

# Sayote (*Sechium edule*) Fiber Isolated at Varying Acid Hydrolysis Time and Reinforcement to Starch/PVOH Composite Blends

Jennifer Bandao-Antonio,<sup>a,\*</sup> and Jose Mario A. Diaz<sup>b</sup>

The chemo-mechanical extraction of sayote (*Sechium edule*) fibers and their use as reinforcement to biodegradable starch/polyvinyl alcohol composite blends were studied. Fourier transform infrared analysis revealed the removal of hemicelluloses from the fiber surface after 7 and 10 h of acid hydrolysis time. Scanning electron micrographs show the removal of surface impurities during chemical-mechanical treatment. There was a more exposed fiber surface after 7 and 10 h of acid hydrolysis time. However, fibers acid hydrolyzed for 10 h revealed the presence of more cracks on the fiber surface. X-ray diffraction analysis showed that 7 h acid hydrolyzed fiber had the highest relative crystallinity index of 64.9% as compared to the fiber that was acid hydrolyzed for 10 h with a relative crystallinity index of 58.6%. Both 7 h and 10 h acid hydrolyzed fibers gave 20% yield after extraction. The fiber that was hydrolyzed for 7 h was used as reinforcement to starch/polyvinyl alcohol composite and gave a bending and tensile strength of 5.36 MPa. The unreinforced composite gave a bending and tensile strength of 2.85 MPa. The scanning electron micrograph of the reinforced composite revealed a more homogeneous surface and lesser starch granule exposure as compared to the unreinforced composite with a rough and bumpy surface. The onset of degradation and carbonization of the fiber reinforced composite was seen at around 280 and 580 °C respectively.

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*Keywords:* Sayote (*Sechium edule*) nanofiber; Starch/PVOH composite blend; Chemo-mechanical treatment; Acid hydrolysis

*Contact information:* a: Department of Physical Sciences, College of Science, University of the Philippines Baguio, 2600, Baguio, Philippines; b: Department of Chemistry, School of Science and Engineering, Ateneo de Manila University, Loyola Heights, 1108, Quezon City, Philippines;

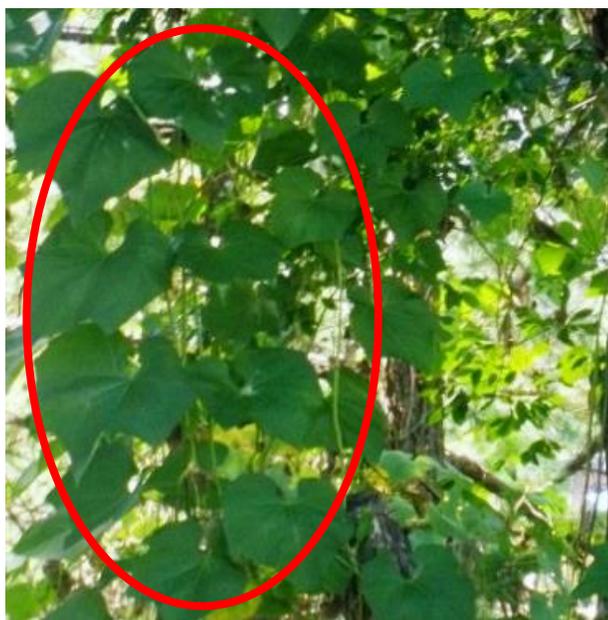
\* Corresponding author: jbantonio4@up.edu.ph

## INTRODUCTION

Sayote vine is found in every corner of Baguio City in the Philippines and its neighboring areas in the Cordillera Administrative Region (CAR). It is a tropical perennial vine that can reach as high as 12 m. The root stock is tuberous. The leaves are hairy, broadly cordate, and angled. The leaf petioles are 15 cm long. Staminate flowers are yellow to pale green with a length of 12 to 20 cm. Pistillate flowers are solitary with a corolla of 12 to 17 mm. Its fruit is the most sustainable food produced throughout the year. The cultivated plant can reach a lifespan of 20 years with proper application of fertilizers. The sayote plant can also survive in empty spaces and stonewalls even without proper nurture. It can also climb up a tree as high as 12 m, as shown in Fig. 1.

Biodegradable composites are being developed in response to an increasing awareness of the negative impacts of non-biodegradable products to the environment. Fiber

reinforcement is used to improve the mechanical properties of some biodegradable composites, since it was observed in previous studies that the fabrication of biodegradable composites alone showed poor mechanical properties. The use of natural fibers as reinforcement gives researchers and material scientists high hopes on the possibility of fabricating materials that are cheap and have comparable mechanical properties to that of petroleum-based composites. Natural plant fibers are cheap, abundant, lightweight, and are strong reinforcement to biodegradable matrices. The presence of hydroxyl (-OH) groups on the fiber enables the formation of a very strong patterns of intermolecular hydrogen bonds between fiber and biodegradable matrix, making fiber reinforcement possible. The strong bonding interaction between the cellulosic fiber and biodegradable polymers such as starch and polyvinyl alcohol (PVOH) makes the fiber improve the properties of the composite blend.



