OPTIMIZATION OF BIODEGRADABLE PLASTIC BASED ON WATER HYACINTH (EICHHORNIA CRASSIPES) WASTE CELLULOSE WITH THE ADDITION OF SORBITOL PLASTICIZER, CHITOSAN FILLER, AND CALCIUM SILICATE

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ABSTRACT

The global increase in plastic production, reaching 322 million tons in 2015 and projected to multiply by 2050, underscores the urgent need for sustainable alternatives. Biodegradable plastics, derived from renewable resources like starch, protein, and cellulose, offer an eco-friendly solution. Water hyacinth, rich in cellulose, presents potential as a raw material for biodegradable plastics. However, its rigidity, strength, and hydrophilicity necessitate improvements through additives. This study investigates the effects of sorbitol plasticizer, chitosan, and calcium silicate fillers on water hyacinth cellulose-based biodegradable plastics. Key tests include tensile strength, elongation, solubility, and biodegradability. Optimal compositions (cellulose 33.68%, chitosan 4.41%, calcium silicate 7.99%) achieved tensile strength of 15.97 MPa, 10.97% elongation, 52.5% solubility, and 21-day degradation. Characterization by FTIR, SEM, and XRD confirmed functional group changes, smoother surfaces, and reduced crystallinity. Further research should refine bioplastic molding methods.

KEYWORDS: Water hyacinth waste, cellulose, biodegradable plastic, chitosan, sorbitol, calcium silicate, FTIR, SEM, XRD

1. INTRODUCTION

Water hyacinth (Eichhornia crassipes) is a floating aquatic plant commonly found in water bodies. Its rapid growth has led to its classification as a weed in several regions of Indonesia, including lakes and other water reservoirs. This plant is often referred to as the most problematic aquatic weed globally due to its capacity to rapidly dominate and cover extensive water surfaces. Its rapid growth causes certain impacts on open waters such as clogging irrigation systems and hydropower plants, limiting livestock access to water, blocking waterways and canals, dominating water areas, changing water conditions (oxygen levels, pH and temperature) and reducing sunlight infiltration). Water hyacinth easily adapts to its environment, reproduces rapidly, and is able to compete strongly so that in a short time it will be abundant and fill the waters. Water hyacinth contains higher cellulose than non-cellulose (lignin, ash, fat, and other substances). The high fiber content of water hyacinth consists of 72.63% cellulose, 17% lignin, and 8% hemicellulose [1]. Therefore, water hyacinth has the potential to be utilized due to its abundant availability and high cellulose content.

Water hyacinth, with its high cellulose content, holds promise as a raw material for producing biodegradable plastics [2]. Unlike conventional plastics, biodegradable plastics can perform similar functions but degrade through microbial activity [3]. However, cellulose-based biodegradable plastics have drawbacks such as rigidity, strength, and hydrophilicity [2]. To address these limitations, the addition of fillers is necessary to enhance their properties. This research focuses on investigating the use of sorbitol as a plasticizer, along with chitosan and calcium silicate as fillers, to improve the quality of biodegradable plastics derived from water hyacinth cellulose.

2. MATERIALS AND METHODS

2.1 Materials

The materials used for this research include water hyacinth obtained from Lake Rawa Pening, sorbitol as a plasticizer, tapioca starch, chitosan, and calcium silicate as fillers, as well as NaOH and NaClO for the water hyacinth cellulose extraction process.

2.2 Methods

2.2.1 Pretreatment of water hyacinth

The water hyacinth pretreatment process refers to research conducted by [1]. In this study, water hyacinth was obtained from Lake Rawa Pening. The water hyacinth was then pretreated by drying, cutting, and pulverizing. Water hyacinth was chopped into small sizes of 1-2 mm using a knife and grinder. Then, it was dried in an oven at 105oC for 2 hours.

2.2.2 Cellulose extraction

The cellulose extraction process from water hyacinth was carried out by referring to the research of [3] with some modifications. The first step was to take water hyacinth powder from pretreatment and dissolve it in 20% NaOH solution by weight. The mixture was then heated at 100°C for 60 minutes using a hot plate while stirring with a magnetic stirrer. After heating, the mixture was filtered using a sieve to obtain water hyacinth extract. This extract was then dried in an oven at 105°C for 2 hours. The process was followed by bleaching using a solution of 3.5% NaOCl by weight and distilled water in a ratio of 1:1, which was heated at 90°C for 60 minutes while stirring. After that, the mixture was filtered using a 100-mesh sieving device, washed with distilled water, and dried again in an oven at 105°C for 2 hours.

