

## Production of Nanocrystalline Cellulose from Bleached Soda Bagasse Pulp

Sabah Ashrafi Birgani,<sup>a</sup> Mohammad Talaeipour,<sup>a,\*</sup> Amir Hooman Hemmasi,<sup>a</sup> Behzad Bazyar,<sup>a</sup> and Kambiz Larijani<sup>b</sup>

The cellulose used in this study was prepared from bleached soda bagasse obtained from the Pars paper factory. To prepare nanocellulose, the sample was subjected to alkaline pretreatment and then acid hydrolysis using 54% sulfuric acid at several temperatures (35, 50, 60, and 65 °C) and different times (30, 60, 90, and 120 min). Then, they were prepared using a centrifuge, dialysis bag, ultrasound, and freezer, respectively. The produced nanocellulose was characterized by transmission electron microscopy (TEM), field-emission scanning electron microscopy (FE-SEM), Fourier transform infrared spectroscopy (FTIR), and X-ray diffraction (XRD). According to the results, temperatures of 50 and 90 °C were selected for the preparation of nanocellulose. The crystallization index of the hydrolyzed pulp and produced nanocellulose was 53 and 61%, respectively. The produced nanocellulose had a fibrillar shape.

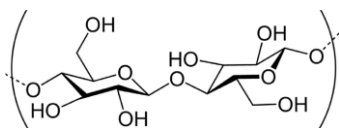
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*Contact information:* a: Department of Wood and Paper Science and Technology, Faculty of Natural Resources and Environment, Science and Research Branch, Islamic Azad University, P. O, Box 14515/775, Tehran, Iran; b: Department of Chemistry Science and Research Branch, Islamic Azad University, Tehran, Iran; P. O, Box 14515/775; \*Corresponding author: m.talaeipoor@srbiau.ac.ir

### INTRODUCTION

Cellulose is the most abundant polymeric raw material found in nature (Henriksson and Berglund 2007; O'Connell *et al.* 2008; Gama *et al.* 2012; Faruk *et al.* 2012; Vazquez *et al.* 2015; Zhu *et al.* 2016; Trache *et al.* 2016a,b; Santos *et al.* 2017; Ribeiro *et al.* 2019);  $10^{10}$  to  $10^{12}$  tons are produced annually (Postek *et al.* 2013). Only a small part of it ( $10^9 \times 6$  tons) is used in various industrial fields for example, paper, textile, industries, and chemistry (Trache *et al.* 2020 a, b). In general, cellulose is a fibrous, water-insoluble, strong substance that plays a key role in maintaining the structure of plant cell walls (Habibi *et al.* 2010). The properties of cellulosic fibers are greatly influenced by factors such as their internal fiber structure, microfibril angle, chemical composition, and cell dimensions, which vary between parts of the same plant and between plant species (Siqueira *et al.* 2010). Ignoring its source, Cellulose is a linear polysaccharide with D-glucopyranose bond joined by  $\beta$ -1,4 glycoside bonds (Henriksson and Berglund 2007; O'Connell *et al.* 2008; Faruk *et al.* 2012) (Fig. 1). There are three hydroxyl groups in cellulose. The primary hydroxyl is in the methylol group (-CH<sub>2</sub>OH) at C-6, and there are secondary hydroxyl groups (-OH) on C-3 and C-4. Each monomer unit rotates 180° relative to its adjacent monomers. Though it is a linear polysaccharide containing hydrophilic hydroxyl groups, it does not dissolve in water and common solvents due to the multiplicity of hydrogen bonds between the cellulose chains. A combination of hydrogen bonds between cellulose chains and van der Waals forces between glucose units lead to the formation of crystalline regions

in cellulose (O'Connell *et al.* 2008). The cell wall of wood cells is formed by the accumulation of cellulose, lignin, and hemicellulose. Cellulose fibers bind to each other with the help of lignin, and hemicellulose acts as a binder between cellulose and lignin. The number of hydrogen bonds plays an important role in the formation of cellulose and cellulose fibers.



**Fig. 1.** The structure of cellulose

Cellulose structure is composed of two parts: crystalline and amorphous. The amorphous region is the most susceptible for an acid or enzyme hydrolysis (Dufresne 2013; Kargarzadeh *et al.* 2017; Tarchoun *et al.* 2019a, b, c; Ribeiro *et al.* 2019). Cellulose can be obtained from a variety of sources, such as wood, herbaceous plants, grass, agricultural products and by-products, animal sources, algae, and bacteria (Trache *et al.* 2017; Nandi and Guha 2018; Kumar *et al.* 2020). Cellulose is a natural, biodegradable, environmentally friendly, and renewable material that has different physical, chemical, optical, magnetic, and biological properties due to its constituent raw materials (Jin *et al.* 2014; He *et al.* 2019). Cellulose crystals, nanocrystals, viscose, nanofibrils, and nanofibers have been produced (Siro and Plackett 2010). The properties of nanocelluloses including surface-to-volume ratio (high specific surface area), high modulus and tensile strength, low thermal expansion weakness, high hydrogen bonding capacity, biodegradability, high specific surface area, high rigidity, and non-toxicity (Siro and Plackett 2010; Habibi *et al.* 2010; Moon *et al.* 2011; Klemm *et al.* 2011; Lavoine 2012; Zhou *et al.* 2014; Salimi *et al.* 2019) have made this unique nanomaterial useful for research (Foster *et al.* 2018; Salimi *et al.* 2019). This provides the basis for its entry into more advanced applications (Yassin *et al.* 2019). In general, nanocellulose is classified into two main groups: nanofibers (bacterial cellulose, cellulose nanofibrils and cellulose nanocrystals) and nanostructured materials (including nanofibrillated cellulose and microfibrillated cellulose) (Trache *et al.* 2017; Hussin *et al.* 2019; Pennells *et al.* 2020). Different forms of nanocelluloses can be prepared using different methods and from various cellulosic sources (Phanthong *et al.* 2018; Pires *et al.* 2019; Salimi *et al.* 2019). Characteristics of each nanocellulose class such as size and morphology depend on the processing conditions, the isolation, and cellulose origin. (Li *et al.* 2018; Vilarinho *et al.* 2018; Pires *et al.* 2019; Köse *et al.* 2020). The preparation of nanocellulose usually requires two steps (Trache *et al.* 2017; Nandi and Guha 2018; Xie *et al.* 2018). In the first stage, the extracted materials (monomers, dimers, and lipid polymers, sugars, tannins, resins, flavonoids, terpenoids, terpenes, waxes, fatty acids, *etc.*), hemicelluloses, and lignin are partially or completely eliminated from the source by special pretreatment methods (Taipina 2012; Kargarzadeh *et al.* 2017). Second, the removal of amorphous regions of crude cellulose results in production of cellulose nanocrystal (CNC) (Dufresne 2013). After the second step, other treatments, such as solvent removal, neutralization, washing, purification, centrifugation, ultrasound, dialysis, stabilization, and drying (freezing and spray drying) can be performed (Lavoine *et al.* 2012; Taipina 2012; Trache *et al.* 2020). The most common process for producing nanocrystals uses strong acids such as hydrochloric acid, which produce highly crystalline nanoparticles (Klemm *et al.* 2018). An example of nanocellulose production from hardwood is shown in Fig. 2.

