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Synthesis of Cellulose Straw and Rice Husk as Raw Materials for Bioplastic Production

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Abstract

The development of environmentally friendly materials such as bioplastics is a solution to reduce the negative impact of conventional plastics on the environment. This study presents a novel approach by utilizing hemicellulose extracted from rice straw and husks—abundant agricultural waste materials—as the main raw materials for bioplastic production. Hemicellulose was extracted using the alkalization method, and its characterization was carried out through FTIR analysis to identify typical polysaccharide functional groups. The results of the analysis showed the presence of O-H, C-H, and C-O-C groups, which indicate the glycosidic structure of hemicellulose, as well as indications of residual lignin with C=C groups. The resulting hemicellulose was then processed into bioplastics with the addition of plasticizers and reinforcing materials, incorporating glycerin, CMC, and TiO_2 to enhance mechanical properties and biodegradability. Mechanical and degradation tests demonstrated superior performance, highlighting the synergy between these additives. This study provides an innovative strategy for transforming agricultural waste into high-value bioplastics, contributing to sustainability and advancing green material technology.

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INTRODUCTION

The One of the most widely used materials among Indonesians is plastic. Due to its various benefits, such as its strength, flexibility, light weight, transparency, ability to be mixed with other materials, and lack of corrosion, plastic is frequently employed as a container material. (Mukhlisien, Suhendrayatna, Montazeri, & Amar, 2021). The amount of rubbish produced in Indonesia in 2021 was 28.6 million tons. The amount of plastic debris polluting the ocean has risen to 1.29 million tons annually. According to the Ministry of Environment and Forestry's SIPSN website (2022), 17.86% of the garbage mounds contained plastic waste. Compared to 2019, the amount of plastic waste generated rose by 2% (Kementerian Lingkungan Hidup dan Kehutanan, & Direktorat Jenderal Pengelolaan Sampah Limbah dan Bahan Berbahaya Beracun, 2022).

Additionally, more environmentally friendly and biodegradable plastic substitutes can be employed in place of commonly used synthetic polymers. Biodegradable plastic is a kind of bioplastic with biodegradable qualities that is derived from biological polymers. It is possible to obtain this biopolymer from agricultural products. Biodegradable plastic can be made from agricultural goods such as food crop waste that has a high starch or cellulose content (Purwandari, Susanti, Suparno, & Aji, 2019). Various plant species that provide essential nutrients like proteins and carbohydrates—which provide people with energy—are referred to as food crops (Morris & Mohiuddin, 2023). These food crops are divided into three categories: cereals (rice, wheat, and corn), tubers (sweet potatoes and cassava), and legumes (peanuts, soybeans, and long beans) (Rozi, et al., 2023).

This study introduces a novel approach by utilizing hemicellulose extracted from agricultural waste, specifically straw and rice husk, as the primary material for biodegradable plastic production. Unlike conventional methods that rely on synthetic or food-based biopolymers, this research leverages abundant and underutilized agricultural residues, offering a sustainable and eco-friendly solution. Additionally, the synthesis process incorporates innovative techniques to maximize hemicellulose yield while maintaining its structural integrity, paving the way to produce high-performance renewable plastics with enhanced biodegradability.

METHOD

The Materials and Equipment

Indramayu agricultural products provide the raw ingredients for straw and rice husk. After being cleaned and dried, the straw is pulverized and sieved through an 80-mesh screen. The following chemicals are utilized in the process: glycerol, CMC, hydrogen peroxide, and NaOH. A 25 cm x 15 cm mold, a hotplate, a magnetic stirrer, an 80-grit filter, and a blender were among the equipment utilized in this investigation.

Research Procedures

There are three steps involved in turning straw and husk waste into bioplastic: pretreatment, delignification, dehemicellulose, bleaching, and sample printing. In order to speed up the delignification process, straw is crushed during the preparation phase. A blender is used to grind (Wang, Asano, Kudo, & Hayashi, 2020). The straw is ground until its mesh size reaches 80. The alkali delignification process is then used to delignify the refined straw and husks. 25 grams of straw/husk are weighed and processed right away in the hydrolysis step using a 15% NaOH solution (30 grams of NaOH dissolved in 200 mL of distilled water) at 100 °C for one hour. Following that, the cellulose solids are dried for three hours at 50 °C in an oven (Ashgar, et al., 2015). Bioplastics are made by soaking cellulose in 10% acetic acid (200 ml) and 20 grams of NaCl for 60 minutes at 60°C to make the fibers smoother and cleaner (Mohammed, et al., 2022). The cellulose is filtered, rinsed with clean distilled water, and dried in an oven.

The cellulose bleaching procedure is then carried out for 1 hour at 60°C with 7% H_2O_2. Bioplastic is formed by combining cellulose, CMC, and glycerol in a 1:1:1 ratio. The solution is agitated and heated at 90°C for 10-15 minutes, until it thickens. This bioplastic solution is then moulded to the size of 25 cm x 15 cm with a thickness of 0.2 cm and dried at 60°C till a bioplastic sheet form.

Morphology and Functional Group Test

Scanning Electron Microscope (SEM) test was conducted to determine the surface morphology of cellulose samples. The resulting cellulose samples were then characterized by FTIR spectrophotometer and tested to analyze their functional groups.

RESULTS AND DISCUSSION

Cellulose Delignification Process from Rice Straw

To dissolve cellulose from plant sources, the lignin in the plant cell walls must first be removed through a process called delignification. Delignification facilitates the separation of cellulose by destroying the lignin that binds it. In the isolation of cellulose from rice straw, the use of bases, namely using NaOH with a concentration of 15%, is carried out. Recent research on delignification focuses on straw; the use of NaOH solution in the delignification process can dissolve cellulose to produce alpha-cellulose (Rivai, et al., 2021).

The delignification process consists of three stages based on cellulose retention. In the early stage (delignification rate <0.4), the cellulose loss is very small, which is around 0.1 or lower. This loss is mainly due to noncrystalline cellulose. At this stage, rice straw and husk experience slow lignin removal, with minimal cellulose loss. This cellulose loss is mainly from the noncrystalline cellulose part, which is more easily degraded than crystalline cellulose. In the middle stage (delignification rate 0.4–0.65), delignification proceeds with little cellulose loss, with cellulose retention in the narrow range of 0.80–0.85 (entries 1, 3–9). This pattern indicates the progress of selective delignification. At this stage, delignification becomes more selective, with more efficient lignin removal without much impact on cellulose. Cellulose retention ranges from 80–85%. At the final stage (delignification rate >0.65), delignification is accompanied by significant cellulose loss (Asano, University, & Kudo, 2020).

At this stage, the remaining lignin tends to be in areas that are difficult to access, such as in small pores or tightly bound to cellulose. As a result, oxidizing species also attack cellulose, causing significant cellulose loss. In the delignification process of rice straw and rice husk, control of the delignification rate is critical to ensure maximum lignin removal while maintaining cellulose retention. The middle stage is usually the optimum point for obtaining a material with a high cellulose and low lignin content, which is ideal for applications such as bioplastic production. The final stage needs to be avoided or minimized to prevent significant losses of cellulose, which is the main component in bioplastics (Wang, Asano, Kudo, & Hayashi, Deep Delignification of Woody Biomass by Repeated Mild Alkaline Treatments with Pressurized O2, 2020). Based on the research results, it was found that NaOH with a concentration of 15% has great effectiveness in isolating cellulose with a cellulose yield of 33.63%.

Addition of Acetic Acid and NaCl

The dehemicellulose process was carried out using a 10% acetic acid solution and NaCl, because hemicellulose is easily dissolved and hydrolyzed by mineral acids. Furthermore, the sample was filtered to separate cellulose in solid form from hemicellulose. Hemicellulose is easier to process and more flexible than cellulose, so it can provide more flexible properties to bioplastics. Hemicellulose can also function as a good binder in bioplastic formulations and can increase the ability of bioplastics to absorb air (Stepan, 2013).

Bleaching Process using H_2O_2

As seen in Figure 1, throughout this process, the solvent's color shifts from clear to white, and the sample's color changes from dark brown to white.

(a) (b) (c)

Figure 1. Prior to the bleaching procedure, (a) rice straw hemicellulose, (b) rice husk hemicellulose, and (c) rice straw and husk hemicellulose following the bleaching process.

H_2O_2 is introduced along with oxygen and NaOH during the alkaline extraction, or EOP, step so that it reacts in a basic environment. The goal of this extraction procedure is to dissolve the color-causing substances, which are probably soluble at medium-to-high temperatures in alkaline solutions. Using the characteristics of the chemicals, this is accomplished as part of the bleaching process. Hydrogen peroxide also has a number of benefits, such as producing bleached pulp that is extremely durable and only slightly weakens the fibers (Çiçekler, Tutus, & Beram, 2023).

Hydrogen peroxide is very stable in acidic conditions, but easily decomposes in alkaline conditions. The decomposition process is faster and produces oxygen that expands and creates foam with increasing temperature according to figure 2. The decomposition reaction of H_2O_2 that occurs is as follows:

$$H_2O_2 \rightarrow \frac{1}{2} O_2 + H_2O$$

Figure 2. Bleaching Process using H_2O_2

Characteristics of Rice Straw and Rice Husk Cellulose

The surface shape of rice husk and straw prior to the delignification process is shown in Figure 3. The surface of rice husk and rice straw seems to be solid, smooth, and non-hollow. Plant cell walls are filled with lignin, which is shown by this surface (Balk, Sofia, Neffe, & Tirelli, 2023). Following the delignification process, the product's lignin content dissolves in NaOH, resulting in a loose, hollow, and rough surface, as seen in Figures 3(a), 3(c) (pineapple leaves) and 4(b), 4(c) (straw). According to the findings of the SEM examination, delignification breaks down and eliminates the cellulose's exterior layer while exposing its interior.

The results and discussions contain the results of research findings and their discussion scientifically. Write down scientific *findings* obtained from the research results that have been carried out but must be supported by adequate data. The scientific findings referred to here are not the data of the research results obtained.

An overview of the chemical structure of hemicellulose, which is dominated by polysaccharide groups, may be seen in the FTIR spectrum displayed in Figure 4. There is a large peak in the 3000–3500 cm^{-1} range that shows absorption from the O–H group (stretching). The hydrophilic nature of hemicellulose, which is abundant in intramolecular and intermolecular hydrogen bonds—a common property of polysaccharides—is reflected in this hydroxyl group.

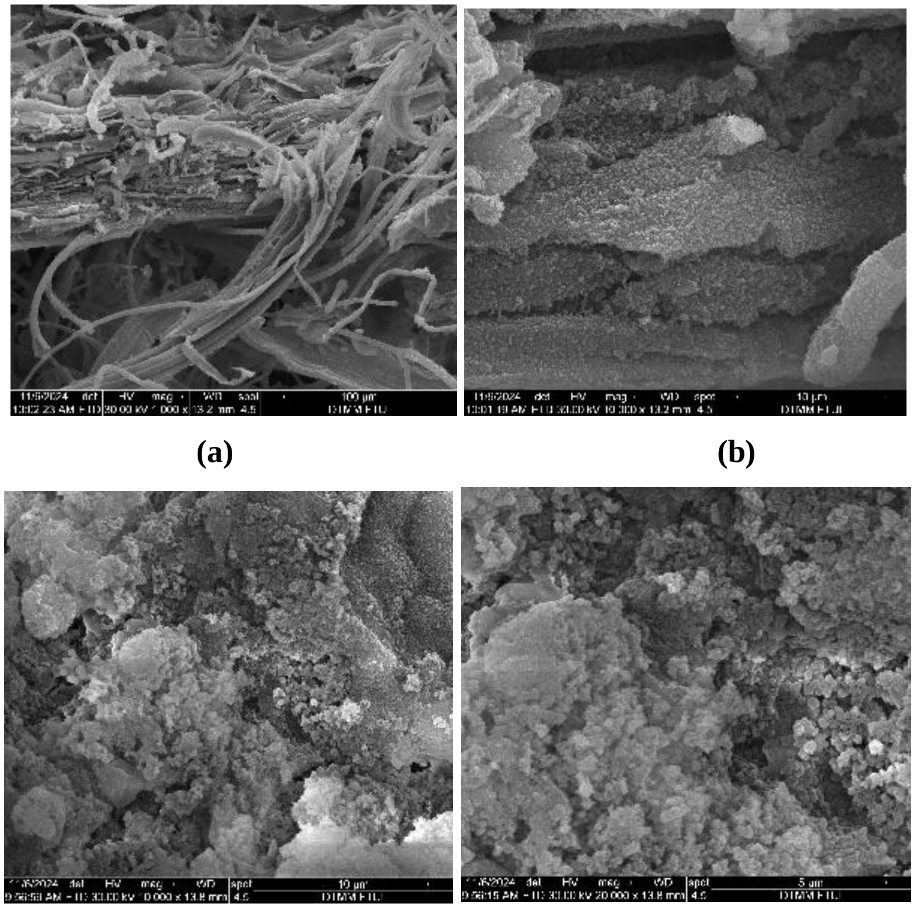

Figure 3. SEM Results (a) Rice Straw Hemicellulose Magnification 100µ and (b) Magnification 10µ, (c) Rice Husk Hemicellulose Magnification 10µ (d) Rice Husk Hemicellulose Magnification 5µ

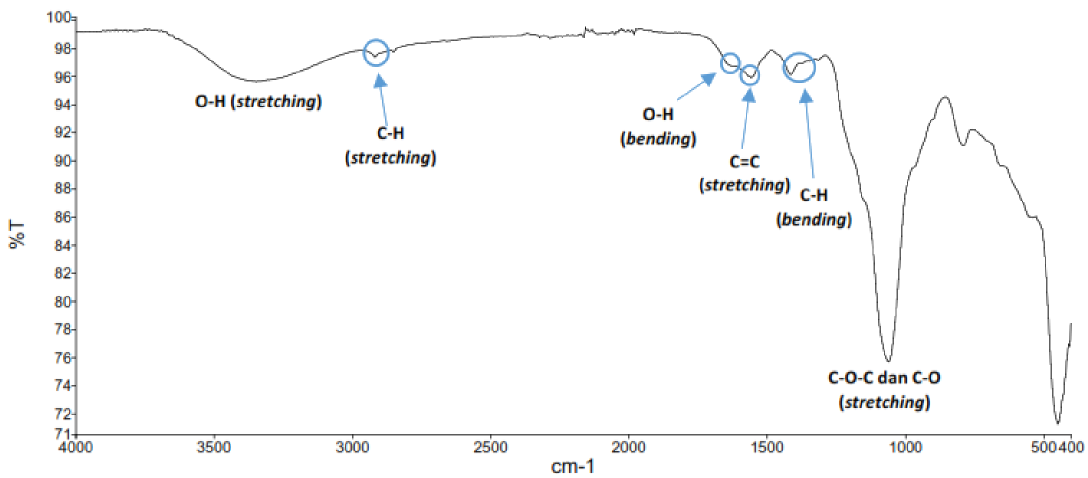

Figure 4. FTIR Graph of Rice Husk Cellulose

In the region of 2800–2900 cm^{-1}, a small peak appears related to the C-H group (stretching). This group usually comes from an alkane group, such as methyl or methylene, which is part of the carbon chain in hemicellulose. Around 1600–1650 cm^{-1}, there is a peak reflecting O-H (bending). This indicates the presence of bound water in the hemicellulose structure, indicating that the sample has hygroscopic properties, which are common in lignocellulosic materials. The peak at 1500–1600 cm⁻¹ indicates C=C (stretching), which refers to the possible presence of residual aromatic structures from lignin.

Table 1. FTIR Frequency Data of Rice Husk Hemicellulose

Functional Groups	Group	Frequency area (cm^{-1})	Intensity
Alcohol	O-H (stretching)	3000 – 3600	Medium, Wide
Alkane	C-H (stretching)	2800 – 2900	Very Weak, Medium
H_2O	O-H (bending)	1600 - 1650	Weak, Wide
Aromatic	C=C (stretching)	1500 – 1600	Medium, Medium
Alkane	C-H (bending)	1300 – 1450	Medium, Medium
Polysaccharides	C-O-C dan C-O (stretching)	900 – 1200	Strong, Sharp

Although hemicellulose has been isolated, lignin which has a complex aromatic structure is often difficult to separate completely. The region of 1300–1450 cm^{-1} shows C-H bending, which reflects alkane groups that are common in organic structures. Meanwhile, at 900–1200 cm^{-1}, there are very sharp peaks, reflecting C-O-C (stretching) and C-O (stretching), which are characteristic of glycosidic bonds in polysaccharides. This region is a major indicator of the structure of hemicellulose, which consists of monosaccharide units connected by glycosidic bonds.

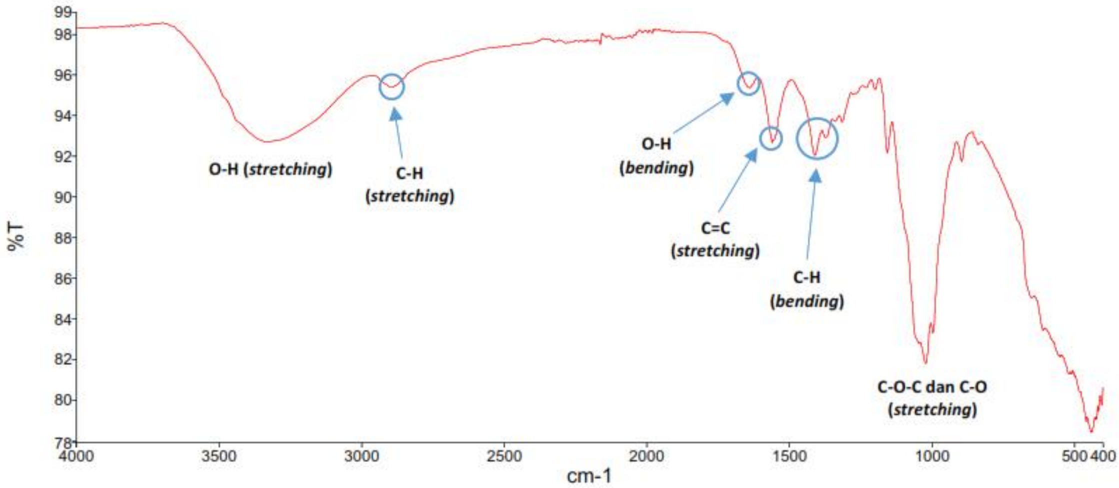

Figure 5. FTIR Graph of Rice Straw Cellulose

The FTIR spectrum shown in Figure 5 of hemicellulose isolated from rice straw shows several main peaks reflecting the chemical characteristics of this compound. In the region of 3000–3500 cm^{-1}, there is a strong broad peak, indicating the presence of hydroxyl groups (O-H stretching). This group is the main characteristic of hemicellulose because of its nature as a polysaccharide with many hydroxyl groups that play a role in forming intermolecular hydrogen bonds. Around 2800–2900 cm^{-1}, there is a relatively small peak, identified as C-H stretching of the alkane group. This group is likely derived from the carbon chain of hemicellulose or other related compounds. In the region of 1600–1650 cm^{-1}, there is a moderate peak reflecting O-H bending, indicating the presence of bound water in the hemicellulose structure. This indicates the hygroscopic nature of hemicellulose, which is able to absorb and retain water molecules. The sharp peak at 1500–1600 cm^{-1} indicates C=C stretching, indicating the presence of aromatic structures. In this context, this peak may originate from the remaining lignin in the hemicellulose sample.

Table 2. FTIR Frequency Data of Rice Straw Hemicellulose

Functional Groups	Group	Frequency area (cm^{-1})	Intensity
Alcohol	O-H (stretching)	3000 – 3500	Medium, Wide
Alkane	C-H (stretching)	2800 – 2900	Very Weak, Medium
H_2O	O-H (bending)	1600 - 1650	Weak, Moderate
Aromatic	C=C (stretching)	1500 – 1600	Medium, Sharp
Alkane	C-H (bending)	1300 – 1450	Medium, Wide
Polysaccharides	C-O-C dan C-O (stretching)	900 – 1200	Strong, Sharp

Lignin is a complex aromatic compound that is often difficult to remove completely from lignocellulosic materials. The 1300–1450 cm^{-1} region shows a moderate peak identified as C-H bending, which is related to the bending of the alkane groups in the polysaccharide structure. The very prominent sharp peaks in the 900–1200 cm^{-1} region reflect C-O-C and C-O stretching, which are characteristic of glycosidic bonds and alcohol groups in hemicellulose. This region is very important for identifying polysaccharides because the basic structure of hemicellulose consists of monosaccharide units connected by glycosidic bonds.

Analysis of Bioplastics Made from Straw and Rice Husks

The rice straw-based bioplastic shown in Figure 6 has a surface that tends to be rough and uneven, with a fairly transparent yellowish colour. The surface shows the presence of straw fibres that do not appear to be evenly distributed in the bioplastic matrix. This can occur due to the mixing of materials that is not homogeneous during the manufacturing process. In certain parts, there are visible discoloration or brownish stains, which are likely the result of thermal reactions or material degradation during processing (Rahmatullah, et al., 2024).

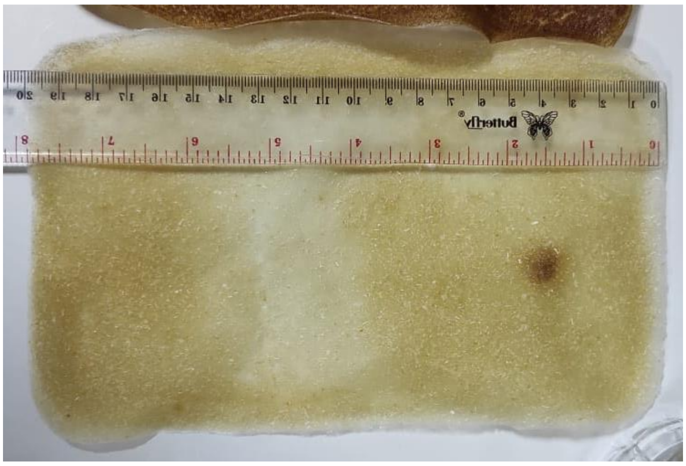

Figure 6. Bioplastic made from rice straw

In terms of structure, the thickness of the bioplastic appears to be non-uniform in some areas, which can affect its mechanical properties and water absorption. This varying thickness may be caused by less-than-optimal moulding or drying techniques. In addition, the rough texture can increase the porosity of the material, which ultimately has the potential to increase water absorption and reduce its resistance to moisture (Boey, Keong, & Tay, 2022).

Overall, this bioplastic shows potential for use, but its quality can still be improved. The homogeneity of straw distribution and control of the thermal process are important aspects that need to be improved to produce more uniform bioplastics with better mechanical properties and resistance. Additional tests such as mechanical tests, water absorption tests, and thermal resistance tests can provide further information on the performance of this material.

Figure 7. Bioplastic made from rice husks

Based on Figure 7, rice husk-based bioplastic, here is the morphological analysis:

The surface of the bioplastic appears to have a rough texture with an uneven pattern, which is caused by the less homogeneous distribution of rice husk particles in the bioplastic matrix. This factor occurs due to imperfect mixing of materials or varying rice husk particle sizes. The rough texture of this bioplastic indicates a challenge in creating a uniform structure, which can affect the mechanical properties and water absorption of the bioplastic (Boey, Keong, & Tay, 2022).

Visually, the bioplastic has a yellowish color that reflects the natural character of rice husk and the influence of additional materials such as glycerin which functions as a plasticizer. This color can also be influenced by thermal reactions during the bioplastic formation process, such as heating, which may cause chemical changes to the material. The thickness of the bioplastic appears to vary, indicating irregularities in the molding or drying process. This can affect the mechanical properties of the bioplastic, such as stiffness and flexibility. The bioplastic appears to be quite thick, which may indicate good mechanical strength, although its flexibility could be improved with the addition of plasticizers. However, the uneven texture and uneven particle distribution may cause weak points in the material, potentially reducing its durability. In addition, the porosity resulting from the rough morphology may increase water absorption, so it is worth considering in applications that require resistance to moisture.

Overall, the morphology of this bioplastic requires further processing of bioplastic production related to rice husk-based materials to achieve homogeneous distribution. Further analysis through mechanical tests, water absorption, and thermal stability are needed to provide a deeper understanding of the performance of this material and how improvements can be made to improve the quality of bioplastics.

CONCLUSION

This study effectively revealed the huge potential of rice straw and rice husk as primary raw materials for the manufacturing of environmentally friendly bioplastics. The alkalinization procedure was used to extract hemicellulose, resulting in materials with typical polysaccharide chemical properties such as O-H, C-H, and C-O-C groups, as well as signs of the existence of lignin residues. Bioplastics were created by processing hemicellulose and adding glycerine, CMC, and TiO_2 to enhance mechanical performance and biodegradability. The study's findings revealed that bioplastics have both outstanding mechanical performance and improved biodegradability, making them a viable alternative to traditional plastics. This strategy provides a unique option for utilizing agricultural waste, such as rice straw and rice husks, adding value to previously regarded useless resources. Furthermore, this work contributes significantly to the development of green material technology that benefits the environment while tackling the issues posed by rising plastic waste levels.

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