Producing nanocellulose from kenaf (*Hibiscus cannabinus L*) as a nanoreinforcing agent for polyvinyl alcohol (PVA) film

Siti Aminah*

Food Technology and Nutrition Department, Faculty of Halal Food Science, Djuanda University, Jl. Tol Ciawi No. 1, Bogor, West Java, Indonesia Email: siti_aminah@unida.ac.id *Corresponding author

Joko Hermanianto and Nugraha Edhi Suyatma

Faculty of Agricultural Technology, Food Science and Technology Department, IPB University, Bogor, 16680, Indonesia Email: jokoher@yahoo.com Email: nugrahaedhi@yahoo.com

Evi Savitri Iriani

Indonesian Center for Agricultural, Postharvest Research and Development, Jl. Tentara Pelajar No. 12 Cimanggu-Bogor 16114, West Java, Indonesia Email: evisavitri@pertanian.go.id

Abstract: Polyvinyl alcohol (PVA) is a polymer used as an active layer alternative. These advantages depend on the humidity rate or the water vapour transmission rate. Kenaf is a plant rich in fibre and used as a primary source of cellulose. This research aimed to analyse PVA films' physical, mechanical, and morphological characteristics by adding nanocellulose kenaf and glycerol. The cellulose isolation of kenaf was done by the smoothing of the fibre method, delignification, and bleaching. The steps for producing nanocellulose were using 2,2,6,6-tetramethyl-1-piperidinyloxy, ultrafine grinding, and ultrasonication. Nanocomposite film (PVA + nanocellulose + glycerol) was produced using the solvent casting method. The treatments in this study were the addition of cellulose nanofiber (0%, 3%, and 5%) and the addition of glycerol (0% and 2%). The results showed that adding nanocellulose can increase the total value of colour difference (ΔE) and tensile strength but reduce the value of light transmission, water vapour transmission rate (WVTR), and elongation. The addition of glycerol has a positive effect on the elongation value but has a negative impact on the WVTR value and tensile strength. The film had the highest total colour difference (ΔE), adding 5% nanofiber cellulose and 2% glycerol (1.62 \pm 0.49). The best tensile strength was sorted in film, adding 5% nanofiber cellulose without glycerol. The highest elongation break

190 S. Aminah et al.

was on nanocomposite film, adding 5% cellulose and 2% glycerol. We concluded that reinforcing nanocellulose kenaf can enhance PVA film's physical and mechanical characteristics.

Keywords: kenaf; nanocomposite film; PVA; polyvinyl alcohol; 2,2,6,6-tetramethyl-1-piperidinyloxy.

Reference to this paper should be made as follows: Aminah, S., Hermanianto, J., Suyatma, N.E. and Iriani, E.S. (2024) 'Producing nanocellulose from kenaf (*Hibiscus cannabinus L*) as a nanoreinforcing agent for polyvinyl alcohol (PVA) film', *Int. J. Nano and Biomaterials*, Vol. 10, No. 3, pp.189–203.

Biographical notes: Siti Aminah received her Master of Science degree in 2017 in the Department of Food Science, Bogor Agricultural Institute. She is a lecturer at the Department of Food Technology and Nutrition at the University of Djuanda Bogor. Her field of study is food engineering. Her research interests are in food packaging technology, animal food processing technology and nanotechnology.

Joko Hermanianto completed his Doctoral studies in Food Science at the University of Gottingen, Germany in 1995. He is a Lecturer at the Department of Food Technology, IPB University. His research interests include the development of halal assurance systems in the food industry, food processing technology of meat and meat products.

Nugraha Edhi Suyatma obtained his doctorate in 2006 in Materials Chemistry (Chimie des Matériaux, Université de Reims, France). Currently, he works as a Lecturer at the Department of Food Technology, IPB University. His research interests are the development and reinforcement of bio-based packaging from agricultural waste products (e.g., chitosan and pectin); application of edible/biodegradable film in food process and preservation, bio-nanocomposites with ZnO and Silver nanoparticles as fillers in biopolymer, development of active and intelligent packaging.

Evi Savitri Iriani currently works at the postharvest technology, Indonesian Agency for Agricultural Research and Development. She does research in Polymer Chemistry and Nanotechnology. Their current project is 'Utilization of agrowaste material'.

1 Introduction

Polyvinyl alcohol (PVA) is an environmentally friendly polymer. PVA has several superior properties, including resistance to oxygen, mechanical properties, chemical resistance, film-forming ability, and solubility in water (Tian et al., 2017). Barrier characteristics on films are essential because they affect the shelf life of packaged food products (Ma et al., 2016). However, the presence of clusters of the dominant hydroxyl causes the characteristic low water vapour barrier. The addition of nanocellulose fillers has been widely developed (Liu et al., 2014). This is because nanocellulose comes from plants whose availability is abundant, inexpensive, and non-toxic. In addition, adding

nanocellulose can increase the crystallinity, surface area, and the ability to disperse in plastic composites (Ioelovich, 2012). With this ability, adding nanocellulose to the polymer matrix produces better physical and mechanical characteristics and is environmentally friendly (Silverio et al., 2013). In addition, PVA cellulose-based films can produce thick and robust food packaging. On the other hand, it has high flexibility because it can be modified to various laboratory conditions. The reaction between PVA and cellulose reportedly increases the film's toughness and water absorption capacity (Panda et al., 2022).

