Pertanika J. Sci. & Technol. 31 (S1): 21 - 38 (2023)

PERTANIKA JOURNALS

SCIENCE & TECHNOLOGY

Journal homepage: http://www.pertanika.upm.edu.my/

Environmental Properties of Coconut Fiber/Reinforced Thermoplastic Starch/Beeswax Hybrid Composites

Khuganeshwaran Mogan[1], Ridhwan Jumaidin[1]*, Rushdan Ahmad Ilyas[2,3,4,5] and Zatil Hafila Kamaruddin[6,7]

[1]Fakulti Teknologi dan Kejuruteraan Industri dan Pembuatan, Universiti Teknikal Malaysia Melaka, Hang Tuah Jaya, 76100 Durian Tunggal, Melaka, Malaysia
[2]Faculty of Chemical and Energy Engineering, Universiti Teknologi Malaysia, 81310 UTM, Johor, Malaysia
[3]Centre for Advanced Composite Materials, Universiti Teknologi Malaysia (UTM), Johor Bahru 81310, Malaysia
[4]Institute of Tropical Forest and Forest Products (INTROP), Universiti Putra Malaysia, 43400 UPM Serdang, Selangor, Malaysia
[5]Centre of Excellence for Biomass Utilization, Universiti Malaysia Perlis, 02600, Arau, Perlis, Malaysia
[6]Faculty of Mechanical Engineering, Universiti Teknikal Malaysia Melaka, Hang Tuah Jaya, 76100 Melaka, Malaysia
[7]German-Malaysian Institute, Jalan Ilmiah Taman Universiti, Kajang 43000, Malaysia

ABSTRACT

The creation of degradable biocomposites is anticipated to alleviate the challenges of worldwide environmental contamination and resource exhaustion. The study investigates the effect of coconut fiber on the environmental properties and water affinity behavior of thermoplastic starch/beeswax composite. The biocomposites were fabricated by incorporating the coconut husk fiber range from 10 to 50 wt%. The thermoplastic starch contains cassava starch, glycerol, and beeswax. The modification of the mixture became efficient when the mixing was determined to be stronger when used as a high-pace blender to aid the mixing process. The mixture then underwent a hot compression molding method to form the mixture into the desired sample form. We can conclude from the results that samples with high fiber content absorb less water than those with no fiber content. For moisture absorption, when the fiber content increases, the ability of the fiber to moisture absorption is decreased. The thickness swelling results show that the sample shows less swelling as the fiber percentage increases. For the soil burial test, incorporating 50 wt% coconut fiber decreases the weight reduction for 4 weeks.

ARTICLE INFO

Article history:
Received: 06 March 2023
Accepted: 24 July 2023
Published: 27 October 2023

DOI: https://doi.org/10.47836/pjst.31.S1.02

E-mail addresses:
khuganmoghan98@gmail.com (Khuganeshwaran Mogan)
ridhwan@utem.edu.my (Ridhwan Jumaidin)
ahmadilyas@utm.my (Rushdan Ahmad Ilyas)
zatilhafila@gmail.com (Zatil Hafila Kamaruddin)
*Corresponding author

ISSN: 0128-7680
e-ISSN: 2231-8526

For the water solubility test, the solubility of 50 wt% is the best. Based on the findings, integrating coconut fiber into the modified thermoplastic cassava starch increases the composite properties relative to the non-reinforcement matrix material starch.

Keywords: Biodegradation, coconut fiber, soil burial, thermoplastic starch

INTRODUCTION

Due to the fast development of technology, the environment was exposed to many types of pollution, including microplastic pollution due to synthetic petroleum-based polymer plastics. Thus, engineers and scientists are collaborating to introduce novel biomaterials to reduce environmental harm. Currently, biopolymers reduce pollution, which can be decomposed easily. Recent research focuses on producing a substance composed of starch biopolymers (Abotbina et al., 2022). Starch is one of the best-promised raw materials for biodegradable growth, which can be naturally available carbohydrates from various cultivations (Punia Bangar et al., 2021). Native starch usually exists in a common granule structure with 15–45% crystallinity. Besides that, starch is one of the most important materials used to produce thermoplastic starch with the help of plasticizers as the reinforcement to enhance the mechanical properties, physical properties, and thermal properties of the biopolymer (Diyana et al., 2021; Fuqua et al., 2012; Tarique et al., 2021). The starch can be transformed into thermoplastic, TPS, under high temperature and shear (Yoksan et al., 2022). Thermoplastic starch can produce a naturally reinforced composite using natural fibers (Diyana et al., 2021). The chemical similarities of starch and plant fiber provide strong compatibility when the natural fibers combined with TPS enhance their mechanical properties.

One disadvantage of thermoplastic starch (TPS) for packaging is that it is not as strong as other plastic materials, such as polyethylene, and may break down over time (Ilyas et al., 2018). Additionally, TPS is biodegradable only under certain conditions, such as industrial composting, which may not be readily available. As a result, TPS packaging may not be as durable or sustainable as other plastic packaging options. Many materials can reinforce the composite material (Fuqua et al., 2012). These criteria of the natural fiber encouraged the researchers to develop natural fiber to become natural composites. Due to the low economic development and improving rural areas, natural fiber-reinforced composites have become the prime factor in reducing the world's need for petroleum-based materials (Fuqua et al., 2012).

Adding commercial cellulose fiber or micro-fiber improves the water resistance of the TPS (Ma et al., 2005). Recent studies have shown that the use of natural fiber in the composite will increase the capacity of traditional thermoplastics; it is identified that coconut husk fiber can be used to produce a better and improved quality of reinforced composite with the aid of a plasticizer in the mixture (Mościcki et al., 2012; Thinkohkaew

et al., 2020). In a previous study reported by Jústiz-Smith et al. (2008), the tensile strength values of coconut fibers are comparable to those reported in the literature (Eichhorn et al., 2001), although in the case, the mean value of coconut fiber was smaller than 175 MPa. Previous research suggested that natural fiber, by physical and mechanical characteristics such as tensile strength, is stronger than synthetic fiber. The cellulose content accounts for the high tensile strength of composite materials; thus, materials fabricated from these fibers show good mechanical properties and can be used when the strength is important (Jústiz-Smith et al., 2008). Natural composites are given huge attention for the development of the perceived negative impact of plastics in solid waste disposal (Willett, 2009). Improving bio-natural composite material is supposed to reduce the environmental emissions caused by traditional thermoplastics and introduce better materials that are safer and pollution-free for the users and nature. Several studies were conducted to analyze the potential of natural fiber-reinforced composites. According to a study by Jumaidin et al. (2021), thermoplastic starch was reinforced with banana leaf fiber, significantly enhancing its mechanical properties. Meanwhile, a study conducted by Hazrati et al. (2021) utilized thermoplastic starch and *Dioscorea hispida* fiber to demonstrate that the modification of thermoplastic starch resulted in enhanced material properties. The water solubility of the biocomposite has decreased while its mechanical properties have increased.

