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Effect of Addition Elephant Grass Cellulose and $CaCO_3$ Oyster Shell Waste as Bioplastic Composites

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ABSTRACT. The effect of adding cellulose and $CaCO_3$ as a bioplastic filler was studied. The source of cellulose is obtained from elephant grass plants, while $CaCO_3$ is obtained from oyster shell waste. The primary raw material for bioplastics is tapioca starch with glycerol as a plasticizer using the solution casting method. The resulting bioplastics are thin and transparent but not very elastic, with a thickness is 1 mm. The mechanical properties test of bioplastics obtained tensile strength between 1-3 MPa and elongation between 1-4.4%. Physical properties test results obtained density between 0.313-0.33 g/mL and water absorption between 31.94-81.16%. The morphological test showed that the bioplastic surface was getting more uneven with more $CaCO_3$ filler. The use of cellulose fillers without the combination obtained better results than cellulose and $CaCO_3$ fillers.

Keywords: Bioplastic, $CaCO_3$, cellulose, elephant grass, oyster shell.

INTRODUCTION

It is difficult enough to stop excessive plastic usage in modern society. Global industrial plastic manufacturing hits 100 million tons per year (Sulistyono, 2016). Conventional plastics are often derived from petroleum and are not biodegradable; as a result, they harm the environment and become garbage. Thirteen percent of the plastic garbage gets recycled (Sulistyono, 2016). Thus, one of the solutions to this issue is the production of eco-friendly plastics (Setiawan et al., 2015).

Bioplastics can be made using several plant-derived materials such as starch, cellulose, lignin, and animal-derived materials such as protein, casein, and lipids (Darni et al., 2014). The choice of starch as a raw material for making bioplastics is because the extraction process is easy, the price is low, and its availability is abundant in Indonesia (Ernita et al., 2020). One type of starch that can be used is tapioca starch. Tapioca starch is non-toxic, biodegradable, inexpensive, and readily available in nature (Wahyuningtiyas & Suryanto, 2017). Tapioca starch has an amylose content of 12.28-27.38% and an amylopectin content of 72.61-87.71%. Amylose plays a role in the mechanical properties of bioplastics, while amylopectin gives sticky properties to the resulting bioplastics (Susanti et al., 2015).

However, tapioca starch as a material for making bioplastics still has drawbacks, especially mechanical properties. Therefore, many studies have been carried out to obtain better mechanical properties. One way to improve the mechanical properties of this tapioca starch-based bioplastic is by adding fillers. Filler is helpful for hardening or strengthening bioplastics. Fillers work based on the principle of adhesion, which is the attraction between molecules of different materials (Melani et al., 2017). Haryanto & Titani (2017) reported that the tensile strength of bioplastic from tapioca starch without filler was 0.37 N/mm2, and elongation was 19.41%. Susanti et al. (2015) reported the bioplastic tensile strength of tapioca starch with glycerin of 0.039 MPa and Young's modulus of 0.90 MPa. In another study, Syafri et al. (2017) reported that the tensile strength of tapioca starch-based bioplastics was 1.65 MPa without the addition of filler. Based on the tensile strength of these studies, it can be concluded that tapioca starch-based bioplastics need to be added with fillers to improve their mechanical properties.

Many fillers are often used in making bioplastics, including cellulose and $CaCO_3$. The cellulose used in this study came from elephant grass, while $CaCO_3$ came from oyster shell waste. Agustin et al. (2014) reported the results of tensile strength, percent elongation, and Young's modulus from CNC (Cellulose Nano Crytals) fillers on corn starch-based bioplastics of 10-26 MPa; 3.6-33.1% and 3.27-8.96 MPa. It can be known that cellulose can increase the tensile strength of bioplastics. However, at the percent elongation, the yield decreased with the increase in cellulose. In another study conducted by Syafri et al. (2017) using PCC (Precipitated Calcium Carbonate)

as a filler in tapioca starch-based bioplastics, the results obtained tensile strength, percent elongation, and Young's modulus of 1.65-3.38 MPa; 39.91-53.14% and 1.75-6.45 MPa. Their research indicates that the value of tensile strength will grow with the addition of PCC up to 4 percent, but then decrease, while the percent elongation will continue to decrease as the quantity of PCC increases. Based on the benefits of cellulose and $CaCO_3$ outlined in this study, cellulose and $CaCO_3$ were chosen as fillers for bioplastics derived from tapioca starch.

Of the many studies conducted on bioplastics, no studies have reported the effect of combining cellulose and $CaCO_3$ in the bioplastic matrix. Therefore, cellulose and $CaCO_3$ fillers will be combined to observe the bioplastic's physical, mechanical, and morphological properties produced in this research.

EXPERIMENTAL SECTION

Materials and Equipment

The main ingredients used in this study were tapioca starch (food grade) as a matrix, elephant grass taken in Sukamakmur District, Aceh Besar, and oyster shells taken in Alue Naga Banda Aceh. While the supporting materials used are 98% NaOH (Merck), NaOCl (Technical), Glycerol (Merck), Ammonium Carbonate (Technical), and HCl (Merck).

Raw Material Preparation

Elephant grass

Elephant grass is reduced to 1-2 cm, then washed with distilled water and dried in the sun. After drying, the elephant grass was mashed with a blender and sieved with a 50 mesh sieve (Holik et al., 2014).

