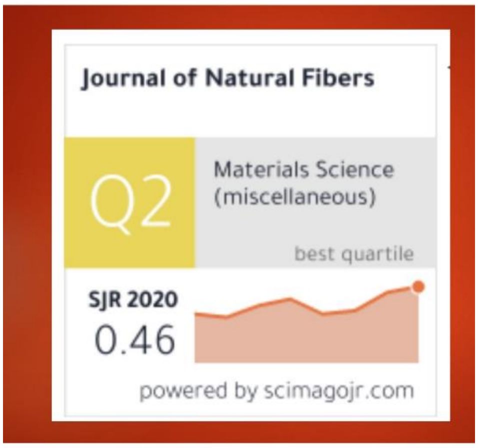
Bukti Korespondensi

Paper 15

Cellulose nanofibers isolation and characterization from ramieusing a chemical-ultrasonic treatment*Pengusul*

Dr. Edi Syafri, ST, M.Si



Citation metrics

- **3.507 (2021)** Impact Factor
- **Q1** Impact Factor Best Quartile
- **3.760 (2021)** 5 year IF
- **5.6 (2021)** CiteScore (Scopus)
- **Q1 (2021)** CiteScore Best Quartile
- **1.270 (2021)** SNIP
- **0.480 (2021)** SJR

1. Proses Submission

Journal of Natural Fibers - Manuscript ID WJNF-2017-0179 has been submitted online Kotak Masuk ×

Journal of Natural Fibers <onbehalfof+izabela.maciejowska+iwnirz.pl@manuscriptcentral.com> Sel, 4 Jul 2017, 20.49

kepada saya

Inggris > Indonesia Terjemahkan pesan Nonaktifkan untuk: Inggris ×

04-Jul-2017

Dear Mr Syafri:

Your manuscript entitled "Cellulose Nanofiber Isolation and Characterization from ramie fibers using a chemical-ultrasonic treatment" has been successfully submitted online and is presently being given full consideration for publication in Journal of Natural Fibers.

Your manuscript ID is WJNF-2017-0179.

Please mention the above manuscript ID in all future correspondence or when calling the office for questions. If there are any changes in your street address or e-mail address, please log in to ScholarOne Manuscripts at https://mc.manuscriptcentral.com/jnf and edit your user information as appropriate.

You can also view the status of your manuscript at any time by checking your Author Center after logging in to https://mc.manuscriptcentral.com/jnf.

Thank you for submitting your manuscript to Journal of Natural Fibers.

Sincerely,
Journal of Natural Fibers Editorial Office

Journal of Natural Fibers - Manuscript ID WJNF-2017-0179 has been submitted online Kotak Masuk ×

Journal of Natural Fibers <onbehalfof+izabela.maciejowska+iwnirz.pl@manuscriptcentral.com> Sab, 8 Jul 2017, 06.49

kepada saya

Inggris > Indonesia Terjemahkan pesan Nonaktifkan untuk: Inggris ×

07-Jul-2017

Dear Mr Syafri:

Your manuscript entitled "Cellulose Nanofiber Isolation and Characterization from ramie fibers using a chemical-ultrasonic treatment" has been successfully submitted online and is presently being given full consideration for publication in Journal of Natural Fibers.

Your manuscript ID is WJNF-2017-0179.

Please mention the above manuscript ID in all future correspondence or when calling the office for questions. If there are any changes in your street address or e-mail address, please log in to ScholarOne Manuscripts at https://mc.manuscriptcentral.com/jnf and edit your user information as appropriate.

You can also view the status of your manuscript at any time by checking your Author Center after logging in to https://mc.manuscriptcentral.com/jnf.

Thank you for submitting your manuscript to Journal of Natural Fibers.

Sincerely,
Journal of Natural Fibers Editorial Office

Journal of Natural Fibers - Manuscript ID WJNF-2017-0179 has been submitted online

Kotak Masuk ×

Journal of Natural Fibers <onbehalfof+izabela.maciejowska+iwnirz.pl@manuscriptcentral.com>

kepada saya

Min, 16 Jul 2017, 22.00

Inggris > Indonesia Terjemahkan pesan

Nonaktifkan untuk: Inggris ×

16-Jul-2017

Dear Mr Syafri:

Your manuscript entitled "Cellulose Nanofiber Isolation and Characterization from ramie fibers using a chemical-ultrasonic treatment" has been successfully submitted online and is presently being given full consideration for publication in Journal of Natural Fibers.

Your manuscript ID is WJNF-2017-0179.

Please mention the above manuscript ID in all future correspondence or when calling the office for questions. If there are any changes in your street address or e-mail address, please log in to ScholarOne Manuscripts at https://mc.manuscriptcentral.com/jnf and edit your user information as appropriate.

You can also view the status of your manuscript at any time by checking your Author Center after logging in to https://mc.manuscriptcentral.com/jnf.

Thank you for submitting your manuscript to Journal of Natural Fibers.

Sincerely,
Journal of Natural Fibers Editorial Office

From: ryszard.kozlowski@escorena.net

To: edisyafri11@gmail.com

CC:

Subject: Journal of Natural Fibers - Decision on Manuscript ID WJNF-2017-0179

Body: 15-Feb-2018

Dear Mr Syafri:

Your manuscript entitled "Cellulose Nanofibers Isolation and Characterization from ramie using a chemical-ultrasonic treatment", which you submitted to Journal of Natural Fibers, has been reviewed. The reviewer comments are included at the bottom of this letter.

The reviews are in general favourable and suggest that, subject to minor revisions, your paper could be suitable for publication. Please consider these suggestions, and I look forward to receiving your revision.

When you revise your manuscript please highlight the changes you make in the manuscript by using the track changes mode in MS Word or by using bold or colored text.