Although production of nanocellulose from various natural sources has been extensively studied, the use of the bleached soda bagasse pulp as a source of natural fiber has not been investigated fully. Bagasse is a by-product of the sugarcane factories. Sugarcane bagasse contains about 40 to 50% cellulose in its composition, much of which is in the crystalline structure (Wulandari *et al.* 2016). Due to its characteristics such as availability, low lignin content, and low cost, it is a promising raw material for extraction and production of nanocellulose. This study extracted CNC from bleached soda bagasse pulp using alkaline pretreatment followed by acid hydrolysis ( $\text{H}_2\text{SO}_4$ ).

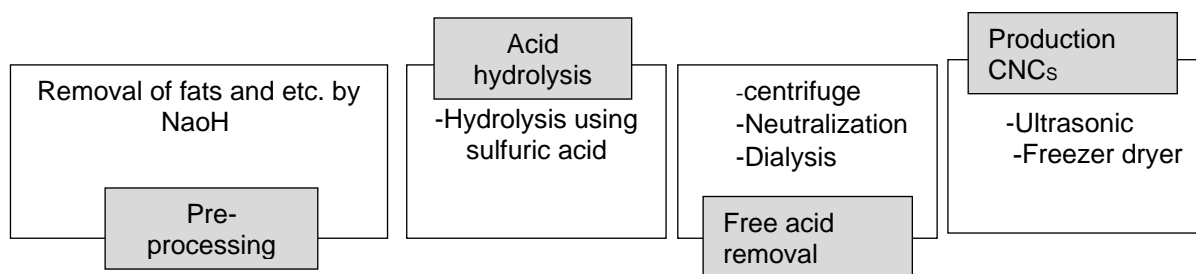


Fig. 2. An example of CNC production steps from hardwood (Lin *et al.* 2019)

## EXPERIMENTAL

### Materials

Bleached soda bagasse pulp was prepared from the Pars Company. Chemicals such as potassium hydroxide, acetic acid, sodium hydroxide, and sulfuric acid were purchased from Merck (Darmstadt, Germany). The sample was subjected to alkaline hydrolysis and acid hydrolysis, followed by the other treatments including solvent removal, washing, purification, filtration, centrifugation, dialysis, sonication, and freeze-drying.

### Alkaline Hydrolysis (Production of Alpha Cellulose)

For removing pigments and producing alpha cellulose, 20 g of the bleached soda bagasse pulp was mixed with 8% caustic soda in a ratio of 20: 1 (relative to dry weight to acid) by a constant mixer for 90 min at 30 °C. The pulp was drained and washed with 100 mL of 2% potassium hydroxide, 200 mL of hot distilled water, 200 mL of acetic acid, and 2000 mL of cold distilled water.

### Preparation of Nanocellulose

Alpha cellulose produced using 54% sulfuric acid was hydrolyzed at a 13:1 ratio. Cellulose hydrolysis was performed at several temperatures (35, 50, and 65 °C) and times (30, 60, 90, and 120 min). The reaction was stopped by adding 10 times distilled water (250 mL) to the solution. The suspension was centrifuged at 6500 rpm for 30 min and then placed in a dialysis bag for one week to neutralize and reach a pH level of 5. Neutral suspension was exposed to ultrasound for 10 min to produce homogeneous nanocellulose, which was then freeze-dried.

### Scanning Electron Microscope (FE-SEM)

A TESCSN VEGAII model (Czech Republic) was operated with a voltage of 15 kW. For this purpose, a sample of carrier powders was placed on a carbon plate and was

coated with gold with a thickness of 2 to 3 nm for 4 min.

### Transmission Electron Microscope (TEM)

The dimensions of nanocelluloses were measured on a Zeiss-EM10C-100 KV TEM (Germany) with a voltage of 100 KW. For preparing the sample, the nanocellulose suspension was mildly ultrasonicated in a water bath for 10 min, and 1 mL of the solution was dropped on a grid. After a few min, the sample was ready for imaging.

### Fourier Transform Infrared (FT-IR) Spectroscopy and Identification of Functional Groups

The FTIR analysis was performed using a Perkin Elmer RXI spectrometer (USA) at room temperature. The freeze-dried samples were mixed with KBr (Ratio 1 to 1000 mg) and pressed into tablets. The sample was scanned in a spectral range of 400 to 4000  $\text{cm}^{-1}$ .

### X-Ray Diffraction (XRD) Spectroscopy

X-ray diffraction patterns were recorded by Philips MPD X-ray generator (Netherlands) at 40 kV and 30 mA with X-ray radiation of  $\lambda=1/54056$  in the  $2\theta$  range of 5 to  $50^\circ$  with a scan step of  $0.05^\circ$ . The crystallinity index (CrI) was determined by the peak-height method (Segal *et al.* 1959) using Eq. 1,

