Degradable Bioplastic Developed from Pine-Wood Nanocellulose as a Filler Combined with Orange Peel Extract

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Abstract: *This research presents the degradable bioplastics developed from pinewood nanocellulose as a filler in PVA matrices. The steps involve the isolation and characterization of cellulose and nanocellulose. Meanwhile, the manufacturing of degradable bioplastic involves the combination of PVA, nanocellulose, and with or without orange peel extract. The effect of bioplastics without the addition of citric acid and orange peel extract is also reported as a comparison. It is found that orange peel extract improves the tensile strength (1708.54 kPa), elastic modulus (42.71 kPa), elongation (40%), and degradability (78.44% in 2 weeks) compared to bioplastic without the orange peel extract. These results indicate that orange peel extract acts as a reinforcing agent in PVA-nanocellulose bioplastic.*

Keywords: *pinewood-nanocellulose; degradable-bioplastic; orange peel extract*

INTRODUCTION

Plastic waste has reached 6.9 billion tons on a global scale, and every year 34 million tons of plastic waste are produced, with up to 93% of that waste ending up in a landfill or the ocean [1-2]. Fan et al. predicted in the year 2040, it is estimated that 710 Mt of plastic waste will be disposed of and pollute the surface and groundwater sources [3]. Java Island produces 189,349 tons per month of plastic waste, and the Malang regions generate 4,829.77 tons per month [4]. Plastic waste takes nearly a million years to naturally decompose in the environment due to the non-degradable characteristic of plastic materials [5-6]. Thus, it severely threatens the environment and living matters, such as sewage blocking up and animal and human health problems [7]. As an alternative strategy, degradable bioplastics must be developed to replace the petroleum-based plastic used in packaging.

Generally, bioplastics can be made from several components, such as reinforcing agents, crosslinkers, and additives components. According to references, cellulose and nanocellulose can be used as reinforcing agents [8-9], combining with polyhydroxyalkanoate (PHA), polyvinyl alcohol (PVA), or polylactic acid (PLA) [10-11]. The cellulose or nanocellulose can be isolated from forest and agricultural wastes. In addition, the other component reported as a linker, i.e., citric acid, sorbitol, glyoxal, dicarboxylic acid, and organic acid reagents like maleic acid and ascorbic acid [12-14]. Meanwhile, additive materials are generally added to improve the degradability characteristic and/or appearance of bioplastic [15]. The natural plant extract has been reported previously to increase the strength of bioplastic [16-17]. Nasihin et al. used pine flower waste cellulose combined with starch, PVA, and turmeric extract. The resulting bioplastic has a gradation color from yellow to colorless with the decreasing turmeric extract concentration [18]. However, the strength and elasticity of bioplastic are lower.

According to the report from the Ministry of Industry of the Republic of Indonesia, the number of wood industries in Indonesia has increased by 62.7%. As an outcome, a large amount of biomass contained-cellulosic material such as sawdust, tree bark, and other forest by-product is wasted [19-20], and only 40% of waste from the wood industry or agriculture is processed [21]. Similar data also reported that pine forests in East Java (a total area of about 800 kha) also provide biomass as waste. The pine flower waste still composes its important volatile terpenoid compounds [22], including cellulosic material (22.22%) [23]. Meanwhile, pine wood contains 54.9% of cellulose, hemicellulose (14%), lignin (24.3%), ash (1.1%), and silica (0.2%) [24]. The treatment process of the pine wood by using acid hydrolysis and bleaching produces pure cellulose. The step removes polyphenolic and lignin from the matrix [25]. This step generally provides cellulose in micrometric size. Further hydrolysis process to produce cellulose in nano-size is performed by cutting the β-glycosidic bonds of cellulose polymer [26]. The resulting nanocellulose can form fiber or crystalline nanocellulose, which depends on the original source of cellulose and the strategy to hydrolyze it [27]. For example, nanocrystalline cellulose can be isolated after acidic hydrolysis using citric acid, sulfuric acid, and hydrochloric acid [18,28]. Enzymatic hydrolysis of cellulose also produces crystalline nanocellulose [25]. Meanwhile, oxidation following hydrolysis generally provides amorphous nanocellulose [29]. Besides that, nanocrystalline cellulose is also produced through hydrolysis under mechanical and ultrasonication-assisted hydrolysis [30]. According to Agustin et al. applying nanocellulose as a filler for bioplastic affect the physical performance of bioplastics, such as tensile strength and modulus value [31]. Another paper reported an increase in the transparency of bioplastic [32].