To start the revision, please click on the link below:

*** PLEASE NOTE: This is a two-step process. After clicking on the link, you will be directed to a webpage to confirm. ***

https://mc.manuscriptcentral.com/jnf?URL_MASK=b55adf32be794268a003781320ddf249

This will direct you to the first page of your revised manuscript. Please enter your responses to the comments made by the reviewer(s) in the space provided. You can use this space to document any changes you made to the original manuscript. Please be as specific as possible in your response to the reviewer(s).

This link will remain active until you have submitted your revised manuscript. If you begin a revision and intend to finish it at a later time, please note that your draft will appear in the “Revised Manuscripts in Draft” queue in your Author Center.

IMPORTANT: Your original files are available to you when you upload your revised manuscript. Please delete any redundant files before completing the submission.

Because we are trying to facilitate timely publication of manuscripts submitted to Journal of Natural Fibers, your revised manuscript should be uploaded by 08-Mar-2018. If it is not possible for you to submit your revision by this date, we may have to consider your paper as a new submission.

Once again, thank you for submitting your manuscript to Journal of Natural Fibers and I look forward to receiving your revision.

Sincerely,
Professor Kozlowski
Editor in Chief, Journal of Natural Fibers
ryszard.kozlowski@escorena.net

Reviewer(s)' Comments to Author:

Reviewer: 1

Comments to the Author
in this manuscript, cellulose nanofibers was isolated from ramie (Boehmeria nivea (L.) Gaud) with the combination of chemical method and ultrasonication treatment, and the characterization of the morphology and crystal structure, thermal stability and size. However, the abstract is so simple, and English level is bad, so please improve it.

Reviewer: 2

Comments to the Author
Please, include in the text the yield of nanocellulose obtained of this process.

Editor's Comments to Author:

Associate Editor: 1
Comments to the Author:
(There are no comments.)

Date Sent: 15-Feb-2018

Dari: Journal of Natural Fibers <onbehalfof@manuscriptcentral.com>
Tanggal: 15/02/2018 14:49 (GMT+07:00)
Ke: edisyafri11@gmail.com
Subjek: Journal of Natural Fibers - Decision on Manuscript ID WJNF-2017-0179

15-Feb-2018

Dear Mr Syafri:

Your manuscript entitled "Cellulose Nanofibers Isolation and Characterization from ramie using a chemical-ultrasonic treatment", which you submitted to Journal of Natural Fibers, has been reviewed. The reviewer comments are included at the bottom of this letter.

The reviews are in general favourable and suggest that, subject to minor revisions, your paper could be suitable for publication. Please consider these suggestions, and I look forward to receiving your revision.

When you revise your manuscript please highlight the changes you make in the manuscript by using the track changes mode in MS Word or by using bold or colored text.

To start the revision, please click on the link below:

*** PLEASE NOTE: This is a two-step process. After clicking on the link, you will be directed to a webpage to confirm. ***

https://mc.manuscriptcentral.com/jnf?URL_MASK=b55adf32be794268a003781320ddf249

This will direct you to the first page of your revised manuscript. Please enter your responses to the comments made by the reviewer(s) in the space provided. You can use this space to document any changes you made to the original manuscript. Please be as specific as possible in your response to the reviewer(s).

This link will remain active until you have submitted your revised manuscript. If you begin a revision and intend to finish it at a later time, please note that your draft will appear in the “Revised Manuscripts in Draft” queue in your Author Center.

IMPORTANT: Your original files are available to you when you upload your revised manuscript. Please delete any redundant files before completing the submission.

Because we are trying to facilitate timely publication of manuscripts submitted to Journal of Natural Fibers, your revised manuscript should be uploaded by 08-Mar-2018. If it is not possible for you to submit your revision by this date, we may have to consider your paper as a new submission.

Once again, thank you for submitting your manuscript to Journal of Natural Fibers and I look forward to receiving your revision.

Sincerely,
Professor Kozlowski
Editor in Chief, Journal of Natural Fibers
ryszard.kozlowski@escorena.net

Reviewer(s)' Comments to Author:
Reviewer: 1

Comments to the Author
in this manuscript, cellulose nanofibers was isolated from ramie (Boehmeria nivea (L.) Gaud) with the combination of chemical method and ultrasonication treatment, and the characterization of the morphology and crystal structure, thermal stability and size. However, the abstract is so simple, and English level is bad, so please improve it.

Reviewer: 2

Comments to the Author
Please, include in the text the yield of nanocellulose obtained of this process.
Editor's Comments to Author:

Associate Editor: 1
Comments to the Author:
(There are no comments.)

Respon utk Reviewer

Cellulose Nanofibers Isolation and Characterization from ramie using a chemical-ultrasonic treatment

Running head : Characterization of ramie (*Boehmeria nivea* (L.) Gaud)

Abstract
The nature of environmentally friendly, low-cost, lightweight and strengths of natural fibers make them an excellent choice to replace synthetic fibers. In this study, cellulose nanofibers was isolated from ramie (*Boehmeria nivea* (L.) Gaud) using chemical method–ultrasonication. The characterization of the morphology and crystal structure of the material were examined by scanning electron, transmission electron, and particle size analyzer. The results showed that cellulose nanofibers isolation successfully achieved with diameter of 9.9-89.1 nm and 1 μm length. The Fourier transform infrared spectroscopy analysis confirmed that hemicellulose and lignin levels decrease during the pulping and bleaching process, otherwise the cellulose levels increase. The X-ray powder diffraction observed that the ultrasonic treatment was increased the crystallization index from 62.5% to 73.65%. It is found that the ultrasonication is effective method in the production of nanocellulose from ramie with 89, 35% yield.

Keywords

Ramie; cellulose; nanofiber; natural fibers; chemical treatment; ultrasound

INTRODUCTION

Recently, the depletion of petroleum-based resources and serious environmental problems, such as climate change and global warming have lead researcher’s focus to develop the promising field of natural fibers. (Silitonga et al. 2016; Vignesh et al. 2016). This is largely owing to their biodegradability, environmental friendly, low cost, light weight and excellent specific strength (Wahono et al. 2018; Senthamaraikannan et al. 2016; Abral et al. 2014). Therefore, renewable source-based environmental has developed can be used to replace petroleum-based materials and conventional fibers such as glass by natural fibers in reinforced composites as a means of overcoming environmental problems (Manimaran et al. 2017; Cadena Ch et al. 2017).

