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Abstract

The objective of this study is to produce nanocellulose sheets from oil palm empty fruit bunches (OPEFBs). Nanocellulose sheets from OPEFBs were prepared through the dewatering process using vacuum filtration. Subsequently, the produced sheets were treated with NaOH solution to improve the mechanical stability of their structure, causing them to be firm in water. The water absorption of nanocellulose sheet increased with increasing NaOH concentrations up to 20 wt%, increasing tensile strength, and Young's modulus, but decreased when they were treated with 30 wt%. The diameter shrinkage of nanocellulose sheets was directly proportional to the NaOH concentration and soaking time. The images of scanning electron microscope show that the porous structure of the nanocellulose sheets appeared swollen when using NaOH. NaOH treatment above 10 wt% could change the crystalline form from cellulose I to cellulose II. The tensile strength and Young's modulus of nanocellulose sheets could improve with NaOH treatment. The optimum tensile strength and Young's modulus were achieved in a six-hour immersion process with 20 wt% NaOH.

Keywords

Alkali treatment, Cellulose nanofiber, Nanocellulose, Oil palm empty fruit bunch (OPEFB)

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Cover Page Footnote

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1. Introduction

Indonesia is one of the largest crude palm oil (CPO) producers in the world, alongside Malaysia, producing more than 31.28 million tons in 2012 [1]. With such CPO production, waste in the form of oil palm empty fruit bunches (OPEFBs) is produced in abundance every year, which is a problem for the environment. The OPEFBs contain 37.3%–46.5% of cellulose, 25.3%–33.8% of hemicellulose, and 27.6%–32.5% of lignin [2]. Exploration of the use of OPEFBs is interesting and increases their value.

Cellulose is a biopolymer, abundantly found in nature and is explored for its use in the paper industry. It also has specific properties that are different from synthetic polymers and other natural polymers, including biodegradability, hydrophilicity, chirality, versatile semicrystalline fiber morphologies, and broad chemical modifying capacity [3–5].

Cellulose is a homopolysaccharide formed by the repetition of D-glucopyranose units or anhydroglucose units (AGU) joined together by (1–4) glycosidic linkages [6]. The three hydroxyl groups in each AGU allow the cellulose chain to converge to form fibrillar species. Individual cellulose molecules are combined into larger units, namely elementary fibrils or microfibrils, which have a nanometer-sized diameter. In general, we can also call cellulose-based elementary fibrils or microfibrils with nanocellulose, which can be produced by mechanical (cellulose nanofibers/CNF) or chemical treatments (cellulose nanocrystals/CNC) [7–14].

Nanocelluloses have superior physical and mechanical properties, such as high aspect ratio, high surface-to-volume area, entangled web-like structure, high Young's modulus along the longitudinal direction (138 GPa in the crystal region), and very low longitudinal thermal expansion coefficient (10^{-7} K^{-1}) [15]. We have successfully isolated nanocelluloses from OPEFBs with an average diameter of $27.23 \pm 8.21 \text{ nm}$ and a length of several micrometers using mechanical treatment [10,12,16]. Shanmugarajah et al. [17] also succeeded in isolating NCC from OPEFBs using 64% (w/w) H_2SO_4 at 40°C . The diameter of the resulting NCC was approximately 499.2 nm. Meanwhile, Lani et al. [18] successfully prepared NCC with diameters ranging 4–15 nm by 64 wt% sulfuric acid hydrolysis.

Cellulose chains are packed closely by both inter-molecular and intra-molecular hydrogen bonds.

Meanwhile, alkali treatment is reported to increase the tensile strength of plant fibers [19]. Not many publications explain the production of nanocellulose sheets from biomass using a NaOH treatment. Abe and Yano [20] report that a simple NaOH solution treatment produces a nanocellulose sheet-based hydrogel from Radiata Pine. The obtained sheets had a swelling degree and tensile strength of 12.1 and 3.0 MPa, but 13.4 and 5.4 MPa when treated with NaOH 9 and 15 wt%, respectively. This NaOH treatment is also known as maceration. Zhang et al. [21] also used maceration to prepare nanocelluloses sheet-based hydrogel from bamboo.

The maceration of natural fibers using NaOH has been widely studied by researchers around the world. Maceration can be used to remove pectin, lignin, hemicellulose, and natural oils covering the outer surface of the fibers. This process can also be used to modify the surface of natural fibers to increase roughness, break some microfibril bundles, and defibrillate and individualize microfibrils [22,23]. Alkali treatment also improves the tensile strength of plant fibers [24,25].

In this study, we prepare nanocellulose sheets from OPEFBs and treat them with an NaOH solution. We explore the effects of NaOH solution on the properties of the obtained nanocellulose sheets.

2. Materials and methods

OPEFBs were supplied by PTPN VIII in West Java, Indonesia. The chemicals used were sodium hydroxide (NaOH/99.6% purity), sodium hypochlorite (technical grade), potassium hydroxide (KOH/86.9% purity), and acetic acid (100% purity). All chemicals were from Merck, Germany. The OPEFBs were cut into small pieces with a length of 0.5–1 cm and subsequently boiled in water (100°C) for 1 h to remove impurities. The pieces were then dried in an oven at 55°C for 2 days. The dried OPEFBs were soaked in 6% KOH solution at room temperature for 12 h and then washed with deionized water. The fibers were then bleached for 5 h using a sodium hypochlorite solution and rinsed with deionized water until they reached a neutral pH (pH 7).

