Journal of Engineering and Technological Advances (JETA)
https://jeta.segi.edu.my/index.php/segi
2025 Vol. 10 No. 1

SYNTHESIS AND PERFORMANCE OF DEGRADABLE CELLULOSE-BASED PLASTICS FROM COCOA PODS (*THEOBROMA COCOA. L*) WITH POLYLACTIC ACID BLENDS

[1,2]Dewi, R.*., [2,3]Ananda, A., [1,2]Sylvia, N., [4]Riza, M., [5]Subhan, M., [6]Cionita, T., [7,8]Siregar, P.J., [9]Kusuma, S.B.

[1] Department of Chemical Engineering, Faculty of Engineering, Malikussaleh University, 24353 Lhokseumawe, Indonesia.
[2] Center of Excellence Technology Natural Polymer and Recycle Plastics, Faculty of Engineering, Malikussaleh University, 24353 Lhokseumawe, Indonesia.
[3] Department of Renewable Energy Engineering, Faculty of Engineering, Malikussaleh University, 24353 Lhokseumawe, Indonesia.
[4] Department of Chemical Engineering, Faculty of Engineering, Syiah Kuala University, 23111 Banda Aceh, Indonesia.
[5] Department of Entrepreneurship, Malikussaleh University, Faculty of Economics and Business, 24353 Lhokseumawe, Indonesia.
[6] Faculty of Engineering, Built Environment & Information Technology, SEGi University, Selangor 47810, Malaysia.
[7] Faculty of Mechanical and Automotive Engineering Technology, Universiti Malaysia Pahang Al-Sultan Abdullah (UMPSA), 26600 Pekan, Pahang, Malaysia.
[8] Centre for Automotive Engineering, Universiti Malaysia Pahang Al-Sultan Abdullah (UMPSA), 26600 Pekan, Pahang, Malaysia.
[9] Department of Industrial Engineering, Faculty of Engineering, University of Medan Area, 20000 Medan, Indonesia.
* Corresponding Author: rozanna.dewi@unimal.ac.id TEL: +62-81262315868

Received: 10 May 2025; Accepted: 20 June 2025; Published: 30 June 2025

doi: 10.35934/segi.v10i1.137

Highlights:

- *Making cellulose from cacao pods waste through a delignification process.*
- *Synthesis of degradable plastic from cellulose fibers modified with PLA matrix using a thermopressing method.*
- *Degradable plastic can completely decompose within 60 days.*

Abstract: The idea of degradable plastic has been studied abundantly in the past few decades. The goal is to find a way to replace non-degradable plastic. Cellulose is a promising material for making degradable plastic. This study presents a simple method for preparing degradable plastic using pure cellulose from cocoa pods. Experiments were conducted to study the mechanical strength, heat resistance, water absorption, and biodegradability of cellulose degradable plastics. Cellulose-based degradable plastic exhibits tensile strength that is nearly equivalent to that of the pure polymer poly lactic acid (PLA). Infrared spectroscopy (FTIR) analysis of the degradable plastic reveals the presence of O-H (hydroxyl group), CH_2 (alkane group), C=C (alkene group), and C-O (carboxylic acid group) bonds, indicative of a diverse molecular structure. Furthermore, the material exhibits significant thermal stability and thermal conductivity, suggesting its applicability in thermal-responsive applications. In the process of making bioplastics, the use of less PLA and more cellulose has been shown to make the material soak up water more quickly. These changes have made the material more waterproof. The degradable plastic decomposed quite well, breaking down completely within 68 days, indicating that cellulose from cocoa pods can be used to make degradable plastics.

Keywords: degradable plastic; cocoa pods cellulose; mechanical strength, heat resistance; biodegradability

1. Introduction

The ongoing demand for plastics and the subsequent deleterious environmental consequences have highlighted the pressing need for a sustainable alternative. Degradable plastics present a viable solution to address this critical issue (Shafqat et al, 2021). So far, about 8.3 billion tons of polymers have been produced, and about 6.3 million tons of polymer waste have been accumulated. Only 9% of this plastic waste was recycled. In the other cases, it was either burned or left over. The majority of it, about 79%, ends up either in disposal sites or the natural habitat. By 2050, experts estimate that 12000 metric tons of plastic will be discarded. In order to address the growing needs of consumers and to replace conventional plastics currently in the market, degradable plastic must be produced in a variety of forms and at a competitive cost of production (Nigam et al, 2022). A variety of raw materials are utilized in the production of bioplastics, including starch, polyhydroxybutyrate, polylactic acid, and other raw materials. Cellulosic biomass, a plentiful natural resource, is a promising material for the production of environmentally sustainable plastics.

Cellulose is a material that exhibits a range of beneficial characteristics, including its abundance, low cost, suitability for biological applications, minimal toxicity, lightweight nature, effective barrier properties against oxygen, excellent performance in stiffness and elasticity, and its capacity to absorb water (Darmenbayeva et al, 2025). Cocoa (*Theobroma cocoa L.*) is considered to be one of the globally most significant crops to be cultivated, with an approximate worldwide annual product output of five million tons of dry cocoa beans (Sánchez et al, 2023). A by-product, known as cocoa shell, is a result of the cocoa production process and has great potential for further research due to its composition. It contains fibre, protein, polyphenols, antioxidants, vitamins, and phenolic compounds (Bariši´c et al, 2020). Cocoa by-products consist of cocoa pods husk. It has been determined that the fruit is composed of approximately 70% cocoa pods, with the residual 30% comprising cocoa bean shell and pulp (Anoraga et al, 2024). The classification of biobased polymers utilized as bioplastics encompasses three distinct categories: plant-based (thermoplastic starch, TPS), polymerized monomers from biomass (PLAs), and bio-extracted polymers (PHBs) (Martin-Gamboa et al, 2023).

The research by Dewi et al (2024) employed starch from avocado seeds and PLA and utilized the Response Surface Methodology (RSM) to ascertain optimal operating conditions and the weight of materials utilized. This study provides a comparison with commercial and conventional plastics, paving the way for the use of these materials in housewares that are both sustainable and healthy. O'Loughlin et al (2023) have concluded that PLA is a brittle material with a Young's modulus of 2996-3750 MPa and an elongation at break of 1.3-7%. This is the temperature region where the polymer transitions from a hard and glassy state to a soft and rubbery state. This is the temperature at which the polymer starts changing its form. The water absorption of PLA can degrade the material. The properties of these materials are enhanced by the addition of biopolymers and by composite blends (Hussain et al, 2024).

