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Synthesis of Water Hyacinth/Cassava Starch Composite as An Environmentally Friendly Plastic Solution

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ABSTRACT. Conventional plastics made from petroleum polymers were the largest contributor to solid waste on earth. Environmentally friendly bioplastic fabricated by compositing starch and natural fibers were proposed to be a solution to this problem. The purpose of this research was to synthesize bioplastic from water hyacinth fiber composite with cassava starch and test its mechanical properties. Bioplastic fabrication was using melt intercalation method using water hyacinth fibers (WHF) with and without alkali treatment mixed with cassava starch (CS) and glycerol as plasticizer. The resulting bioplastic were characterized with FT-IR and tested for mechanical properties. The mechanical test results showed that water hyacinth fiber alkaline treated/cassava starch (WHF-AT/CS) bioplastic has tensile strength, % elongation, and water absorption values of 1.226 MPa, 3.33%, and 10.26%, respectively. While the bioplastic water hyacinth fiber untreated/cassava starch (WHF-UT/CS) has a tensile strength test value, % elongation, and water absorption of 0.306 MPa, 1.67%, and 11.39, respectively. Therefore it can be said that WHF-AT/CS bioplastic has better mechanical properties when compared to WHF-UT/CS bioplastic

1. INTRODUCTION

Today it is undeniable that in our daily lives we are not far from using conventional petroleum-based plastics as food and non-food packaging. Behind the economical price, light weight, and easy to find, there is a serious threat to the ecosystem and the environment. The consumptive lifestyle of today's society has resulted in the accumulation of waste generated, especially conventional plastic waste. Its nature, which is difficult to be degraded by microorganisms, makes conventional plastic the largest contributor to solid waste [1]–[3]. Based on the United Nations Center for Regional Development (UNCRD) report, Indonesia is ranked second in terms of plastic waste mismanagement, with 88% of plastic waste ending up in the waters [4]. Various efforts have been made to overcome the problems caused by conventional plastics, one of which is bioplastic.

Bioplastic is a type of plastic made from natural materials that can be decomposed by microorganisms and does not leave toxins so that it is considered more friendly to the environment [1]. Several studies have been conducted to produce starch-based bioplastic [5]–[7]. Indonesia has great potential to use starch as a raw material for making bioplastic. This is because there are various starch-producing plants, such as cassava, corn, potatoes, sago, and yams. However, starch-based bioplastic have several disadvantages, including low mechanical properties and lack of water resistance. Therefore, it needs to be fabricated into a composite by adding fillers.

The addition of fillers is intended to produce high mechanical properties, such as tensile-strength, elongation, elasticity, and resistance to water and heat [8]. Filler in the manufacture of bioplastic composites can be inorganic or organic materials. One of the fillers made from organic materials is natural fiber. Natural fibers can be obtained from plant parts, namely bark/stem, leaves, fruit, grass/vine, and wood [9], [10].

Natural fibers generally contain cellulose, hemicellulose, lignin, as well as other accompanying impurities. Cellulose is the main component that gives strength to the fiber [11], [12]. The presence of impurities such as lignin, hemicellulose, oil, and waxy substances affects the interfacial interaction between natural fibers and starch. This hinders the adhesion force between hydroxyl groups of cellulose and hydroxyl groups of starch. Thus, it is necessary to treat natural fibers that will be composited with starch, one of which is alkaline treatment [13], [14].

A number of researchers have reported success in compositing starch with cellulose. Yang et al. [15] have fabricated bioplastic from oil palm empty fruit bunch cellulose with cassava starch and have a tensile-strength

value of 1.99 MPa and a water absorption of 19.95%. Jumaidin et al. [16] successfully synthesized bioplastic from cogon grass cellulose with cassava starch and had a tensile strength value of 5 MPa and a water absorption of 16.61%. Asrofi et al. [17] also reported that sugarcane bagasse cellulose bioplastic and cassava starch have a tensile strength value of 2.5 MPa and a water absorption capacity of 26.09%.