The cellulose fibers were dispersed in deionized water using a warring blender. The concentration of cellulose suspension was diluted to 2 wt%. The suspension was passed through an ultrafine grinder 37

times (Masuko Sangyo Co., Ltd.) operating at 1500 RPMs. The suspension was then sonicated using an ultrasonic homogenizer (amplitude of 40%, 41 W) for 30 min to liberate nanocelluloses.

A nanocellulose sheet was prepared by vacuum filtration of cellulose nanofibers suspension using a polytetrafluoroethylene (PTFE) membrane. The obtained wet sheets were immersed in NaOH solution with a concentration of 5, 10, 20, or 30 wt% at 50 °C for 6 or 12 h. Afterward, the sheets were neutralized in diluted acetic acid (2 wt%) and washed with deionized water.

The morphology of nanocelluloses and their sheets were analyzed by scanning electron microscopy (SEM, Zeiss EVO MA10). Changes in crystal form of the nanocellulose sheets due to NaOH treatment were evaluated by X-ray diffraction (XRD). The measurements were carried out on an X-ray diffractometer (Bruker D8 Advance) equipped with a Cu K α radiation source operated at 40 kV and 35 mA. The samples for XRD were prepared as freeze-dried sheets. The diameter shrinkage of obtained nanocellulose sheets was measured with a caliper, as described in a previous report [16]. Meanwhile, the water absorption of each nanocellulose sheet was defined as the ratio between the weight of the swollen sheet and the weight of the freeze-dried sheet.

3. Results and discussion

The cellulose nanofibers obtained in this study had a mean diameter of 26.8 ± 5.1 nm and several micrometers in length (Fig. 1). The length of the cellulose nanofibers could not be determined with certainty because the ends of the fibers could not be detected in the SEM images. In general, cellulose nanofibers

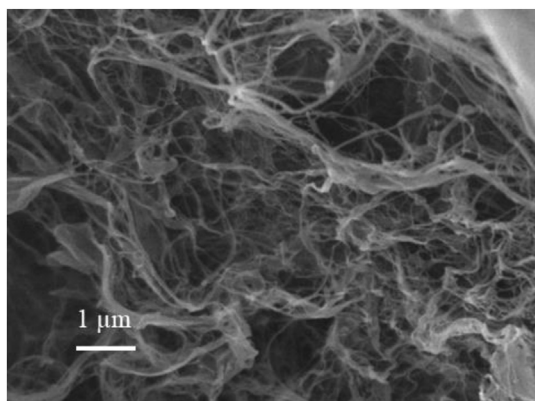


Fig. 1. Cellulose nanofibers from OPEFBs.

obtained by mechanical treatment are several micrometers long [10,26–28]. This is different from cellulose nanofibers obtained by chemical treatment, which generally range from 100 to 500 nm long [29–31].

The nanocellulose sheets were formed through the dewatering process using vacuum filtration and a PTFE membrane. Without NaOH treatment, the nanocellulose sheets after the dewatering process were easily ruined in water. Non-immersive nanocellulose sheets in NaOH solution were extremely fragile when given little pressure. Fig. 2 shows that the obtained nanocellulose sheets were white and exhibited a gel texture. Immersing the sheets in an NaOH solution made them durable. Gel-like sheets were formed by molecular self-assembly through ionic or hydrogen bonds [32].

Diffusion occurred through osmosis when the nanocellulose sheets were soaked in a NaOH solution. Due to the high concentration of NaOH, the OH⁻ ions from the NaOH solution penetrated the nanocelluloses and swelled them. The concentration of the NaOH solution affected the texture of the nanocellulose sheets. The higher concentration of NaOH solution resulted in a more rigid texture. The sheets soaked in NaOH solution with a concentration of 5 or 10% remained flat; in contrast, the sheets soaked in NaOH solution with a concentration of 20 or 30% curled and wrinkled (Fig. 2).

Fig. 3 indicates that the diameter shrinkage of nanocellulose sheets was directly proportional to the concentration of the NaOH solution. Prolonged soaking also induced this effect. The sheets soaked for 12 h exhibited greater diameter shrinkage than the sheets soaked for 6 h. Thus, the diameter shrinkage correlated with the soaking time. Diameter shrinkage occurred because of the diffusion process from high concentration (NaOH solution) into low concentration (inside the nanocellulose sheets). The diffusion was accelerated by an increase in temperature, which could increase the entropy–elastic force, thus causing a longitudinal contraction. Therefore, longitudinal shrinkage occurs. This result was consistent with the result reported by Abe and Yano [20]: an increase in the concentration of NaOH increased the diameter shrinkage.