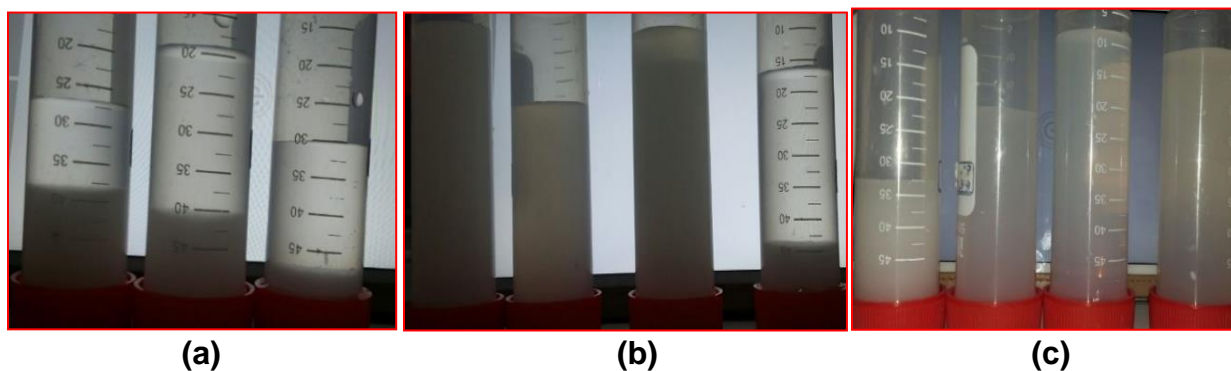
$$\text{CrI} = [(I_{002} - I_{\text{am}}) / I_{002}] \times 100 \quad (1)$$

where  $I_{002}$  is the maximum diffraction intensity corresponding to  $2\theta$  (about  $22^\circ$ ) related to crystalline regions, and  $I_{\text{am}}$  is the minimum diffraction intensity at  $2\theta$  (about  $18^\circ$ ) related to amorphous region.

## RESULTS AND DISCUSSION

### Stability of the Suspension

The samples were studied for stability of the prepared suspensions. As shown in Fig. 3, all treatments were precipitated at  $35^\circ\text{C}$ . One of the treatments was precipitated at  $50^\circ\text{C}$  (30 min) after 5 min, indicating that these suspensions were in micro dimensions, while other suspensions remained stable until the end of the first week of the test.



**Fig. 3.** Images obtained by digital camera regarding suspension stability. Treatments at (a)  $35^\circ\text{C}$ , (b)  $50^\circ\text{C}$ , and (c)  $65^\circ\text{C}$

The stability of nanofiber suspensions is attributed to factors such as high specific surface area of nanofibers and high irregular and random movement of nanofibers in excess volume. During acid hydrolysis, temperature is a very important factor influencing acid activity. For example, at low temperatures (35 °C), mild acid activity occurs on cellulose chains. At the right temperature and time, the acid destroys amorphous regions of the cellulose chains, which are removed, causing formation of more crystals in the cellulose chains, as well as greater stability of the suspension. To select a treatment with stable suspension to produce nanocellulose, FE-SEM and FTIR analyses were performed.

### Scanning Electron Microscope

In order to evaluate the size of nanocellulose produced in different samples and select the optimal conditions, electron microscopy was used. The FESEM images are shown in Fig. 2. The average length of nanocellulose was equal to 21.31, 13.82, 16.58, 17.64, 17.65, and 19.11 nm for the treatments studied under following conditions: 50 °C and 60 min, 50 °C and 90 min, 50 °C and 120 min, 30 °C and 65 min, 60 °C and 65 min, 90 °C and 65 min, and 120 °C and 65 min, respectively. The results showed that the sample obtained from 50 °C and 90 min were the smallest size.

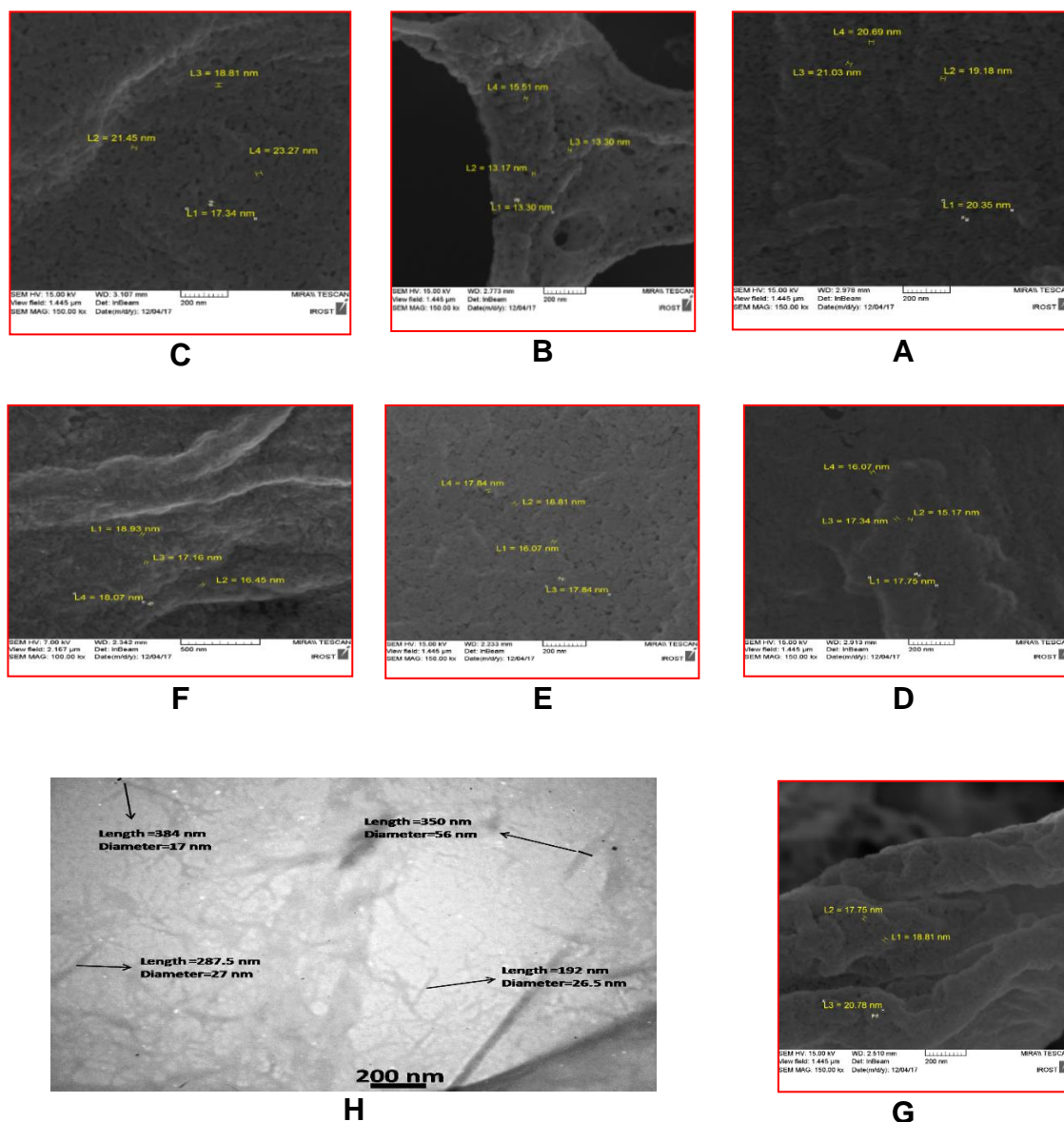
### Transmission Electron Microscope (TEM)

The TEM micrograph showed a very dilute suspension of nanocellulose from bleached soda bagasse. As shown in Fig. 4, nanocellulose particles accumulated in some places, while they were dispersed in the others. The particles were of fibrillar shape.