2.2.3 Preparation of biodegradable plastics

The process of making biodegradable plastic was carried out by referring to the research of [4]. The steps began with weighing 10 grams of tapioca flour using a digital scale, then putting cellulose into a 250 mL erlenmeyer. Sorbitol, chitosan, cellulose, and $CaSiO_3$ were added according to the specified variations, followed by the addition of 60 mL of distilled water to dissolve the mixture. The mixture was heated using a hot plate while stirring with a magnetic stirrer at 150 rpm and 70°C for 30 minutes. Next, the solution was molded using 20 cm diameter Teflon as a film mold, leveled the surface with a spatula, and allowed to dry for two days. After the drying process, the biodegradable films formed were examined to ensure the results were as expected.

2.2.4 Preparation of biodegradable plastics

Tensile strength represents the highest stress a material can endure under stretching or pulling forces. This parameter was evaluated in a study by [4] using an MPY testing machine. The film sheets used for testing measured 2.5×15 cm and were conditioned at 50% relative humidity (RH) for 48 hours. The test utilized an Instron device configured with an initial grip separation of 50 mm, a crosshead speed of 50 mm/min, and a load cell capacity of 50 kg. The tensile strength was calculated using the following formula:

$$\tau = \frac{F_{max}}{A} \tag{1}$$

where:
τ = Tensile strength (MPa)
$F\ max$ = Tensile strength force (N)
A = Cross-sectional area (mm^2)

2.2.5 Percent elongation analysis

Percent elongation refers to the change in the maximum length of a film before breaking. This test, as outlined in the research by [4] was conducted using a testing machine. The film samples, measuring $2.5 \times$

15 cm, were conditioned at 50% relative humidity (RH) for 48 hours. The Instron equipment was configured with a crosshead speed of 50 mm/min, an initial grip separation of 50 mm, and a load cell capacity of 50 kg. The measurement was performed at the point of film rupture or tearing and calculated using the following formula:

$$\text{Percent Elongation} = \frac{(l_1 - l_0)}{l_0} \qquad \text{x} \qquad 100\%$$

(2)

Where:

l_0 = Initial length

l_1 = Length after breaking

2.2.6 Solubility analysis

The solubility test was conducted following the method described by [5]. Plastic film samples measuring 2×10 cm were immersed in a container filled with water and stirred manually. Solubility was determined as the percentage of the film that dissolved in water after being soaked for one week. The solubility percentage was calculated using the formula:

$$\text{Percent solubility} = \left(\frac{a - (c - b)}{a} \right) \times 100 \ \%$$

(3)

where:

a = Initial sample weight (grams)

b = Weight of filter paper (grams)

c = Dry weight of filter paper and sample (grams)

2.2.7 Biodegradability analysis

Biodegradability refers to the ability of a material, whether organic or inorganic, to undergo decomposition through the activity of microorganisms. To assess this property, a biodegradability test was performed to determine the time required for the plastic film sample to break down. This test employed the soil burial method, as described by [5] in which a sample measuring 4×1 cm² was buried in soil within a pot and left exposed to air. Observations were conducted weekly until the sample completely degraded or the bioplastic sheet was no longer visible.

3. RESULTS AND DISCUSSIONS

3.1 Experiment results data

The experimental data obtained were processed using Design Expert 13.0.5 program to optimize the best quantity of cellulose, chitosan, and calcium silicate for tensile strength, percent elongation, solubility,

biodegradability of biodegradable plastics. There are 15 research treatments and responses presented in Table 1. In addition, the relationship of each independent variable to the response is also presented.