Oyster shell

The oyster shells were washed thoroughly and then dried in the sun for seven days. Then reduce the size to a powder. After that, the oyster shell powder was sifted through a 50 mesh sieve (Handayani & Syahputra, 2017).

Extraction of Cellulose and $CaCO_3$

Extraction of cellulose from elephant grass

Figure 1 shows a diagram of the process of making cellulose from elephant grass. This cellulose extraction method was adapted from Holik et al. (2014). Delignification: Cellulose is produced from elephant grass powder by adding 10 grams of elephant grass powder to 200 mL of 4% NaOH solution and then heated at a temperature of 80±5 °C for 1 hour with stirring. Then filtered, and the residue was washed with distilled water until pH neutral (pH 7). Then the residue was dried in an oven at a temperature of 80±5 °C to a constant weight. Hydrolysis: 2 grams of the results from the delignification stage were added to 36 mL of 0.2 N HCl solution. Then it is heated at a temperature of 80±5 °C for 2 hours. The results of this stage are then filtered and washed with distilled water until pH neutral. Bleaching: residue from the hydrolysis step, which was neutral, was added to 12% NaOCl solution until the color of the residue turned white. Then the residue is filtered and washed until the chlorine smell disappears. The residue was dried in an oven at a temperature of 80±5 °C until the weight was constant.

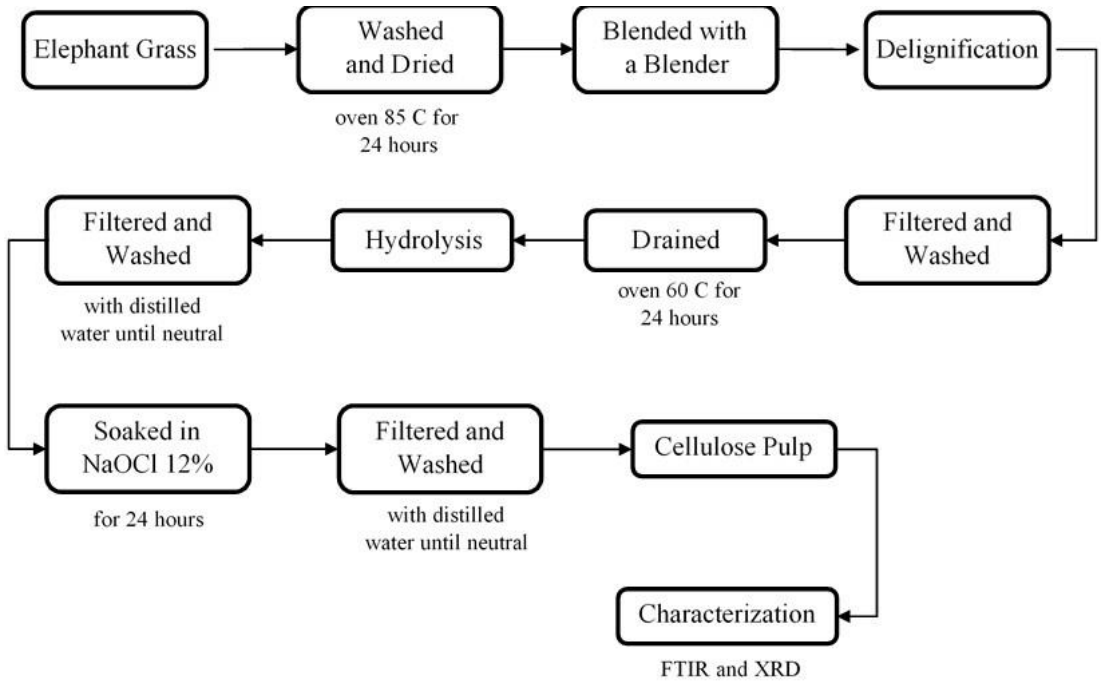

Figure 1. Diagram process of extracting cellulose from elephant grass

Extraction of $CaCO_3$ from oyster shells

Figure 2 shows a diagram of the $CaCO_3$ extraction process from oyster shells. This $CaCO_3$ extraction method was adapted from research conducted by Cahyono et al. (2019). A total of 30 grams of oyster shell powder was added to 250 mL of 3 N HCl solution and stirred without heating. It was then filtered to separate solids and solutions. A 1.2 N ammonium carbonate solution was added to this solution and allowed to stand for 24 hours to form a $CaCO_3$ precipitate. Then the $CaCO_3$ solid was filtered and dried in an oven at 60 °C until the weight was constant.

Bioplastic film making

Figure 3 shows a schematic process of making bioplastic films. This bioplastic film is made using the method from Tamiogy, Kardisa, Hisbullah, & Aprilia (2019). The method used is the phase inversion method. Ten grams of tapioca starch was mixed with 100 mL of distilled water and stirred using a magnetic stirrer until homogeneous. Then 3 mL of glycerol was added as a plasticizer. Furthermore, cellulose was added with variations of 3, 5, and 7 grams. The solution was heated on a hot plate at 70 °C until it thickened. The solution was poured on a glass plate as a mold and dried at room temperature for five days (until the bioplastic was easy to open from the mold). After that, the bioplastic was removed from the glass plate and analyzed for its physical, mechanical, and morphological properties. The best variation was taken based on its mechanical properties, and then the same treatment was carried out but with the addition of $CaCO_3$ with variations of 1.5, 3, and 4.5 grams. **Table 1** shows a research design for making bioplastics. Considerations in preparing the research design, as shown in **Table 1**, are based on previous research on cellulose and $CaCO_3$ fillers. In a study conducted by Holik et al. (2014), it was found that the use of 20% cellulose obtained better mechanical properties than 5, 10, and 15% cellulose. According to a study Rakhman & Darni (2018), the mechanical characteristics of materials containing $CaCO_3$ at concentrations of 0, 2, 4, and 6% increased. In this study, the filler composition was used more than what has been done in previous studies to obtain the best results.