### FT-IR Spectroscopy and Identification of Functional Groups

The FTIR spectra of different treatments are shown in Figs. 5 and 6. All samples exhibited a wide band in the range of 3200 to 3500  $\text{cm}^{-1}$ , which is due to stretching vibration of O-H from OH groups of cellulose molecules (Khalil *et al.* 2001; Xu *et al.* 2005; Sain *et al.* 2006; Viera *et al.* 2007; Mandal and Chakrabarty 2011; Li *et al.* 2012; Lu and Hsieh 2012; Hokkanen *et al.* 2013; Rosli *et al.* 2013; Maiti *et al.* 2013; Kumar *et al.* 2014; Maryana *et al.* 2014; Li *et al.* 2014; Saelee *et al.* 2016; Almasian *et al.* 2016; Wulandari *et al.* 2016; Lam *et al.* 2017). The peak at 2894  $\text{cm}^{-1}$  is attributed to stretching modes of C-H in methyl and methylene functional groups, representing the main functional groups of cellulose (Mandal and Chakrabarty 2011; Lam *et al.* 2017). This range was not observed for samples with temperature and time conditions of 65 °C and 60, 90, and 120 min, but it was observed for the other samples. The peak from 1649 to 1641  $\text{cm}^{-1}$  was also related to O-H vibration of the adsorbed water (Ramo *et al.* 2000; Alemdar and Sain 2008; Mandal and Chakrabarty 2011; Li *et al.* 2012; Hokkanen *et al.* 2013; Almasian *et al.* 2016; Wulandari *et al.* 2016). The peak at 1250  $\text{cm}^{-1}$  represents aryl groups in lignin, found only in the bleached soda bagasse pulp sample, but it was decreased in the other samples. The peak from 890 to 820  $\text{cm}^{-1}$  is related to the vibration of COS and CS bond (Wei *et al.* 2014; Niu *et al.* 2017).

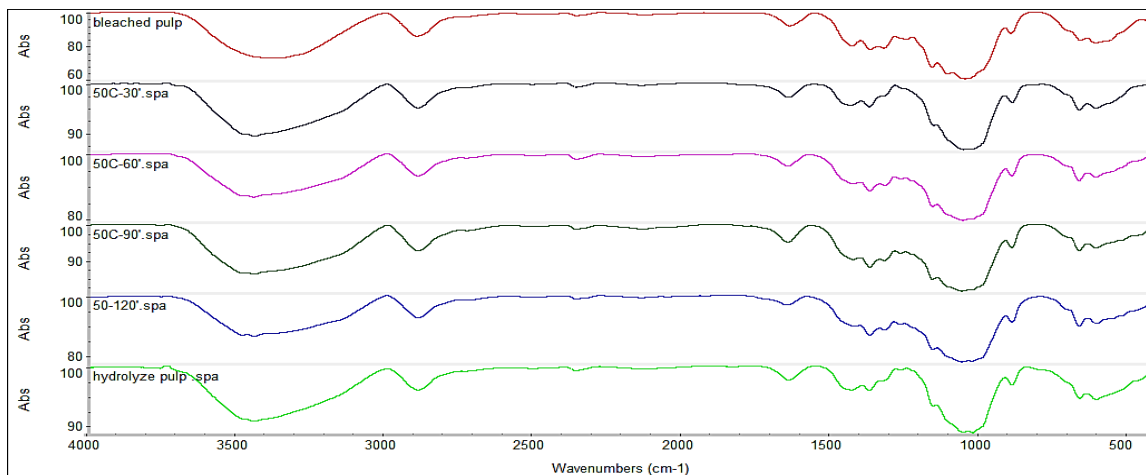
The results showed that carboxyl groups and sulfate esters were present in all samples. The slight increase of the adsorbing band at 1108 to 1100  $\text{cm}^{-1}$ , attributed to sulfate, supported the possibility of sulfate ester formation (Wei *et al.* 2014). Moreover, one peak at 894  $\text{cm}^{-1}$  is attributed to glycoside CH deformation by ring vibration and O-H bending, which is related to b-glycoside bonds between anhydro-glucose units in cellulose (Nelson and O'Connor 1964; Alemdar and Sain 2008; Mandal and Chakrabarty 2011; Wulandari *et al.* 2016). This peak was seen only in the bleached soda bagasse pulp sample.



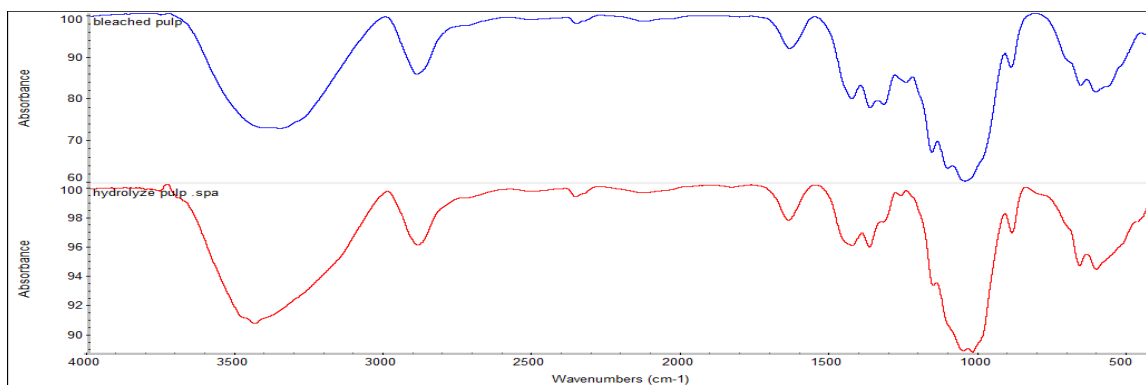
**Fig. 4.** Images obtained by FE-SEM of produce nanocellulose for different temperatures and time period at (A) 50 °C and 60 min, (B) 50 °C and 90 min, (C) 50 °C and 120 min, (D) 65 °C and 30 min, (E) 65 °C and 60 min, (F) 65 °C and 90 min, (G) 65 °C and 120 min, and (H) TEM image of the produced nanocelluloses at 50 °C and 90 min

