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Research Article

Investigation on Reinforcement Effects of Nanocellulose on the Mechanical Properties, Water Absorption Capacity, Biodegradability, Optical Properties, and Thermal Stability of a Polyvinyl Alcohol Nanocomposite Film

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This paper presents the reinforcement of nanocellulose (NC) in polyvinyl alcohol (PVA) to examine the effect of the amount of reinforcement on the properties of PVA. The nanocellulose was successfully extracted by sulfuric acid hydrolysis method and ultrasonication, and successively reinforced with polyvinyl alcohol by the solvent-casting method. After incorporating nanocellulose into the PVA matrix, the effect of nanocellulose on the tensile strength, elongation at break, water absorption capacity, transmittance, thermal stability, and biodegradability of PVA was investigated. The tensile strength increased from 24.5 ± 0.53 MPa to 35.5 ± 0.55 MPa and 40.6 ± 0.73 MPa with the addition of 2%NC and 5%NC, respectively. The elongation at break increased from $40 \pm 0.53\%$ to $45.7 \pm 0.53\%$ with 2%NC, and after the reinforcement of 7%NC, it decreased to $32.2 \pm 0.75\%$. The water absorption capacity result reveals that neat PVA absorbs the highest amount of water which is $84.6 \pm 0.56\%$ and is reduced to $73 \pm 0.78\%$ by adding 2%NC. By increasing the nanocellulose loading to 7%, the water intake capacity was reduced to $61 \pm 0.59\%$ which illustrates the water intake was reduced linearly with the increment of NC. The ultraviolet-visible (UV-Vis) result implies that the transmittance of neat PVA and PVA-2%NC composite film was 85.4% and 78.2% at 600 nm, respectively, which indicates the decrement in transmittance. The thermogravimetry analysis (TGA) reveals that the thermal stability of polyvinyl alcohol after incorporating nanocellulose particles was reduced. The weight loss of neat PVA is 70.7 ± 1.7% after 90 days while the weight loss of the PVA composite films reinforced with 1%, 3%, 5%, 7%, and 9% was $65 \pm 1.85\%$, $57 \pm 1.57\%$, $55.6 \pm 0.64\%$, $52 \pm 1.73\%$, and $53.1 \pm 1.72\%$, respectively. The scanning electron microscopy micrograph for the PVA-6%NC nanocomposite film reveals a dispersion of nanocellulose in a matrix.

1. Introduction

There have been increased challenges in society when it comes to needs such as food, fuel, energy, and materials. These necessities are commonly reliant on the depletion of fossil resources, which is caused by advances in science and technology that enable mankind to consume more fossil resources [1]. Many materials based on petroleum products have had major environmental consequences, such as loss of natural nonrenewable resources, energy shortages, and global warming [2]. Due to its many applications, plastic materials are one of the segments that are growing fast in the waste stream. As petroleum-based synthetic polymer products like plastics do not decompose upon disposal,

ecological concerns have resulted in interest in a new natural and compostable material to improve biodegradability and environmental safety [3]. To meet the rapid demand for environmental safety and sustainability, many researchers have developed different plastics that can be degraded. But these plastics lack some properties such as good mechanical and water intake properties which limit their daily application in different areas. At present, the production of biocomposite from a renewable source is attracting the attention of industrialists and researchers instead of using petrochemical resources.

Biocomposites are a promising alternative to further improve material properties while maintaining their biodegradability. They are materials made up of two or more different constituents with the properties of the resultant material being superior to the properties of the individual material that made them [4]. They have improved properties such as low density, biodegradablity, and good mechanical properties. It is important to reinforce fillers or other particles in the polymer matrix because most of the plastics by themselves are not suitable for load-bearing applications due to their lack of sufficient strength, dimensional stability, and stiffness. However, fibers retain high stiffness and strength but are difficult to use in load-bearing applications because they have a fibrous structure [3]. In fiber-reinforced composites, the function of the fiber is strengthening by giving stiffness and strength to the structure, and the plastic matrix serves as the adhesive to hold the fibers in place so that appropriate structural constituents can be made [5].

Nanocelluloses are highly crystalline particles with high aspect ratios, have crystalline domains, and can be extracted from renewable cellulosic sources. The acid hydrolysis method is the most well-known and extensively used of the various ways for making nanocellulose. This process is simple and quick for producing nanocellulose with desirable properties. This technique breaks down the cellulose's irregular and amorphous portions, removing single, well-defined crystals [6]. Nanocellulose with small particle size and high crystallinity has better and outstanding mechanical properties. It has many applications in various fields like a barrier in the separation process of hazardous waste, food wrappers which replace the nonbiodegradable plastics, and a nanocomposite to improve properties such as mechanical, thermal, and ionic conductivity properties of the polymer [7]. Teff is the widely cultivated cereal crop in Eritrea and Ethiopia for food, and it is a primary food grain mostly used to make Injera [8]. In 2019/2020, 5.7 million tons of teff grain has been produced, and 6.4 million tons of teff straw has been reported as waste [9]. Teff straw is the residue of teff and has around 38% cellulose, 27% hemicellulose, 18% lignin, and 10% extractives content, which makes it a competitive secondary source of cellulose with sugarcane bagasse, wheat straw, rice husk, etc. [10]. In Ethiopia, it is available on large scale and used for local uses such as bedding, mulching, and mud composite to make a house in rural, and more than 5 million tons of teff straw is disposed of as waste each year [11]. In this work, it is selected due to its high content of cellulose as the source of nanocellulose.

