



The Heat Resistant Capabilities of Bioplastic Composites on Sago Hampas Starch- Al_2O_3

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Abstract

Sago hampas are a waste with high starch content. The starch from sago hampas can be used as a raw material for bioplastics. However, because bioplastics have a lower heat resistance than conventional plastics, additives are required to increase heat resistance. Aluminum oxide can be used as a metal compound that acts as an additive to increase heat resistance. Bioplastics were created using a weight percentage of 0, 1, 3, and 5% Al_2O_3 . Analysis was carried out with DTA to determine the melting point of bioplastics. Furthermore, bioplastics tested the mechanical properties, density, and water resistance. The best results were identified using FTIR and SEM. The results showed that adding Al_2O_3 at 1, 3, and 5% increased the heat resistance of bioplastics with melting points of 270, 274, and 280 °C. Bioplastic obtained the best results except for mechanical properties with a melting point of 280 °C, tensile strength of 3.41 Mpa, elongation of 38.66%, density of 5.52 g cm^{-3} , and 80.28% water resistance for bioplastic with 5% Al_2O_3 that suitable on Indonesian National Standard 7188.7:2016. The FTIR analysis revealed that bioplastic containing Al_2O_3 experienced physical interactions. Morphological analysis revealed that Al_2O_3 was evenly distributed on the bioplastic's surface.

Keywords: Bioplastic, composite, heat resistance, sago hampas, starch

1. Introduction

Bioplastics are made from biomass, which microorganisms can decompose in the environment. Natural polymers such as lignin, protein, natural rubber, starch, cellulose, and chitin can be used to make bioplastics (Kabasci, 2014). Because of its abundance, starch is widely used as a material for the production of bioplastics. Starch can be obtained from tubers, seeds, nuts, and the trunks of sago trees. Several studies have used cassava starch, sago starch, a mixture of durian seed starch and sago starch, jackfruit seed starch, and other starch sources as the main ingredients for bioplastics (Wahyuningtyas and Suryanto, 2017; Amni et al., 2019).

Each type of starch contains varying amounts of amylose and amylopectin, such as corn starch containing 26% amylose and 74% amylopectin, cassava starch containing 20-27% amylose and 77-80% amylopectin and sago starch containing 27% amylose and 73% amylopectin (Eliasson, 2004).

Indonesia has 5.4 million ha of sago (*Metroxylon* sp.) land, with Papua accounting for 95% (Kementan, 2020). In 2007, a modern sago mill was established to aid sago

management. As a result, more waste is produced. Waste from sago processing includes bark and hampas. The extracted sago hampas account for approximately 14% of the total wet weight of sago stalks (Flach, 1997). Sago hampas is currently used in animal feed, biosorbent, compost, and fermentation (Husin et al., 2019). Despite the abundance of sago hampas, it still has a reasonably high starch content of 52.98%. As a result, sago hampas can be recycled to produce starch.

Several studies have addressed these shortcomings by adding additives such as ZnO, which can improve the mechanical and antimicrobial properties of cassava starch bioplastics because starch-based bioplastics have several shortcomings, including low physical properties, brittleness, and low water resistance (Hidayat et al., 2019). TiO_2 is used as reinforcement and heat resistance in corn starch bioplastic (Amin et al., 2019), and rice husk nanosilica as reinforcement and reduces water vapor transmissibility. Aside from these characteristics, the heat resistance of bioplastics derived from natural polymers varies depending on the source of starch and other supporting materials. According to Abdullah et al. (2019), increasing the ratio of cassava starch and glycerol can improve mechanical properties and melting point.

According to Amin et al. (2019), TiO₂ can improve the mechanical properties and heat resistance of bioplastics made from corn starch, glycerol, and acetic acid, resulting in a melting point of 291 °C without TiO₂ and 303 °C with TiO₂.