Many researcher have isolated the highly-purified cellulose nanofibers from several various plant such as ramie (Lu et al. 2006), wood (Abe et al. 2007), sisal (Morán et al. 2008), pineapple leaf (Cherian et al. 2010), brazilian satintail (Benini et al. 2016), and Ipomoea staphylina (Santhanam et al. 2016). The essential common process for producing of cellulose nanofibers from plant fibers are pre-treatment and Nano fibrillation. There are a lot of pre-treatment to remove non cellulosic materials such as alkali treatment (Yongvanich 2015), enzymatic pre-treatment (Tibolla et al. 2017), ionic liquids (Han et al. 2013), TEMPO (2,2,6,6-tetramethylpiperidine-1-oxyl) mediated oxidation (Quintana et al. 2017) and steam explosion treatment (Chirayil et al. 2014). Then Nano fibrillation technologies such as grinders, and ultrasonic method (Khawas and Deka 2016) were used to isolated the fibrils from the purified cellulose fibers.

Nano fibrillation method has a high intensity ultrasonication (HIU) is suggested as method to isolate micro/nanofibers (Khawas and Deka 2016). The powerful mechanical oscillating power and high intensive waves in ultrasound will produce implosive collapse of bubble in liquid. That phenomenon called acoustic cavitation that provided 10-100 kJ mol-1 energy, which include most energy of a hydrogen bond scale. When the rapidly forming and collapsing microbubbles interact with and impact on the cellulose surface, they erode and denude the surface, which leads to fibrillation and formation of nanofibers (Kusumo et al 2017; Chen et al. 2011). Hence, the objective of this study aims to isolate cellulose nanofibers from ramie fibers by using a combination between chemical and high-intensity ultrasonication process to increase the efficiency of hemicellulose and lignin removal, and to reduce its size into Nano-order. Cellulose nanofibers produced in ramie fibers characterization and analyzed with SEM, TEM, XRD, FTIR, PSA and TGA.

MATERIALS AND METHODS

Materials

The ramie plant was collected from Politeknik Pertanian Payakumbuh, Indonesia with a land position of about 700 meters above sea level. The ramie fiber is taken by removing the bark (decortication) of the main stem. The reagent used in this process are as follows: sodium hydroxide (NaOH, purity >98%), potassium hydroxide (KOH, purity :99%), sodium chlorite ($NaClO_2$) and Acetic acid (CH_3COOH).

Isolation

Ramie fibers were first cut in small pieces (10 - 20mm) and dried under the sun for 3 days in order to reduce the water content to become 9-11%. The 500 g dried ramie fibers and 18 w/v % NaOH loaded into digester and cooked for 2 hours at 170 ^{o}C and 7-9 kg/cm^{2}bar, in order to remove the lignin and hemicellulose (Syafri et al. 2016). Then the obtained pulp ramie fibers were washed with distilled water until became neutralized pH. The two stages of different bleaching process were performed to remove the residual lignin and hemicellulose. In the first stage, the obtained pulp was added 5 w/v % natrium chloride ($NaClO_2$) and 3 ml acetic acid (CH_3COOH) for 2 hours at 170^{o}C until the pH value was 5. The obtained pulp remained after first bleaching treatment was washed using distilled water until the pH value was 7 and the second bleaching treatment was given using 4 w/v % potassium hydroxide (KOH) at 80^{o}C for 1 hours. The chemically purified cellulose ramie fibers loading with 2 w/v % of deionized water, and then sonicated using 1.5 cm cylindrical probe at a frequency of 22 kHz with an output power of 400W for 1 hours at temperature 70 ^{o}C. The details steps of chemical treatment combine with ultrasonication is isolated process of cellulose nanofiber from ramie are illustrated in Figure 1. The yield of nanocellulose was calculated using gravimetrically Eq. (1).

$$\text{Yield (\%)} = (\text{Weight of nanocellulose/Initial weight of native cellulose}) \times 100 \qquad (1)$$

Characterization

The chemical composition of the raw ramie (RR), after pulping treatment ramie (PP) and after bleaching ramie (BR) fibers was measured in accordance with the standards of the ASTM D 1104-56 (hemicellulose), TAPPI Standard T9M-54 (cellulose) and TAPPI Standard T3M-54 (lignin). The FT-IR analysis was performed by a Perkin Elmer FT-IR spectrometer frontier in the range of 400 - 4,000 cm^{-1}. Crystallinity was obtained with an PANalytical's X-ray diffractometers using Cu Kα radiation (λ=1.5406 A) at 40 kV and 30 mA, over the angular 2θ range 2-100^o and a step time of 20^o/min. The crystallinity index (CI) was calculated from the heights of the 200 peaks (I200, 2Θ = 22.6^o) and the intensity minimum between the 200 and 110 peaks (I_{am}, 2h = 18^o) using the Segal method (Gopinath et al. 2016;Segal 1959), as shown in Eq. 1, I_{002} represents both crystalline and amorphous material, whereas I_{am} represents the amorphous material.