Table 1: Effects of cellulose, chitosan, and calcium silicate addition on tensile strength, percent elongation, percent solubility, and biodegradability of biodegradable plastics

Run	Cellulose (%w/w)	Chitosan (%w/w)	Calcium Silicate (%w/w)	Tensile Strength (MPa)	Percent Elongation (%)	Percent Solubility (%)	Biodegradability (day)
1	20	4	4	10	14.5	64.5	25
2	25	4	4	11.3778	13.5	62	24
3	30	4	4	15	12.5	58.5	24
4	35	4	4	12.4444	11.5	54	21
5	40	4	4	19	10.5	52.5	21
6	20	10	7	13.6667	10	73.5	23
7	30	7	7	16.5714	9.5	59	24
8	40	10	7	22.2222	8.5	57.5	26
9	20	4	7	13.5	12	61.5	26
10	40	7	4	17.6667	9	56	22
11	30	7	7	14.4706	9.5	61	24
12	40	7	10	19.5	8	52.5	23
13	30	10	10	14.3333	8.5	62	27
14	20	7	10	13	11	64.5	26
15	20	7	4	12.7647	12.5	72.5	25

3.2 Biodegradable plastic optimization

After analyzing the data on the influence of each independent variable on the response variable. Optimization stages were carried out to obtain the best composition of cellulose, chitose, and calcium silicate to obtain the best response. The best response criteria were maximizing strong pull, targeting an

elongation percentage of 5%, setting the solubility percentage in the range of experimental results, and minimizing degradation time. The 3D plot of each response was obtained as follows.
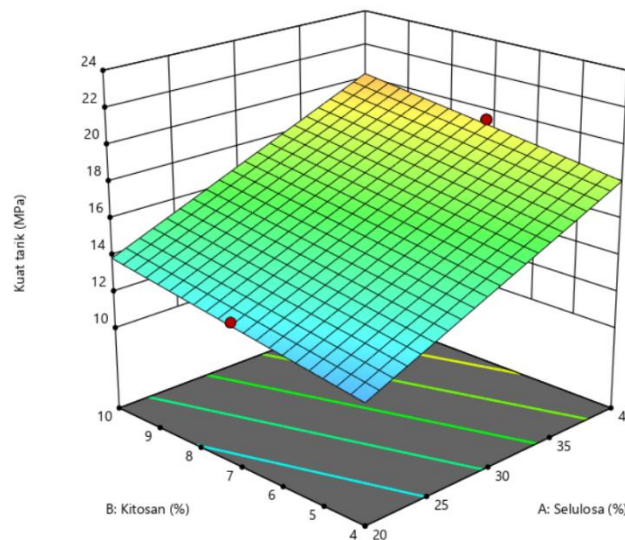

Figure 1: 3D plot of interaction of independent variables on tensile strength response (10% calcium silicate)

As the cellulose composition increases, the tensile strength of the bioplastics also rises. This aligns with the study conducted by [6], which investigated the effect of cellulose addition on tensile strength. Cellulose is known as a reinforcing component in composite materials, enhancing their mechanical strength. The increase in tensile strength is due to enhanced attractive interactions between molecules in the thin layers. This is caused by hydroxyl groups forming inter- and intramolecular hydrogen bonds, resulting in thin layers composed of fibers that mutually reinforce each other.
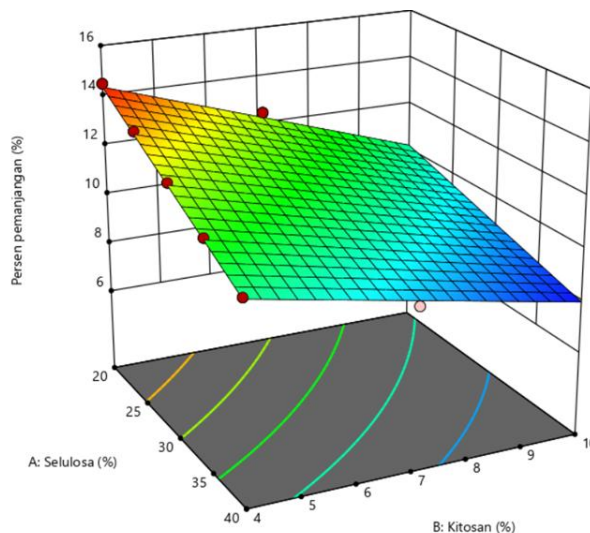
Figure 2: 3D plot of interaction of independent variables on the response of percent elongation (4% calcium silicate)