Furthermore, Chueangchayaphan et al. (2019), discovered a melting point of 280 °C in a thermoplastic cassava starch-TiO₂-Al₂O₃ and glycerol using variations in particle size of Al₂O₃. Al₂O₃ is a chemical that is widely used in industry. Because of its high heat capacity, this compound is used as a high-temperature insulator because it is a good electrical insulator. Al₂O₃ has a high melting point of 2050 °C (Selvam et al., 2021).

Starch, such as cassava starch and cornstarch, is still required as an ingredient in daily consumption so that its use as a bioplastic material is limited. The high starch content of sago hampas can be reused as a raw material for bioplastics to reduce industrial waste. The amount of starch and glycerol used in bioplastics can affect their heat resistance properties (Abdullah et al., 2019). On the other hand, adding additives can increase heat resistance (Amin et al., 2019). The utilization of Al₂O₃ as additives in bioplastic is still less. This study aimed to investigate the effect of Al₂O₃ on increasing the heat resistance of sago starch-based bioplastics.

2. Methodology

2.1. Materials

The materials used in this study were sago hampas from the sago processing industry in Tanah Baru Bogor, glycerol (PT. Brataco, Bogor, Indonesia), acetic acid (p.a 99%), Al₂O₃ powder (Merck, Germany), aquades, and water.

2.2. Starch Separation

The method for separating starch from sago hampas was that sago hampas were mixed with water in a container. The sago hampas were squeezed three times. The juice was left for 24 hours to allow starch deposits to form. Following that, the starch precipitate was sundried. The dried precipitate was ground and sieved through a sieve with 60 mesh.

2.3. Preparation of Bioplastic

A total of 10 g of sago hampas starch was combined with Al₂O₃ at 0, 1, 3, and 5% by weight of sago hampas starch and filled into a

beaker glass, respectively. 100 mL of distilled water, 10 mL of acetic acid, and 3 mL of glycerol mixed into sago hampas starch and Al₂O₃. The mixture was heated to 90 °C using a hot plate magnetic stirrer and stirred at 800 rpm. After the mixture was homogeneous, the sample was poured into a glass mold and dried at room temperature for 24 hours.

2.4. Analysis and Characterization of Bioplastic

The melting point of bioplastics was performed using a differential thermal analysis (DTA Hitachi STA7300). A sample of bioplastic was placed in an alumina pan holder. The sample was then vacuumed to a pressure of 1 bar. The nitrogen gas was then introduced at 100 mL/min rate. Thermal testing of bioplastics was performed at temperatures as high as 1000 °C. The DTA test results were evaluated in the form of an endothermic or exothermic peak.

Bioplastics were tested using a universal testing machine (Zwickroel) for tensile strength and elongation. Each bioplastic sample was cut to a specific length and clamped on both sides. The test of tensile strength and elongation according to ASTM D882.

A density test was performed using a pycnometer to determine the density of bioplastics. A density test was performed using a pycnometer to determine the density of bioplastics by using Equation 1.

$$d_s \text{ (g cm}^{-3}\text{)} = \frac{(m_{ps} - m_p)}{(m_{pl} - m_p) - (m_{pls} - m_{ps})} \times (d_1 - d_a) + d_a \quad (1)$$

Remark:

d_s (g cm⁻³) : density
 m_p (g) : mass of pycnometer
 m_{ps} (g) : mass of pycnometer and sample
 m_{pl} (g) : mass of pycnometer and water
 m_{pls} (g) : mass of pycnometer + water + sample
 d_1 (g/mL) : water density
 d_a (g/mL) : air density

The water resistance test was used to determine the amount of water that was not absorbed by the bioplastic. The test was carried out by cutting each bioplastic sheet and weighing the sample. The sample was then placed in a container containing distilled water at room temperature and allowed to stand for 24 hours. After settling, the sample was cleaned with a dry cloth and weighed. Equations calculated the test results:

$$WA (\%) = \frac{W - W_0}{W_0} \times 100 \quad (2)$$

$$WR (\%) = 100\% - \text{water absorption rate} \quad (3)$$

Remark:

WA (%) : water absorption

W (g) : wet weight

W₀ (g) : conditioned weight

WR (%) : water resistance

Functional groups analysis was carried out using fourier transform infrared spectroscopy (FTIR Perkin Elmer Spectrum 2). Bioplastic was prepared with KBr and then pressed with a hydraulic jack and measured using FTIR. The test results, which included the wavenumber and percent transmittance, were then interpreted to determine the functional group.

