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Utilization of Nanocellulose from Red Onion Skins as Nanofiller in Polyvinyl Alcohol-Based Film

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Abstract

Research has been conducted to apply nanocellulose from red onion skins as a nanofiller on polyvinyl alcohol (PVA) based films. It aims to determine the effect of red onion skin nanocellulose on PVA-based films in improving mechanical and biodegradation properties. Nanocellulose was produced from the skin of red onions through an acid hydrolysis process with a temperature of 45°C for 40 minutes. The resulting nanocellulose was characterized using FTIR, XRD, and SEM. Nanocellulose, with concentrations of 10%, 20%, 30%, 40%, and 50%, was then used as a filler in PVA-based films. The results were characterized to determine the mechanical and biodegradation properties. The nanocellulose obtained from the red onion skins in this study was 12.615 nm with a crystallinity index of 78.668%. The optimum tensile strength was achieved at 14.7573 MPa in PVA-based film filled with 20% nanocellulose extracted from red onion skins. The greatest elongation percentage of 118.3265% was observed in a PVA-based film containing 50% nanocellulose from red onion skins. Moreover, optimal biodegradation occurred in PVA films incorporating 50% nanocellulose from red onion skins, resulting in a weight loss of 12.86% over 14 days.

1. Introduction

Nano-sized material has become one of the foundations for developing science and technology. One of the nanomaterials that is being widely studied is nanocellulose. Nanocellulose is a new type of nanomaterial with a 2-20 nm diameter and reaches hundreds to thousands of nanometers in length [1]. These nanomaterials can be produced using various techniques, specifically acid hydrolysis, mechanical processes, and enzymatic methods [2]. The sources of nanocellulose are various natural fibers from plants or marine animals [3].

Natural fibers from plants that have not been widely used and can be a source of nanocellulose are the skin of red onions. The foodstuff waste material had α-cellulose content of 41-50% in a dry state [4]. The cellulose from red onion skins can be transformed into nano-sized particles. This certainly causes changes in the cellulose's characteristics, such as increased surface area, crystallinity and dispersion, and enhanced biodegradability [2]. This improved biodegradation property makes nanocellulose a valuable filler for bioplastic materials [2].

The addition of nanocellulose fillers proves useful in producing environmentally friendly and biodegradable plastics, particularly as synthetic plastics continue to rise in tandem with the growing global population. Presently, global plastic production stands at an estimated 100 million tons per year, relying on petroleum-derived raw materials, which are finite in supply. Consequently, the resulting plastic products pose significant challenges for natural degradation. Moreover, synthetic plastics in food packaging may contribute to various health issues, including disruptions to hormonal and reproductive systems and the potential for cancer triggers, owing to the transfer of plastic monomers into packaged food items. Thus, solutions must be found to make plastic packaging safe for health and the environment [5]. One good alternative solution is to develop packaging using

bioplastics that are biodegradable and environmentally friendly.

The polymer that is often used as a bioplastic material is polyvinyl alcohol (PVA). This material is a synthetic polymer with odorlessness, tastelessness, non-toxicity, and biodegradability properties. Additionally, PVA exhibits insolubility in oil or fat, along with high tensile strength and flexibility, causing PVA to be widely used as a promising alternative packaging material. However, the tensile strength and flexibility of PVA are affected by humidity. As humidity increases, PVA tends to absorb more water, leading to a reduction in its mechanical properties. An alternative solution to the PVA problem is adding nanocellulose fillers into the materials [6]. This research utilizes nanocellulose from red onion skins as a nanofiller in PVA-based bioplastic films. The addition of nanocellulose from red onion skins to PVA-based films is useful to improve mechanical properties and degradation time in the soil.

2. Experimental

In this study, nanocellulose was synthesized using the acid hydrolysis method. This method was chosen because it is easy and simple to form nano-sized cellulose. Meanwhile, PVA film was produced using the solvent casting method with the principle of gelatinization. This method ensures the production of films with uniform thickness and high optical clarity.