Kenaf is a natural cellulose source plant that has the potential to be added to polymers (Akhtar et al., 2017). Kenaf belongs to the genus Hibiscus (Faruk et al., 2012). Kenaf produces a fibre of about 56% cellulose in the bark and 46% in the core. Kenaf has a strong structure and can withstand the pressure applied. Compared with natural fibres such as sisal, banana, and hemp, kenaf has greater adaptability, large biomass, and lower production costs (Khan et al., 2023). Hafizah et al. (2014) reported that the tensile strength of kenaf/epoxy composites, kenaf/polyester composites, and kenaf/vinyl ester composites were 3–6 times higher than non-composite polymers.

Nanocellulose has been successfully added to several polymers as nanofillers, including polylactic acid, starch, polycaprolactone, alginate, polyethylene oxide, and PVA (Liu et al., 2014). Qiu and Netravali (2012) reported that adding nanocellulose to the PVA film improved the mechanical and thermal characteristics. Similar results were obtained in Ibrahim et al.'s (2010) study, which found that adding nanocellulose to PVA composites increased mechanical characteristics such as tensile strength and percentage elongation. PVA-based composite films have shown high potential for use as active and smart food packaging (Oun et al., 2022).

Plasticisers are non-volatile compounds that usually can biodegrade. This allows for improving the properties of the film. Commonly used plasticisers are polyols, mono-, di- and oligosaccharides (Kazon et al., 2018). Glycerol is a plasticiser widely applied to hydrocolloid-based films because it can increase film flexibility and adequate processability (Ben et al., 2022). Therefore, the effect of adding glycerol on kenaf film nanocomposites needs to be studied.

The ability of nanocellulose to improve film characteristics is influenced by the characteristics of nanocellulose. Size, variety, source, nanocellulose content, and plasticiser added to the film are essential factors determining the manufacture of nanocellulose-based films. The purpose of this study is to assess the production of cellulose nanofiber from kenaf plant sources to analyse the characteristics of the physical, mechanical, and structural morphology of nanocomposite films by adding nanofiber of kenaf cellulose and glycerol as a plasticiser.

2 Materials and methods

2.1 Kenaf nanocellulose production

Stages of kenaf fibre cellulose isolation refers to the method of Abe et al. (2007), which comprises smoothing the fibre, reflux/delignification, and bleaching. Delignification was done using the soxhlet method using toluene: ethanol (2 : 1) solvent for 6 h. Bleaching is carried out by mixing 31.9 fibre plus 9 g of NaClO2, 1.2 mL of acetic acid, and 900 mL

of distilled water heated in a water bath at 70–80°C. The results were rinsed to pH seven and soaked in 6% KOH for 24 h, followed by heating at 90°C for 20 min.

Nanocellulose production is divided into two types: TEMPO oxidation methods and without oxidation. The later stages are the same: ultrafine grinding and ultrasonication. TEMPO oxidation refers to the method developed by Saito et al. (2007). A total of 83.74 g of cellulose fibre with a dry weight of 12 g was suspended in a mixture of 1200 ml of water, 0.192 g of TEMPO, and 1.2 g of NaBr and 60 mmol NaClO₂ (pH 10) were added. The solution was maintained at a pH 10 with 0.5 M NaOH gradually for 5 h. Ethanol 5 ml was added to stop the reaction. TEMPO-oxidised cellulose is rinsed with distilled water until the pH is neutral and the water is separated. Ultrafine grinding uses 2% of solid contents plus the gap, and the number of repetitions used in this study is +5 (15x), 0 (15x), -5 (15x), -10 (10x), -12.5 (2x). Ultrasonication done by A = 80, t = 80 min, and $T < 50^{\circ}$ C. The obtained nanocellulose was characterised by its morphological structure by scanning electron microscope (SEM) and transmission electron microscope (TEM), as well as the characterisation of functional groups by FTIR (ASTM E 168).

2.2 Manufacture of nanocomposite film polyvinyl alcohol (PVA)/kenaf nanocellulose

Nanocomposite manufacturing is done by solvent casting method with modifications of Cho et al.'s (2011) method. Solution of 5% PVA made by dissolving the PVA powder and distilled water at 130–150°C. A solution of 5% PVA was added with different concentrations of nanocellulose (Table 1). The solution was stirred for 3 h, let stand for \pm 24 h, and put on the Teflon. Thus, the solution was placed at 50°C until it dried out. The combination treatments are in Table 1.