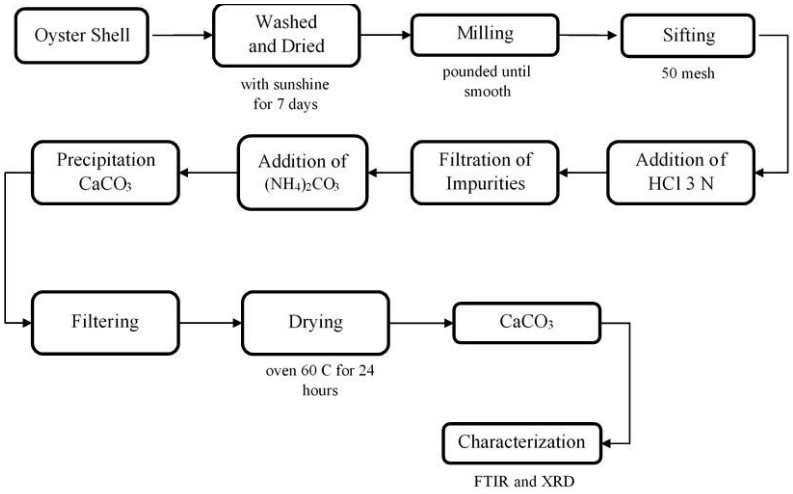

Figure 2. Diagram process of extracting $CaCO_3$ from oyster shells

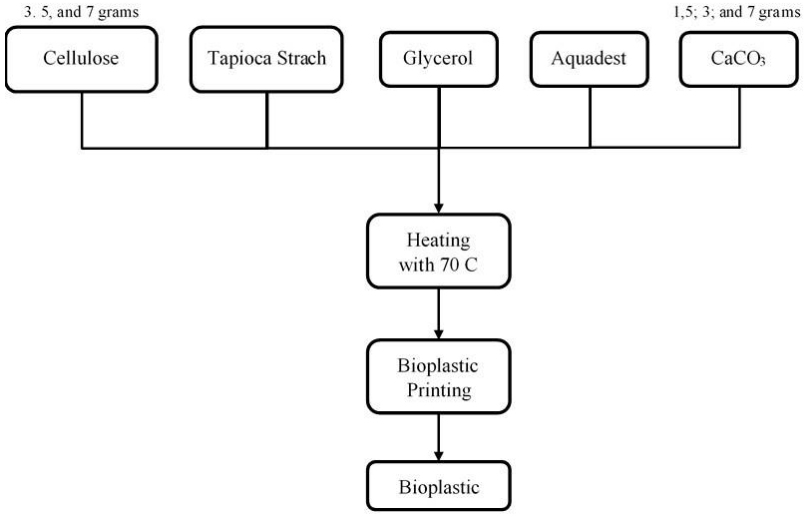

Figure 3. Diagram process of making bioplastic film

Table 1. Research Design

No	Variable	Tapioca Starch Mass (g)	Glycerol Volume (mL)	Aquadest Volumes (mL)	Cellulose Mass (g)	$CaCO_3$ Mass (g)
1	A1	10	3	100	3	-
2	A2	10	3	100	5	-
3	A3	10	3	100	7	-
4	B1	10	3	100	5	1,5
5	B2	10	3	100	5	3
6	B2	10	3	100	5	4,5

Bioplastic Film Characterization

Physical properties

Density test

The method to find the density of bioplastic was adapted from Darni et al. (2014). The mass (m) of bioplastics was weighed. Then the 10 mL measuring cup was filled with up to 5 mL of water, and the weighed bioplastic was put into the measuring cup. After 15 minutes, the new water volume (v) was recorded to calculate the actual bioplastic volume by finding the difference between the initial and final water volumes. Then the density (ρ) can be obtained from Equation (1).

$$\rho = \frac{m}{v} \quad (1)$$

Where: ρ (g/mL) : Density of bioplastic
m (g) : Mass of bioplastic
v (mL) : Difference between the initial and final volumes of water

Water absorption

The water absorption procedure follows the procedure carried out by Tamiogy et al. (2019). The weight of the initial bioplastic to be tested is weighed, then put into a beaker filled with water. After 10 seconds, the bioplastic was removed and weighed. Repeat until the bioplastic weight is constant. The water absorbed is calculated by Equation (2).

$$\text{Air (\%)} = \frac{W - Wo}{Wo} \times 100 \quad (2)$$

Where: W (g) : Weight of bioplastic after soaking
Wo (g): Initial weight of bioplastic

Mechanical properties

Testing the mechanical properties of bioplastics was to get the value of tensile strength, percent elongation, and Young's modulus. This test shows how strong the bioplastic is when pulled (Juandi & Haekal, 2016). This mechanical property test follows the ASTM 638 standard with a standard sample size, as shown in **Figure 4**.

