Fabrication and characteristics of [cellulose](http://cn.bing.com/dict/clienttranslate?mkt=zh-CN&setLang=ZH&form=BDVEHC&q=%E7%BA%B3%E7%B1%B3%E7%BA%A4%E7%BB%B4%E7%B4%A0%E7%9A%84%E5%88%B6%E5%A4%87%E5%92%8C%E8%A1%A8%E5%BE%81" \l "#" \t "_blank) nanofibril films from [Coconut](http://cn.bing.com/dict/clienttranslate?mkt=zh-CN&setLang=ZH&form=BDVEHC&q=%E6%A4%B0%E5%AD%90%E6%A0%91%E6%9E%9D%E5%8F%B6##) palm petiole prepared by different mechanical processings

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**ABSTRACT**

This paper focused on extraction of cellulose nanofibrils (CNFs) from coconut (*Cocos nucifera L.*) palm petioles falling off naturally, which if not properly processed constitute an environmental hazard. CNFs were isolated by chemical pretreatments and different mechanical processings, including grinding (G), grinding followed by ultrasonication (GU) and grinding followed by homogenization (GH). As one of the applications of this biodegradable and renewable CNFs, fabrication of films without using an organic solvent has been attempted using the received CNFs. Fourier transform infrared spectroscopy (FTIR) spectra analysis showed that the chemical treatment removed most of hemicellulose and lignin from the palm petioles, remaining only cellulose. SEM observations showed that diameter of the CNFs from GU method was between 50-100 nm, and the aspect ratio of CNFs was over 1,000. Tensile properties and transmittance of CNFs films with different mechanical treatments were also studied. Compared to grinding treatment, the CNFs film prepared by grinding/ultrasonication and grinding/homogenization treatments presented better tensile properties and transmittance. This work provides a new approach for more effective utilization of coconut palm petiole as potential feedstock for CNFs.

**Keywords:** Cellulose nanofibrils; Coconut palm petiole; Film; Grinding; Homogenization; Ultrasonication.

**1. Introduction**

With the decline of natural resources and the rising cost of raw fibre materials, the use of non-traditional sources such as forest and agricultural residues as raw materials has become more and more necessary for industry (Xing et al., 2006, 2007). The renewable, low-cost, lightweight and high specific strength and stiffness have identified these natural fibres as ideal reinforcement materials for polymer composites (Wu et al., 2010). Coconut palm (*Cocos nucifera L.*) is spread widely in tropical and sub-tropical regions as an important plant for local economy. The nutritious nut provides large amounts of food for people, but at the same time, million tons of residues cause serious solid waste pollution and air pollution because of they are either dispersed in fields and cities without rotting, or burnt. With the increasing attention on sustainable and environmental friendly materials, there are more and more studies about using natural fibers as reinforcement in polymer-matrix composites (Bledzki et al., 2010; Agunsoye et al., 2012). In order to explore new approaches for more effective utilization of coconut agricultural residues, Maheswari et al. (2012) extracted cellulose microfibrils with diameters in the range of 10–15μm from the agricultural residue of coconut palm leaf sheath using chlorination and alkaline extraction process method. Rosa et al. (2010) also prepared cellulose nanowhiskers from coconut husk fibers under different preparation conditions. However, there are very few reports in the literature about the extraction and applications of cellulose micro and/or nanofibrils from coconut palm petiole residues.