On the other side, Indonesia is also the main producer of orange fruits. It is sold and consumed directly as fresh fruit or converted into packaging juice. The global waste of orange peel is projected to be 15×10^6 tons annually [33]. Meanwhile, Indonesia produces 6.8×10^8 tons of orange peel waste [34]. Hassan et al. reported that orange peel contains ascorbic acid of around 59 mg/100 g (w/w) [35]. Ascorbic acid is an organic acid containing polyhydroxy and ester functional groups. This substituent is predicted to be applied as a linker to bind with nanocellulose and PVA [12-13]. Thus, it could improve the binding inter-molecular component of bioplastic. This paper reports the contribution of orange extract containing ascorbic acid to improve the performance of degradable bioplastic developed with nanocellulose from pinewood as a filler.

■ EXPERIMENTAL SECTION

Materials

Pinewood (*Pinus merkusii*) waste was used as a natural source of cellulose taken from the Brawijaya University Forest, and the pinewood waste was ground into powder. The chemical reagents used in this research were sodium hypochlorite (99% purity Merck, Germany), acetic acid (99% purity SMART LAB, Indonesia), sodium hydroxide (99% purity Merck, Germany), hydrogen peroxide (30% purity SMART LAB, Indonesia), hydrochloric acid (37% purity Merck, Germany), polyvinyl alcohol (99% purity Merck, Germany), glycerin (85% purity Merck, Germany), and citric acid (99% purity Merck, Germany).

Instrumentation

Instruments used for research include ultrasonicator (DELTA D150H, 48 kHz, 220-240 volt), Fourier Transform Infrared (Shimadzu FTIR-8400S), X-ray Diffractometer (XRD PANanalytical- type X'pert Pro), Scanning Electron Microscope (FEI SEM Inspect-S50), Tensile Strength Apparatus (ZP-50 N).

Procedure

Preparation of cellulose

The procedure to produce cellulose followed the reference [36] with minor modifications. Pinewood waste was washed under running water to remove the impurity from the pinewood waste and dried under the sunlight for 2–3 days. Then, the clean pinewood waste was ground into powder. The powder was weighed to 50 g and put into a 1000 mL beaker glass. Then, the delignification process continued with the addition of sodium hypochlorite 6% and acetic acid 1% solution with a ratio of powder to solution was 1:10 (w/v). The mixture was stirred and heated at 70 °C for 1 h. After that, the alkaline bleaching process was done with the addition of sodium hydroxide 4% and hydrogen peroxide 24% (1:1 v/v) with the ratio of powder to the mixture was 1:10 (w/v). The mixing under stirring was conducted for 2 h at 50 °C, and this process was repeated up to 3 times until the white cellulose was obtained. Then, it was neutralized and dried in an oven at 50 °C for 24 h. Characterization was undergone by means of FTIR, XRD, and SEM.

Preparation of nanocellulose

The isolated cellulose from the previous step was further hydrolyzed following reference [28] with modification. The modification procedure in this step involves hydrolysis assisted by ultrasonication. The detailed procedure is reported elsewhere separately. The drying process was undertaken in an oven at 50 °C for 1 h. Nano cellulose was produced as a white-fine powder after separation under centrifugation, and further characterization was taken place using FTIR, XRD, and SEM.

Preparation of orange peel waste extract

Orange peel waste samples were obtained from the industrial waste of orange juice production in Malang, East Java, Indonesia. Samples were washed and cut into small sizes. Then, it was dried in an oven at 50 °C for 48 h. The dried peel was ground to get a powder. Then, orange peel powder was extracted with ethanol 50%, using a powder-to-solvent ratio of 1:2 (w/v) [37]. The extraction process was assisted by ultrasonication (ultrasonicator D150H and 220–240 V power and 48 kHz) for 1 h at room temperature. The extract was filtered using a Buchner funnel, and the resulting filtrate was evaporated under reduced pressure using a rotary evaporator to yield crude extract.

Fabrication of bioplastics

The procedure to fabricate bioplastics was done following Hussein et al. [38] with some modifications. The composition ratio of each component of bioplastic is displayed in Table 1. The steps to fabricate bioplastic involves the mixing of 10% PVA solution with 1.5% nanocellulose in a 50-mL Erlenmeyer. This mixture was stirred for 1 h at 90 °C, then 2% citric acid solution or 2% orange peel extract was added to the mixture. The mixture was further stirred for 1 h at the same temperature. One mL of glycerin was added to the mixture until a viscous solution was formed. Then, it was poured into a molding glass (20 × 20 cm^2) and left in an oven at 50 °C for 24 h.

Characterization

Fourier transform infrared (FTIR) spectroscopy analysis. Fourier Transform Infrared Spectroscopy (FTIR) analysis of nanocellulose and bioplastic was done to determine the absorption bands of functional groups composed in the nanocellulose and bioplastic. The measurement was performed by scanning in a range of 400–4000 cm^{-1}.