The novelty of this research work conducted lies in the fact that the utilisation of degradable plastics extracted from cocoa pods shell cellulose and PLA can help reduce greenhouse gas emissions when compared to conventional plastic manufacture technologies. Polylactic acid (PLA) is a type of polymer that comes from natural resources like plants. It's very easy to use and breaks down quickly, usually within two years (Naser et al, 2021). One potentially promising solution is lignocellulose-based materials. This is an eco-friendly option that overcomes the main limitation of bioplastics, which is primarily due to their availability. The materials are biodegradable and biocompatible (Abe et al, 2021). Chemical and nutritional composition of cocoa bean shells: moisture (4-13.1 g/100 g); ash (6.0-9.1 g/100 g); carbohydrates (13.2-70.3 g/100 g); protein (18.2-27.4 g/100 g); lipids (2.3-6.5 g/100 g); dietary fibre (13.8 8-65.6 g/100 g); total phenolic content (22-100 mg GAE/g); total flavonoid content (7.5-21.8 mg RU/g1.6-43.9 mg CE/g), and total tannin content (2.3-25.3 mg CE/g) (Sánchez et al, 2023). Meanwhile, Campos-Vega et al (2018) found that fresh cocoa pods samples less than 1 cm in size contained 87% organic matter, consisting of 55.7% fiber, 20.6% nitrogen-free extract, 8.4% crude protein, and 2.5% fat. It has been demonstrated that cocoa fibre contains physicochemical and antioxidant characteristics, rendering it a viable option for the production of products necessitating high fiber content (Zhang et al, 2021). Cocoa is a distinguished product due to its high polyphenol content, specifically flavonoids, the health benefits of which have been extensively researched (Tušek et al, 2024).

Polymer films are produced through two primary methodologies. The initial method, designated as solvent casting, involves the introduction of a plastic injection molding machine solution into a mold and the subsequent removal of the moisture from the solvent. The second procedure is linked to the incorporation of the thermoplastic characteristics of certain bioplastics, for instance, the extruder method. A range of processes have been identified as being related to the given topic, including compression molding, injection molding, and blow molding (Shamloo et al, 2018). Previous investigations have indicated the feasibility of producing degradable plastics from cocoa pods cellulose with PLA modification through the application of the casting method, moulded using a hot press. The PLA concentrations utilized in this study ranged from 2, 4, 6, and 8 g, with cellulose varying from 1, 3, 5, and 7 g. A variety of PLA ratios were employed in an analysis to optimize the properties of the degradable plastics, including mechanical and thermal characteristics, chemical compounds, water absorption, and biodegradability.

2. Materials and Methods

2.1. Materials

Cocoa pods shells were taken from cocoa plantations and chocolate raw material producers located in Cot Girek Village, North Aceh District, Nisam Subdistrict, and cellulose was processed from cocoa pods. The aquadest is derived from PT Bratachem, a company based in Surabaya, Indonesia. Polylactic acid (PLA) ($C_3H_6O_3$) has a molecular weight of 90.08 g/mol (Sigma-Aldrich, 38534, 1G). Maleic anhydride (MA) has a purity level of 99 % (Sigma-Aldrich, 8.00408.1000) and Xylene from Merck (Supelco 108297). Polypropylene (PP), hydrogen peroxide (H_2O_2) 30 % 500 mL Merck-1.07209.0500, Polyethylene Glycol Millipore-8.17003 No CAS - 25322-68-3, Sodium Hydroxide (NaOH) Merck-1310-73-2 and Sodium Hypochlorite (NaOCl) Merck- 2828 90 00.

2.2. Methods

2.2.1. Preparation of Cocoa Pods Powder

The cocoa pods underwent a thorough cleansing process involving the use of clean water. Subsequently, the pods were meticulously cut into segments measuring 0.5 cm. Following this preparation, the cocoa pods were exposed to the sun for a period of 24 h to reduce their water content. The dried cocoa pods were subsequently cut into small pieces to facilitate the grinding process. Next, the cocoa beans are processed by grinding them in a blender, which produces flour. This flour was created with an objective of increasing the surface area available for extraction. The subsequent step involved the sifting of cocoa powder using an 80-mesh sieve, a process that ensures the separation of particles based on their size. Subsequently, the powder was stored in a closed container.

2.2.2. Cellulose Extraction

Cocoa powder is then added to distilled water at a ratio of 1:10, followed by thorough mixing and heating at 100°C for a duration of 60 minutes. Thereafter, the material was subjected to filtration and then dried in an oven at 105 °C for 60 minutes. The addition of 5 % NaOH was continued until the complete submersion of the substrate. Following this, the sample was autoclaved at 121℃ for a duration of 60 minutes, during which it was mixed with 5 % H_2O_2. After this, the specimen was further autoclaved at 70 ℃ for a duration of 30 minutes. The mixture was subsequently filtered, separation of the solid. Thereafter, 10 % NaOCl was incorporated, and the mixture was heated for a duration of 30 minutes. The solids are thoroughly washed with distilled water and dried in an oven heated to 105 °C. This resulted in the formation of dry powder of alpha cellulose.

2.2.3. Synthesis of Degradable Plastics

The initial step in the procedure entails the incorporation of 10 ml of xylene into a glass beaker, which contains 1 g of polyactic acid (PLA), 1 g of polyethylene glycol, 0.5 g of polypropylene, and 1 g of maleic anhydride. The mixture was heated and stirred at 125 °C until it became homogeneous. The addition of 1 g of cellulose was followed by a second stirring of the mixture at 150 °C until the desired homogeneity was achieved. Following the mixing process, the resultant material was shaped using a hot press at 80 °C, under a pressure of 130 kg/cm^2, and with a holding time of 15 minutes. After the pressing process, the material is left to cool at room temperature. The resulting degradable plastic was subjected to a variety of analytical procedures, including mechanical, chemical, water absorption, and biodegradability tests, to ascertain its characteristics.

2.2.4. Characterization and Analysis

Mechanical properties were tested in accordance with ASTM D-638. Dumbbell-shaped specimens 50 mm long are used in this test. Tensile strength and elongation were then measured using a Universal Testing Machine. The tensile strength and elongation values can be calculated using the following equations (1) and (2).

$$\sigma = \frac{F_{max}}{A} \quad (1)$$

Symbol σ shows the tensile strength (MPa), F_{max} the maximum stress (N), and A the area of the films (mm^2).

$$\varepsilon = \frac{\triangle l}{l_0} \quad (2)$$

ε is the strain (MPa), l is the gauge length, l_0 is the gauge length of the pre-sample, and l is the gauge length of the test sample after elongation.