Polyvinyl alcohol (PVA) is a water-soluble synthetic biopolymer made by saponification of polyvinyl acetate. Transparency, high-thermal characteristics, and resistance to oxygen permeation are all notable advantages of this biodegradable material [12]. Because of its benefits of being nontoxic and highly durable, it is considered as a suitable source of materials for composite production [13]. Besides that, PVA is a polymer that is biodegradable in the soil and semicrystalline with outstanding chemical resistance [14]. Nevertheless, their high cost, high water absorption capacity, and low mechanical properties limit the applications of PVA materials [15]. To solve these problems, PVA is combined with other low-cost and decomposable polymers like cellulose, nanocellulose, nanoclay, and nanosilica. Many biopolymers are used as the matrix for composite preparation such as poly(vinyl) alcohol (PVA), poly(ethylene glycol) (PEG), poly(caprolactone) (PCL), and polylactic acid (PLA). Polyvinyl alcohol is selected due to its optical transmission, water-solubility, hydrophilicity, and good compatibility with nanocellulose and the fact that it does not need other modifications to use in the composite formulation with nanocellulose.

Intensive research has been done to develop different composite materials by extracting nanocellulose and nanofibrillated cellulose from different materials like bagasse [16], sisal fiber [17], garlic straw residue [18] coconut fiber [19], cotton [20], and pineapple [21]. [22] and [13] extracted NFC and reinforce it with PVA to study the tensile strength, thermal stability, and morphology of the polymer. [23, 24], and [25] reinforce polyvinyl alcohol with nanocellulose extracted from oil palm empty fruit bunches (OPEFB), MCC, and cotton stalk, respectively, to study its mechanical, thermal, and water absorptivity. But it needs to explore the influence of NC loading on PVA properties.

Thus, this study was made to investigate the improvement of properties of PVA by incorporating nanocellulose and to identify the effect of filler loading on the tensile strength, elongation at break, water absorption, thermal stability, optical properties, and biodegradability of PVA. The nanocellulose was extracted by sulfuric acid hydrolysis, and PVA-NC composite films were produced by the solvent-casting method. Finally, the properties of composite films and neat PVA were determined, and the effects of nanocellulose loading were investigated.

2. Materials and Methods

2.1. Materials. Polyvinyl alcohol (PVA) powder (CH_2CHOH), sodium hydroxide (NaOH, 97%), sulfuric acid (H_2SO_4, 98%), sodium chlorite ($NaClO_2$, 80%), acetic acid glacial (CH_3COOH, 96%) were purchased from Alkane PLC, Addis Ababa. Glycerol, filter paper (90 mm in diameter), and Petri dish were purchased from Ranchem Industry and Trading PLC, Addis Ababa. All chemicals are analytical grade and used as received. Different solutions were prepared according to the required concentrations in a laboratory, and the experimental works for production were conducted at Addis Ababa Institute of Technology, School of Chemical and Bioengineering Laboratory.

2.2. Methods

2.2.1. Extraction of Nanocellulose.

The nanocellulose used in this work was prepared from teff straw by acid hydrolysis, and the method used was adopted from Mandal and Chakrabarty [26] with little modification. The ground teff straw was kept in a water bath for 2 hours with an acidified sodium chlorite solution (0.7% *w/v*) at 75°C with continuous mechanical stirring (400 rpm). Glacial acetic acid (5% *v/v*) was used to adjust the pH of sodium chlorite to four. The fiber-to-acidified sodium chlorite solution ratio was 1 : 25 g/ml. The residue was vacuum-filtered, washed with distilled water, and the process was repeated until the pH of the filtrate was neutral. The residue was then oven-dried overnight at 50°C and kept for later use. For 1 hour, the dried holocellulose was immersed in a hot 4% *w/v* NaOH solution at 60°C under constant mechanical stirring (400 rpm), and the fiber-to-solution ratio was 1 : 20 g/ml. In a water bath, 10 grams of cellulose was hydrolyzed with a sulfuric acid solution while being continuously stirred (500 rpm). The cellulose-to-sulfuric-acid solution ratio was kept constant at 1 : 25 g/ml. To halt the hydrolysis process, tenfold distilled water (2500 ml) was added to the reaction mixture, and the suspensions were centrifuged for five minutes at 3000 rpm to remove acids. The suspensions collected from the centrifugation process are then dialyzed to remove the remaining acids. The dialysis process is performed with a dialysis tube (12-14 kDa MWCO) in distilled water for 10 days, with the water changed daily until the pH of the product was near neutral. After that, it was sonicated for five minutes at LIDI (Leather Industry and Development Institute), Ethiopia, with a probe ultrasonicator at 25 kHz to obtain a uniformly dispersed nanocellulose suspension and avoid agglomeration of the nanocellulose particles.