$$C_I(\%) = \left(\frac{I_{002}-I_{am}}{I_{002}}\right) x100 \qquad (2)$$

Thermal degradation characteristic of CNFR was evaluated using Thermogravimetric Analysis (TGA4000, Perkin Elmer, USA) with a heating rate of 10^{o}C/min in range 50-500 ^{o}C under a flow of 60 mL/min of nitrogen gas. The surface morphology of the ramie fibers was observed using scanning electron microscopy (SEM, HITACHI 3400N) at 20 kV. The SEM specimens are prepared by placing a hemp fibers particle that has undergone some chemical treatment over a double-sided tape carbon in high vacuum mode. The surface morphology of the ramie fibers was observed using scanning electron microscopy (SEM, HITACHI 3400N) at 20 kV. The SEM specimens are prepared by placing a hemp fibers particle that has undergone some chemical treatment over a double-sided tape carbon in high vacuum mode. TEM (Transmission Electron Microscope) JEOL 1400 was used to evaluate the morphology of the cellulose nanofiber. The specimens were prepared by taking a few drops of an ultrasonified cellulose nanofibers suspension. The suspension is then placed on a thin carbon-coated copper housing. The CNFR cellulose suspension is allowed to dry above the container at room temperature then observed at 80 kV voltage acceleration. Beckman Coulter DelsaNano C instrument was performed to measure to cellulose ramie particles. Photon Correlation Spectroscopy (PCS) was used to determine the particle size by measuring the degree of fluctuation in the intensity of the laser beam spread by particles that spread through the liquid. The sample was measure at 25^oC, viscosity 0.88 cP, scattering intensity; 9581 cps, with refractive index; 1.3332 for 2 min. DelsaNano software used to process the particle measurement data.

RESULT AND DISCUSSIONS

Chemical analysis of ramie fibers

The chemically purified cellulose content successfully extracted by removal of lignin, hemicellulose and other impurities from raw ramie (RR) fibers using a chemically method (pulping and bleaching). The degradation of chemical composition of raw ramie (RR) fibers, pulping ramie (PR) fibers and bleaching ramie (BR) fibers were presented in Table 1. From table 1, it showed that the pulping and bleaching treatment increase 17.16% of cellulose content from 79.45% (RR), to 93.09 % (RB), whereas hemicellulose and lignin decreased 50 % from 9.63 % to 6.42 %. It is obviously evidence that $NaClO_2$, CH_3COOH, and KOH were found efficiently in removing most of lignin and hemicellulose in the fibers. This is due to the bond damage to the cell wall structure in pulping process. (C.S et al. 2016). Due to when the lignin reacts with $NaClO_2$will form a soluble lignin chloride compound (Chirayil et al. 2014), the remaining hemicellulose and lignin removed by $NaClO_2$and KOH in bleaching process. The content of lignin, hemicellulose and cellulose after bleaching process was 1.76 wt%, 6.42 wt% and 86.69 wt%, respectively.

Morphological property of chemically purified cellulose fibers analysis

SEM images of the raw ramie (RR), after pulping treatment ramie (PP), after bleaching ramie (BR) and cellulose nanofibers (CNFR) from chemical treatments and ultrasonication are presented in Figure 2. Figure 2a showed that the original ramie fibers has a large bundle with diameter 70-150 µm and intact structures covered with other impurities (e.g. hemicellulose, lignin, pectin and waxy substances). The morphological structure of the ramie fibers after pulping treatment (PR) differed significantly from the RR. After 2 h processing using pulping method, the fiber bundles were separated into individual micro-sized fibers and amorphous region termination which is marked by a significant decreasing diameter from 70-150 µm into 20-50 µm (Figure 2b).

After bleaching treatment, the fiber was separated into individual micro-sized cellulose fibers, as shown in Figure 2c. The average diameter size of bleached pulp fibers was reduced to 1-5 µm. The surface of bleached pulp fibers also found smoother. Thus, the removal impurities such as lignin and hemicellulose by delignification and bleaching processes could reduce the diameter of fiber from 20-50 µm to 1-5 µm, as well as disintegrated the single sized into individual micro-sized cellulose fibers. The separation of fasciculus induced by chemical treatment (pulping and bleaching) is due to the removal of lignin in the middle lamella that cased the disintegration of the compact ramie structure. It was evident that the chemical treatment was efficient in defibrillation of ramie fibers.

From the figure 2d it showed the individual long fibers entangled together forming like a web structure the presence of individual nanofibers (Figure 2d), it is evidence that Nano fibrillation force introduced by the ultrasonicated can break the hydrogen bonds and disintegrate microfibers into Nano fibrils in order to defibrillate the cellulose fibers which were obtained by the previous chemical treatment (Khawas and Deka 2016). It can be seen that fibrils associated with one another and formed a large aggregated bundle with a diameter of about 20-50 nm with 1µm length.

Particle Size Analyzer (PSA) analysis.

The PSA test data showed a uniform diameter of the haemoglobin diameter of CNFR with a diameter range between 9.9-89.1 nm, with an average diameter of 19 nm. Figure 3 shows that the diameter of 15.6 nm with the highest intensity number is 34.2%. This uniform distribution of cellulose diameter size for ramie fibers is similar with the study by Chen et al, on flax fibers with an ultrasonic-chemical process (Chen et al. 2011).

Fourier transform infrared (FT-IR) analysis

The FTIR spectra of the raw ramie (RR), after pulping treatment ramie (PP), after bleaching ramie (BR) and cellulose nanofibers (CNFR) from chemical treatments and ultrasonication are presented in Figure 4. The peaks at 3400-3300 cm^{-1}represents the stretching vibration of OH which indicate the hydrophilic properties of the material, while the peak of 2900-2800 cm^{-1} is attributed to the asymmetric stretching caused by the C-H group (Jonoobi et al. 2009).

The new functional group does not appear on the FTIR graph after the pulping and bleaching treatment. The peak 1625-1642 cm^{-1} is represented the bending mode of the absorbed water (O-H). In this study, the chemical treatment and ultrasonication was successfully performed to reduce the lignin and some of the hemicellulose due to the disappearance of this peak after treatment. In Figure 4., there is an increase in intensity at the peak of 898 cm^{-1} in ultrasonication treatment compared with chemical treatment and raw ramie. This indicates that the cellulose content is increased because in uptake of wave at the top position of 898 cm^{-1} is a cellulose glucose ring (Chirayil et al. 2014).