A technique used to determine the composition of materials is Fourier Transform Infrared (FTIR) spectroscopy. Initially, a test specimen was exposed to infrared light. Subsequently, an analysis of the sample's chemical characteristics was conducted. The molecular structure of a sample is revealed by the wavelengths of light it absorbs. The FTIR analysis in the present study was performed in the frequency range from 550 to 4000 cm^{-1}. Thermal degradation, which is defined as polymer degradation, caused by excessive heat, is a major problem in many fields. During this process, hydrogen atoms are released from the polymer chain. In the context of polymers, the upper temperature limit is determined by the onset of thermal degradation. Thermal stability analysis for degradable plastics in this study was carried out through the application of Thermogravimetric Analysis (TGA). Water absorption testing is used to analyze how well degradable plastics can resist water. The swelling was measured according to ASTM D2765. Samples were first weighed and soaked in the solvent for 24 hours. The samples were reweighed after they had expanded, then dried and reweighed. The swelling was calculated using the following equation (3).

$$\text{Swelling} = \frac{\text{Weight of expanded sample} - \text{Weight of pre sample}}{\text{Pre} - \text{weight sample}} \times 100\ \% \quad (3)$$

The burial in soil test was used for the purposes of evaluating the biodegradability of the material. ASTM G-21-70 was strictly adhered to, causing the implementation of direct contact between the degradable plastic and the soil. The experiment was conducted by measuring and cutting 2 cm × 2 cm samples from the plastic. Prior to incorporation into the soil at a depth of 30 cm, the weight of the samples should be measured. Subsequent to a 72-h period, the samples were again examined. Subsequently, the samples were extracted from the soil and re-weighed. The biodegradability of plastics is calculated using equation (4).

$$\text{Biodegradabillity (\%)} = \frac{M0 - M1}{M0} \times 100\% \quad (4)$$

Where M_0 is the original weight (in grams) and M_1 is the finished weight (in grams).

3. Results and Discussion

3.1. Mechanical Characteristics Analysis

In the present study, the tensile strength of the material was evaluated by using a structural analysis device. Tensile strength is defined as the highest load that a polymer can withstand before failure. Excessive stress or structural deformation has been demonstrated to result in breakage. The experiment incorporated PLA 8 g & 1, 3, 5, and 7 g cellulose (P8C1; P8C3; P8C5; P8C7).

Table 1. Mechanical characteristics of degradable plastics from cocoa pods cellulose and PLA

Degradable Plastic	Tensile Strength (MPa)	Elongation (%)	Young's modulus (MPa)
P8C1	2.0749	0.66	301.0597
P8C3	2.0579	0.78	250.0710
P8C5	4.2549	1.08	390.7441
P8C7	3.3403	0.80	385.470
Pure PLA	5.00 - 42.0	15.0 – 100	2960 - 3600

Matweb (material property data) is used for pure PLA data.

As illustrated in Table 1, the mechanical property characteristics of the degraded plastics under investigation underwent significant alterations. The amount of PLA and cellulose in the material determines the tensile strength of the polymer. Tensile strength, defined as the maximum force required to cause a rupture in the film, is a critical measure of film durability. Elongation is defined as the extent to which a film can be stretched before it reaches its breaking point. Young's modulus is defined as the ratio of the change in stress to the change in strain in linear rheological models of stress/strain curves (Marichelvam et al, 2019).

As illustrated in Table 1, the tensile strength, elongation, and Young's modulus of the samples were found to range from (2.0579 to 4.2549 MPa), (0.66% to 1.08%), and (250.0710 to 390.7441 MPa), respectively. This observation indicates that the incorporation of (P8C1; P8C3; P8C5; P8C7) into the degradable plastic resulted in a significant enhancement of mechanical properties. The mechanical characteristics with the highest values, as determined by the analysis of the data, include tensile strength of 4.2549 MPa, elongation of 1.08%, and Young's modulus of 390.7441 MPa, which were obtained with the addition of P8C5. Previous studies have demonstrated the potential of synthesizing potato starches and blending them with PLA to enhance the properties of the resulting composites (Farshbaf Taghinezhad et al, 2025). The incorporation of a hydrophilic group into starch chains has been demonstrated to enhance the interfacial compatibility of the resultant polymer blend, thereby increasing the tensile strength of PLA-starch composites (Zuo et al, 2014). Chu et al (2024) found that adding 1% nanofibrillated cellulose to a material increased its tensile strain by 87.9% compared to the pure PLA. The tensile strength reached its optimum level depending on the amount of PLA and cellulose present in the material. This is similar to the tensile strength of cocoa pods cellulose degradable plastics; the tensile strength tends to increase with increasing concentration of cellulose used.

Modification of PLA was identified to perform as a filler, thereby increasing the tensile strength of cellulose-based degradable plastics derived from cocoa pods. The tensile strength (the force required to break the material) of the moderate properties group ranged from 10 to 100 MPa. According to the findings of the Standard Nasional Indonesia (SNI) standard, the tensile strength of plastic was determined to be within the range of 24.7 to 302 MPa (Dewi et al, 2024). The modified cellulose demonstrated that the degradable plastic film exhibited a reduced elongation value at break in comparison to the conventional plastic film. The SNI value for degradable plastic elongation is estimated to range between 21% and 220%. The elongation values from this study are not good enough to meet the requirements of the SNI standard. Additionally, it is relevant to note that the elongation values obtained from the present study have not been comparable to those of PET, with a range of 15 to 165% (Dewi et al, 2024) (Tian et al, 2024). The Young's modulus that was measured was similar in range to that of PE Braden RA 2-63 foil (222.73 MPa to 298.24 MPa) (Kubik & Zeman, 2014).

However, our study is similar to other PLA biopolymers in the database. These biopolymers have a tensile strength of 5 to 42 MPa, an elongation of 15 to 100%, and a Young's modulus of 2.96 to 3.60 GPa. It has been shown that the addition of filler composition results in an improvement in the modulus of elasticity value (Dewi et al, 2024). The aforementioned phenomenon is evidenced by a decline in tensile strength and, in many cases, a considerable reduction in deformability (Dewi et al, 2024).

3.2. FTIR Analysis

The specific wavelengths that a sample absorbs depend on the molecular structure of the sample. Figure 1 presents the results of an FTIR test on a degradable plastic made from P8C7.

