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Cellulose nanowhiskers extracted from TEMPO-oxidized jute fibers

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ABSTRACT

Cellulose nanowhiskers is a kind of renewable and biocompatible nanomaterials evoke much interest because of its versatility in various applications. Here, for the first time, a novel controllable fabrication of cellulose nanowhiskers from jute fibers with a high yield (over 80%) via a 2,2,6,6-tetramethylpiperidine-1-oxyl radical (TEMPO)/NaBr/NaClO system selective oxidization combined with mechanical homogenization is reported. The versatile jute cellulose nanowhiskers with ultrathin diameters (3–10 nm) and high crystallinity (69.72%), contains C6 carboxylate groups converted from C6 primary hydroxyls, which would be particularly useful for applications in the nanocomposites as reinforcing phase, as well as in tissue engineering, pharmaceutical and optical industries as additives.

1. Introduction

In recent years, cellulose nanofibers which have abundant and renewable natural resources have attracted wide attention (Deng et al., 2010; Mao, Ding, Wang, & Yin, 2010) because of their super functionalities for various applications (e.g. green nanocomposites, optically transparent functional materials, tissue engineering scaffolds, catalysts, textiles, surface coatings, drug delivery, food package), due to their extremely large and active surface areas (Cao, Dong, & Li, 2007; Das et al., 2010; Dong et al., 2012; Lu & Hsieh, 2009).

Up till now, many fabrication methods for isolating cellulose nanowhiskers from various plant fibers have been attempted. One of the most common methods is strong acid hydrolysis, through which cellulose nanocrystals with a comparatively low yield of about 30% and having lengths of 100–200 nm and widths of 20–40 nm could be extracted from many kinds of cellulose resources including cotton (Teixeira et al., 2010), ramie (Menezes De, Siqueira, Curvelo, & Dufresne, 2009), sisal (Morán, Alvarez, Cyras, & Vázquez, 2008), mengkuang leaves (Sheltamia, Abdullaha, Ahmada, Dufresne, & Kargarzadeh, 2012), wood pulp (Dong, Kimura, Revol, & Gray, 1996), jute (Das et al., 2011), banana leaves (Elanthikkal, Gopalakrishnapanicker, Varghese, & Guthrie, 2010), and mulberry (Li et al., 2009). In addition, high shear refining and cryocrushing process could obtain cellulose nanofibers from several wood pulps with a comparatively uneven diameter in the range from several to hundreds nanometers (Chakraborty, Sain, & Kortschot, 2005).

Recently, the 2,2,6,6-tetramethyl-1-piperidinyloxy (TEMPO) radical, which was found to selectively oxidize the primary alcohol groups in aqueous media, was applied to several wood pulps and cotton linters (Fukuzumi, Saito, Iwata, Kumamoto, & Isogai, 2009; Saito, Nishiyama, Putaux, Vignon, & Isogai, 2006). The oxidation proceeded throughout the fibers but occurred only at the surface of the microfibrils (Saito, Shibata, Isogai, Suguri, & Sumikwa, 2005). Combined with mechanical disintegration, aqueous dispersions of individual cellulose nanofibers can be obtained from the TEMPO-oxidized cellulose fibers with a high yield of over 80% (Isogai, Saito, & Fukuzumi, 2011). However, this method seems unsuccessful to directly apply to the non-wood fibers with high crystalline and polymerization degree such as sisal, jute and hemp.

For environmental protection as well as government regulations in some countries, the great importance and developmental prospect of rationally utilizing non-wood fibers has become increasingly significant. Accordingly, the study in the field concerned has also made more and more progress. As a typical kind of non-wood fibers, jute is widely cultured in some Asian countries (e.g. China, India, and Bangladesh) and can be harvested two or three times annually, thereby resulting in significant high yield and low cost. In our previous work, we mainly focused on the study

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of jute fibers modification and their reinforced biodegradable polymer composites (Liu, Yu, Cheng, & Qu, 2009; Liu, Yu, Cheng, & Yang, 2009; Xia, Yu, Cheng, Liu, & Wang, 2009). The higher cellulose content (45–71.5%) (Li, Tabil, & Panigrahi, 2007) and crystallinity (especially higher than other non-wood fibers) of jute fibers indicates its suitability in the preparation of cellulose nanowhiskers (Jahan, Saeed, He, & Ni, 2009). To our knowledge, most researches on jute cellulose nanocrystals extraction were based on acid hydrolysis (Das et al., 2011, 2010; Maiti, Ray, Mitra, & Misra, 2012) and steam explosion (Wang, Huang, & Lu, 2009) in previous literature, and there was no report on the preparation of jute cellulose nanowhiskers by TEMPO-oxidization combined with mechanical treatment.