In recent years, a particular natural fibre derivative, CNFs has received major attention for its superior mechanical properties and has been used as promising candidates for reinforcement materials in nanocomposites (Abe and Yano, 2009; Bras et al., 2010; Chirayil et al., 2014). Cellulose is an abundant and naturally occurring polymer that mainly comes from wood and agricultural plants. Much effort has been made to develop adequate and commercially viable processes for disintegrating cellulose fibres into their structural components at nanoscale (Fan et al., 2009). Grinding, high-pressure homogenization, high-intensity ultrasonication, and enzymatic methods have been the principal applied procedures for the fibrillation of plant fibers. Iwamoto et al. (2005) prepared CNFs from pulp fiber by a high-pressure homogenizer treatment and a grinder treatment, and found that the grinder treatment resulted in the successful fibrillation of wood pulp fibers into nanofibers. Li et al. (2012) isolated CNFs with 10–20 nm in diameter from sugarcane bagasse by high-pressure homogenization in an ionic liquid (1-butyl-3-methylimidazolium chloride. Zhao et al. (2013) also extracted cellulose nanofibrils with diameters mainly ranged from 16 to 28 nm from dry softwood pulp through high shear homogenization. Chen et al. (2011) individualized cellulose nanofibers ranged from 5 to 20 nm from poplar wood using high-intensity ultrasonication combined with chemical pretreatments, and found that the diameter distributions of the resulting nanofibers were highly dependent on the output power of the ultrasonic treatment. From the literature, it is difficult to identify the most appropriate treatment method for producing CNFs among the grinding, homogenization and ultrasonication methods.

In cell walls, cellulose microfibril bundles exist encased by the embedding matrix such as hemicellulose and lignin, and the removal of such matrix substances is necessary for the fibrillation process. In this paper, chemical pretreatments were used at first to remove most of wax, extractives, hemicelluloses, and lignin of coconut palm petioles, and then different mechanical treatments, including grinding, grinding followed by ultrasonication and grinding followed by homogenization were introduced to disintegrate cellulose fibres into nano scale fibrils. There is also an urgent need for local government in south China to deal with the large amount of palm petioles wastes. Therefore, the objective of this paper was to convert the unfriendly falling off coconut palm petioles residues into biodegradabe and renewable value-added products, CNFs, and to explore the feasibility of fabricating CNFs films without organic solvents. The structure and properties of prepared CNFs films were characterized by tensile test, scanning transmission electron microscopy (STEM), Fourier transform infrared (FTIR) analysis and optical performance test.

**2. Experimental details**

*2.1. Materials*

Woody powder from coconut palm petioles (coming from Hainan province, China), cleaned with water and air dried, broken to the size of 2-3 mm × 6-7 mm with a L-905 shredder, grinded into powders with a FZ102 miniature plants grinder (TAISITE instrument Co., Tianjin), and sieved under 100 mesh (Zhang Xing Sand Screen Factory, Zhejiang province), was used for this study. The chemical composition of the coconut palm petioles was preliminarily investigated, and the content of cellulose, hemicelluose, lignin and ash were 33.29 wt%, 33.61 wt%, 19.87 wt%, 5.5 wt% (on a dry weight basis), respectively (Zhu et al., 2014). Chemical agents, including benzene, ethanol, sodium chlorite, hydrogen peroxide and potassium hydroxide, were purchased from Nanjing Chemical Reagent Co., Ltd.

*2.2. Preparation of CNFs*

Firstly, coconut palm petiole powder (10 g) was dewaxed with 2:1 (v/v) mixture of benzene/ethanol for 6 hours in a Soxhlet apparatus (SXT-06, Shanghai Hongji Instrument LO., Ltd.). The main purpose of this step is to remove off waxes and extractives according to open literatures (Abe and Yano 2009; Chen et al. 2011; Pettersen R. C., 1984). Secondly, the dewaxed powder, as shown in Fig.1-a, was bleached using 1 wt% acidified sodium chlorite water solution (400 mL) at 75 °C for an hour, and the process was repeated for four times, resulting in a white mixture, and then the mixture was washed with distilled water until the residue was neutral (shown in Fig.1-b). This step mainly removed off the lignin in the fibres according to Abe et al. (2009). Thirdly, in order to purify the cellulose by removing hemicellulose, residual starch and pectin, the bleached sample was further treated using 6 wt% potassium hydroxide water solution (400 mL) at 90 °C for 2 hours, and then filtered and rinsed with distilled water until the residues were neutralized, as shown in Fig. 1-c. Finally, the slurry of 1 wt% purified cellulose in water was processed by grinding (G), grinding followed by ultrasonication (GU) and grinding followed by homogenization (GH) treatment, respectively, resulting in cellulose suspension. Grinding process was done for 15 times with a MKCA6-3 grinder (Masuko Corp., Japan) at 1,500 rpm. Ultrasonication process of the ground cellulose was conducted in an ice/water bath for 40 minutes at 20-25 kHz frequency with an output power of 960 W by an ultrasonic generator (XO-1200, Xianqu Biological Technology Co., Ltd., China) with a cylindrical titanium alloy probe tip (2.5 cm in diameter). A 250 mL glass beaker with 5 cm in diameter and 10.8 cm in height was used as the recipient. At the same time the ground cellulose was subsequently processed twice with a homogenizer (EmulsiFlex-C3, AVESTIN, Inc, Canada) at pressure of 105 KPa.