Bioplastic morphology

The morphological characterization of bioplastics includes functional group analysis and surface analysis using FTIR (Fourier Transform Infrared) and SEM (Scanning Electron Microscopy). For the functional group analysis, the sample in the form of a film is placed in a sample container to be tested. The sample spectrum is recorded on the monitor screen at a wavelength of 400-4000 cm^{-1}. The FTIR used is Shimadzu Prestige 8400. For the morphology analysis, the sample is cut into 1 x 1 cm pieces to be attached to the holder (a sample container to be tested). The sample is inserted into the SEM USED JEOL JSM 6360 equipment chamber for position setting and image recording. SEM photos were taken with a magnification of 300 times.

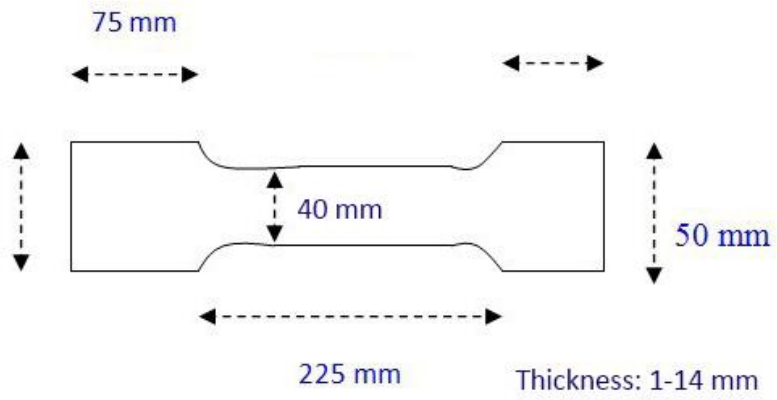

Figure 4. Standard size of mechanical properties test (Ernita et al., 2020)

RESULTS AND DISCUSSION

Bioplastic Density

A density test can perform to determine a material's density/density (mass/volume). The higher the density of a material, the better its mechanical properties (Darni et al., 2014). The density of bioplastics can see in **Figure 5**.

The density of a material is related to the type of substance contained in the material. In addition, the amount of a substance used as a filler is an important factor in the density to be obtained (Darni et al., 2014). The high-density value of the bioplastic indicates that the bioplastic is more resistant to water vapour because every volume in the bioplastic is already filled (Rakhman & Darni, 2018). From **Figure 5**, it can see that the more filler is added, the density of the resulting bioplastic will increase. In samples A1, A2, A3, B1, B2 and B3 obtained a density value of 0.313; 0.314; 0.317; 0.330; 0.331 and 0.332 g/mL. The results obtained from sample A have a lower density value than the density value of sample B. This result is because sample B uses two fillers, cellulose and $CaCO_3$, where $CaCO_3$ will fill the empty spaces in the matrix and result in a higher density value than cellulose filler alone. Density is also the amount of mass that occupies a volume, so the combination of two fillers increases the mass that occupies a volume (Rakhman & Darni, 2018).

Bioplastic Water Absorption

Water absorption is also an important indicator in determining the quality of bioplastics. The results of the water absorption test from this study can see in **Figure 6**.