In the present investigation, cellulose nanowhiskers were extracted from chemically pretreated jute fibers by combination of TEMPO/NaBr/NaClO system selective oxidization and mechanical homogenization. The effects of different chemical treatments on jute were then investigated by Fourier transform infrared (FTIR) spectroscopy, solid-state ^{13}C nuclear magnetic resonance (NMR), X-ray diffraction (XRD), and thermogravimetric analysis (TGA). The optical transmittance of TEMPO-oxidized cellulose nanowhiskers suspension was measured by UV–vis spectrometer. The detailed dimensions of the isolated cellulose nanowhiskers were investigated by transmission electron microscopy (TEM).

2. Experimental

2.1. Raw materials

Jute bast fibers used in this study were supplied by Redbud Textile Tech. Inc., Jiangsu, China, which sieved under 60 mesh were vacuum dried. Sodium hydroxide (NaOH), dimethylsulfoxide (DMSO), 2,2,6,6-tetramethylpiperidine-1-oxyl radical (TEMPO, 98%), sodium bromide (NaBr), 12 wt% sodium hypochlorite (NaClO) solution, ethanol and other chemicals were of laboratory grade (Shanghai Aladdin Chemical Regent Inc., China) and used without further purification.

2.2. Pretreatment of jute fibers

To remove most noncellulosic constituents, grinded jute fibers powder with 0.3 mm particle size was soaked in a 15 wt% NaOH solution and heated at 60 °C for about 4 h. After fully washing with distilled water, the mass was dried at 70 °C for 24 h. The dried mass was then dipped in DMSO (liquor ratio 1:15) followed by heating at 70 °C on a water bath. Again after washing several times with distilled water, it was oven-dried at 70 °C for 24 h.

2.3. Preparation of cellulose nanowhiskers suspension

Preparation of cellulose nanowhiskers suspension was performed according to the procedure described by (Ma, Burger, Hsiao, & Chu, 2011). The reaction between TEMPO/NaClO/NaBr system and jute cellulose is shown in Fig. 1. The C6 primary hydroxyls of cellulose would be oxidized to C6 carboxylate groups by TEMPO/NaBr/NaClO in water at pH 10–10.5 (Isogai et al., 2011). The aforementioned jute sample (10 g) was dispersed in the solution with water (190 g), NaBr (0.20 g) and TEMPO (0.02 g). The reaction was started by the addition of 12% NaClO solution (18 g) under stirring. The pH was kept at 10–10.5, monitored with a pH meter, by adjusting with 2 wt% NaOH aqueous solution. The reaction was stopped by adding 5 mL ethanol while no NaOH consumption, followed by stirring for another 20 min. The final product was washed with deionized water by successive centrifugations (5000 rpm for 10 min) until neutral. Oxidized cellulose slurry (1.0 g) was dispersed in 100 g of water and sonicated for 5 min at 13,000 rpm with an IKA T25 homogenizer (IKA Works, Shanghai, China). The suspension was centrifuged at 5000 rpm for 30 min, and the recovered supernatant became the cellulose nanowhiskers aqueous suspension. The solid content of the supernatant was measured by drying at 105 °C for 3 h to determine the yield (%) of cellulose nanowhiskers based on the dry weight of the oxidized cellulose.

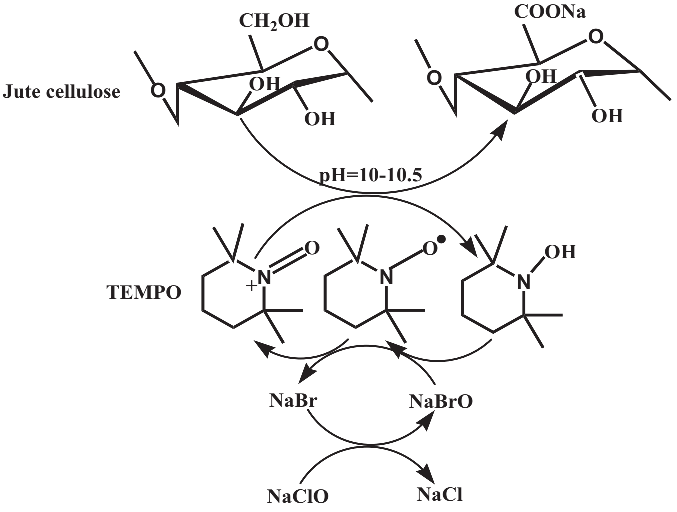

Fig. 1. Selective oxidation of C6 hydroxyls of jute cellulose to C6 carboxylate groups by TEMPO/NaBr/NaClO system.

2.4. Characterization

2.4.1. Optical transmittance of cellulose nanowhiskers suspension

Cellulose nanowhiskers suspension was introduced into a poly(methyl methacrylate) disposable cuvette, and the transmittance was measured from 300 to 900 nm using a UV–vis spectrometer (Lambda 35, Perkin Elmer, U.S.A.). The spectrum of a cuvette filled with water was used as a reference and to correct the transmittance of the suspension sample.

2.4.2. Optical microscopy of jute fibers

The dimensions of untreated jute fibers and grinded jute fibers were observed using an optical microscope (Olympus BX50) with a digital camera.