**Fig. 1.** A sayote plant (encircled) up an *Alnus* tree

The fabrication of composite materials using biodegradable materials such starch/PVOH with fiber reinforcement can find applications to the food bio-packaging industry. There is too much waste material generated from non-biodegradable food packages that piles up in our land and oceans. Studies on the assembly of novel materials from the environment such as fibers and biopolymers could help solve the environmental impacts of using petroleum based polymers.

### Fiber Extraction Studies

Studies on the isolation of plant fibers considering the acid hydrolysis time were done by some researchers. The fiber crystallinity index (CI) was also investigated as an indication of a successful isolation of micro- and nano-crystalline cellulosic fiber from some plants. The study on the alkaline treatment, bleaching, and hydrolysis of banana fiber (Elanthikkal *et al.* 2010) resulted in the removal of pectin, hemicelluloses, and lignin after extraction. However, the increase in acid concentration caused loss of fibrous character due to surface etching and erosion. In another study, the acid hydrolysis of chardonnay grape skins processed with 64% to 65% H<sub>2</sub>SO<sub>4</sub> concentration at 45 °C for 30 min was performed.

Cellulose nanocrystals with a crystallinity of 64.3% were formed that appeared mostly as spherical nanoparticles with diameters ranging from 10 nm to 100 nm (Lu and Hsieh 2012). The isolation of cellulose micro- and nano- fibers using acid hydrolysis furnished cellulose fibers as smaller size aggregates and smaller diameter with increased hydrolysis time (Frone *et al.* 2011a). Cellulose nanofibers from wood, bamboo, wheat straw, and flax were isolated by chemical treatments. Nanofiber diameters ranging from 10 nm to 40 nm were produced after ultrasonic fibrillation. There was a partial removal of lignin and hemicelluloses during chemical process, and the CI of the fibers were found out to be 60% (Chen *et al.* 2011). Whiskers from sugarcane bagasse (Teixeira *et al.* 2011) at a mild temperature of 45 °C and at a shorter extraction time of 30 min were also done. The extracted nanowhiskers had a length around 255 nm and an average diameter of 4 nm, a thermal stability of 255 °C, and crystallinity at 87.5%. The sugarcane bagasse whiskers obtained at longer extraction time were found to exhibit damaged cellulose structure. Thus, they exhibited decreased thermal stability. Cellulose microfibrils were also produced from coconut palm leaf sheath (Maheswari *et al.* 2012), and it was found that the extracted microfibrils had higher cellulose content and lower hemicelluloses and lignin than the original leaf sheath. Most of the lignin and hemicelluloses were removed during the chemical process, and the extracted cellulose was thermally stable up to 300 °C. Alkali and bleaching treatments of rice husks were performed, and it was found that alkali treatment was efficient in removing hemicelluloses while bleaching was able to remove lignin. The chemical treatments increased the CI from 46.8% to 59% of the extracted fiber and decreased the diameter from 170 μm to 7 μm (Johar *et al.* 2012). The successful isolation of nanocrystals from mengkuang leaves was also studied. Upon alkali bleaching and H<sub>2</sub>SO<sub>4</sub> treatment, the cellulose content, thermal stability, and CI were increased. The final CI was found to be 69.5% (Sheltami *et al.* 2012). Fibers from oil palm empty fruit bunches were also extracted using 20% formic acid and 10% hydrogen peroxide, which yielded 64% of cellulose with α-cellulose content of 93.7% (Nazir *et al.* 2013). The chemical method of extraction caused a total removal of waxes, lignin, and hemicelluloses from the fiber and the crystallinity of the extracted cellulose was 70%. The effective extraction of cellulose from agave fibers (Rosli *et al.* 2013) by 4% NaOH treatment to remove hemicelluloses and 1.7% NaClO<sub>2</sub> to remove lignin were also performed. The nanocellulose was finally isolated by using acid hydrolysis. The NaOH and NaClO<sub>2</sub> treatments were found to reduce the fiber diameter from 230 μm to 7 μm and improved thermal stability. The CI of the extracted agave nanofibers was 82%.

### Composite Fabrication Studies

Several research studies have already been done on the use of plant fibers for composite fabrication. A study on starch, sugarcane bagasse fiber, and polyvinyl alcohol effects on extruded foam properties (Mali *et al.* 2010) resulted in findings where a decrease in the compression strength of starch/PVOH under 90% relative humidity was recorded. However, the addition of fibers became significant. Fibers acted as a reinforcing agent since they did not interact with water at high relative humidity. Scanning electron micrographs also revealed homogeneous surfaces with the addition of sugarcane bagasse fibers. The investigation on the physico-mechanical properties of the jute micro/nanofibril reinforced starch/PVOH biocomposite films (Das *et al.* 2011) resulted in a conclusion that the jute nanofibrils were good reinforcement to starch/PVOH. The incorporation of jute nanofibrils decreased the interchain force of attraction and increased the % strain value. The PVOH composite with cellulose nanofiber that was obtained by ultrasonication was

