPP, 20SPP, 30SPP, 40SPP and 50SPP, respectively. ANOVA results show that all samples are significantly different from each other at 4, 6, 8, 10 and 24 hour readings. In contrast, ANOVA shows no significant difference of SPP loading for the first 2 hours, which means that during this period almost all samples absorb similar amounts of water regardless of sugar palm particles loading. But then, Fig. 4 reveals that the control film actually absorbs more water compared to other blended films during the first 2 hours of immersion.



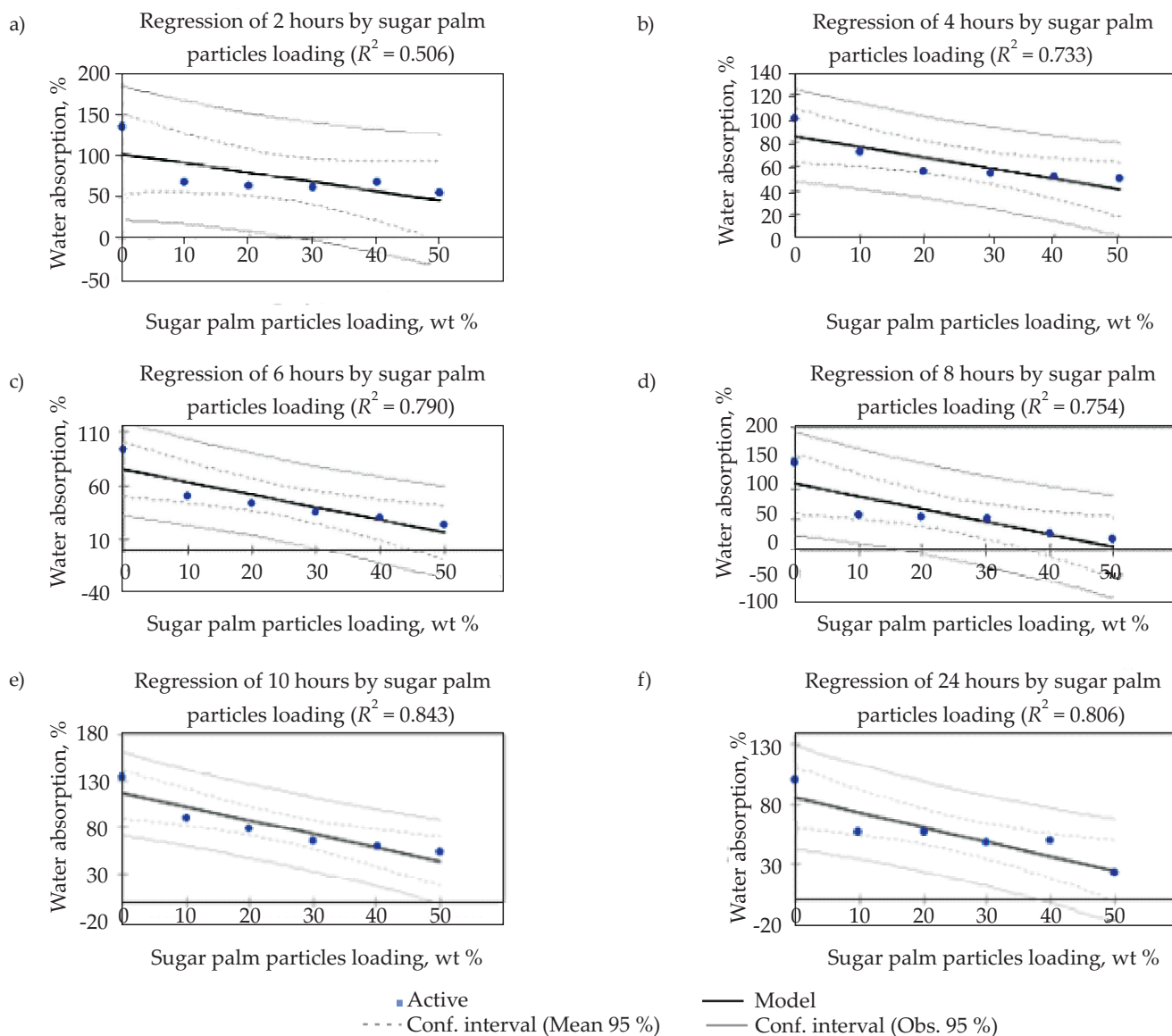


Fig. 4. Water absorption after 2, 4, 6, 8, 10 and 24 hours of immersion as a function of sugar palm particles loading (0–50 wt %)

Agglomerative hierarchical cluster analysis (HACA) is used to establish a set of clusters in which the objects in the same cluster are similar to each other, thus, provides intuitive similarity of the relationship between the sample and the entire dataset. Water absorption periods of 2, 4, 6, 8, 10 and 24 hours are classified by unique nature assigned to them. Temporal variation shows that the data can be grouped into three classes where 2 and the 10 hours both are classified as first class which represents the highest water absorption; while the 4, 6 and 24 hours immersion are grouped in the second class, which represents moderate water absorption. On the other hand, the 8 hours immersion is categorized as the third class which denotes the lowest water absorption. During the 8 hours immersion, all samples show minimal hydration before bounce back at the 10 hours immersion. Table 7 shows classification of water absorption according to hours of immersion.