2.4.3. Transmission electron microscopy (TEM) of cellulose nanowhiskers

A 10 μL drop of the 0.1 wt% cellulose nanowhiskers dispersion was mounted on a glow-discharged carbon-coated Cu grid. The excess liquid was absorbed by filter paper, and one drop of 2 wt% uranyl acetate negative stain was added before drying. Excess solution was blotted out with filter paper and allowed to stand for drying by natural evaporation. The sample grid was observed by a Philips CM 120 transmission electron microscope with an acceleration voltage of 60 kV. Micrograph was taken on Fuji FG films.

2.4.4. Fourier transform infrared (FTIR) spectroscopy

The FTIR spectra of all samples were recorded on an attenuated total reflection fourier transform infrared (ATR-FTIR) instrument (Nicolet Nexus 670, Thermo Fisher, U.S.A.) equipped with smart iTR module in the range of 400–4000 cm^{-1} with a resolution of 4 cm^{-1}.

2.4.5. X-ray diffraction (XRD) measurement

In order to investigate the crystallinity of untreated jute, alkali treated jute and jute cellulose nanowhiskers, the milled sample powders were analyzed at ambient temperature by step scanning on a X-ray diffractometer (D/max-2550 PC, Rigaku, Japan) using a monochromatic Cu K_{α} radiation ($\lambda = 1.5406$ Å) in the range of $2\theta = 6$–$40°$ with a step size of 0.04° and a scanning rate of 5.0°/min. MDI Jade 5.0 software was used to calculate the crystallinity of each sample.

2.4.6. Solid-state ^{13}C nuclear magnetic resonance (NMR)

Solid-state ^{13}C NMR spectra of untreated jute fibers and TEMPO-oxidized jute nanowhiskers were recorded on Avance 400 Bruker spectrometer (Bruker Inc., Switzerland) with a cross polarization/magic angle spinning (CP/MAS) unit at ambient temperature.

2.4.7. Thermal gravimetric analysis (TGA)

Thermogravimetric analyses were performed using a TA Instruments (TG209 F1, NETZSCH, Germany). In a typical experiment, about 5 mg of samples were placed in a clean platinum pan and heated from 20 to 600 °C at a rate of 10 °C/min.

3. Results and discussion

3.1. Morphology of jute fiber and cellulose nanowhiskers

The jute fibers are long filaments composed of numbers of short ultimate cells which are cemented by lignin and hemicellulose (Xia et al., 2009). Fig. 2a and b shows the optical micrograph of untreated jute and grinded jute, respectively. It could be seen that the diameter of jute fiber was approximately 50 μm, and after mechanically grinded, fibers became thinner and shorter, even some in diameter of 10 μm. The jute nanowhiskers prepared by TEMPO-oxidization were examined by TEM to determine their size and shape. The TEM micrograph of a 0.1 wt% suspension of cellulose nanowhiskers from jute (Fig. 3) reveals that most fibrils are defibrillated to nanofibers from the cell walls. It can be seen that the cellulose rods are agglomerated in some places due to the high specific areas and strong hydrogen bonds established between the nanofibrils, while in some other places they are separated well. The dimension of elongated cellulose nanowhiskers are 3–10 nm in width and 100–200 nm in length, which is extremely thinner than that prepared by acid hydrolysis (Jahan et al., 2009; Maiti et al., 2012). One important thing has to note that the cellulose nanowhiskers yield is calculated to over 80%, which is similar with that of dried sulfite pulps prepared by (Saito et al., 2006).