X-ray diffraction (XRD) analysis. The crystallinity of cellulose and nanocellulose was measured from the crystallinity index (CI) value. It was conducted using PanAnalytical type X'pert Pro with a range of 2θ = 10–90° and a scan rate of 2°/min. The CI was calculated following the equation [39].

$$CI = \frac{I_{002} - I_{amorph}}{I_{002}} \times 100\% \quad (1)$$

Scanning electron microscope (SEM) analysis. Scanning Electron Microscope (SEM) was used to observe morphological surface cellulose, nanocellulose, and bioplastic. The samples were put into a specimen holder and analyzed using field emission scanning electron microscope (FEI SEM) Inspect-S50.

Mechanical properties analysis. The mechanical properties of bioplastic were determined using the Tensile Strength apparatus model: ZP-50N. The tensile strength, elongation break, and elastic modulus parameters were measured following the ASTM-D1708-18 standard method (ASTM, 2018). The bioplastic was

Table 1. The composition of PVA, nanocellulose, citric acid, and orange peel waste extract

Sample code	PVA 10% (mL)	Nanocellulose 1.5% (mL)	Citric acid (g)	Orange peel waste extract (g)	Glycerin (mL)	Total volume (mL)
PVA/NC/Orange peel waste extract	8	2	-	0.2	1	11
PVA/NC/citric acid	8	2	0.2	-	1	11
PVA/NC	8	2	-	-	1	11

cut into 2.2 × 0.5 cm of width sizes, each side of the bioplastic was made wider. The wider part was placed into the grip part of the tensile strength tools. Then, the tensile strength tools stretched the bioplastic. The time began when the bioplastic was stretched, and the time ended when the sample was broken.

Bioplastic degradation test. A degradation test of manufactured bioplastic was performed following the procedure of soil burial test [17]. The bioplastic was cut into 2 × 2 cm^2 sizes and weighed. Then, it was buried in the soil at a 5 cm depth. The soil was watered every 3–4 days for 15 days to keep the humidity of the soil. Then, the bioplastic was taken from the soil, washed, and dried in an oven at 50 °C until dry. Then, it was weighted until a constant value resulted. The percentage of degradation was calculated following Eq. (2):

$$\text{Degradation (\%)} = \frac{\text{Mi} - \text{Ma}}{\text{Mi}} \times 100\% \quad (2)$$

while Mi was the initial mass of the bioplastic before the degradation test, and Ma was the final mass of the bioplastic after the biodegradation test.

RESULTS AND DISCUSSION

Cellulose and Nanocellulose Pinewood

The extraction of cellulose from pinewood waste was done by the acid pre-treatment and alkaline process. The acid pre-treatment process provides 97.16% of dark brown powder. This step eliminates hemicellulose and non-cellulosic components such as wax, chlorophyll, and other acidic soluble compounds. Additionally, it is also used to increase the surface area of cellulose [40]. Meanwhile, the alkaline process is the step to delignification, i.e., removing lignin from cellulose. The delignification process uses sodium hydroxide and hydrogen peroxide as reagents to accelerate the breaking down of the bonds between lignin and cellulose [25]. The yield from this step was 61.86%, and white powder was isolated (Fig. 1). Fig. 1(a) and 1(b) show the appearance of pine wood waste before and after turning. An acid pre-treatment process was carried out by reacting pinewood powder with sodium hypochlorite and acetic acid. This step changes the color of pinewood powder to dark brown (Fig. 1(c)). The delignification process was completed by stirring the pinewood resulting from acid pre-treatment with sodium hydroxide and hydrogen peroxide. This process produces white cellulose (Fig. 1(d)). Further acid hydrolysis of cellulose with hydrochloric acid 10% assisted by ultrasonication provide a white nanocellulose. It has a softer form (Fig. 1(e)). Previous research stated that nanocrystalline cellulose could be obtained by an acid hydrolysis process using hydrochloric acid [28]. Cheng et al. stated that after 30 min of ultrasonication for nanocellulose extraction, the cellulose particles break into smaller fibrils and produce fibrils with a dimension of 100 nm [41]. Nanocellulose was reported to have functional properties such as high degradability, low toxicity, biocompatibility, and excellent mechanical strength. It was also easily adapted into polymeric matrices and blended easily, thus producing cost-effective and durable materials [32,42]. A further study stated that

