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Effects of Chitosan on the Characteristics of Sorbitol Plasticised Cellulose Acetate/Starch Bioplastics

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Due to the natural origins of its primary components, such as plant fibers, bioplastics are materials that disintegrate quickly. Typically, plasticizers are used to create cellulose-based bioplastics. Plasticizers soften hard, inflexible structures, improve elasticity and flexibility, and lessen bioplastics' tendency to break. The manufacturing of bioplastics in this study includes the incorporation of chitosan and sorbitol plasticizers as a variable. Chitosan and sorbitol were utilized in the following mass ratios: 0.5 g, 1.0 g, and 20 %, 30 %, and 40 %. The optimal chitosan and sorbitol concentrations produced the most desirable bioplastic properties. At a ratio of 20 % sorbitol and 0.5 g chitosan, the greatest bioplastic value for tensile strength (845.68 kPa) and Young's modulus (67.65 x 102 KPa) was attained. With values of 44.94 % and 81.28 %, respectively, the ratio of 30 % sorbitol to 1 g chitosan produced the best mass and water absorption values. At a ratio of 30 % sorbitol and 0.5 g chitosan at 0.94 g/mL and a ratio of 40 % sorbitol and 1 g chitosan at 1.75 %, the highest density and elongation values were identified.

1. Introduction

Plastic waste is a problem on a worldwide scale. According to the Global Plastics Outlook 2022, a mere nine percent of plastic trash is effectively recycled, 19 % is burnt, and over 50 % is dumped in landfills. The other 22 % is discarded or burned outside. Due to its capacity to organically degrade by microorganisms and leave no compounds hazardous to living beings, bioplastic is environment-friendly (Melani et al., 2017). Kapok plants, which may be converted into raw materials for cellulose-based biopolymers to manufacture bioplastics, are abundant in Indonesia. The kapok plant (*Ceiba pentandra*) is an ideal plant for usage as a biopolymer plastic since its fiber contains a high cellulose content, which can vary from 35 to 64 % (Chaiarrekij et al., 2011). Cellulose-based bioplastics have drawbacks such as low water resistance, brittleness, low stability (which makes them susceptible to deterioration), and poor mechanical qualities (Abe et al., 2021). Due to the hydrophilic nature of cellulose, modifications to the manufacturing of bioplastics with the inclusion of additional natural components are required to overcome fragility and increase mechanical qualities. Chitosan is an antibacterial copolymer that appears in the form of thin sheets that are white or yellow in color and odorless. The use of chitosan as an additive in the manufacturing process of bioplastics helps to reduce the rate of water absorption, improve mechanical characteristics, and reduce the moisture content of bioplastic films (Saputro and Ovita 2017). Sorbitol plasticizer increases elasticity by decreasing the degree of hydrogen bonding and increasing the spacing between polymer molecules. Sorbitol as a plasticizer may be more successful in producing bioplastic films with reduced oxygen permeability (Asngad et al., 2020). Author et al. (2022) conducted previous research on bioplastics derived from kapok fiber. regarding the influence of the kind and concentration of a plasticizer on the properties of cellulose acetate-based bioplastics from kapok fiber and the effect of polypropylene filler with glycerol and sorbitol plasticizers on bioplastics from kapok fiber. They discovered that adding the plasticizer sorbitol increased the density of bioplastics, percent elongation, Young's

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modulus, and degradation. Sofia et al. (2017) investigated the use of chitosan in the production of bioplastics, comparing pumpkin skin-chitosan bioplastics with plasticizers derived from various glycerol sources. Rahadi et al. (2020) did another study on the properties of bioplastics generated from liquid waste from tofu (whey) with the inclusion of chitosan and glycerol. Owing to the results obtained in this study, chitosan improves the mechanical characteristics of bioplastics by forming hydrogen bonds between chains, causing bioplastics to become tight and stiff. There has never been any research on the production of cellulose-based bioplastics using chitosan and sorbitol. The purpose of this study is to see how differences in the addition of chitosan and sorbitol plasticizers affect the properties of cellulose-based bioplastics made from kapok fiber.