previously reported (Frone *et al.* 2011b). The tensile strength and modulus of the material were increased with the addition of small concentration of cellulose fibers due to the strong hydrogen bonding of the OH- groups of cellulose fibers with the matrix. Thermal stability of the resulting material was also increased. Wang and Li (2015) reported a high reinforcing efficiency of cellulose nanofiber from peanut shells in PVOH. The addition of cellulose nanofibers produced an optically transparent, thermally stable, and flexible biocomposite. Mechanical properties such as tensile strength also increased when fiber content was up to 80 wt%. Heidarian *et al.* (2017) conducted an investigation on starch and PVOH biocomposites and the effect of cellulose nanofibril from aspen wood sawdust. The cellulose nanofiber at 5 wt% was seen to be well distributed in the PVOH/starch composite. Mechanical properties, moisture absorption, biodegradability, and barrier properties of the cross-linked composite were all improved on the addition of the wood fiber. The incorporation of cellulose nanofibers extracted from sisal into glycerol-plasticized cassava starch-based films was found to be feasible in the production of nanocomposites with improved mechanical, barrier, and thermal properties. The Young's modulus and maximum tensile force were increased by the cellulose nanofiber addition and increasing concentration (Santana *et al.* 2017).

In light of findings reported by various researchers, this study was conceptualized. Initial experimentation and characterization were done on the chemo-mechanically extracted sayote fibers. The more mature and sun-dried sayote vines were chosen for isolation based on their known desirable properties such as fewer amorphous structures and fibrous and orderly features. The isolated sayote fiber was used to reinforce the starch/PVOH composite blend.

It was the aim of this study to produce a material with good mechanical properties from starch/PVOH composite reinforced using sayote (*Sechium edule*) fiber.

## EXPERIMENTAL

### Preparation of Fibers from Sayote Vine

Sayote (*Sechium edule*) vines were collected from Baguio City, Philippines. The mature vines were sun dried for 5 d before chemical-mechanical treatment. Figure 2 shows the mature and sun-dried sayote vine, it was the best sought material for extraction and blending with starch and PVOH.

The mature and sun-dried sayote vines were chemo-mechanically treated using the procedure described in Fig. 3. Before acid hydrolysis, the sayote vines were pre-treated with NaOH for 3 h. Then the acid hydrolysis time for the extraction of the fiber was varied to identify the best parameter for fiber isolation. Two groups of sayote vines, 200 g each, were subjected to 7 and 10 h acid hydrolysis time. After acid hydrolysis, the fibers were treated with NaOH for 2 h and H<sub>2</sub>O<sub>2</sub> for 12 h. The chemo-mechanically treated fibers were subjected to a high-pressure defibrillation for 1 min using an Oster Osterizer (Sunbeam, Florida, USA) blender. The defibrillated fiber was ultrasonicated for 5 h. The final product was washed 10 times with distilled de-ionized water to neutralize the pH. The fibers were frozen for 1 d then dried for 2 d using a SIM International Freeze Dryer (SIM International Group, Beijing, China) at -86.4 °C and 2 mTorr. The yield for both 7 and 10 h acid hydrolyzed fibers was 20% after extraction.



Fig. 2. Mature and sun-dried sayote vines for extraction

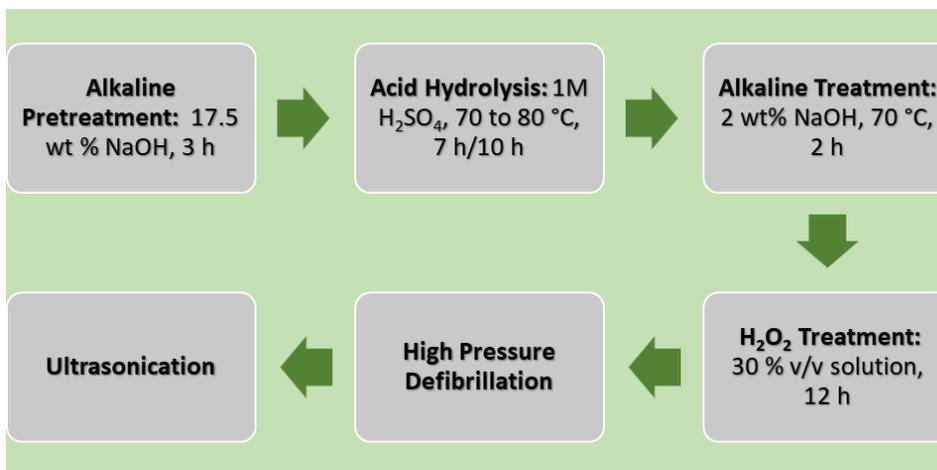


Fig. 3. Chemo-mechanical treatment of sayote fiber

### Preparation of Fiber Reinforced and Unreinforced Starch/PVOH Blends

Two types of samples were prepared for mechanical testing. One sample was fiber reinforced and another sample without fiber reinforcement. The sayote fiber used for reinforcement was the sample acid hydrolyzed for 7 h, since it had a higher crystallinity index and lesser surface cracks, and should provide better reinforcement. Initially, 57.50 g PVOH was gelatinized with 1.45 mL glycerol and 60 mL water for 30 min at 50 °C. Then 62.50 g of starch was plasticized with 1.58 mL glycerol and 65 mL water for 30 min at 100 °C with constant stirring. The gelatinized PVOH and plasticized starch were mixed