Water hyacinth is one of the most abundant aquatic plants in Indonesia. This plant is often considered a weed due to its rapid growth and often disrupts water transportation traffic. Behind the problems caused, water hyacinth has the potential as a filler because its cellulose content is quite high, which is around 61% [10], [14]. Based on this, this article will review the results of composite fabrication from water hyacinth fibers treated and without alkali treatment with tapioca starch as an environmentally friendly plastic solution. The selection of cassava starch is based on its abundant availability, low price, and environmental friendliness. The main contents of cassava starch are amylose (15%) and amylopectin (85%).

2. MATERIALS AND METHODS

2.1 Materials

Water hyacinth was obtained from the Kedukan River, Palembang Indonesia. Water hyacinth fiber (WHF) was obtained by removing the leaves and roots of water hyacinth, leaving only the stems. Next, wash the water hyacinth stems and knit them into small pieces. Then crushing the clean water hyacinth and transferring it into a tray and then drying it in the sun until dry, called water hyacinth fiber untreated (WHF-UT). Local commercially produced Cap Pak Tani brand of cassava starch (CS) was used as a matrix for the bioplastic. Other chemical reagents used were sodium hydroxide (p.a, Merck Millipore), acetic acid (p.a, Merck Millipore) glycerol (food grade), distilled water from the Integrated Laboratory of UIN Raden Fatah Palembang.

Figure 1. (a) wet WHF, (b) dried WHF, and (c) Cap Pak Tani brand of CS

2.2 Preparation of Water Hyacinth Fiber

The dried WHF were soaked into a 2,5%, 5%, and 10% NaOH solution at 40°C for 2 hours. It was then neutralized with acetic acid and washed with water. The fibers were then dried in the sun until dry, called water hyacinth fiber alkaline treated (WHF-AT). WHF-UT and WHF-AT were then characterized using Fourier Transform Infrared (Bruker, Alpha Series).

2.3 Fabrication of WHF/CS Bioplastic Film

CS was dissolved in distilled water (500 grams: 3000 cc) and then heated to boiling. Next, 2 mL of glycerol and WHF were mixed into it slowly with stirring (ratio of CS:WHF = 60:40). After homogeneous composite was molded and dried in the sun until dry.

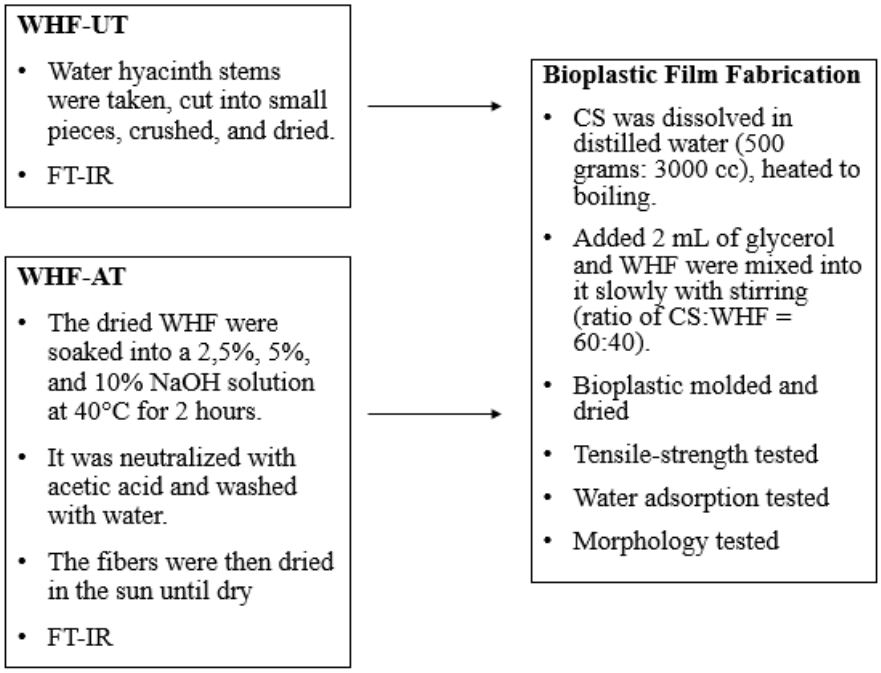


Figure 2. Experimental design

2.4 Mechanical properties testing

Testing of the mechanical properties of bioplastic, including tensile strength and elasticity. Bioplastic samples were cut with a size of 12 cm x 1 2m then test was conducted using Hung Ta Instrument (HT-8172) with speed 55 mm/min.