2. Methodology

The manufacturing of bioplastics in this present research employed materials for instance, kapok plants (Indralaya City), starch (PT Budi Strach and Sweetener Tbk), aquadest (SMARTLAB, A-1078), alkaline water pH 8 (PT Enagic. Co. Ltd.), sorbitol (Technical), sodium hydroxide (NaOH) (Merck), glacial acetic acid (CH_3COOH (by Merck), acetic anhydride ($(CH_3CO)_2O$) (Merck), sulfuric acid (H_2SO_4) (Merck), chitosan (Technical). Additionally, the procedure of synthesis and purification of cellulose acetate in kapok fiber was carried out using the research technique developed by Author et al. (2022). with adjustments to the manufacturing of bioplastics. 1.5 g of starch was dissolved in 9 mL of distilled water before being cooked at 70 °C for 15 min until gelatin was produced. The starch solution was then treated with 1 g of cellulose acetate. Chitosan (0.5 g and 1.0 g) and sorbitol (20 %, 30 %, and 40 %) concentrations were added based on the variations of each sample. The solution was heated at 50 °C for a period of fifteen minutes while stirring for thickening purposes. The bioplastic solution was then printed on a petri dish that had been previously coated with aluminum foil and allowed to cure at room temperature to generate bioplastic sheets.

2.1 Density Analysis

The density value of bioplastics was determined using the approach described by Darni et al. (2014), in which the mass (g) of the sample to be evaluated was weighed with a digital balance. A 10 mL measuring cup was filled with 5 mL of water, and the bioplastic composite sample was placed in the measuring cup. The final volume of the biocomposite plastic can be determined by dividing the final volume by the beginning volume of sample water. The density of a sample may be estimated using Eq(1), where was the density (g/mL), m was the mass (g), and v was the volume (mL).

$$\rho = \frac{m}{v} \tag{1}$$

2.2 Scanning Electron Microscopy (SEM) Test

The surface morphology of bioplastics was observed using a scanning electron microscope (SEM). The Tescan Vega 3 SEM instrument with a firing energy of 25 kV and a firing range of 14-15 mm was used to examine the surface of bioplastics at 1000 x magnification.

2.3 Tensile Strength and elongation test

The CMT-10 Computer Control Electronic Universal Testing Machine was used to do tensile testing. Tensile strength and elongation were calculated using stress-strain curves extrapolated from force-distance data acquired at room temperature and a strain rate of 2 mm/min. All mechanical testing of bioplastics has been performed according to the ASTM-D638-14 standard procedure. The narrow section sample width was 13 mm, the narrow section length was 57 mm, the overall length was 165 mm, the thickness was 3.5 mm, and the gage length was 50 mm. In each sample, a total of five specimens were tested.

2.4 Biodegradation test

The ability of degradation of bioplastics was investigated and shown using the approach developed by Martucci and Ruseckaite (2015). The decomposition time with loose soil with breakdown qualities and an acidity degree of roughly pH 6 - 7 must be checked regularly within 4 d, with three replications for each sample. The test begins with weighing the bioplastic sample before to performing the deterioration test and again after 4 d of degradation. The soil samples were removed, carefully brushed, sprayed multiple times with distilled water, and dried at 50 °C to constant weight. Eq(2) may be utilized for calculating the percent breakdown of bioplastic samples, where A1 was the original mass (g) and A2 was the final mass after drying (g).

$$\text{Biodegradation (\%)} = \frac{A1 - A2}{A1} \times 100\ \% \tag{2}$$

2.5 Water Absorption Test

The water absorption test was performed according to Martucci and Ruseckaite (2015) by cutting a bioplastic sample with a diameter of 50 mm and then weighing the bioplastic. Eq(3) can be used to calculate the quantity of water absorption, where W0 is the initial sample weight (g) and W is the end sample weight (g).

