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PRELIMINARY STUDY ON THE PHYSICAL AND MECHANICAL PROPERTIES OF TAPIOCA STARCH / SUGARCANE FIBER CELLULOSE COMPOSITE

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ABSTRACT
This study is to investigate the potential application of Tapioca Starch (TS) and Sugar Cane Fiber Cellulose (SCFC) as a green composite for disposable packaging food container. The experiment is started by preparing and characterizing the SCFC through various characterization tools, whereby the effect of various SCFC loading to the fabricated TS composites was then studied to find the best formulation for TS/SCFC composites. Various engineering properties for TS/SCFC composites were tested out to determine the mechanical and physical properties, beside the fracture morphology by using the optical microscope observation. Through the morphological view on the mechanical and physical testing fractured surfaces, it had significantly indicates the reinforcement role of various fiber loading into the resulted properties of produced composites. Although the results have shown good performance of composites for both mechanical and physical properties, but the adhesion between SCFC and TS matrix were not well attached. In overall, the development of this new material for food packaging application will provide great potential solution to the environmental friendly and safe packaging medium, either for food, consumer or environmental, as a whole.

Keywords: tapioca starch, sugarcane fiber cellulose composites, properties, disposable food packaging, fiber loading.

INTRODUCTION
This research is focused on the fabrication of two natural components that were combined as biodegradable polymer matrix composites for the application of disposable food packaging. The considerable interest in this research is to produce an alternative material by compounding tapioca starch and sugar cane fiber cellulose to replace the existing non-biodegradable plastic material in the market. By using sugar cane fiber cellulose (SCFC) as a bio based natural fiber and tapioca starch (TS) as biodegradable matrix material, the development of this product is based on natural green raw materials due to its characteristics of biodegradable, non-toxic and non-allergenic bio environmental resources. Sugar cane is used due to its properties as a natural filler reinforcement that has played an important role in enhancing the composites performance. By combining sugar cane with tapioca starch that acts as a matrix, it will give many advantages to the environment. This is due to the advantages of renewability, low density, and high specific strength as well as biodegradable and recyclable at a very reasonable cost [1]. Besides, it is expected to give the benefit to the environment due to non-degradable waste of plastic food packaging caused by uncontrolled solid waste disposal.

The fibers outstanding properties such as high specific strength and stiffness, impact resistance, flexibility, and modulus make them an attractive alternative over the traditional materials [2]. Specifically, towards the properties of sugar cane fiber cellulose, which includes good specific strengths and modulus, economical viability, low density and low weight that make them a promising reinforcement of choice by the industry. Thus, natural fiber, like sugarcane, that can be used as a replacement to the conventional fiber since the global environmental issues, have led renews interest in the development of bio-based industrial products [3].

MATERIALS AND METHODS
The raw materials used in this study are tapioca starch (TS) as matrix material, sugar cane fiber cellulose (SCFC) as the reinforcement material, and glycerol as plasticizer. By applying several characterization approaches, the SCFC properties were tested for density test, water absorption test, drying characteristic test, and the microscopic study at the first stage. Second, TS/SCFC composites were pressed by using hot compression molding machine. To select the best fabricated composites based on mechanical properties, the test such as tensile testing, flexural testing, and impact testing are carried out according to ASTM D638, ASTM D790, and ASTM D256, respectively. While for the physical properties evaluation, the water absorption test (ASTM D570-98) was carried out in order to get the suitability of produced composites for any commercial application. In addition, the thickness swelling test also is required as well as the morphological investigation against the mechanical and physical specimens by using the optical microscope.

SCFC drying profile study
The drying process is by using oven (MEMMERT UFB 400). The investigation of the SCFC drying behavior over the periods of time is required through the reading (taken by three times per day) of the weight and moisture profile changes of SCFC.

Water absorption

Water absorption test against SCFC fiber is used to determine the amount of water absorbed under the specified conditions. The samples were weighed before and after the water absorption process. The total percentage of weight is labeled as Ww, which refer to the percentage of water absorbed.

$$\% Ww = [(Wf - Wi) / Wi] \times 100 \quad (1)$$

Density measurement

The density measurement of SCFC was performed by using an Electronic Densimeter model MD-300S. The measurement was repeatedly done for five times for different sample of SCFC. Average value was then calculated.

Microscopy study

To measure the SCFC sizes and the morphological behavior of the fabricated composites, the optical microscope with Vis Pro software is used for the microscopy observation. This is also conducted for dimensional study against the length of each single fiber measured and microscopy study regarding the fractography of composites surface specimens to understand the reinforcement characteristic of the fabricated composites.

Compounding of TS / SCFC composites

In order to compound the TS, SCFC and glycerol, the formulations were divided into five main compositions of weight ratios such as 53:0:47, 50:3:47, 47:6:47, 44:9:47 and 41:12:47 of fiber loading addition. The weight percentage (wt %) was computed by using the following formula;

$$Wtp \% = [(Vp.\rho p) / (Vp.\rho p + Vf.\rho f)] \times 100 \quad (2)$$

$$Wtf \% = [(Vf.\rho f) / (Vp.\rho p + Vf.\rho f)] \times 100 \quad (3)$$

Where Wtp % and Wtf % is the weight percentage of the TS and SCFC; Vp represent the volume fraction of TS and Vf represent the volume fraction of SCFC. ρp and ρf is density of TS and SCFC, respectively.

Hot compression molding

The compounded TS/SCFC/glycerol composites were compressed by using hot compression molding machine. The compression period is about 10 minutes at 130ºC and the overall cooling period is about 5 minutes.

Tensile test

The tensile test was carried out in accordance to ASTM D638 by utilizing the Universal Testing Machine (UTM) model Instron. Three measurements were tested at 23±2ºC with 50 % of relative humidity and each obtained values is averaged. The dimension for this test is about 165mm x 19mm x 3.2mm, which are Type 1 specimens at a crosshead speed rate of 3.75 mm/min with a gauge length of 50 mm.

Impact test

Notched Charpy impact strength was performed in order to determine the Impact Strength (J/m²) values (as representing energy loss per unit specimen thickness) on an Impact Tester according to ASTM method of D256. Three measurements were tested at 23±2ºC with 50 % of relative humidity and each obtained values is averaged.

Flexural test

The flexural test was performed by using a three-point bending method in accordance to ASTM D790, utilizing the Universal Testing Machine UTM. The purpose of this test is to measure the flexural strength and flexural modulus (Young's Modulus) of the specimens. The load applied to the specimens is on two supports with constants speed of 2.5 mm/min and standard span length of 160mm.

Water absorption test

This study is to determine the water absorption behavior of TS/SCFC composites according to ASTM D570-98. Samples was prepared in the form of bar with dimension of 76.2 x 25.4 x 3.2 mm and dried in a vacuum oven at 50±5°C for 24 hours and then immersed in distilled water to get weigh to the nearest 0.001 gram. The value of Ww is an average value of three samples measurements. The percentage increase in weight from the solution is then calculated to the nearest 0.01 %, where %wf is the final percentage increase in weight of the tested sample.

$$\% wf = [(Ww - Wc) / Wc] \times 100 \quad (4)$$

Thickness swelling test

Thickness swelling test is interrelated to water absorption test. The thicknesses of the samples (3 samples) were recorded after 24 hours of soaking period by using a digital vernier caliper. The calculation of thickness swelling is as follows:

$$\text{Thickness Swelling} = [(T_f - T_i) / Ti] \times 100 \quad (5)$$

Where: T_i is the thickness of specimens (mm) before immersion and T_f is the thickness of specimen after 24 hours of immersion.

RESULTS AND DISCUSSIONS

Drying characteristic of SCFC

Drying characteristics is important criteria to determine the utilizing of any natural fiber for composites fabrication. In this study, the drying process was employed as to investigate the drying behavior and kinetic of moisture release from SCFC raw materials against the initial and final weights of fibers. Figure-1 shows the daily weight losses of the fibers. In overall, the percentages of weight losses from SCFC showed the fluctuating pattern up until 5^{th} days of observation, before plateau formation for another three following days.

In addition, to investigate the influences and correlation of moisture to the resulted morphology and properties of natural fiber, the study was also carried out against the rate of moisture moves in SCFC (that depends on relative humidity of surrounding air), the steepness of the moisture gradient, and the internal temperature of the SCFC. The higher the temperature of the SCFC, the faster moisture will move from the wetter interior to the drier surface. However, if the temperature is too high, strength reduction may occur and influenced to the mechanical properties of fabricated composites.