Morphology analysis was carried out using scanning electron microscopy (SEM JEOL JSM-6510LA). The bioplastic sample was placed in a set holder with two adhesive strips. Following that, it is vacuum-coated with gold metal and inserted into the sample holder of the SEM. SEM magnified the sample and morphological images were taken.

3. Results and Discussion

3.1. Bioplastic

The gelatinization process occurs during heating treatment, resulting in a thick and chewy solution (Adawiyah et al., 2021). The bioplastics properties obtained have a smooth and transparent texture, as shown in Figure 1. The addition of glycerol results in smooth properties, caused reduces the rough texture of the bioplastics (Lusiana et al., 2019). The transparency is due to colorless materials. Al₂O₃ is a white solid, the more Al₂O₃ added, the whiter the color of the bioplastic and the lower transparency.

3.2. Melting Point

The DTA results shown in Figure 2 revealed that bioplastics with 0, 1, 3, and 5% Al₂O₃ added had melting points of 266, 270, 274, and 280 °C, respectively. The melting point of sago starch bioplastic without Al₂O₃ is higher than that of cassava starch bioplastic at 120 °C by Abdullah et al. (2019). Because sago starch contains less amylopectin than cassava starch, its amylose content is higher. Amylose has a higher melting point than amylopectin (Delcour and Hosney, 2010). As a result, sago starch bioplastics have a higher melting point than cassava starch bioplastics. The addition of Al₂O₃ raises the melting point from

4 to 14 °C. The raises are due to its high density (Alaerts et al., 2018).



Figure 1. Bioplastic with Al₂O₃ addition (a) 0%, (b) 1%, (c) 3%, (d) 5%.

In this study, the highest yield was bioplastic with 5% Al₂O₃ addition with a melting point of 280 °C. The melting point of bioplastics differs only slightly between variations because the amount of Al₂O₃ differs slightly. The resulting melting point represents starch decomposition (Chueangchayaphan et al., 2019). Bioplastics with Al₂O₃ added can be used as a wrap/cover for heat materials.

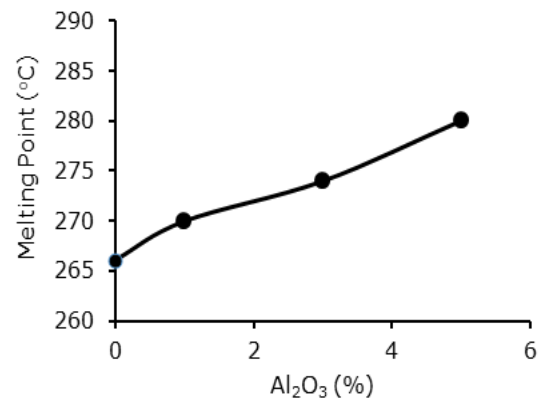
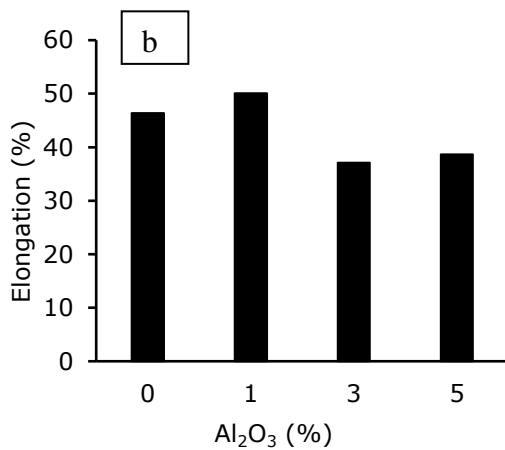
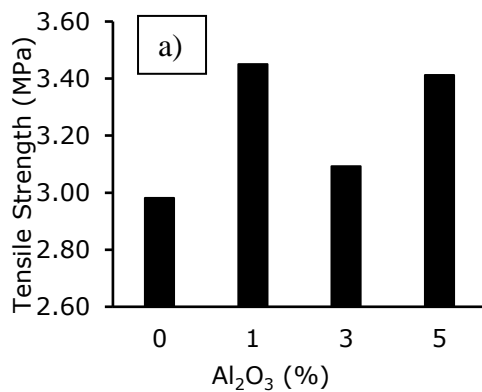