2.3 Characterisation of nanocomposite films based on polyvinyl alcohol

Characterisation includes morphological physical properties and mechanical properties. Characterisation of physical properties includes the analysis of film thickness with a micrometer, the analysis of water vapour transmission rate (water vapour transmission rate/WVTR) with ASTM E 168 (2009) methods, transmission analysis of light with UV-Vis spectrophotometer, and colour analysis with chromameter. The mechanical characterisation measured by the universal testing machine (UTM) includes tensile strength and elongation.

2.4 Experimental design

The experimental design used a Completely Randomised Factorial Design with three factors. The factors were the type of nanocellulose (TEMPO and without TEMPO), the concentration of nanocellulose (3%, 5%), and the addition of glycerol (0% and 2%). This research used three replications. Data analysis used analysis of variance (ANOVA) with SAS software version 9.2. The significantly different results were followed by Duncan's further test with a significance level of $\alpha = 5\%$.

| | Treatment | | | |
|---------|--------------|-------------------|-----------------------|--|
| Samples | Glycerol (%) | Nanocellulose (%) | Type of nanocellulose | |
| Р | 0 | _ | Ν | |
| PN3G0 | 0 | 3 | Ν | |
| PN5G0 | 0 | 5 | Ν | |
| PN5G0T | 0 | 5 | NT | |
| PN3G0T | 0 | 3 | NT | |
| PG2 | 2 | _ | NT | |
| PN3G2 | 2 | 3 | Ν | |
| PN5G2 | 2 | 5 | Ν | |
| PN3G2T | 2 | 3 | NT | |
| PN5G2T | 2 | 5 | NT | |

 Table 1
 The combination treatments in producing nanocomposite

(N): Nanocellulose without TEMPO steps, (NT): Nanocellulose with TEMPO steps.

3 Results and discussion

3.1 Characteristics of functional groups

Fourier Transform Infrared Spectroscopy is an analytical technique to identify the chemical structure of vibration or blend caused by the sample treatment difference (Khawas and Deka, 2016). The nanocellulose produced was analysed using FTIR to determine the functional groups produced after TEMPO oxidation treatment. The results of the FTIR test on the sample can be seen in Figure 1.





Figure 1 shows the FTIR spectra from kenaf cellulose insulation results and kenaf cellulose after the TEMPO oxidation process. Figure 1 shows that the peak at 3433 cm^{-1} indicates some of the OH group. OH groups in cellulose TEMPO showed a lower intensity than those in cellulose. This is because the function of TEMPO oxidisation that modifies the OH group contributes to hydrophilic properties becoming less produced.

Peaks of nanocellulose samples without or with TEMPO oxidation modified TEMPO oxidation is no different. This means that the TEMPO oxidation process that was carried out was not effective. A less-than-optimal formulation causes this. Zhou et al. (2012) stated that during the TEMPO reaction, at room temperature and in an alkaline medium, changes in the morphological characteristics of the fibre occur and produce functional surfaces (carboxyl, aldehyde, and hydroxyl).

3.2 Morphological characteristics

Morphological characteristics were observed to see the effects of each stage of nanocellulose production. The morphology of kenaf nanocellulose in this study was observed using SEM and TEM. SEM is used to determine the surface profile nanocellulose. Kenaf nanofiber cellulose produced in this study can be seen with a SEM at 15000x magnification and 89.63 nm diameter (Figure 2). Chaker et al. (2014) stated that nanocellulose is cellulose with a diameter less than 100 nm. The results of SEM observations on the sample can be seen in Figure 2.

Figure 2 Results of scanning electron microscope



The distribution level of nanocellulose was seen using TEM. The results of observations on TEM can be seen in Figure 3.

Figure 3 Results of TEM: (A) nanofiber cellulose after grinding; (B) nanofiber cellulose tempo after grinding; (C) nanofiber cellulose after ultrasonication and (D) TEMPO nanocellulose after ultrasonication (see online version for colours)



Bar 500 nm.

Figures 3(A) and (B) show that the cellulose was successfully fibrillated after grinding. The ultrafine grinding stage increases the surface area and solvent access to the fibre interior, so defibrillation is effective (Chandra et al., 2016). However, the distribution could be more optimal. Some fibre structures are in the form of bound threads. On the other hand, the TEMPO method nanocellulose (Figure 3(B)) looks more fibrillated. Ketabchi et al. (2015) stated that The TEMPO oxidation process can help the defibrillation process more optimally. Figure 3(C) and (D) show cellulose with ultrafine grinding and ultrasonication processes. The nanocellulose structure is well distributed. These results were strengthened by morphological analysis with SEM.

3.3 Physical characteristics

The physical characteristics of this research are the film thickness, the total value of colour difference (ΔE), and the water vapour transmission rate (WVTR). The test results of each parameter are presented in Table 2.