Figure 3. Tensile-strength testing instrument (Hung Ta Instrument, HT-8172)

2.5 Water adsorption testing

Bioplastic samples were cut with a size of 1 cm x 1 cm and weighed first. Next, the samples were immersed

in water at room temperature for 24 hours. The sample was drained and weighed again. The percentage of water absorbed was calculated using the formula:

$$\%Wa=\frac{Wh-Wo}{Wo} \times 100\% \quad \ldots (1)$$

where Wh is weight of adsorb sample and Wo initial weight of dried sample.

2.6 Morphology of bioplastic film testing

Bioplastic samples were cut with a size of 1 cm x 1 cm. Furthermore, it was placed on a glass preparation and analyzed using an optical microscope (Video Digital Microscope HIrox RH2000).

3. RESULTS AND DISCUSSION

3.1 Water hyacinth fiber

Chemically, natural fibers contain the main components of cellulose, hemicellulose, and lignin. It is not uncommon to find impurities such as pectin, waxes, and water-soluble materials. Cellulose is the main component that gives strength to the fiber [11], [12]. Therefore, before being fabricated into composites, water hyacinth fibers are alkali treated to separate cellulose from other components. This can be confirmed by comparing the FT-IR spectra of water hyacinth fibers without and with alkaline treatment, as presented in Figure 4.

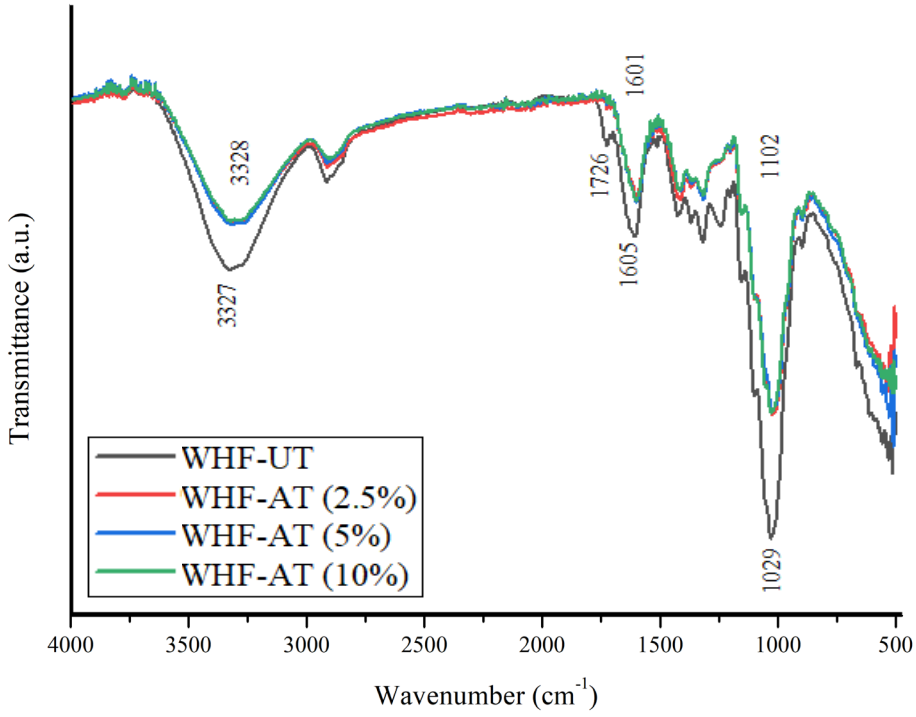


Figure 4. FT-IR spectra of WHF

The FT-IR spectra for WHF-UT shows an absorption at a wave number of 1726 cm^{-1} which indicates the presence of a C=O stretching of esters. In addition, there is also an absorption at wave number 1605 cm^{-1} which reinforces that the carbonyl group is near the aromatic ring. These typical absorptions indicate the presence of lignin. The absence of C=O stertching of esters absorption in the FT-IR spectra for WHF-AT confirms that delignification has occurred.