Beeswax is wax that occurs naturally and can be obtained from a bee's hive. It is generally known as the most effective hydrophobic organic molecule that may be utilized for reducing water sensitivity. Examining raw beeswax quality is based on its physical color appearance, which indicates its product value. After manipulation, the freshly produced beeswax initially appears light or white but transforms to yellow, dark yellow, and brownish tones. However, the incorporation of beeswax into thermoplastic starch resulted in a reduction of the mechanical properties of the bio-composite due to poor compatibility (Zhang et al., 2018), attributed to the formation of a heterogeneous film and structural discontinuities upon the addition of beeswax. Therefore, the issue of incompatibility must be addressed when combining hydrophilic thermoplastic starch and hydrophobic beeswax. Incorporating natural fiber reinforcement within the thermoplastic starch/beeswax matrix presents a promising solution to address this issue.

Even though studies have reported using thermoplastic starch/beeswax as the matrix in composites, none has reported the utilization of coconut fiber as the reinforcement via the hot compression molding method. Thus, this study aims to investigate the effect of coconut fiber on the environmental properties and water affinity behavior of thermoplastic starch/beeswax composite.

MATERIALS AND METHODS

Materials

The coconut husk was obtained from the coconut farm and soaked in water for 2 weeks. The soaking process helped to extract the fiber as the filler. Then, the coconut husk has isolated the fiber from the rest of the coconut husk. The collected coconut fiber was sun-dried for 1 day to ensure the fiber dried completely. The fiber is then ground to produce a short fiber of approximately 1 cm long with a diameter range of 125 to 300 microns. The fiber is then put in an oven at a temperature of 105°C for 5 hours to eliminate the moisture content of the fiber. Cassava starch obtained was purchased from Antik Sempurna Sdn. Bhd., Shah Alam; *Glycerol* was obtained from QReC (Asia) Sdn. Bhd., Rawang with an AR grade of 99.5% and beeswax was obtained from Aldrich Chemistry.

Sample preparation

Prior to the composite's preparation, thermoplastic cassava starch (TPCS) was mixed with 5% beeswax to reduce the moisture sensitivity of the material. Then, TPCS/Beeswax with coconut fiber was modified by incorporating different amounts of coconut fiber (0, 10, 20, 30, 40, 50 wt.%) into the polymer matrix. The mixture was pressed at 145°C for approximately 1 hour and a cooling process at 30°C for about 15 minutes. The hot pressing was carried out using Go Tech hydraulic testing.

Scanning Electron Microscopy (SEM)

A scanning electron microscope (SEM), Zeiss Evo 18 model, and a 10 kV acceleration voltage were used to analyze tensile fractured surfaces' morphology.

Fourier-Transform Infrared Spectroscopy (FTIR)

The Fourier-Transform Infrared Spectroscopy, FTIR, was used to analyze and detect the presence of the functional groups. The results of this test will be obtained using the JASCO FT/IR-6100 IR spectrometer Japan.

Thickness Swelling

The swelling analysis of the samples was conducted using the enhanced method developed by Jawaid et al. (2011). Five (10 mm × 10 mm × 3 mm) samples were cut and oven dried for 24 h at 105°C ± 2. The samples were immersed in distilled water at room temperature of 23°C ± 1, ranging from 30 minutes to 2 hours. The initial thickness of the sample was recorded as (T_i), and the final thickness of the samples was recorded as (T_f).

$$Thickness\ Swelling\ \%: [\{T_f - T_i\} / T_i]\ X\ 100\%$$

Moisture Content

The moisture content refers to the amount of water present in a sample. The water uptake capacity of each sample was assessed by measuring the weight loss according to the ASTM D 644-94 (1994). Moisture content was evaluated for five samples with measurement (10 mm x 10 mm x 3 mm). The weight of the specimens was registered for the initial weight, W_i, and placed into an oven to undergo the drying process for 24 hours at a temperature of 105°C. The final weight, W_f of the specimen, is recorded immediately after the specimen is removed from the oven to ensure the surrounding moisture will not be absorbed into the specimen while weighing the specimen.

$$Moisture\ Content\ \%: [\{W_f - W_i\} / W_i]\ X\ 100\%$$

Water Absorption

The investigation involved the analysis of water absorption using the ASTM D 570-98 (1998) approach. Five specimens, each measuring (10 × 10 × 3 mm), were subjected to a drying process for 24 hours in an air-circulating oven at a temperature of 105°C ± 2 to remove any residual moisture from the samples. Then, the samples were dipped into the water at room temperature of 23°C ± 1 with a range of time from 30 minutes to 2 hours. The specimens were weighed before, W_i, and after, W_f the immersion to obtain the water absorbed by the samples.

$$Water\ Absorption\ \%: [\{W_f - W_i\} / W_i]\ X\ 100\%$$

Moisture Absorption

Five (5) samples, each with dimensions of 10 x 10 x 3 mm, were prepared. The samples were dried in a vacuum oven at 105°C for 24 hours. After drying, the samples were cooled in a desiccator until they reached constant weight and practical equilibrium. The samples were placed in a relative humidity chamber at 75% and 25°C. The humidity chamber was obtained from GOTECH, GT-7005, China. For the removal of the existing moisture from the samples, the samples were dried in a circulating oven for 24 hours at a temperature of 10°C. The mass of the sample was reported before Mi and after Mf in the humidity chamber phase.

$$Moisture\ Absortion\ \%: [\{M_f - M_i\} / M_i]\ X\ 100\%$$

Density Test

The composite's density measurement was conducted per the ASTM D1895 standard. Five

specimens, each measuring 10 x 10 x 3 mm, were prepared and subjected to drying for 24 hours in an oven maintained at a temperature of 105°C to remove the existing moisture content in the specimen. Then, the densimeter weighed the samples, and the reading was recorded. Later, the specimens were inserted into the water inside the densimeter to identify the specimen's density and volume.

$Density$: $Mass$ / volume

Environmental Testing

Environmental testing is the performance measurement of the material under a specified condition caused by the environment. Two tests were conducted, such as water solubility and soil burial, to analyze the weight loss of the sample after undergoing the testing accordingly.