Figure 2. Effect of Al₂O₃ addition to melting point of bioplastic (a) 0%, (b) 1%, (c) 3%, (d) 5%.

3.3. Tensile Strength and Elongation

The tensile strengths of bioplastics with 0, 1, 3, and 5% Al₂O₃ were 2.98, 3.45, 3.09, and 3.41 MPa, respectively (Figure 3a). The low tensile strength for all bioplastics is due to the addition of glycerol. The addition of glycerol reduces in the strength of the intermolecular forces so that the mobility between the molecular chains increases (Hamzah et al., 2021). However, these findings suggest that adding Al₂O₃ to bioplastics can increase their tensile strength. Bioplastics containing 1, 3, and 5% Al₂O₃ produced varying tensile strength values. The elongation values of bioplastics with 0, 1, 3, and 5% Al₂O₃ addition were 46.39, 50.06, 37.10, and 38.66%, respectively (Figure 3b). However, the elongation value of bioplastics decreased with the addition of 3 and 5% Al₂O₃. The uneven thickness of the bioplastic causes fluctuating

tensile strength and elongation results. Bioplastic containing 1% Al_2O_3 increased elongation value compared to bioplastic without Al_2O_3 . The more Al_2O_3 is added, the stronger its physical properties will be. Because Al_2O_3 is a crystalline compound with a stable structure, it forms a strong nature. In this study, the highest yield obtained by bioplastic containing 1% Al_2O_3 can increase tensile strength from 2.98 to 3.45 MPa and elongation from 46.39 to 50.06%. Tensile strength has a standard of 24.7-302 MPa and elongation of 21-220% according to SNI 7188.7:2016 plastics, so the mechanical properties of bioplastics do not meet the standards.



3.4. Density

Figure 4 showed the density of bioplastics with 0, 1, 3, and 5% Al_2O_3 were 2.86, 3.70, 4.89, and 5.52 g cm^{-3} , respectively. Due to the hollow space in the bioplastic inserted by Al_2O_3 , the addition of Al_2O_3 can increase the density. Density value can raise the melting point of bioplastics. However, the density did not differ significantly with the addition of Al_2O_3 . The difference because the amount of Al_2O_3 added varies slightly. In this study,

bioplastic with 5% Al_2O_3 produced the highest yield. Overall, the density of bioplastics without and with Al_2O_3 conforms to SNI 7188.7:2016 with a plastic density of 0.95 g cm^{-3} .

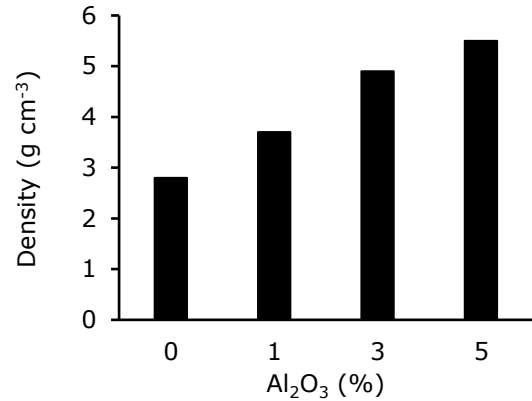


Figure 4. Effect of bioplastic with Al_2O_3 addition to density.