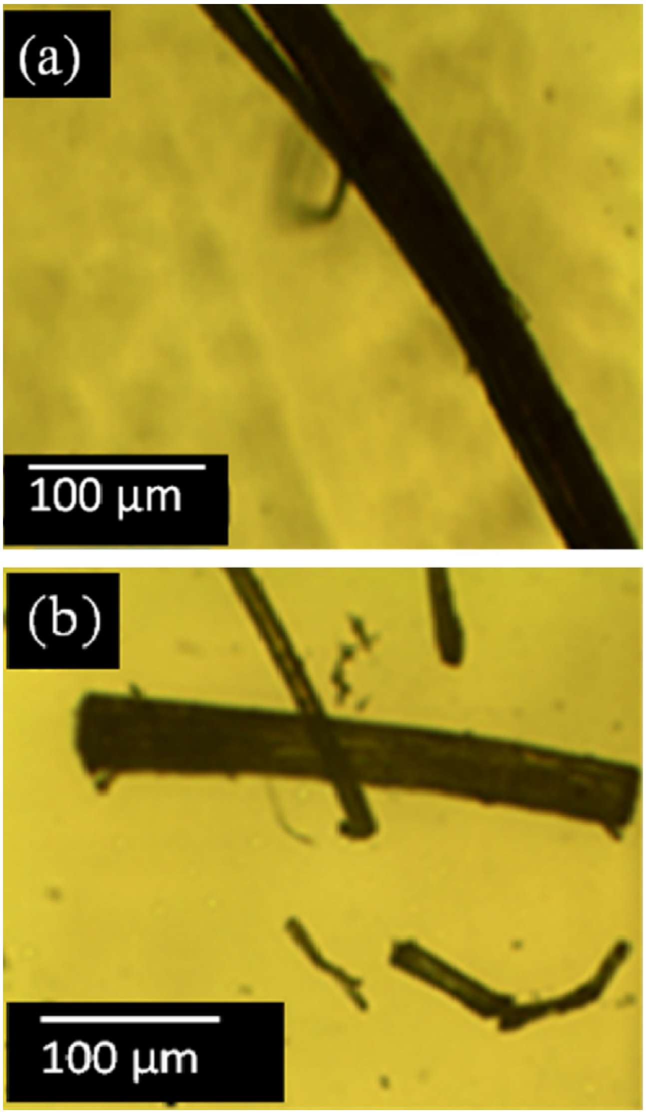

Fig. 2. Optical micrograph of (a) jute fibers and (b) grinded jute fibers.

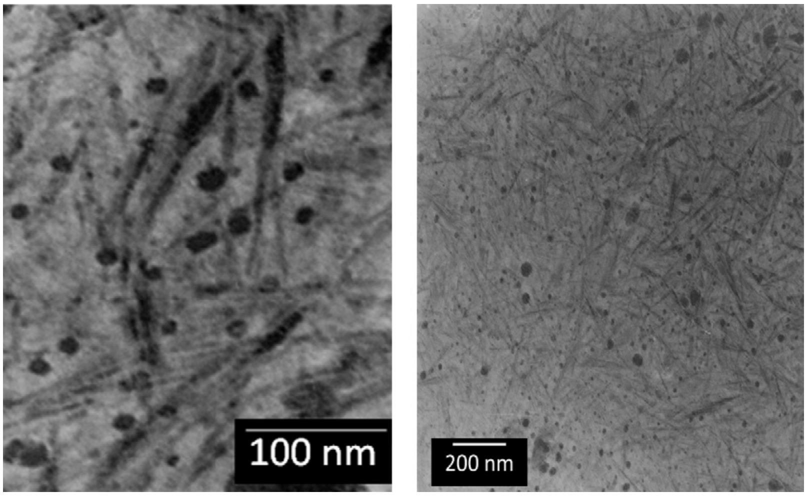

Fig. 3. TEM micrograph of jute cellulose nanowhiskers (at two magnifications).

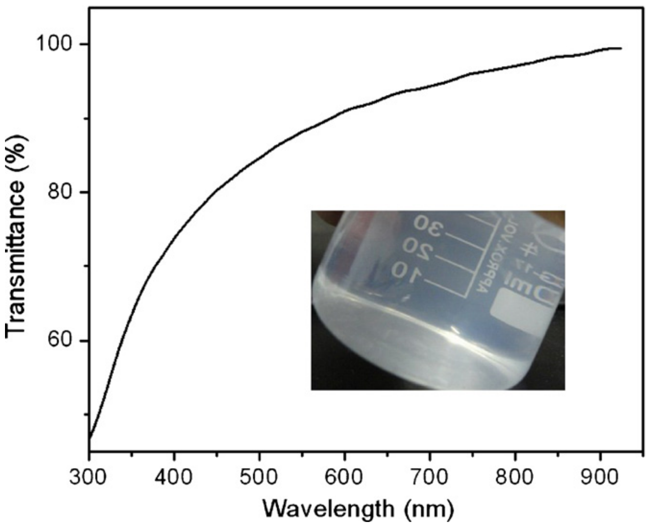

Fig. 4. UV–vis transmittance of 0.1 wt% cellulose nanowhiskers suspension prepared from TEMPO-oxidized jute. The inset photograph shows light transmittance behavior of the cellulose nanowhiskers suspension.

3.2. *Optical transmittance of nanowhiskers suspension*

The UV–vis transmittance of 0.1 wt% cellulose nanowhiskers suspension prepared from TEMPO-oxidized jute fibers and the digital photographs of the suspensions of the obtained nanowhiskers in a beaker are shown in Fig. 4. The transmittance of the 0.1 wt% cellulose nanowhiskers suspension at 600 nm was approximately 90%. The transmittance is wavelength-dependent and is lower at shorter wavelengths, as light scatters more when the wavelength approaches the diameter of the particles. For suspended fibrils or rods thinner than the wavelength, the light scattering is proportional to the mass/length ratio or the cross section area (Marcus, Linus, & Jan, 1977). Treating jute fibers with TEMPO/NaBr/NaClO aqueous solution involved oxidization of hydroxyl groups and introduction of carboxylate groups along the surface of the crystallites, which would result in a negative charge of the surface. This anionic stabilization via the attraction/repulsion forces of the electrical double layers at the crystallites is probably the reason for the stability of the transparent nanowhiskers suspension (Marchessault, Morehead, & Koch, 1961).