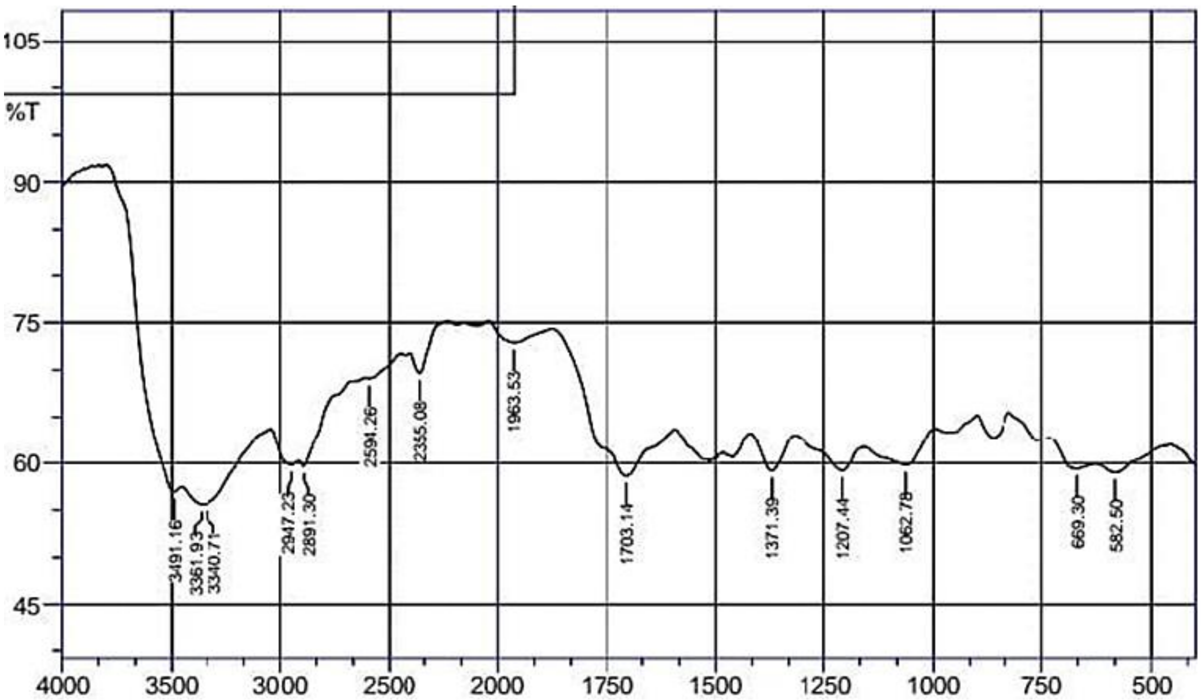

Figure 1. FTIR analysis of cellulose-based cocoa pods degradable plastics with PLA blends

In this study, FTIR analysis was used to chemically characterize a degradable plastic sample. The objective of the FTIR analysis was to ascertain the presence of specific functional groups within the sample under investigation. As illustrated in Figure 1, it was observed that the sodium hydroxide chemical compound present within the cellulose sample exhibited O-H bending vibrations, with these being recorded at specific wavenumbers, including 3491.16, 3361.93, and 3340.71 cm^{-1} (Singh et al, 2020). The presence of a CH_2 bond is indicative of a glucose content in cocoa pods cellulose, with wave numbers 2947.23, 2891.30, 2594.26, and 2335.08 cm^{-1} correlating with this finding (Razali et al, 2022). The wave numbers of 1963.53 and 1703.14 cm^{-1} are attributed to C=C and C-O bond vibrations present in xylene and polypropylene-graft-maleic anhydride compositions, with the potential for cross-linking with PLA (Dewi et al, 2024). The composition of various functional groups has the potential to influence the interfacial bond between cellulose and maleic anhydride monomers within degradable plastics, thereby contributing to the determination of their mechanical and physical properties (Dewi et al, 2024). An exothermic chemical reaction involving the combination of maleic anhydride and sodium hydroxide is observed at specific wave numbers, namely 11371.39 and 1207.78 cm^{-1}, indicating the presence of an N-H group in amines. This reaction is a critical step in the manufacturing process of degradable plastics (Navasingh et al, 2023).

In their research, Czechowski et al (2022) showed that the most common types of PLA can be found at 2994 and 2944 cm^{-1} (this is the stretching vibration of the C-H bond), 1382 cm^{-1} (this is the bending vibration of the C-H bond), and 866 cm^{-1} (this is the stretching vibration of the C-C bond). Yang et al (2019) found that a particular type of stretching vibration occurred at a frequency of 1745 cm^{-1} in pure PLA, and this vibration was found in the C=O bond. Gbadeyan et al (2023) studied bioplastics made from snail shells and bagasse cellulose using PLA. The FTIR spectra of these bioplastics showed the main group of C-O, C=O, and carboxylic acid-bending C-H stretching.

Degradable plastics from cocoa pods contain cellulose, which is hydrophilic (water-binding) and has hydroxyl and carbonyl groups. Degradable plastics are easily broken down by soil in the presence of these groups. This also indicates that degradable plastics have a higher amount of unbound hydroxyl groups. This is due to the fact that there are fewer atoms that can form hydrogen bonds.

3.3. TGA Analysis

The heat stability of degradable plastics is defined by the effects of polymer degradation, or break down, when heated. The TGA test was performed using a blend of P8C7 in the degradable plastic. As demonstrated in Figure 2, the curve resulting from the TGA analysis illustrates the change in mass versus temperature.

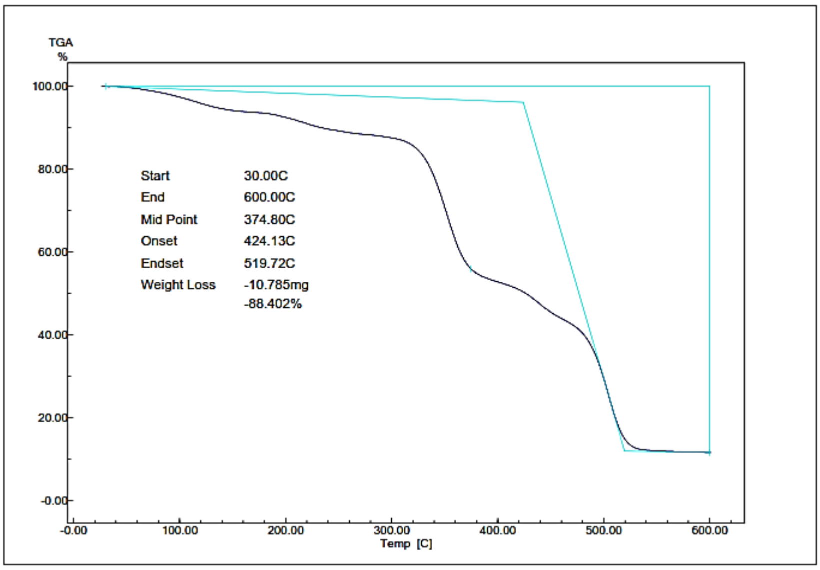

Figure 2. TGA analysis of cellulose-based cocoa pods degradable plastics with PLA blends