2.2.2. Preparation of PVA/NC Composite.

Solvent casting was used to make the nanocellulose–PVA composite films. The 5% *w/v* PVA aqueous solution was obtained by dissolving 5 g of PVA powder in distilled water at 80°C for 2 hours in a water bath with mechanical agitation (400 rpm) until the PVA was fully dissolved and a clear jelly-like solution was created. Various amounts of NC suspensions (1%, 2%, 3%, 4%, 5%, 6%, 7%, 8%, 9%, and 10%) were added to the PVA solution under constant manual stirring until the solution became viscous [14]. The ultrasonication operation was then performed for 15 minutes in water using a sonicator to obtain a homogenous mixture and avoid nanocellulose agglomeration in the PVA matrix at Addis Ababa Institute of Technology, Department of Chemical and Bioengineering. Then, the solutions were kept for 24 hours to obtain bubble-free solutions before casting on the Petri dish. Then, different films were cast on the Petri dish according to their perspective ratio and left for 7 days to obtain dry films.

2.2.3. Characterization of Nanocellulose/PVA Composite

(1) Water Absorption Capacity.

The water absorption tests were done by the water immersion method and carried out by following the procedures described by [14] with slight modifications. The composite films' water absorption capacity was determined by drying them in a vacuum oven at 50°C for 24 hours. The film was then cooled and weighted instantly. For 5 minutes, the prepared film was soaked in distilled water. The remaining water on the film was then removed, and the film was immediately weighted. The water intake capacity of each sample was calculated using Equation (1), and the average result is reported as a mean value with average deviation values of three replicates.

$$\text{Water Absorption Capacity} = \frac{W_t - W_o}{W_o} \times 100\%. \quad (1)$$

(2) Mechanical Properties.

To study if the properties of PVA improved after reinforcing the nanocellulose particles, the mechanical characteristics of composite films were measured. The tests were carried out according to ASTM D 882-92 standards (ASTM 2012). From the films, rectangular samples were cut into 60 mm by 13 mm strips. Before measuring mechanical characteristics, the cut films were conditioned at temperatures and relative humidity of 23 ± 2°C and 50 ± 10%, respectively. Equations (2) and (3) are used to determine the tensile strength and elongation at the break of the films, respectively, three measurements were taken, and the mean value with standard deviation was reported.

$$\text{Tensile strength} = \frac{\text{Maximum breaking load applied}}{\text{Crossectional area of the film}}, \quad (2)$$

$$\text{Elongation at break} = \frac{\text{Final length at break-Initial length}}{\text{Initial length}}. \quad (3)$$

(3) Thermal Stability.

Thermogravimetry analysis was performed in this study to understand how reinforcing nanocellulose particles affect the thermal property of PVA, and it was conducted at the Jimma Institute of Technology's School of Material Science and Engineering using the Perkin Elmer TGA-4000 equipment. Under a nitrogen gas flow, the PVA and composite samples were heated from 25 to 500°C. The nitrogen gas flow rate was 20 ml/min, with a heating rate of 10 ml/min. The data from the derivative thermogravimetry system was also recorded. The data obtained were then analyzed using the Origin Pro 8.5 version.

(4) Biodegradability Test.

The degradability test was conducted to determine the rate of degradability of the composite and PVA films by the soil-burying method ASTM D 5988 [14]. The films were weighted after being divided into 50 mm by 50 mm strips. The degradation test was conducted by burying the films in a pot filled with soil to a depth of 5 cm. The films were carefully removed at 15-day intervals, and the residual soil on the film's surface was scraped carefully. Then, the weight loss with time was determined using Equation (4), three measurements were taken, and the mean value with standard deviation was reported.

$$\text{Degradation rate} = \frac{W_i - W_f}{W_i} \times 100\%, \qquad (4)$$

where W_i is the initial and W_f is the final weight of the films.

(5) Optical Properties. UV-Vis spectroscopy is an analytical technique that measures the amount of discrete wavelengths of UV or visible light that are absorbed by or transmitted through a sample in comparison to a reference or blank sample. Optical transmission properties of the neat PVA and PVA reinforced with nanocellulose were determined using the SPECORD 200 Plus UV-Vis spectroscopy (Analytic Jena, AG, and Germany) instrument in the wavelength range of 200 nm to 800 nm.

3. Result and Discussion

3.1. Characterization of PVA-NC Composite Films

3.1.1. Mechanical Properties. The result of the mechanical properties of PVA and PVA-NC composite films obtained is presented in Table 1. The result illustrates that the addition of NC to PVA increases the tensile strength up to an addition of 5%NC and increases elongation at break at a 2%NC loading rate. The addition of 2%NC and 5%NC increases the tensile strength from 24.5 MPa to 35.5 MPa and 40.6 MPa, respectively, and then decreased to 36.5 MPa with 7%NC. The elongation at break was increased from 40% to 45.7% with 2%NC and reduced to 36.5% by incorporating 5%NC. After the NC loading reaches 7%, it declined to 32.2%.