Water Solubility

The water solubility analysis of the samples was performed using the methodology developed by Zhang et al. (2018) with minor adjustments. Five samples measuring (10 mm x 10 mm x 3 mm) were cut, and the initial weight of the specimens was recorded. Secondly, the specimens were immersed in a container of 30 ml of distilled water and stirred slowly. Once the process was complete, the remaining sample was removed from the container, and the surface water of the sample was cleaned by covering filter paper on the specimen's surface. The samples were dried again for 24 hours at 105°C ± 2 to assess the final weight of the specimens.

$Water\ Solubility$ %: $[\{W_f - W_i\} / W_{i]}\ X\ 100\%$

Soil Burial

The biodegradation test was conducted using the methodology adopted by Jumaidin et al. (2020). The soil mixture was 50% sand and 50% soil obtained. The water content of the soil and the mixture were kept within the range of 30–40% by adding 400 ml of water per day to 1250 g of the mixture. A plastic green mesh was used to cover the sample before burial to ease the removal of the specimen from the soil, which will ensure there is an allowance for the microorganisms and moisture to be in contact with the samples. Firstly, the specimens were weighed to get the initial weight, W_i, before the testing. Secondly, the specimens were covered with soil and stapled together with plastic green mesh before the specimens were buried. Then, the specimens were buried into the polybag with a depth of 10 cm. Fourthly, the specimens were removed from the polybag at specified intervals and

cleaned with distilled water to remove impurities from the sample surface. The specimens were dried for 24 hours in a circulating oven at a temperature of 105°C. The specimens were dried for 24 hours in a circulating oven at a temperature of 105°C and weighed to get the final sample weight, $W_{f.}$

$$Soil\ Burial\ \%: [\{W_f - W_i\} / W_i]\ 100\%$$

RESULT AND DISCUSSION

Scanning Electron Microscopy (SEM)

SEM images of thermoplastic starch reinforced with coconut fiber (0, 10, 20, 30, and 50 wt%) are displayed in Figure 1. In the 0 wt% specimen, the surface of the specimen was observed to be in glossy granular patterns due to the presence of beeswax changes in the composition of the starch. For the specimen with 10 wt% coconut fiber, a crack line can be found on the specimen, acting as phase separation. Thus, the strength between the specimens is weak, and the specimen has no reinforcement. Specimen with 20 wt% fiber, there is no separation between the fiber and the matrix as the fiber mixed homogeneously with the matrixes. Specimen with 30 wt% shows a good adhesion of the matrix and coconut fiber, and specimen with 50 wt% shows the presence of void and fiber breakage as the percentage of fiber increases.

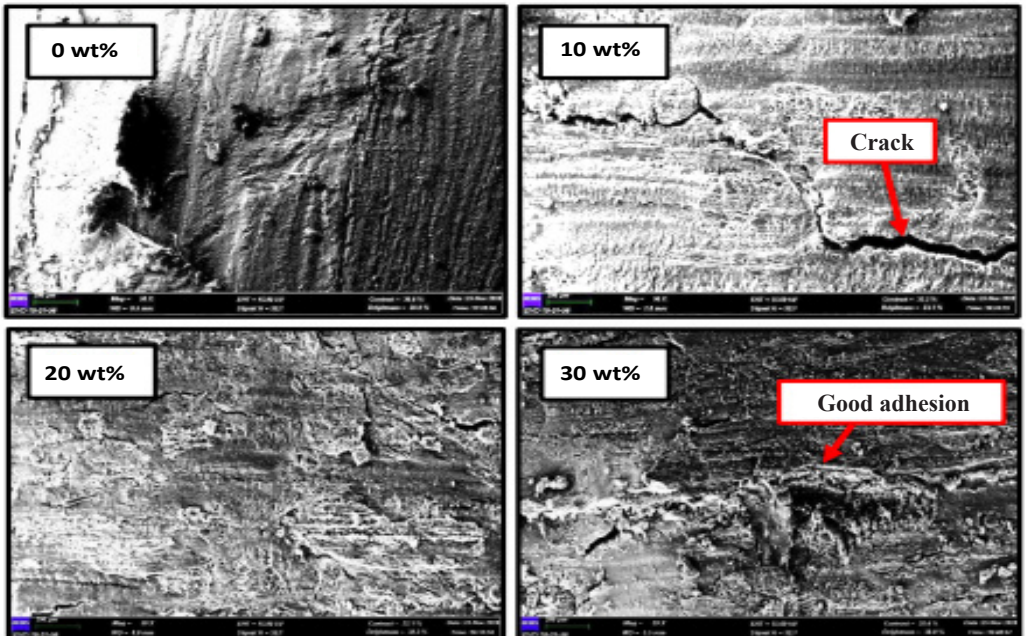

Figure 1. SEM micrograph of TPCS reinforced with coconut fiber

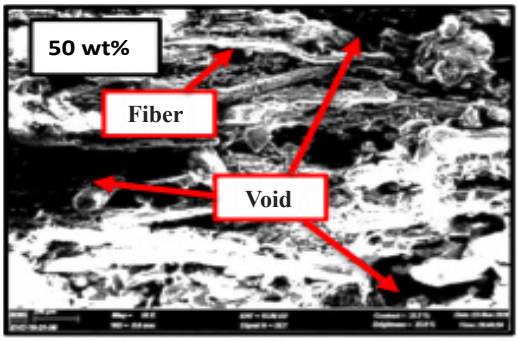

Figure 1. Continue

Fourier-Transform Infrared Spectroscopy (FT-IR)

The pattern of the composites was similar to other fiber composites, even though they had different percentages of fiber. It indicates that combining other materials with starch to form a composite will not be influenced by the starch's chemical composition. The main goal of the FT-IR test was to characterize the chemical bonding of TPCS with cassava starch.

Figure 2 shows the results obtained from the FTIR test on the TPCS and its composite reinforced with coconut fiber. The starch and coconut fiber have contributed to the presence of a hydrogen-bonded hydroxyl group, O-H, which was 3400–3200 cm^{-1}. According to Venkatachalam et al. (2016) findings, the involvement of amylose and amylopectin in jute fiber has been shown to affect the presence of the O-H group in the spectrum. The specimen was also present in the C-H range of aliphatic hydrocarbon groups found in the 2936–2916 cm^{-1} range. The C-H bonding found with 20 to 50 wt% found a decrease from 2919.9 cm^{-1} to 2919.0 cm^{-1}. These findings agree with the study by Alawar et al. (2009), which reported that the presence of the C-H band comes from natural fiber components such as cellulose and hemicellulose. Other than that, the N-H range of hydrocarbon groups was found in