Fig. 4 exhibits the SEM images of the cross-section of the freeze-dried nanocellulose sheets. The control sheet (non-immersive nanocellulose sheet in NaOH) was densest among all sheets. Even without immersion in NaOH, the control sheet seemed to have a porous structure (Fig. 4a) and the nanocellulose did not appear to swell (Fig. 4b). The sheet-making method seemed to

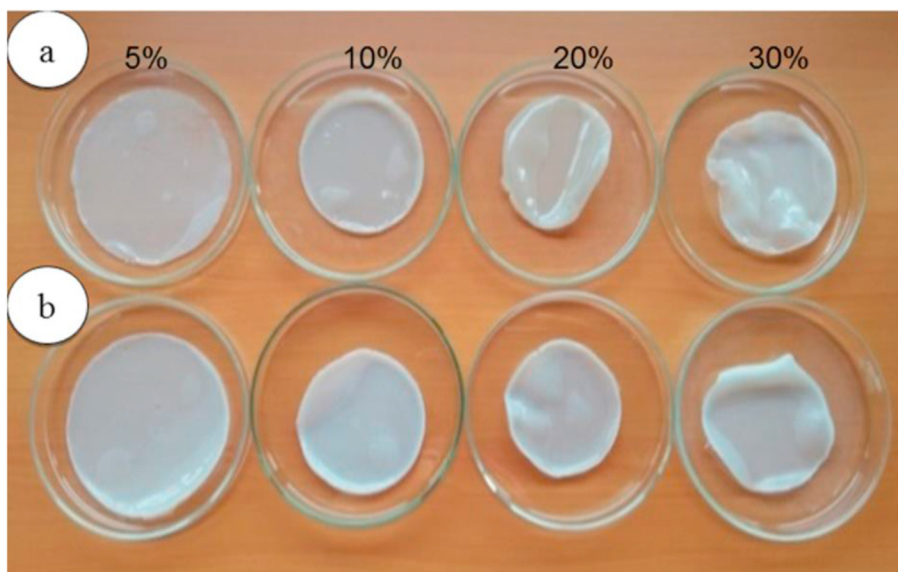


Fig. 2. Nanocellulose sheets after NaOH treatment for (a) 6 h and (b) 12 h in 5, 10, 20, and 30 wt% of NaOH solution (left–right).

affect the structure of the sheet produced. In this study, the nanocellulose sheets were manufactured by the vacuum filtration method. This filtration process might cause the porous structures of the obtained sheets. With immersion in the NaOH solution, the porous structure of nanocellulose sheets appeared swollen (Fig. 4c–j). The swelling increased as the NaOH solution concentration increased (up to 20 wt%) and decreased with 30 wt% NaOH concentration. According to Oushabi et al. [33], an increase in the percentage of NaOH could increase the number of pores on the fiber surface. However, this is not clearly seen in SEM images. From Fig. 4j, the structure of nanocellulose sheets was damaged from the severe reaction with 30 wt% NaOH.

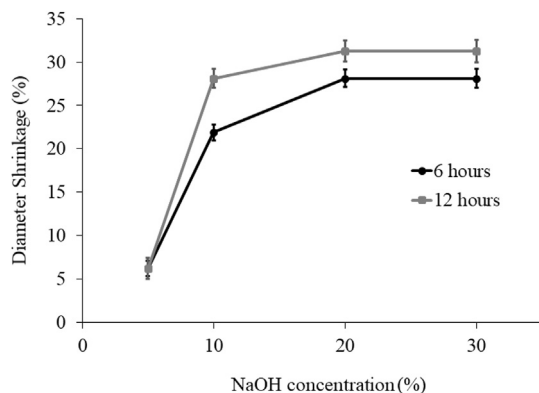


Fig. 3. Diameter shrinkage of nanocellulose sheets treated with different NaOH concentrations.

The damage had an impact on the ability of nanocellulose sheets to absorb water. The high NaOH concentration (30 wt%) was also thought to destroy a small crystalline region of the sheet, which caused a decrease in crystallinity after treatment with 30 wt% NaOH (Table 1).

Water absorption is an important parameter in assessing the ability of a sheet to retain water or solution in its structure. Fig. 5 shows the water absorption of nanocellulose sheets soaked for different times in NaOH solutions with different concentrations. This figure shows that the sheets soaked for 6 and 12 h exhibited the same tendencies. The water absorption increased with increasing concentration of NaOH, up to 20 wt%. Thereafter, it decreased at a concentration of 30 wt%. These results were consistent with SEM observations where the pore size appeared to shrink on nanocellulose sheets soaked in 30 wt% NaOH.

Alkali treatment with a concentration of up to 20% could swell the porosity of the nanocellulose structure so that it could optimally absorb water. Pores in the nanocellulose structure were able to hold water molecules. However, water absorption decreased with 30 wt% NaOH. The 30 wt% alkali treatment caused a break in hydrogen bonds in the nanocellulose structure, forming a hydrophobic group [34].

Fig. 6 shows that high-concentration alkaline treatments could alter the crystalline structure of native nanocellulose. Dipping nanocellulose sheets in an NaOH solution with a concentration above 10% could