2.1. Instrument and Materials

The instruments used in this study were 15 mL centrifuge tubes, volume pipettes, hot plates and magnetic stirrers, Petri dishes, centrifuges, measuring flasks, desiccators, electric stoves, pH paper, analytical balance, glassware, scanning electron microscopy (SEM) (Inspect-S50), X-ray diffraction (XRD) (E'xpert pro), Fourier transform infra-red (FTIR), and universal testing machine. The research materials included red onion skins, distilled water, 96% ethanol, 50% sulfuric acid, 24% hydrogen peroxide, 4% sodium hydroxide, 5% sulfuric acid, filter paper, and PVA crystals.

2.2. Preparation of Cellulose from Red Onion Skins

Approximately 20 grams of red onion skins were prepared, followed by adding ethanol and distilled water in a 1:1 ratio, totaling 100 mL. The solution was heated for 2 hours, and the process was repeated twice. Subsequently, it was drained and added with 200 mL of 4% NaOH, stirred with a magnetic stirrer at 60°C for 2 hours. The solution was washed with warm distilled water until neutral and separated between the filtrate and the residue. The residue was added with 32 mL of 24% H_2O_2 and 128 mL of 4% NaOH and stirred with a magnetic stirrer for 1.5 hours at 60°C. The extract was filtered to separate the residue and the filtrate. Following this, the residue was washed until reaching a neutral pH.

2.3. Nanocellulose Preparation

The cellulose derived from red onion skins was subjected to acid hydrolysis by adding 50% H_2SO_4 at a ratio of cellulose to H_2SO_4 of 1:5. This process was conducted at 45°C and stirred with a magnetic stirrer for 40 minutes. Subsequently, the obtained product was added with 100 mL of cold water and centrifuged at a speed of 5000 rpm for 15 minutes until reaching a neutral pH. The obtained nanocellulose samples were characterized using XRD, FTIR, and SEM.

2.4. Formation of PVA-based Bioplastic Films with the Addition of Nanocellulose Fillers

Each 1.5 gram of PVA crystal was dissolved with 15 mL of distilled water for 2 hours at 80°C. After cooling, each solution was added with nanocellulose in concentrations of 10%, 20%, 30%, 40%, and 50% relative to the dry weight of PVA. The mixtures were stirred using a magnetic stirrer at 40°C for 1 hour. Subsequently, they were printed onto a 90 mm diameter petri dish and roasted for 5 hours at 65°C. The resulting films were characterized using SEM and universal testing machine, and followed by a biodegradation test.

3. Results and Discussion

3.1. Characterization of Nanocellulose Produced from Red Onion Skins

Nanocellulose was produced through acid hydrolysis at 45°C for 40 minutes. This process selectively destroys the amorphous component of cellulose, leaving behind the crystalline portion, as illustrated in Figure 1. A 50% concentration of sulfuric acid was utilized for the hydrolysis process because of the presence of a sulfur group, which facilitates easy dispersion in water compared to other strong acids [7]. A hydrolysis time of 40 minutes was employed. Exceeding this time limit during hydrolysis can result in the breakdown of cellulose into monomeric components.

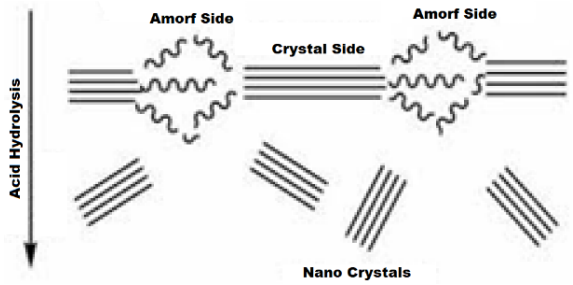

Figure 1. Mechanism of acid hydrolysis [7]