3.3. *FTIR spectroscopy analysis*

The investigation of chemical structure change in the jute fibers after alkali and TEMPO oxidized treatment was carried out through FT-IR absorption spectroscopy. FTIR-ATR spectra of the untreated jute, alkali treated jute and jute nanowhiskers are shown in Fig. 5. Both the jute control sample and alkali treated jute have no absorption band from 1400 to 1700 cm^{-1}, but two strong absorption bands at 1601 and 1410 cm^{-1} derived from the carbonyl groups present in response to the TEMPO-mediated oxidation of cellulose nanowhiskers, indicating that hydroxyl groups at the C6 position of cellulose molecules are converted to sodium carboxylate (Ifuku, Tsuji, Morimoto, Saimoto, & Yano, 2009). Meanwhile, all samples exhibits a broad band in the region of 3500–3200 cm^{-1} due to the free O–H stretching vibration of hydroxyl groups in cellulose molecules, again indicating not all O–H groups joining the reaction but a selective oxidation of the primary OH units.

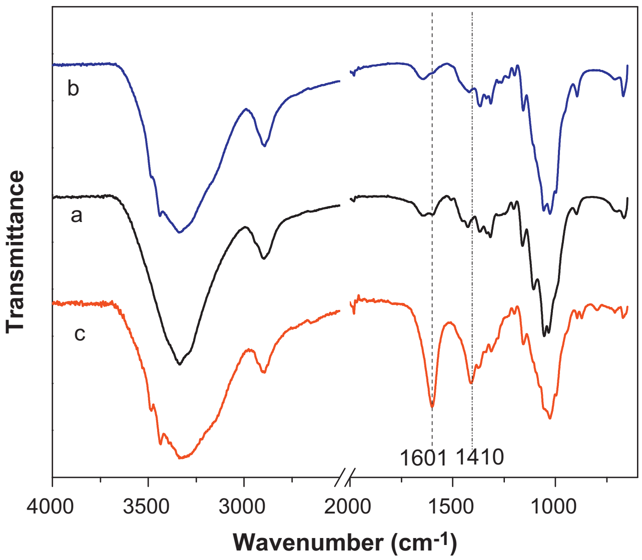

Fig. 5. FTIR spectra of (a) untreated jute; (b) alkali treated jute; and (c) jute cellulose nanowhiskers.

3.4. ^{13}C *NMR spectrum analysis*

Solid-state ^{13}C NMR spectra of jute fibers and TEMPO-oxidized cellulose sample are shown in Fig. 6. For the control jute, the typical signals of cellulose appear at 104.8 (C-1), 88.5 and 83.5 (C-4), 74.5 and 72.1 (C-2, C-3, and C-5), and 64.8 and 62.7 ppm (C-6). It can be seen that there is a new peak due to sodium carboxylate carbons in the oxidized cellulose nanowhiskers at 174.7 ppm compared with the control jute (Saito et al., 2005). The TEMPO

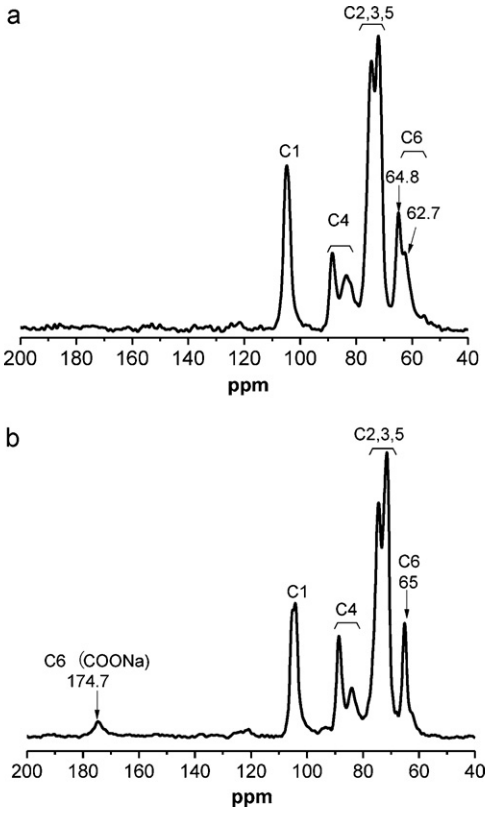

Fig. 6. Solid-state ^{13}C NMR spectra of (a) untreated jute fibers and (b) jute cellulose nanowhiskers.