According to Figure 1, there is an increase in tensile strength of the films up to nanocellulose loadings reaching 5%. This is aroused by the existence of fine particles with a high aspect ratio and dispersion of nanocellulose in the PVA matrix and indicates the presence of intermolecular interactions between nanocellulose and PVA [27]. The mutual attraction of the PVA matrix and NC with hydroxyl-rich surfaces and the strong bonding between the hydroxyl groups of NC and matrix could explain the improved mechanical properties of composites [23].

The addition of 7% nanocellulose to the PVA solution resulted in a reduction in tensile strength (Figure 1). The explanation for this decrease is due to an increase in intermolecular contacts caused by the extra amount of nanocellulose, which may compete with the interactions of polyvinyl alcohol and nanocellulose. Therefore, compatibility between PVA and nanocellulose decreased, which results in the decrement of tensile strength at a higher loading amount of nanocellulose [25]. Another reason could be because the number of flaws in nanocomposites, such as weak borders between particles and bubbles trapped during sample preparation, increases with increasing reinforcement, resulting in a decrease in composite tensile strength. The amount of nanocellulose used enhances inhomogeneous dispersion.

When the nanocellulose loading reaches 2%, there is an increase in elongation at a composite break (Figure 2). However, after adding 5%NC, a decrease was noticed, which could be due to the rigidity of the fibers. Strong contacts

Table 1: Tensile strength and elongation at break of neat PVA and composite films with different NC loading.

Composite samples	Tensile strength (MPa)	Elongation at break (%)
PVA only	24.5 ± 0.53	40 ± 0.52
PVA/1%NC	30 ± 0.61	42.3 ± 0.73
PVA/2%NC	35.5 ± 0.55	45.7 ± 0.53
PVA/3%NC	37 ± 0.57	43.8 ± 0.64
PVA/4%NC	38.7 ± 0.65	39.7 ± 0.57
PVA/5%NC	40.6 ± 0.73	36.5 ± 0.79
PVA/6%NC	39.2 ± 0.52	34 ± 0.52
PVA/7%NC	36.5 ± 0.47	32.2 ± 0.75
PVA/8%NC	33.4 ± 0.72	27 ± 0.46
PVA/9%NC	32 ± 0.54	25.57 ± 0.43
PVA/10%NC	29.5 ± 0.71	22.67 ± 0.56

NB: the average result is reported as a mean value with average deviation values of three replicates.
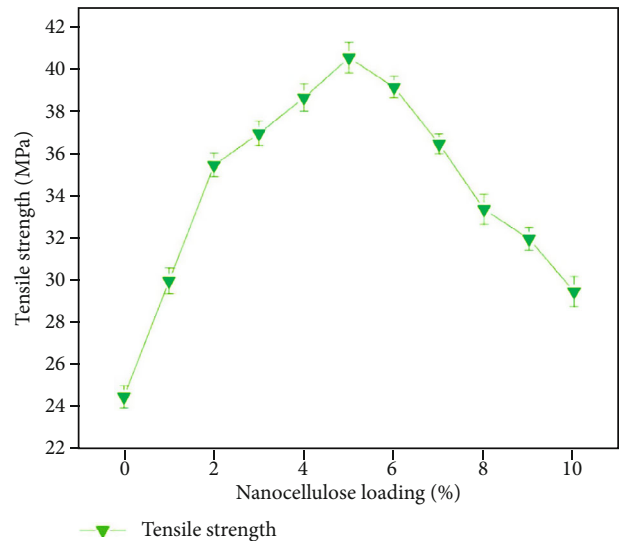

Figure 1: Tensile strength of PVA and PVA-NC composite films.

between nanocellulose and PVA chains may also limit chain mobility, reducing ductility and preventing it from stretching.

[28] reported that the tensile strength of nanocellulose-reinforced PVA composite films rose up to 6% nanocellulose loading and then decreased after that. The elongation at break rose linearly as the amount of NC added to the PVA matrix increased, with a reduction when the NC loading reached 10%. This reduction could be attributed to an increase in the viscosity of the film solution as the amount of nanocellulose in the composite films increases, the orientation of the nanocellulose in the composite films, void formation, and severe filler agglomeration during processing [29].
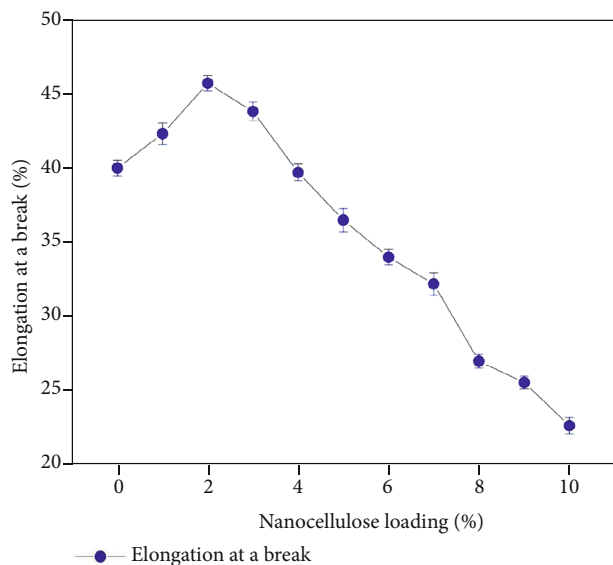
FIGURE 2: Elongation at break of PVA and PVA-NC composite films.