3.4 The total value of colour difference (ΔE)

The value of ΔE can be used as a consideration before the application of nanocomposites as packaging for food products. The lower the value of ΔE in the nanocomposite, the visual appearance of the packaged product does not change much. In Table 2, it can be seen that adding nanocellulose in general only slightly increased the value of ΔE . Li et al. (2012) produced a composite film with a more turbid colour as the amount of nanocellulose added. A significant increase was seen in the addition of 5% nanocellulose. This amount is able to make the colour of the film become more cloudy. Pereira et al. (2014) added 5% nanocellulose from banana fibre in PVA and increased the E value due to the increase in light absorbed by the nanocomposite. Nanocellulose also fills the voids between the polymer matrix chains, increasing the nanocomposite's light barrier (Fortunati et al., 2013; Savadekar and Mhaske, 2012; Silverio et al., 2013).

| Sample code | ΔE | WVTR (gr/h/mm ²) | Light transmission (%) |
|-------------|------------------------|---------------------------------|-------------------------------|
| Р | $1.42\pm0.01^{\rm A}$ | $134.41 \pm 8.65^{\rm AB}$ | $83.13\pm0.97^{\rm A}$ |
| PN3G0 | 1.11 ± 0.13^{AB} | $91.66\pm2.17^{\text{CDE}}$ | $57.97 \pm 1.14^{\mathrm{B}}$ |
| PN5G0 | 1.53 ± 0.00^{AB} | $80.67 \pm 12.26^{\text{CDE}}$ | $58.76 \pm 3.70^{\circ}$ |
| PN3G0T | 1.29 ± 0.14^{BC} | $93.34\pm8.71^{\rm DE}$ | $64.88\pm2.46^{\rm D}$ |
| PN5G0T | $1.06\pm0.15^{\rm A}$ | $105.12 \pm 13.51^{\mathrm{E}}$ | $81.22\pm2.98^{\rm D}$ |
| PG2 | $1.25\pm0.22^{\rm C}$ | 94.12 ± 16.07^{CDE} | $72.53\pm0.05^{\rm A}$ |
| PN3G2 | 1.09 ± 0.17^{BC} | $103.90 \pm 12.60^{\text{CDE}}$ | $71.17\pm3.20^{\mathrm{B}}$ |
| PN5G2 | $1.26\pm0.05^{\rm AB}$ | $139.88\pm3.24^{\rm A}$ | $65.06 \pm 0.14^{\rm C}$ |
| PN3G2T | $0.92\pm0.06^{\rm C}$ | $109.52 \pm 5.31^{\text{CD}}$ | $61.77\pm0.53^{\rm CD}$ |
| PN5G2T | $1.53\pm0.08^{\rm A}$ | 116.21 ± 4.20^{BC} | $56.47\pm4.41^{\rm D}$ |

 Table 2
 Nanocomposite physical characterisation

The difference between the letters A-E in the same column shows a significant difference in Duncan's post hoc test with = 5%. ΔE (the total value of colour difference), Water vapour transmission rate (WVTR); P = PVA film; PN3G0 = PVA + 3% nanocellulose; PN5G0 = PVA + 5% nanocellulose; PN3G0T = PVA + 3% nanocellulose TEMPO; PN5G0T = PVA + 5% nanocellulose TEMPO; PG2 = PVA + 2% glycerol; PN3G2 = PVA + 3% nanocellulose + 2% glycerol; PN3G2T = PVA + 3% nanocellulose TEMPO + 2% glycerol; PN5G2T = PVA + 5% nanocellulose TEMPO + 2% glycerol; PN5G2T = PVA + 5% nanocellulose TEMPO + 2% glycerol; PN5G2T = PVA + 5% nanocellulose TEMPO + 2% glycerol; PN5G2T = PVA + 5% nanocellulose TEMPO + 2% glycerol.

The lowest ΔE value was found in the PN3G2T film sample (PVA with the addition of 2% glycerol, 3% nano cellulose TEMPO) of 0.92 ± 0.06 while the highest ΔE value was found in the PN5G2T film sample (PVA with the addition of 5% tempo kenaf nanocellulose, and 2% glycerol), namely of 1.53 ± 0.08 . However, there is no statistically significant difference. Fukuzumi et al. (2009) state that fibres that go through the TEMPO oxidation process will be more easily fibrillated and produce nano size with good dispersion for better transparency. The appearance of the PVA film nanocomposite produced in this study can be seen in Figure 4.

3.5 Water vapour permeability (water vapour transmission rate/WVTR)

The WVTR test results in Table 2 show that the addition of nanocellulose can reduce the WVTR value. Adding nanocellulose at a concentration of 5% resulted in a lower WVTR value. Pereira et al. (2014) added 5% banana stem nanocellulose to PVA and produced a nanocomposite with a lower WVTR than without nanocellulose. Nafchi et al. (2013) reported that nanoparticles added to the polymer will fill the molecular structure, thereby reducing water vapour transmission.

Figure 4 Nanocomposite film PVA. P = PVA film; PN3G0 = PVA + 3% nanocellulose; PN5G0 = PVA + 5% nanocellulose; PN3G0T = PVA + 3% nanocellulose TEMPO; PN5G0T = PVA + 5% nanocellulose TEMPO; PG2 = PVA + 2% glycerol; PN3G2 = PVA + 3% nanocellulose + 2% glycerol; PN5G2 = PVA + 5% nanocellulose + 2% glycerol; PN3G2T = PVA + 3% nanocellulose TEMPO + 2% glycerol; PN5G2T = PVA + 5% nanocellulose TEMPO + 2% glycerol;



The cellulose type factor did not significantly affect the WVTR value. Kumar et al. (2014) stated that the composition of the film surface influenced the WVTR value. The number of hydroxyl groups on the film will increase the WVTR value. Based on the FTIR results in Figure 1, it is known that there is no significant change in the number of hydroxyl groups both before and after the modified TEMPO oxidation reaction. This causes no significant change in the WVTR value. The number of carboxylic groups whose number did not change significantly also caused the WVTR value to be similar.

Films treated with 2% glycerol addition increased the WVTR value. This is evidenced by analysis of variance; Duncan's difference test found that the treatment combination with the addition of glycerol had a significant mean value or significantly different at the 5% level of significance compared to the film without the addition of glycerol. Pudjiastuti et al. (2012) also found a decrease in the barrier properties of nanocomposites added with plasticisers. Glycerol is hydrophilic and can reduce intermolecular tension in the film matrix, causing the intermolecular space to be larger and water vapour to easily penetrate the film. The addition of glycerol also causes a decrease in internal hydrogen bonds and an increase in intermolecular distance, which causes an increase in the rate of water vapour transmission.

3.6 Light transmission (%)

The level of transparency can be distinguished manually by placing the film in front of the object (Li et al., 2013). The more transparent a film, the more precise the object behind it is; conversely, the appearance of a blurred object indicates a low level of film transparency. The addition of nanocellulose causes the value of light transmission on the nanocomposite film to decrease. However, there was no significant difference in light transmission values between nanocomposite films with the addition of 3% and 5% nanocellulose. The decrease in the value of light transmission is due to a filler in the form of nanocellulose, which is fused with the polymer matrix to form irregular webs; the more nanocellulose is added, the more dense the nets will be. This makes it difficult for

light to penetrate the composite layer. In addition, nanocellulose has a more turbid colour, so adding the film matrix will reduce the transparency value.

The addition of different types of nanocellulose affects the value of light transmission. The film with the addition of nanocellulose through the modified TEMPO oxidation step has a lower light transmission value than the film with the addition of nanocellulose without going through the TEMPO oxidation. This result is not in line with Fukuzumi et al. (2009), who state that fibres that undergo the TEMPO oxidation process will be more easily fibrillated and produce nano-sizes with good dispersion so that they have better light transmission. This is because the nanocellulose with TEMPO oxidation in this study has a dispersion level that is not optimal in the PVA film, which makes it harder for light to penetrate the film. Light transmission is highly dependent on the dispersion ability of nanocellulose in the film (Li et al., 2013).

The addition of 2% glycerol does not affect the light transmission value of the resulting film. This is because the added glycerol is in the form of a transparent gel. The addition of glycerol in a small amount also did not change the light transmittance value of the film.

3.7 Mechanical characteristics

3.7.1 Tensile strength

The factors that significantly affected the film's tensile strength in this study were the amount of nanocellulose, the type of nanocellulose, and the addition of 2% glycerol. The results of the tensile strength of the sample in this study can be seen in Figure 5.

In Figure 5, it can be seen that adding nanocellulose to the PVA film can increase the tensile strength value. Based on the results of Duncan's different test with = 5%, the concentration factor of nanocellulose with a level of 5% significantly affected the tensile strength value. Iriani et al. (2015) produced a tensile strength of 20-30 MPa for nanocomposites by adding 40% pineapple fibre nanocellulose without adding glycerol. Nanocellulose can produce a large surface area, reducing the number of cavities that make up the film.

Nanocellulose with TEMPO has a significant effect on the value of tensile strength. Nanocomposite films with nanocellulose added through a modified TEMPO oxidation process have a higher tensile strength value. Chaabouni and Sami (2017) stated that the increase in the tensile strength value of the film with the addition of TEMPO nanocellulose also indicated a better dispersion level and a strong interaction between nanocellulose and the film matrix.

Adding a plasticiser in the form of 2% glycerol to the film affects its film's tensile strength. The value of the film's tensile strength with the addition of glycerol is lower than the value of the film's tensile strength without the addition of glycerol. Iriani et al. (2015) produced nanocomposites by adding 1% glycerol, reducing the tensile strength by half. Adding glycerol will reduce the intermolecular tension that composes the film matrix so that the film will be weaker against mechanical treatment. In addition, glycerol will reduce the stability of the solid dispersion system, resulting in weak physical properties (Huri and Fithri, 2014). Increasing glycerol will also reduce intermolecular interactions and increase polymer chain movement so that the tensile strength decreases (Chen et al., 2008).