X-ray diffraction (XRD) analysis

The XRD diffraction patterns of raw ramie in every step of isolation into cellulose nanofibers processes were performed to investigate the crystallinity behavior of the fibers. The crystallinity index calculated using Equation 1. The result of the crystallinity was 55.48, 57.06, 62.50 and 73.65%.for RR, PR, BR and CNFR respectively (Table 2). It was observed that the crystallinity is affected by each process. The value of crystalline index increased to 57.06% after pulping treatment, which might have resulted from the dispersion and removal of non-cellulosic components including lignin and hemicelluloses that present in amorphous regions of raw fibers (C.S et al. 2016). After bleaching treatment, the crystalline index was increased to 62.50%, due to the elimination of retained lignin in the liquefied residues was complete. During ultrasonication the crystalline index increase to 73.64%. This is showed that the ultrasonication contribute to homogenize and remove part of amorphous portion of CNFR (Wen Shuai Chen et al. 2011). The result can be shown by the FTIR analysis and it is similar with the study on cellulose nanofibers from wood which reported by Chen et al (Chen et al. 2011).

Figure 5 illustrates the X-ray diffractogram of raw fibers, pulp, bleached pulp, and cellulose nanofibers of ramie. The peak of 2Ө: 15.710, 22.340, 32.00 for RR represent the typical cellulose I structure (Liu et al. 2007). According to JCPDS (Join Committee on Powder Diffraction Standards) it is seen that the resulting cellulose has a monoclinic crystal structure as follows: a = 6.74 (Å): b = 5.93 (Å): and c = 10.36(Å), angle γ:810. After chemical and ultrasonication treatment, angel 2 Ө changed to be 12.010, 19.830, and 21.960 represent the typical cellulose II structure. It observed obviously that the chemical and ultrasonication treatment transform the cellulose of raw ramie fibers from cellulose type I to cellulose type II (Liu et al. 2007). According to JCPDS the cellulose has crystal structure as follows: a = 7.87(Å):, b = 10.31 (Å): and c = 10.13(Å), angle γ: 900 .

Thermogravimetric analysis (TGA)

TGA analysis of reinforcing material is important to determine their applicability for bio composite processing at high temperature. TGA thermograms of ramie fibers from each treatment process are shown in Figure 6. From the figure, it showed that the CNFR has the highest thermal stability follows by BR, PR and RR. It is characterized by under the higher temperature which ramie residual fibers mass reduced and it is applicable to the temperature below 350 ^{o}C. In general, the thermal decomposition of cellulosic material components is at temperature 200 ^{o}C - 400 ^{o}C.

Initial decomposition for lignocellulose temperature of all cellulose nanofibers under various treatment levels had a little effect thermal decomposition of cellulose. The temperature of initial weight loss was found at range of 50-200 ^{o}C, it might be because of the evaporation and removal of bound water (Wang et al. 2017). The carbon residue was obtained at 500 ^{o}C of about 27% for RR, PR and BR, whereas for CNFR the carbon residue is less than 10%. The existence of the material residue is higher in the un-treatment ramie compared with the treatment ramie fibers due to the raw ramie contain a lots of ash, lignin which has a low degradation rate (Manfredi et al. 2006; Morán et al. 2008).

Nanocellulose Yield

Product yields, which were calculated gravimetrically using Eq. (1), are tabulated in Table 3. From Table 3 it showed that cellulose nanofibers using ultrasonic probe for 30 minutes and 60 minutes gives cellulose nanofibers yield 83,90% and 89,35% repectively. This result is 4.35% greater than yield achieved by habibi et al (85%) which makes cellulose nanofibers from ramie using PCL-grafted method (habibi et al. 2008).

CONCLUSION

The study revealed that the cellulose nanofibers is successfully isolated from ramie (Boehmeria nivea (L.) Gaud) using chemical method – ultrasonication with average diameter of 19 nm and 1 μm length. The chemical composition analysis showed that the hemicelluloses and lignin was significant decrease during the process. The FTIR analysis showed that the absorbent peak revealed at 897 cm-1 indicating the increasing of cellulose. From the TGA analysis, it showed that the cellulose nanofiber from ramie has the increasing thermal stability and the crystalline index was increased from 55.4 % to 73.65 %. It could be concluded that the chemical process (e.g. pulping followed with the two stages of different bleaching process) combined with ultrasonication method was successfully isolated cellulose nanofibers from ramie (Boehmeria nivea (L.) Gaud) with 89,39% yield.

References

Abe, K., S. Iwamoto, and H. Yano. 2007. Obtaining Cellulose Nanofibers with Uniform Width of 15 Nm from Wood, *Biomacromolecules* 8: 3276-3278.

Abral, H., D. Kadriadi, A. Rodianus, P. Mastariyanto, Ilhamdi, S. Arief, S. M. Sapuan and M. R. Ishak. 2014. Mechanical Properties of Water Hyacinth Fibers – Polyester Composites before and after Immersion in Water, *Materials & Design* 58: 125-129. doi:10.1016/j.matdes.2014.01.043.

Benini, K. C. C. C., H. J. C.Voorwald, M. O. H. Cioffi, A. C. Milanese, and H. L. Ornaghi, JR. 2016. Characterization of a New Lignocellulosic Fiber from Brazil: Imperata brasiliensis (Brazilian Satintail) as an Alternative Source for Nanocellulose Extraction, *Journal of Natural Fibers* 14 (1): 112-125. doi: 10.1080/15440478.2016.1167647.

C.S, J. C., N. George and S. K. Narayanankutty. 2016. Isolation and Characterization of Cellulose Nanofibrils from Arecanut Husk Fibre, *Carbohydrate Polymers* 142: 158-166. doi:10.1016/j.carbpol.2016.01.015.

Cadena Ch, E. M., J. M. Vélez R, J. F. Santa and V. Otálvaro G. 2017. Natural Fibers from Plantain Pseudostem (Musa Paradisiaca) for Use in Fiber-Reinforced Composites, *Journal of Natural Fibers*: 1-13. doi:10.1080/15440478.2016.1266295.