The results of this study demonstrate that cellulose-based degradable plastics maintained optimal thermal stability at high thermal loads. The degradation of cellulose from cocoa pods and PLA-degradable plastics exhibited multiple weight reduction phases, indicative of effective biodegradation. The phase of degradation of cellulose is associated with the most considerable weight loss, which is observed between 374.80 °C and 424.13 °C. Under this condition, the majority of the substance passes through a process of decomposition, reaching a state of complete depletion at an approximate temperature of 600 °C. The mass reduction of the areca nutshell-degradable plastic specimen was measured at 88.402 %, indicating a mass loss of -10.785 mg. This stage is characterized by the depolymerization and cracking of carbon chains within the cellulose structure, as well as the loss of hydrogen groups (Sanyang et al, 2015), (Mohammed et al, 2022), Gbadeyan et al. (2023). Bioplastic film was fabricated using a combination of 99.5 wt.% PLA and 0.5 wt.% sugarcane bagasse cellulose fibers (SBCF). This combination was found to offer peak thermal stability at 263 °C, suggesting that this optimal ratio of PLA and SBCF is conducive to enhancing the heat-related physical characteristics of films. The results of the present study were similar to those of other studies that reported the loading of SBCF as a method to improve thermal stability (Wang et al, 2020), (Lafia-Araga et al, 2021).

The degradation of fibers of lignocellulosic origin has been observed to occur at temperatures ranging from 400 °C to 500 °C, as evidenced by the presence of minor fluctuations in the resulting data set (Khotsaeng et al, 2023). In their research, Steven et al. (2022) investigated bioplastics derived from *Cladophora* sp. algae and epichlorohydrin (ECH). The cellulose bioplastic lost about 20% of its weight when it was heated to between 80 and 230 ºC. This could be due to the loss of moisture and the continued presence of residual ECH. Also, the ECH compound is slightly soluble in water. The process of evaporation occurs simultaneously with the loss of moisture content (Zainal et al, 2020).

3.4. Water Absorption Analysis

The degree of water resistance exhibited by degradable plastics was determined by the extent of their swelling when immersed in water. The degree of swelling is indicative of the amount of liquid the material absorbs, resulting in its expansion. As illustrated in Table 2, the water resistance test data of cocoa pods cellulose, when utilized in a blend with PLA, were examined through a series of swelling tests.

Table 2. Water absorption with swelling test of degradable plastics from cocoa pods, cellulose, and PLA variations

PLA (g)	Cellulose (g)	Degradable Plastic	Initial Mass (g)	Final Mass (g)	Swelling Degree (%)
2	1	P2C1	0.326	0.331	12.63
	3	P2C3	0.214	0.230	10.20
	5	P2C5	0.286	0.358	8.57
	7	P2C7	0.198	0.232	6.03
4	1	P4C1	0.502	0.560	18.22
	3	P4C3	0.322	0.381	12.11
	5	P4C5	0.399	0.450	10.43
	7	P4C7	0.431	0.472	7.60
6	1	P6C1	0.350	0.393	19.91

PLA (g)	**Cellulose (g)**	**Degradable Plastic**	**Initial Mass (g)**	**Final Mass (g)**	**Swelling Degree (%)**
	3	P6C3	0.374	0.428	16.56
	5	P6C5	0.516	0.633	14.54
	7	P7C7	0.421	0.470	9.76
8	1	P8C1	0.336	0.371	22.31
	3	P8C3	0.226	0.250	19.35
	5	P8C5	0.434	0.500	16.86
	7	P8C7	0.294	0.330	12.02

As demonstrated in Table 2, the interaction effect of cocoa pods cellulose and PLA mass incorporation on water absorption is evident. The swelling values obtained for the degradable plastic of cocoa pods cellulose with PLA are as follows: 2% PLA mass (6.03–12.63%); 4% PLA mass (7.60–18.22%); 6% PLA mass (9.76–19.91%); and 8% PLA mass (12.02–22.31%). The utilization of P8C1 has been demonstrated to offer the most substantial value in comparison to other swelling values. Conversely, the lowest value was attained when employing P2C7, yielding a value of 6.03%. The utilization of a reduced quantity of PLA, accompanied by an augmented amount of cellulose during the degradable plastics manufacturing process, has been shown to expedite the rate of water absorption. These modifications have led to an enhancement in the observed low water permeability. The addition of PLA improved the swelling properties due to an increased rate of absorption. Cellulose has hydroxyl (O-H) bonds; these bonds replace the bonds between the polymers that make up the degradable plastic. This results in a mechanically strong bioplastic. The hydroxyl (O-H) groups can bond with water (Hendrawati et al, 2023).

Fillers are added to degradable plastics to make them stronger. They are made from renewable materials that can be easily broken down and are easy to find. Adding fillers to bioplastics can make them stiffer, stronger, and more resistant to gas, melting, and heat (Coppola et al, 2021). In accordance with the EN ISO 317 standard, the maximum permissible deformation for plastic is 1.44% (Putranti et al, 2023). The results of this study indicate that the degradable plastic has not conformed to the established quality standard EN ISO 317.

3.5. Biodegradability Analysis

Soil microbes assist in the degradation of plastic when it is disposed of in a buried condition. In order to ascertain the optimal burial duration for a particular plastic product, the samples must first be buried in the soil of a defined weight for a predetermined time interval. As shown in Figure 3, the present study utilized cocoa pods cellulose (1, 3, 5, and 7%) as a principal compound, incorporating PLA fillers (2, 4, 6, and 8%) to ascertain the impact of the resultant degradation rate.