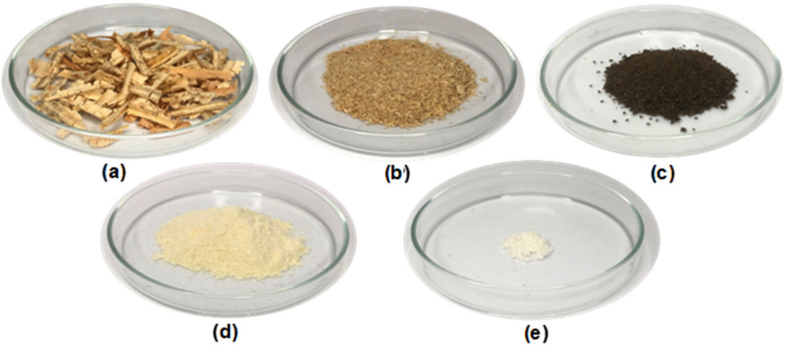

Fig 1. Pictures: (a) Pinewood waste, (b) Pinewood waste powder, (c) Acid pre-treatment result, (d) Cellulose, (e) Nanocellulose

nanocellulose could be used as a reinforcing filler in bioplastic fabrication and improve the tensile strength and elastic modulus of a bioplastic produced [31].

Fourier Transform Infrared (FTIR) Analysis of Cellulose and Nanocellulose

The FTIR spectra of pinewood, cellulose, and nanocellulose were measured to examine the changes in the functional groups composed in the sample. The FTIR result of pinewood waste, cellulose, and nanocellulose was explained in Table 2. From Table 2, there was the absorption of O–H stretching at a wavenumber of 3387.25, 3398.66, and 3345.89 cm^{-1} in pinewood waste, cellulose, and nanocellulose. Then, the absorption of C–H stretching vibration was found at 2898.06, 2900.91, and 2899.49 cm^{-1}. However, the absorption of C=O stretching and vibration of C=C were only found in pinewood waste at a wavenumber of 1732.85 and 1510.36 cm^{-1}, which were assigned as hemicellulose and lignin. Similar findings were also reported by Popescu et al. and Sukmawan et al. that C=O and C=C groups assigned as hemicellulose and lignin could be found at a wavenumber at 1700 and 1510 cm^{-1} [43-44].

X-Ray Diffraction (XRD) Analysis of Cellulose and Nanocellulose

X-Ray Diffraction (XRD) is a technique used to assess the degree of crystallinity in various materials. Among the wood components, only cellulose has a crystallinity phase, and the other polymer has a non-crystalline phase [43]. The X-Ray diffractogram is explained in Fig. 2. The XRD analysis of cellulose and nanocellulose was recorded at $2\theta = 22.5°$ and the peaks for cellulose and nanocellulose characteristics with grid plane 200 [45].

Table 3 describes the crystallinity index of cellulose and nanocellulose, resulting in a crystallinity index of 58.49% for cellulose and 60.32% for nanocellulose at 2θ degrees of 22.59° and 22.62°. The crystallinity of nanocellulose is higher than cellulose, which implies that the acid hydrolysis process on cellulose reduces the amorphous phase of cellulose and increases the crystalline phase [46].

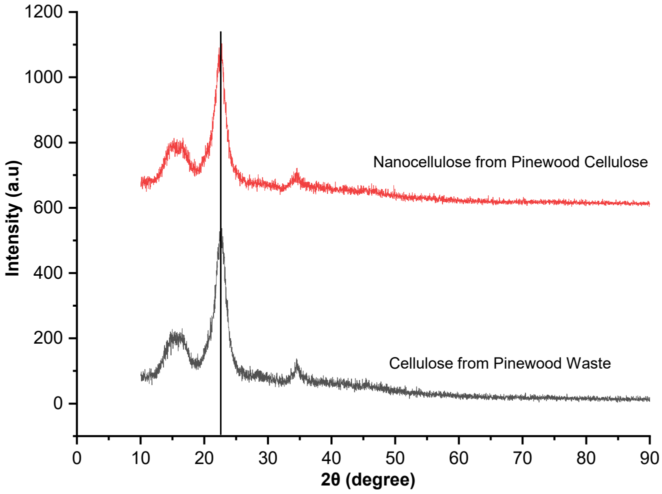

Fig 2. X-Ray diffractograms pattern of cellulose and nanocellulose from pinewood waste

Table 2. The FTIR result of pinewood waste, cellulose, and nanocellulose

Wavenumber (cm^{-1})			Functional group
Pinewood	Cellulose	Nanocellulose	
3387.25	3398.66	3345.89	O–H stretching
2898.06	2900.91	2899.49	C–H stretching
1732.85	-	-	Hemicellulose, C=O stretching
1640.14	1644.42	1647.27	Water absorption, O–H bonding
1510.36	-	-	Lignin, C=C
-	1427.64	1429.06	CH_2 bending from carboxylate
1374.87	1372.02	1372.02	C–O–C cellulose amorph and crystalline
1317.82	1317.82	1319.25	Cellulose, C–O–C
1059.68	1062.53	1062.53	C–O stretching in glycoside bond
897.09	898.51	898.51	C–O stretching

Table 3. Crystallinity index of cellulose and nanocellulose

	Cellulose	Nanocellulose
Peak (2θ)	22.59°	22.62°
Crystallinity index (%)	58.49	60.32