$$\text{Water (\%)} = \frac{W - W0}{W0} \times 100\ \% \tag{3}$$

3. Results and Discussion

3.1 Bioplastic Characterization

Scanning Electron Microscopy (SEM) of bioplastics seeks to evaluate the surface and morphology of bioplastics. Figure 1 depicts the surface and shape of bioplastics as determined by SEM. The findings of the SEM test on the surface of bioplastics with 1.5 g of starch, 1 g of kapok fiber-based cellulose acetate, and 0.5 g of chitosan are shown in Figure 1. The concentration of plasticizer in the form of sorbitol in the three samples differs by 20 %, 30 %, and 40 %, respectively. Each sample generated has an uneven surface due to the film morphology of bioplastics. This is because the components in the bioplastic solution are not homogeneous (Darni et al., 2014). The less homogenous mixing is brought about by the stirrer not rotating entirely throughout the stirring phase; this is further affected by the mixing of chitosan powder and cellulose acetate, which occurs after the starch has achieved the gelatinization temperature. The surface of the samples containing 20 % and 30 % sorbitol was still rough, however, the 40 % sample had a smoother surface. The amount of sorbitol used changes the surface structure of the bioplastic, making it smoother. Figure 1 shows spherical granules of starch. The starch employed is derived from cassava, and the size of cassava starch granules varies from 4 to 35 μm. Bioplastics with a smoother and denser surface will find it more difficult to absorb water, resulting in greater water resistance. This phenomenon occurs because the denser surface of the bioplastic makes it difficult for water to be absorbed and fill the voids in the bioplastic (Setiawan et al., 2021). SEM mechanical characteristics studies revealed that bioplastics with 40 % sorbitol content had the best surface. Since chitosan is distributed homogeneously in bioplastics and fills empty areas, bioplastics with 40 % sorbitol plasticizer have a finer structure.

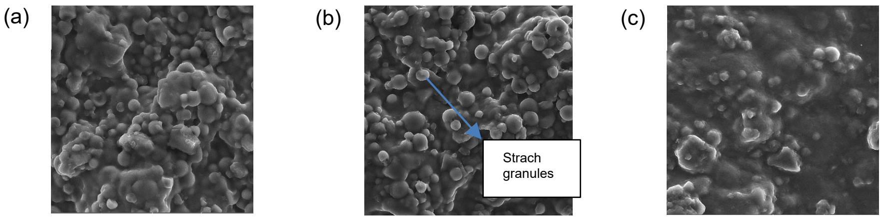

Figure 1: SEM-EDX analysis results at 1000 X chitosan 0.5 magnification with variations: (a) sorbitol 20 %, (b) sorbitol 30 %, (c) sorbitol 40 %.

3.2 Bioplastic Density Test and Water Resistance

High-density bioplastics will have an impact on the increase in mechanical characteristics. Because the higher the ability to absorb water, the more readily damaged bioplastics will be. Figure 2 depicts the results of a density and water absorption measurement of bioplastics.

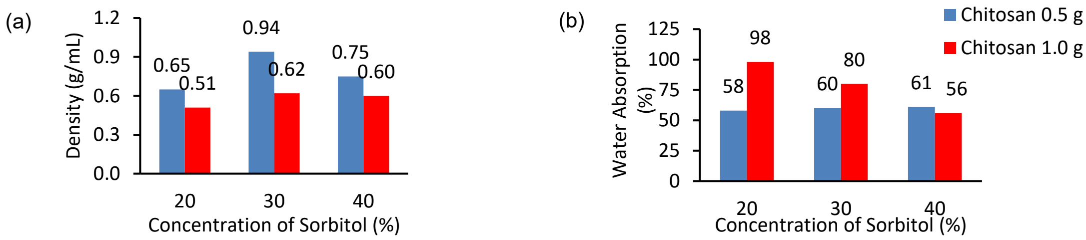

Figure 2: (a) The Effect of Sorbitol Concentration and Chitosan Loading on Density of Bioplastics, (b) The Effect of Sorbitol Concentration and Chitosan Loading on Water Absorption of Bioplastics