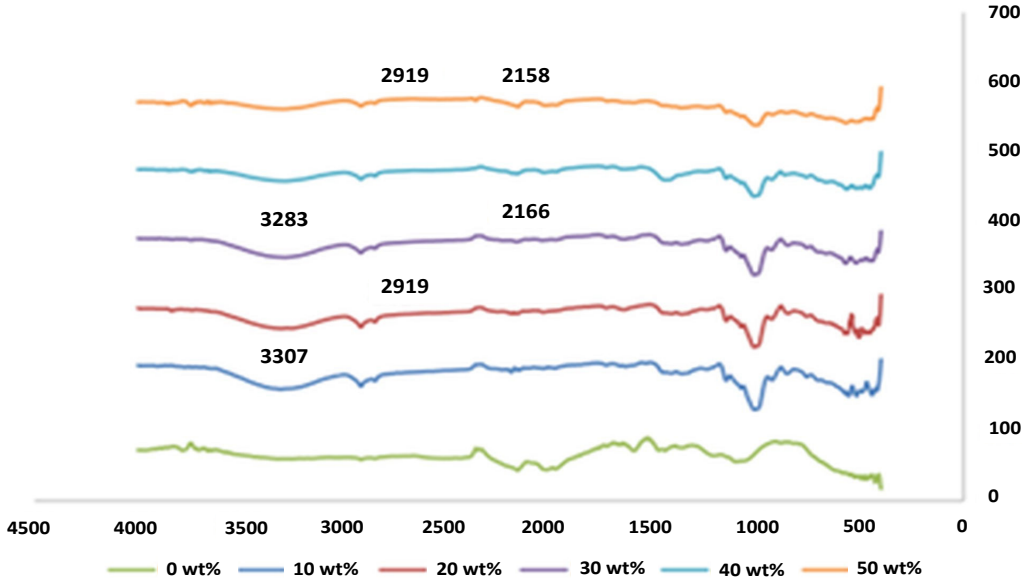

Figure 2. Results obtained from conducting the FTIR test on the TPCS reinforced with coconut fiber

the 2806-2000 cm^{-1} range. The peak found a continuous decrease from specimen 30 wt% to 50 wt% from 2166.6 cm^{-1} to 2158.0 cm^{-1}.

Meanwhile, a small change in the rotation of the O-H bands could be observed as the percentage of fiber loading increases. Specimen with 10 wt% fiber peaks at 3307 cm^{-1}; as the percentage increases, the peak changed to lower the wavenumber as observed with 30 wt% fiber specimens, the peak at 3283.2 cm^{-1}. According to the study conducted by Alawar et al. (2009), the increase of intermolecular hydrogen bonding has been clarified to influence the peak of the spectral band. The results reveal that the TPCS/BW matrix is compatible with the coconut fiber.

Thickness Swelling

Figure 3 shows the thickness swelling for TPCS and its composites. The specimen with a higher fiber proportion was found to swell less. For 30 minutes and 2 hours, the amount of thickness swelling of the specimen with 0 wt% fiber is 21.69% and 39.32%, respectively, and for the specimen with 50 wt% fiber is 6.96% for 30 minutes and 14.17% for 2 hours, respectively. The thickness swelling value for 30 minutes keeps decreasing from 30 wt% to 50 wt% while the swelling value for 2 hours, from 30 to 50 wt%, overall decreasing. This finding concludes that the swelling of the specimen decreases with the rise in the percentage of fiber loading due to the presence of a void in the specimen (Masoodi & Pillai, 2012). A void in the composite influences the specimen's dimensional stability and causes the specimen to start to delaminate because it absorbs water (Masoodi & Pillai, 2012).

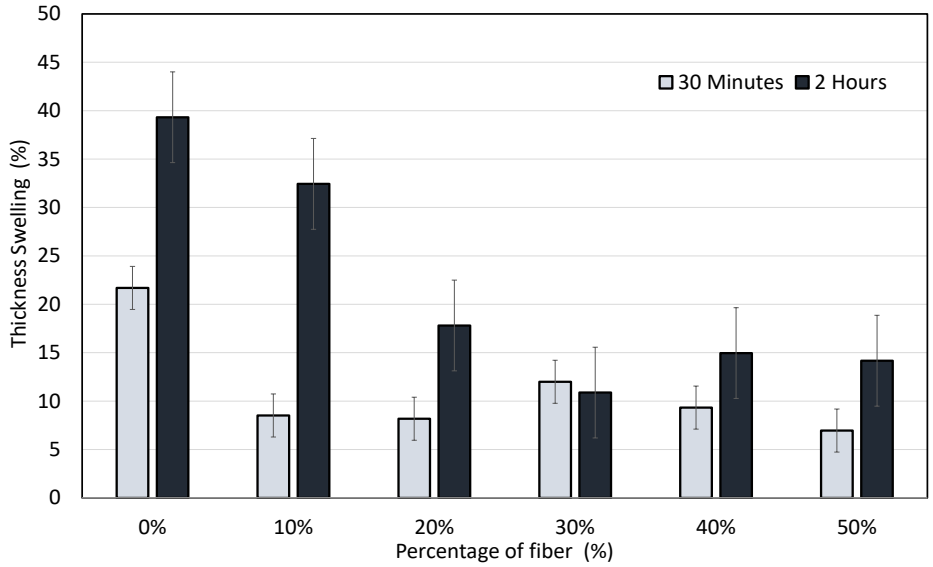

Figure 3. Thickness swelling results from TPCS reinforced with coconut fiber

The observed outcome can be ascribed to fiber in the composites, which exhibits a stiffer configuration than starch, thus imparting greater dimensional stability to the composites (Kamaruddin et al., 2023).

Moisture Content

The moisture content testing is important to conduct in composites' consistency, development, and degradation. Determination of the moisture content is also important for the uniform measurement of the content of other composite materials. The moisture content was tested to determine the amount of moisture in the TPCS matrix specimen and its composite. The moisture content in the composite is influenced by the form of hydrophilic material used, such as glycerol, starch, and fiber (Mubashar et al., 2009).

On the other hand, the moisture content in the composite can impact physical and environmental properties such as water absorption, moisture absorption, water solubility, and biodegradation (Wang et al., 2006). The moisture content analysis indicates that fiber incorporation raises the composite's moisture content due to the hydrophilicity of coconut husk fiber. According to the study conducted by Wang et al. (2006), composites with 40%, 50%, 55%, and 60% fiber loads did not demonstrate the level of which signals the equilibrium moisture absorption.

Figure 4 shows that the specimen with the highest moisture content is 0 wt%, and the lowest moisture content is 40 wt%. The results show that fiber integration reduces the moisture content of the composite, but only for 50 wt% fiber content shows rise, attributed to void. According to the study by Joshi et al. (2004), it was found that the moisture content of TPCS is lower compared to the specimen with natural fiber mixed with TPCS. It proves that the presence of natural fiber increases the moisture content of the composites.