The absorption at wave number 3327 cm^{-1} for WHF-UT and 3328 cm^{-1} for WHF-AT indicates the -OH stretching. The absorption at wave number 1029 cm^{-1} in WHF-UT and 1102 cm^{-1} in WHF-AT indicates the C-O-C asymmetric stretching. In natural fibers, -OH and C-O-C groups are found in cellulose and hemicellulose. The increase in the transmittance value of the absorption of -OH and C-O-C stretching in the FT-IR spectra for WHF-AT indicates that hemicellulose removal has occurred.

Based on the confirmation of FT-IR spectra, it can be said that alkali treatment of water hyacinth fibers has delignified and removed hemicellulose. This is in line with the results of research [11]) that the absence of lignin

and hemicellulose is characterized by the disappearance of typical lignin absorption peaks at wave numbers 1715-1730 cm^{-1} and the increase in absorption transmittance value at wave numbers 3200-3350 cm^{-1}. Thus it can be said that cellulose in water hyacinth fiber has separated from other components.

3.2 WHF/CS bioplastic film

The FT-IR confirmation results of the bioplastic formed are presented in Figures 5 and 6. In the FT-IR spectra in Figure 5 there is an absorption at wave number 3295 cm^{-1} for the WHF-UT/CS bioplastic, while in Figure 6 there is an absorption at 3284 cm^{-1} for the WHF-AT bioplastic. This indicates the presence of -OH. When examined, the intensity of the absorption is higher when compared to WHF and CS. This indicates the addition of the -OH group.