Scanning Electron Microscope (SEM) of Cellulose and Nanocellulose

SEM is an important instrument for determining the morphological behavior of the bioplastic surface [17]. The surface morphological images of cellulose and nanocellulose are shown in Fig. 3. The result indicates that both cellulose and nanocellulose have a crystalline shape. Both are different in size. The diameter and length of cellulose and nanocellulose were calculated with ImageJ software [47]. The cellulose has a larger size than the nanocellulose (Fig. 3). The diameter and length of cellulose were 101.65 and 173.21 nm (Table 4), respectively. Meanwhile, nanocellulose has a diameter and a length of 16.36 and 29.44 nm, respectively. The breakdown process of a polymeric chain of cellulose by acidic hydrolysis combined with an ultrasonication strategy forms the cellulose into a smaller size. Some research also reported an increase in the surface area of cellulose [47-48].

Bioplastic Film's Appearance

The bioplastic made from orange peel waste extract had a slightly yellow color, while the bioplastic made with the addition of citric acid gave a transparent and glossy surface (Fig. 4). Meanwhile, the appearance of bioplastic produced without the addition of citric acid or orange peel waste extract has a transparent and doff surface.

Table 4. The particle size of cellulose and nanocellulose

Sample	Dimension	
	Diameter (nm)	Length (nm)
Cellulose	101.65	173.21
Nanocellulose	16.36	29.44

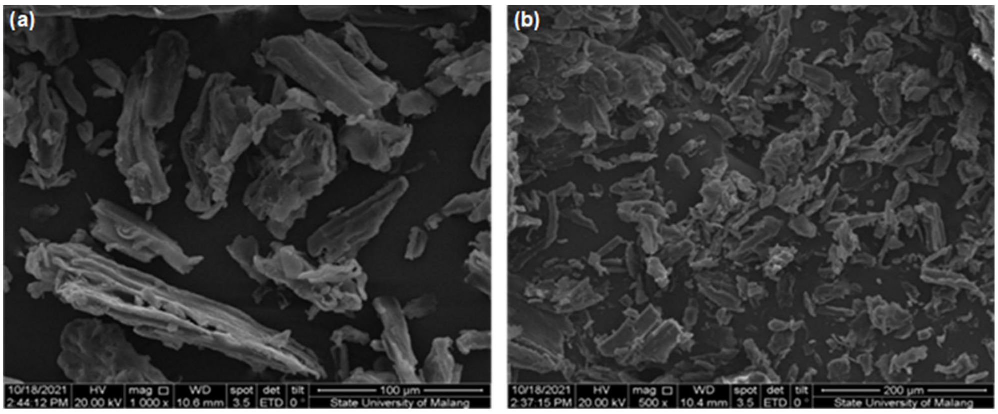

Fig 3. Morphological of (a) Cellulose (1000×), (b) Nanocellulose (500×)

PVA/NC/Orange peel extract PVA/NC/Citric Acid PVA/NC

Fig 4. Degradable bioplastics

Fourier Transform Infrared (FTIR) Analysis of Bioplastics

The result of FTIR analysis shows that stretching vibration of the hydroxyl (O–H) group of bioplastics with the addition of citric acid and orange peel waste extract gives absorption band at 3274.58 and 3271.73 cm^{-1}, whereas bioplastic without the addition of both has a hydroxyl group (O–H) stretching band at 3271.73 cm^{-1}. Meanwhile, the bending vibration H–O–CO composed of bioplastic prepared with the addition of citric acid and orange peel waste extract appears at 1650.13 and 1645.85 cm^{-1}, respectively. The shift of bending vibration to a lower value implies that bioplastic with citric acid and orange peel waste extract has a new atmosphere. And it is predicted that this shifting (from 1651.55 to 1650.13 cm^{-1} and 1645.85 cm^{-1}) indicates a hydrogen bond interaction between functional groups in each component [49]. Furthermore, the C–O group stretching vibration in C–O–H from all bioplastics showed a peak of 1036.86 cm^{-1}.

The only peak that comes from C=O stretching vibration (1712.88 cm^{-1}) was given from bioplastic with the addition of citric acid. It indicates a free carbonyl group in the bioplastic does not interact (Fig. 5). Additionally, bioplastic without the addition of citric acid and orange peel extract changes the intensity of hydroxyl group stretching vibration. It is detected at wave number 3271.73 cm^{-1}. Besides, C–H stretching vibration from the alkyl group was recorded at 2939.42 cm^{-1}. This finding has similar results to Hussein et al. and Popescu et al. [38,43]. The illustration of the interaction between nanocellulose and citric acid is described in Fig. 6(a), and the illustration reaction between nanocellulose and orange peel extract is described in Fig. 6(b).