3.5. Water Resistance

Figure 5 showed the water resistance values for bioplastics with 0, 1, 3, and 5% Al_2O_3 were 67.32, 77.86, 78.49, and 80.28%, respectively. The addition of Al_2O_3 increases the value of water resistance. Due to hydrophilic, starch is one of the factors that can reduce water resistance (Seligra et al., 2015). Starch binding to water can be reduced by the higher embedding of Al_2O_3 in bioplastics. High density also contributes to increase water resistance. Bioplastics with Al_2O_3 additions of 1, 3, and 5% only differ slightly in water resistance because the Al_2O_3 addition is slightly different. As a result, the water resistance was not significantly different with the addition of Al_2O_3 .

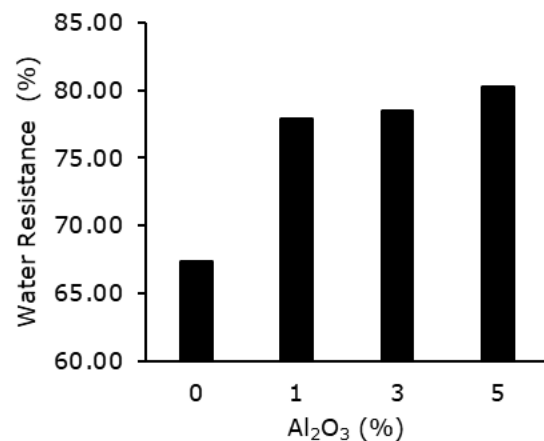


Figure 5. Effect of bioplastic with Al_2O_3 addition to water resistance.

In this study, the highest yield is obtained by bioplastic containing 5% Al₂O₃, which has a water resistance value of 80.28%. According to Indonesian National Standard 7188.7:2016, plastic's maximum water absorption capacity is 21.5% or water resistance of at least 78.5%, indicating that bioplastics with Al₂O₃ 3 and 5% have met Indonesian National Standard. As a result, bioplastics with the addition of Al₂O₃ can be used as water-resistant wrapping/covering materials.

3.6. Functional Grup

Figure 6 shows the FTIR spectrum of bioplastics with 0 and 5% Al₂O₃ addition. Table 1 showed that bioplastics containing 0% Al₂O₃ have an absorption band with a wavenumber of 3272 cm⁻¹ and a broad peak corresponding to the -OH group. The absorption of wavenumber 2927 cm⁻¹ represents the C-H group. The C-O-C (glycoside) group of starch

exhibits strong absorption at wavenumber 1003 cm⁻¹. The FTIR spectra of bioplastics containing 5% Al₂O₃ revealed the presence of a -OH group at 3273 cm⁻¹ absorption with a broad peak and lower transmittance due to the strong hydrogen bonding effect (Coates, 2006).

The absorption wavenumber of 2925 cm⁻¹ indicates the presence of a C-H group with a slight shift. The C-O-C (glycoside) group exhibits a sharp absorption at a wavenumber of 1003 cm⁻¹, with no shift in wavenumber, indicating that Al₂O₃ does not affect the breaking of glycosidic bonds in starch. In bioplastics containing 5% Al₂O₃, there is an absorption group with a wavenumber of 665 cm⁻¹ and a very small peak identified as the Al-O group of Al₂O₃ (Tong et al., 2015). The fact that the wavenumber shift does not change significantly indicates that the interaction is physical.