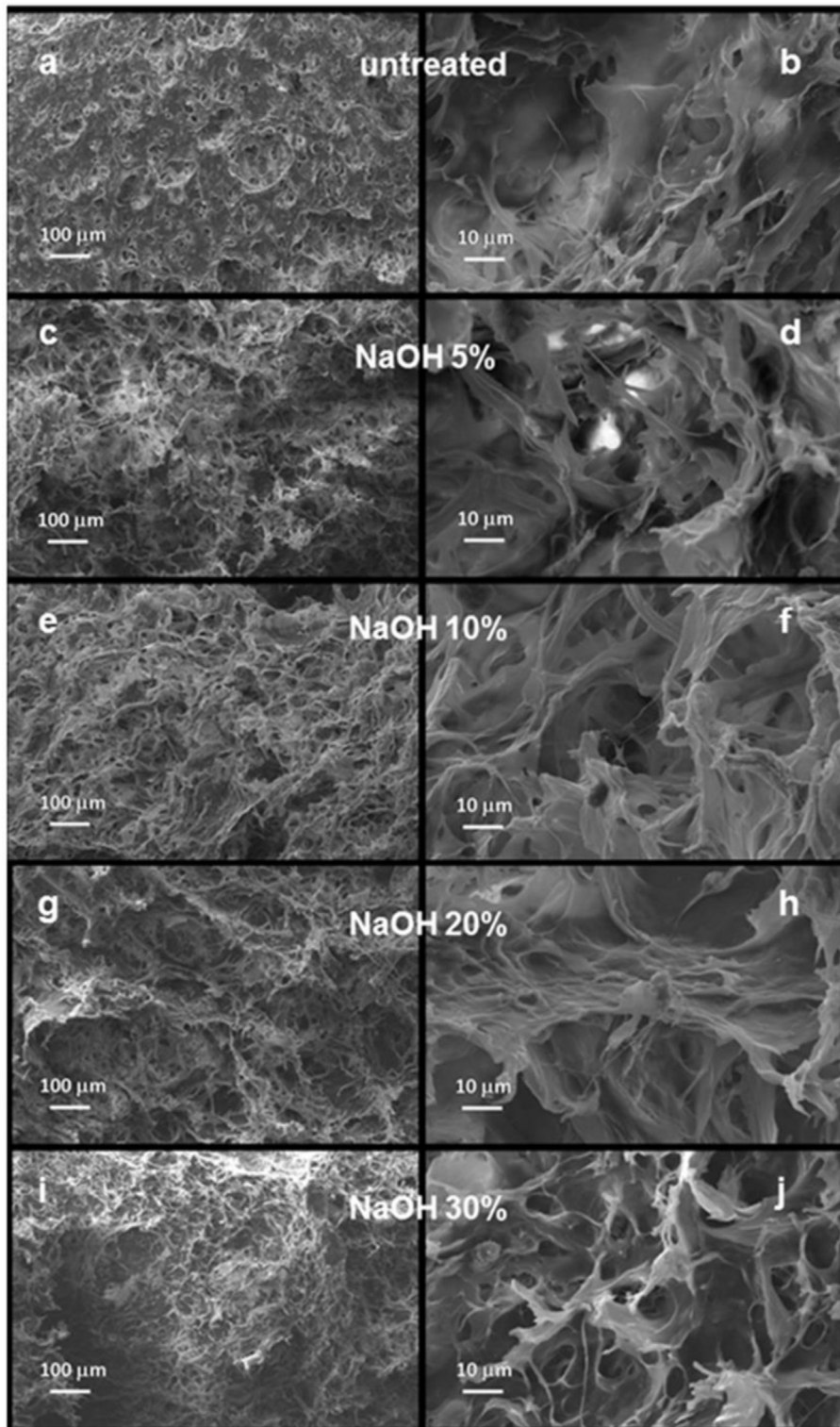


Fig. 4. Morphology of freeze-dried nanocellulose sheets treated with different NaOH concentrations for 6 h.

Table 1
Crystallinity of nanocellulose sheet treated with different NaOH concentrations.

Sample treated with-	Crystallinity (%)
Untreated	66.4 ± 0.8
5 wt% NaOH	50.1 ± 1.2
10 wt% NaOH	54.4 ± 1.1
20 wt% NaOH	75.8 ± 1.5
30 wt% NaOH	68.2 ± 0.9

change the crystalline form from cellulose I to cellulose II [35]. Cellulose I (native cellulose) occurs naturally and is structured in parallel strands without any intersheet hydrogen bonds. Meanwhile, cellulose II is thermodynamically more stable and exists in an anti-parallel strain with hydrogen bonds between sheets. The differences in the properties of cellulose I and II arise from the changes in the crystal structure [36]. The crystallinity of nanocellulose sheets with cellulose II was higher than those with cellulose I. During maceration, the alkaline solution enters the amorphous region of the nanocelluloses, which is the interface region between the crystallites. The NaOH solution may affect the crystalline region even with slight segment movement of cellulose chains. Alkali solution, when entering into nanocelluloses, can swell the cellulose so that the mobility of segments of chains in the swollen area was high. This allowed the direction to change from the parallel cellulose (cellulose I) to anti-parallel cellulose (cellulose II) [37].

The diffraction patterns of untreated and treated nanocellulose sheets are shown in Fig. 6. Two peaks at $2\theta = 14.8^\circ$ (broad peak) and 22.3° confirm that only cellulose I existed for untreated nanocellulose sheets and sheets treated with NaOH up to 10 wt%. The

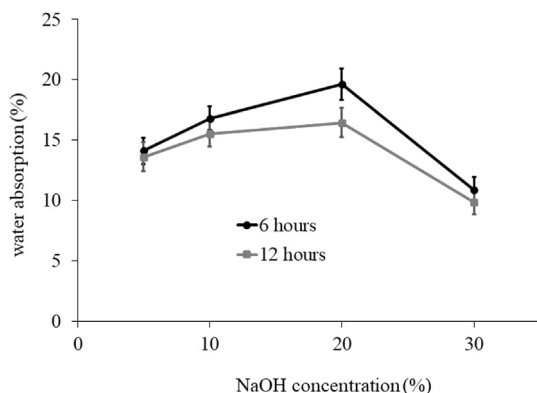


Fig. 5. Water absorption of nanocellulose sheets treated with different NaOH concentrations.

presence of two peaks in the diffraction pattern showed the material to have a low crystallinity. A cellulose material with high crystallinity generally shows a diffraction pattern with 3 peaks at around $2\theta = 14^\circ$ ($1\bar{1}0$), 16° (110) and 20° – 22° (200) [38]. In Fig. 6, it can be seen that crystallinity decreased with the NaOH treatment up to 10 wt%. In the treatment of 20 and 30 wt% NaOH, the peak with index $1\bar{1}0$ shifted to $2\theta = 12.1^\circ$ and the crystalline peak was divided into two peaks located at $2\theta = 20.4^\circ$ ($1\bar{1}0$) and $2\theta = 22.3^\circ$ ($2\bar{0}0$), indicating the formation of the cellulose II structure. Three distinct processes are known during mercerization: microfibril swelling, disruption of crystalline areas, and formation of a new crystalline lattice [39].

During maceration, the inter-fibrillar region in nanocellulose sheets tends to be less rigid, which allows for the rearrangement of cellulose nanofibers [39]. The crystalline structure of nanocellulose sheets swelled increasingly with the increasing NaOH concentrations. When the structure of the nanocellulose sheet swelled, hydroxide ions became easier to penetrate into the internal crystals and react thoroughly with cellulose nanofibers, which caused a reduced crystallinity. The hydroxide ion penetration rate is slowed by increasing the viscosity of NaOH solutions at high concentrations (20 and 30 wt%) [39]. In this study, new crystalline lattices formed with concentrations of 20% and 30% NaOH (Fig. 6). With increasing NaOH concentrations, the crystallinity of nanocelluloses increased (Table 1). Table 1 shows that nanocellulose sheets without NaOH treatment had 66.4% crystallinity. The crystallinity decreased quite dramatically to 50.1% with a treatment of 5% NaOH. Then, increasing NaOH concentration up to 20% crystallinity also increased the crystallinity but decreased it when the nanocellulose was treated by 30 wt% of NaOH. According to Mwaikambo and Ansell [40], a high crystallinity tends to produce strong and attractive fibers in the formation of plant fiber composites.

Figs. 7 and 8 and Table 2 show the tensile strength and Young's modulus of nanocellulose sheets. Both the tensile strength and Young's modulus of nanocellulose sheets increased with increasing NaOH concentration up to 20 wt%. The optimum tensile strength of sheets immersed in NaOH solution for 6 and 12 h was reported at 20 wt% NaOH, reaching 2.14 and 1.87 MPa, respectively. The tensile strength obtained in this study was smaller than that obtained by Abe and Yano [20]. They produced nanocellulose sheets from wood powder immersed in 15% NaOH for

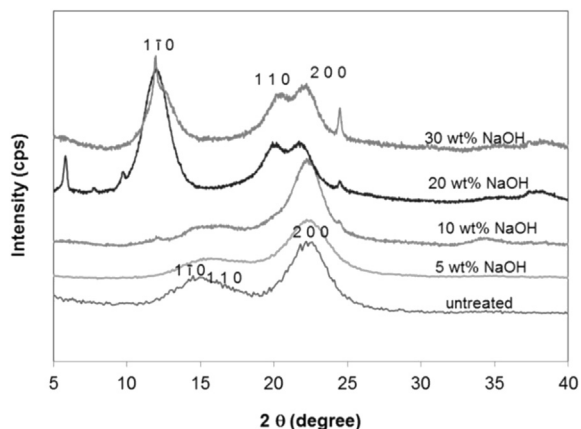


Fig. 6. X-ray profiles of nanocellulose sheets treated with different NaOH concentrations.

12 h, resulting in tensile strength of 5.4 MPa. The different values might be due to the difference in the raw material used.

The untreated nanocellulose sheet was very fragile for tensile tests. Cellulose nanofibers have a high density of –OH groups, which bond with adjacent –OH groups by hydrogen bonding. However, without NaOH treatment, the hydrogen bonds between cellulose nanofibers might not be powerful enough to support a high tensile force of nanocellulose sheets. Abe and Yano [20] reported that the nanocellulose sheet with cellulose II structure had higher tensile properties than that of cellulose I structure. The high tensile properties of the nanocellulose sheets can be attributed to their crystalline structure. While the nanocellulose sheet was treated with 30% NaOH, it was found that the tensile strength and Young's modulus decreased. This might be due to the applied

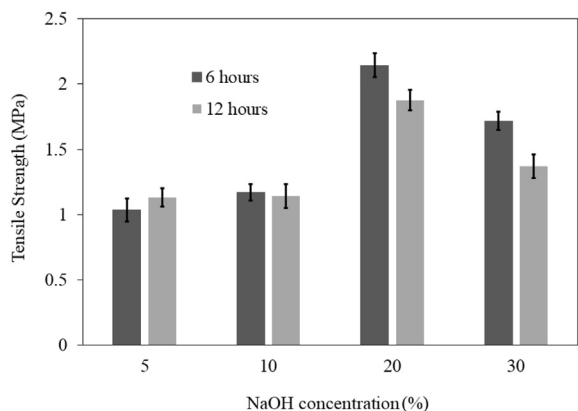


Fig. 7. Tensile strength of nanocellulose sheets treated with different NaOH concentrations.

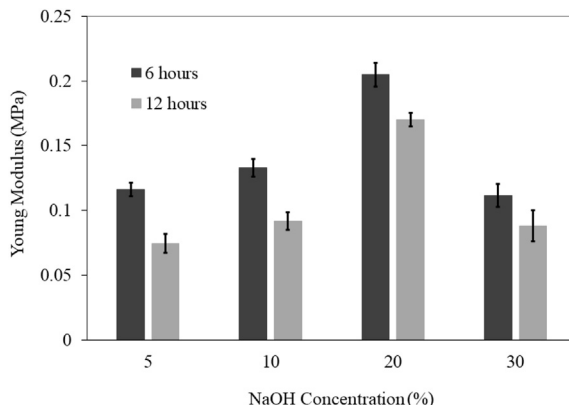


Fig. 8. Young's modulus of nanocellulose sheets treated with different NaOH concentrations.

Table 2

Recapitulate data of diameter shrinkage, water absorption, tensile strength, and Young's modulus of nanocellulose sheets.

Sample ^a	Diameter shrinkage	Water absorption	Tensile strength (MPa)	Young's modulus (MPa)
6h5%	6.25 ± 0.91	14.11 ± 1.11	1.03 ± 0.09	0.12 ± 0.005
6h10%	21.87 ± 0.95	16.81 ± 0.98	1.17 ± 0.06	0.13 ± 0.007
6h20%	28.13 ± 0.97	19.62 ± 1.31	2.14 ± 0.09	0.20 ± 0.009
6h30%	28.16 ± 1.05	10.86 ± 1.12	1.72 ± 0.07	0.11 ± 0.009
12h5%	6.25 ± 1.21	13.61 ± 1.21	1.13 ± 0.07	0.07 ± 0.007
12h10%	28.13 ± 1.16	15.52 ± 1.04	1.14 ± 0.09	0.09 ± 0.007
12h20%	31.25 ± 1.22	16.44 ± 1.24	1.88 ± 0.08	0.17 ± 0.005
12h30%	31.25 ± 1.31	9.84 ± 0.99	1.37 ± 0.09	0.09 ± 0.012

6h5%: a sample was treated for 6 h, immersed in 5% NaOH.

^a Sample notation.

high concentration that caused the crystalline structure of the nanocellulose sheets to be attacked and destroyed [33].

Recapitulate data of all properties analyzed in this study (diameter shrinkage, water absorption, tensile strength, and Young's modulus) can be seen in Table 2.

4. Conclusions

In conclusion, nanocellulose sheets prepared from the cellulose nanofibers from OPEFBs using an alkaline treatment process showed a more stable form. Higher NaOH concentrations resulted in an increase in the diameter shrinkage. The water absorption, tensile strength, and Young's modulus increased with increasing concentration of NaOH up to 20 wt%, but decreased when the nanocellulose sheet was treated with 30 wt% NaOH. By immersing in 20% NaOH solution for 6 and 12 h, the tensile strength of the

sheets were 2.14 and 1.87 MPa, respectively. This might be due to the increase of crystallinity (until 75.8%) and the change of the cellulose structure form from cellulose I to cellulose II caused by NaOH treatment. It could be concluded that the mechanical stability of nanocellulose sheets could be improved by NaOH solution treatment.

Conflicts of Interest

The authors declare no conflict of interest.

