

The Effect of Cellulose Succinate Concentration on the Mechanical Properties of Bioplastics

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Abstract: The principle of bioplastic film formation is based on hydrogen bonds between the constituent molecules of bioplastics. Adding carboxylic groups to cellulose molecules into cellulose succinate (CS) can increase bond regularity, impacting bioplastic's mechanical properties. This study aimed to determine the effect of CS concentration on tensile strength and elongation of bioplastics. The stages of this research are the synthesis of bioplastics by mixing method and variation of CS concentration, characterization of bioplastics to determine the effect of CS concentration on the mechanical properties of bioplastics, analysis of functional groups with FTIR, and bond regularity with XRD. The results obtained in this study show that CS concentration affects the characteristics of bioplastics. The best bioplastic was found at 3.2% CS concentration with a tensile strength value of 10.48 MPa and elongation of 4.38%. The peaks in FTIR analysis are typical groups of cellulose, namely O-H, C-H, and C-O. CS bioplastics also consist of amorphous and crystalline phases.

Keywords: Bioplastic, Cellulose, Cellulose succinate

INTRODUCTION

Bioplastics are plastics derived from biomass materials (Badan Standardisasi Nasional, 2016). The raw materials of bioplastics can be starch (Santana et al., 2018), protein (Araújo et al., 2018), and cellulose (Sharif et al., 2018). Cellulose-based bioplastics still have weaknesses, especially in terms of mechanical properties. Jannah et al. (2019) report that bioplastics with cellulose as raw material still have a low tensile strength, namely 5 MPa and elongation of 4.75%.

Factors that influence the mechanical properties of bioplastics are fillers and plasticizers. Apart from that, the primary polymer also determines the success of bioplastic synthesis. According to Darni et al. (2018) and Ahmad et al. (2022), cellulose modification can improve the characteristics of bioplastics. Modifications can be performed mechanically (Etikaningrum et al., 2016) and chemically (Leszczyńska et al., 2019). Chemically, cellulose can be modified through etherification and esterification reactions.

One of the cellulose ester derivatives is cellulose succinate (CS). The carboxylic group contained in the cellulose succinate molecule can form a cross-linking reaction with the monomers that make up other bioplastics. This is very beneficial for improving the mechanical properties of bioplastics.

Based on this, the author wants to know how much influence the chemically modified cellulose has on the mechanical properties of bioplastics. The effect was observed through

variations in cellulose succinate concentration in bioplastic synthesis. The bioplastic synthesis was accompanied by adding chitosan as filler and sorbitol as a plasticizer.

RESEARCH METHODS

Materials and Tools

The instruments used in this research were a Hydraulic Universal Testing Machine (UTM), Shimadzu Fourier Transform Infra-Red (FT-IR) Spectrophotometer, PanAnalytical X-Ray Diffraction (XRD), GenLab oven, hotplate, magnetic stirrer, analytical balance, and glassware.

The materials used in this research were distilled water (H₂O), acetic acid (CH₃COOH) 1%, chitosan (C₆H₁₁NO₄)_n, cellulose succinate (C₆H₁₁O₅COOH)_n, and technical sorbitol (C₆H₁₁O₆).

Procedures

Synthesis of Cellulose Succinate Bioplastics (CSB)

A total of 1.2 grams of chitosan was dissolved in 50 mL of 1% CH₃COOH (Hayatun et al., 2020). The chitosan solution was added to cellulose succinate with varying concentrations of 0.8% (CSB1); 1.6% (CSB2); 2.4% (CSB3); 3.2% (CSB4); and 4.0% (CSB5). The mixture was homogenized with a magnetic stirrer for 1.5 hours. After that, the solution was added with 1 mL of sorbitol. The solution was homogenized until it formed a gel. The gel is poured into the mold and allowed to rest. After that, the mold containing the bioplastic gel is dried in an oven at 60°C (Jannah et al., 2019).

RESULTS AND DISCUSSION

Mechanical Properties of CSB

The result bioplastic was brownish-yellow (Figure 1) and had a slightly rough surface on one side. The yellow color of bioplastic film was formed because bioplastic production took place under acidic conditions (Xavier Neves et al., 2019), while the brown color came from the primary color of cellulose. The rough surface of bioplastics is caused by cellulose succinate, which has not completely dissolved in the solution. This also affected the tensile strength of bioplastics.