As cellulose increases, the elongation percentage will decrease. This is in accordance with research conducted by [7] that the addition of cellulose mass as a filler can reduce the elongation percentage of bioplastics. Cellulose has a high crystal area and a regular structure and has molecules with very long and strong bonds so that it can make bioplastic products hard and stiff which results in decreased elongation.
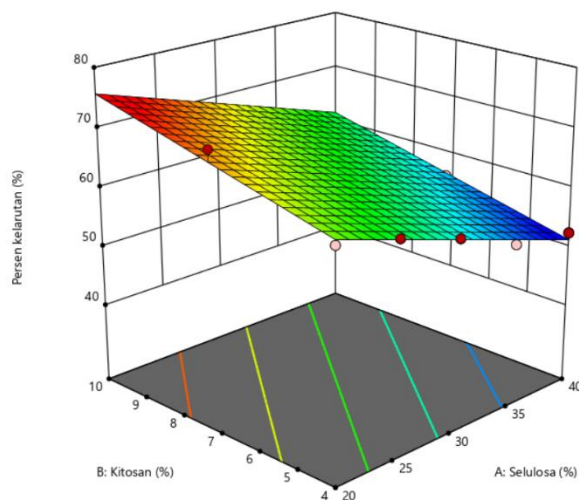

Figure 3: 3D plot of interaction of independent variables on the response of solubility percentage (Calcium silicate 4%)

Water hyacinth cellulose has a long and strong linear structure so that a crystalline area is formed that is difficult for water to penetrate [7]. The crystalline part of cellulose tends to be more resistant to water penetration than the amorphous part. Research shows that the more crystalline cellulose, the lower the solubility of the material in air because it is more difficult for water to penetrate the dense crystalline network. Therefore, increasing cellulose can reduce the solubility of biodegradable plastic in water [8].
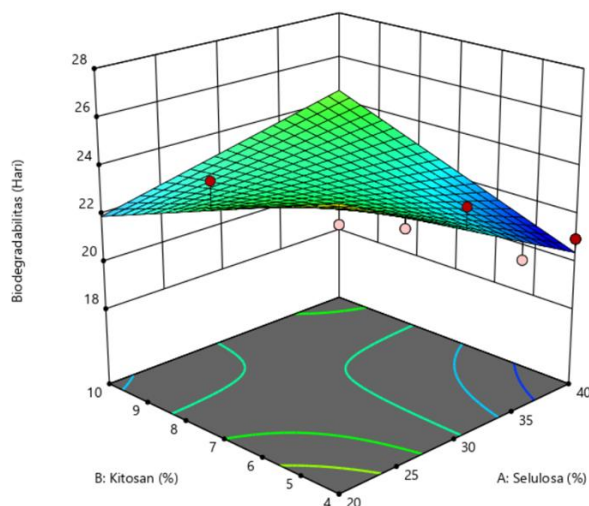

Figure 4: 3D plot of interaction of independent variables on biodegradability response (4% calcium silicate)

Through the results of Design Expert optimization using predetermined criteria, the optimization of cellulose composition was obtained by 40%; chitosan by 6.088%; and calcium silicate by 6.448%. This composition is predicted to have a tensile strength value of 18.380 MPa which is in accordance with SNI, namely a minimum of 13.7 MPa, an elongation percentage value of 8.804% which still does not meet SNI, namely a maximum of 5%, a solubility percentage of 52.5%, and the time required to decompose for 22 days.
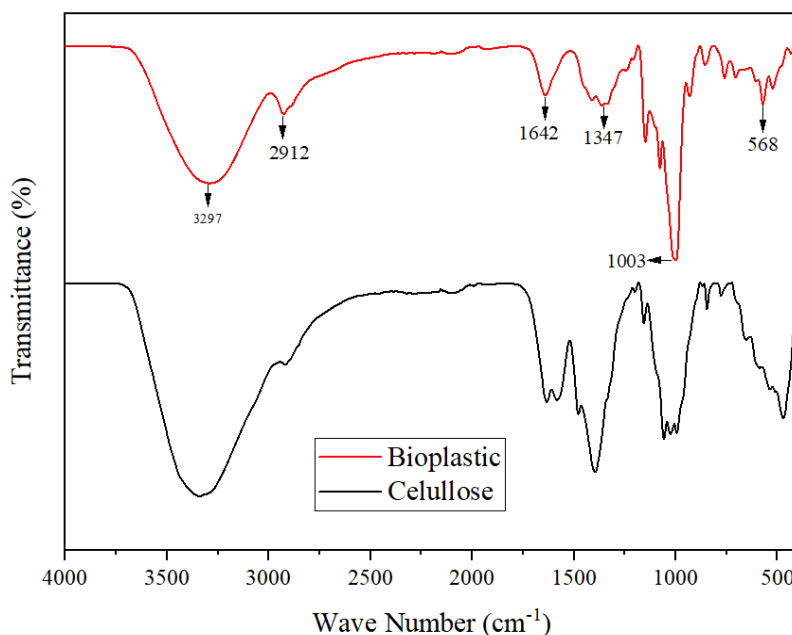
3.3 Fourier transform infrared (FTIR)