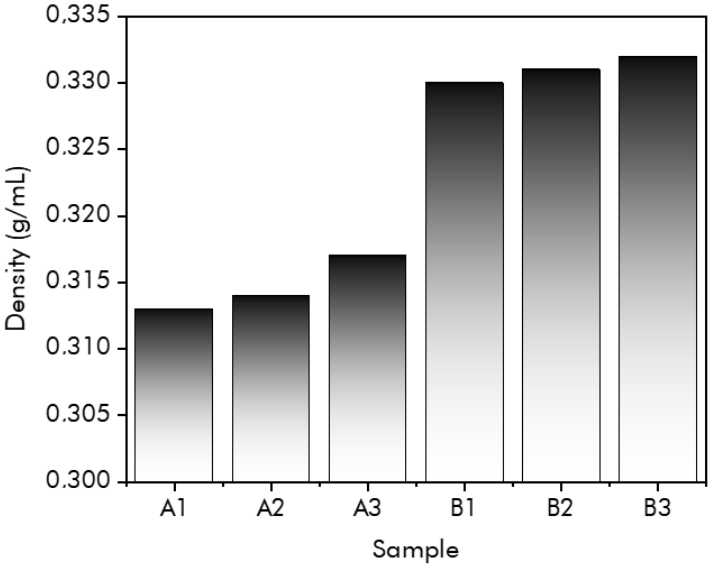

Figure 5. Density of Bioplastic

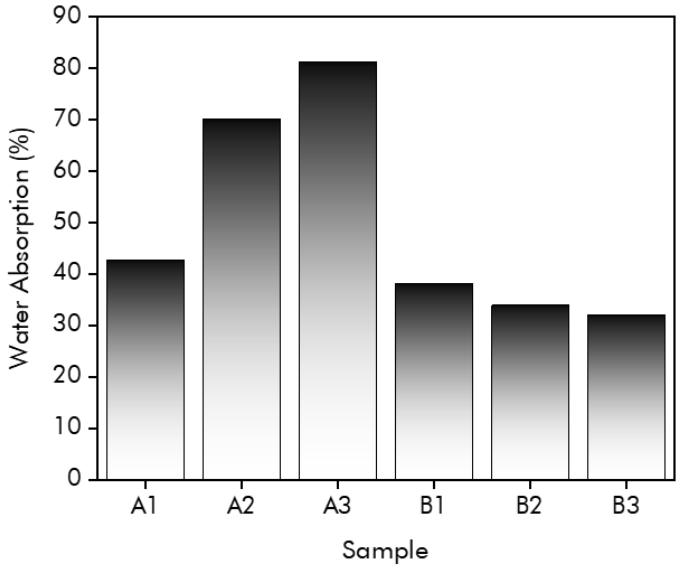

Figure 6. Water absorption of bioplastics

From **Figure 6**, it can see that the results of the water absorption test for samples A1, A2, A3, B1, B2, and B3 were 42.65; 70; 81.16; 38.03; 33.87, and 31.94%. The water absorption capacity was increasing for sample A. According to research by Sutan et al. (2019), hydrogen bonds in cellulose are strong, so it will not be easy to absorb water. Therefore cellulose is also insoluble in water. However, if too much cellulose is used or almost equals the base polymer (starch), the water absorption capacity of the bioplastic will increase. This increase is because the excess cellulose can make hydrogen bonds form intramolecular hydrogen bonds in water (Sutan et al., 2019).

In sample B, with $CaCO_3$, the results decreased with the increase in $CaCO_3$ used. According to Kisku et al. (2014), this decrease was due to the strong interfacial adhesion between $CaCO_3$ and the polymer matrix. The addition of $CaCO_3$ will reduce water absorption because the $CaCO_3$ particles inhibit the diffusion of water into the matrix structure (Dawale et al., 2018).

Tensile Strength of Bioplastic

Tensile strength is the maximum stress that a material can withstand before breaking (Arini et al., 2017). The results of the tensile strength test can see in **Figure 7**.

Figure 7 showed an increase in tensile strength followed by the filler used. The tensile strength for sample A was 1, 3, and 3 MPa, while for sample B, it was 1, 2, and 2 MPa. These results showed differences in tensile strength between samples A and B. Bioplastic samples added with $CaCO_3$ filler have a lower tensile strength value than bioplastic samples that only use cellulose filler. This result happens because of differences in the structure of tapioca starch, cellulose, and $CaCO_3$. This different structure makes them difficult to bond (Darni et al., 2014). Cellulose and starch are organic compounds, whereas $CaCO_3$ is inorganic. This discrepancy causes the bonds between the components to be weak. The low force required to break bioplastics is a result of the weak strength of bioplastics' molecular bonds (Hutabalian et al., 2020). The tensile strength of a material is its response to a force (Tamiogy et al., 2019), hence the bioplastics' tensile strength will decrease as the force decreases.

Percent Elongation of Bioplastic

The elongation test compares the increase in length that occurs with the length of the material before the tensile test is done. **Figure 8** shows the result of the percent elongation.

From **Figure 8**, it can see that there was an increase in the percent elongation for samples A and B. In sample A, percent elongation is increasingly significant, while in sample B, it was not too significant, and the increase was in sample B3. Elongation changes the maximum length of a material if the material is pulled before the material breaks. The percent elongation value for samples A1, A2, A3, B1, B2 and B3 is 1; 3.8; 4.4; 2,6; 1,3 and 1%. It can say that the addition of cellulose filler is directly proportional to the percent elongation, where the more filler used, the higher the percent elongation. The increase was due to the cellulose and tapioca starch forming hydrogen bonds to make the chain longer, which affected the percent elongation value (Darni et al., 2014). In addition, the contribution of $CaCO_3$ to sample B lowered the percent elongation of the bioplastic. This drop is attributed to the calcium concentration of $CaCO_3$, which stiffens bioplastics. This rigidity will affect when pulling on bioplastics (Rakhman & Darni, 2018). Therefore, the percentage of elongation will decrease as $CaCO_3$ increases.