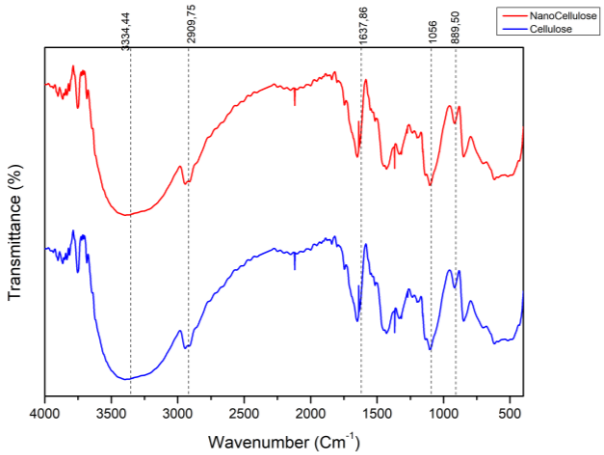

Figure 2. FTIR spectra of (a) cellulose and (b) nanocellulose

Characterization of functional groups to ensure that the resulting nanocellulose from the red onion skins was done using FTIR. The spectroscopic results are presented in Figure 2. In the FTIR spectrum of cellulose, a prominent peak at the wavenumber 3344.44 cm^{-1} was observed, corresponding to the -OH stretch region (3300-3400 cm^{-1}). A peak at wavenumber 2909.75 cm^{-1} was also identified, associated with the -CH vibration of cellulose, hemicellulose, and lignin [8].

Another notable peak at wavenumber 1637.86 cm^{-1} indicated the water absorption region and C-H deformation of cellulose and lignin [9, 10]. The regions around 1056 cm^{-1}, denoting C-O and C- H, affirmed the cellulose structure. The presence of a peak in the 850-900 cm^{-1} range indicated β-glycosidic bonds. Notably, the FTIR results for nanocellulose did not exhibit significant deviations from those of cellulose. Treatment with sulfuric acid was found to preserve the functional groups of cellulose, inducing only a discontinuity in the glucose ring and variations in absorption intensity [11].

XRD characterization aims to determine the crystallinity index and the crystal size of the nanocellulose. The diffractogram of nanocellulose from red onion skins is shown in Figure 3. The peaks produced by nanocellulose at 2θ are 16.5786 and 22.6298. The crystallinity index was calculated using the Segal method (Equation 1) [12].

$$Crystallinity\ index = \frac{I_{crystalline} - I_{amorf}}{I_{crystalline}} \times 100\% \quad (1)$$

$$Crystal\ size = \frac{k\ .\lambda}{\beta\ .\cos\theta} \quad (2)$$

The size of the nanocellulose crystals is calculated using the Scherrer equation (Equation 2) [13]. The crystallinity index obtained from cellulose acid hydrolysis on red onion skins is 78.668%. The size of the crystalline nanocellulose obtained is 12.615 nm.

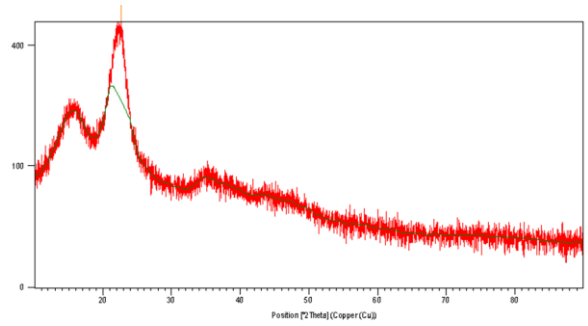

Figure 3. XRD diffractogram of nanocellulose from red onion skins

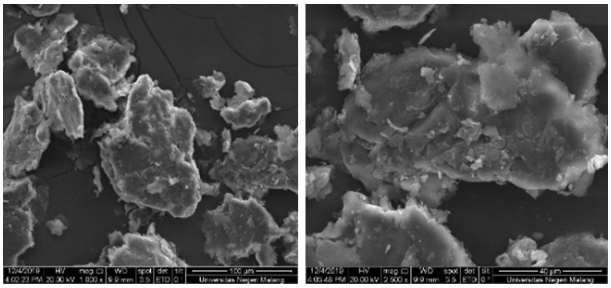

Figure 4. Morphology of nanocellulose from red onion skins

The SEM analysis, depicted in Figure 4, illustrates the morphology of nanocellulose derived from red onion skins. The micro-scale presentation showcases the presumed morphological size of the nanocellulose, influenced by the physical treatment, specifically the freeze-drying process. During freeze-drying, the nanocellulose paste undergoes rapid cooling, leading to the sudden formation of water-containing lumps resembling crystals. These lumps aggregate and manifest morphologically in micro-sized sections [14].