Figure 5: FTIR spectrum of bioplastic and cellulose

FTIR characterization is a characterization to analyze what functional groups are formed in bioplastics from water hyacinth cellulose and tapioca flour. In Figure 5 there are two curves with their respective peaks shown in Table 2.

Table 2: FTIR spectrum data of bioplastic

Wave Number (cm⁻¹)	Functional Group
3297	Hydroxyl (O-H)
2912	Alkyl (-CH)
1642	Carbonyl (C=O)
1347	Alkane (C-H)
1003	Alcohol (C-O)
568	Chloro (C-Cl)

Bioplastics are made from tapioca flour and cellulose as the main ingredients. In its manufacture, sorbitol plasticizer and chitosan filler along with calcium silicate are also added. The resulting hydroxyl functional group (O-H) indicates the presence of sorbitol which is used as a plasticizer [9]. The alkyl group indicates the presence of cellulose and the carbonyl group indicates the presence of hemicellulose. The alcohol

group indicates the presence of chitosan as a filler. Finally, there is a chloro group which probably comes from sodium hypochlorite from the cellulose bleaching process [10].

In general, the functional groups in cellulose and bioplastics are not significantly different. There is only a weakening and strengthening of the peaks which may occur due to the addition of tapioca flour, filler, and plasticizer so that there is a change in the concentration of the hydroxyl, alkane, and alcohol functional groups [9]. Therefore, it can be concluded that no chemical reaction occurs during the manufacture of bioplastics. The additional materials only strengthen or weaken the characteristics of water hyacinth cellulose to achieve the expected mechanical properties and biodegradability.