Scanning Electron Microscope (SEM) Analysis of Bioplastics

The surface morphology and internal microstructure of the films are revealed by SEM analysis [50]. Fig. 7(a) shows the smooth surfaces of bioplastic with citric acid. However, there is a part of nanocellulose that does not blend perfectly. While in Fig. 7(b), the surfaces of bioplastic with the addition of orange peel extract seem smoother than those using citric acid, and the cross-section SEM analysis (a1) looks more pore than the bioplastic with the addition of orange peel extract (b1). It is proven by another paper report that the

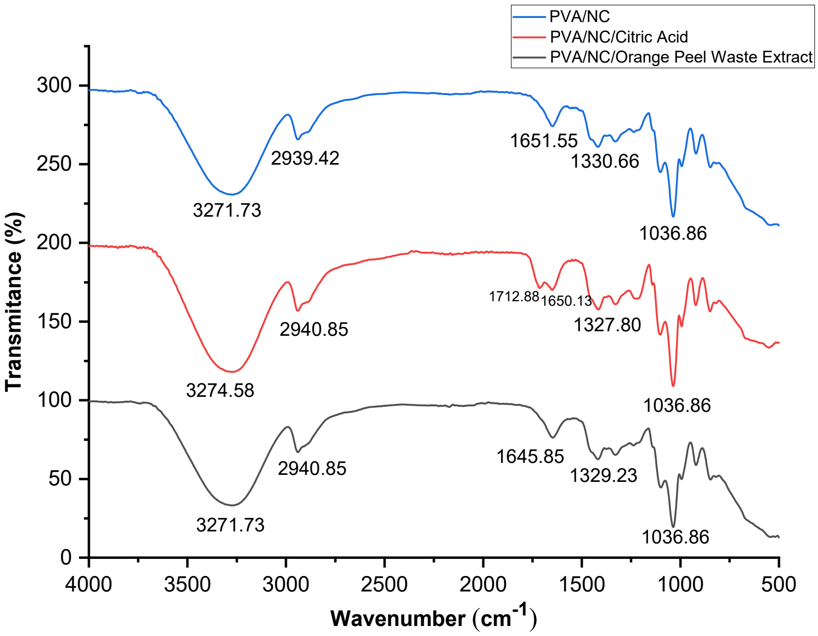

Fig 5. FTIR spectra of biodegradable plastics pinewood nanocellulose based, bioplastic without citric acid and orange peel waste extract (blue line), bioplastic with citric acid (red line), bioplastic with orange peel waste extract (black line)

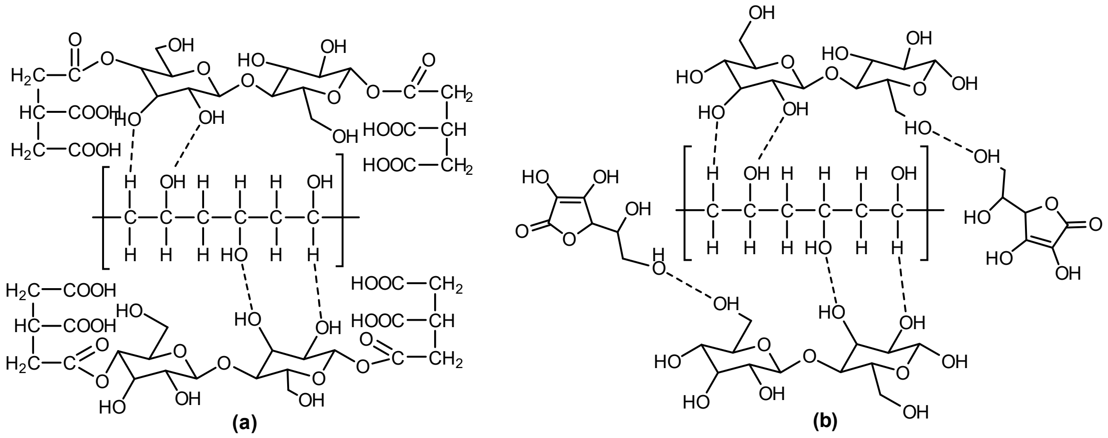

Fig 6. The illustration reaction of bioplastic pinewood nanocellulose based using citric acid (a) and orange peel waste extract (b)