Chen, W., H. Yu, Y. Liu, P. Chen, M. Zhang and Y. Hai. 2011. Individualization of Cellulose Nanofibers from Wood Using High-Intensity Ultrasonication Combined with Chemical Pretreatments, *Carbohydrate Polymers* 83 (4): 1804-1811. doi:10.1016/j.carbpol.2010.10.040.

Cherian, B. M., A. L. Leão, S. F. de Souza, S. Thomas, L. A. Pothan and M. Kottaisamy. 2010. Isolation of Nanocellulose from Pineapple Leaf Fibres by Steam Explosion, *Carbohydrate Polymers* 81 (3): 720-725. doi:10.1016/j.carbpol.2010.03.046.

Chirayil, C. J., J. Joy, L. Mathew, M. Mozetic, J. Koetz and S. Thomas. 2014. Isolation and Characterization of Cellulose Nanofibrils from Helicteres Isora Plant, *Industrial Crops and Products* 59: 27-34. doi:10.1016/j.indcrop.2014.04.020.

Gopinath, R., K. Ganesan, S. S. Saravanakumar and R. Poopathi. 2016. Characterization of New Cellulosic Fiber from the Stem of Sida Rhombifolia, *International Journal of Polymer Analysis and Characterization* 21 (2): 123-129. doi:10.1080/1023666x.2016.1117712.

Habibi, Y., A.-L. Goffin, N. Schiltz, E. Duquesne, P. Dubois and A. Dufresne. 2008. Bionanocomposites Based on Poly (Caprolactone) Grafted Cellulose Nanocrystals by Ring Opening Polymerization, *Journal of Materials Chemistry* 18 (41): 5002-5010. doi:10.1039/b809212e.

Han, J., C. Zhou, A. D. French, G. Han and Q. Wu. 2013. Characterization of Cellulose Ii Nanoparticles Regenerated from 1-Butyl-3-Methylimidazolium Chloride, *Carbohydrate Polymers* 94 (2): 773-781. doi:10.1016/j.carbpol.2013.02.003.

Jonoobi, M., K. O. Niska, J. Harun, and M. Misra. 2009. Chemical Composition, Crystallinity, and Thermal Degradation of Bleached and Unbleached Kenaf Bast (Hibiscus Cannabinus) Pulp and Nanofibers, *BioResources* 4 (3): 629 - 639.

Khawas, P. and S. C. Deka. 2016. Isolation and Characterization of Cellulose Nanofibers from Culinary Banana Peel Using High-Intensity Ultrasonication Combined with Chemical Treatment, *Carbohydrate Polymers* 137: 608-616. doi:10.1016/j.carbpol.2015.11.020.

Kusumo F, Silitonga AS, Ong HC, Masjuki HH and Mahlia TMI. 2017. A comparative study of ultrasound and infrared transesterification of Sterculia foetida oil for biodiesel production. *Energy Sources, Part A: Recovery, Utilization, and Environmental Effects*. : 39(13):1339-46.

Liu, Z.-T., Y. Yang, L. Zhang, Z.-W. Liu and H. Xiong. 2007. Study on the Cationic Modification and Dyeing of Ramie Fiber, *Cellulose* 14 (4): 337-345. doi:10.1007/s10570-007-9117-0.

Lu, Y., Lihui Cao, and Xiaodong. 2006. Morphological, Thermal and Mechanical Properties of Ramie Crystallites Reinforced Plasticized Starch Biocomposites, *Carbohydrate Polymers* 63 (2): 198-204. doi:10.1016/j.carbpol.2005.08.027.

Manfredi, L. B., E. S. Rodríguez, M. Wladyka-Przybylak and A. Vázquez. 2006. Thermal Degradation and Fire Resistance of Unsaturated Polyester, Modified Acrylic Resins and Their Composites with Natural Fibres, *Polymer Degradation and Stability* 91 (2): 255-261. doi:10.1016/j.polymdegradstab.2005.05.003.

Manimaran. P, P.Senthamaraikannan, K.Murugananthan and M.R.Sanjay. 2017. Physicochemical properties of new cellulosic fibers from Azadirachta indica plant. *Journal of Natural Fibers.* DOI:10.1080/15440478.2017.1302388

Morán, J. I., V. A. Alvarez, V. P. Cyras and A. Vázquez. 2008. Extraction of Cellulose and Preparation of Nanocellulose from Sisal Fibers, *Cellulose* 15 (1): 149-159. doi:10.1007/s10570-007-9145-9.

Quintana, E., M. B. Roncero, T. Vidal and C. Valls. 2017. Cellulose Oxidation by Laccase-Tempo Treatments, *Carbohydrate Polymers* 157: 1488-1495. doi:10.1016/j.carbpol.2016.11.033.

Santhanam, K., A. Kumaravel, S. S. Saravanakumar and V. P. Arthanarieswaran. 2016. Characterization of New Natural Cellulosic Fiber from the Ipomoea Staphylina Plant, *International Journal of Polymer Analysis and Characterization* 21 (3): 267-274. doi:10.1080/1023666x.2016.1147654.

Segal, L., J. J. Creely, A. E. Martin, Jr., and C. M. Conrad. 1959. An Empirical Method for Estimating the Degree of Crystallinity of Native Cellulose Using X-Ray Diffractometer, *J. Text. Res* 29: 786–794.

Senthamaraikannan, P., S.S. Saravanakumar, V.P. Arthanarieswaran, and P. Sugumaran. 2016. Physicochemical properties of new cellulosic fibers from bark of *Acacia planifrons. International Journal of Polymer Analysis and Characterization* 21: 207-213.

Silitonga, A. S., H. H. Masjuki, H. C. Ong, T. Yusaf, F. Kusumo and T. M. I. Mahlia. 2016. Synthesis and Optimization of Hevea Brasiliensis and Ricinus Communis as Feedstock for Biodiesel Production: A Comparative Study, *Industrial Crops and Products* 85: 274-286. doi:10.1016/j.indcrop.2016.03.017.