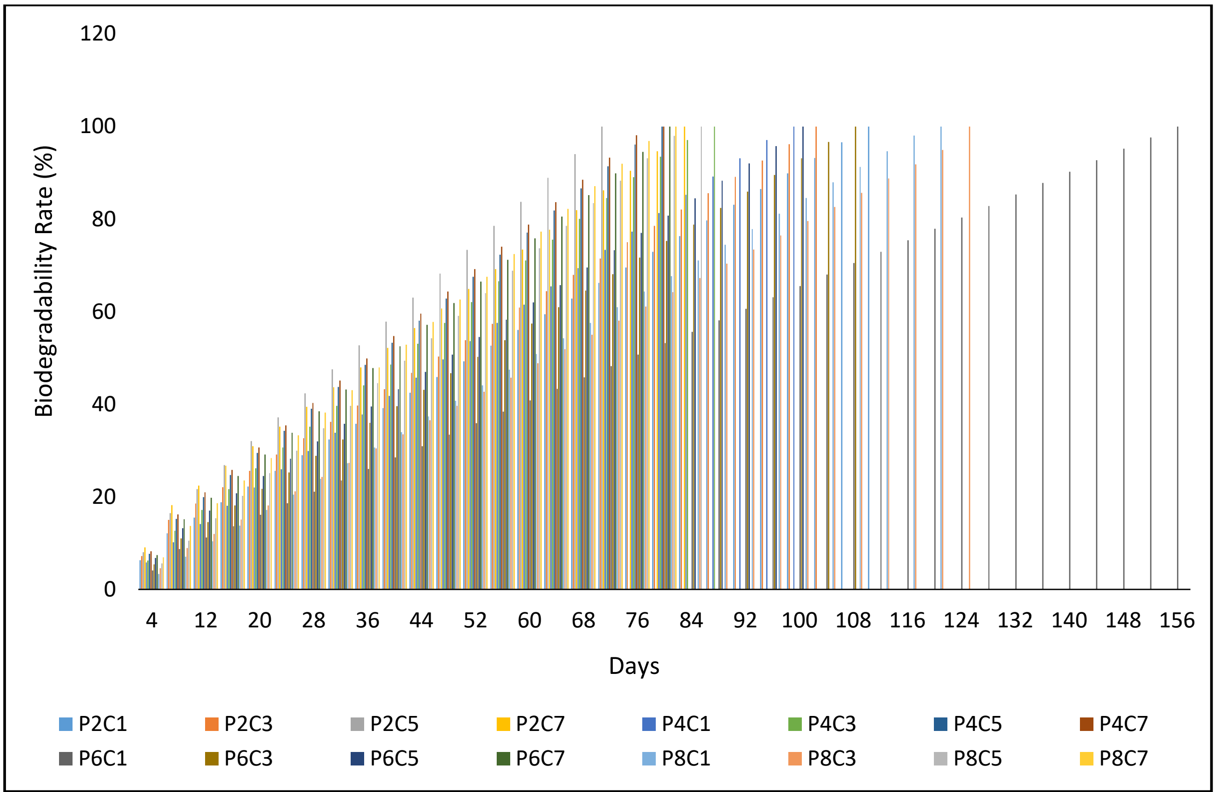

Figure 3. Biodegradability rate analysis of degradable plastic cocoa pods cellulose-based and PLA variations

The results presented in Figure 3 demonstrate that degradable plastic derived from cocoa pods cellulose, reinforced with PLA of 68 - 156 days, exhibits complete decomposition within the soil environment. The incorporation of P2C5 based plastic material led to a substantial enhancement in the rate of decomposition, which was found to be fully completed approximately 68 days after the initiation of the experiment. The specimens degraded more slowly with the increase in the PLA filler content. The complete degradation of P6C1 occurred on day 156 of the experiment. Brunšek et al (2023) conducted a study on bioplastics derived from cellulose fibers and PLA. The study revealed that flax fibers experienced the greatest mass loss following a 11-day exposure to microorganisms. Jute and sisal fibers experienced the second most significant decrease in mass. Jute fiber demonstrated a 44.51 % decrease in mass, while sisal exhibited a 7.92 % reduction. The chemical composition of the fibers appears to influence the rate of their biodegradation. Mahmoud et al (2022) in their study made biocomposites from apricot shell, PLA-modified walnut, a 38% reduction in composite weight was obtained after 12 months in soil burial tests.

According to ASTM D-6002, a standard that provides guidance for evaluating the degradability of environmentally friendly plastics in relation to ASTM D-20.96. The products in the aforementioned category are composed of a single polymer (homopolymer or linear copolymer). A minimum of 60% of the organic carbon contained within these products is required to be reduced to carbon dioxide within a period of 180 days, in order to meet the established standard (Dewi et al, 2024). It is imperative to comprehend the processes and rates of polymer degradation in diverse environments, both natural and controlled, in order to ascertain the behavior of polymers in changing conditions (Baidurah et al, 2022). According to a recent study, the addition of starch, cellulose, or derivatized lignocellulosic compounds to polyvinyl alcohol (PVA) has been shown to enhance the dissolution and biodegradation of the resultant polymer in soil environments (Elgharbawy et al, 2024).

4. Conclusion

In this research, we were able to produce degradable plastics from cocoa pods cellulose by hot-pressing method and adopting a simple fabrication procedure. The mechanical properties of cellulose degradable plastics from cocoa pods peels have values (tensile strength 4.2549 MPa, elongation 1.08%, and Young's modulus 390.7441 MPa) that are not yet comparable to the tensile strength of pure PLA. The FTIR analysis of the degradable plastic from cocoa pods cellulose revealed a greater abundance of free -OH groups. This is due to the fact that there are fewer atoms that can form hydrogen bonds. The phase of thermal decomposition of degradable plastic cellulose is correlated with the greatest loss of mass, which is measured within the temperature range of 374.80 °C to 424.13 °C. In swelling analysis, our research showed by using P8C1 gave the highest value of 22.31% compared to the other swelling values. The smallest value was found in the group that used P2C7, which resulted in 6.03 % swelling. The addition of P2C5 led to a significant increase in the decomposition rate. The experiment was fully completed in 68 days after it began.

The findings of this research have potential benefits, cellulose, the principal ingredient in the production of degradable plastics, is a natural material that is easily available and can be produced in large quantities from waste resources. This approach has the potential to enhance the economic viability of cocoa fruit, thereby generating increased income for traders and agricultural producers. In addition, the environment has an aesthetic value that can reduce environmental pollution. PLA-modified cocoa fruit peel cellulose-based degradable plastics and other materials have a wide range of applications. They can be used as household appliances, in automotive applications, and for welding, among other uses.

Conflict of Interest

The authors declare no conflict of interest.

Acknowledgement

The present study was performed within the Chemical Engineering Research Lab at the Faculty of Engineering, Malikussaleh University, Aceh.

Credit Author Statement

Conceptualization, D.R. and A.A.; methodology, S.N. and S.M.; validation, R.M., C.T. and S.P.J.; formal analysis, A.A.; investigation, S.N. and S.M.; resources, D.R. and R.M.; data curation, D.R.; writing—original draft preparation, A.A.; writing—review and editing, D.R. and S.P.J.; visualization, K.S.B.; supervision, C.T. and K.S.B.; project administration, D.R.