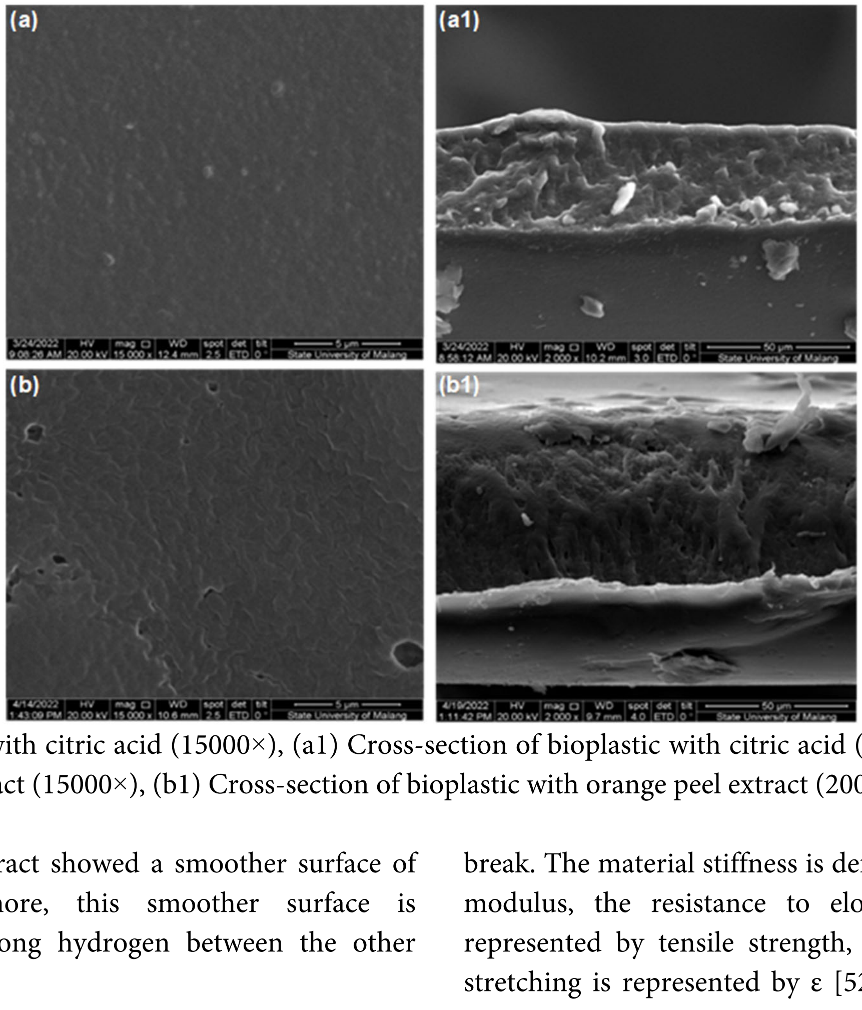

Fig 7. (a) Bioplastic with citric acid (15000×), (a1) Cross-section of bioplastic with citric acid (2000×), (b) Bioplastic with orange peel extract (15000×), (b1) Cross-section of bioplastic with orange peel extract (2000×)

addition of plant extract showed a smoother surface of bioplastic. Furthermore, this smoother surface is indicated by the strong hydrogen between the other components [17,51].

Mechanical Properties Analysis of Bioplastics

The mechanical properties studied were elastic modulus, tensile strength, and percentage elongation at break. The material stiffness is defined by EM or elastic modulus, the resistance to elongation at break is represented by tensile strength, and the capacity for stretching is represented by ε [52]. From Table 5, the elastic modulus of bioplastic without the addition of citric acid and orange peel waste extract is lower than bioplastic that fabricates with citric acid and orange peel waste extract. The tensile strength and elastic modulus of

Table 5. Elastic modulus (EM), tensile strength (TS), percentage of elongation at break (EB) of bioplastic

Samples	EM (kPa)	TS (kPa)	%ε
PVA/NC/Orange peel waste extract	42.71	1708.54	40.00
PVA/NC/Citric acid	36.12	1203.85	33.33
PVA/NC	31.71	317.11	10.00

bioplastic with the addition of orange peel waste extract have the highest value at 1708.54 and 40.86 kPa. The elongation at break of bioplastics with the addition of citric acid and orange peel extract was 33.33% and 42.71%, respectively. It proved that orange peel extract makes bioplastic stiffer than when it uses citric acid. A similar finding was reported by Marsi et al. that the addition of orange peel powder increases the mechanical properties of bioplastic [15]. Another report on the use of plant extract also increases the mechanical properties and provides a smooth surface morphology of bioplastic. It is due to the amount of hydrogen bonding interaction between the components [17].

Fig. 8 shows the tensile strength curve of the bioplastic with and without the addition of citric acid and orange peel waste extract. From the tensile strength curve, the fabrication of bioplastics without the addition of citric acid and orange peel waste extract had the lowest tensile strength values and the lowest breaking time for the resulting bioplastics. Furthermore, the fabrication of bioplastics with the addition of orange peel waste extract has a higher tensile strength value and the highest breaking time.