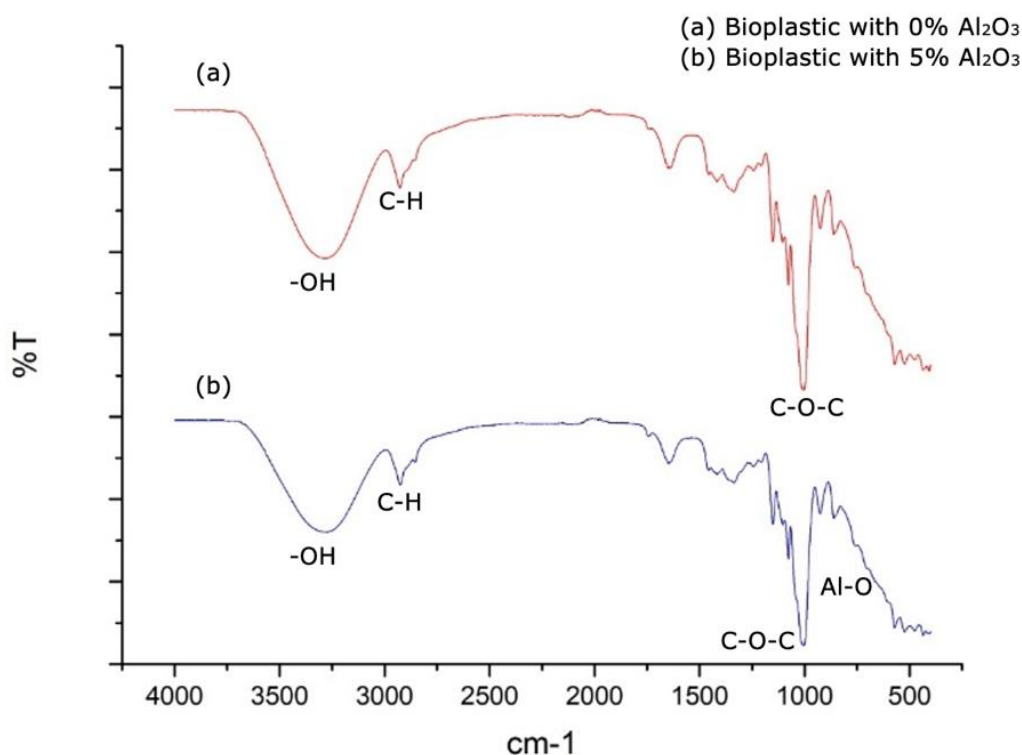


Figure 6. FTIR spectrum of bioplastic with 0 and 5% Al₂O₃ addition.

Table 1. Functional group of bioplastic with 0 and 5% Al₂O₃ addition.

Functional group	Wavenumber (cm ⁻¹)	
	Bioplastic with 0% Al ₂ O ₃ (cm ⁻¹)	Bioplastic with 5% Al ₂ O ₃ (cm ⁻¹)
-OH	3272	3273
C-H	2927	2925
C-O-C	1003	1003
Al-O	-	665

3.7. Morphology

SEM analysis of bioplastic without Al₂O₃ revealed a smooth surface, indicating a homogeneous mixture of bioplastics (Figure 7a). There are small grains with an uneven surface in bioplastic with 5% Al₂O₃ addition (Figure 7b). The granules are thought to be Al₂O₃ incorporated into the bioplastic. On the surface of the bioplastic, Al₂O₃ is evenly distributed. According to Chueangchayaphan et al. (2019), Al₂O₃ is homogeneously dispersed evenly in the composite.

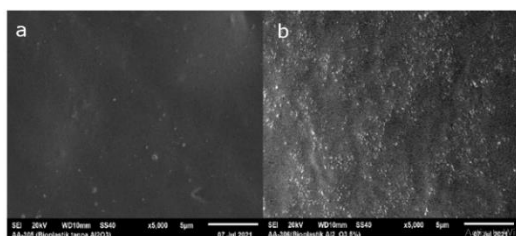


Figure 7. SEM (5000x) of bioplastic with Al₂O₃ addition (a) 0% and (b) 5%.

4. Conclusion

Adding Al₂O₃ to bioplastics can improve heat resistance and increase melting point value than bioplastics that do not contain Al₂O₃. Due to the uneven thickness, the mechanical properties of bioplastics with Al₂O₃ addition of 0, 1, 3, and 5% do not meet the standards. The addition of Al₂O₃ to bioplastics can increase density and water resistance. The density of bioplastics with Al₂O₃ addition of 0, 1, 3, and 5% have met the standard, according to Indonesian National Standard 7188.7:2016. However, the water resistance of bioplastics containing 3 and 5% Al₂O₃ only meets the standard. The interaction in bioplastics with the addition of Al₂O₃ is a physical interaction and Al₂O₃ can be evenly distributed on the bioplastic's surface.

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