TABLE 2: Result of water absorption capacity of PVA composites with different NC loadings.

Nanocellulose loading	Water absorption capacity (%)
PVA/0%NC	84.6 ± 0.56
PVA/1%NC	79.4 ± 0.62
PVA/2%NC	73 ± 0.78
PVA/3%NC	71 ± 0.76
PVA/4%NC	68.5 ± 0.77
PVA/5%NC	66 ± 0.67
PVA/6%NC	62 ± 0.84
PVA/7%NC	61 ± 0.59
PVA/8%NC	65.6 ± 0.72
PVA/9%NC	67 ± 0.82
PVA/10%NC	72.7 ± 0.88

NB: the average result is reported as a mean value with average deviation values of three replicates.

3.1.2. Water Absorption Capacity. The result obtained from the experiment in Table 2 indicates the water intake capacity of the composite films decreased linearly with increasing the NC loading. For neat PVA, the highest amount of water absorption percentage was obtained which was 84.6% and reduced to 73% by the addition of 2%NC. By increasing the nanocellulose loading to 7%, the water absorption capacity was reduced to 61%.

Figure 3 illustrates the relationship between the nanocellulose loading rate and water absorption capacity. The reduction of water absorption capacity was observed, and this result reveals that the nanocellulose-reinforced films have lower water absorption which means they have good water preventive ability against water. The observed improvement in the properties of the films after the reinforcement of NC was related to several reasons. The first reason is the formation of a network structure between NC and PVA components which prevents the water molecules from dissolving the films and improves their water resistance [14]. The other is that the hydrogen bond formation between the polymer matrix and NC prevents the hydrogen bonding sites for water molecules. The water absorption test is the most important character in the evaluation of composite effectiveness. Mechanical properties and moisture absorption behavior are related to each other. Films that absorb less water will show better mechanical properties. The other thing is that the usage of polyvinyl alcohol in food packaging applications was limited because of its properties to absorb water. The packaging materials that absorb water are suitable for the growth of bacteria and microorganisms [30]. These properties affect and lead to the deterioration of food under storage. So, the addition of NC shows better improvement in the water preventability of the PVA matrix.

The same results were reported by different authors. [31] produced a vinyl-ester eco nanocomposite by reinforcing with it nanoclay and reported that the addition of 5% nano-
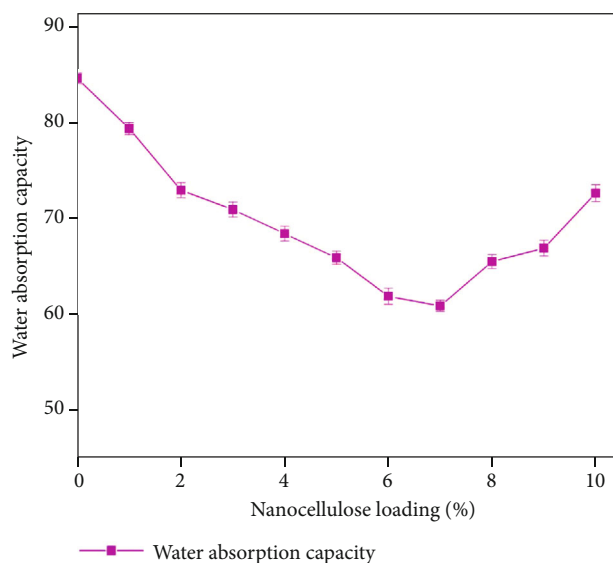

FIGURE 3: Water absorption capacity of PVA-NC composite films at various amounts of nanocellulose loading.

cellulose reduces the water intake capacity by 7.5%. Lani et al. [32] reported that increasing the nanocellulose content up to 10% reduces the water intake capacity of PVA by 20%. The water intake capacity results in this work were in agreement with the works of literature.