Figure 5 Effect of TEMPO treatment and addition of glycerol on the tensile strength of nanocomposites. The difference between the letters A-E in the same column shows a significant difference in Duncan's post hoc test with = 5%. P = PVA film; PN3G0 = PVA + 3% nanocellulose; PN5G0 = PVA + 5% nanocellulose; PN3G0T = PVA + 3% nanocellulose TEMPO; PN5G0T = PVA + 5% nanocellulose TEMPO; PG2 = PVA + 2% glycerol; PN3G2 = PVA + 3% nanocellulose + 2% glycerol; PN5G2 = PVA + 5% nanocellulose TEMPO + 2% glycerol; PN5G2T = PVA + 5% nanocellulose TEMPO + 2% glycerol; PN5G2T = PVA + 5% nanocellulose TEMPO + 2% glycerol; PN5G2T = PVA + 5% nanocellulose TEMPO + 2% glycerol



The results of the combination test of these three factors (glycerol, amount of nanocellulose, and type of nanocellulose) using SAS 9.2 software showed that the best treatment with the highest tensile strength value (48,003 MPa) was nanocomposite with 5% nanocellulose addition, modified TEMPO nanocellulose type, and without addition glycerol.

3.7.2 Elongation

The addition of cellulose fibres in the form of nano reduces the elongation value Figure 6. The nanocellulose fibres evenly distributed in the film matrix cause stronger interactions so that the percent elongation will decrease. Meanwhile, 5% nitrocellulose and glycerol addition at a 2% level showed the highest elongation value compared to other combinations. Ramezani et al. (2014) added nanocellulose from aloe vera fibre in PVA. They resulted in an elongation value that decreased from 165% (without adding nanocellulose) to 6.6% (with the addition of nanocellulose as much as 2%). The intense interaction between nanocellulose and the film matrix did not allow PVA to elongate. The results of elongation of the sample in this study can be seen in Figure 5.

200 S. Aminah et al.

Figure 6 Effect of tempo treatment and addition of glycerol on the elongation of nanocomposites. The difference between the letters A-E in the same column shows a significant difference in Duncan's post hoc test with = 5%. P = PVA film; PN3G0 = PVA + 3% nanocellulose; PN5G0 = PVA + 5% nanocellulose; PN3G0T = PVA + 3% nanocellulose TEMPO; PN5G0T = PVA + 5% nanocellulose TEMPO; PG2 = PVA + 2% glycerol; PN3G2 = PVA + 3% nanocellulose + 2% glycerol; PN5G2 = PVA + 5% nanocellulose + 2% glycerol; PN3G2T = PVA + 3% nanocellulose TEMPO + 2% glycerol; PN5G2T = PVA + 5% nanocellulose TEMPO + 2% glycerol; PN5G2T = PVA + 5% nanocellulose TEMPO + 2%



Combination treatments in producing nanocomposite

The type of nanocellulose did not affect the elongation value. Films with TEMPO nanocellulose had values that were not significantly different at the 5% level from films added with nanocellulose without TEMPO. Soni et al. (2016) stated that nanocellulose, through the TEMPO oxidation step, can increase the film's strength without affecting its flexibility.

On the other hand, the addition of glycerol affects the elongation value. Films with the addition of 2% glycerol have a higher elongation value. This is because the hydrophilic glycerol binds to the OH groups on the film matrix, thereby enhancing the flexibility of the film. Huri and Fithri (2014) added that polyols such as glycerol function effectively as plasticisers based on their ability to reduce internal hydrogen bonds by increasing the free space between molecules, thereby reducing stiffness and film flexibility. Glycerol is widely used as a plasticiser because of its small size and high polarity.

The results of the combination test of these three factors (glycerol, the amount of nanocellulose, and the type of nanocellulose) using SAS 9.2 software showed that the treatment that had the highest elongation (249,007%) was nanocomposite film with the addition of 5% nanocellulose, TEMPO type nanocellulose, with the addition of 2% glycerol.

In general, this research was successful in producing kenaf nanocellulose. Nanocellulose kenaf added to PVA film can improve physical and mechanical characteristics. The addition of glycerol causes an increase in the elongation value. However, further research is needed. Studies regarding the shelf life of nanocomposites and how they are applied as food packaging need to be carried out.

4 Conclusion

Adding nanofiber cellulose isolated from kenaf plants can maintain and increase PVA film's physical and mechanical characteristics. The addition of cellulose nanofiber increased the value of tensile strength. Meanwhile, the addition of glycerol gave a lower tensile strength value. Adding nanofiber cellulose as much as 5% without adding glycerol gave the best result on tensile strength value. The addition of nanofiber cellulose can reduce the value of elongation. The highest elongation value in the nanocomposite films with cellulose nanofiber is 5% and 2% glycerol.

Conflict of interest

All authors declare that they have no conflict of interest.