**Fig. 1.**

*2.3. Fabrication of CNF films*

CNF films were fabricated according to the method by Iwamoto et al. (2005). The obtained cellulose nanofibres were dispersed in water at a fiber content of 0.1 wt% and stirring for 24 hours with a magnetic agitator. Fig. 1-d, 1-e and 1-f were the water suspensions of cellulose nanofibrils obtained by gringing (G), grinding/ultrasonic (GU) and grinding/homogenizing (GH), respectively. A 500 mL water suspension was vacuum filtered using a polytetrafluoroethylene membrane filter with a mesh of 0.2 μm and a diameter of 40 mm, resulting in a thin mat. The mat was then sandwiched between two water cellulose filter membranes, which were loaded between two smooth glass plates, and dried at 60 °C for 48 hours in an oven (DZF-6090, Jinghong Laboratory Equipment Co., Ltd., China). Three types of films with CNFs treated by G, GU, and GH methods, respectively, were designated as F1, F2 and F3, shown in Fig. 2-a, 2-b and 2-c.

**Fig. 2.**

*2.4. Fourier transform infrared spectroscopy*

In order to investigate the chemical changes induced by the successive chemical treatments, the FTIR spectra of the samples was conducted on a Nicolet iS10 spectrometer (Thermo Scientific) to collect detailed information of the functional groups of the freeze-dried samples after chemical treatments. Attenuated total reflectance (ATR) was recorded from 4000 to 400 cm−1 at a resolution of 4 cm-1. A total of 32 spectra were acquired at room temperature to yield 64 spectra for each sample.

*2.5. Tensile testing*

The tensile test was performed with a universal testing machine (SANS, Sans Materials Testing Co., Ltd., China) at a constant crosshead speed of 1 mm/min and a load cell of 100 N. The film sample size was 35×5 mm (length × width). All measurements were done in at least four replicates, and the average values of the Young’s modulus and the tensile strength were calculated from the four replicates.

*2.6. UV-visible spectrum*

Regular transmittance of the prepared films F1, F2 and F3 were measured at wavelengths from 190 to 1000 nm using a UV-visible spectrometer (U-4100, Hitachi High-Tech. Corp, Japan) with an integrating sphere 60 mm in diameter. Regular transmittance was measured by placing the specimens 25 cm from the entrance port of the integrating sphere. Three replicas were conducted for each type of the films.

*2.7. Scanning electron microscopy testing*

The suspension of GU was frozen at -40 °C for 24 hours with a DW-FL200A (Meiling Cryogenic Technology Co., Ltd., China), followed by freeze-drying for 48 h by Xianou-10 (Xianqu Biological Technology Co., Ltd., China), resulting in freeze-dried CNFs with GU treatment. After being coated with a thin layer of gold for 30-60 s, the fibrils and the surface of the corresponding film F2 were scanned by a scanning electron microscopy (S-4800, HITACHI, Japan) at an accelerating voltage of 3 kV.

**3. Results and Discussion**

*3.1. FTIR spectroscopic analysis*

Table 1 lists the typical functional groups and the corresponding bands for cellulose, hemicelluloses and lignin (Nelson and O´Connor, 1964; Oh et al., 2005; