

DOI <http://dx.doi.org/10.36722/sst.v8i3.1885>

# Physical Properties of Bioplastic Composite Synthesized from Sago Waste and Betung Bamboo Activated Carbon

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**Abstract** – Bioplastics are expected to reduce the use of commercial plastics. Several efforts have been made to improve the properties of bioplastics, including the addition of antioxidants or antimicrobial agents. One of the antimicrobial agents is activated charcoal or activated carbon. This study was aimed to investigate the effect of activated carbon addition to the bioplastic composite. Sago waste was utilized as the matrix in the composite, while activated carbon was as filler. Sago waste is the leftover biomass resulted from the sago extraction. The proximate analysis showed that the composition of sago waste was mostly dominated by the fibrous materials followed by protein and fat. The activated carbon was obtained from the treatment of Betung bamboo. The Fourier Transform Infrared (FTIR) spectroscopy indicated that the thermal treatment of Betung bamboo at 175 °C for 5 h resulted. The treated sago waste was immersed in 5% KOH for 24 h and further dried and then pyrolyzed at 750 °C for 90 min to yield activated carbon. The activated carbon was dominated by the carbonaceous materials. This was also supported by the proximate data indicating the presence of 91% carbonaceous materials in activated carbon. The Brunauer Emmett Teller (BET) analysis showed that the activated carbon has surface area of 460 m<sup>2</sup>/g. This activated carbon was then used as filler at bioplastic composites. Two levels of activated carbon percentage on composite were tested, which were 3-5% (% m/v). The mechanical analysis using universal testing machine indicated that the addition of activated carbon reduces the tensile strength and modulus of elasticity. The scanning electron microscopy (SEM) images clearly indicated the presence of activated carbon on the surface of bioplastics. The data obtained in this study showed the potential of composite sago waste and activated carbon as bioplastics. Further, the activated carbon obtained in this study can serve as an adsorbent, providing possible applications as antibacterial agent.

**Keywords** – Activated Carbon, Biomass, Bioplastic, Composite, Pyrolysis, Sago Waste.

## INTRODUCTION

Commercial plastics are mostly synthesized through petrochemical route involving fossil fuel. Thus, they are considered as environment unfriendly. Bioplastic can be an alternative for this as it is synthesized from natural polymer that can be easily degraded by microorganisms [1]. Bioplastics are divided into three categories based on their raw materials, namely hydrocolloids (proteins, polysaccharides), lipids and composites [2]. Sago, the agricultural products, contains hydrocolloids. Sago flour usually consumed is produced from sago plants through an extraction process, the result is

starch (from the polysaccharide class) which will be processed into flour and sago waste known as repu. Utilization of repu is still very limited and sometimes discarded so that it can become waste that causes environmental pollution. The repu content component of the proximate analysis results has a crude protein content of 2.1%; fat 1.8%; crude fiber 20.3%; gray 4.6%; cellulose 36.3%; hemicellulose 14.6%; lignin 9.7%; and 3.3% silica [3].

The hydrocolloid has thermoplastic, elastomeric, thermoset, and adhesive functional properties [4]. Hydrocolloid based bioplastics generally have selective gas barrier properties (such as O<sub>2</sub> and

CO<sub>2</sub>), efficient inhibitors of oils and lipids, as well as good mechanical properties [5]. However, hydrocolloid-based materials are categorized as water sensitive films with low moisture resistance performance [5]. In the application of bioplastics for food packaging, these characters will negatively influence the packaging property as this condition can provide opportunities for the growth of pathogenic microorganisms [6]. Modification of bioplastics with the addition of antioxidants or antimicrobial agents into the film solution is expected to minimize this probability. Modified bioplastics will have properties as active coatings that can protect food products from oxidation and decomposition by microbes, the result improves visual quality as well as improved product safety [7]. One of the natural materials that has the potential to overcome the performance of films that are low in resistance to moisture is activated charcoal which has hygroscopic properties or is able to absorb water.

Activated charcoal is carbon or charcoal that has undergone an enlarged pore or surface area through the activation process. Activated charcoal from oil palm shells to act as a good adsorbent for aqueous solutions and some gaseous pollutants [8]. This property is obtained from the activation process of activated charcoal through physical methods or chemical methods with chemical activators. The hygroscopic nature of activated charcoal used as an adsorbent can also overcome the low performance of hydrocolloid-based bioplastics in terms of moisture resistance, so that they can make bioplastics as active coatings that have the potential to have antibacterial properties.

Several studies have reported that the adsorbent properties of activated charcoal are influenced by its porosity and surface area. Pore size and morphology were reported to have an effect on the adsorption of toxins and *E. coli* using the diarrhea drug metronidazole in combination with activated charcoal from the palm fronds [9]. The selection of bioplastic composition by determining the appropriate amount of activated charcoal addition is deemed necessary to be considered, in order to achieve the goal of adding activated charcoal as an antibacterial in bioplastics. The use of bamboo charcoal concentrations of 2% and 5% in solution during incubation of mutant *Streptococcus* showed suppression of bacterial growth [10]. These results indicate that the nature of the adsorbent already possessed by bamboo charcoal from the carbonization stage. The initial process with hydrothermal carbonization is good enough to bring

out the hygroscopic properties of the activated charcoal raw material [11]. Charcoal absorptivity can be increased based on the increase in the pore volume of the charcoal surface through chemical activation [12]. Based on these studies, the addition of activated charcoal will be carried out with several variations in concentration, namely 3%; 4 %; and 5% to test the antibacterial activity of bioplastic composites and activated charcoal from bamboo against gram-negative *E. coli* and gram-positive *S. aureus*.

## METHOD

### Materials

The main raw material of this research is sago waste (Riau, Indonesia). Another material is carrageenan from PT Agro Permata Indah 21 (Bogor, Indonesia), sembilang bamboo from RC for Biomass and Bioproducts-BRIN (Cibinong, Bogor), sorbitol, surfactant tween-80, aquades, NaOH, HCl and KOH. The other materials for antibacterial testing are *S. aureus* and *E. coli* from BRIN Biomaterial (Cibinong, Bogor).

### Methods

#### Sago waste protein extraction

The protein extraction process is by making a solution of 40 g of sago waste powder which has gone through a 74-mesh sieve process and then dissolved in 360 ml of 25 mM NaOH. This mixing was carried out using a hot plate stirrer at a speed of 120 rpm for 8 hours at a temperature of 25 °C. After 2 h stirring process, the pH of the solution was adjusted to 10 by adding 2 M NaOH. Further, the solid liquid separation was carried out through centrifugation of the solution for 15 min with a rotating speed of 10,000 rpm. This process will produce protein containing supernatant (liquid phase), which was the result of extraction, and precipitate (solid phase). The supernatant was further stored at the fridge for further bioplastic synthesis.

#### Bamboo based activated charcoal production

Activated charcoal was obtained through two step processes which were hydrothermal carbonization and activation [8], [9]. Before going through the hydrothermal carbonization process, the bamboo was firstly cleaned first and then dried in an oven at a temperature of 105 °C for 24 h to remove the moisture then ground and mashed. The precursor in the form of bamboo powder was then placed in the reactor. For this, the reactor was filled with distilled

water up to a third of the reactor volume. While the precursor was weighted at 15% of the weight of the water. This set up was conducted in order to ensure that the pressure in the reactor was approximately 1 atm. The hydrothermal carbonization was conducted at 225 °C for 8 h at autogenous pressure (pressure generated independently by water vapor in the reactor). This step resulted what so called hydrochar. To obtain activated carbon, this hydrochar was further immersed in immersed in 5% sodium hydroxide solution with a mass ratio of hydrochar to NaOH is 1:20 for 24 hours. Further, it was rinsed until close to neutral pH. This immersed hydrochar was further activated through pyrolysis carbonization at 750 °C for 90 min.

The activated charcoal obtained from previous pyrolysis carbonization was then involved in bioplastic synthesis. The activated charcoals were varied on their concentrations and particle size. To obtain the intended particle size, the activated carbon was ground and sieved at 120 and, 170230 mesh. These sizes correspond to particle size of 125 and 88 µm, respectively.

### Bioplastic synthesis

Bioplastic was synthesized through casting solution method [13]. First, as much as 2 g carrageenan powder was added into 1% acetic acid. Second, this mixture was stirred at room temperature, 300 rpm, for 3 h. Third, as much as 40 ml of this solution was mixed with 60 ml of protein containing supernatant (See Section sago waste protein extraction). The mixing was conducted at 40 °C with 200 rpm. After 20 min of mixing, a 4% sorbitol and activated charcoal was added to this mixture. In this study, the effect of concentration and particle size of activated charcoal on bioplastic properties was investigated (**Table 1**).

Table 1. Sample code of bioplastic

Sample Code	Activated Carbon Concentration (%)	Activated Carbon Particle Size (µm)
A00	0	-
A11	3	125
A12	3	88
A21	4	125
A22	4	88
A31	5	125
A32	5	88

Then 3% tween-80 was added to the solution and then sonicated using an ultrasonic processor for 60 minutes at a frequency of 20 kHz and an amplitude of 40%. The sonicated film solution was heated for 10 minutes at 50 C. The film solution was then

molded using the solution casting method. A total of 10 ml of sonicated film emulsion was then poured onto the mold. This process is repeated up to three pours every 5 hours until the entire solution has been poured. Limiting the number of layers is very necessary, because the more volume and number of layers will affect the physical and mechanical properties of the film to be produced [13], [14]. Synthesis of bioplastic films without the addition of activated charcoal will also be carried out as standard bioplastic samples.

### Properties of sago waste and activated carbon

Approximate testing was performed to determine the properties of sago residue and also tested the quality of activated charcoal according to SNI 06-3730-1995 on the properties of technical activated charcoal.

### Physical properties characterization

Physical properties were evaluated by Fourier Transform Infra Red (FTIR) and Scanning Electron Microscopy (SEM). The FTIR analysis was conducted to identify the functional groups that exist in bioplastic. The SEM analysis was to observe the surface structure and homogeneity of the bioplastic produced. All bioplastic samples were prepared in a 1.5 cm x 1.5 cm for the characterization.

### Mechanical properties characterization

The mechanical test aims to determine the value of tensile strength, modulus young and percentage of elongation. Edible films were tested using Toorsee's Electronic System Universal Testing Machine with ASTM D 882 standard and a speed of 5 mm/minute.

### The Brunauer Emmett Teller (BET) analysis

The Brunauer Emmett Teller (BET) analysis was conducted to determine the value of surface are of activated carbon.

### Antibacterial properties analysis

Tests of antibacterial activity of edible films were performed on two types of bacteria, namely gram negative (*E. coli*) and gram positive (*S. aureus*). That method used is the disc diffusion assay method.

## RESULT AND DISCUSSION

### Properties of Sago Waste and Activated Carbon

Proximate testing of sago waste was carried out to determine the content of fiber, protein, and fat content. At the same time, the proximate test on Betung bamboo and its charcoal was carried out to determine the characteristics of the charcoal, which

consisted of water, ash, volatile, fixed carbon, and iodine adsorption capacity, and methylene blue adsorption capacity. The results of the proximate test on sago waste are shown in **Table 2**, while the proximate test on Betung bamboo and its charcoal is in **Table 3**.

Table 2. Proximate test results of sago waste

No	Proximate Analysis	Value (% wt)
1	Fat	4.675
2	Protein	5.46
3	Fiber	7.90

Based on the results of the proximate test (**Table 2**), sago waste is dominated by fiber (7.90%), followed by protein (5.46%) and fat (4.675%). This study is in line with previous study stating that the content of sago waste is dominated by fiber (12.44%), followed by protein (3.38%), and fat (1.01%) [15].

Proximate analysis was carried out to estimate the performance of the fuel during the heating and combustion process [16]. The results showed that the water content of Betung bamboo, hydrochar, and activated charcoal, respectively, was 9.17 %; 4.91% and 1.79% met the Indonesian standard for activated charcoal properties (<15%).

The lowest water content value was obtained in activated charcoal samples, indicating the influence of the higher temperature and activation process during the activated charcoal synthesis process. The ash content of Betung bamboo, hydrochar, and activated charcoal was 0.71%, 0.21 % and 5.27 %, respectively, and met the Indonesian standard for activated charcoal properties (<10%). Just like water, ash is a non-flammable component. With a low ash content, activated charcoal can be good for fuel needs.

The levels of volatile matter of Betung bamboo, hydrochar, and activated charcoal are 83.73%, 81.94%, and 31.40%, respectively, while fixed carbon content was 15.57 %, 17.85%, and 91.55%, respectively. All of the materials did not met the standard for volatile matter, and only activated charcoal met the standard of fixed carbon (>65%). Both volatile matter and fixed carbon are flammable components. With a high fixed carbon content, it is expected to produce materials that have good fuel properties.

The carbonization process in bamboo to convert it into hydrochar and activated charcoal is carried out as a thermal process, in which dehydration or reduction of water (H<sub>2</sub>O) occurs, breaking bonds, and in the end, what remains is the element carbon [11]. It is reinforced by the results of Fourier Transform Infrared (FTIR) testing, as shown in **Figure 1**.

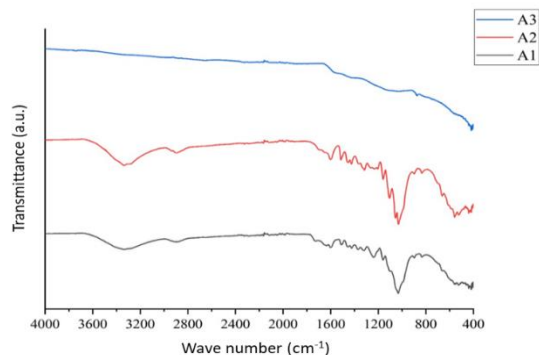


Figure 1. Infrared spectrum of Betung bamboo (A1), hydrochar (A2) and activated carbon (A3)

In **Figure 1**, the changes in functional groups significantly in activated charcoal (A3) compared to Betung bamboo (A1) and hydrochar (A2). Based on research (Budiman et al., 2019), there is a reduction of the -OH group at a wave number of 3600-3000 cm<sup>-1</sup>, and the C=O strain bonds are mainly from esters, carboxylic acids, or aldehydes from cellulose and hemicellulose, and lignin at numbers wave 1800-1650 cm<sup>-1</sup>.

The iodine number shows the porosity of the material and its ability to adsorb other materials, especially those that are a pollutant. On the other hand, the methylene blue number indicates the ability of a material to adsorb dyes. The iodine number of Betung bamboo, hydrochar, and activated charcoal were 313.01mg/g, 308.75 mg/g, and 636.72 mg/g %, respectively. On the other hand, the methylene blue number of Betung bamboo methylene blue, hydrochar, and activated charcoal were 10 mg/g, 19 mg/g, and 197.39 mg/g %, respectively. It shows that activated charcoal has the highest ability to adsorb pollutants and dyes. Activated charcoal has reasonably good porosity, although the iodine number still does not meet the standard.

Table 3. Proximate test result of Betung bamboo and its charcoal

Materials	Water Content (%)	Ash Content (%)	Wolatile Matter (%)	Fixed Carbon (%)	Iodine Number (mg/g)	Methylene Blue Number (mg/g)
Betung bamboo	9.17	0.71	83.73	15.57	313.01	10.00
Hydrochar	4.91	0.21	81.94	17.85	308.75	19.00
Activated carbon	1.79	5.27	31.40	91.55	636.72	197.39
<b>*SNI No. 06.3730-1995</b>	<b>&lt;15</b>	<b>&lt;10</b>	<b>&lt;25</b>	<b>&gt;65</b>	<b>&gt;750</b>	<b>&gt;120</b>

\*SNI: Indonesian National Standard

The results of iodine number and methylene blue number are related to the BET surface area of Betung bamboo activated carbon (460.5309 m<sup>2</sup>/g) compared with the raw Betung bamboo (0.4646 m<sup>2</sup>/g) and Betung bamboo hydrochar (0.7556 m<sup>2</sup>/g). Similar to BET surface area, the nanoparticle size of activated charcoal is also the best (13.03 nm) compared with Betung bamboo and hydrochar's nanoparticle size (12914 nm and 7940 nm, respectively).

Table 4. Brunauer Emmett Teller (BET) analysis of Betung bamboo and its chars

No	Materials	Surface Area (m <sup>2</sup> /g)	Nanoparticle Size (nm)
1	Betung bamboo	0.4646	12914.01
2	Hydrochar	0.7556	7940.34
3	Activated carbon	460.5309	13.03

**Mechanical properties of biofilm**

The mechanical properties of the biofilm in the form of tensile strength and modulus of elasticity are shown in **Table 5**. **Table 5** shows that the biofilm without the addition of activated charcoal has the highest tensile strength and modulus of elasticity compared to the biofilm with the addition of activated charcoal. It occurs because the addition of charcoal causes the reduction of hydrogen bonds in the biofilm, resulting in lower strength, both tensile strength and modulus of elasticity.

Table 5. Mechanical properties of biofilm

No	Code Sample	Tensile Strength (MPa)	Modulus Elasticity (GPa)
1	A00	0.38	0.1
2	A11	0.22	0.0020
3	A12	0.18	0.0029
4	A21	0.28	0.0055
5	A22	0.30	0.0045
6	A31	0.26	0.0041
7	A32	0.19	0.0029

When compared between biofilms with the addition of activated charcoal, it can be seen that there is a tendency for biofilms with the addition of finer-sized activated charcoal to have higher strength. It happens because activated charcoal with a small size has a larger surface area that binds to other biofilm-forming materials.

**Antibacterial properties analysis**

Table 6 indicated the antibacterial properties of three bioplastics to *E. coli* and *S. aureus*. The involvement of activated carbon showed the potential of antibacterial property, particularly for A11. This sample was showed to be able to inhibit the growth of *E. coli* and *S. aureus*. While these phenomena were absence in sample A00.

Table 6. The zone of inhibition of bioplastic

Sample Code	Zone of Inhibition (cm)	
	<i>E. coli</i>	<i>S. aureus</i>
A00	0	0
A11	0.50 ± 0.07	0.79 ± 0.07
A12	0.47 ± 0.06	0

**Morphological properties of bioplastic**

The morphological appearance of the biofilm surface with the addition of activated charcoal in various compositions and sizes are shown in Figure 2.

Based on Figure 2, it can be seen that the surface of the biofilm without activated charcoal (A00-1 and A00-2) looks smoother and more uniform than the surface of the biofilm with the addition of activated charcoal. In addition, the appearance of biofilm with smaller activated charcoal addition (A11, A21, and A31) also has a more even appearance when compared to the surface of the biofilm with the addition of more oversized activated charcoal (A12, A22, and A32). It happens because the activated charcoal particles with a finer size can be more evenly distributed during biofilm manufacturing.

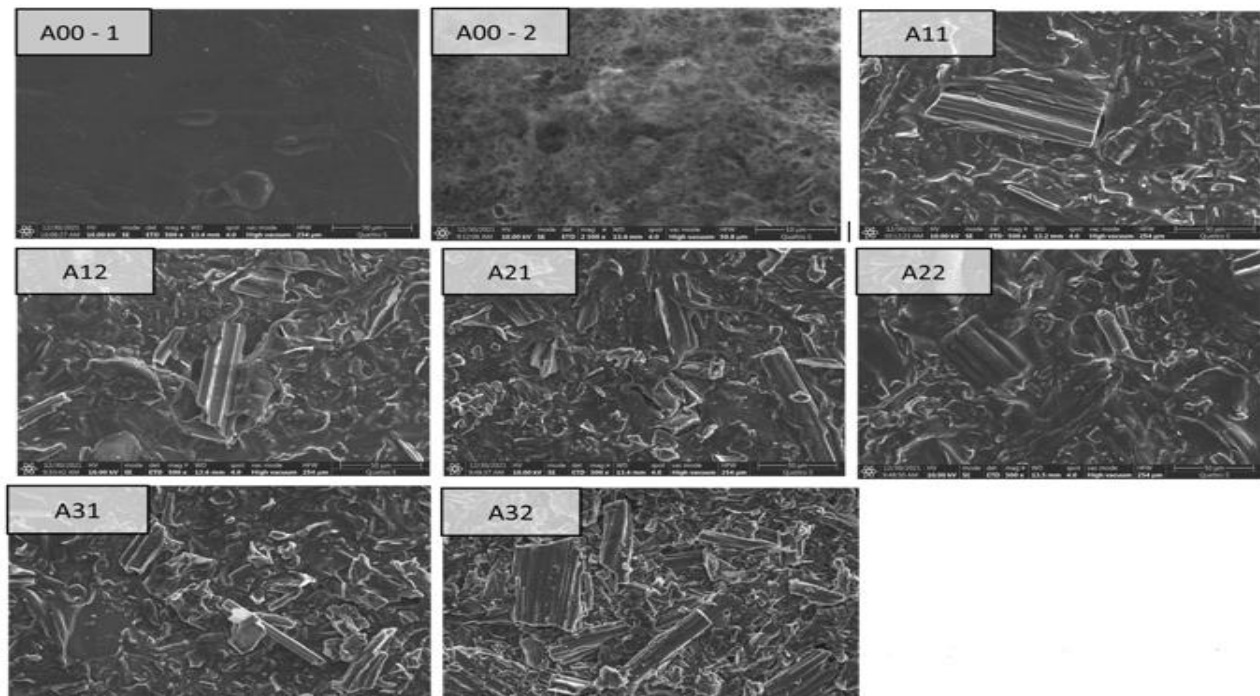


Figure 2. Morphological of biofilm surface obtained form

## CONCLUSION

Activated charcoal made from Betung bamboo through a hydrothermal process and activation using 5% NaOH has good properties by meeting most of the standards for activated charcoal properties, namely water content, ash content, fixed carbon, and methylene blue number. Activated charcoal addition to the biofilm can increase the roughness on the surface of the biofilm. Activated charcoal with a finer size has well dispersed than the coarser size in the biofilm. Meanwhile, the addition of activated charcoal can reduce the biofilm's tensile strength and modulus of elasticity because the hydrogen bonds that contribute to the strength of the biofilm decrease with the addition of activated charcoal. The resulted bioplastic, particularly the one containing activated charcoal with concentration of 3% (wt) and 125  $\mu\text{m}$  in particle size showed potential antibacterial property against *E. coli* and *S. aureus*.

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