Acknowledgments

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References

- [1] Directorate General of Plantation, Statistik Perkebunan Indonesia: Kelapa Sawit, Directorate General of Estate Crops, Jakarta, 2015.
- [2] Y. Sudiyani, D. Styarina, E. Triwahyunia, Sudiyarmantoa, K.C. Sembiringa, Y. Ariatiawana, H. Abimanyua, M.H. Han, Utilization of biomass waste empty fruit bunch fiber of palm oil for bioethanol production using pilot – scale unit, *Energy Procedia* 32 (2013) 31–38, <https://doi.org/10.1016/j.egypro.2013.05.005>.
- [3] D. Klemm, D. Schumann, F. Kramer, N. Heßler, D. Koth, B. Sultanova, Nanocellulose materials – different cellulose, different functionality, *Macromol. Symp.* 280 (2009) 60–71, <https://doi.org/10.1002/masy.200950608>.
- [4] B. Priya, V.K. Gupta, D. Pathania, A.S. Singha, Synthesis, characterization and antibacterial activity of biodegradable starch/PVA composite films reinforced with cellulosic fibre, *Carbohydr. Polym.* 109 (2014) 171–179, <https://doi.org/10.1016/j.carbpol.2014.03.044>.
- [5] V.K. Suhas, P.J.M. Gupta, R. Carrott, Singh, M. Chaudhary, S. Kushwaha, Cellulose: a review as natural, modified and activated carbon adsorbent, *Bioresour. Technol.* 216 (2016) 1066–1076, <https://doi.org/10.1016/j.biortech.2016.05.106>.
- [6] Y. Habibi, L.A. Lucia, O.J. Rojas, Cellulose nanocrystals: chemistry, self-assembly, and applications, *Chem. Rev.* 110 (2010) 3479–3500, <https://doi.org/10.1021/cr900339w>.
- [7] M. Nasir, R. Hashim, O. Sulaiman, M. Asim, Nanocellulose: preparation methods and applications, in: M. Jawaid, S. Boufi, A.H.P.S. Khalil (Eds.), *Cellulose Reinforced Nanofibre Composites*, Woodhead Publishing, Oxford, 2017, pp. 261–276.
- [8] D. Klemm, E.D. Cranston, D. Fischer, M. Gama, S.A. Kedzior, D. Kralisch, F. Kramer, T. Kondo, T. Lindström, S. Nietzsche, K. Petzold-Welcke, F. Rauchfuß, Nanocellulose as a natural source for groundbreaking applications in materials science: today's state, *Mater. Today* 21 (2018) 720–748, <https://doi.org/10.1016/j.mattod.2018.02.001>.
- [9] C.H. Gómez, A. Serpa, J. Velásquez-Cock, P. Gañán, C. Castro, L. Vélez, R. Zuluaga, Vegetable nanocellulose in food science: a review, *Food Hydrocolloids* 57 (2016) 178–186, <https://doi.org/10.1016/j.foodhyd.2016.01.023>.
- [10] F. Fahma, Sugiarto, T.C. Sunarti, S.M. Indriyani, N. Lisdayana, Thermoplastic cassava starch-PVA composite films with cellulose nanofibers from oil palm empty fruit bunches as reinforcement agent, *Int. J. Polym. Sci.* 2017 (2017) 1–5, <https://doi.org/10.1155/2017/2745721>.
- [11] N. Lavoine, L. Bergström, Nanocellulose-based foams and aerogels: processing, properties, and applications, *J. Mater. Chem. A* 5 (2017) 16105–16117, <https://doi.org/10.1039/C7TA02807E>.
- [12] N. Lisdayana, F. Fahma, T.C. Sunarti, E.S. Iriani, Thermoplastic starch–PVA nanocomposite films reinforced with nanocellulose from oil palm empty fruit bunches (OPEFBs): effect of starch type, *J. Nat. Fibers* 17 (2020) 1069–1080, <https://doi.org/10.1080/15440478.2018.1558142>.
- [13] I.W. Arnata, S. Suprihatin, F. Fahma, N. Richana, Cationic modification of nanocrystalline cellulose from sago fronds, *Cellulose* 27 (2020) 3121–3141, <https://doi.org/10.1007/s10570-019-02955-3>.
- [14] D. Sartika, K. Syamsu, E. Warsiki, F. Fahma, Optimization of sulfuric acid concentration and hydrolysis time on crystallinity of nanocrystalline cellulose: a response surface methodology study, *IOP Conf. Ser. Earth Environ. Sci.* 355 (2019), 012109, <https://doi.org/10.1088/1755-1315/355/1/012109>.
- [15] W. Chen, H. Yu, Q. Li, Y. Liu, J. Li, Ultralight and highly flexible aerogels with long cellulose I nanofibers, *Soft Matter* 7 (2011) 10360–10368, <https://doi.org/10.1039/c1sm06179h>.
- [16] F. Fahma, N. Lisdayana, Z. Abidin, D. Noviana, Y.W. Sari, R.R. Mukti, M. Yunus, A. Kusumaatmaja, G.T.M. Kadja, Nanocellulose-based fibres derived from palm oil by-products and their in vitro biocompatibility analysis, *J. Text. Inst.* 111 (2020) 1354–1363, <https://doi.org/10.1080/00405000.2019.1694353>.
- [17] B. Shanmugarajah, P.L. Kiew, I.M.L. Chew, T.S.Y. Choong, K.W. Tan, Isolation of nanocrystalline cellulose (NCC) from palm oil empty fruit bunch (EFB): preliminary result on FTIR and DLS analysis, *Chem. Eng. Trans.* 45 (2015) 1705–1710, <https://doi.org/10.3303/CET1545285>.
- [18] N.S. Lani, N. Ngadi, A. Johari, M. Jusoh, Isolation, characterization, and application of nanocellulose from oil palm empty fruit bunch fiber as nanocomposites, *J. Nanomater.* 2014 (2014) 1–9, <https://doi.org/10.1155/2014/702538>.
- [19] C. Chang, L. Zhang, Cellulose-based hydrogels: present status and application prospects, *Carbohydr. Polym.* 84 (2011) 40–53, <https://doi.org/10.1016/j.carbpol.2010.12.023>.
- [20] K. Abe, H. Yano, Cellulose nanofiber-based hydrogels with high mechanical strength, *Cellulose* 19 (2012) 1907–1912, <https://doi.org/10.1007/s10570-012-9784-3>.
- [21] X. Zhang, Y. Wang, C. Lu, W. Zhang, Cellulose hydrogels prepared from micron-sized bamboo cellulose fibers, *Carbohydr. Polym.* 114 (2014) 166–169, <https://doi.org/10.1016/j.carbpol.2014.08.012>.
- [22] M. Machaka, H. Basha, H.A. Chakra, A. Elkordi, Alkali treatment of fan palm natural fibers for use in fiber reinforced concrete, *Eur. Sci. J.* 10 (2014) 1857–7881, 10.10(1212):1857-7881.
- [23] E.B.C. Santos, C.G. Moreno, J.J.P. Barros, A.Ad Moura, FdC. Fim, A. Ries, R.M.R. Wellen, L.Bd Silva, Effect of alkaline and hot water treatments on the structure and