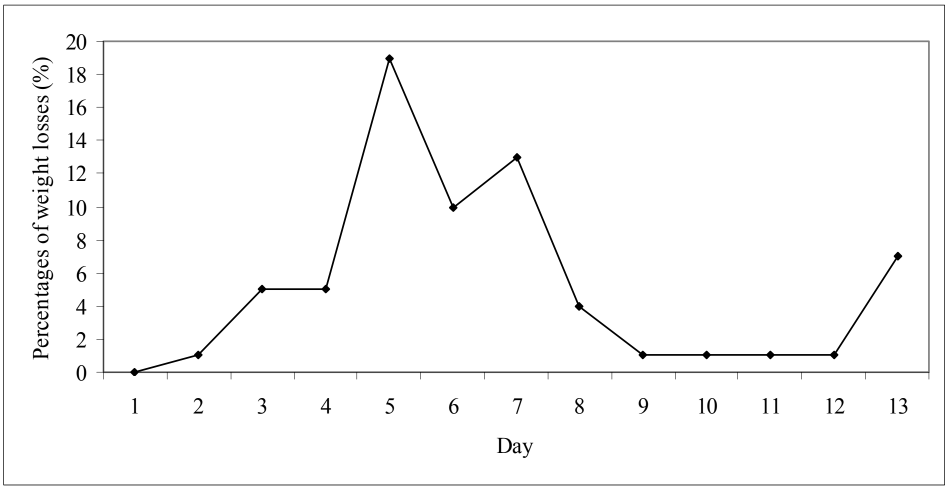

Figure-1. Percentage of weight losses for SCFC.

Water absorption behavior of SCFC

Understanding about the water absorption by SCFC during the soaking period is one of practical importance, since it will affects the mechanical properties of resulted product. The characteristic of water absorption for sugar cane fiber cellulose (SCFC) was determined through the samples measured against the weight, before and after the distilled water processing. Figure-2 shows that the SCFC natural fiber is very prone to absorb water. Due to water absorption, the average percentage of hydrophilic characteristic of SCFC (that is 87%) must be given higher consideration because it will greatly affect to the structural integrity of resulted composites. This is attributed to the natural capillaries present in the natural fiber, which is quickly attaining equilibrium with hydration medium by capillary inhibition [4]. Hence, it can be assumed that water concentration at the surface of natural filler is raised to saturation almost instantaneously.

The periodic water absorption has also give the negative impact on the quality of fabricated composites. The amount of absorbed water in fiber is depends on density and water diffusivity of fiber. Data obtained from water absorption or sorption behavior could provide good approximation of average liquid water diffusivity of the material [5, 6].

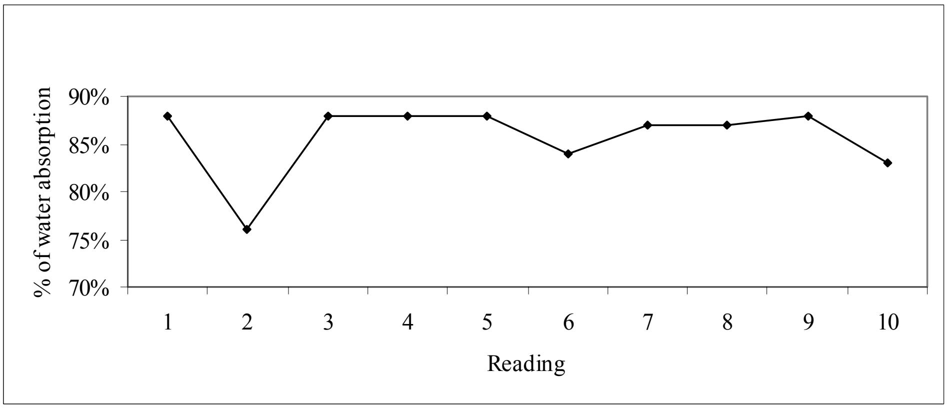

Figure-2. Water absorption of sugarcane fiber cellulose (SCFC).