Syafri, E., A. Kasim, H. Abral, A. Asben, and S. Wahono. 2016, Pengembangan Digester Pulp Untuk Menghasilkan Partikel Selulosa Serat Rami Sebagai Filler Material Bionanokomposit, *International Journal on Advanced Science, Engineering and Information Technology* 7(5):1950-1956. http://dx.doi.org/10.18517/ijaseit.7.5.1292

Syafri, E., A. Kasim, H. Abral and A. Asben. 2017. Effect of Precipitated Calcium Carbonate on Physical, Mechanical and Thermal Properties of Cassava Starch Bioplastic Composites

Tibolla, H., F. M. Pelissari, M. I. Rodrigues and F. C. Menegalli. 2017. Cellulose Nanofibers Produced from Banana Peel by Enzymatic Treatment: Study of Process Conditions, *Industrial Crops and Products* 95: 664-674. doi:10.1016/j.indcrop.2016.11.035.

Vignesh, V., A. N. Balaji and M. K. V. Karthikeyan. 2016. Extraction and Characterization of New Cellulosic Fibers from Indian Mallow Stem: An Exploratory Investigation, *International Journal of Polymer Analysis and Characterization* 21 (6): 504-512. doi:10.1080/1023666x.2016.1175206.

Wahono, S., A. Irwan, E. Syafri and M. Asrofi. 2018. Preparation and Characterization of Ramie Cellulose Nanofibers/c_aco_3 unsaturated polyester resin composites, *ARPN Journal of Engineering and Applied Sciences* 13 (2): 746 – 751.

Wang, L.-F., S. Shankar and J.-W. Rhim. 2017. Properties of Alginate-Based Films Reinforced with Cellulose Fibers and Cellulose Nanowhiskers Isolated from Mulberry Pulp, *Food Hydrocolloids* 63: 201-208. doi:10.1016/j.foodhyd.2016.08.041.

Chen, W.S., H. P. Yu., P. Chen, N. X. Jiang, J. H. Shen, Y. X. Liu, and Q. Li. 2011. Preparation and Morphological Characteristics of Cellulose Micro/Nano Fibrils, *Materials Science Forum* 675-677: 255-258.

Yongvanich, N. 2015. Isolation of Nanocellulose from Pomelo Fruit Fibers by Chemical Treatments, *Journal of Natural Fibers* 12 (4): 323-331. doi:10.1080/15440478.2014.920286.

ecision Letter (WJNF-2017-0179.R1)

From: ryszard.kozlowski@escorena.net

To: edisyafri11@gmail.com

CC:

Subject: Journal of Natural Fibers - Decision on Manuscript ID WJNF-2017-0179.R1

Body: 15-Mar-2018

Dear Mr Syafri:

Ref: Cellulose Nanofibers Isolation and Characterization from ramie using a chemical-ultrasonic treatment

Our reviewers have now considered your paper and have recommended publication in Journal of Natural Fibers. We are pleased to accept your paper in its current form which will now be forwarded to the publisher for copy editing and typesetting. The reviewer comments are included at the bottom of this letter, along with those of the editor who coordinated the review of your paper.

You will receive proofs for checking in due course.

Once the Taylor & Francis production department receives and performs an initial check on your article, they will send you a link to complete your online article publishing agreement. This is an essential step. Your completed agreement must be accepted by the publisher before we can publish any version of your paper.

The publisher also requests that proofs are checked through the publisher's tracking system and returned within 48 hours of receipt.

Thank you for your contribution to Journal of Natural Fibers and we look forward to receiving further submissions from you.

Sincerely,
Professor Kozlowski
Editor in Chief, Journal of Natural Fibers
ryszard.kozlowski@escorena.net

Reviewer(s)' Comments to Author:

Editor's Comments to Author:

Associate Editor
Comments to the Author:
The manuscript files have been checked. The replies to the reviewer are satisfactory. The article seems to be corrected in a good manner and ready for publication. Please, be ready to send to the publisher the tables and figures in separate files, having good production quality.

/.

Date Sent: 15-Mar-2018

Reminder: Your Revision for Journal of Natural Fibers is pending Kotak Masuk ×

Journal of Natural Fibers <onbehalfof@manuscriptcentral.com> Jum, 23 Feb 2018, 12.52

kepada saya

Inggris > Indonesia Terjemahkan pesan Nonaktifkan untuk: Inggris ×

23-Feb-2018

Dear Mr Edi Syafri:

Recently, you received a decision on Manuscript ID WJNF-2017-0179, entitled "Cellulose Nanofibers Isolation and Characterization from ramie using a chemical-ultrasonic treatment." This email is simply a reminder that your revision is due on 08-Mar-2018.

To start the revision, please click on the link below:

*** PLEASE NOTE: This is a two-step process. After clicking on the link, you will be directed to a webpage to confirm. ***

https://mc.manuscriptcentral.com/jnf?URL_MASK=ae158742bffe4036a5a965ec8f0bd1dc

This will direct you to the first page of your revised manuscript in your Author Center. The manuscript and decision letter are located here also.

This link will remain active until you have submitted your revised manuscript. If you have already begun a revision, you can click on the link to continue your revision. Please note that your draft will appear in the "Revised Manuscripts in Draft" queue in your Author Center.

If it is not possible for you to submit your revision by 08-Mar-2018, we will consider your paper as a new submission.

Please contact the Editorial Office if you are unable to submit within this time.

From: **Journal of Natural Fibers** <onbehalfof@manuscriptcentral.com>
Date: 2018-03-15 20:38 GMT+07:00
Subject: Journal of Natural Fibers - Decision on Manuscript ID WJNF-2017-0179.R1
To: edisyafri11@gmail.com

15-Mar-2018

Dear Mr Syafri:

Ref: Cellulose Nanofibers Isolation and Characterization from ramie using a chemical-ultrasonic treatment

Our reviewers have now considered your paper and have recommended publication in Journal of Natural Fibers. We are pleased to accept your paper in its current form which will now be forwarded to the publisher for copy editing and typesetting. The reviewer comments are included at the bottom of this letter, along with those of the editor who coordinated the review of your paper.