3.1.3. Thermogravimetry Analysis. The TGA and DTG curves obtained are given in Figure 4. TGA curves of both films reveal that three regions of mass loss, in the first degradation stage occur between the region of 30°C-180°C which is related to the evaporation of absorbed water and accounts for 10.86% for neat PVA and 7.4% for PVA composite (Figure 4(a)). The second region was from 200°C to 370°C,
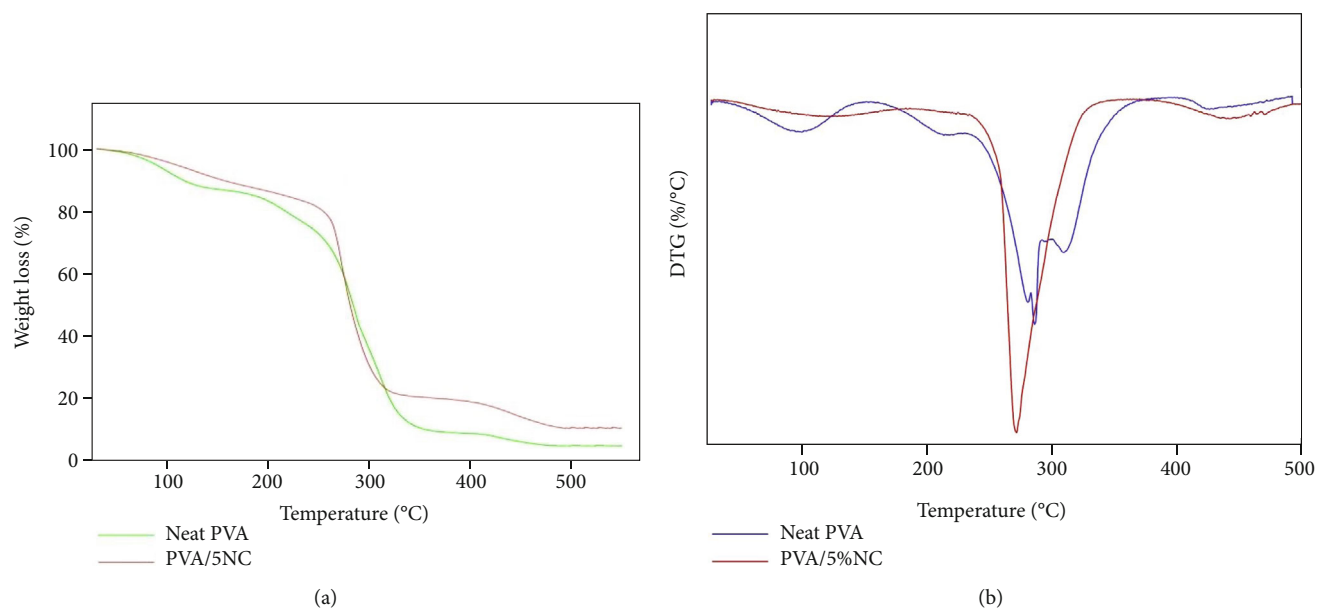
(a)

(b)

Figure 4: Thermal degradation of PVA-0%NC and PVA-5%NC composite films. (a) TGA curves and (b) DTGA curves.
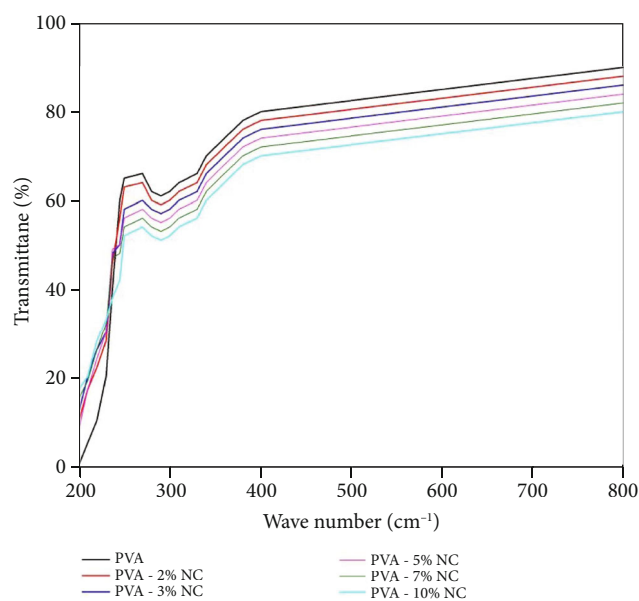

Figure 5: Result of UV-Vis spectrometer for neat PVA and PVA-NC composite films.

Figure 6: Biodegradability result of neat PVA and PVA-NC composite films.

and it is due to the degradation of PVA intermolecular hydrogen bonding decomposition of PVA by a dehydration reaction on the polymer chain and by the degradation of the main backbones. The third region is above 400°C which indicates the carbonization of organic materials. As observed in Figure 4(b), the DTG curves of the neat PVA film reached its maximum peak at a temperature of 290°C, and the nanocellulose-reinforced PVA film reached its maximum peak at 275°C. The thermal degradation temperature of the PVA film shifted to lower temperatures after reinforcement. The char residue is high for PVA composite film than that of neat PVA. This shows the reduction in thermal
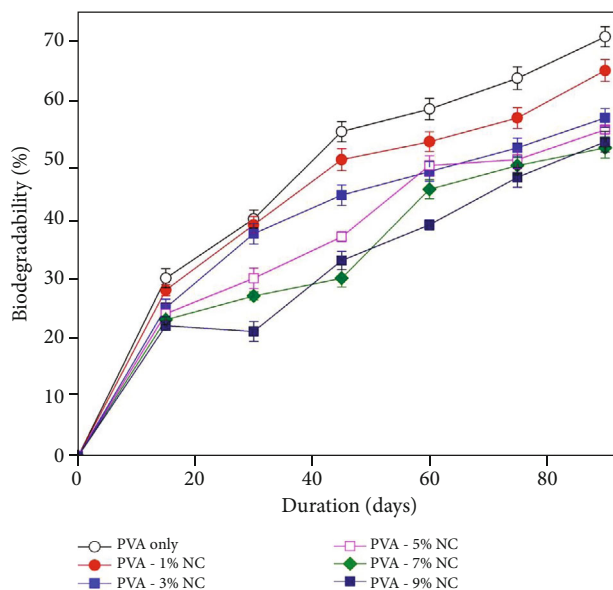
stability of PVA after incorporating nanocellulose particles, and this is aroused by the lower thermal stability of NC than PVA polymer. The nanocellulose extracted by sulfuric acid shows lower stability than that extracted by other methods because of the existence of sulfate groups created in nanocellulose during the acid hydrolysis process, and the thermal degradation of sulfate groups starts at lower temperatures [33].