SCFC density measurement

The density of sugar cane fiber cellulose (SCFC) was determined by using an electronic densimeter. The following Figure-3 shows the comparison of density measurement for other natural and industrial commercial fibers with sugar cane fiber cellulose (SCFC). SCFC shows the lowest density compared to other fibers such as coir, jute, aramid, carbon, flax, sisal, soft wood craft and cotton. Based on these results, it can be declared that the SCFC is very promising to be used for any light weight application due to its lowest density value than other type of fiber.

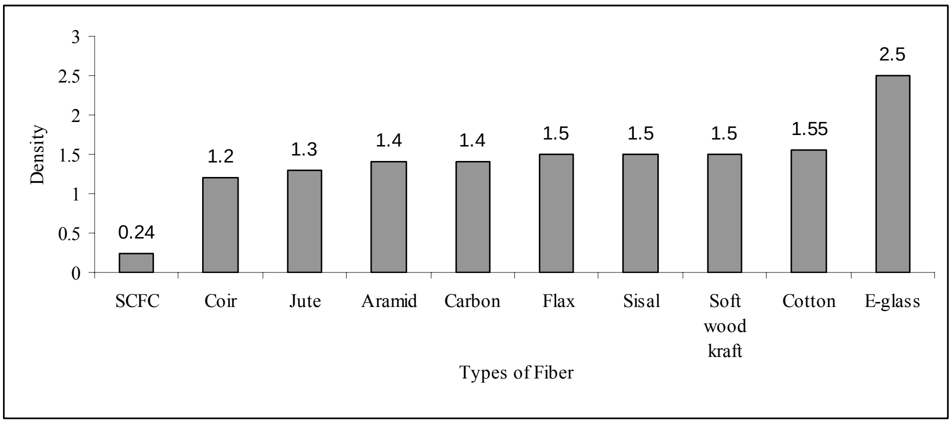

Figure-3. Comparison of density measurement with other fibers.

Microscopic observation of SCFC

In this study, the SCFC were crushed into the smaller length by using a rotor mill machine that caused the fibers lengths varied into wide range of size, within 890.13 to 2881.46 microns. This parameter will influence the result of the final properties of the fabricated composites. The fiber length is critical factor that need to be considered due to its necessity for effective strengthening and stiffening of the composites material [7]. Figure-4 shows the picture of morphology of sugar cane fiber cellulose (SCFC) by using the optical microscope.