References

Abe, M. M., Martins, J. R., Sanvezzo, P. B., Macedo, J. V., Branciforti, M. C., Halley, P., Botaro, V. R., & Brienzo, M. (2021). Advantages and disadvantages of bioplastics production from starch and lignocellulosic components. *Polymers*, *13*(15), 2484. https://doi.org/10.3390/polym13152484

Anoraga, S. B., Shamsudin, R., Hamzah, M. H., Sharif, S., & Saputro, A. D. (2024). Cocoa by-products: A comprehensive review on potential uses, waste management, and emerging green technologies for cocoa pod husk utilization. *Heliyon*, *10*(16), e35537. https://doi.org/10.1016/j.heliyon.2024.e35537

Brunšek, R., Kopitar, D., Schwarz, I., & Marasović, P. (2023). Biodegradation properties of cellulose fibers and pla biopolymer. *Polymers*, *15*(17), 3532. https://doi.org/10.3390/polym15173532

Baidurah, S. (2022). Methods of analyses for biodegradable polymers: A review. *Polymers*, *14*(22), 4928. https://doi.org/10.3390/polym14224928

Barišić, V., Jozinović, A., Flanjak, I., Šubarić, D., Babić, J., Miličević, B., Doko, K., & Ačkar, Đ. (2020). Difficulties with use of cocoa bean shell in food production and high voltage electrical discharge as a possible solution. *Sustainability*, *12*(10), 3981. https://doi.org/10.3390/su12103981

Campos-Vega, R., Nieto-Figueroa, K. H., & Oomah, B. D. (2018). Cocoa (Theobroma cacao L.) pod husk: Renewable source of bioactive compounds. *Trends in Food Science & Technology*, *81*, 172–184. https://doi.org/10.1016/j.tifs.2018.09.022

Chu, H., Chen, Z., Chen, Y., Wei, D., Liu, Y., & Zhao, H. (2024). Mechanical properties and crystallinity of specific pla/cellulose composites by surface modification of nanofibrillated cellulose. *Polymers*, *16*(17), 2474. https://doi.org/10.3390/polym16172474

Coppola, G., Gaudio, M. T., Lopresto, C. G., Calabro, V., Curcio, S., & Chakraborty, S. (2021). Bioplastic from renewable biomass: A facile solution for a greener environment. *Earth Systems and Environment*, *5*(2), 231–251. https://doi.org/10.1007/s41748-021-00208-7

Czechowski, L., Kedziora, S., Museyibov, E., Schlienz, M., Szatkowski, P., Szatkowska, M., & Gralewski, J. (2022). Influence of uv ageing on properties of printed pla containing graphene nanopowder. *Materials*, *15*(22), 8135. https://doi.org/10.3390/ma15228135

Darmenbayeva, A., Zhussipnazarova, G., Rajasekharan, R., Massalimova, B., Zharlykapova, R., Nurlybayeva, A., Mukazhanova, Z., Aubakirova, G., Begenova, B., Manapova, S., Bulekbayeva, K., & Shinibekova, A. (2024). Applications and advantages of cellulose–chitosan biocomposites: Sustainable alternatives for reducing plastic dependency. *Polymers*, *17*(1), 23. https://doi.org/10.3390/polym17010023

Dewi, R., Sylvia, N., Zulnazri, Z., Riza, M., Siregar, J. P., Cionita, T., Kusuma, B. S. (2024). Characterization of sago starch-based degradable plastic with agricultural waste cellulose fiber as filler. *Environmental Science*. 11(2), 304-323. 10.3934/environsci.2024014.

Dewi, R., Sylvia, N., Zulnazri, Z., Riza, M., Siregar, J. P., Cionita, T. (2024). Use calcium silicate filler to improve the properties of sago starch based degradable plastic. *Environmental Science*. 12(1), 1-15. 10.3934/environsci.2025001.

Dewi, R., Sylvia, N., Zulnazri, Z., Fithra, H., Riza, M., Siregar, J. P., Cionita, T., Fitriyana, D. F., & Anis, S. (2024). The optimization of avocado-seed-starch-based degradable plastic synthesis with a polylactic acid (PLA) blend using response surface methodology (RSM). *Polymers*, *16*(16), 2384. https://doi.org/10.3390/polym16162384

Elgharbawy, A. S., El Demerdash, A.-G. M., Sadik, W. A., Kasaby, M. A., Lotfy, A. H., & Osman, A. I. (2024). Synthetic degradable polyvinyl alcohol polymer and its blends with starch and cellulose—A comprehensive overview. *Polymers*, *16*(10), 1356. https://doi.org/10.3390/polym16101356

Farshbaf Taghinezhad, S., Mansourieh, M., Abbasi, A., Major, I., & Pezzoli, R. (2025). Improved compatibilized TPS/PLA blends: Effects of singular and binary compatibilization systems. *Carbohydrate Polymer Technologies and Applications*, *10*, 100819. https://doi.org/10.1016/j.carpta.2025.100819

Gbadeyan, O. J., Linganiso, L. Z., & Deenadayalu, N. (2023). Assessment and optimization of thermal stability and water absorption of loading snail shell nanoparticles and sugarcane bagasse cellulose fibers on polylactic acid bioplastic films. *Polymers*, *15*(6), 1557. https://doi.org/10.3390/polym15061557

Hussain, M., Khan, S. M., Shafiq, M., & Abbas, N. (2024). A review on PLA-based biodegradable materials for biomedical applications. *Giant*, *18*, 100261. https://doi.org/10.1016/j.giant.2024.100261

Hendrawati, Liandi, A. R., Ahyar, H., Maladi, I., Azhari, A., & Cornelia, M. (2023). The influence of the filler addition of rice husk cellulose, polyvinyl alcohol, and zinc oxide on the characteristics of environmentally friendly cassava biodegradable plastic. *Case Studies in Chemical and Environmental Engineering*, *8*, 100520. https://doi.org/10.1016/j.cscee.2023.100520

Kubík, Ľ., & Zeman, S. (2014). Mechanical properties of polyethylene foils. *Journal of Central European Agriculture*, *15*(1), 138–145. https://doi.org/10.5513/JCEA01/15.1.1425

Khotsaeng, N., Simchuer, W., Imsombut, T., & Srihanam, P. (2023). Effect of glycerol concentrations on the characteristics of cellulose films from cattail (Typha angustifolia L.) flowers. *Polymers*, *15*(23), 4535. https://doi.org/10.3390/polym15234535

Lafia-Araga, R. A., Sabo, R., Nabinejad, O., Matuana, L., & Stark, N. (2021). Influence of lactic acid surface modification of cellulose nanofibrils on the properties of cellulose nanofibril films and cellulose nanofibril–poly(Lactic acid) composites. *Biomolecules*, *11*(9), 1346. https://doi.org/10.3390/biom11091346

Marichelvam, M. K., Jawaid, M., & Asim, M. (2019). Corn and rice starch-based bio-plastics as alternative packaging materials. *Fibers*, *7*(4), 32. https://doi.org/10.3390/fib7040032

Martin-Gamboa, M., Allegue, L. D., Puyol, D., Melero, J. A., & Dufour, J. (2023). Environmental life cycle assessment of polyhydroxyalkanoates production by purple phototrophic bacteria mixed cultures. *Journal of Cleaner Production*, *428*, 139421. https://doi.org/10.1016/j.jclepro.2023.139421

Mohammed, A. A. B. A., Hasan, Z., Omran, A. A. B., Elfaghi, A. M., Khattak, M. A., Ilyas, R. A., & Sapuan, S. M. (2022). Effect of various plasticizers in different concentrations on physical, thermal, mechanical, and structural properties of wheat starch-based films. *Polymers*, *15*(1), 63. https://doi.org/10.3390/polym15010063