with a stirrer for 30 min at 100 °C. Next, 5 g of sayote fibers were added to the mixture and was sonicated for 1 h. The blend was placed in a petri dish and was dried in a Gallenkamp oven (London, United Kingdom) at 40 °C for 12 h.

The sample without fiber reinforcement was prepared using the procedure described above except for the addition of 5 g sayote fiber. The starch/PVOH fiber reinforced and unreinforced composite blends were placed in a desiccator for one week before characterization. The masses of starch and sayote fibers were weighed using an analytical balance with an uncertainty of 0.0001 g.

### Characterization of Extracted Fiber at Varied Processing Conditions

The Fourier transform infrared (FTIR) spectra were obtained using a Nicolet 6700 FTIR (Kyoto, Japan) spectrometer to determine the functional groups present in the fiber. A 1 mg sample was mixed with KBr in a 1:10 ratio and was mounted in a KBr disc. The transmittance was measured at 100 scans from 400 to 4000  $\text{cm}^{-1}$ .

The scanning electron microscopy (SEM) micrographs obtained from Hitachi TM-1000 (Tokyo, Japan) was used to determine the surface morphology of the extracted fibers. The fibers were gold sputtered for 1 min before scanning at different magnifications.

The X-ray diffraction (XRD) diffractograms were obtained using Siemens Kristalloflex 760 X-ray generator (Munich, Germany) with copper x-ray tube (wavelength = 1.54056Å) and Philips 1080 vertical goniometer (Amsterdam, Netherlands) at a high voltage of 34 kV and 20 mA current was used to determine the crystallinity index (CI) of the extracted fibers. The fibers were ground to form a very fine substance so that all atomic planes were randomly represented before placing the sample in the sample holder. The samples were scanned using Cu  $K\alpha$  radiation at  $2\theta$  (°) at a range of 5.0° to 85° at a scanning rate of 0.02°.

The CI was computed using the peak-height method or Segal method, where the peak intensity of the amorphous portion was subtracted from the peak intensity of the crystalline portion divided by the peak intensity of the crystalline portion times 100% in Eq. 1,

$$CI = \frac{I_{(002)} - I_{(am)}}{I_{(002)}} \times 100 \quad (1)$$

where CI is the crystallinity index,  $I_{(002)}$  is the peak intensity of the crystalline portion, and  $I_{(am)}$  is the peak intensity of the amorphous portion.

### Characterization of Fiber Reinforced and Unreinforced Starch/PVOH Blends

The SEM micrographs were obtained using the Hitachi TM-1000 to determine the surface morphology of the extracted fibers. The fibers were gold sputtered for one minute before scanning at different magnifications.

The tensile and bending strengths were determined with the use of a Tinius Olsen (0-2000 lbs capacity) (Pennsylvania, USA) low-capacity universal testing machine. Samples were cut into 1.27 × 10.16 cm rectangular shaped composites and were subjected to tensile and bending tests. Trials were done in triplicate.

Thermogravimetric analysis was performed using a Shimadzu TA 50 (Kyoto, Japan) device. The sample weight was about 5 to 10 mg. Samples were heated from 20°C to 800°C at a heating rate of 10°C/min. All experiments were carried out in a liquid nitrogen atmosphere at a flow rate of 50 mL/min.

## RESULTS AND DISCUSSION

### Extracted Fiber

The FTIR analysis on the non-acid hydrolyzed fiber, 7 h acid hydrolyzed fiber and 10 h acid hydrolyzed fiber were done to compare the functional groups that were removed from the fiber surface in the process of chemo-mechanical treatment.