Table 7. Classification of water absorption according to hours of immersion

Observation	Class
2 hours	1
4 hours	2
6 hours	2
8 hours	3
10 hours	1
24 hours	2

#### Water solubility

Table 8, Table 9 and Figure 5 show the ANOVA results for water solubility, the correlation results for water solubility and regression of water solubility for 24 hours, re-

**Table 8.** ANOVA for water solubility as a function of sugar palm particles loading (0–50 wt %)

Source	DF	Sum of squares	Mean squares	F	Pr > F
Model	1	310.398	310.398	12.983	0.023
Error	4	95.633	23.908		
Corrected					
Total	5	406.031			

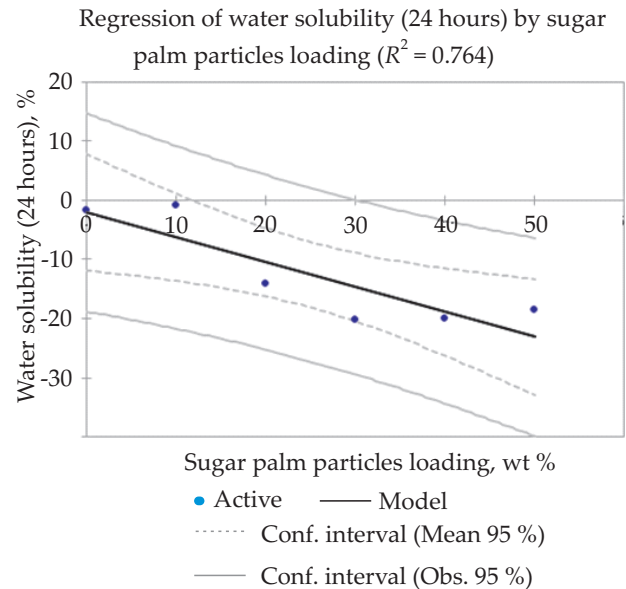
Computed against model  $Y = \text{mean}(Y)$ ,  $DF$  – degrees of freedom.

spectively, as sugar palm particles loading varies from 0 to 50 wt %. The results show that higher sugar palm particles loading results in a lower water solubility for blend films with  $R^2 = 0.764$ . Moreover, ANOVA analysis shows that different SPP loadings has significantly different effect on water solubility property,  $p = 0.023$ , while correlation analysis indicates a strong negative relationship between SPP loading to water solubility with  $r = -0.874$ , which means the higher filler is loaded, the lower water solubility property for the films is observed.

## Discussion

### Water absorption

The results show that water absorption decreases with an increase in sugar palm particles loading. As it can be seen in Fig. 3, increasing the sugar palm particles loading to 50% reduces 2.48 times water absorption during 2 hours of immersion. Sago starch blended with 10 wt % sugar palm particles is capable to reduce water uptake only 2.06 times compared to control film. Further readings show similar trend when sago starch blended with 50 wt % sugar palm particles lowers the water absorption as much as 1.99 times in contrast to 1.44 times for sago starch blended with 10 wt % sugar palm particles. Other readings also exhibit the trend of water absorption that lowers as sugar palm particles loading increases, for the 6, 8, 10 and 24 hours of immersion. The decline in water uptake is most probably due to a good compatibility between the two phases; sago starch matrix and sugar palm particles filler, where visual observation confirms homogeneous appearance. The homogeneity in sugar palm particles distribution means that the ash is well wetted by sago starch matrix. However, for 40 wt % and 50 wt % sugar palm particles loading, excessive filler that does not bind with sago starch matrix accumulates at the bottom of the blend films. On the other hand, there is no void or crack visible in the film, which is probably due to the size of the sugar filler; as it is very fine and smaller than 0.15 mm. The absence of voids may lower water penetration as it is one of the indicators for a good interaction between both components. This is due to the fact that previous studies have found that good interaction between starch and natural fiber has lowered the void formation and decreased water uptake of the composites from 1.27% for 10 wt % flax fiber reinforced starch to 0.97%, as flax fiber content increased to 30 wt % [12].

