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Exploring the Potential of Natural Material: A Study Bioplastics from Cornstarch and Coconut Shell Cellulose

Sulistyaningsih^a, Yusri Hajjo Dwipa^a, Muzayyanah Rizqy Fikriani^a,
Retno Aliyatul Fikroh^{a*}

Abstract. This study aimed to develop cornstarch-based bioplastics with the addition of coconut shells as a natural reinforcing material. The coconut shells first underwent a delignification process using NaOH and bleaching with H₂O₂ to remove lignin and increase cellulose content. The characterization results obtained from FTIR showed that the cellulose structure was maintained, with an increase in hydroxyl groups that played an important role in the formation of the bioplastic structure. Bioplastics were then synthesized with three different formulations based on variations in the amount of coconut shells and were tested for mechanical properties and water absorption. The test results showed that increasing the amount of coconut shells increased tensile strength, with the highest value of 6.75 MPa observed in the third formulation. However, the first formulation, with 2.5 grams of coconut shells, exhibited the best water resistance, with an average absorption of 37.21%. All formulations met the JIS standard for edible films. This study concluded that coconut shells had the potential to serve as a reinforcing material in environmentally friendly bioplastics.

Keywords : Bioplastic, Cellulose, Coconut Shell, Cornstarch

^aUIN Sunan Kalijaga, Jl. Laksda Adisucipto, Papringan, Caturtunggal,
Depok, Sleman, Yogyakarta 55281, Indonesia

Correspondence and requests for materials should be addressed to Retno Aliyatul Fikroh
(email: retno.fikroh@uin-suka.ac.id)

Introduction

Plastic waste is difficult to decompose, taking 10-100 years to break down completely. The impact of this accumulation is highly destructive to the environment, ranging from land pollution to marine ecosystems. Indonesia's seas are heavily polluted, with 60%-80% of marine waste being plastic. This leads to the death of marine life and the destruction of coral reefs, which serve as important habitats [1]. One of the most devastating impacts of plastic waste is the destruction of ecosystems and the death of marine life. Indonesia's seas are highly polluted by waste, with 60%-80% of the waste that ends up in the sea being plastic [2].

Plastic pellets can be recycled to become raw materials for various products that have useful value according to the needs of the industry [3]. However, it should be noted that plastic products around us are mostly made from petroleum and natural gas, both of which are categorized as B3 (Toxic and Hazardous) materials. Plastics are a type of long-chain polymer, making them very difficult to decompose naturally. Various efforts have been made to reduce the use of conventional plastics that cause environmental pollution; one such effort is to develop and use bioplastics [4].

Bioplastics are plastics or polymers that can be easily degraded naturally, both through microorganisms and by weather (moisture and sunlight radiation) [5]. Bioplastics are made from natural sources such as corn crops, sugarcane, or starch that can decompose naturally in the environment [6], [7]. Bioplastics composed of cellulose can be developed using natural materials that are no longer used. The use of waste as a bioplastic base material is an alternative to reduce the use of conventional plastics. One of the potential wastes for the manufacture of bioplastics is coconut shells because they produce a fairly high amount of cellulose.

Coconut shells are actually rich in benefits, both as raw materials for various products such as bioplastics, building materials, textiles, or animal feed, and as a source of biomass energy [8]. However, the lack of awareness regarding the economic and environmental value of coconut waste, combined with insufficient infrastructure and technology for its treatment, has hindered the maximum utilization of this waste in

the country. By combining technological innovation with the sustainable use of natural resources, the utilization of coconut shells in the manufacture of bioplastics becomes an important step towards a more sustainable solution in addressing the problem of environmental pollution by plastics [9].

Coconut shells in Indonesia are often only considered as waste, even though they have great potential to be used to their fullest [10]. Indonesia's coconut fruit production averages 15.5 billion grains/year, equivalent to 3.02 million tons of copra, 3.75 million tons of water, 0.75 million tons of shell charcoal, 1.8 million tons of coir fiber, and 3.3 million tons of coir dust [11]. However, most of this waste has not been properly utilized. Ironically, most of the coconut waste is exported to other countries without being utilized locally. Coconut shells contain chemical compounds, specifically 27.31% cellulose and 33.30% lignin. Cellulose has inter- and intramolecular bonds, which form microfibrils, making it insoluble in most solvents, including water, and giving it a high resistance to enzymatic processes and hydrolysis [12]. Because cellulose has stronger hydrogen bonds than starch, it is more resistant to hydrolysis. True cellulose will break down before melting, just like starch, so it is not a thermoplastic polymer [13].

The use of coconut shells as a bioplastic material is due to their characteristics of being antimicrobial, resistant to mold and decay, non-flammable, unaffected by moisture, sturdy and durable, elastic, returning to their original shape with the same consistency after use, and having the ability to absorb water up to three times their weight [14]. Cornstarch can be used as a bioplastic material because its main ingredient is starch, which is a natural, environmentally friendly polymer [15]. Starch has a molecular structure in the form of long chains of glucose that are able to form intermolecular bonds when heated and mixed with a plasticizer, such as glycerol [16].

In the manufacture of bioplastics, chitosan functions as a stabilizer and thickener, as well as forming a clear protective layer during film manufacturing, which can improve the mechanical properties of bioplastics [17]. Chitosan acts as a mixer to increase the strength of plastics. This is because chitosan can form hydrogen bonds with the chains of amylose and amylopectin in starch. Chitosan is composed of amine functional groups, primary and secondary hydroxyl groups, which cause chitosan to have high chemical reactivity [18]. Chitosan is hy-

drophobic and is able to form membranes and films well [19].

Bioplastics made from coconut shells and cornstarch are an interesting innovation, especially when used to replace the role of packaging that is difficult to decompose, unlike conventional plastics that take a long time to break down in the soil. Bioplastics from coconut shells can be reused even after their initial purpose is served, for instance, by being repurposed as handicrafts or as compost, because they are natural materials that are easily decomposed in the soil [20]. In addition, the use of coconut shells and cornstarch as bioplastic materials can reduce waste in the surrounding environment. This aligns with the SDGs principle, where the exploration and exploitation of natural resource components in the development process must be balanced with the sustainable management of natural products and wastes. The principle of environmental preservation must be the foundation for development and change in order to achieve welfare for humans and the sustainability of nature [21].

The use of coconut shells as a material for making bioplastics has not been widely researched, so researchers are interested in conducting studies on their use for environmentally friendly bioplastics. The use of coconut shells offers an alternative for creating environmentally friendly bioplastics, aiming to overcome the plastic problem prevalent in the current environment. Therefore, the researcher is interested in conducting research entitled 'Exploration of the Potential of Natural Materials: Bioplastics from Cornstarch and Coconut Shell Cellulose,' aimed at developing packaging through the use of coconut shell cellulose as a means to reduce conventional plastic waste that is difficult to decompose in the soil and causes pollution of the natural environment.

Experimental

Method. The research method used in this study is comparative. This method was chosen to identify the best formulation ratio that can be used to develop bioplastics from cornstarch and coconut shells. The variation in this study lies in the amount of coconut shells, with formulations of 2.5 grams, 5 grams, and 7.5 grams respectively. This research was carried out from September 2024 to January 2025 at the Chemis-

try Laboratory of UIN Sunan Kalijaga Yogyakarta. The tools, materials, and work procedures in this study are as follows:

Material. The tools used in this study are hotplates, analytical balances, beakers, measuring cups, 20 cm × 20 cm acrylic molds, a mortar, aluminum foil, a magnetic stirrer, a thermometer, a spatula, a 140-mesh sieve, and an oven. The materials used are coconut shells, H₂O₂ cornstarch, NaOH, deionized water, chitosan (as a filler), glycerol (as a plasticizer), and litmus paper.

Instrumentation. Testing of cellulose has been performed using the FTIR method. Fourier Transform Infrared (FTIR).

Sample preparation. Cellulose isolated from coconut shells refers to research Nurwidiyani & Triawan (2022) [22]. Cellulose isolation begins with the preparation of a coconut shell sample. Twenty grams of coconut shell powder are weighed and then infused in 14.4% H₂O₂ and 9% NaOH. The mixture is heated and stirred with a magnetic stirrer at 55°C for 2 hours. The coconut shells are then filtered and rinsed three times using deionized water. The sample is dried using an oven. After drying, the sample is mashed using a mortar and pestle, then filtered using a 140-mesh sieve.

Bioplastic manufacturing process. Bioplastics are polymers that can be easily degraded in the soil by microorganisms, weather, moisture, and sunlight radiation [5]. The process of making this bioplastic uses the Melt Intercalation method [23] Specifically, the phase inversion technique through solvent evaporation after the printing process.

The manufacture of bioplastics is carried out by preparing two solutions: starch from cornstarch and chitosan. The preparation of the starch solution involves dissolving 15 grams of cornstarch, then stirring it using a magnetic stirrer for 10 minutes until it becomes a starch suspension. The chitosan solution is then made by dissolving 2.5 grams of chitosan in 50 grams of 1% acetic acid and stirring at a temperature of 100°C until a chitosan gel is obtained. After that, the starch suspension is stirred and heated at 65°C for 5 minutes.

The thickened starch solution is added to the previously prepared chitosan solution, followed by the addition of coconut shells with variations of 2.5 grams, 5 grams, and 7.5 grams. Also, 0.5 grams of montmorillonite are added while stirring and heating at 90°C for 5 minutes. After that, 5 grams of

glycerol are added and stirred at 65 °C for 10 minutes. The finished bioplastic solution is poured into a 20x20 cm acrylic mold and leveled on its surface. The wet bioplastic is then dried in the oven at 50°C for approximately 1.5 hours.

Bioplastic Results Test. The resulting bioplastics then undergo a testing process. The tests that will be carried out are the tensile test and the water resistance test. Tensile testing is performed using a spring balance. The procedure for this test is as follows: One end of the bioplastic sample is clamped onto one of the spring balances, gently pull the other end until the sample breaks, record the maximum force value indicated by the spring balance before the sample breaks (in Newtons). Data analysis is carried out using equation 1.

$$\text{Tensile Strength} = \frac{F}{A} \quad (1)$$

Description: F = Force; A=cross-sectional area.

The water resistance test is carried out by soaking the bioplastic sample in a container filled with water, then weighing the sample every 10 minutes until its weight becomes constant. The water resistance test aims to measure the amount of water absorbed by the bioplastic [24]. Water absorption will cause an increase in the weight of the bioplastic polymer, which can indicate whether there is a bond in the polymer and the bond structure within the bioplastic [25]. Its water absorption capacity is calculated using equation 2.

$$\text{WC (\%)} = \frac{W1 - W0}{W0} \quad (2)$$

Description : Wc = Water absorption; W0 = Starting weight; W1 = Weight after soaking.

Result and Discussion

The manufacture of bioplastics using cornstarch and coconut shells begins with the preparation of coconut shells. Coconut shells need to be delignified and bleached. Delignification aims to reduce the level of lignin in lignocellulose. Delignification will open up the lignocellulose structure, making cellulose more accessible. The delignification process will dissolve the lignin content in the material, thereby facilitating the process of separating lignin from fibers [26]. The

delignification process causes damage to the structure of lignin and releases carbohydrate compounds. The process of destroying the structure of materials with lignocellulose content is one of the steps to convert lignocellulose into sugar compounds. The delignification process in this study uses NaOH. This process aims to reduce the lignin content, the natural "glue" that makes coconut shells hard and rigid. By using a strong basic solution of NaOH (sodium hydroxide), the chemical bonds in the lignin molecules are broken down. Once the lignin bonds are destroyed, the lignocellulose structure opens up, allowing the cellulose fibers to separate from the lignin and hemicellulose. The result is a purer, more accessible cellulose fiber, ready to be processed into bioplastic polymers. Without this process, the lignin would hinder processing and molding, making delignification an essential step to obtain an optimal bioplastic raw material.

As for the bleaching process, the color of the coconut shell changed from dark brown to light brown. This is because the bleaching process involves the breakdown of lignin by H₂O₂ in an alkaline atmosphere. Lignin causes a dark brown color in the coconut shell; thus, a reduced amount of lignin in the coconut shell causes its color to fade [27].

The bleached sample is then subjected to FTIR testing to identify the functional groups of chemical compounds contained in bioplastic materials, thereby providing in-depth insights into their chemical structure. FTIR analysis displays the specific wavenumber of a functional group, which is then matched with the number of waves in the spectrum according to the theory of the special IR absorption region of the cellulose constituent group. The testing process begins with preparing bioplastic samples that are cut to the size required for analysis. The sample is then analyzed using FTIR spectroscopy at room temperature. This method generates data in the form of a graphic spectrum that illustrates the relationship between wavelength and the rate of light transmission through the sample. From this spectrum, information about the types and intensities of functional groups contained in bioplastics, such as carbonyl, hydroxyl, or methyl, can be accurately determined. This analysis is essential to evaluate the quality of the material and its potential application as an environmentally friendly material.

Figure 1 shows the results of the FTIR test on coconut fiber samples. The results of this FTIR test reveal several peaks that represent the func-

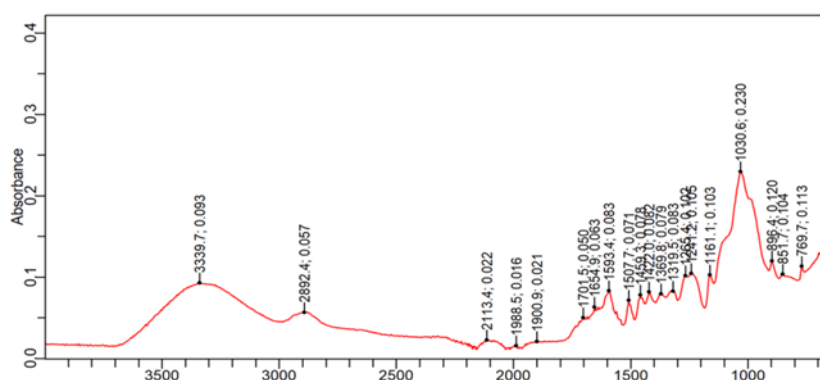


Figure 1. Coconut shell FTIR test results

tional groups contained in the sample. The hydroxyl group (-OH) appears at a wavelength of 3339.69470 nm, indicating the O-H stretch vibration characteristic of cellulose. A wavelength of 1030.60891 nm indicates the peak of C-O stretch vibration in the glycosidic structure of cellulose. At a wavelength of 1265.43120 nm, the peak of C-O stretch vibration of aromatic lignin appears. The low intensity of this peak indicates that lignin has been largely degraded during the bleaching process, although a small amount is still present. A wavelength of 1507.70816 nm indicates aromatic C=C vibrations. The presence of a peak at this wavelength suggests that there are still traces of lignin left in the material. The C=O vibration of hemicellulose is seen at a wavelength of 1701.52972 nm, which indicates residual hemicellulose contained in the material. C-H vibrations are seen at wavelengths of 289.41416 nm, which indicates the presence of an aliphatic C-H stretch in cellulose.

Peaks at 3339.69470 nm (O-H vibration) become sharper after bleaching. This indicates an increase in hydroxyl group content, which is typical of cellulose structures. These groups are important for intermolecular interactions, such as the formation of hydrogen bonds, which provide strength and mechanical stability to bioplastics. Peaks at 1030.60891 nm and 289.41416 nm confirm that the glycosidic structure and aliphatic framework of cellulose are not damaged during the bleaching process, which means the material is still suitable for use in the manufacture of bioplastics. The spectrum results showed that the cellulose from coconut shells had reached a sufficient level of purity to become a basic material for bioplastics. The remaining lignin content, even in small amounts, can provide certain benefits, such as strengthening the structure of bioplastic materials, but it needs to be

Table 1. Bioplastic Formulation

Material	F1	F2	F3
Cornstarch (gr)	15	15	15
Coconut shell (gr)	2,5	5	7,5
Chitoosan (gr)	2,5	2,5	2,5
Acetic Acid (gr)	50	50	50
Glycerol (gr)	5	5	5
Montmorillonit (gr)	0,5	0,5	0,5
Aqueducts (mL)	100	100	100

monitored so as not to interfere with the natural degradation process.

The process of making bioplastics in the study is based on three formulations. Each formulation is given a different variety of coconut shells. The formulations for making this bioplastic shown at Table 1. The appearance is as in Figure 2.

Following synthesis, the bioplastics underwent several tests, including tensile strength and water absorption measurements. These tests aimed to determine the optimal ratio of cornstarch to coconut shell for bioplastic formulations exhibiting desired characteristics. The effect of the cornstarch and coconut shell formulation on the bioplastic characteristics is presented in the Table 2.