**Table 1.** FTIR Summary for Sayote Macro/microfiber

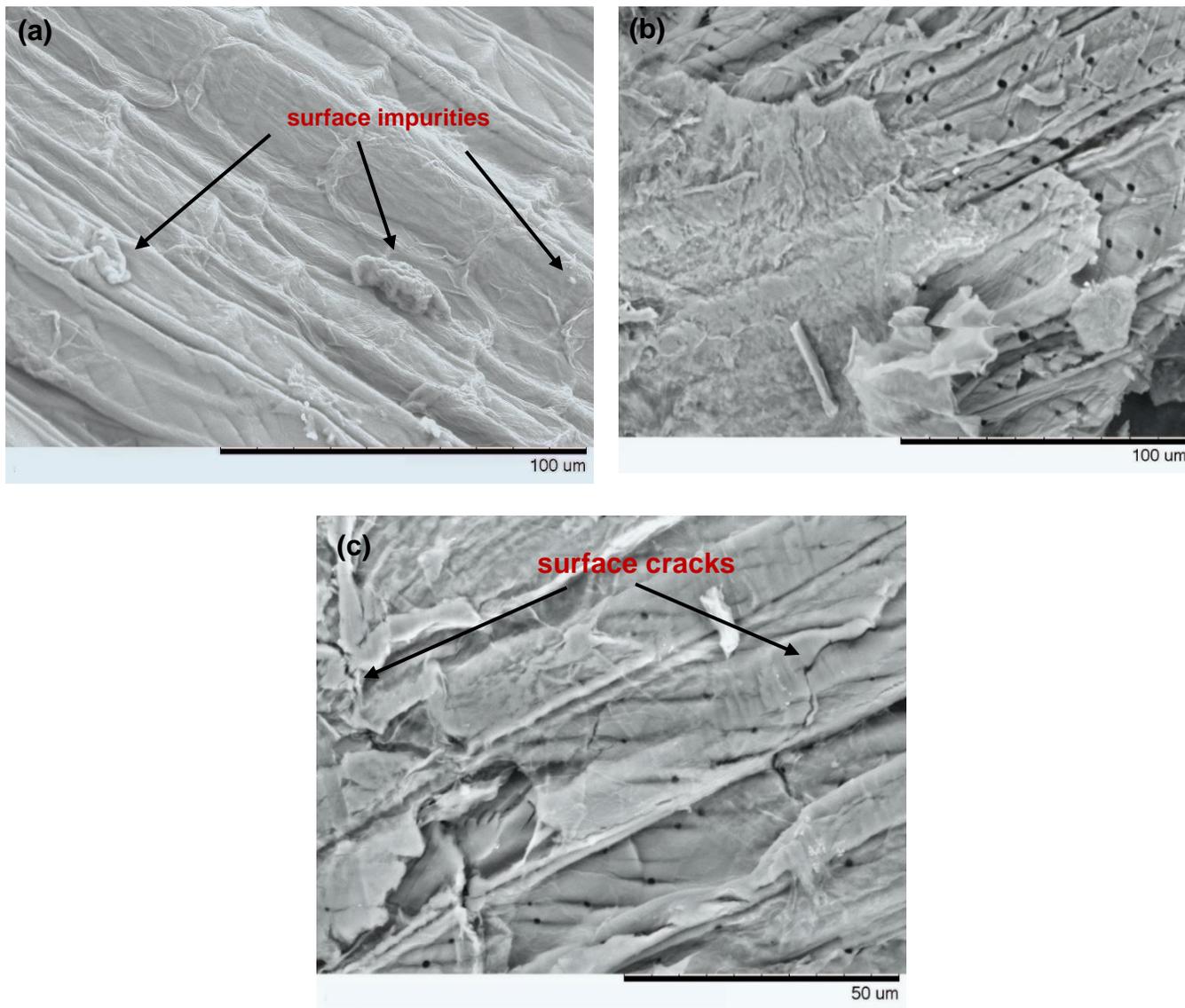
Characteristic	Non-acid Hydrolyzed	7 h Acid Hydrolyzed	10 h Acid Hydrolyzed
OH Stretching	3411.47	3421.18	3414.68
CH Symmetrical Stretching	2920.24	2919.89	2919.35
Aromatic Stretching of Lignin	1201.04 (C-O-C Symmetric Stretching) 1232.21 (COH bending at C6) 1266.49 (Gring stretching)	1201.04 1225.97 1266.49	1232.21 1266.95
C=O Stretching of Lignin and Hemicelluloses	1729.53	None	None
C-C,C-OH,C-H Vibration	1063.79	1061.18	1062.21

Table 1 shows the summary of the FTIR spectra for sayote fibers prepared using different hydrolysis time. The table shows the removal hemicelluloses and partly of lignin after chemo-mechanical treatment. The peaks at 1201.0, 1232.2, and 1266.5  $\text{cm}^{-1}$  were characteristic of the aromatic stretching of lignin (Maheshwari *et al.* 2012). The peak at 1201.0 for C-O-C symmetrical stretching in the aromatic ring of lignin disappeared in the fibers that were acid hydrolyzed for 10 h. This means that at 10 h acid hydrolysis time, fragmentation of the aromatic ring of lignin starts.

The peak at 1730.9 was characteristic of C=O stretching of the acetyl and uronic ester linkage of carboxylic groups for the ferulic and p-coumeric acids of lignin and hemicelluloses (Abraham *et al.* 2011). It was present on the non-acid hydrolyzed fiber but was not present on the fiber extracted after 7 and 10 h of acid hydrolysis. This means that lignin bonds were destroyed due to the cleavage of the  $\alpha$ - and  $\beta$ - ether linkages and the weaker hemicelluloses was completely degraded.