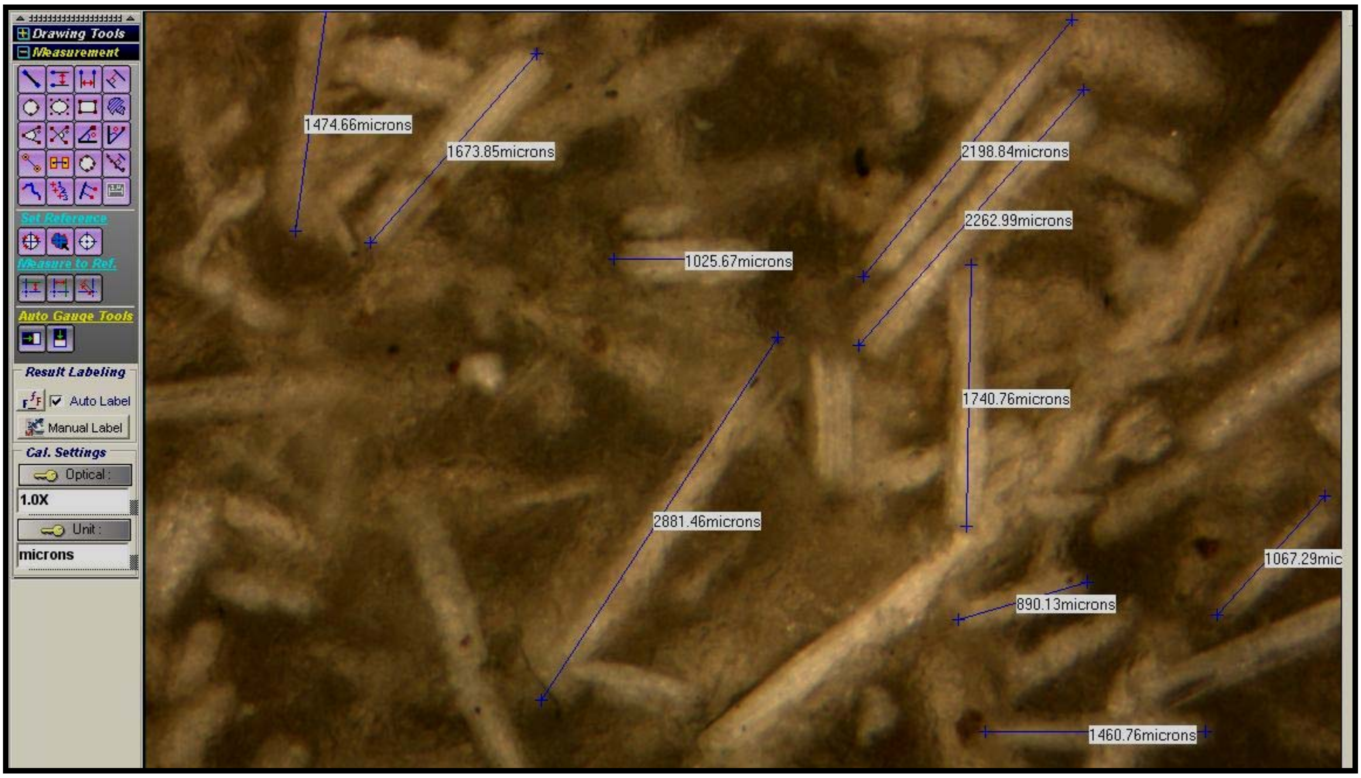

Figure-4. Morphology of sugar cane fiber cellulose (SCFC) at 20x of magnification.

Tensile properties of TS/SCFC composites

The fabricated composites samples were tested with tensile test in order to obtain the tensile strength and tensile modulus by following the ASTM D638 testing standard. However, the value of the maximum stress and Young's modulus for each composites formulation cannot be obtained from the testing due to early failure phenomena at the sample grip. This is due to the nature of soft sample texture which caused early damages during the gripping stage. However, this is contradicted to

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Sirikhajornnam and Danwanichakul (2006), that in their study for the preparation of biodegradable film from starch, tensile strength is about 21MPa until 35MPa for various composites formulation. They stated that, a film with higher ratio of glycerol (which is typical plasticizer) is poorer in its tensile strength, but show better flexibility [8]. This is because the plasticizers increased the film flexibility by reducing hydrogen bonding between the polymer chains.

Impact properties of TS/SCFC composites

The Charpy impact test was done as to show the ability of specimens to absorb the energy of pendulum force. All of the specimens did not break off due to their soft nature. These samples had been categories as non-break failure or totally ductile failure. The impact energy results of the fabricated composites samples were shown as in the following Table-1. The impact energy and impact strength are proportionally increased with the increasing of SCFC loading and glycerol percentages addition. The impact strength data for TS/SCFC/Glycerol composite produced in this research was integrated in the Figure-5, as to compare the resulted properties with the other fiber type's composites [9].

The impact response of the fiber composites is highly influenced by the interfacial bond strength between the matrix and the fiber. The impact energy is dissipated by debonding fiber or matrix fracture and fiber pull-out [10]. In this study, the failure mechanism was not obtained and cannot be fully understood because of the soft nature of the fabricated composites. Figure-6 depicts the surface observation for TS/SCFC composite which undergone the impact test.

Table-1. Impact properties of pure TS and TS/SCFC composites with the presence of glycerol.

Composition of TS/SCFC/glycerol (Wt %)	**Impact energy** (J)	**Impact strength** (J/m²)
53/0/47	0.17	0.0034
50/3/47	0.27	0.0062
47/6/47	0.47	0.0092
44/9/47	0.50	0.0096
41/12/47	0.67	0.0145
Average of impact strength		0.0086