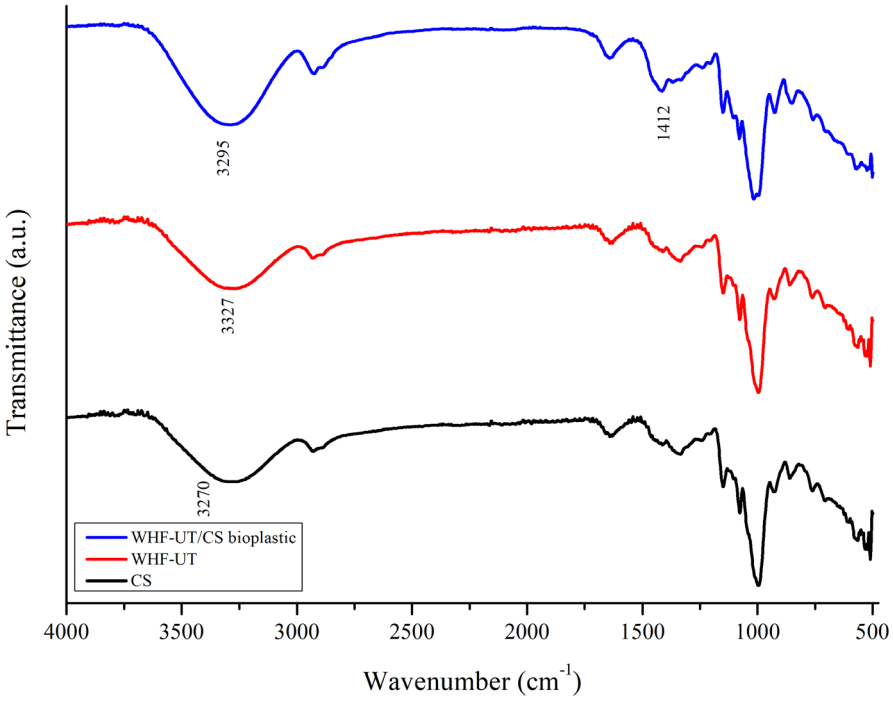


Figure 5. FT-IR spectra of WHF-UT/CS bioplastic

In addition, there is a shift in wave number to the right when compared to the absorption of the -OH group for WHF which indicates the formation of hydrogen bonds between the -OH groups of WHF and CS. In addition, the absorption at wave numbers 1412 cm-1 and 1424 cm-1 which is the -OH bending also strengthens the formation of hydrogen bonds. This is in line with the results of research by Edhirej et al. [18] that the interaction between natural fibers and starch occurs through hydrogen bonds which is indicated by a shift in wave numbers from 3330 cm-1 to 3280-3290 cm-1. The success of bioplastic synthesis is also indicated by the increase in absorption intensity at wave numbers 1000-1030 cm-1 which indicates C-O bending derived from WHF and CS.

Fabrication of WHF composites with SC resulted in sheets as shown in Figure 7. WHF-UT/CS bioplastic film are not evenly dispersed, while WHF-AT/CS is evenly dispersed. This difference is due to the adhesive interaction between WHF and CS which occurs through hydrogen bonds. Hydrogen bonds are formed in a good adhesive so that WHF-AT/CS bioplastic are evenly dispersed. While in WHF-UT/CS bioplastic, hydrogen bonds are formed in a bad adhesive due to the presence of lignin thus blocking the -OH group from WHF interacting with the -OH group from CS.

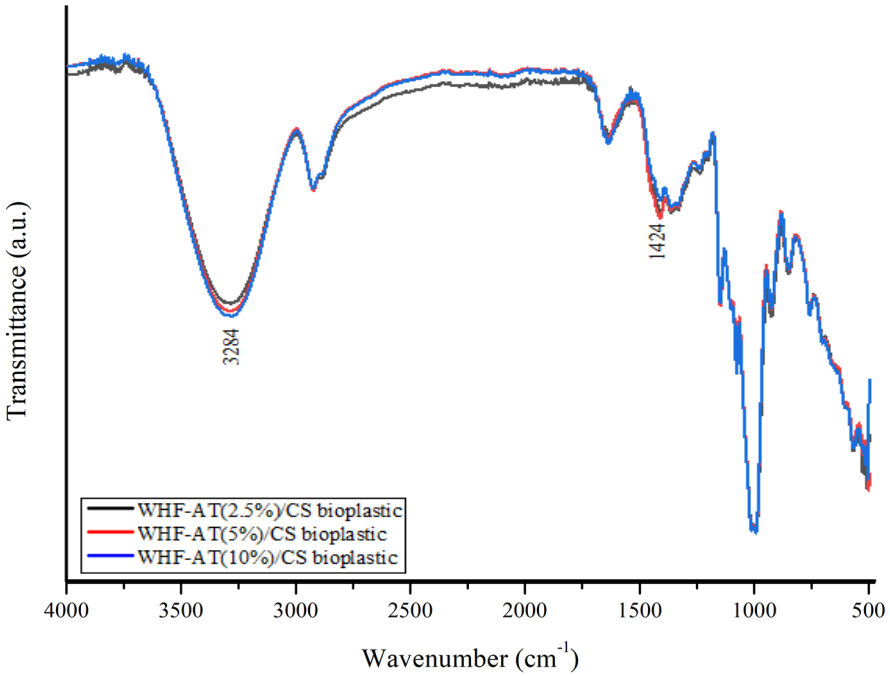


Figure 6. FT-IR spectra of WHF-AT/CS bioplastic.

WHF-UT/CS WHF-AT/CS

Figure 7. Bioplastic film

3.3 Mechanic Properties and water adsorption of WHF/CS bioplastic film

The mechanical properties tested on WHF/CS include tensile-strength and elongation as presented in Figure 8 and 9. Based on the data in Figure 8, the tensile-strength of WHF-AT/CS bioplastic increased four times when compared to WHF-UT/CS bioplastic. In addition, the concentration of alkaline in WHF soaking also affects the tensile strength of the bioplastics produced. The higher the alkaline concentration indicates that more lignin and hemicellulose are lost, so the interaction between cellulose in WHF and CS is getting better. This is what results in the higher concentration of alkaline used to soak WHF, the bioplastic tensile strength produced also increases. In other hand, the minimum tensile strength value criteria given by SNI 7818:2014 is 13.7 MPa. Thus, the resulting tensile strength value does not meet the SNI.