3.4 Scanning electron microscopy (SEM)

SEM (Scanning Electron Microscopy) analysis was conducted to analyze the morphological characteristics of cellulose (Figure 6) and biodegradable plastic (Figure 7) with magnifications of x500, x1000, x2500, and x5000. The Scanning Electron Microscope (SEM) is an advanced and versatile tool primarily used for studying the surface characteristics of materials. During SEM analysis, samples are bombarded with high-energy electrons, and the emitted electrons or X-rays are detected and analyzed. This emission provides valuable insights into the material's topography, morphology, composition, grain orientation, and crystallographic properties. Topography reveals the surface features, including texture, smoothness, or roughness, while morphology describes the shape and size of the material [11].
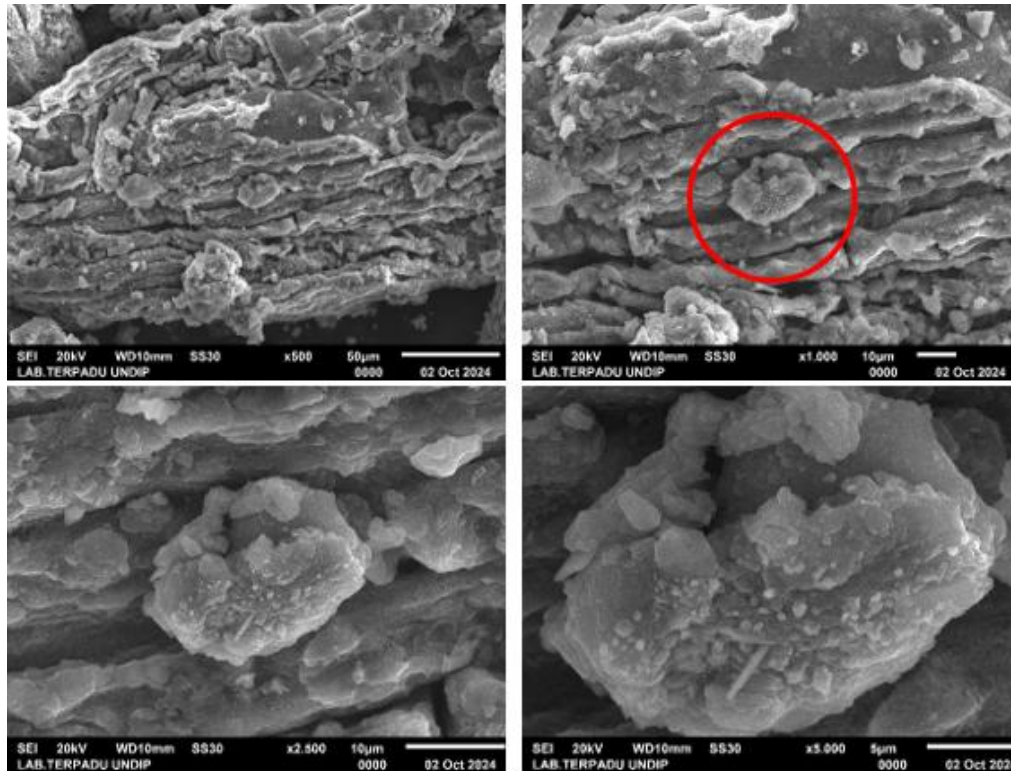
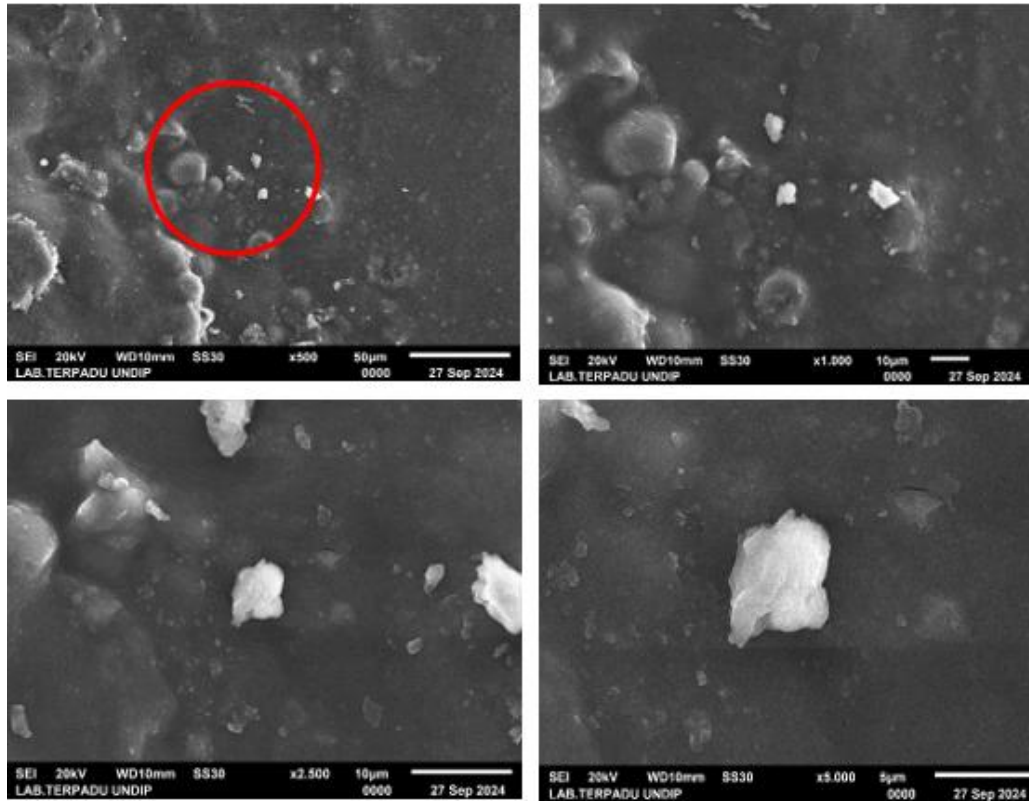**Figure 6: Results of SEM characterization of water hyacinth cellulose**

Figure 7: SEM characterization results of biodegradable plastics

Scanning Electron Microscopy (SEM) analysis of water hyacinth cellulose and biodegradable plastic showed significant differences in the surface structure and composition of the material. Water hyacinth cellulose has a layered and wrinkled structure with scattered crystal grains, indicating crystallinity that affects its mechanical and thermal properties. This layered structure shows good porosity potential. Overall, it appears that the resulting cellulose structure is not completely uniform, where there are areas with finer particles and areas with larger structures. Meanwhile, biodegradable plastic shows a smoother surface with uneven particle distribution. There are some small lumps and particle fragments scattered throughout the surface. This may indicate variations in composition or initial degradation of the plastic material. At higher magnification (x2,500 and x5,000), it is more obvious that there are larger and prominent grains in some areas. This may be the result of the undegraded polymer phase or the aggregation of additives used in the biodegradable plastic manufacturing process.