The peak intensity was decreased to  $894\text{ cm}^{-1}$  by increasing hydrolysis time, indicating that the glycosidic bond had been broken and more OH groups had been released (Niu *et al.* 2017). The peak at  $1054\text{ cm}^{-1}$  associated with tensile vibration of C-O-C bond and glycoside bridge (Mandal and Chakrabarty 2011) was observed only in the bleached soda bagasse pulp due to hydrolysis and gradual reduction of molecular weight in the other samples. The band at  $1730\text{ cm}^{-1}$  was related to C=O stretching vibration for ester and acetyl linkage in hemicellulose or lignin (Nelson and O'Connor 1964; Alemdar and Sain 2008; Mandal and Chakrabarty 2011; Li *et al.* 2014; Wulandari *et al.* 2016; Lam *et al.* 2017). The peaks from 1602 to 1604 and 1505 to  $1511\text{ cm}^{-1}$  are attributed to C=C stretching vibration of aromatic ring in lignin (Kumar *et al.* 2014), and tension is related to the band at  $1245\text{ cm}^{-1}$  resulting from C-O stretching vibration of aryl group in lignin (Mandal and

Chakrabarty 2011; Rosli *et al.* 2013; Saelee *et al.* 2016). None of these groups were found in the FTIR spectrum of the studied cellulose fibers, confirming the removal of lignin. According to the results of FE-SEM and FTIR, a sample studied at 50 °C and 90 min was selected to determine crystallinity and capture TEM images.



**Fig. 5.** FTIR spectra of the bleached soda bagasse pulp and alkaline hydrolysis-subjected pulp, and acid hydrolysis-subjected pulp at 50 °C and 30 min, 60 min, 90 min and 120 min



**Fig. 6.** FTIR spectra bleached soda bagasse pulp and alkaline hydrolysis-subjected pulp

### X-ray Diffraction Spectroscopy

XRD microscopic images of the hydrolyzed pulp and produced nanocellulose are shown in Fig. 7. With the increase in amount of non-cellulosic polysaccharides and dissolution of amorphous regions, more orientation occurs along a specific axis (Mandal and Chakrabarty 2011). Both diffraction diagrams showed two well-defined peaks at  $\Theta_2=18$  and  $\Theta_2=22$ . Diffraction pattern related to the hydrolyzed pulp and produced nanocellulose was almost identical and similar to previous results (Mandal and Chakrabarty 2011; Guilherme *et al.* 2015; Wulandari *et al.* 2016; Niu *et al.* 2017). The crystallization index of the hydrolyzed pulp and produced nanocellulose was obtained as 53% and 61%, respectively. The increased degree of crystallinity was attributed to removal of hemicelluloses and lignin.



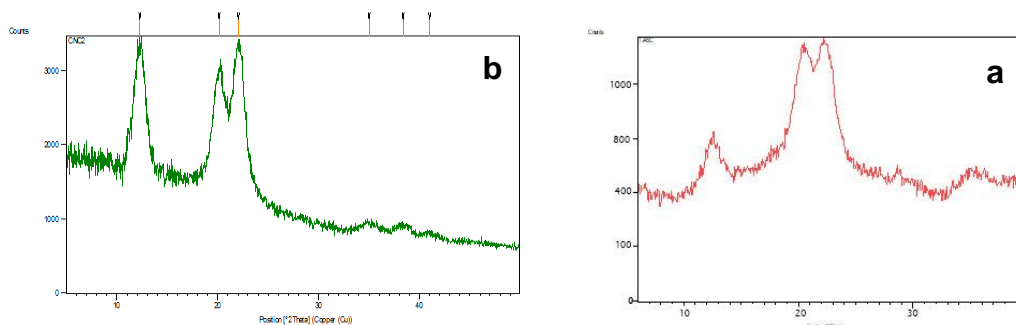


Fig. 7. XRD diagram of the hydrolyzed pulp (a) and produced nanocellulose (b)

## CONCLUSIONS

1. The crystallization index of the hydrolyzed pulp and produced nanocellulose was 53% and 67%, respectively. The increased crystallinity is attributed to the removal of hemicelluloses and lignin. Relatively high crystalline degree of nanocellulose produced from bagasse makes it very promising in new industrial applications, for example, as a reinforcer in matrix of polymers produced by various industries.
2. Based on the results of TEM analysis, nanocellulose had a fibrillar shape. Nanocellulose particles accumulated in some places but were dispersed in others.
3. FTIR analysis showed the absence of bands in the ranges of  $1725$  to  $1730\text{ cm}^{-1}$ ,  $1602$  to  $1604\text{ cm}^{-1}$ , and  $1511$  to  $1505\text{ cm}^{-1}$ , indicating the removal of lignin and hemicellulose from the suspension.
4. At higher temperatures and at right time, the acid destroys amorphous regions of the cellulose chains. Destruction and removal of these regions causes crystallization of the cellulose chains as well as stability of the suspension.

## REFERENCES CITED

- Almasian, A., Chizari Fard, Gh., Parvinzadeh Gashti, M., Mirjalili, M., and Mokhtari Shourijeh, Z. (2016). "Surface modification of electrospun PAN nanofibers by amine compounds for adsorption of anionic dyes," *Desalination and Water Treatment* 57(12), 10333-10348. DOI: 10.1080/19443994.2015.1041161
- Alemdar, A., and Sain, M. (2008). "Isolation and characterization of nanofibers from agricultural residues: Wheat straw and soy hulls," *Bioresour Technol.* 99(6), 1664-1671. DOI: 10.1016/j.biortech.2007.04.029
- Börjesson, M., and Gunnar, W. (2015). "Crystalline nanocellulose — Preparation, modification, and properties," *Cellulose - Fundamental Aspects and Current Trends*, Matheus Poletto and Heitor Luiz Ornaghi Junior, IntechOpen, 159-191. DOI: 10.5772/61899. Available from: <https://www.intechopen.com/chapters/49666>
- Dufresne, A. (2013). "From nature to high performance tailored materials," in: *Textbook of Nanocellulose*, De Gruyter, Berlin. DOI:10.1515/9783110254600