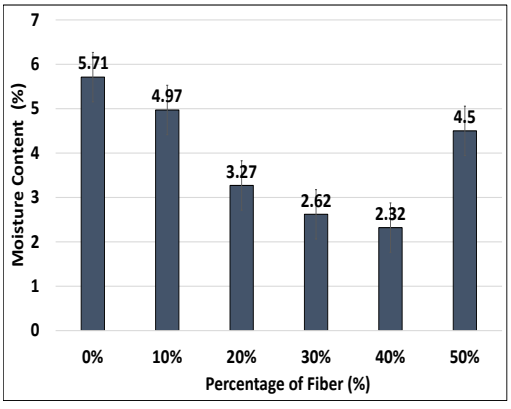

Figure 4. Moisture content results for TPCS reinforced with coconut fiber

Water Absorption

Water absorption of natural composites is a significant concern for their future outdoor applications. Coconut husk fiber water absorption reinforced unsaturated polyester composites following so-called pseudo-Fickian conduct were found (Akil et al., 2009).

Based on testing results, reducing water absorption depended mainly on matrix affinity. Compared to water, the fiber results in a better interface bonding between the

fiber matrix and the bonding created by the higher fiber loading, which prevents water from spreading into the matrix (Akil et al., 2009).

The water absorption ability of the TPCS matrix and its composite when immersed in water for 30 minutes and 2 hours are shown in Figure 5. For most specimens, water absorption results in 40 to 46% absorption when submerged in water for 30 minutes, which shows no substantial difference.

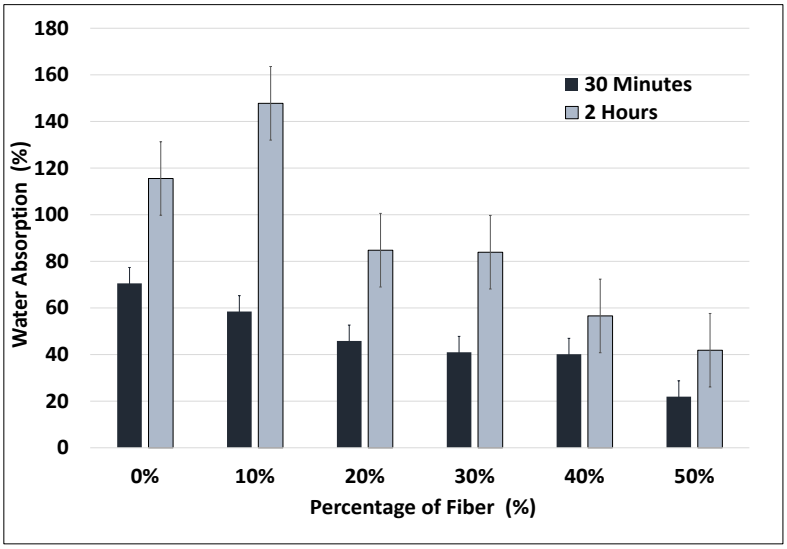

Figure 5. Water absorption result for TPCS reinforced with coconut fiber

As for the specimens submerged in water for 2 hours, it was observed that as fiber content increases, the amount of water absorbed decreases, but only for 10 wt% fiber; there is a rise due to phase separation, as mentioned in SEM micrograph analysis.

The lowest water absorbed in the specimen is fiber, with 50 wt% valued as 41.84% water absorbed. Generally, the 30-minute and 2-hour water absorption graphs show a constant decrease in the amount of water absorbed. Specimen with 30 wt% fiber for 30 minutes absorbed 40.97% of water while absorbed 83.9% of water for 2 hours testing time. According to the study conducted by van Bavel (1996), higher cellulose content in natural fiber can help to increase water absorption.

Moisture Absorption

According to the study conducted by Kim et al. (2016), the rate of absorption of moisture is the rate of transport of water molecules through molecules that can be caused by certain conditions, such as the diffusion coefficient, the state of the atmosphere or equilibrium, and even the dimension of the substance itself. A moisture absorption test is important to be carried out in a composite to identify the rate of moisture absorption by the composite.

This finding is in agreement with the study by Kim et al. (2016), which reported that the absorption of moisture in thermoplastic material is one of the major causes of thermoplastic failure because the presence of moisture absorbed from the environment in the TPS causes the formation of hydrogen intermolecular bonding with the fiber, which weakens the matrix and the fiber interfacial bonding. According to Kim et al. (2016), a weaker matrix and fiber interfacial bonding will cause the rate of water absorption to increase dramatically.

Figure 6 shows the results obtained by conducting a moisture absorption test. Overall results show that the sample completely absorbed the moisture to its potential, suggesting that the sample was extremely hygroscopic. However, the specimen with 0 wt% fiber has the least moisture absorption in terms of fiber loading, and the specimen with 10 wt% has the highest moisture absorption within 5 days. It was found that composite with 20 wt% to 50 wt% fiber has a lower percentage of moisture absorption by 14.17% for 20 wt% fiber, 12.47% for 30 wt% fiber, 10.81% for 40 wt% fiber, and 11.14% for 50 wt% fiber, respectively. The results obtained for 0 wt% are lower than the other fiber loading specimens due to the low interfacial bonding of the specimens' matrix and the presence of the void in the specimen.

This result is correlated with the water absorption test, proving that the interfacial bonding of the specimen's matrix and fiber is better with an increasing percentage of fiber in the specimen. The hydrophilic properties of the fiber cause the moisture absorption of the specimen. This finding agrees with the study by Wang et al. (2006); due to the strong hydrophilic characteristics of bamboo, the increasing amount of bamboo fiber in the TPS and the bamboo composite increase the absorption rate.