Figure 2(a) depicts the varying density of cellulose acetate-based bioplastics derived from kapok fiber. The inhomogeneous mixing process between cellulose acetate and starch causes the fluctuating density of bioplastic films (Afidin, 2021). The density values of the bioplastic product ranged from 0.51 to 0.94 g/mL, with the lowest density obtained from samples containing 1 g of chitosan and 20 % sorbitol. The greatest density bioplastics were produced from samples containing 0.5 g chitosan and 30 % sorbitol. The denser the molecular structure, the greater the density value of a substance. Water molecules will find it challenging to permeate bioplastic films with tight structures (Darni et al., 2014). Low-density bioplastics have an open structure, allowing fluids such as water, oxygen, and carbon dioxide to easily permeate. The higher the chitosan content, the larger the thickness, density, and tensile strength, and the lower the value of water absorption and elongation of bioplastics. According to Figure 2(a), the density test results revealed that when the chitosan concentration grew, the density values decreased. Increasing the concentration of chitosan can enhance thickness while decreasing density. The additional plasticizers will diminish melt viscosity, making starch more difficult to plasticize since its mobility slows during the process. The uniformity of the bioplastic structure is a sign of an improvement in the value of bioplastic mechanical strength (Brahmana et al., 2021). Figure 2(a) shows that at 40% sorbitol concentration, the density value of bioplastics begins to decrease. The extra chitosan filler makes homogenizing the bioplastic structure problematic. Figure 2(b) depicts the impact of sorbitol and glycerol addition on water absorption. Chitosan is more water resistant since it is a water-insoluble chemical, reducing the hydrophilic character of starch bioplastics. Furthermore, the inclusion of sorbitol plasticizer helps strengthen the film matrix, making it more resistant to water damage. According to (Setiawan et al., 2021), adding a greater plasticizer (sorbitol) increases the adhesive qualities between molecules, lowering the rate of both air absorption and moisture in bioplastic films.

3.3 Tensile Strength, Elongasi, and Young's Modulus of Bioplastic

Tensile strength testing is used to measure a bioplastic's capacity to bear a specific force before it breaks. To assess the percentage change in length of the bioplastic before it breaks, the percent elongation test of cellulose acetate-based bioplastics from kapok fiber was performed. The Young's modulus test on cellulose acetate-based bioplastics derived from kapok fiber is designed to measure the elasticity of the produced bioplastics. Young's modulus is calculated by comparing tensile strength and % elongation. Figure 3 depicts the tensile strength, elongation, and Young's modulus values of bioplastics.

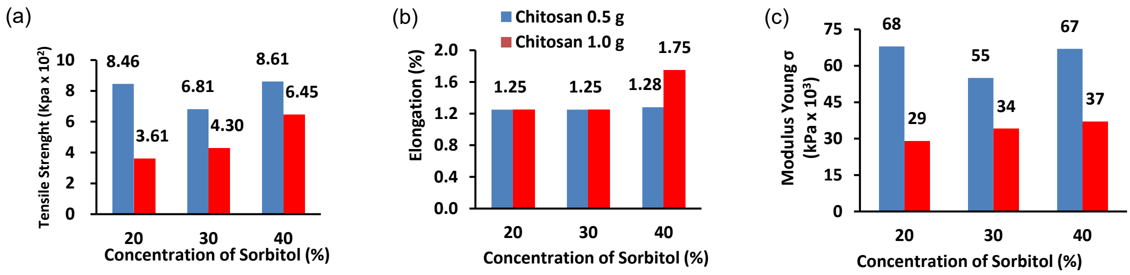

Figure 3: The Effect of Sorbitol Concentration and Chitosan Loading on Mechanical Properties of Bioplastics, (a) Tensile Strength (b) Elongation at break (c) Young's Modulus.