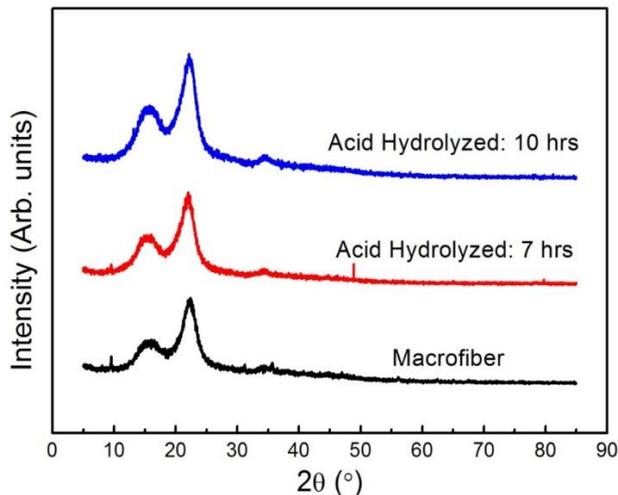
Figure 4 shows the surface morphology of the non-acid hydrolyzed fiber and the chemomechanically treated fiber that were acid hydrolyzed for 7 and 10 h. The presence of impurities was seen on the fiber surface in Fig. 4a. The extracted fiber for 7 and 10 h acid hydrolysis time showed more exposed surface. This was attributed to the removal of hemicelluloses, lignin, and other substances or impurities present on the fiber surface. The more exposed surfaces shown in Figs. 4b and 4c were very much favorable to intermolecular interaction with biodegradable polymers. However, it can be noted that the

fiber acid hydrolyzed for 7 h exhibited lesser cracks on the fiber surface as compared to the fibers acid hydrolyzed for 10 h.



**Fig. 4.** SEM micrograph of sayote: (a) non-acid hydrolyzed fiber, (b) fiber acid hydrolyzed for 7 h, and (c) fiber acid hydrolyzed for 10 h at 1000 X

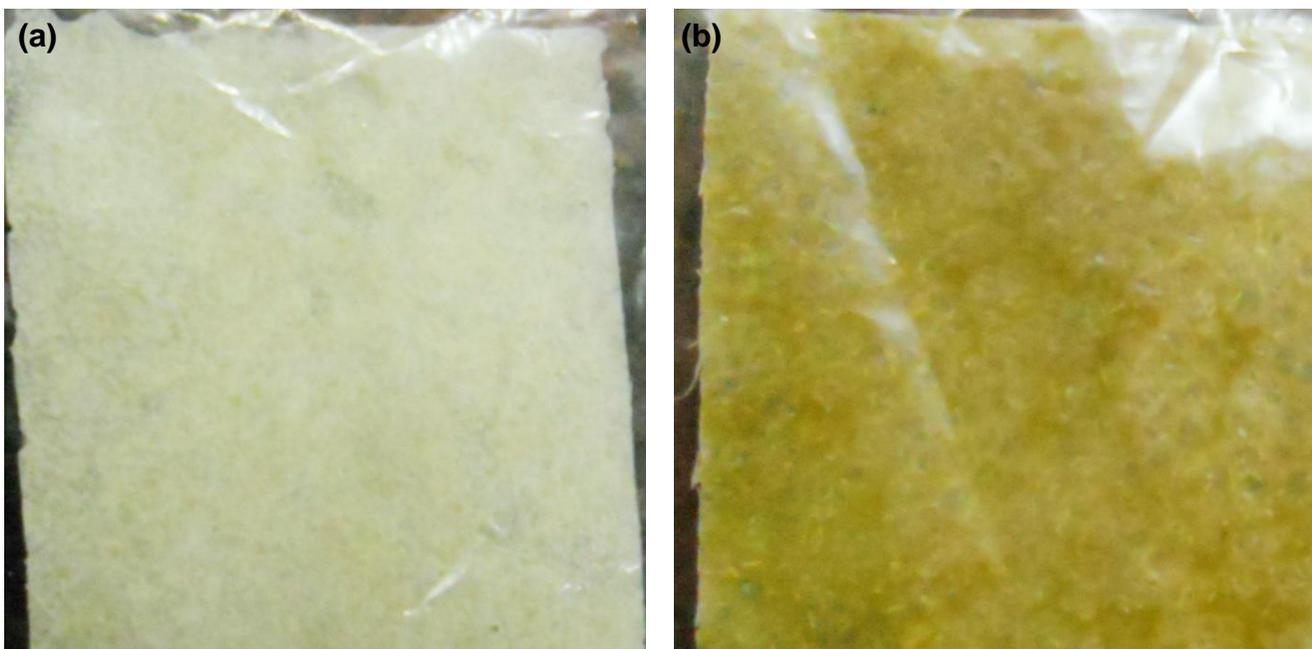
The XRD study showed the crystallinity of the extracted fibers. Comparing the crystallinity index of the fiber not subjected to acid hydrolysis with the 7 h acid hydrolyzed fibers, there was an increase in the relative crystallinity index from 60.0% to 64.9% (Fig. 5). This means that the removal of hemicelluloses, lignin, and other impurities on the fiber surface exposed the more crystalline portions of the fiber. Looking at the crystallinity index of sayote fiber which was acid hydrolyzed for 10 h, the crystallinity index at 58.6 was lower than that of the fiber acid hydrolyzed for 7 h. This is attributed to the presence of cracks as seen on the SEM micrograph of sayote fibers acid hydrolyzed for 10 h. The presence of cracks may have been responsible for the decrease in the crystallinity of the fiber structure.