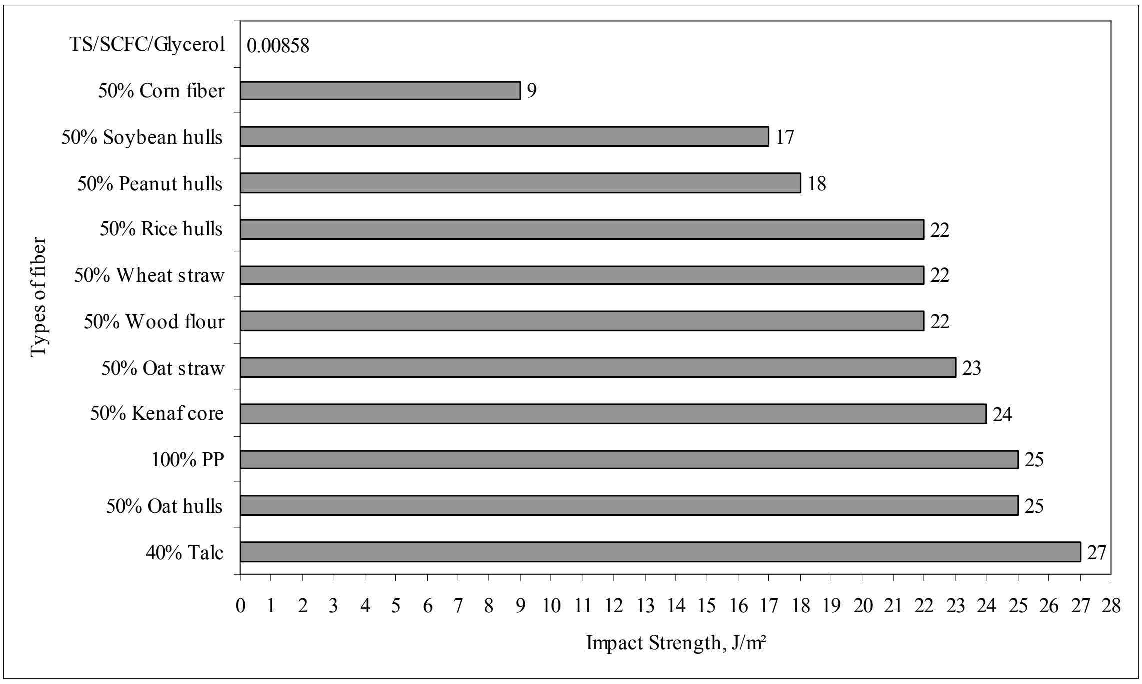

Figure-5. Charpy impact properties of various fibers (adapted from [9]).

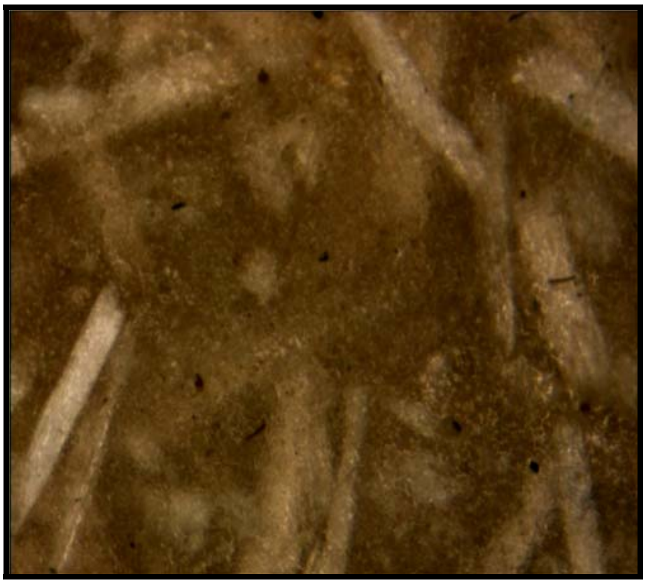

Figure-6. The surface of TS/SCFC composite with 41% tapioca starch, 12% SCFC and 47% glycerol at the 20x of magnification.

Flexural properties of TS/SCFC composites

The flexural test measures the required force to bend a beam under the three point loading conditions in accordance to ASTM D790 standard. Flexural modulus is used as an indication of a material stiffness. The increasing of flexural modulus results with the filler loading increment showed that the movement of tapioca starch (TS) molecules was more difficult with the increasing of filler content. This trend is consistent with the study conducted by [9] and [11]. Figure-7 shows the results of flexural modulus test for five compositions of the TS/SCFC composites.