3.2. Physical and Chemical Characteristics of PVA-based Films with the Addition of Nanocellulose Filler from Red Onion Skins

Nanocellulose is added as a filler in PVA-based films to improve the mechanical properties, specifically tensile strength and elongation of the film. PVA, a water-soluble polymer due to its prevalent hydroxyl group, is highly susceptible to moisture absorption, reducing its mechanical properties [15]. Therefore, additional materials are needed as fillers to improve the properties of PVA-based films. Nanocellulose is very well used as a filler to improve the mechanical properties of films because nanocellulose has a high enough stiffness and a good dispersal ability in a polymer [16]. The ability of nanocellulose to enhance these properties depends on the source, size, and percentage of addition.

PVA-based films produced by adding nanocellulose fillers from red onion skins are shown in Figure 5. Mechanical testing of PVA-based films with nanocellulose fillers includes tensile strength and percent elongation using a Universal Testing Machine that refers to the ASTM-D638 standard. Tensile strength is the maximum pull achieved by a material before breaking up. Tensile strength values can be calculated based on Equation (3) [17].

$$Tensile\ strength = \frac{P_{max}}{A} \quad (3)$$

Where, P_{max} is the maximum force in Newton units and A cross-sectional area in m^2.

Tensile strength measurements were carried out twice for each percent addition of nanocellulose to obtain the average tensile strength results. The tensile strength results of PVA-based films with the addition of nanocellulose fillers are shown in Figure 6.

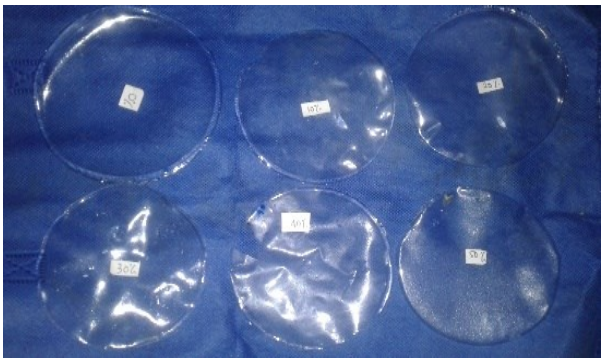

Figure 5. The PVA-based film with the addition of nanocellulose fillers from red onion skins

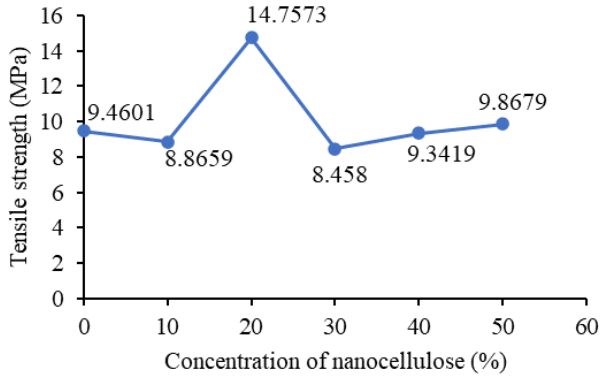

Figure 6. The graph between tensile strength and the addition of nanocellulose filler concentration

PVA-based films without the addition of nanocellulose fillers have a tensile strength value of 9.4601 MPa. The tensile strength fluctuates with the addition of concentrations of nanocellulose from the skin of red onions. At the addition of a nanocellulose concentration of 20%, the tensile strength value is at the optimum condition of 14.7573 MPa. This is included in the standard film category with good tensile strength, which has a tensile strength of 10-100 Mpa [18]. The decrease in tensile strength value with the addition of higher filler concentration is attributed to agglomeration and aggregation between the filler (nanocellulose from red onion skin) and the matrix (PVA film). These phenomena result in strengthened bonds between fillers and weakened interactions between the filler and the matrix [19].

Percent elongation is an extension of the material before breaking up. The higher the elongation percent, the more elastic the material. The material becomes stretchable [20]. Percent elongation is calculated using Equation (4) [17].

$$Percent\ Elongation = \frac{L_2 - L_1}{L_1} \times 100\% \qquad (4)$$

Where L_1 is the length of the film before being drawn (m), and L_2 is the length of the film after being drawn (m). The graph of the percent elongation value of the film against the concentration of fillers in the form of nanocellulose from the skin of red onions is shown in Figure 7.

The highest elongation percentage of 118.3265% is indicated by the addition of 50% nanocellulose. This value aligns with the standards established by SNI [21], falling within the range of 21-220%. A higher elongation percentage within this standard range indicates improved film quality, characterized by increased resistance to tearing. The enhanced percent elongation is attributed to the biocompatibility of nanocellulose, fostering the formation of hydrogen bonds between the matrix and filler. The resulting hydrogen bonding strengthens the PVA film, facilitating smoother stretching and increased elongation.

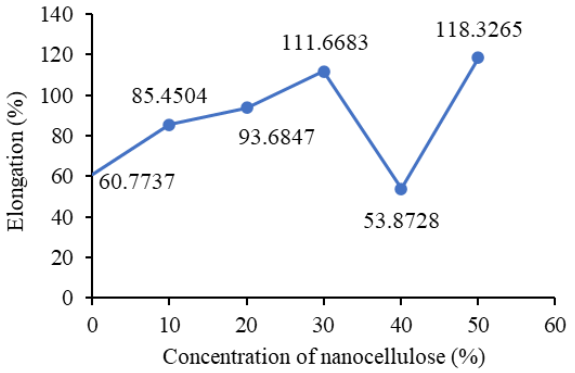

Figure 7. The graph between elongation and addition of nanocellulose concentration

The morphological properties of a PVA-based film with a 20% nanocellulose filler were analyzed using SEM, and the results are depicted in Figure 8. As observed in Figure 8, the addition of nanocellulose effectively conceals cavities within the formed film. This can be attributed to the small size of nanocellulose, contributing to a larger surface area. Figure 8 illustrates agglomeration, increasing interface adhesion force between the filler and the matrix. This enhanced force facilitates the formation of hydrogen bonds within the material.

The purpose of conducting functional group analysis using FTIR on PVA-based films with the incorporation of nanocellulose is to confirm the successful mixture of PVA and nanocellulose in the film. The spectra resulting from FTIR analysis are depicted in Figure 9. Figure 9 indicates that the observed mixing was physical, as no new functional groups were identified in the FTIR analysis results. The variations observed in absorption intensity in PVA-based films are attributed to the combination of PVA and nanocellulose. This aligns with the findings of Kharazmi *et al.* [22], who identified PVA functional groups using FTIR, revealing peaks at 3280, 2917, 1690, 1425, 1324, 1081, and 839 cm^{-1}. These peaks correspond to the O-H stretching vibration of the hydroxy group, CH_2 asymmetric stretching vibration, C=O carbonyl stretching, C-H stretching on CH_2 vibration, and C-H deformation vibration.

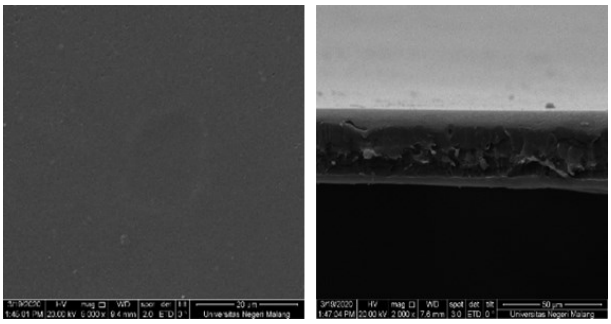

Figure 8. Morphology of PVA-based films with the addition of nanocellulose from red onion skins

Table 1. The results of the measurement of percent weight loss from PVA-based films with the addition of nanocellulose from red onion skins

Filler (%)	Initial weight (g)	Day 7th (g)	Weight loss (%)	Day 14th (g)	Weight loss (%)
0	0.15	0.091	5.9	0.0865	0.45
10	0.15	0.088	6.2	0.0745	1.35
20	0.15	0.0701	7.99	0.0596	1.05
30	0.15	0.0657	8.43	0.0465	1.92
40	0.15	0.0478	10.22	0.03376	1.404
50	0.15	0.036	11.4	0.0214	1.46