Figure 3(a) demonstrates that the tensile strength of bioplastics with varying sorbitol concentrations varies from 360.66 to 845.68 kPa. Samples containing 0.5 g chitosan and 40 % sorbitol had the greatest tensile strength value, able to bear a force of 860.68 kPa. Samples containing 1 g of chitosan and 20 % sorbitol had the lowest tensile strength value and possessed the ability to withstand 360.66 kPa. According to (Afidin, 2021), the tensile strength of bioplastics is still fluctuating since the combining of starch and cellulose acetate is not homogenous. For each chitosan addition, the greatest tensile strength value was found at a sorbitol concentration of 40 %. The increase in the strength of biodegradable plastics is due to the addition of sorbitol concentration, which causes the plastic molecular weight to increase and the resulting structure of the biodegradable plastic to become tighter, requiring a large enough force to break the polymer chain bonds. The tensile strength of biodegradable polymers has improved in tandem with their molecular weight. Chitosan can improve tensile strength qualities by increasing the amount of hydrogen bonds in bioplastics, thus it is more difficult for chemical bonds to break (Kalsum et al., 2020). Similarly found in the result presented in Figure 3(a), the addition of chitosan mass tends to enhance the tensile strength value in each sample. Figure 3(b) demonstrates that the elongation values are mainly identical and an increase with the addition of 40 % sorbitol in the two bioplastic samples has been noticed. The sample with 1 g chitosan and 40 % sorbitol concentration showed a substantial variation in value, with the greatest percent elongation value, 1.75 %. The elongation value of bioplastics experienced an increase accompanied by a rise in plasticizer content. Sorbitol can promote elongation because it can diminish intermolecular connections between amylose and amylopectin and starch or cellulose, which affects the hydrogen bonding of starch and sorbitol molecules (Sanyang et al., 2015). The elongation value has an impact on the quality of the bioplastic; a greater elongation value implies that the bioplastic has superior mechanical qualities. A previous study by Hartatik and Nuriyah (2014) found that increasing the chitosan content reduced bioplastic elongation. Nevertheless, tests on samples of cellulose acetate-based bioplastics derived from kapok fiber revealed that increasing the concentration of chitosan tends to improve the percent elongation value. The Young's modulus test results for each bioplastic sample are shown in Figure 3(c). The greatest Young's modulus value was found in bioplastic samples with a mass of 0.5 g chitosan and a sorbitol content of 20 %, with Young's modulus of 67655.17 kPa. The lowest Young's modulus 24452.94 kPa was reported for bioplastic samples containing 1 g of chitosan and 20 % sorbitol content. Bioplastics have high Young's modulus values and high tensile strength values. This is due to the fact that Young's modulus is directly related to tensile strength while being inversely proportional to elongation. Increased sorbitol concentration can enhance the elasticity of bioplastics, resulting in a decline in the value of Young's modulus (Author et al., 2022). According to (Firmansyah et al., 2021), chitosan affects the value of Young's modulus of bioplastics. The existence of more chitosan raises the value of young's modulus of bioplastics. However, it differs from the test findings, which tend to diminish with increasing chitosan and sorbitol concentrations.

3.4 Biodegradable Bioplastic Test

The capacity of biodegradable bioplastics based on cellulose acetate from kapok fiber to disintegrate entirely when discarded into nature was investigated. The test for biodegradation of bioplastics may be performed in two ways: covered with soil and not covered with soil. The bioplastic mass degradation estimates were carried out by accumulating soil and were tested every day until the fourth day. The soil utilized for this test is humus soil, which may be found in a variety of locations and is often used for plants. Humus soil was used as the testing medium since microorganisms in humus soil may breakdown organic content in bioplastics. Figure 4 depicts the effect of chitosan and sorbitol additions on the biodegradability of bioplastics with and without landfilling.