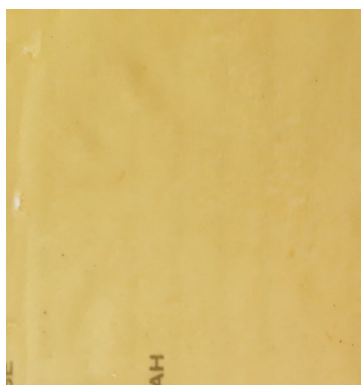


Figure 1. Cellulose Succinate Bioplastic Film

The results of the characterization of the mechanical properties of bioplastics in Figure 2 showed an increase in the tensile strength value at CSB3. However, it decreased again in the following variation up to CSB5. Meanwhile, elongation increased as the cellulose succinate concentration increased. This followed Gabriel et al.'s (2021) statement that tensile strength was opposite to elongation. The decrease in the tensile strength value

of bioplastics at higher concentrations of cellulose succinate is caused by an imbalance in the polymer composition. In this case, the concentration of chitosan functions as a strengthening agent and sorbitol as a constant plasticizer. This condition causes the cohesive strength of the polymer chain to decrease (Abdullah et al., 2019). This has an impact on the strength and elasticity of bioplastics.

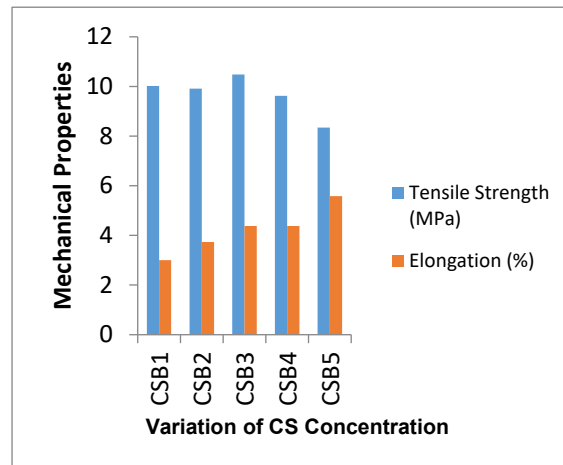


Figure 2. The Effect of CS Concentration on the Tensile Strength and Elongation of Bioplastics

Based on CSB tensile strength and elongation data, the optimum conditions were taken at CSB3 with tensile strength values of 10.48 MPa and elongation of 4.38%. Determining optimum conditions was based on the theory, which states that the mechanical properties of suitable bioplastics are solid and flexible, so an increase in tensile strength must also be accompanied by an increase in elongation (Yuliatun & Marfitania, 2022).

The bioplastic sheets obtained in this research were still classified as fragile bioplastics. However, cellulose-modified bioplastics had better mechanical properties than unmodified cellulose bioplastics. Jannah et al. (2019) reported that cellulose bioplastic's tensile strength and elongation were 5 MPa and 4.75%. Compared with several bioplastic standards in Table 1, CSB3 meets SNI in tensile strength but does not meet elongation standards.

Table 1. Comparison of CSB Tensile Strength and Elongation with Several Standards

Parameters	PLA Japan	PCL England	SNI	CSB
Tensile Strength (MPa)	2050	190	2,47-302	10.48
Elongation (%)	9	500	21-220	4.38

FTIR

The principle of bioplastic formation was based on physical mixtures and chemical interactions (Yang et al., 2010). The formation of bioplastics was based on hydrogen bonds between the molecules that make up the bioplastic film (Bagheri et al., 2019). These bonds can occur inter and intra-molecularly. When two or more substances are mixed, it will reflect changes in the FTIR spectrum peak. These changes can be seen in the cellulose succinate (CS) spectrum and CSB, and based on the results of characterization with FTIR, the peaks that appear in Figure 3a have similarities with those in Figure 3b. The O-H group had shifted from 3421.72 cm⁻¹ to 3415.93 cm⁻¹. The same thing was shown in the C-H vibration peak at 2924.09 cm⁻¹. Then, the C-O group derived from the glycosidic bond of

cellulose molecules was still shown in the vibration area of 1078.21 cm^{-1} . The shift in wave number was likely due to the presence of hydrogen bonds between the constituent molecules of bioplastics (Kane et al., 2016).