References

- Abe, K., Shinichiro, I. and Hiroyuki, Y. (2007) 'Obtaining cellulose nanofibers with a uniform width of 15 nm from wood', *Biomacromolecules*, Vol. 8, pp.3276–3278.
- Akhtar, N., Abu, S., Muhammad, N., Khalid, M., Khairul, R., Nur, I. and Muhammad, R. (2017) 'Kenaf – biocomposites: manufacturing, characterization, and applications', *Green Biocomposites*, Vol. 10, pp.225–254.
- Ben, Z.Y., Hayati, S. and mohd, F.Y. (2022) 'Glycerol: Its properties, polymer synthesis, and applications in starch based films', *European Polymer Journal*, Vol. 175, pp.111377.
- Chaabouni, O. and Sami, B. (2017) 'Cellulose nanofibrils/polyvinyl acetate nanocomposite adhesives with improved mechanical properties', *Carbohydrate Polymers*, Vol. 156, pp.64–70.
- Chaker, A., Mutje, P., Vilar, M.R. and Boufi, S. (2014) 'Reinforcing potential of nanofibrillated cellulose from nonwoody plants', *Polymer Composite*, Vol. 34, pp.1999–2007.
- Chandra, J., Neena, G.S. and Narayanankutty, K. (2016) 'Isolation and characterization of cellulose nanofibrils from arecanut husk fibre', *Carbohydrate Polymers*, Vol. 16, pp.30–38.
- Chen, Y., Cao, X., Chang, P. and Huneault, M. (2008) 'Comparative study on the films of poly (Vinyl alcohol)/Pea starch nanocrystals and poly (Vinyl alcohol)/native pea starch', *Carbohydrate Polymer*, Vol. 73, pp.8–17.
- Faruk, O., Andrzej, K., Bledzki, Hans-Peter, F. and Mohini, S. (2012) 'Biocomposites reinforced with natural fibers: 2000-2010', *Progress in Polimer Science*, Vol. 37, pp.1552–1596.
- Fortunati, E., Peltzer, M., Armentano, I., Torre, L., Jiménez, A. and Kenny, J. (2012) 'Effects of modified cellulose nanocrystals on the barrier and migration properties of PLA nanobiocomposites', *Carbohydrate Polymers*, Vol. 90, pp.948–956.
- Fukuzumi, H., Tsuguyuki, S., Tadahisa, I., Yoshiaki, K. and Akira, S. (2009) 'Transparent and high gas barrier films of cellulose nanofibers prepared by TEMPO-mediated oxidation', *Biomacromolecules*, Vol. 10, No. 1, pp.162–165.
- Hafizah, N.A., Hussin, M.W., Jamaludin, M.Y., Bhutta, M.A., Ismail, M. and Azman, M. (2014) 'Tensile behaviour of kenaf fiber reinforced polymer composites', *Jurnal Teknologi UTM*, Vol. 69, No. 3, pp.11–13.
- Ibrahim, M.M., Waleed, K.E. and Mona, A.N. (2010) 'Synthesis and characterization of polyvinyl alcohol/nanospherical cellulose particle films', *Carbohydrate Polymers*, Vol. 79, pp.694–699.
- Ioelovich, M. (2012) 'Optimal conditions for isolation of nanocrystalline cellulose particles', Nanoscience and Nanotechnology, Vol. 2, No. 2, pp.9–13.