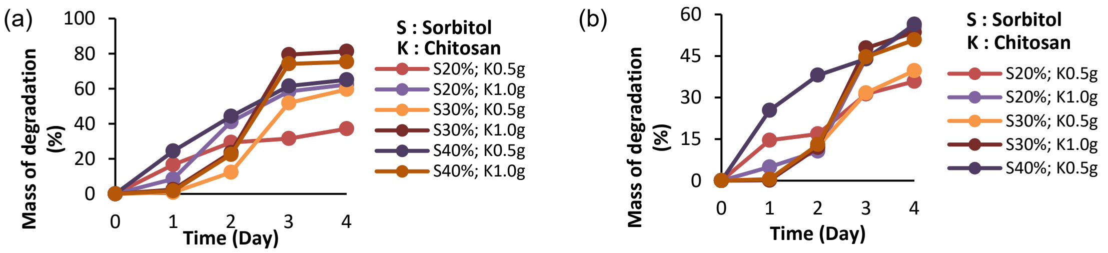

Figure 4: (a) Mass Percent Yield of Degraded Bioplastics with Land Stockpiling; (b) Mass Percent Yield of Degraded Bioplastics Without Landfill

Figure 4 depicts the results of a 4-day biodegradation test with and without stockpiling soil. According to graph 4(a), the maximum degradation mass percent value was observed in samples containing 30 % sorbitol and 1 g of chitosan, with a degradation of about 81.28 %. Samples with a sorbitol concentration of 20 % and 0.5 g chitosan concentration of 37.17 % had the lowest deteriorated mass percent result. In Figure 4(b), the sample with a sorbitol concentration of 40 % and 0.5 g chitosan concentration of 56.41 % had the highest degraded mass percent without being stored with soil. Samples with a sorbitol concentration of 20 % and 0.5 g chitosan concentration of 35.84 % had the lowest deteriorated mass percentage result. The percentage yield of decomposed mass remained varied with and without stockpiling, possibly be attributed to the homogeneity factor when combining sorbitol and chitosan with cellulose acetate and starch. Non-homogeneous mixing processes might lead bioplastics to be easily destroyed by soil and vice versa. Due to the inhomogeneity of the mixing process, holes and gaps emerge disregarded bioplastics, allowing environmental variables such as water absorption in the bioplastics to occur more readily and expedite the degradation process (Author et al., 2022). The combination of starch and chitosan for bioplastics produces good results because microorganisms grow more easily because of starch's capacity to bind water and make the surface of bioplastics wet (Sofia et al., 2017). Due to its greater molecular weight and stronger intermolecular interactions, sorbitol in bioplastics can slow down the degradation time (Melani et al., 2017).

4. Conclusion

According to the results of the characteristic tests of cellulose acetate-based bioplastics from kapok fiber in this study, the higher the concentration of sorbitol may alter the properties of bioplastics by increasing elongation.

The addition of 40 % sorbitol resulted in the greatest increase in elongation (40 %). The quantity of chitosan in bioplastics can impact their properties by increasing the percentage of water absorption capacity. The addition of 1 g of chitosan resulted in the greatest improvement in the percentage of water absorption capacity, around 69 %, in this investigation. On the other hand, the higher the concentration of chitosan, the lower the tensile strength, Young's modulus, and density of bioplastics. Furthermore, the addition of 1 g of chitosan resulted in the greatest loss in tensile strength (57.39 %), the greatest fall in Young's modulus (57.35 %), and the biggest decline in density value (34.04 %). Different concentrations of chitosan and sorbitol produced the optimum bioplastic characteristics. At a ratio of 20% sorbitol and 0.5 g chitosan, the maximum bioplastic yields for tensile strength values of 845.68 kPa and Young's modulus of 67.65 x 102 KPa were obtained. The best water absorption and mass degradation values were obtained with a ratio of 30 % sorbitol and 1 g chitosan, with values of 44.94 % and 81.28 %, respectively. The highest density and elongation values occurred at a 30 % sorbitol and 0.5 g chitosan ratio of 0.94 g/mL and a 40 % sorbitol and 1 g chitosan ratio of 1.75 %.

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