Degradability Test Result of Bioplastics

The degradability studies of bioplastics were obtained from the biodegradability test using the soil burial method. The soil-buried bioplastics were collected at 15 days to measure the degradation. After the bioplastic was taken out from the soil, bioplastics were washed with water to remove the soil residue and dried in an oven at 50 °C for 2 h. The results showed a weight loss of the bioplastics in 15 days. Fig. 9 shows the biodegradability analysis of bioplastic without citric acid and orange peel waste extract, bioplastic with citric acid, and bioplastic with orange peel waste extract, and it shows bioplastic can degrade well in soil. From Table 6, the degradation result of bioplastic without citric acid and orange peel waste extract had the highest result at 86.27%. Furthermore, the degradation result of bioplastic with citric acid at 77.27%, and the degradation result of bioplastic with orange peel waste extract at 78.44%.

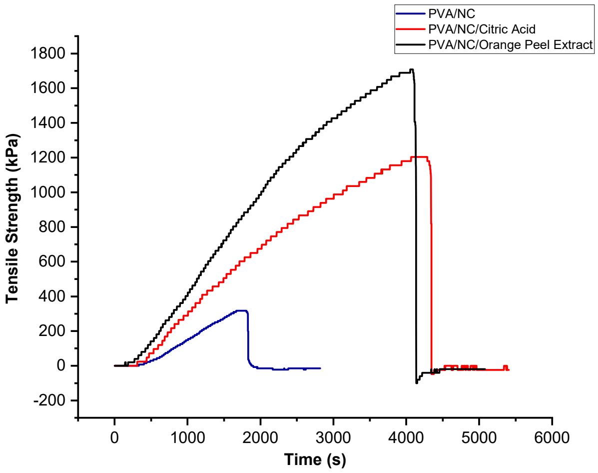

Fig 8. Tensile strength curves of bioplastic without the addition of citric acid and orange peel waste extract (blue line), bioplastic with citric acid (red line), and bioplastic with orange peel waste extract (black line)

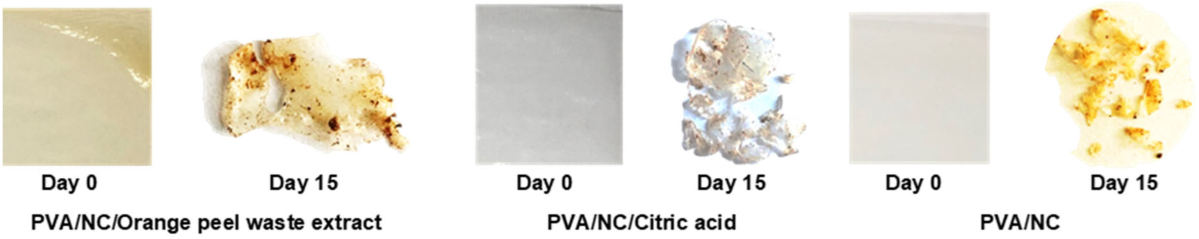

Fig 9. Biodegradation analysis on days 0 and 15

Table 6. Degradation percentage of bioplastic

Sample	% Degradation after 15 days
PVA/NC/Orange peel waste extract	78.44
PVA/NC/Citric acid	77.27
PVA/NC	86.27

The results of the degradation analysis of bioplastics without the addition of citric acid and orange peel waste extract had the highest degradation results, while bioplastics without the addition of citric acid and orange peel waste extract had a large rate of degradation analysis of the PVA and nanocellulose by the water is due to the leaching process of PVA and nanocellulose by the water. It caused the hydroxyl groups in PVA to have higher water sorption and solubility in water [49]. Furthermore, the bioplastic with the addition of citric acid and extract had a slower degradability result due to the antimicrobial effect of citric acid and extract [53]. The results of the analysis of the degradation of bioplastics with citric acid and orange peel extract had a lower value than the degradation analysis of bioplastics without the addition of citric acid and orange peel waste extract. The degradation of bioplastic with orange peel extract had a higher result than the bioplastic with citric acid. It is because the orange peel waste extract act as a biodegradation component that can give a higher oxygen permeability to microorganism [15].

■ CONCLUSION

Pinewood waste has an important application. Cellulose and nanocellulose can be extracted from it and used as an environmentally friendly bioplastic material. In this study, pinewood waste was bleached using the alkaline bleaching method to obtain cellulose, and nanocellulose was isolated after hydrolysis assisted by ultrasonication. Nanocellulose is used as a filler material with PVA to produce bioplastics. The integration of PVA, nanocellulose, and orange peel waste extract increases the tensile strength, modulus of elasticity, and elongation at the break of bioplastics compared to bioplastics without the addition of citric acid and orange peel waste extract. Tensile strength, modulus of elasticity, and elongation at break prove that bioplastic with the addition of orange peel extract has the highest performance. The addition of orange peel waste extract also creates the degradability of bioplastics slower than that without the addition of it.

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