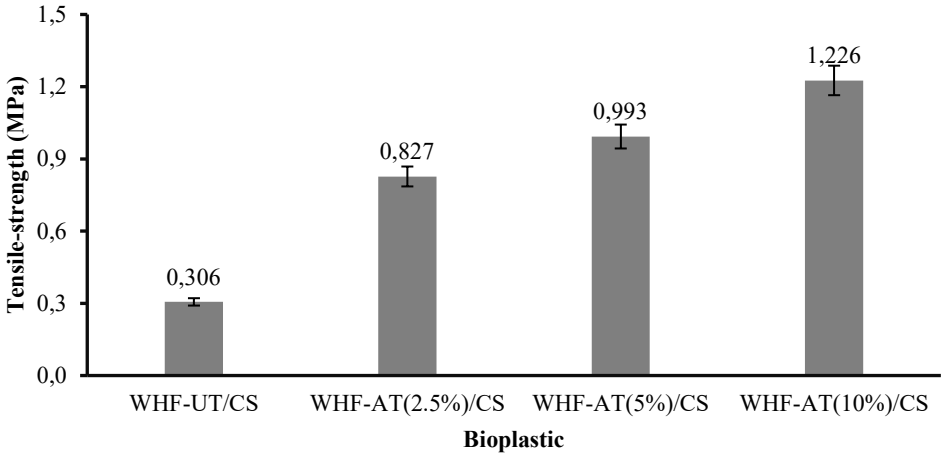


Figure 7. Tensile-strength of bioplastic film

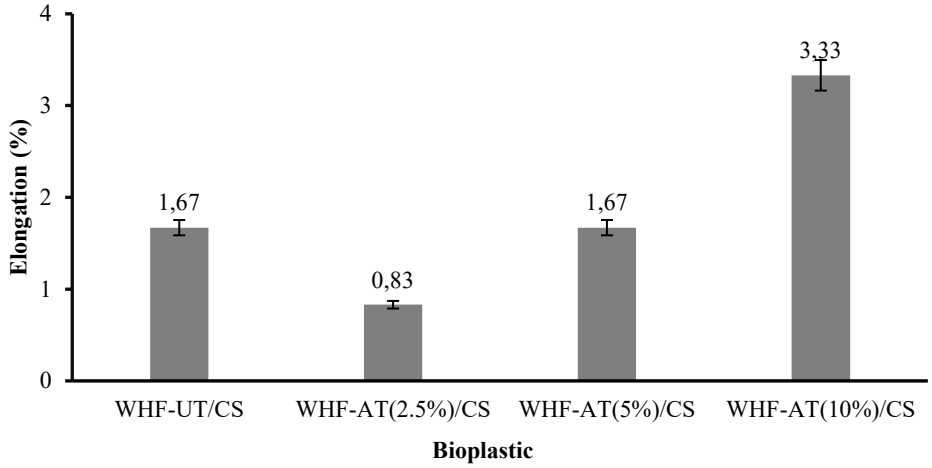


Figure 8. Elongation of bioplastic film

Likewise, the % elongation (Figure 9) is in line with the tensile strength value. The absence of lignin in WHF doubles the % elongation of the bioplastic produced. This indicates that the absence of lignin further strengthens the interface interaction between WHF in this case is cellulose with CS. On the other hand, in the presence of lignin, the tensile-strength and % elongation of bioplastic produced are low. This is due to the bad adhesive that occurs on the surface of WHF and CS.

One of the indicators of environmentally friendly plastics is to have the ability to absorb water at a certain limit, because the more easily absorb water causes the quality of bioplastic to decrease. Based on the data in Table 1, it can also be seen that the absence of lignin in WHF decreases water absorption. Hydrogen bonds on the surface of WHF-AT and CS cellulose are so strong that they make it difficult to combine with water molecules. According to Mastrolia et al. [1] environmentally friendly bioplastic are bioplastic that are physically and chemically degradable into biomass, CO2, and water.