Mahmoud, Y., Belhanche-Bensemra, N., & Safidine, Z. (2022). Impact of microcrystalline cellulose extracted from walnut and apricots shells on the biodegradability of Poly (Lactic acid). *Frontiers in Materials*, *9*, 1005387. https://doi.org/10.3389/fmats.2022.1005387

Naser, A. Z., Deiab, I., & Darras, B. M. (2021). Poly(Lactic acid) (Pla) and polyhydroxyalkanoates (Phas), green alternatives to petroleum-based plastics: A review. *RSC Advances*, *11*(28), 17151–17196. https://doi.org/10.1039/D1RA02390J

Navasingh, R. J. H., Gurunathan, M. K., Nikolova, M. P., & Królczyk, J. B. (2023). Sustainable bioplastics for food packaging produced from renewable natural sources. *Polymers*, *15*(18), 3760. https://doi.org/10.3390/polym15183760

Nigam, S., Das, A. K., Matkawala, F., & Patidar, M. K. (2022). An insight overview of bioplastics produced from cellulose extracted from plant material, its applications and degradation. *Environmental Sustainability*, *5*(4), 423–441. https://doi.org/10.1007/s42398-022-00248-3

O'Loughlin, J., Doherty, D., Herward, B., McGleenan, C., Mahmud, M., Bhagabati, P., Boland, A. N., Freeland, B., Rochfort, K. D., Kelleher, S. M., Fahy, S., & Gaughran, J. (2023). The potential of bio-based polylactic acid (Pla) as an alternative in reusable food containers: A review. *Sustainability*, *15*(21), 15312. https://doi.org/10.3390/su152115312

Putranti, L. N., & Nugraheni, P. S. (2023). Effect of carboxymethyl cellulose addition on the characteristic of chitosan-based bioplastic. *IOP Conference Series: Earth and Environmental Science*, *1289*(1), 012038. https://doi.org/10.1088/1755-1315/1289/1/012038

Razali, N. A. M., Mohd Sohaimi, R., Othman, R. N. I. R., Abdullah, N., Demon, S. Z. N., Jasmani, L., Yunus, W. M. Z. W., Ya'acob, W. M. H. W., Salleh, E. M., Norizan, M. N., & Halim, N. A. (2022). Comparative study on extraction of cellulose fiber from rice straw waste from chemo-mechanical and pulping method. *Polymers*, *14*(3), 387. https://doi.org/10.3390/polym14030387

Singh, A. A., Genovese, M. E., Mancini, G., Marini, L., & Athanassiou, A. (2020). Green processing route for polylactic acid–cellulose fiber biocomposites. *ACS Sustainable Chemistry & Engineering*, *8*(10), 4128–4136. https://doi.org/10.1021/acssuschemeng.9b06760

Sánchez, M., Laca, A., Laca, A., & Díaz, M. (2023). Cocoa bean shell: A by-product with high potential for nutritional and biotechnological applications. *Antioxidants*, *12*(5), 1028. https://doi.org/10.3390/antiox12051028

Sanyang, M., Sapuan, S., Jawaid, M., Ishak, M., & Sahari, J. (2015). Effect of plasticizer type and concentration on tensile, thermal and barrier properties of biodegradable films based on sugar palm (Arenga pinnata) starch. *Polymers*, *7*(6), 1106–1124. https://doi.org/10.3390/polym7061106

Shafqat, A., Al-Zaqri, N., Tahir, A., & Alsalme, A. (2021). Synthesis and characterization of starch based bioplatics using varying plant-based ingredients, plasticizers and natural fillers. *Saudi Journal of Biological Sciences*, *28*(3), 1739–1749. https://doi.org/10.1016/j.sjbs.2020.12.015

Shamloo, A., Fathi, B., Elkoun, S., Rodrigue, D., & Soldera, A. (2018). Impact of compression molding conditions on the thermal and mechanical properties of polyethylene. *Journal of Applied Polymer Science*, *135*(15), 46176. https://doi.org/10.1002/app.46176

Steven, S., Fauza, A. N., Mardiyati, Y., Santosa, S. P., & Shoimah, S. M. (2022). Facile preparation of cellulose bioplastic from cladophora sp. Algae via hydrogel method. *Polymers*, *14*(21), 4699. https://doi.org/10.3390/polym14214699

Tian, J., Kong, Y., Qian, S., Zhang, Z., Xia, Y., & Li, Z. (2024). Mechanically robust multifunctional starch films reinforced by surface-tailored nanofibrillated cellulose.

Composites Part B: Engineering, *275*, 111339. https://doi.org/10.1016/j.compositesb.2024.111339

Tušek, K., Valinger, D., Jurina, T., Sokač Cvetnić, T., Gajdoš Kljusurić, J., & Benković, M. (2024). Bioactives in cocoa: Novel findings, health benefits, and extraction techniques. *Separations*, *11*(4), 128. https://doi.org/10.3390/separations11040128

Wang, Q., Ji, C., Sun, L., Sun, J., & Liu, J. (2020). Cellulose nanofibrils filled poly(Lactic acid) biocomposite filament for fdm 3d printing. *Molecules*, *25*(10), 2319. https://doi.org/10.3390/molecules25102319

Yang, Z., Li, X., Si, J., Cui, Z., & Peng, K. (2019). Morphological, mechanical and thermal properties of poly(Lactic acid) (Pla)/cellulose nanofibrils (Cnf) composites nanofiber for tissue engineering. *Journal of Wuhan University of Technology-Mater. Sci. Ed.*, *34*(1), 207–215. https://doi.org/10.1007/s11595-019-2037-7

Zainal, S. H., Mohd, N. H., Suhaili, N., Anuar, F. H., Lazim, A. M., & Othaman, R. (2021). Preparation of cellulose-based hydrogel: A review. *Journal of Materials Research and Technology*, *10*, 935–952. https://doi.org/10.1016/j.jmrt.2020.12.012

Zhang, B., Yang, X., Liu, L., Chen, L., Teng, J., Zhu, X., Zhao, J., & Wang, Q. (2021). Spatial and seasonal variations in biofilm formation on microplastics in coastal waters. *Science of The Total Environment*, *770*, 145303. https://doi.org/10.1016/j.scitotenv.2021.145303

Zuo, Y., Gu, J., Yang, L., Qiao, Z., Tan, H., & Zhang, Y. (2014). Preparation and characterization of dry method esterified starch/polylactic acid composite materials. *International Journal of Biological Macromolecules*, *64*, 174–180. https://doi.org/10.1016/j.ijbiomac.2013.11.026