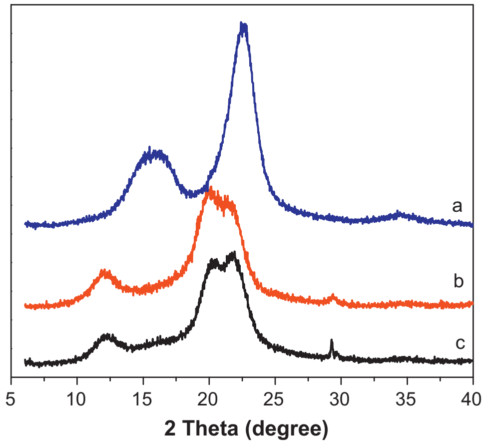

Fig. 7. X-ray diffraction patterns for (a) untreated jute; (b) alkali treated jute; and (c) jute cellulose nanowhiskers.

oxidized jute also has large resonance peaks due to the original C6 primary alcohol groups of the glucose unit at about 65.2 ppm, little or only slight oxidation occurred at the C6 primary alcohol groups of jute in TEMPO/NaBr/NaClO system (Isogai & Kato, 1998). Meanwhile, the signal at 62.7 ppm sharply decreases compared with the spectrum of the control jute, indicating a selective oxidation of the primary OH-units (Vieira, Heinze, Antonio-Cruz, & Mendoza-Martinez, 2002).

3.5. X-ray diffraction analyses

The crystallinity of untreated jute fibers, alkaline treated jute, and oxidized jute cellulose nanowhiskers has been analyzed by X-ray diffractometry. It can be noted from Fig. 7 that the fibers show increasing orientation along a particular axis as the noncellulosic polysaccharides are removed and the amorphous zones are dissolved. The crystallinity thus goes on increasing in going from the untreated jute (63.73%) to alkali-treated jute (66.08%), and subsequently to oxidized jute cellulose nanowhiskers (69.72%). The little increasing in the crystallinity after TEMPO oxidization, which is due to partial loss of the disordered regions during the washing process because of their increased water-solubility, was also observed by (Saito & Isogai, 2004). The high crystalline jute cellulose nanowhiskers could be more effective in providing better reinforcement for composite materials because the high Young's modulus (138 GPa) of the crystal region along the longitudinal direction (Sakurada, Nukushina, & Ito, 1962).

It can be also found that the XRD pattern of the control jute fibers exhibits a sharp high peak at $2\theta = 22.5^\circ$, and two overlapped weaker diffraction peaks respectively at $2\theta = 14.8^\circ$ and 16.4°, which are assigned to cellulose I. However, for the 15 wt% NaOH treated jute, two additional diffraction peaks appear at 12.2° and 20.2°, which are assigned to cellulose II. Alkali treatment results a lattice transformation from cellulose I to cellulose II, which was also observed by (Liu & Hu, 2008).

3.6. Thermogravimetric analysis (TGA)

Thermal decomposition properties were determined from the TG and DTG curves as described below at a heating rate of 10 °C/min. Fig. 8 shows the TG and DTG curves of untreated jute, alkali-treated jute and TEMPO-oxidized cellulose nanowhiskers. All the TG curves showed an initial small drop between 30 and 150 °C, which corresponded to a weight loss of approximately 5% absorbed moisture on the surfaces of these materials including chemisorbed water and/or the intermolecularly H-bonded water. Thermal degradation of the TEMPO-oxidized jute cellulose nanowhiskers started at approximately 200 °C in a N_2 atmosphere, while for the untreated jute and alkali-treated jute, degradation began at approximately 270 °C. Thus, the formation of sodium carboxylate groups from the C6 primary hydroxyls of cellulose microfibril surfaces by TEMPO-mediated oxidation leads to a significant decrease in the thermal degradation point (Fukuzumi, Saito, Okita, & Isogai, 2010).

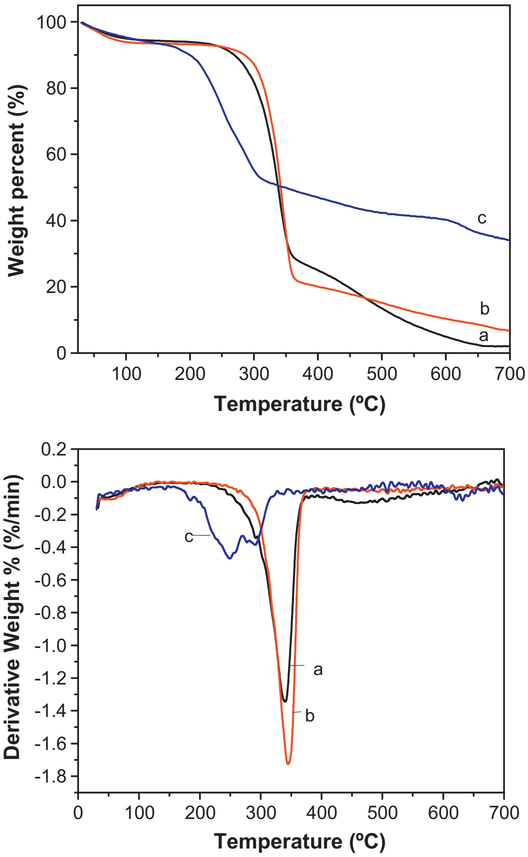

Fig. 8. TG and DTG curves of (a) untreated jute fibers; (b) alkali-treated jute; and (c) jute cellulose nanowhiskers.