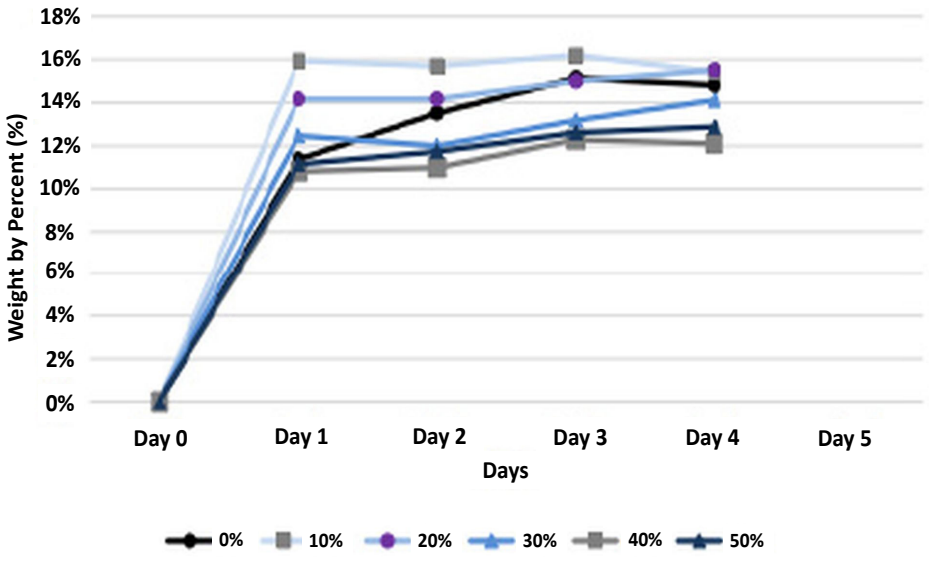

Figure 6. Moisture absorption of TPCS reinforced with coconut fiber

Density Test

According to the study conducted by Kilinç et al. (2017)**,** the strength of the natural composite directly affects the fiber density. Polymer molecular weight, fiber percentage, fiber processing processes, and fiber stretching specifically influence fiber density. Due to the ability of natural fiber to be low cost and low density, the mechanical properties of natural fibers are enhanced with fiber treatment (Bhatnagar et al., 2015).

Figure 7 shows the results obtained from the density test. The results overall increase when the percentage of the fiber increases. The density of the 0 wt% specimen is 1.401. The density of the 10 to 50 wt% was 1.303, 1.250, 1.232, 1.111, and 1.232 kg/m^3, respectively. The densimeter was used to obtain the density of the specimens. The density value of specimens increases as the fiber percentage increases.

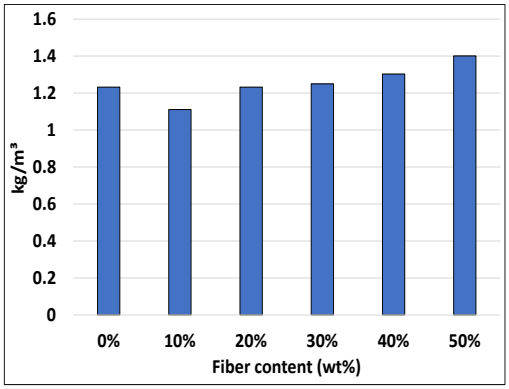

Figure 7. Density of TPCS reinforced with coconut fiber

This result was similar to the moisture content of the coconut fiber composite. This finding agrees with the study of Bhatnagar et al. (2015); the packing reduces due to the presence of long fibers in the composites, interrupting fiber flow and resulting in high void spaces. With the rise in fiber content, the void content in the composite increases. In natural fibers, large concentrations of the hydroxyl group make them polar and hydrophilic. This polar nature in natural fiber-based polymer composites also results in high moisture absorption, leading to fiber swelling and voids in the fiber-matrix interface.

Water Solubility

Another significant quality of using fibers as polymer reinforcement is the tendency of natural fiber to be soluble in water. To effectively soluble in water from natural fibers, it is important to understand the water solubility mechanisms in these materials (Väisänen et al., 2017). From the obtained results, it can be concluded that when the fiber content increases, the water solubility levels are lower.

Figure 8 shows the results of the water solubility of TPCS/BW and its composite. The results show a continuous decrease in the water solubility of the specimen as the fiber percentage increases. The specimen with 0 wt% values water solubility percentage of 46.31%, the specimen with 10 wt% fiber values water solubility percentage of 64%, while 20 to 50 wt% values at 59.38%, 57.95%, 48.22%, and 28.57%, respectively. These

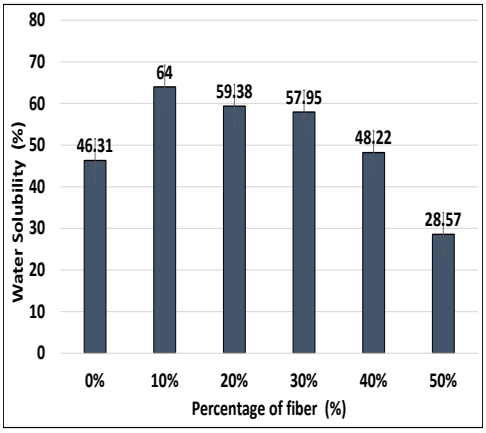

Figure 8. Results of the water solubility of TPCS reinforced with coconut fiber

results are similar to moisture absorption testing. According to the study by Ma et al. (2005), the increasing percentage of fiber in a composite may decrease the solubility of the composite.

This finding is in agreement with the study by Akil et al. (2011), which reported that it is likely that the interfacial bonding of the matrix and fiber is stronger with a higher percentage of fiber, similar to the results obtained from the moisture absorption test. The high fiber usage in the specimen could impair the excluded outcome. Additionally, this observation is consistent with prior research on the development of cornhusk/ sugar palm fiber reinforced corn starch-based hybrids, which indicated that adding sugar palm fiber resulted in reduced sample solubility (Ibrahim et al., 2020).

Soil Burial

"Biodegradation" in the biomedical sector refers to hydrolyzes and oxidations, the main mechanisms of polymer degradation (Mumtaz et al., 2010). Based on Kalka et al. (2014), polymer biodegradation can involve various steps, including polymer degradation by decomposing organisms.

Referring to the results obtained, a mass reduction for the specimens showed a decreasing trend with the increased fiber loading. The soil burial test was similar to the water absorption test result (Mansor et al., 2015). A study by Mumtaz et al. (2010) showed that certain natural fiber composites slow the degradation rate as the fiber percentage increases. This finding agrees with the study by Kalka et al. (2014), which reported that the growth in date palm and flax fiber in the composite reinforced corn starch indicates a decline in the mass reduction percentage. It can be assumed that the higher the fiber content in the samples, the lower the sample's percentage of mass reduction.

Figure 9 shows the results gathered from 4 weeks of soil burial tests. The overall result shows a reduction in the mass of the specimen. For the specimen with 0 wt%, the mass reduction is 45.11%, and the lowest is a specimen with 20 wt% values at 40.33%. In addition, the specimen with fiber content from 30 to 50 wt%, values 50.39%, 50.12%, and 38.85% respectively. This overall result can be obtained compared to the water absorption result. The specimen with 10 wt% fiber mass reduction is higher than the other fiber content specimen due to a void in the specimen.