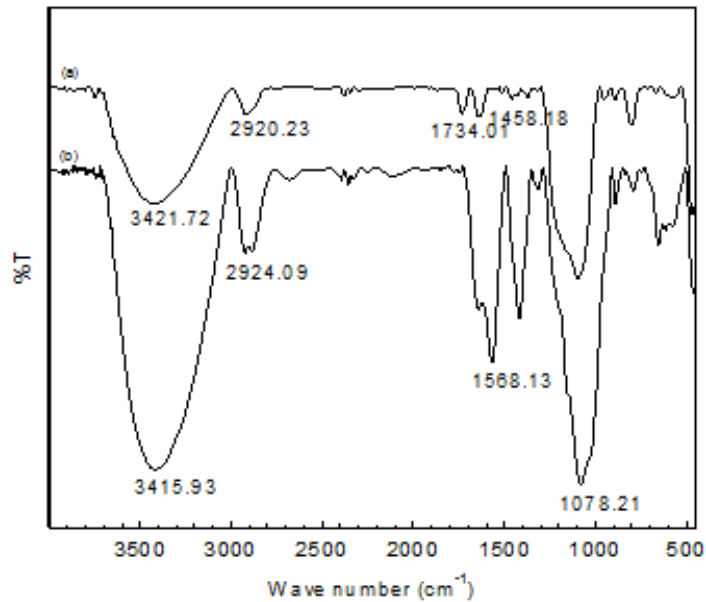


Figure 3. FTIR Spectra of (a) CS (b) CSB

The difference between the spectrum of raw materials and bioplastics was the presence of new groups derived from chitosan molecules. In the $3500\text{--}3100\text{ cm}^{-1}$ region, a peak should appear from the N-H stretching group derived from the primary amine group of chitosan. However, in this study, the peak was likely to overlap with O-H due to hydrogen bonding. This was reinforced by N-H bending and C-N peaks at 1568.13 and 1315.45 cm^{-1} . The overlap between O-H and N-H stretching peaks was also reported by Kane et al. (2016).

XRD

In Figure 4, it can be observed that several sharp peaks and intensities appear at 44.0675° ; 37.8277° ; and 64.4372° . The highest peak was at 2θ 44.0675° . Sharp distinctive peaks were ascribed to cellulose with hydrogen bond interactions with other constituent polymers (Alashwal et al., 2020). Based on the diffractogram, it can be observed that the cellulose succinate bioplastic formed consists of an amorphous phase and a crystalline phase. This condition had similarities with the research reported by (Warsiki et al., 2020). The amorphous phase dominates in favor of the bioplastic biodegradation process.

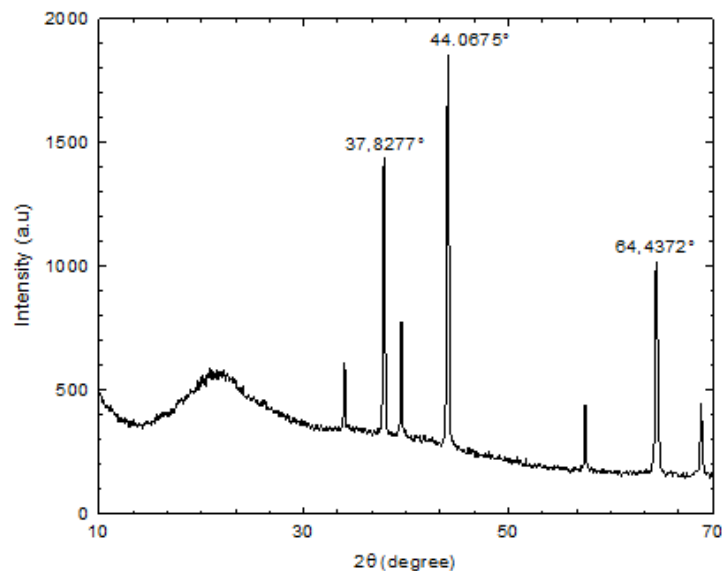


Figure 4. XRD Diffractogram Spectra of CSB

The distance between crystal lattices (d) can indicate the regularity of the matrix structure (Ramakrishnan et al., 2018). The d values obtained were 0.2053 nm, 0.2376 nm, and 0.1444 nm. These values indicate that CSB had a regular structure. If it was related to the mechanical properties, namely tensile strength and elongation, it was in line because tensile strength was also related to the regularity between the constituent molecules of bioplastics (Syafri et al., 2018).

CONCLUSIONS

This research concludes that cellulose succinate can enhance the tensile strength and elongation properties of bioplastics. The optimal concentration of cellulose succinate in the bioplastics examined was determined to be 3.2%, achieving a tensile strength of 10.48 MPa and an elongation of 4.38%. Characterization using Fourier-transform infrared spectroscopy (FTIR) revealed that the C-O group is indicative of the glycosidic bonds present in cellulose molecules. The X-ray diffraction (XRD) patterns indicated that the structure of cellulose succinate bioplastics is both amorphous and crystalline. Furthermore, the measured crystal lattice distance (d -spacing) underscores the structural regularity, which significantly influences the mechanical properties of the bioplastics.

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