**Fig. 5.** XRD diffractogram for macrofiber without acid hydrolysis: CI-60.02%, fiber acid hydrolyzed for 7 h: CI-64.89%, and fiber acid hydrolyzed for 10 h: CI-58.65%

### Sayote Microfiber Reinforced Starch/PVOH Composite

The 7 h acid hydrolyzed fiber was selected as reinforcement in the fabrication of the composite due to its higher crystallinity and fewer cracks. Figure 6 shows the unreinforced and fiber reinforced starch/PVOH composite blends. For the reinforced material, Fig. 6b differed in color with that of the unreinforced blend. The sayote fiber reinforced blend was golden in color due to the presence of the fiber reinforcing material in the blend.



**Fig. 6.** Starch/PVOH composite blend: (a) unreinforced material and (b) reinforced with sayote fiber

### Tensile and Bending Strength Tests

Table 2 shows the tensile and bending strength tests for starch/PVOH composite blends with and without microfiber reinforcement. It can be seen that fiber reinforcement

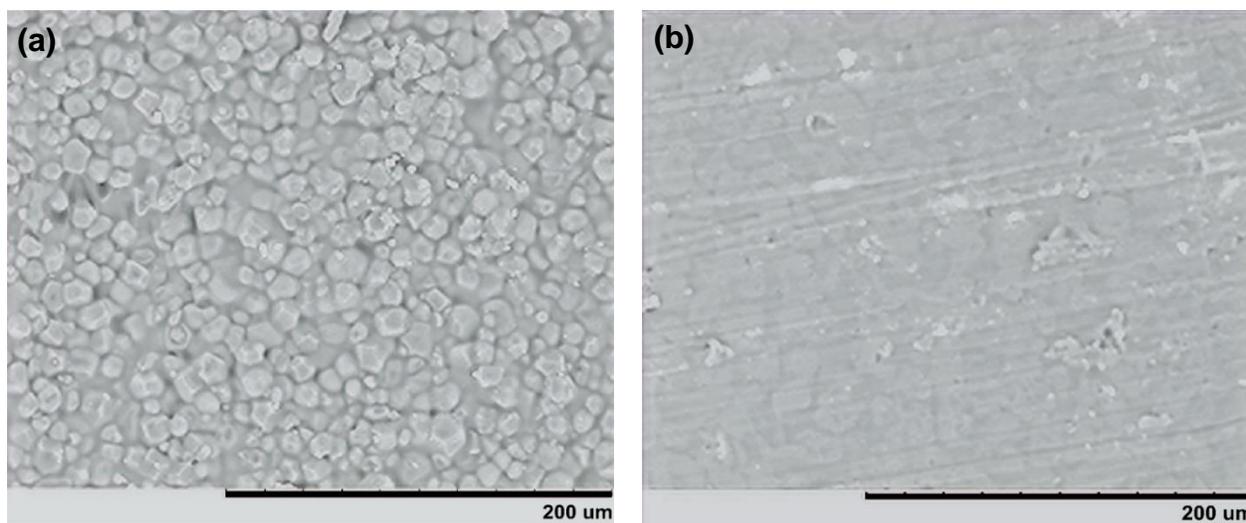
to starch/PVOH composite blend significantly increased the tensile and bending strength up to 53%. This was due to the reinforcing effect of sayote fibers, the fibers were able to increase the intermolecular and intramolecular interaction between starch and PVOH, thus improving their mechanical properties in terms of tensile and bending strengths.

**Table 2.** Tensile and Bending Strength Test Results for Composite Blends with Accompanying Standard Deviations in the Measurements

Composite Blend	Tensile Strength (MPa)	Bending Strength (MPa)
Plasticized starch/PVOH	2.85 (0.50)	2.85 (0.74)
Plasticized starch/PVOH/sayote fiber	5.36 (0.36)	5.36 (0.76)

### SEM Analysis

Figure 7 reveals the SEM micrographs of fiber reinforced starch/PVOH composite blends. The surface of unplasticized starch/PVOH composite blend shows a rough surface with exposure of the granular structure of starch. The plasticized starch/PVOH also shows a rough surface with some cracks indicating a brittle material with poor mechanical property. However, the addition of sayote microfiber to plasticized starch/PVOH revealed a more uniform surface and the granular structure of starch was not exposed because the microfibrils were able to cause a stronger intermolecular bonding between starch and PVOH.