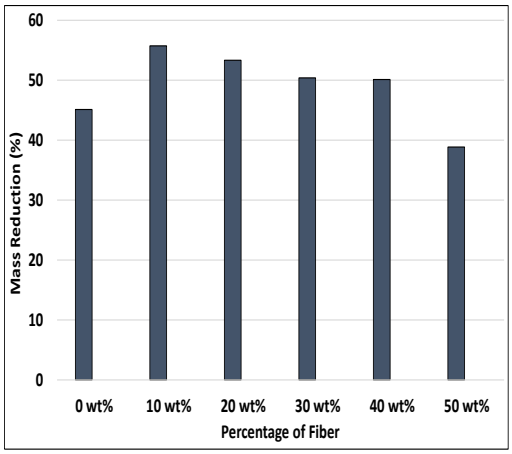

Figure 9. Mass reduction (%) from the soil burial test for TPCS reinforced with coconut fiber

CONCLUSION

Thermoplastic starch reinforced with coconut fiber was successfully prepared through dry mixing and hot-pressing. It was found that incorporating coconut fiber has reduced the water absorption and moisture content of the thermoplastic starch matrix. A decrease in the material's thickness, swelling, and solubility accompanied this finding. Composites with 50 wt% fiber show the least water absorption and swelling percentage. This result can be associated with improving the material's dimensional stability, which is less hydrophilic than the neat thermoplastic starch matrix. Soil burial results show that higher fiber content led to lower degradation activity of the material, where 50% fiber shows the lowest weight reduction. Overall, incorporating coconut fiber has reduced the hydrophilicity characteristic of the material, increasing the potential of this composite as an alternative material for more environmentally friendly bioplastic products.

ACKNOWLEDGMENTS

The author thanks Universiti Teknikal Malaysia Melaka, Malaysia for the financial support from research grant PJP/2021/FTKMP/S01815.

REFERENCES

Abotbina, W., Sapuan, S. M., Sultan, M. T. H., Alkbir, M. F. M., & Ilyas, R. A. (2022). Extraction, characterization, and comparison of properties of cassava bagasse and black seed fibers. *Journal of Natural Fibers*, *19*(16), 14525-14538. https://doi.org/10.1080/15440478.2022.2068103

Akil, H. M., Cheng, L. W., Mohd Ishak, Z. A., Abu Bakar, A., & Abd Rahman, M. A. (2009). Water absorption study on pultruded jute fibre reinforced unsaturated polyester composites. *Composites Science and Technology*, *69*(11-12), 1942-1948 https://doi.org/10.1016/j.compscitech.2009.04.014

Akil, H. M., Omar, M. F., Mazuki, A. A. M., Safiee, S., Ishak, Z. A. M., & Abu Bakar, A. (2011). Kenaf fiber reinforced composites: A review. *Materials and Design*, *32*(8–9), 4107–4121. https://doi.org/10.1016/j.matdes.2011.04.008

Alawar, A., Hamed, A. M., & Al-Kaabi, K. (2009). Characterization of treated date palm tree fiber as composite reinforcement. *Composites Part B: Engineering*, *40*(7), 601-606 https://doi.org/10.1016/j.compositesb.2009.04.018

Bhatnagar, R., Gupta, G., & Yadav, S. (2015). A Review on composition and properties of bagasse fibers. *International Journal of Scientific & Engineering Research*, *6*(5), 143-147.

Diyana, Z. N., Jumaidin, R., Selamat, M. Z., Ghazali, I., Julmohammad, N., Huda, N., & Ilyas, R. A. (2021). Physical properties of thermoplastic starch derived from natural resources and its blends: A review. *Polymers*, *13*(9), 5–20. https://doi.org/10.3390/polym13091396

Eichhorn, S. J., Baillie, C. A., Zafeiropoulos, N., Mwaikambo, L. Y., Ansell, M. P., Dufresne, A., Entwistle, K. M., Herrera-Franco, P. J., Escamilla, G. C., Groom, L., Hughes, M., Hill, C., Rials, T. G., & Wild, P. M. (2001). Current international research into cellulosic fibres and composites. *Journal of Materials Science*, *36*(9), 2107–2131. https://doi.org/10.1023/A:1017512029696

Fuqua, M. A., Huo, S., & Ulven, C. A. (2012). Natural fiber reinforced composites. *Polymer Reviews*, *52*(3–4), 259–320. https://doi.org/10.1080/15583724.2012.705409

Hazrati, K. Z., Sapuan, S. M., Zuhri, M. Y. M., & Jumaidin, R. (2021). Preparation and characterization of starch-based biocomposite films reinforced by *Dioscorea hispida* fibers. *Journal of Materials Research and Technology*, *15*, 1342–1355. https://doi.org/10.1016/j.jmrt.2021.09.003

Ibrahim, M. I. J., Sapuan, S. M., Zainudin, E. S., & Zuhri, M. Y. M. (2020). Preparation and characterization of cornhusk/sugar palm fiber reinforced corn starch-based hybrid composites. *Journal of Materials Research and Technology*, *9*(1), 200–211. https://doi.org/10.1016/j.jmrt.2019.10.045

Ilyas, R. A., Sapuan, S. M., Ishak, M. R., & Zainudin, E. S. (2018). Development and characterization of sugar palm nanocrystalline cellulose reinforced sugar palm starch bionanocomposites. *Carbohydrate Polymers*, *202*, 186–202. https://doi.org/10.1016/j.carbpol.2018.09.002

Jawaid, M., Khalil, A. H. P. S., Khanam, N. P., & Bakar, A. A. (2011). Hybrid composites made from oil palm empty fruit bunches/jute fibres: Water absorption, thickness swelling and density behaviours. *Journal of Polymers and the Environment*, *19*(1), 106–109. https://doi.org/10.1007/s10924-010-0203-2

Joshi, S. V., Drzal, L. T., Mohanty, A. K., & Arora, S. (2004). Are natural fiber composites environmentally superior to glass fiber reinforced composites? *Composites Part A: Applied Science and Manufacturing*, *35*(3), 371–376. https://doi.org/10.1016/j.compositesa.2003.09.016

Jumaidin, R., Ahmad Diah, N., Alamjuri, R. H., Ahmad Rushdan, I., & Yusof, F. A. (2021). Processing and characterisation of banana leaf fibre reinforced thermoplastic cassava starch composites. *Polymers*, *13*(9), 1420. https://doi.org/https://doi.org/10.3390/ polym13091420

Jumaidin, R., Khiruddin, M. A. A., Asyul Sutan Saidi, Z., Salit, M. S., & Ilyas, R. A. (2020). Effect of cogon grass fibre on the thermal, mechanical and biodegradation properties of thermoplastic cassava starch biocomposite. *International Journal of Biological Macromolecules*, *146*, 746–755. https://doi.org/10.1016/j.ijbiomac.2019.11.011