- morphology of piassava fibers, *Mater. Res.* 21 (2018), e20170365, <https://doi.org/10.1590/1980-5373-mr-2017-0365>.
- [24] A. Alawar, A.M. Hamed, K. Al-Kaabi, Characterization of treated date palm tree fiber as composite reinforcement, *Compos. B Eng.* 40 (2009) 601–606, <https://doi.org/10.1016/j.compositesb.2009.04.018>.
- [25] M. Li, G. Han, J. Yu, Microstructure and mechanical properties of *Apocynum venetum* fibers extracted by alkali-assisted ultrasound with different frequencies, *Fibers Polym.* 11 (2010) 48–53, <https://doi.org/10.1007/s12221-010-0048-2>.
- [26] S. Iwamoto, A. Nakagaito, H. Yano, Nano-fibrillation of pulp fibers for the processing of transparent nanocomposites, *Appl. Phys. A* 89 (2007) 461–466, <https://doi.org/10.1007/s00339-007-4175-6>.
- [27] L. Berglund, M. Noël, Y. Aitomäki, T. Öman, K. Oksman, Production potential of cellulose nanofibers from industrial residues: efficiency and nanofiber characteristics, *Ind. Crop. Prod.* 92 (2016) 84–92, <https://doi.org/10.1016/j.indcrop.2016.08.003>.
- [28] Y. Okahisa, Y. Furukawa, K. Ishimoto, C. Narita, K. Intharapichai, H. Ohara, Comparison of cellulose nanofiber properties produced from different parts of the oil palm tree, *Carbohydr. Polym.* 198 (2018) 313–319, <https://doi.org/10.1016/j.carbpol.2018.06.089>.
- [29] N. Yongvanich, Isolation of nanocellulose from pomelo fruit fibers by chemical treatments, *J. Nat. Fibers* 12 (2015) 323–331, <https://doi.org/10.1080/15440478.2014.920286>.
- [30] S. Naduparambath, T.V. Jiniitha, V. Shaniba, M.P. Sreejith, A.K. Balan, E. Purushothaman, Isolation and characterisation of cellulose nanocrystals from sago seed shells, *Carbohydr. Polym.* 180 (2018) 13–20, <https://doi.org/10.1016/j.carbpol.2017.09.088>.
- [31] K.S. Prado, M.A.S. Spinacé, Isolation and characterization of cellulose nanocrystals from pineapple crown waste and their potential uses, *Int. J. Biol. Macromol.* 122 (2019) 410–416, <https://doi.org/10.1016/j.ijbiomac.2018.10.187>.
- [32] A.K.A. Silva, C. Richard, M. Bessodes, D. Scherman, O.W. Merten, Growth factor delivery approaches in hydromel, *Biomacromolecules* 10 (2009) 9–18, <https://doi.org/10.1021/bm801103c>.
- [33] A. Oushabi, S. Sair, F.O. Hassani, Y. Abboud, O. Tanane, A. El Bouari, The effect of alkali treatment on mechanical, morphological and thermal properties of date palm fibers (DPFs): study of the interface of DPF-polyurethane composite, *S. Afr. J. Ind. Eng.* 23 (2017) 116–123, <https://doi.org/10.1016/j.sajce.2017.04.005>.
- [34] S. Mouhoubi, M.E.H. Bourahli, H. Osmani, S. Abdeslam, Effect of alkali treatment on alfa fibers behavior, *J. Nat. Fibers* 14 (2016) 239–249, <https://doi.org/10.1080/15440478.2016.1193088>.
- [35] E. Oudiani, Y. Chaabouni, S. Msahli, F. Sakli, Crystal transition from cellulose I to cellulose II in NaOH treated *Agave americana* L. fibre, *Carbohydr. Polymers* 86 (2011) 1221–1229, <https://doi.org/10.1016/j.carbpol.2011.06.037>.
- [36] R.C.R. Nunes, Rubber nanocomposites with nanocellulose, in: S. Thomas, H.J. Maria (Eds.), *Progress in Rubber Nanocomposites*, Woodhead Publishing, Oxford, 2016, pp. 463–494.
- [37] T. Okano, A. Sarko, Mercerization of cellulose II alkali-cellulose intermediates and a possible mercerization mechanism, *J. Appl. Polym. Sci.* 30 (1985) 325–332, <https://doi.org/10.1002/app.1985.070300128>.
- [38] U.J. Kim, S.H. Eom, M. Wada, Thermal decomposition of native cellulose: influence on crystallite size, *Polym. Degrad. Stabil.* 95 (2010) 778–781, <https://doi.org/10.1016/j.polymdegradstab.2010.02.009>.
- [39] Y. Yue, G. Han, Q. Wu, Transitional properties of cotton fibers from cellulose I to cellulose II structure, *BioResources* 8 (2013) 6460–6471, <https://doi.org/10.15376/biores.8.4.6460-6471>.
- [40] L.Y. Mwaikambo, M.P. Ansell, Chemical modification of hemp, sisal, jute, and kapok fibers by alkalization, *J. Appl. Polym. Sci.* 84 (2002) 2222–2234, <https://doi.org/10.1002/App.10460>.