It is quite interesting to note that the DTG curve of the TEMPO-oxidized jute cellulose nanowhiskers was broad, and seemed to consist of two peaks around 249 °C and 290 °C. The former was probably due to thermal degradation point of the sodium anhydroglucuronate units. The latter DTG peak was obviously lower than that of the untreated jute (338.8 °C), indicating that the crystalline cellulose chains decreased in DTG peaks by the effect of thermally more unstable anhydroglucuronate units present in the TEMPO-oxidized jute. The decrease in thermal degradation point might be one of the drawbacks in practical applications of TEMPO-oxidized cellulose fibers.

4. Conclusions

In summary, cellulose nanowhiskers with superthin diameter (3–10 nm) in the form of a stable and transparent dispersion were prepared from chemically pretreated jute fibers by TEMPO selectively oxidized pretreatment combined with mechanical homogenization. Additionally, the versatile cellulose nanowhiskers with a high crystallinity, carboxylate functional groups and high surface areas, would be particularly useful for applications such as green nanocomposites ultrafiltration, medicine, and catalyst supports. We believe that the method described here with a high yield and much lower energy consumption could be easily extended to provide a route to produce nanowhiskers with smaller widths from other natural cellulose materials with high cellulose content and crystallinity.

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References

Cao, X. D., Dong, H., & Li, C. M. (2007). New nanocomposite materials reinforced with flax cellulose nanocrystals in waterborne polyurethane. *Biomacromolecules*, *8*(3), 899–904.

Chakraborty, A., Sain, M., & Kortschot, M. (2005). Cellulose microfibrils: A novel method of preparation using high shear refining and cryocrushing. *Holzforschung*, *59*(1), 102–107.

Das, K., Ray, D., Bandyopadhyay, N. R., Sahoo, S., Mohanty, K. A., Misra, M., et al. (2011). Physicomechanical properties of the jute micro/nanofibril reinforced starch/polyvinyl alcohol biocomposite films. *Composites: Part B*, *42*(3), 376–381.

Das, K., Ray, D., Banerjee, C., Bandyopadhyay, N. R., Sahoo, S., Mohanty, A. K., et al. (2010). Physicomechanical and thermal properties of jute nanofiber reinforced biocopolyester composites. *Industrial and Engineering Chemistry Research*, *49*(6), 2775–2782.

Deng, H., Zhou, X., Wang, X., Zhang, C., Ding, B., Zhang, Q., et al. (2010). Layer-by-layer structured polysaccharides film-coated cellulose nanofibrous mats for cell culture. *Carbohydrate Polymers*, *80*(2), 475–480.

Dong, H., Strawheckera, K. E., Snydera, J. F., Orlicki, J. A., Reiner, R. S., Rudie, A. W., et al. (2012). Cellulose nanocrystals as a reinforcing material for electrospun poly(methyl methacrylate) fibers: Formation, properties and nanomechanical characterization. *Carbohydrate Polymers*, *87*(4), 2488–2495.

Dong, X. M., Kimura, T., Revol, J., & Gray, D. G. (1996). Effects of ionic strength on the isotropic chiral nematic phase transition of suspensions of cellulose crystallites. *Langmuir*, *12*(8), 2076–2082.

Elanthikkal, S., Gopalakrishnapanicker, U., Varghese, S., & Guthrie, J. (2010). Cellulose microfibres produced from banana plant wastes: Isolation and characterization. *Carbohydrate Polymers*, *80*(3), 852–859.

Fukuzumi, H., Saito, T., Iwata, T., Kumamoto, Y., & Isogai, A. (2009). Transparent and high gas barrier films of cellulose nanofibers prepared by TEMPO-mediated oxidation. *Biomacromolecules*, *10*(1), 162–165.

Fukuzumi, H., Saito, T., Okita, Y., & Isogai, A. (2010). Thermal stabilization of TEMPO-oxidized cellulose. *Polymer Degradation and Stability*, *95*(9), 1502–1508.

Ifuku, S., Tsuji, M., Morimoto, M., Saimoto, H., & Yano, H. (2009). Synthesis of silver nanoparticles templated by TEMPO-mediated oxidized bacterial cellulose nanofibers. *Biomacromolecules*, *10*(9), 2714–2717.

Isogai, A., & Kato, Y. (1998). Preparation of polyuronic acid from cellulose by TEMPO-mediated oxidation. *Cellulose*, *5*(3), 153–164.

Isogai, A., Saito, T., & Fukuzumi, H. (2011). TEMPO-oxidized cellulose nanofibers. *Nanoscale*, *3*, 71–85.

Jahan, M. S., Saeed, A., He, Z., & Ni, Y. (2009). Jute as raw material for the preparation of microcrystalline cellulose. *Cellulose*, *18*(2), 451–459.

Li, R., Fei, J., Cai, Y., Li, Y., Feng, J., & Yao, J. (2009). Cellulose whiskers extracted from mulberry: A novel biomass production. *Carbohydrate Polymers*, *76*(1), 94–99.