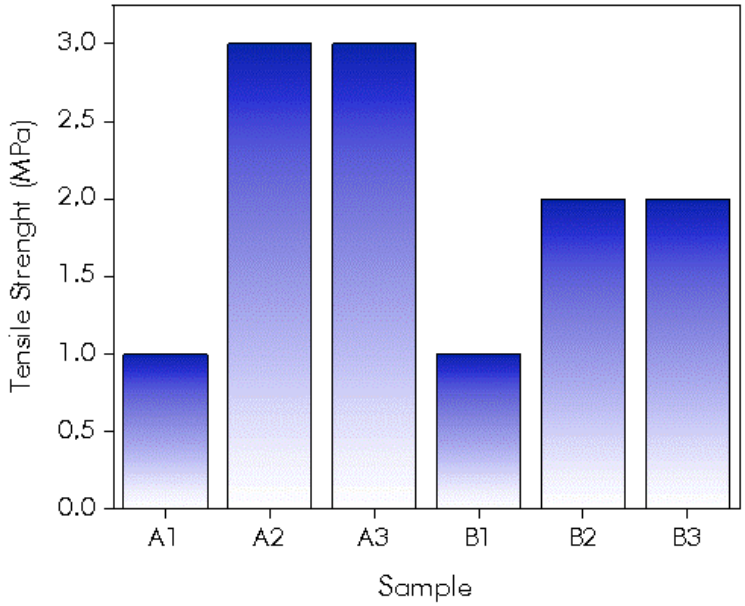

Figure 7. Tensile strength of bioplastics

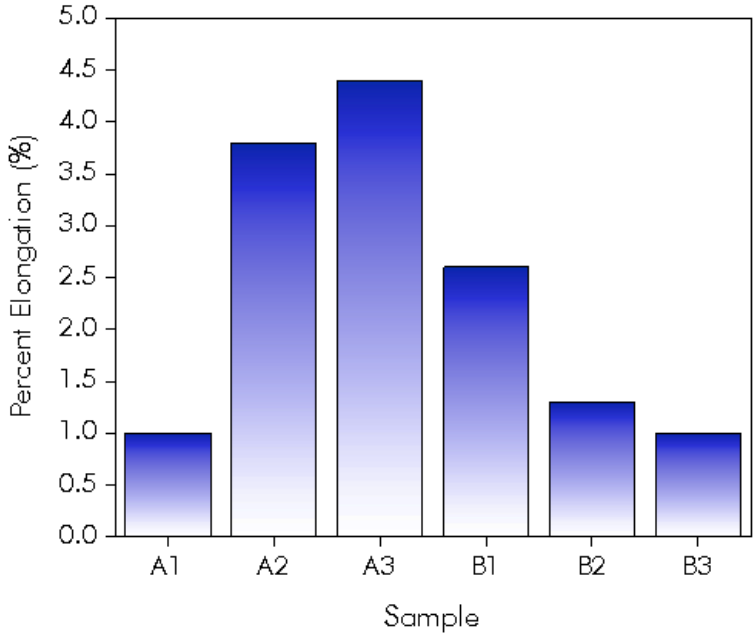

Figure 8. Percent Elongation of Bioplastics

Table 2. Comparison of Bioplastics in This Study with Other Studies

Parameter	Haryanto & Titani (2017)	Haryanto & Saputri (2016)	Harsojuwono et al. (2016)	Susanti et al. (2015)	This study
Tenslie Strenght (MPa)	0.37	2.5	0.51 – 0.93	0.9	3
Percent of Elongation (%)	19.41	40	18.75 – 65.31	-	4.4
Young's Modulus	0.02	0.06	0.01 – 0.05	-	0.68
Water Absorption (%)	-	-	-	-	81.16

Based on **Table 2**, there is an increase in tensile strength results from previous studies using tapioca starch as a bioplastic matrix. Cellulose can increase the stiffness of bioplastics so that the tensile strength increases, but because the tensile strength is always inversely proportional to the percent elongation, there is a decrease in the percent elongation in this study. This study's higher tensile strength value was due to the good bond between cellulose and tapioca starch. Haryanto & Titani (2017) made bioplastics with a mixture of two starches (tapioca and corn) without using fillers. The tensile strength obtained is 0.37 MPa. The use of fillers affects increasing the tensile strength, so making bioplastics without using fillers will lower the tensile strength. Haryanto & Saputri (2016) made bioplastic using tapioca starch with ZnO filler. The tensile strength obtained is 2.5 MPa. This result is lower than the result of this study due to the different properties between starch and ZnO (organic and inorganic). In this study, starch and cellulose, which have the same properties (organic and organic), were used to make the bond between the matrix and filler better. In contrast to the tensile strength obtained, the percent elongation obtained is lower than several previous studies. This lower value is caused by the higher value of tensile strength, which makes the material stiffer and lowers the value of percent elongation. Differences with several other studies are presented in **Table 2**.

Functional Group Analysis

The way to analyze the functional group from a component is using FTIR. FTIR can identify a functional group because the interaction of a molecule with electromagnetic radiation is specific and different for each functional group of a compound (Limbong, 2017). The functional groups of bioplastic samples A and B are compared in **Figure 9.** FTIR analysis is required to back up the physical and mechanical testing results that have already been completed. **Figure 9** displays the FTIR analysis results for bioplastic samples A and B, where the spectra are either the same or different. The spectra at 1557 cm^{-1} reveal the presence of a functional group O–H, whose intensity diminishes as $CaCO_3$ rises. The presence of the O–H group shows the bioplastic's hydrophilicity, and a decrease in the intensity of the O–H group suggests that adding $CaCO_3$ can diminish the bioplastic's hydrophilicity (Christwardana et al., 2021).

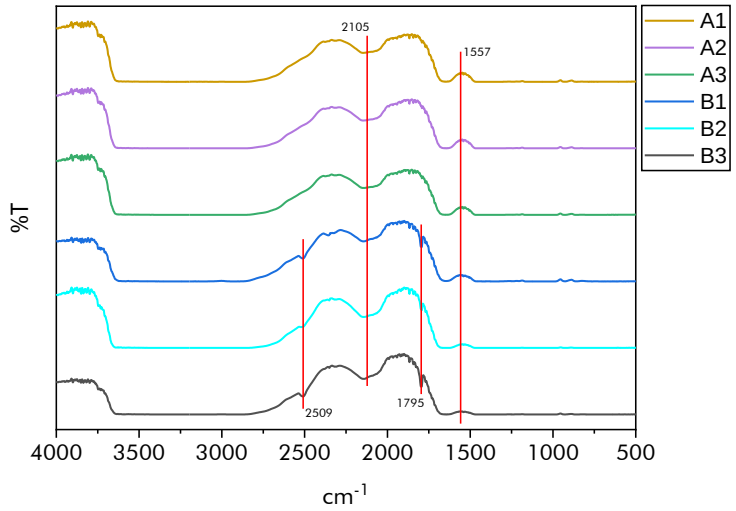

Figure 9. Comparison of the spectrum of bioplastics with their constituent materials