Table 1. Water adsorption of bioplastic film

Bioplastic	Water Adsorption (%)
WHF-UT/CS	11.39
WHF-AT(2.5%)/CS	11.08
WHF-AT(5%)/CS	10.77
WHF-AT(10%)/CS	10.45

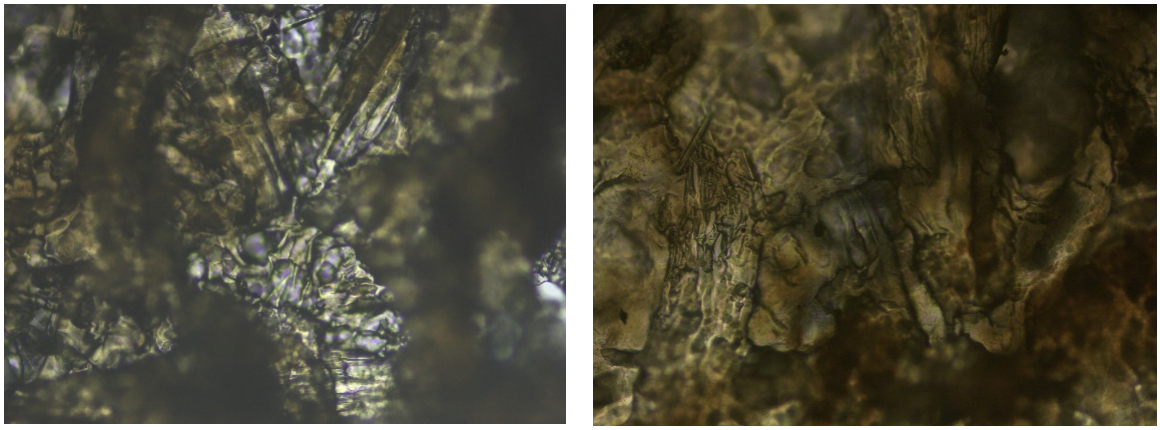
The mechanical properties comparison of WHF/CS bioplastics with similar bioplastics from CS as matrix and various fillers are presented in Table 2.

Table 2. Comparison mechanic properties of WHF/CS bioplastic film with similar CS-bioplastics

Bioplastic	Tensile-strength (MPa)	Elongation (%)	Water Adsorption (%)	Ref
WHF/CS	1.226	3.33	10.45	This Work
OPEFB/CS	0.021	16.36	40.2	[19]
Betel Fiber/CS	14.23	5.44	128.57	[20]
Banana Peel/CS	10.9	29.00	-	[21]
Sea Weeds/CS	4.69	1.92	-	[22]
Cassava Peel/CS	3.259	20.00	-	[23]
Palm Tree Fruit/CS	7.08	22.32	-	[24]
Bamboo Fiber/CS	0.069	-	-	[25]

3.4 Morphology of bioplastic film

Morphological observations of bioplastic were performed using an optical microscope. The results of four times magnification of bioplastic samples are presented in Figure 9.



WHF-UT/CS WHF-AT/CS

Figure 9. Morphology of bioplastic film

Based on Figure 9, it appears that the fiber in WHF-UT/CS bioplastic is quite thick. This may be due to the presence of impurities such as lignin and hemicellulose. Also visible are white cavities that are thought to be bad adhesive between WHF-UT and CS so that the resulting composite is not evenly dispersed. Unlike the WHF-AT/CS bioplastic. Fiber appears thin and there are no visible white cavities. There is good adhesive between the surface of WHF-AT with CS so that the resulting composite is evenly dispersed.

4. CONCLUSION

Bioplastic can be made by compositing WHF and CS. The resulting is bioplastic film. The absence of impurities in WHF such as lignin and hemicellulose has an impact on the mechanical properties of the bioplastic produced. WHF-AT/CS bioplastic have better mechanical properties compared to WHF-UT/CS bioplastic.

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