- Faruk, O., Bledzki, A. K., Fink, H.-P., and Sain, M. (2012). "Biocomposites reinforced with natural fibers: 2000-2010," *Progress in Polymer Science* 37 (11), 1552-1596. DOI: 10.1016/j.progpolymsci.2012.04.003
- Foster, E. J., Moon, R. J., Agarwal, U., Bortner, M. J., Camarero-Espinosa, S., Chan, K. J., Clift, M. J. D., Cranston, E. D., Eichhorn, S. J., Fox, D. M., *et al.* (2018). "Current characterization methods for cellulose nanomaterials," *Chemical Society Reviews* 47(8), 2609-2679. DOI: 10.1039/C6CS00895J
- Gama, M., Gatenholm, P., and Klemm, D. (2012). *Bacterial Nanocellulose: A Sophisticated Multifunctional Material*, CRC Press. DOI:10.1201/B12936
- Guilherme, A. A., Dantas, P. V. F., Santos, E. S., Fernandes, F. A. N., and Macedo, G. R. (2015). "Evaluation of composition, characterization and enzymatic hydrolysis of pretreated sugar cane bagasse," *Brazilian Journal of Chemical Engineering* 32(01), 23-33. DOI: 10.1590/0104-6632.20150321s00003146
- Habibi, Y., Lucia, L. A., and Rojas, O. J. (2010). "Cellulose nanocrystals: Chemistry, self-assembly, and applications," *Chem. Rev.* 110(6), 3479-3500. DOI: 10.1021/cr900339w
- He, X., Deng, H., and Hwang, H. (2019). "The current application of nanotechnology in food and agriculture," *Journal of Food and Drug Analysis* 27(1), 1-21. DOI: 10.1016/j.jfda.2018.12.002
- Henriksson, M., and Berglund, L. A. (2007). "Structure and properties of cellulose nanocomposite films containing melamine formaldehyde," *Journal of Applied Polymer Science* 106 (4), 2817-2824. DOI: 10.1002/app.26946
- Hokkanen, S., Repo, E., and Sillanpää M. (2013). "Removal of heavy metals from aqueous solutions by succinic anhydride modified mercerized nanocellulose," *Chemical Engineering Journal* 223, 40-47. DOI: 10.1016/j.cej.2013.02.054
- Hussin, M. H., Trache, D., Chuin, C. T. H., Fazita, M. N., Haafiz, M. M., and Hossain, M. S. (2019). "Extraction of cellulose nanofibers and their eco-friendly polymer composites," in *Sustainable Polymer Composites and Nanocomposites*, S. Thomas, R. K. Mishra, and A. M. Asiri (eds.), Springer, pp. 653-691. DOI: 10.1007/978-3-030-05399-4\_23
- Jin, L., Wei, Y., Xu, Q., Yao, W., and Cheng, Z. (2014). "Cellulose nanofibers prepared from TEMPO-oxidation of kraft pulp and its flocculation effect on kaolin clay," *Journal of Applied Polymer Science* 131(12), 8-11. DOI: 10.1002/app.40450
- Kargarzadeh, H., Mariano, M., Huang, J., Lin, N., Ahmad, I., Dufresne, A., and Thomas, S. (2017). "Recent developments on nanocellulose reinforced polymer nanocomposites: A review," *Polymer* 132(6), 368-393. DOI: 10.1016/j.polymer.2017.09.043
- Khalil, H., Ismail, H., Rozman, H., and Ahmad, M. (2001). "The effect of acetylation on interfacial shear strength between plant fibres and various matrices," *European Polymer Journal* 37, 1037-1045. DOI: 10.1016/S0014-3057(00)00199-3
- Klemm, D., Kramer, F., Moritz, S., Lindstrom, T., Ankerfors, M., Gray, D., and Dorris, A. (2011). "Nanocelluloses: A new family of nature-based materials," *Angewandte Chemie International Edition* 50(24), 5438-5466. DOI:10.1002/anie.201001273
- Klemm, D., Durand, H., Zeno, E., Balsollier, C., Watbled, B., Sillard, C., Fort, S., Baussanne, I., Belgacem, N., Lee, D., Hediger, S., Demeunynck, M., Bras, J., and Paepe, G. D. (2018). "Nanocellulose as a natural source for groundbreaking applications in materials science: Today's state," *Materials today* 27(7), 720-748. DOI:10.1016/j.mattod.2018.02.001

- Köse, K., Mavlan, M., and Youngblood, J. P. (2020). “Applications and impact of nanocellulose based adsorbents,” *Cellulose* 27, 2967-2990. DOI: 10.1007/s10570-020-03011-1
- Kumar, A., Negi, Y. S., Choudhary, V., Bhardwaj, N. K. (2014). “Characterization of cellulose nanocrystals produced by acid-hydrolysis from sugarcane bagasse as agro-waste,” *Journal of Materials Physics and Chemistry* 2(1), 1-8. DOI:10.12691/jmpc-2-1-1
- Kumar, A., Durand, H., Zeno, E., Balsollier, C., Watbled, B., Sillard, C., Fort, S., Baussanne, I., Belgacem, N., Lee, D., Hediger, S., Demeunynck, M., Bras, J., and Paepe, G. D. (2020). “The surface chemistry of a nanocellulose drug carrier unravelled by MAS-DNP,” *Chemical Science* 11(15), 3868-3877. DOI: 10.1039/C9SC06312A
- Lam, N. L., Chollakup, R., Smitthipong, W., Nimchua, T., and Sukyai, P. (2017). “Characterization of cellulose nanocrystals extracted from sugarcane bagasse for potential biomedical materials,” *Sugar Tech* 19(5), 539-552. DOI :10.1007/s12355-016-0507-1
- Lavoine, N., Desloges, I., Dufresne, A., and Bras, J. (2012). “Microfibrillated cellulose-its barrier properties and applications in cellulosic materials: A review,” *Carbohydrate Polymers* 90(2), 735-764. DOI:10.1016/j.carbp ol.2012.05.026
- Li, J., Cha, R., Mou, K., Zhao, X., Long, K., Luo, H., *et al.* (2018). “Nanocellulose-based antibacterial materials,” *Adv. Healthc. Mater* 7(20). DOI: 10.1002/adhm.201800334
- Li, J., Wei, X., Wang, Q., Chen, J., Chang, G., Kong, L., Su, J., and Liu, Y. (2012). “Homogeneous isolation of nanocellulose from sugarcane bagasse by high pressure homogenization,” *Carbohydrate Polymers* 90(4), 1609-1613. DOI: 10.1016/j.carbpol.2012.07.038
- Lin, K. H., Enomae, T., and Chang, F. C. (2019). “Cellulose nanocrystal isolation from hardwood pulp using various hydrolysis conditions,” *Molecules* 24(20),857-903. DOI: 10.3390/molecules24203724
- Li, M., Wang, L., Li, D., Cheng, Y., and Adhikari, B. (2014). “Preparation and characterization of cellulose nanofibers from de-pectinated sugar beet pulp,” *Carbohydrate Polymers* 102, 136-143. DOI: 10.1016/j.carbpol.2013.11.021
- Lu, P., and Hsieh, Y. (2012). “Cellulose isolation and core-shell nanostructures of cellulose nanocrystals from chardonnay grape skins,” *Carbohydrate Polymers* 87(4), 2546-2553. DOI:10.1016/j.carbpol.2011.11.023
- Moon, J. R., Martine, A., Nairn, J., Simonsen, J., and Youngblood, J. (2011). “Cellulose nanomaterials review: Structure, properties and nanocomposites,” *Chemical Society Reviews* 40(7), 3941-3994. DOI:10.1039/ C0CS00108B
- Maiti, S., Jayaramudu, J., Das, K., Reddy, S. M., Sadiku, R., Ray, S. S., and Liu, D. (2013). “Preparation and characterization of nano-cellulose with new shape from different precursor,” *Carbohydrate Polymers* 98(1), 562-567. DOI: 10.1016/j.carbpol.2013.06.029
- Mandal, A., and Chakrabarty, D. (2011). “Isolation of nanocellulose from waste sugarcane bagasse (SCB) and its characterization,” *Carbohydrate Polymers* 86(3), 1291-1299. DOI: 10.1016/j.carbpol.2011.06.030
- Maryana, R., Marifatun, D., Wheni, A. I., Satriyo, K.W., and Rizal, W. A. (2014). “Alkaline pretreatment on sugarcane bagasse for bioethanol production,” *Energy Procedia* 47, 250-254. DOI: 10.1016/j.egypro.2014.01.221

- Millar, G. J., Rochester, C. H., and Waugh, K. C. (1995). "An FTIR study of the adsorption of formic acid and formaldehyde on potassium-promoted Cu/SiO<sub>2</sub> catalysts," *Journal of Catalysis* 155(1), 52-58. DOI: 10.1006/jcat.1995.1187
- Nandi, S., and Guha, P. (2018). "A review on preparation and properties of cellulose nanocrystal-incorporated natural biopolymer," *Journal of Package Technology and Research* 2, 149-166. DOI :10.1007/s41783-018-0036-3
- Nelson, M., and O'Connor, R. (1964). "Relation of certain infrared bands to cellulose crystallinity and crystal lattice type. Part II. A new infrared ratio for estimation of crystallinity in celluloses I and II," *Journal of Applied Polymer Science* 8(3),1325-1341. DOI:10.1002/app.1964.070080323
- Niu, F., Li, M., Huang, Q., Zhang, X., Pan, W., Yang, J., and Li, J. (2017). "The characteristic and dispersion stability of nanocellulose produced by mixed acid hydrolysis and ultrasonic assistance," *Carbohydrate Polymers* 165, 197-204. DOI: 10.1016/j.carbpol.2017.02.048
- O'Connell, D. W., Birkinshaw, C., and O'Dwyer T., F. (2008). "Heavy metal adsorbents prepared from the modification of cellulose: A review," *Bioresource Technology* 99, 6709-6724. DOI: 10.1016/j.biortech.2008.01.036
- Pennells, J., Godwin, I. D., Amiralian, N., and Martin, D. J. (2020). "Trends in the production of cellulose nanofibers from non-wood sources," *Cellulose* 27, 575-593. DOI: 10.1007/s10570-019-02828-9
- Phanthong, P., Reubroycharoen, P., Hao, X., Xu, G., Abudula, A., and Guan, G. (2018). "Nanocellulose: Extraction and application," *Carbon Resources Conversion*. 1(1), 32-43. DOI: 10.1016/j.crcon.2018.05.004
- Pires, J. R., Souza, V. G., and Fernando, A. L. (2019). "Valorization of energy crops as a source for nanocellulose production—current knowledge and future prospects," *Industrial Crops Products* 140, 111642. DOI: 10.1016/j.indcrop.2019.111642
- Postek, M., Moon, R., Rudie, A., and Bilodeau, M. (2013). *Production and Applications of Cellulose Nanomaterials*, TAPPI Press.
- Ramo, J., Sillanpaa, M., Orama, M., Vickackaite, V., and Niinisto, L. (2000). "Chelating ability and solubility of DTPA, EDTA and b-ADA in alkaline hydrogen peroxide environment," *Journal of Pulp and Paper Science* 26(4), 125-131.
- Ribeiro, R. S. A., Pohlmann, B. C., Calado, V., Bojorge, N., and Pereira Jr, N. (2019). "Production of nanocellulose by enzymatic hydrolysis: Trends and challenges," *Engineering in Life Sciences* 19, 279-291. DOI: 10.1002/elsc.201800158
- Rosli, N., Ahmad, I. and Abdula, I. (2013). "Isolation and characterization of cellulose nanocrystals from *Agave angustifolia* fibre," *BioResources* 8(2), 1893-1908.
- Saelee, K., Yingkamhaeng, N., Nimchua, T., and Sukyai, P. (2016). "An environmentally friendly xylanase-assisted pretreatment for cellulose nanofibrils isolation from sugarcane bagasse by high-pressure homogenization," *Industrial Crops and Products* 82, 149-160. DOI :10.1016/j. indcrop.2015.11.064
- Salimi, S., Sotudeh-Gharebagh, R., Zarghami, R., Chan, S. Y., and Yuen, K. H. (2019). "Production of nanocellulose and its applications in drug delivery: A critical review," *ACS Publications* 7, 15800-15827. DOI: 10.1021/acssuschemeng.9b02744
- Sain, M., and Oksman, K. (2006). "Introduction to cellulose nanocomposites," *Cellulose Nanocomposites: Processing, Characterization and Properties* 938, 2-8. Downloaded via 91.133.215.175 on August 22, 2021 at 18:14:28 (UTC). See <https://pubs.acs.org/sharingguidelines> for options on how to legitimately share published articles.

- Santos, S., Carbajo, J. M., Gómez, N., and Ladero, M. (2017). "Paper reinforcing by in situ growth of bacterial cellulose," *Journal of Materials Science* 52(10). DOI:10.1007/s10853-017-0824-0
- Segal, L., Greely, J. J., Martin, A. E., and Conrad, C. M. (1959). "An empirical method for estimating the degree of crystallinity of native cellulose using X-ray diffractometer," *Textile Research Journal* 29(10), 786-794. DOI :10.1177/004051755902901003
- Siro, I., and Plackett, D. (2010). "Microfibrillated cellulose and new nanocomposite materials: A review," *Cellulose* 17(3), 459-494.
- Siqueira, G., Bras, J., and Dufresne, A. (2010). "*Luffa cylindrica* as a lignocellulosic source of fiber, microfibrillated cellulose, and cellulose nanocrystals," *BioResources* 5(2), 727-740.
- Sugama, T. (1997). "Oxidized potato-starch films as primer coatings of aluminium," *Journal of Materials Science* 32(15), 3995-4003.
- Taipina, M. O. (2012). "Nanocrystals de cellulose: Obtenção, caracterização e modificação de superfície," *Repository from the State University of Campinas, Brazil*. [https://repositorio.unicamp.br/handle/REPOS\\_IP/248724](https://repositorio.unicamp.br/handle/REPOS_IP/248724).
- Tarchoun, A. F., Trache, D., and Klapötke, T. M. (2019a). "Microcrystalline cellulose from *Posidonia oceanica* brown algae: Extraction and characterization," *International Journal of Biological Macromolecules* 138, 837-845. DOI:10.1016/j.ijbiomac.2019.07.176
- Tarchoun, A. F., Trache, D., Klapötke, T. M., Chelouche, S., Derradji, M., Bessa, W., and Mezroua, A. (2019b). "A promising energetic polymer from *Posidonia oceanica* brown algae: Synthesis, characterization, and kinetic modeling," *Macromolecular Chemistry and Physics* 220(22). DOI: 10.1002/macp.201900358
- Tarchoun, A. F., Trache, D., Klapötke, T. M., Derradji, M., and Bessa, W. (2019c). "Ecofriendly isolation and characterization of microcrystalline cellulose from giant reed using various acidic media," *Cellulose* 26, 7635-7651. DOI: 10.1007/s10570-019-02672-x
- Trache, D., Hussin, M. H., Chuin, C. T. H., Sabar, S., Fazita, M. N., Taiwo, O. F., et al. (2016a). "Microcrystalline cellulose: Isolation, characterization and biocomposites application – A review," *International Journal of Biological Macromolecules* 93(Part A), 789-804. DOI: 10.1016/j.ijbiomac.2016.09.056
- Trache, D., Khimeche, K., Mezroua, A., and Benziane, M. (2016b). "Physicochemical properties of microcrystalline nitrocellulose from alfa grass fibres and its thermal stability," *Journal of Thermal Analysis and Calorimetry* 124, 1485-1496. DOI: 10.1007/s10973-016-5293-1
- Trache, D., Hussin, M. H., Haafiz, M. M., and Thakur, V. K. (2017). "Recent progress in cellulose nanocrystals: Sources and production," *Nanoscale* 9, 1763-1786. DOI:10.1039/C6NR09494E
- Trache, D., Tarchoun, A. F., Derradji, M., Mehelli, O., Hussin, M. H., and Bessa, W. (2020a). "Cellulose fibers and nanocrystals: Preparation, characterization and surface modification," in: *Textbook of Functionalized Nanomaterials I*, CRC Press, Boca Raton, FL. DOI:10.1201/9781351021623
- Trache, D., Tarchoun, A. F., Derradji, M., Hamidon, T. S., Masruchin, N., Brosse, N., and Hussin, M. H. (2020b). "Nanocellulose: From fundamentals to advanced applications," *Frontiers in Chemistry* 8. DOI: 10.3389/fchem.2020.00392

- Vazquez, A., Foresti, M. L., Moran, J. I., and Cyras, V. P. (2015). "Extraction and production of cellulose nanofibers," in: *Handbook of Polymer Nanocomposites. Processing, Performance and Application*, J. K. Pandey, H. Takagi, A. N. Nakagaito, and H. J. Kim (eds.), Springer, Heidelberg, pp. 81-118. DOI: 10.1007/978-3-642-45232-1\_57\_
- Viera, R. G. P., Filho, G. R., Assuncao, R. M. N., Meireles, C. S., Viera, J. G., and Oliveira, G. S. (2007). "Synthesis and characterization of methylcellulose from sugar cane bagasse cellulose," *Carbohydr. Polym* 67(2), 182-189. DOI: 10.1016/j.carbpol.2006.05.007
- Vilarinho, F., Silva, A. S., Vaz, M. F., and Farinha, J. P. (2018). "Nanocellulose in green food packaging," *Critical Reviews in Food Science and Nutrition* 58, 1526-1537. DOI:10.1080/10408398.2016.1270254
- Wei, H., Rodriguez, K., Renneckar, S., and Vikesland, P. J. (2014). "Environmental science and engineering applications of nanocellulose-based nanocomposites," *Environmental Science Nano* 1(4), 302-316. DOI: 10.1039/C4EN00059E
- Wulandari, W, T., Rochliadi, A., and Arcan, I, M. (2016). "Nanocellulose prepared by acid hydrolysis of isolated cellulose from sugarcane bagasse," in: *10<sup>th</sup> Joint Conference on Chemistry, Materials Science and Engineering 107*, Solo, Indonesia. DOI:10.1088/1757-899X/107/1/012045
- Xie, H., Du, H., Yang, X., and Si, C. (2018). "Recent strategies in preparation of cellulose nanocrystals and cellulose nanofibrils derived from raw cellulose materials," *International Journal of Polymer Science*. 1-25. DOI: 10.1155/2018/7923068
- Xu, Y., Wang, Z., Ke, R., and Khan, S. U. (2005). "Accumulation of organochlorine pesticides from water using triolein embedded cellulose acetate membranes," *Environmental Science and Technology* 39(4), 1152-1157. DOI:10.1021/es040454m
- Yassin, M. A., Gad, A. A. M., Ghanem, A. F., and Abdel Rehim, M. H. (2019). "Green synthesis of cellulose nanofibers using immobilized cellulase," *Carbohydr. Polym.* 205, 255-260. DOI: 10.1016/j.carbpol.2018.10.040
- Zhou, Y., Fu, S., Zhang, L., Zhan, H., and Levit, M. V. (2014). "Use of carboxylated cellulose nanofibrils-filled magnetic chitosan hydrogel beads as adsorbents for Pb (II)," *Carbohydrate. Polymers* 101, 75-82. DOI: 10.1016/j.carbpol.2013.08.055
- Zhu, H., Yang, X., Cranston, E. D., and Zhu, S. P. (2016). "Flexible and porous nanocellulose aerogels with high loadings of metal-organic-framework particles for separations applications," *Advanced Materials* 28(35),7652-7657. DOI:10.1002/adma.201601351

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