Table 3. Infrared spectrum of bioplastic

No.	Spectrum (cm^{-1})	Standard spectrum (cm^{-1})	Functional group	Reference
1	1557	1500 – 1600	O – H	(Christwardana et al., 2021)
2	1795	1760 – 1800	C = O	(Nandiyanto et al., 2019)
3	2105	2100 – 2140	C ≡ C	(Nandiyanto et al., 2019)

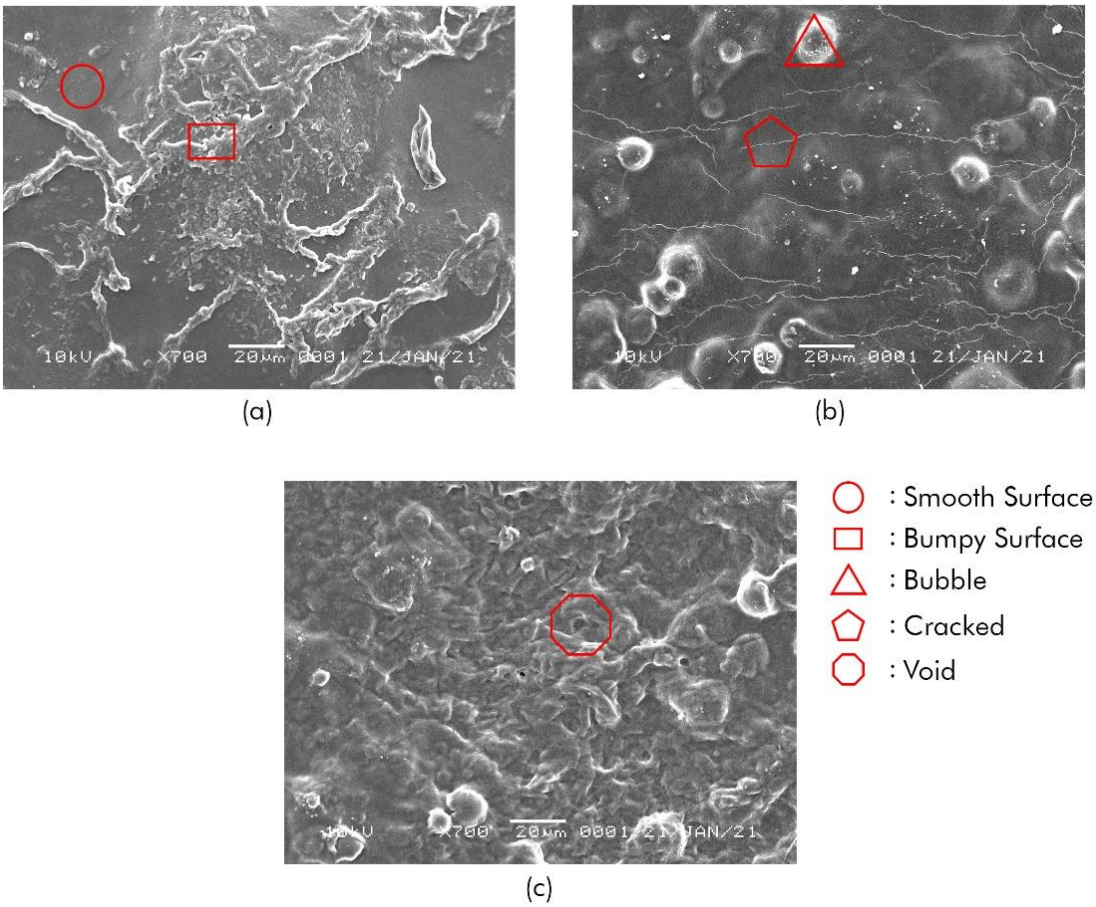

Figure 10. Bioplastic surface with samples A1 (**a**), A2 (**b**), and A3 (**c**)