Jústiz-Smith, N. G., Virgo, G. J., & Buchanan, V. E. (2008). Potential of Jamaican banana, coconut coir and bagasse fibres as composite materials. *Materials Characterization*, *59*(9), 1273–1278. https://doi.org/10.1016/j.matchar.2007.10.011

Kalka, S., Huber, T., Steinberg, J., Baronian, K., Müssig, J., & Staiger, M. P. (2014). Biodegradability of all-cellulose composite laminates. *Composites Part A: Applied Science and Manufacturing*, *59*, 37-44. https://doi.org/10.1016/j.compositesa.2013.12.012

Kamaruddin, Z. H., Jumaidin, R., Kamaruddin, Z. H., Asyraf, M. R. M., Razman, M. R., & Khan, T. (2023). Effect of *Cymbopogan citratus* fibre on physical and impact properties of thermoplastic cassava starch/ palm wax composites. *Polymers*, *15*(10), 2364. https://doi.org/10.3390/polym15102364

Kilinç, A. Ç., Durmuşkahya, C., & Seydibeyoğlu, M. Ö. (2017). Natural fibers. In M. O. Seydibeyoğlu, A. K. Mohanty., & M. Misra (Eds.) *Fiber technology for fiber-reinforced composites* (pp. 209-235). Woodhead Publishing. https://doi.org/10.1016/B978-0-08-101871-2.00010-2

Kim, S., Van Zyl, J., Johnson, J., Moghaddam, M., Tsang, L., Colliander, A., Dunbar, S., Jackson, T., Jaruwatanadilok, S., West, R., Berg, A., Caldwell, T., Cosh, M., Lopez-Baeza, E., Thibeault, M., Walker, J., Entekhabi, D., & Yueh, S. (2016, July 10-15). *Surface soil moisture retrieval using L-band SMAP SAR data and its validation*. [Paper presentation]. International Geoscience and Remote Sensing Symposium (IGARSS), Beijing, China. https://doi.org/10.1109/IGARSS.2016.7729028

Ma, X., Yu, J., & Kennedy, J. F. (2005). Studies on the properties of natural fibers-reinforced thermoplastic starch composites. *Carbohydrate Polymers*, *62*(1), 19–24. https://doi.org/10.1016/j.carbpol.2005.07.015

Mansor, M. R., Salit, M. S., Zainudin, E. S., Aziz, N. A., & Ariff, H. (2015). Life cycle assessment of natural fiber polymer composites. In K. R. Hakeem, M. Jawaid., & O. Y. Alothman (Eds.) *Agricultural biomass based potential materials* (pp.121-141). Springer. https://doi.org/10.1007/978-3-319-13847-3_6

Masoodi, R., & Pillai, K. M. (2012). A study on moisture absorption and swelling in bio-based jute-epoxy composites. *Journal of Reinforced Plastics and Composites*, *31*(5), 285–294. https://doi.org/10.0177/0731684411434654

Mościcki, L., Mitrus, M., Wójtowicz, A., Oniszczuk, T., Rejak, A., & Janssen, L. (2012). Application of extrusion-cooking for processing of thermoplastic starch (TPS). *Food Research International*, *47*(2), 291-299. https://doi.org/10.1016/j.foodres.2011.07.017

Mubashar, A., Ashcroft, I. A., Critchlow, G. W., & Crocombe, A. D. (2009). Moisture absorption-desorption effects in adhesive joints. *International Journal of Adhesion and Adhesives*, *29*(8), 751-760. https://doi.org/10.1016/j.ijadhadh.2009.05.001

Mumtaz, T., Khan, M. R., & Hassan, M. A. (2010). Study of environmental biodegradation of LDPE films in soil using optical and scanning electron microscopy. *Micron*, *41*(5), 430-438. https://doi.org/10.1016/j.micron.2010.02.008

Punia Bangar, S., Nehra, M., Siroha, A. K., Petrů, M., Ilyas, R. A., Devi, U., & Devi, P. (2021). Development and characterization of physical modified pearl millet starch-based films. *Foods*, *10*(7), 1609. https://doi.org/10.3390/foods10071609

Tarique, J., Sapuan, S. M., Khalina, A., Sherwani, S. F. K., Yusuf, J., & Ilyas, R. A. (2021). Recent developments in sustainable arrowroot (*Maranta arundinacea* Linn) starch biopolymers, fibres, biopolymer composites and their potential industrial applications: A review. *Journal of Materials Research and Technology*, *13*, 1191–1219. https://doi.org/10.1016/j.jmrt.2021.05.047

Thinkohkaew, K., Rodthongkum, N., & Ummartyotin, S. (2020). Coconut husk (*Cocos nucifera*) cellulose reinforced poly vinyl alcohol-based hydrogel composite with control-release behavior of methylene blue. *Journal of Materials Research and Technology*, *9*(3), 6602-6611. https://doi.org/10.1016/j.jmrt.2020.04.051

Väisänen, T., Das, O., & Tomppo, L. (2017). A review on new bio-based constituents for natural fiber-polymer composites. *Journal of Cleaner Production*, *149*, 582–596. https://doi.org/10.1016/j.jclepro.2017.02.132

van Bavel, C. H. M. (1996). Water relations of plants and soils. *Soil Science*, *161*(4). 257-260. https://doi.org/10.1097/00010694-199604000-00007

Venkatachalam, N., Navaneethakrishnan, P., Rajsekar, R., & Shankar, S. (2016). Effect of pretreatment methods on properties of natural fiber composites: A review. *Polymers and Polymer Composites*, *24*(7), 555-566. https://doi.org/10.1177/096739111602400715

Wang, W., Sain, M., & Cooper, P. A. (2006). Study of moisture absorption in natural fiber plastic composites. *Composites Science and Technology*, *66*(3), 379-386. https://doi.org/10.1016/j.compscitech.2005.07.027

Willett, J. L. (2009). Starch in polymer compositions. In J. BeMiller., & R. Whistler (Eds). *Starch (pp. 715-743)*. Academic Press. https://doi.org/10.1016/B978-0-12-746275-2.00019-7

Yoksan, R., Boontanimitr, A., Klompong, N., & Phothongsurakun, T. (2022). Poly(lactic acid)/thermoplastic cassava starch blends filled with duckweed biomass. *International Journal of Biological Macromolecules*, *203*, 369–378. https://doi.org/10.1016/j.ijbiomac.2022.01.159

Zhang, Y., Simpson, B. K., & Dumont, M. J. (2018). Effect of beeswax and carnauba wax addition on properties of gelatin films: A comparative study. *Food Bioscience*, *26*, 88–95. https://doi.org/10.1016/j.fbio.2018.09.011