There are different ideas related to this thermal degradation of nanocomposites after reinforcing the nanocellulose extracted by sulfuric acid. [34] reported that the incorporation of 9% nanocellulose into PVA did not improve the

TABLE 3: Weight loss of neat PVA and composite films at different NC loadings after 90 days.

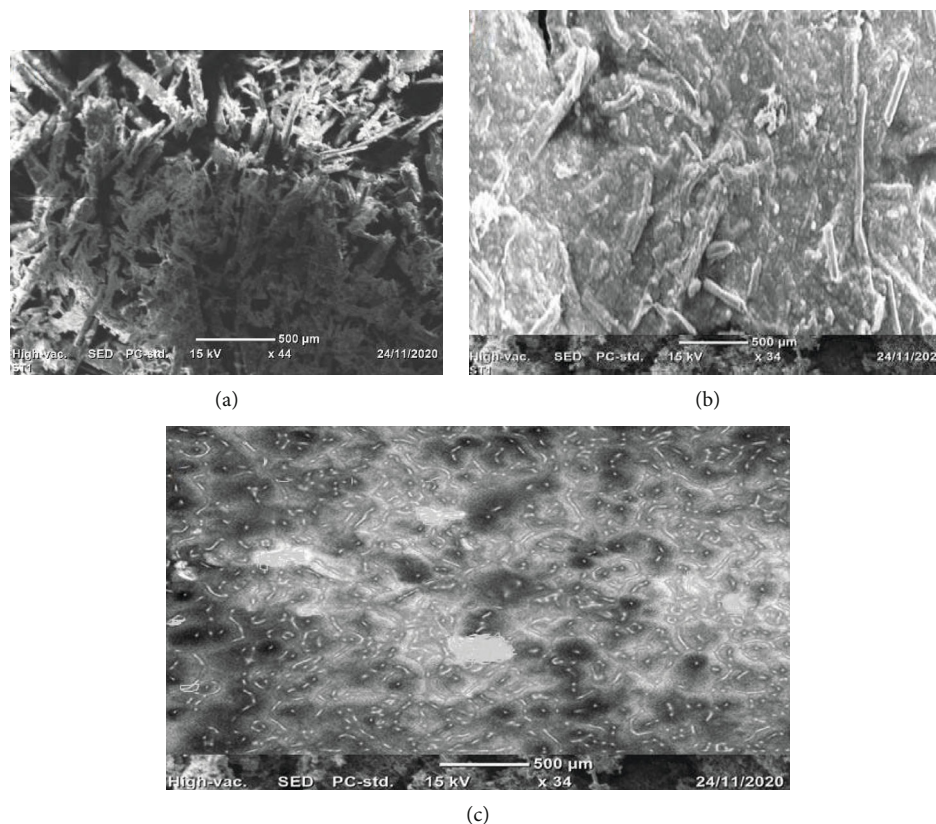| NC loading (%)	0	1	3	5	7	9
Weight loss (%)	70.7 ± 1.7	65 ± 1.85	57.2 ± 1.57	55.6 ± 0.64	52 ± 1.73	53.1 ± 1.72

(a)

(b)

(c)

FIGURE 7: Scanning electron microscopy image for (a) teff straw, (b) nanocellulose, and (c) PVA-6%NC composite films.

thermal stability of the nanocomposite material and discussed that the reduction in polyvinyl alcohol composites is aroused by the existence of the sulfate groups in the nanocellulose. [33] reported the same argument, and the neutralization of nanocellulose suspension by alkaline solution improves the stability, but the suspensions inclined to aggregate due to lacking electrostatic repulsion force between the crystal particles. So, the reduction in thermal stability of composite material observed in this work is in the agreement with that of previous works, and it is possible to say that incorporation of nanocellulose extracted by sulfuric acid into PVA polymer does not improve its thermal properties.

3.1.4. Optical Properties. Figure 5 presents the UV-Vis spectra of neat PVA, PVA-2%NC, PVA-3%NC, PVA-5%NC, PVA-7%NC, and PVA-10%NC to observe the change in transmittance of PVA after incorporating nanocellulose. The neat PVA has the highest transmittance with a value of 85.4%. The transmittance (Tr) value of PVA composite incorporated with 2%NC at 600 nm is 78.2% which shows the decrement, but it has good transmittance when compared to other reinforced composite materials. The Tr value for PVA composites incorporated with 3%NC, 5%NC, 7%NC, and 10%NC was 76.8%, 74.6%, 65.3%, and 63.7%, respectively.