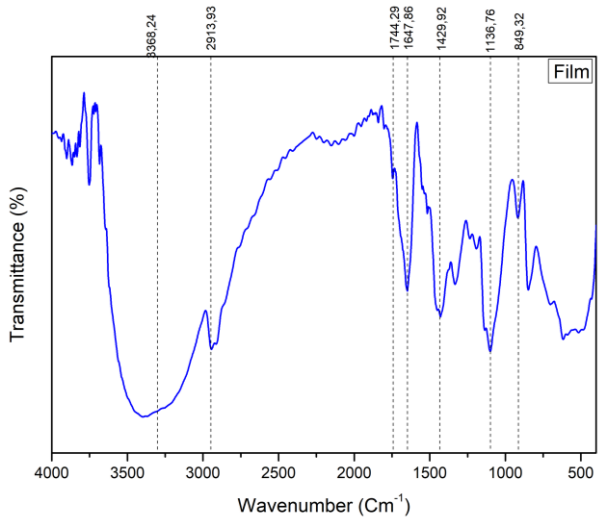

Figure 9. FTIR spectra of PVA-based films with the addition of nanocellulose

In Figure 9, the functional group identification results for the PVA-nanocellulose film reveal an absorption peak at 3368.24 cm^{-1}, indicating the presence of -OH stretching in both nanocellulose and PVA [23]. The absorption at 2913.93 cm^{-1} corresponds to the stretching of C-H bonds. The bending vibration of C-O in the PVA-nanocellulose film is evident at 1744.29 cm^{-1}. An absorption peak at 1647.86 cm^{-1} indicates a weak strain of C=O, while 1429.92 cm^{-1} indicates the flexural CH_2 vibration. The absorption at 1136.76 cm^{-1} represents the stretching of the C-O-C pyranose ring, and absorption at 849.38 cm^{-1} confirms the presence of the glycosidic β-1,4 bond [24]. The observed shift and widening of wavelengths in PVA-nanocellulose are attributed to the contribution of C-O-C and C-O bonds [19], suggesting potential hydrogen bond formation between PVA and nanocellulose molecules [25].

The measurement of biodegradation in PVA-based films with the addition of nanocellulose from red onion skins involved employing a burial technique in the soil at a depth of 30 cm for 14 days. The utilized sample measured 4×1 cm^2 with 0.15 grams in weight. During burial, the sample is intentionally exposed to air and sunlight. The choice of soil as the medium aligns with its relevance as a disposal environment for plastic waste. Additionally, the exposure to air and sunlight is deliberate, considering that films derived from cellulose are susceptible to physicochemical degradation [26]. The measurement of percent weight loss is done by weighing the sample every seven days. The calculation of percent weight loss is determined using Equation (5) [27]. The measurement results of percent weight loss are shown in Table 1.

$$Percent\ lost\ weight = \frac{w_1 - w_2}{w_1} \times 100\% \qquad (5)$$

Where, W1 is the film weight before the biodegradation test, and W2 is the film weight after the biodegradation test.

According to Table 1, the degradation process is most rapid in PVA-based films with the addition of 50% nanocellulose, resulting in an accumulated weight loss percentage of 12.86%. This accelerated degradation can be attributed to physicochemical factors and is further influenced by the inherent biodegradability property of nanocellulose [28]. The biodegradability of PVA-based films with the addition of nanocellulose from red onion skins is influenced by the -OH group on the nanocellulose. PVA and nanocellulose each have a -OH group, which initiates the hydrolysis reaction after absorbing water from the soil [27]. The water absorption from the soil leads to the degradation or deterioration of nanocellulose polymers due to the breaking of bonds between functional groups in the polymer [29] and the decrease in molecular weight [30]. The bonding process causes the PVA-based film to gradually break down into smaller parts in the soil.

4. Conclusion

Nanocellulose produced from red onion skins using acid hydrolysis exhibited a crystal size of 12.615 nm and a crystallinity index of 78.668%. In PVA-based films, the highest tensile strength was achieved by incorporating a 20% nanocellulose filler, resulting in a value of 14.7573 MPa. Additionally, the optimal elongation percentage of the film was observed when adding a 50% nanocellulose filler, with a value of 118.3265%. The film's optimum biodegradation occurred with the addition of a 50% nanocellulose filler, leading to a value of 12.86%.

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