Li, X., Tabil, L. G., & Panigrahi, S. (2007). Chemical treatments of natural fiber for use in natural fiber-reinforced composites: A review. *Journal of Polymers and the Environment*, *15*(1), 25–33.

Liu, L., Yu, J. Y., Cheng, L., & Qu, W. (2009). Mechanical properties of poly(butylene succinate) (PBS) biocomposites reinforced with surface modified jute fibre. *Composites Part A: Applied Science and Manufacturing*, *40*(5), 669–674.

Liu, L., Yu, J. Y., Cheng, L., & Yang, X. (2009). Biodegradability of poly(butylene succinate) (PBS) composite reinforced with jute fibre. *Polymer Degradation and Stability*, *94*(1), 90–94.

Liu, Y. P., & Hu, H. (2008). X-ray diffraction study of bamboo fibers treated with NaOH. *Fibers and Polymers*, *9*(6), 735–739.

Lu, P., & Hsieh, Y. L. (2009). Cellulose nanocrystal-filled poly(acrylic acid) nanocomposite fibrous membranes. *Nanotechnology*, *20*(41), 415604.

Ma, H., Burger, C., Hsiao, B. S., & Chu, B. (2011). Ultrafine polysaccharide nanofibrous membranes for water purification. *Biomacromolecules*, *12*(4), 970–976.

Maiti, S., Ray, D., Mitra, D., & Misra, M. (2012). Study of compostable behavior of jute nanofiber reinforced biocopolyester composites in aerobic compost environment. *Journal of Applied Polymer Science*, *123*(5), 2952–2958.

Mao, X., Ding, B., Wang, M., & Yin, Y. (2010). Self-assembly of phthalocyanine and polyacrylic acid composite multilayers on cellulose nanofibers. *Carbohydrate Polymers*, *80*(3), 839–844.

Marchessault, R. H., Morehead, F. F., & Koch, M. J. (1961). Some hydrodynamic properties of neutral suspensions of cellulose crystallites as related to size and shape. *Journal of Colloid Science*, *16*(4), 327–344.

Marcus, E. C. J., Linus, L. S., & Jan, H. (1977). Mass-length ratio of fibrin fibers from gel permeation and light scattering. *Biopolymer*, *16*(1), 1–15.

Menezes De, A. J., Siqueira, G., Curvelo, A. A. S., & Dufresne, A. (2009). Extrusion and characterization of functionalized cellulose whiskers reinforced polyethylene nanocomposites. *Polymer*, *50*(19), 4552–4563.

Morán, J. I., Alvarez, V. A., Cyras, V. P., & Vázquez, A. (2008). Extraction of cellulose and preparation of nanocellulose from sisal fibers. *Cellulose*, *15*(1), 149–159.

Saito, T., & Isogai, A. (2004). TEMPO-mediated oxidation of native cellulose: The effect of oxidation conditions on chemical and crystal structures of the water-insoluble fractions. *Biomacromolecules*, *5*(5), 1983–1989.

Saito, T., Nishiyama, Y., Putaux, J. L., Vignon, M., & Isogai, A. (2006). Homogeneous suspensions of individualized microfibrils from TEMPO-catalyzed oxidation of native cellulose. *Biomacromolecules*, *7*(6), 1687–1691.

Saito, T., Shibata, I., Isogai, A., Suguri, N., & Sumikwa, N. (2005). Distribution of carboxylate groups introduced into cotton linters by the TEMPO-mediated oxidation. *Carbohydrate Polymers*, *61*(4), 414–419.

Sakurada, I., Nukushina, Y., & Ito, T. (1962). Experimental determination of the elastic modulus of crystalline regions in oriented polymers. *Journal of Polymer Science*, *57*(165), 651–660.

Sheltamia, R. M., Abdullaha, I., Ahmada, I., Dufresne, A., & Kargarzadeh, H. (2012). Extraction of cellulose nanocrystals from mengkuang leaves (*Pandanus tectorius*). *Carbohydrate Polymers*, *88*(2), 772–779.

Teixeira, E. M., Correa, A. C., Manzoli, A., Leite, F. L., Oliveira, C. R., & Mattoso, L. H. C. (2010). Cellulose nanofibres from white and naturally colored cotton fibres. *Cellulose*, *17*(3), 595–606.

Vieira, M. C., Heinze, T., Antonio-Cruz, R., & Mendoza-Martinez, A. M. (2002). Cellulose derivatives from cellulosic material isolated from *Agave lechuguilla* and *fourcroydes*. *Cellulose*, *9*(2), 203–212.

Wang, H., Huang, L., & Lu, Y. (2009). Preparation and characterization of micro- and nano-fibrils from jute. *Fibers and Polymers*, *10*(4), 442–445.

Xia, Z. P., Yu, J. Y., Cheng, L. D., Liu, L. F., & Wang, W. M. (2009). Study on the breaking strength of jute fibres using modified Weibull distribution. *Composites Part A: Applied Science and Manufacturing*, *40*(1), 54–59.