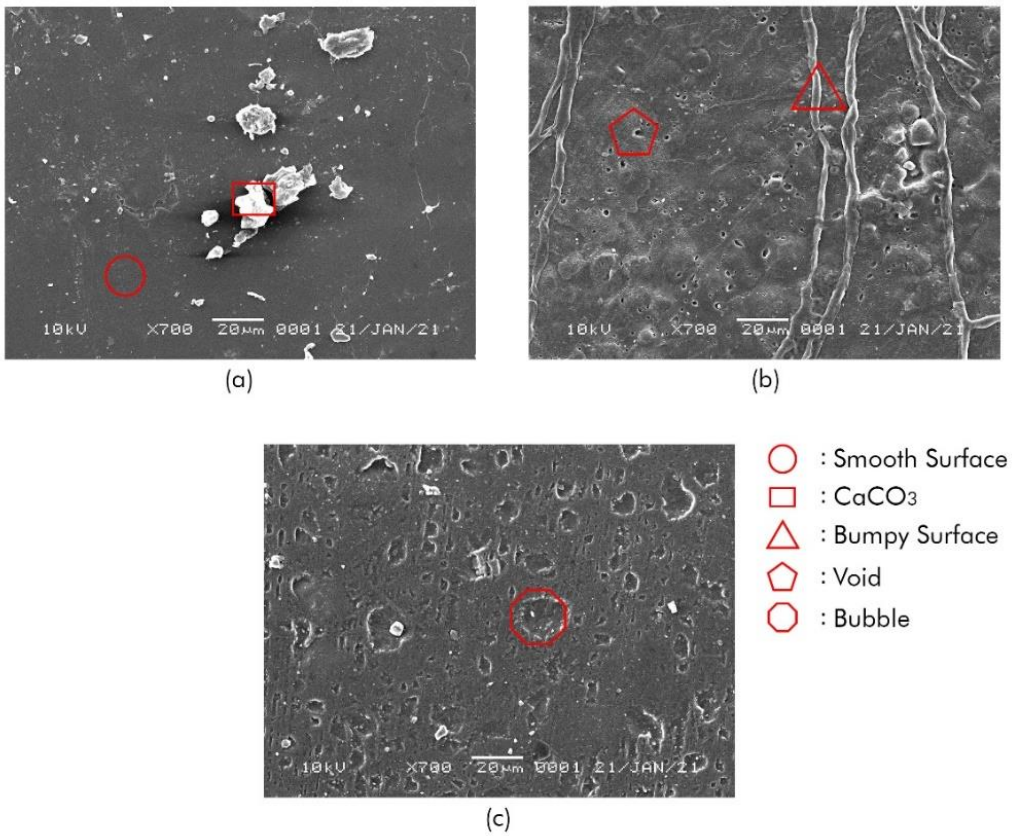

Figure 11. Bioplastic surface with samples B1 (**a**), B2 (**b**), and B3 (**c**)

Hydrogen bonding can improve bioplastics' strength (Nandiyanto et al., 2019). As a result of the lower hydrogen bond intensity in sample B, the tensile strength decreases. The 1795 cm-1 spectrum was only identified in $CaCO_3$ sample B. The C=O group, a carboxyl group present in $CaCO_3$, is seen in this spectrum (Nandiyanto et al., 2019). An alkyne group, A C≡C group, is visible in spectrum 2105 cm^{-1}. Research Harsojuwono et al. (2016) shows that the alkyne group contained in samples A and B comes from tapioca starch. Therefore, alkyne groups are present in all samples A and B.

Morphology Analysis

Addition of one filler (cellulose)

The addition of cellulose into the bioplastic film aims to improve the properties of bioplastics than before. However, whether or not this mixture is perfect will affect the surface of the bioplastic produced. **Figure 10** shows the morphology of bioplastics for sample A.

Figure 10 shows that as the amount of cellulose utilized rises, the surface of the bioplastic becomes uneven. It can be noticed in sample A1 that there are still even bumpy patches. This finding suggests that cellulose in bioplastics is not equally distributed. The presence of cavities can also be seen. Bubbles began to form in samples A2 and A3, and heating can induce fissures in bioplastic. Bubbles and fissures appear since cellulose does not attach to tapioca starch. As cellulose levels rise, the rigidity of bioplastics rises, causing fissures (Tamiogy et al., 2019).

Addition of two fillers (cellulose and $CaCO_3$)

Figure 11 depicts the uneven distribution of cellulose and $CaCO_3$ filler on the surface of the bioplastic. There are still whitish aggregates in sample B1, suggesting insoluble $CaCO_3$. In samples B2 and B3, there are still voids and bubbles, showing that the filler is not fusing with the tapioca starch. The cellulose fibres tangle together and make it challenging to spread uniformly in the tapioca starch mixture. In the meantime, the more $CaCO_3$ is used, the more filler remains visible. The matrix is already filled with filler so that it will not dissolve if it is applied again (Darni et al., 2014).

CONCLUSION

Bioplastics from tapioca starch with elephant grass cellulose and $CaCO_3$ from oyster shell waste as filler were made with 30, 50, and 70% (cellulose) and 15, 30, and 45% ($CaCO_3$) by weight of tapioca starch. Physical properties, mechanical properties, and morphology are tested to evaluate the effects of using the two fillers on these parameters. The use of cellulose as a filler could increase the mechanical properties of bioplastics, but the incorporation of cellulose and $CaCO_3$ fillers produced lower mechanical properties than cellulose fillers alone. The best values for tensile strength, percent elongation,

and Young's modulus were found in samples A3, namely 3 MPa, 4.4%, and 0.68 MPa. Sample B3 had the best density and water absorption, namely 0.33 g/mL and 31.94%. The strength of the O-H group in the bioplastic sample decreases as the amount of $CaCO_3$ increases, as shown by FTIR measurement. According to SEM morphological study, adding more $CaCO_3$ caused the surface of bioplastics to become broken and uneven. Natural materials (starch, cellulose, $CaCO_3$) have the potential to become bioplastics' raw materials, so it is quite intriguing to research further to achieve the greatest outcomes. In addition to having the potential to replace synthetic plastics, using natural materials (oyster shell $CaCO_3$) could reduce unutilized aquatic waste.

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