From this result, the decrement observed in transmittance as the amount of nanocellulose loading increased is aroused from the agglomerations of nanocellulose that acted as a light-blocking barrier. The higher the nanomaterial loading, the more likely they were to interact and aggregate within the PVA matrix, resulting in a reduction in light transmission.

A similar result was reported by [34]; nanocomposite films were produced with various amounts of NC loadings, their transmittance was compared to that of the neat PVA, and the neat PVA has a higher transmittance than that of the reinforced composites. The research concludes that the decrease in transmittance of the composites after the reinforcement possibly rose from the agglomeration of NC as the amounts of loadings increased.

[23] also conducted the UV-Vis spectrometer test for PVA-NC and PVA-nanosilica composite films. The neat

PVA shows high transmittance than that of the reinforced ones. The decrement was observed after incorporating 3%NC, and further decrement was observed after adding 0.5% nanosilica to the PVA-3%NC composite and concluded that the reinforcement of nanocellulose and nanosilica reduces the transmittance of PVA.

3.1.5. Biodegradability. The influence of varying amounts of nanocellulose fiber on PVA film weight loss rate was investigated in this study using a soil burial test, and the results were provided in Figure 6. The test was conducted with various amounts of nanocellulose addition for 90 days. PVA is a biodegradable polymer and can be decomposed naturally by algae, fungi, water vapor, and other microorganisms. The biodegradability of neat PVA is 70.7% after 90 days while the weight loss of the PVA composite films reinforced with 1%, 3%, 5%, 7%, and 9% were 65%, 57.2%, 55.6%, 52%, and 53.1%, respectively, presented in Table 3.

From this result, PVA shows good degradability over other reinforced samples. This is because the water absorbed by the films caused microorganisms such as algae, bacteria, and fungi to adhere and develop; once the microorganisms get attached to the films, they produce enzymes that break down the composite films.

This result is consistent with the water absorption test of PVA/nanocellulose nanocomposite films, indicating that the reinforcement of nanocellulose fiber prevented water from diffusing into the films, allowing microorganisms to develop slowly.

The result obtained in this work is in agreement with the result reported by [35] in which PVA is reinforced with the modified cellulose fiber. In the soil, all of the films prepared exhibited nearly identical resistance to microbial attack. The composites lose structural integrity when the microorganism attacks. The data provided here illustrate that the film composites will have no adverse environmental impacts which means the film composites are completely biodegradable.

3.1.6. Surface Morphology. The scanning electron microscopy result of teff straw, nanocellulose, and PVA-6%NC is given in Figure 7. The surface of NC shows a smooth surface which suggests the removal of amorphous parts in the straw after the acid hydrolysis process during preparation. The SEM micrograph for the PVA-6%NC composite film showed a network structure that was formed after reinforcement. This network represents the interaction between PVA polymer and nanocellulose by forming a rigid percolation reinforced network within-host matrix and strong interaction between the two constituents through hydrogen bonding.

From (Figure 7(a)), it is identified that there is a uniform distribution of nanocellulose within the host matrix. The white dots were also observed in the picture, and this could arise from bubbles generated during PVA nanocomposite film production. During homogenization with ultrasonication, some bubbles were left in the solution and appeared on the film during the casting process which affects the morphology and mechanical properties of the composite.

4. Conclusion

In this study, nanocellulose was successfully extracted by the sulfuric acid hydrolysis method and reinforced with polyvinyl alcohol by the solvent-casting method. The effect of reinforcement of nanocellulose in the PVA matrix on its mechanical properties, water intake capacity, transmittance, thermal stability, and biodegradability was investigated. The result of this work implies that the properties of PVA in terms of tensile strength, elongation at break, and water absorption capacity were improved due to good compatibility between PVA matrix and nanocellulose. Moreover, the properties of composites were affected by the different content of nanocellulose. The tensile strength and water absorption capacity of PVA increased until the amount of nanocellulose reaches 5% and 7%, respectively, while the elongation at break increased from $40 \pm 0.5\%$ to $45.7 \pm 0.53\%$ with 2%NC and reduced to $36.5 \pm 0.47\%$ with 5%NC. The transmittance of PVA was decreased after incorporating the NC. The neat PVA has the highest transmittance with a value of 85.4% while the transmittance of PVA composite incorporated with 2%NC at 600 nm is 78.2% which shows the decrement in transmittance, but it is good when compared to other reinforced composite materials. The thermal stability of polyvinyl alcohol after incorporating nanocellulose particles was reduced, and this arouses from the lower thermal stability of nanocellulose than PVA polymer. The neat PVA is more degradable than other reinforced samples, but almost all samples show weight loss. The SEM result of 5%-PVA composite film indicates the strong interaction between nanocellulose and PVA which results in improvement of its mechanical and water absorption properties. The reinforcement of nanocellulose into a PVA matrix with high content of nanocellulose affects its properties due to the formation of agglomeration and inhomogeneous dispersion. However, the reinforcement with low nanocellulose content (<7%) improves its mechanical properties and water absorption capacity and does not affect the degradability and optical properties of the matrix. Therefore, nanocellulose extracted from the teff straw is a good source to use as a reinforcing agent of the PVA matrix.

Data Availability

The data used to support the findings of this study are included within the article.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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