3.5 X-ray diffraction (XRD)

XRD characterization is a method to identify the structure of a material. The structure of the material itself is divided into two, namely crystal and amorphous. The crystal structure is a regular arrangement of atoms that repeats in three-dimensional space.
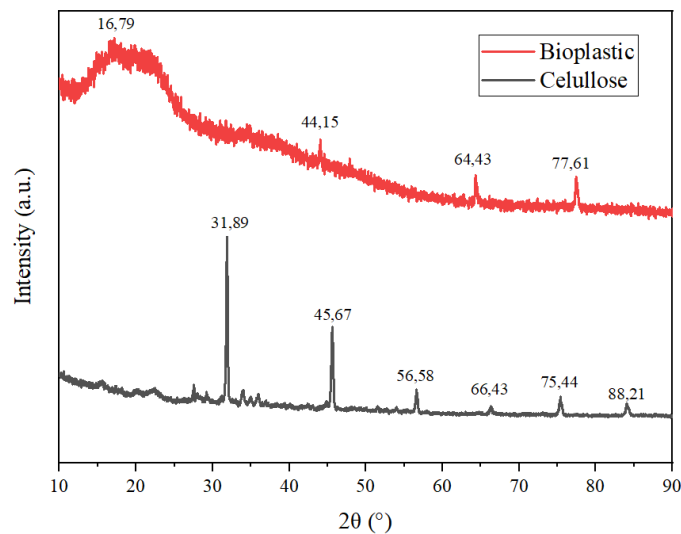

Figure 8: XRD curves of bioplastic and cellulose

Each basic unit is called a unit cell and is repeated periodically to form a crystal lattice. In contrast to the amorphous structure which has an irregular atomic arrangement and does not have a clear crystal structure. Through Figure 8, the crystallinity index (CI) of the material can be calculated to determine the percentage of crystals in a material using the following formula. The crystalline peak is first identified. Then, the identified peak is integrated to obtain the value of the crystalline area. Furthermore, the area of the crystal and amorphous is obtained by integrating the entire graph. Then the CI can be determined using the following formula.

$$\text{CI} = \frac{\text{Area of crystal structure}}{\text{Area of crystal+amorphous structure}} \times 100\% \tag{4}$$

Using Equation (4), the crystallinity index data can be obtained as follows in Table 3 below

Table 3: FTIR spectrum data of bioplastic

Material	CI (%)
Cellulose	28,86187088
Bioplastic	20,48999652

Through Table 3, it can be seen that there is a decrease in crystals from cellulose materials to bioplastic products. Crystalline structures tend to have better mechanical strength, while amorphous structures are more water-absorbing and easily degraded. The decrease in crystallinity can be caused by the addition of tapioca flour and sorbitol which basically have an amorphous structure. [12] found that bioplastics consisting of tapioca flour and glycerol (plasticizer) have low crystallinity values, thus accelerating the degradation ability of bioplastics. This is in accordance with the XRD curve obtained.

4. CONCLUSION

The addition of cellulose increased the tensile strength value but decreased the percent elongation and solubility, and accelerated the degradation time of the plastic. In contrast, the addition of chitosan increased the tensile strength value and percent solubility, but decreased the percent elongation and slowed down the degradation time. Meanwhile, the addition of calcium silicate increased the tensile strength value, decreased the percent elongation and solubility, and prolonged the degradation time. The optimal composition was found at 40% cellulose, 6.088% chitosan, and 6.448% calcium silicate, with tensile strength characteristics of 18.380 MPa, percent elongation of 8.804%, solubility of 52.5%, and degradation time of 22 days. FTIR analysis showed that the bioplastic had similar functional groups to cellulose, with only a change in concentration. SEM results indicated that cellulose had a non-uniform crystal structure, while bioplastics showed a smoother surface with uneven particle distribution. XRD analysis showed a decrease in crystallinity index from cellulose to bioplastics due to the addition of tapioca starch and amorphous sorbitol plasticizer.

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