The results of the tensile strength test showed that the addition of coconut shell powder affected the mechanical strength of the bioplastic. The optimal composition of cornstarch and coconut shells resulted in bioplastics with the highest tensile strength. This occurs because coconut shells provide

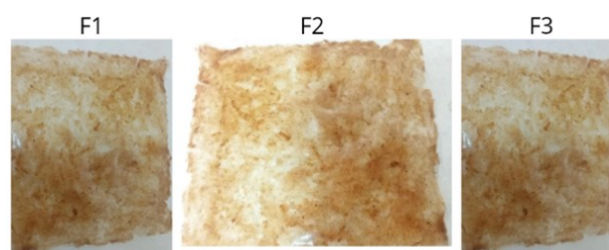


Figure 2. Bioplastic Formulation

Table 2. Traction Test

Results	Formulation		
	1	2	3
Length (m)	0,074	0,074	0,074
Thickness (m)	0,0001	0,0001	0,0001
Style (N)	35	40	50
Spring length change (m)	0,7	0,8	1
A (m ²)	0,0000074	0,0000074	0,0000074
Tensile Strength (Pa)	4729729,7	5405405,4	6756756,7
Tensile Strength (MPa)	4,7297297	5,4054054	6,7567567

additional structure that strengthens the starch matrix in bioplastics. Based on the tensile strength tests that have been carried out, it can be seen that the tensile strength of the three formulations tends to be similar. The highest tensile strength value in Formulation 1 is 4,729,729.73 Pa or 4.73 MPa. Formulation 1 has a composition of 15 grams of cornstarch and 2.5 grams of coconut shells.

According to JIS (Japanese Industrial Standards), an edible film must have a minimum tensile strength of 0.392266 MPa. This confirms that the research results meet the JIS tensile strength standard. This observed tensile strength stems from the cross-bonding between chitosan and glycerol, which consequently reduces the intermolecular forces among the polysaccharide chains, leading to a finer and more flexible plastic sample [28].

The water absorption test was conducted to assess the bonding between polymers within bioplastics. Bioplastics are hygroscopic due to the hydroxyl groups in some of their constituent components, which tend to bind water molecules from the environment. The water absorption test measures the bioplastic's percentage of resistance to water.

Based on this research, bioplastic Formulation 1 exhibited the highest water resistance, with an average absorption percentage of 37.212% and an average water resistance percentage of 62.788%. Meanwhile, Formulation 2 showed an average water absorption percentage of 41.016% and an average water resistance percentage of 58.984%. Formulation 3 had an average water absorption percentage of 46.288% and an average water resistance percentage of 53.712%.

Cellulose expansion aids in diminishing starch's hydrophilic properties, primarily due to cellulose's water insolubility. Chemically, cellu-

lose contains robust hydrogen bonds that impede its interaction with water. Nevertheless, excessive cellulose addition can unexpectedly enhance its absorption capacity. This phenomenon arises because intramolecular hydrogen bonds in cellulose molecules tend to form with water molecules as well. Consequently, a higher water absorption capacity compromises the bioplastic's water protection, leading to rapid degradation or decreased quality [29].

Conclusion

This study shows that coconut shells can be used as an additional raw material in the manufacture of cornstarch-based bioplastics through delignification and bleaching processes. The results of the FTIR analysis showed that the cellulose structure remained intact and that there was an increase in hydroxyl groups, a factor important for the strength and stability of bioplastics. Mechanical testing of three bioplastic formulations showed that the addition of coconut shells increased the tensile strength of the material. The formulation with 7.5 grams of coconut shells gave the highest tensile strength value of 6.75 MPa. All formulations met the minimum tensile strength standards based on JIS for edible films, which is 0.392 Mpa. The water absorption test showed that the formulation with 2.5 grams of coconut shells (Formulation 1) had the best water resistance, with an absorption rate of 37.212%. Overall, the combination of cornstarch and coconut shells produces bioplastics with mechanical properties and water resistance that are good enough for environmentally friendly applications. However, further optimization is still needed for the product to meet higher industry standards.

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Author Contributions

The authors designed and conducted the experiments, analyzed the data, and drafted the manuscript. All authors contributed to data processing, interpretation of results, and final editing of the manuscript. All authors have read and approved the final version of this article.

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