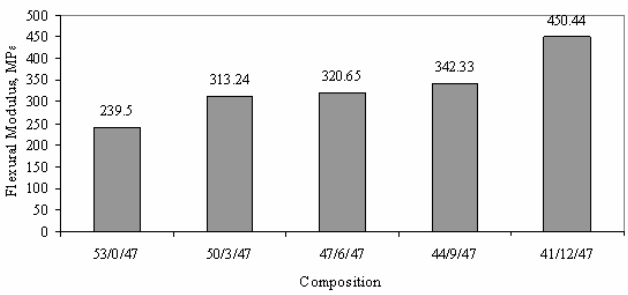

Figure-7. Flexural modulus of pure TS and TS/SCFC composite with the presence of glycerol.

The composites with random distribution of fibers indicate a better performance due to the possibility of homogenous distribution between the fiber and matrix. The stress transferred between the fiber and matrix is more effective and will positively affect to the flexural performance of the fabricated composites. The introduction of glycerol as a plasticizer is also influences to the improvement in the flexural properties for better flexibility. This was also reported by [8]. This is because the plasticizer increased the ability to reduce hydrogen bonding between the polymer chains. However, it cannot be clearly observed the fractured region of the specimens through the optical microscope observation. Although, TS and SCFC was strongly bonded by the glycerol and resulting the increment values of the flexural strength of the fabricated composites. The addition of glycerol gives significant improvement to the sugar cane fiber surface wetting by the tapioca starch matrix. As a result, addition of glycerol will contributes to the formation of stronger adhesion between the SCFC and the TS matrix in composites. The increase of the flexural properties in the composites can be explained via the increasing adhesion between the fiber and its matrix.

Water absorption properties of TS/SCFC composites

Figure-8 shows the percentage of water absorption versus five different TS/SCFC composite specimens which immersed in water at the ambient temperature (23°C). With the addition of sugar cane fiber cellulose (SCFC), the weight of each specimens increases after being immersed in water for 24 hours. This indicates the increasing percentage of water absorption with the increment of SCFC fiber loading that contributes to the negatives effects in term of structural integrity to the fabricated composites. The highest water absorption was in the specimen contained of 12wt% of SCFC with the presence of 41% of tapioca starch and 47wt% of glycerol whereas the formulation 53/0/47 and 50/3/47 of composites absorbed the very minimal amount of water at about 51% of absorption. A similar trend was previously obtained shows that the water absorbability depends to the size of filler particle [8]. The water absorption of composites is relatively high due to hygroscopic nature of the filler [12]. As the filler content is increased, the water absorption is expected to be increased [13]. In case of starch, the water absorbability might be depending to the percentage of fibers that has higher value of water solubility than the other materials [14].

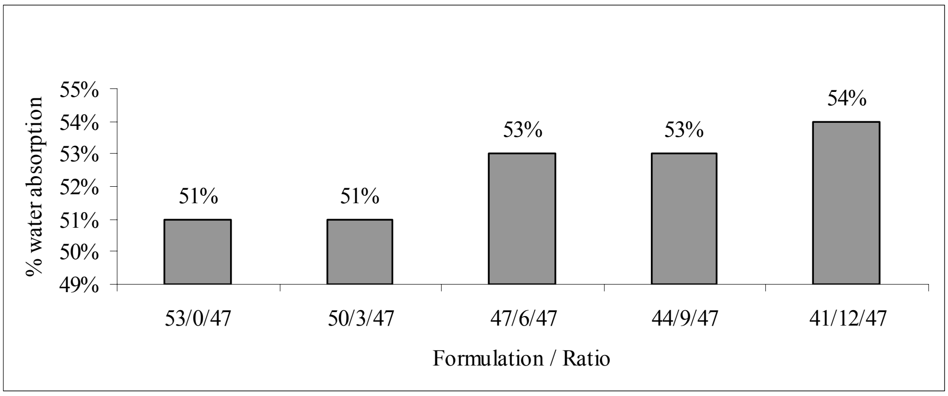

Figure-8. Water absorption characteristic of TS/SCFC/Glycerol composites at different composition.