**Fig. 5.** Water solubility for 24 hours as a function of sugar palm particles loading (0–50 wt %)**Table 9.** Correlation between water solubility and sugar palm particles loading (0–50 wt %)

Correlation matrix (Pearson)	
Variables	% sugar palm ash
Water solubility (24 hours), %	$r = -0.874$ ( $p = 0.023$ )

Water absorption at the first 2 hours shows reduction from 234.61% for the control film to 65.15% as 10 wt % sugar palm particles are added, to 61.35% for 20 wt % sugar palm particles added, to 59.1% at 30 wt % sugar palm particles loading and to 65.26% and 54.31% for 40 wt % and 50 wt % sugar palm particles loadings, respectively. This may be due to the fact that sugar palm particles are less hydrophilic compared to sago starch, which lowers water affinity of the blend films as the loading increases. This is in agreement with work done by Vallejos *et al.* [27]. In fact, the study on the moisture content of *Alfa* cellulose fiber reinforced starch also shows a downward trend in water absorption from 2.62% for the control film to only 2.37% for film with 20 wt % fiber loading [28]. Even though the addition of fiber lowers the water uptake of the starch film, it still insufficiently contributes to the hydrophobicity of the system [29].

In addition, a reduction in water absorption as sugar palm particles loading increases is probably contributed by an increase in system crystallinity. Girones *et al.* report

that the addition of fibers results in an appearance of new signals in X-ray diffraction pattern at  $2\theta \sim 16.6^\circ$  and  $22.5^\circ$  corresponding to the crystalline portion of cellulose [30]. As water permeation occurs mostly in amorphous region which is sago starch matrix, crystallinity introduced by sugar palm particles may reduce the water absorption properties. According to Donhowe and Fennema [31] the higher the ratio of crystalline/amorphous in the film, the lower the water permeability.

On the other hand, visual observation shows white blooming on some of the samples after long immersion in water. The white blooming phenomenon is also found in starch film plasticized with 60 wt % sorbitol when high moisture content favours a movement of starch macromolecules, thus causing physical exclusion of sorbitol plasticizer [32]. The separated sorbitol is then crystallized. This phenomenon occurs mainly in sorbitol-plasticized starch films rather than those containing glycerol, fructose and mannose. In sago starch films blended with sugar palm particles, combination of both sorbitol and glycerol in a ratio of 1 : 1 was applied. This combination reduced the amount of sorbitol plasticizer in the films thus reducing the white blooming. In addition, this technique also decreased glycerol content in the system so that water uptake was reduced. Mali *et al.* [33] report that glycerol-plasticized film absorbs moisture faster and in higher amounts regardless of relative humidity.

During this research, the amount of plasticizer is constant at 30 wt % with respect to dry starch content, which according to literature exceeds compatibility limit of plasticized starch films, namely 20 wt % [34]. The excess of plasticizer that mechanically trapped in the system without strong chemical bond forms a separate phase, as water molecules prefer to form bonds with free plasticizer rather than with starch or fibers. Free plasticizers are small molecules that have high affinity towards water and the fact that they are not bound to the primary hydroxyl of starch make them available for hydrogen bonding. The phase separation occurs only in control film with no filler loading. For the blend films, the excess of plasticizer is likely absorbed by sugar palm particles, therefore, preventing phase separation. In addition, sugar palm particles form a network with starch polymer thus reducing the movement of starch molecules in the system and decreasing the water absorption. The relative influence of these two mechanisms is difficult to determine because absorption of excess plasticizer and starch-fiber network formation happen at the same time [27]. Furthermore, the addition of sugar palm particles may also prevent water molecules from moving downwards following the gravity, thus forced them to move laterally due to resistance provided by the ash. This may also contribute to the reduction of water absorption as sugar palm particles loading is increased.

The absorption of water molecules is linked to the quantity of hydroxyl groups in the molecule [35–37]. Therefore, an increased hydroxyl content as shown by FT-IR analysis in Fig. 2 was initially thought to increase

water absorption as it is proportional to the film hydrophilicity. However, the addition of sugar palm particles which has hydrophobic component inside (lignin) successfully limits water penetration. The control film has a higher water absorption compared to blend films probably due to lower energy barrier for water permeation. Besides, free plasticizers that exist in control film increase affinity toward water molecules, where it helps the absorption of water into the structure of the compact control film [38]. According to Bertuzzi *et al.* [39] when water vapor is brought in contact with the film surface, the most active sites are those first sorbed by water followed by those less active until the film is saturated.

Previous study reveals that starch reinforced with lignocellulosic fiber reaches water equilibrium at different time duration. Water equilibrium is achieved when the percentage of water absorption becomes constant with time. Prachayawarakorn *et al.* report that equilibrium is reached after 3 days when thermoplastic rice starch mixed with 10 wt % cotton fiber shows water absorption reduced by 1.25 times compared to control film [40]. On the other hand, Vallejos *et al.* [27] report that water equilibrium for the composite of starch and bagasse takes a longer duration, which is 12 days. However, as the scope of this experiment is only to record the trend for water absorption when the films are immersed in water for 24 hours, the observation does not extend until water equilibrium is achieved.