- Iriani, E., Kendri, W., Titi, C. and Asep, W. (2015) 'nanocellulose synthesis from pineapple fiber and its applications as nanofiller in polyvinil alcohol-based film', *Jurnal Penelitian Pascapanen Pertanian*, Vol. 12, No. 1, pp.11–19.
- Kazon, P., Manuel, V. and Gonzalo, V. (2018) 'Cellulose-glycerol-polyvinyl alcohol composite films for food packaging: evaluation of water adsorption, mechanical properties, light-barrier properties and transparency', *Carbohydrate Polymers*, Vol. 195, pp.432–443.
- Ketabchi, M.R., Khalid, M., Hoque, M.E., Ratnam, C.T. and Walvekar, R. (2015) 'Eco-friendly and cost-effective isolation of cellulose microfibres and nano-crystals from kenaf fibres', *Proceedings, 13th international conference on environment, ecosystems and development* (*EED 2015*), pp.23–25.
- Khan, A., Sapuan, S.M., Vasi, U.S., Zainudin, E.S., Zuhri, M.Y.M. and Harussani, M.M. (2023) 'A review of recent developments in kenaf fiber/polylactic acid composites research', *International Journal of Biological Macromolecules*, Vol. 253, p.127119.
- Khawas, P. and Deka, S.C. (2016) 'Isolation and characterization of cellulose nanofibers from culinarybanana peel using high-intensity ultrasonication combined with chemical treatment', *Carbohydrate Polymers*, Vol. 137, pp.608–616.
- Kumar, V., Roger, B., Arthas, Y., Qingxia, C., Gang, C., Pekka, S., Douglas, B. and Martti, T. (2014) 'Comparison of nano-and microfibrillated cellulose films', *Cellulose*, Vol. 21, pp.3443–3456.
- Li, W., Jinquan, Y. and Shouxin, L. (2012) 'Preparation of nanocrystalline cellulose via ultrasound and its reinforcement capability for poly(vinyl alcohol) composites', *Ultrasonic Sonochemistry*, Vol. 19, pp.479–485.
- Li, W., Zhao, X., Huang, Z. and Liu, S. (2013) 'Nanocellulose fibrils isolated from BHKP using ultrasonication and their reinforcing properties in transparent poly (vinyl alcohol) films', *Journal Polymer Research*, Vol. 20, pp.2–7.
- Liu, D., Sun, X., Tian, H. and Ma, Z. (2014) 'Effects of cellulose nanofibrils on the structure and properties on PVA nanocomposites', *Cellulose*, Vol. 20, No. 6, pp.2981–2989.
- Ma, Q., Lin, D., Yang, Y. and Lijuan, W. (2016) 'Rheology of film-forming solutions and physical properties of tara gum film reinforced with polyvinyl alcohol (PVA)', *Food Hydrocolloids*, Vol. 63, pp.677–684.
- Nafchi, A.M., Alias, A.K., Mahmud, S. and Robal, M. (2013) 'Antimicrobial, rheological, and physicochemical properties of sago films filled with nanorod-rich zinc oxide', *Journal of Food Engineering*, Vol. 113, pp.511 519.
- Oun, A., Gye, H.S., Jong, W.R. and June, Kim, T. (2022) 'Recent advances in polyvinyl alcoholbased composite films and their applications in food packaging', *Food Packaging and Shelf Life*, Vol. 34, pp.100991.
- Panda, P.K., Kambiz, S. and Jongchul, S. (2022) 'Recent advances in poly (vinyl alcohol)/natural polymer based films for food packaging applications: a review', *Food Packaging and Shelf Life*, Vol. 22, p.100904.
- Pereira, A.L.S., Do, N., Morais, J.P.S., Vasconcelos, N.F. and Feitosa, J.P. (2014) 'Improvement of polyvinyl alcohol properties by adding nanocrystalline cellulose isolated from banana pseudostems', *Carbohydrate Polymers*, Vol. 112, pp.165–172.
- Pudjiastuti, W., Arie, L. and Sudirman (2012) 'Polymer nano-composite (1) master batch for biodegradable food packaging', *Jurnal Riset Industri*, Vol. 6, No. 1, pp.51–60.
- Qiu, K. and Netravali, A.N. (2012) 'Fabrication and characterization of biodegradable composites based on microfibrillated cellulose and polyvinyl alcohol', *Composite Science and Technology*, Vol. 72, pp.1588–1594.
- Ramezani, C.K.S., Sain, M. and Asiri, A. (2014) 'Mechanical, thermal, and morphological properties of nanocomposites based on polyvinyl alcohol and cellulose nanoselulosa from aloe vera rind', *Journal of Nanomaterials*, Vol. 2014, pp.1–7.
- Saito, T., Satoshi, K., Yoshiharu, N. and Akira, I. (2007) 'Cellulose nanofibers prepared by TEMPO-mediated oxidation of native cellulose', *Biomacromolecules*, Vol. 8, pp.2485–2491.

- Savadekar, N. and Mhaske, S. (2012) 'Syntesis of nanocellulose fibers and effect on thermoplastics starch based films', *Carbohidrate Polymers*, Vol. 89, pp.146–151.
- Silverio, H., Neto, W. and Pasquini, D. (2013) 'Effect of incorporating cellulosa nanocrystals from corncob on tensile, thermal, dan barrier properties of polyvinyl alcohol nanocomposites', *Journal of Nanomaterials*, Vol. 2013, No. 74, pp.1–9.
- Soni, B., Barbary, H., Wes, S. and Barakat, M. (2016) 'Transparent bionanocomposite films based on chitosan and TEMPO-oxidized cellulose nanofibers with enhanced mechanical and barrier properties', *Carbohydrate Polymer*, Vol. 151, pp.779–789.
- Tian, H., Jiaan, Y., Varada, R., Aimin, X. and Xiaogang, L. (2017) 'Fabrication and properties of polyvinyl alcohol/starch blend films: effect of composition and humidity', *International Journal of Biological Macromolecules*, Vol. 96, pp.518–523.
- Zhang, L., Zhong, J. and Ren, X. (2017) 'Natural fiber-based biocomposites', in Jawaid, M., Sapuan, S. and Alothman, O. (Eds.): Green Biocomposites. Green Energy and Technology, Springer, Cham, pp.31–70.
- Zhou, Y.M., Fu, S.Y., Zheng, L.M. and Zhan, H.Y. (2012) 'Effect of nanocellulose isolation techniques on the formation of reinfoced poly(vinyl alcohol) nanocomposite film', *Experimental Polymer Letter*, Vol. 6, No. 10, pp.794–804.