The action of water may have resulted in further disruption of the interfacial adhesion between the starch granules and sugar cane fiber cellulose. Subsequently, these will probably lead to the formation of additional voids in the starch based composites, which would then be filled with water (water entrapment in the matrix). Upon drying, these voids will act as stress concentration region, which could then initiate matrix cracking that leading to reduction in both stiffness and strength of the composites. At high water absorption, the interfacial bond strength is reduced and affects both stiffness and strength of the samples [13]. Another factor that is noted to affect the properties from high water sorption is the effect as fibers swell to accommodate the water, resulting in internal stresses or cracks and greater interaction between micro fibrils. Flexural strength and modulus to drop with immersion time and filler content, where the drop in modulus properties was noted to be more significant than the strength properties [13]. This is due to droopiness occurred in intermolecular bonding of the filler.

The following Figure-9 shows the comparison of water absorption characteristics of TS/SCFC/Glycerol with the other natural fibers. It is clearly shown that the TS/SCFC is among the lowest in term of its water absorption properties, in comparison to the types of natural fiber based composites. From this result, we can expect that this bio composites material would be suitable for application like interior panels and food packing material [15]. The rate of water absorption in natural fiber based composites shows two distinct regions; a short term, where the kinetic absorption is noted to be relatively fast, and a long term where the kinetics of absorption is noted to be slow and leads to plateau regions.

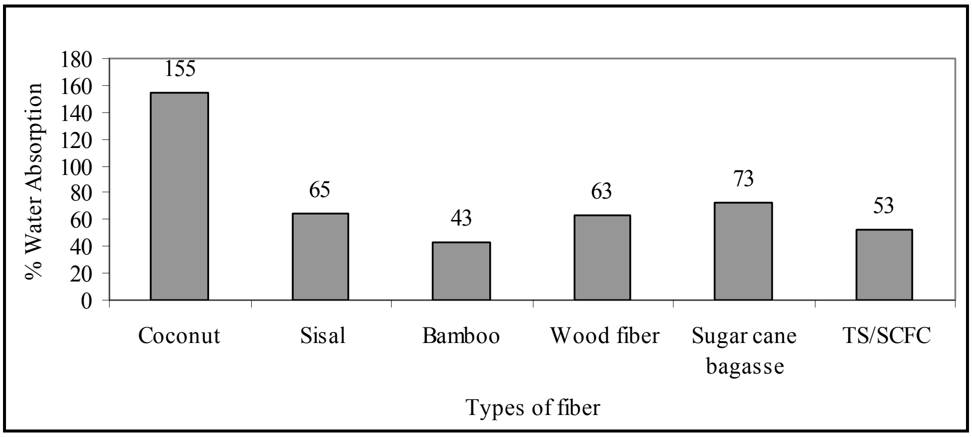

Figure-9. Water absorption characteristic of different types of fiber (adapted from [7]).

Thickness swelling properties of TS/SCFC composites

Thickness swelling testing was employed concurrently with water absorption procedure. Figure-10 shows the increment of thickness swelling of the specimens by increasing the fiber content. Previous study found that the percentage of thickness swelling is increased as the increment content of fibers used in the fabricated composite [16]. The highest percentage value of the thickness swelling was obtained in the specimen of 12wt% SCFC whereas the 53wt% of tapioca starch with the 47wt% glycerol specimen absorbs the very minimal amount of water at around 2% and caused the swelling at 0.09%. Thickness swelling of bio composites occurs when the cell walls in the agro-flour is swelled by water [17]. The larger particle size results in increased water absorption compared to the smaller particle size and, therefore, to greater swelling of the cell wall than in the case of smaller particle size.

Figure-10. Comparison of thickness deflection TS/SCFC/Glycerol composites at different composition.

CONCLUSIONS

The results shown that the mechanical properties of fabricated composites had increased by increment of SCFC content, especially for flexural and impact properties. However, poor performance was obtained to the resulted tensile strength properties. For the physical test, the result showed bad performance on both water absorption and thickness swelling test. This is due to both of the tests showed the increasing absorption and swelling trend by increment of the fiber loading in the resulted composites. Through the morphological study on the mechanical and physical testing of the fractured surfaces, it was clearly found that the adhesion between SCFC and TS matrix is not well adhered. The best compounding formulation of the fabricated composite was observed at the weight fraction of 41 wt% of TS, 12 wt% of SCFC with 47 wt% of glycerol.

The study proves that combination of TS/SCFC/Glycerol could have possibilities to be applied as one of the candidates' alternatives materials for food packaging application. However, further major study is still required to establish and improves the necessary properties for the application of food disposable packaging container.

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