#### Agglomerative hierarchical cluster analysis (HACA)

Table 7, which classifies water absorption percentages according to immersion period reveals that for 2 and 10 hours of film immersion the highest water uptake was recorded regardless of sugar palm particles loading. The increase in water absorption at the tenth hour of water immersion may indicate the occurrence of second stage of water absorption. The first stage probably involves sago starch and plasticizer only due to the fact that both can easily form hydrogen bonds with water as their hydrogen bonding active sites are more accessible. During longer immersion times, more water molecules engage with these active sites thus producing enough energy to facilitate the swelling of sugar palm fiber. Similar to sago starch, cellulose and hemicellulose components in sugar palm particles are also hydrophilic but because cellulose polymer has a high crystallinity, the water needs more time for penetration.

Previous study shows that increase in water diffusion occurs due to the increase in water energy in the starch system [3, 39]. Higher amount of water molecules that exist in sago starch film as immersion period increases probably leads to more frequent collisions between water molecules, consequently their energy may also increase. This high energy acts as driving force that facilitates sugar palm particles fiber to swell. The driving force is actually water potential difference where it determines diffusion and solubility coefficients [39]. The increased

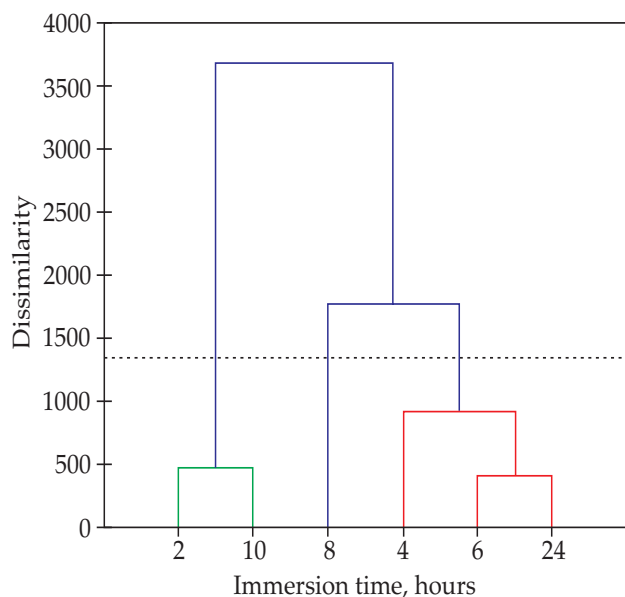


Fig. 6. Dendrogram of water absorption according to period of immersion

influx of water molecules at the 10<sup>th</sup> hour of immersion probably helps more water molecules to penetrate until the fiber achieved saturation.

Figure 6 shows dendrogram for water absorption as classification is made according to immersion period. A dendrogram is a diagram representing a tree that shows the hierarchical relationship between objects. Figure 6 shows that after 4, 6 and 24 hours of immersion moderate amount of water is absorbed, while the lowest absorption was recorded during the eighth hour reading. This fluctuation in the percentage of water uptake is probably because the films have not yet achieved stability in 24-hour water immersion period. The decrease in water absorption between 10 and 24 hours of immersion indicates that most of the active sites for hydrogen bonding have been increasingly saturated; therefore amount of water absorbed is reduced. It is expected that, as immersion period continued, water absorption will show a steady decline until equilibrium is reached.

#### Water solubility

It is understood that water solubility (Table 8, Figure 5) is the measure of water resistance of a material because it determines the amount of solute. In this context, sago starch polymer and plasticizer in the film dissolve in water, as the medium, at certain temperature and pressure. Moreover, it can function as an indicator for film degradation when thrown into water.

#### CONCLUSIONS

This study was carried out to investigate the effect of addition of sugar palm particles (SPP) on the water absorption properties of thermoplastic sago starch biopolymer composite films with 10, 20, 30, 40 and 50 wt % of SPP.

The introduction of sugar palm particles into the sago starch film successfully lowers the percentage of water absorption and solubility of sago starch film. This may be due to the increase of hydrophobic component in the system when the sugar palm particles loading is increased, which contributes to the presence of lignin component. Furthermore, factors such as increased crystallinity and good compatibility between these two phases: matrix and reinforcement also contribute to the reduction in water absorption and solubility. In addition, this study also proposes the phenomenon of second stage water absorption as a result of the increase in water absorption at 10<sup>th</sup> hour of immersion; which probably relates to water penetration into the sugar palm particles. The results of the study indicate a possibility of obtaining a more durable sago starch film by incorporation of sugar palm particles, which is important from the point of view of packaging applications of biopolymer composite films.

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