**Fig. 7.** SEM micrograph of (a) unreinforced starch/PVOH composite blend and (b) sayote fiber reinforced starch/PVOH at 1000 X

### Thermogravimetric Analysis

The thermal degradation of the starch/PVOH samples was affected by the incorporation of the extracted sayote fibers. The first stage of thermal degradation was attributed to the loss of volatiles, H<sub>2</sub>O, and glycerol at 75 to 200 °C. The second stage was due to the main degradation zone of starch and PVOH, which is due to the dehydration of hydroxyl groups and formation of low molecular weight unsaturated and aliphatic carbon species. The third stage is attributed to carbonization at 500 °C (Salgado *et.al.* 2008).

Figure 8 shows the degradation curve for the starch/PVOH and starch/PVOH/fiber composite blends. For starch/PVOH blend, the first degradation step, which was attributed to loss of volatiles, H<sub>2</sub>O, and glycerol, was seen at around 270 °C, while it was at around 280 °C for the fiber reinforced composite blend. This means that fiber reinforcement increased the onset of thermal degradation of the composite blend. The second degradation step, which was attributed to the main degradation zone of starch and PVOH for both unreinforced and fiber reinforced composite blends, was seen at around 370°C. The third degradation rate attributed to carbonization was at around 500 °C for starch/PVOH composite blend and was found to increase to around 580 °C for the starch/PVOH/fiber composite blend. This indicates that fiber reinforcement delayed the carbonization of the composite blend.

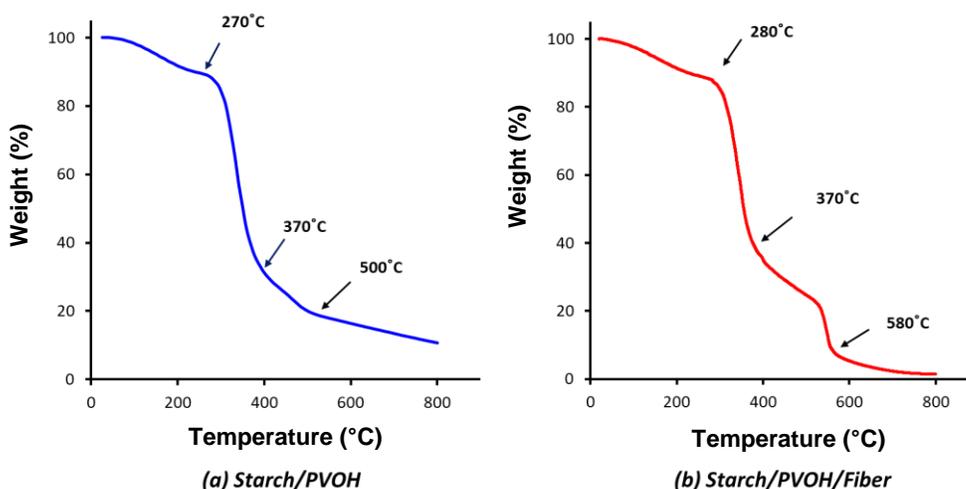


Fig. 8. TGA curves for (a) starch/PVOH and (b) starch/PVOH/fiber composite blends

## CONCLUSIONS

1. Sayote fibers were prepared using the chemo-mechanical extraction at 7 and 10 h acid hydrolysis time. Both acid hydrolysis time resulted to 10% yield of the extracted fiber. Hemicelluloses and lignin were seen to have been removed from the fiber structure. The 7 h acid hydrolysis time produced a more desirable fiber due to the lesser presence of surface cracks and higher crystallinity index.
2. Acid hydrolysis time is one of the important parameters that needs to be manipulated in the isolation of fiber from plants since longer exposure can lead to surface cracks that decrease the crystallinity of the isolated fiber.
3. A more exposed fiber surface is desired, since the OH- groups in the fiber crystalline structure are good for inter-molecular and intra-molecular hydrogen bonding with biodegradable matrices, thus increasing the fiber's reinforcing property. The extracted sayote fiber was able to reinforce the starch/PVOH composite, as evidenced by the increase in tensile and bending strengths when compared to the unreinforced composite.
4. The more crystalline sayote microfiber, that is acid hydrolyzed for 7 hours, was used in the starch/PVOH composite fabrication. Sayote microfiber was found to be a good

reinforcing material to starch/PVOH matrix since it increased the tensile and bending strengths of the composite up to 53% and was able to form a more homogeneous surface with lesser exposure of the granular structure of starch.

5. Fiber reinforcement to biodegradable matrices such as starch and PVOH showed potential of cellulosic plant fibers in the development of mechanically desirable and environment-friendly materials that can be used in the food bio-packaging industry.
6. Fiber reinforcement to starch and PVOH delayed the onset of thermal degradation and the carbonization of the bio-material.

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