You will receive proofs for checking in due course.

Once the Taylor & Francis production department receives and performs an initial check on your article, they will send you a link to complete your online article publishing agreement. This is an essential step. Your completed agreement must be accepted by the publisher before we can publish any version of your paper.

The publisher also requests that proofs are checked through the publisher's tracking system and returned within 48 hours of receipt.

Thank you for your contribution to Journal of Natural Fibers and we look forward to receiving further submissions from you.

Manuscripts with Decisions

ACTION	STATUS	ID	TITLE	SUBMITTED	DECISIONED
	ADM: Maciejowska, Izabela • Accept (15-Mar-2018) • Awaiting Production Checklist view decision letter	WJNF-2017-0179.R1	Cellulose Nanofibers Isolation and Characterization from ramie using a chemical-ultrasonic treatment View Submission	04-Mar-2018	15-Mar-2018
a revision has been submitted (WJNF-2017-0179.R1)	ADM: Maciejowska, Izabela ADM: Mackiewicz-Talarczyk, Maria • Minor Revision (15-Feb-2018) • a revision has been submitted	WJNF-2017-0179	Cellulose Nanofibers Isolation and Characterization from ramie using a chemical-ultrasonic treatment View Submission	03-Oct-2017	15-Feb-2018

Publish

From: **ParagSharma.c.IshwarLalSharma@taylorandfrancis.com** <cats@taylorandfrancis.com>
Date: 2018-03-26 22:04 GMT+07:00
Subject: Your article proofs for review (ID# WJNF 1455073)
To: edisyafri11@gmail.com

26 Mar 2018

Proofs of your article(s) listed below are now available for review through the Central Article Tracking System (CATS) website:

• Journal: WJNF: *Journal of Natural Fibers*

• Manuscript ID: 1455073 (WJNF-2017-0179.R1)

• Manuscript Title: Cellulose Nanofibers Isolation and Characterization from ramie using a chemical-ultrasonic treatment

• By: Syafri; Kasim; Abral; Asben

Please approve these proofs, or return any corrections by 29 Mar 2018. Failure to do so may result in delay of your publication, reallocation to a later issue, or review and approval of your article by the journal's Editor-in-Chief.

• Please follow the link below to view the manuscript through the CATS system. https://cats.informa.com/PTS/in?ut=624C12A2B5084D258D8CC9B1C5BFAF8E

• Your User Name is: SYAFRIE

• If you do not know your password, you may reset it here: http://cats.informa.com/PTS/forgottenPassword.do

Once logged into CATS you will be able to view the 'My Manuscripts' page with the article(s) currently in production where you are listed as an author. Clicking on 'Review Proofs' will allow you to access your article. Please use the web form (if the corrections are fewer than 50 in number) to return any corrections to me. Instructions on how to use the web form can be found by clicking on the 'Proofing Guidelines for Authors' link located at the bottom of the window in which your proofs appear. Alternatively, send your corrections in an email to me or you can print the article and fax or post it to the address listed below.

When the article publishes, each author will receive 50 e-prints to share with colleagues. This will enable you to give any friend, colleague, or contact free access to an electronic version of your article.

Hardcopy reprints of your article are available with special pricing for authors, and authors may also order print copies of the issue in which the article appears. Orders may be placed by logging in to your CATS account and accessing the order form on the "Additional Actions" menu. If you have any questions on this process, please contact me.

PLEASE NOTE: The CATS system only supports Internet Explorer 6 (and later), or Firefox 3 (and later) browser software. Popup blockers should be disabled. If you have any difficulty using CATS, please contact me.

Thank you,

Parag Sharma Ishwar Lal Sharma

Taylor & Francis Group
530 Walnut Street
Suite 850
Philadelphia
PA
19106
Email:ParagSharma.c.IshwarLalSharma@taylorandfrancis.com

JOURNAL OF NATURAL FIBERS
https://doi.org/10.1080/15440478.2018.1455073

Taylor & Francis
Taylor & Francis Group

Check for updates

Cellulose nanofibers isolation and characterization from ramie using a chemical-ultrasonic treatment

Edi Syafri[a], Anwar Kasim[b], Hairul Abral[c], and Alfi Asben[b]

[a]Department of Agricultural Technology, Politeknik Pertanian, Payakumbuh, Indonesia; [b]Department of Agriculture Product Technology, Universitas Andalas, Limau Manis, Indonesia; [c]Department of Mechanical Engineering, Universitas Andalas, Kampus Limau Manis, Indonesia

ABSTRACT

The novelty of this work lies in isolating cellulose nanofibers from ramie (*Boehmerianivea* (L.) Gaud) using chemical-ultrasonication treatment. The cellulose nanofibers were successfully isolated from ramie with a diameter and length of 9.9–89.1 nm and 1 μm, respectively. The samples were characterized by scanning electron microscopy, transmission electron microscopy, particle size analysis, Fourier transform infrared spectroscopy, X-ray diffraction analysis, and thermogravimetric analysis. Indeed, there is a decrease in the hemicellulose and lignin content while the cellulose content increases due to the pulping and bleaching processes. The chemical-ultrasonication treatment results in a high yield of cellulose nanofibers (89.35%).

摘要

本文的新颖之处在于从苎麻中分离纤维素纳米纤维（Boehmerianivea（L.）Gaud）使用化学超可听化处理。分别从直径为9.9～89.1 nm和1μm的苎麻中分离出纤维素纳米纤维。通过扫描电镜、透射电镜、粒度分析、傅立叶变换红外光谱、X射线衍射分析、热重分析等手段对样品进行了表征。事实上，半纤维素和木质素含量减少，而纤维素含量的增加由于制浆和漂白过程。在高产量的纤维素纳米纤维化学超声治疗结果（89.35%）。

KEYWORDS

Ramie; cellulose; nanofibers; natural fibers; chemical treatment; ultrasonication

关键词

纤维素；纳米纤维；天然纤维；化学处理；超声波处理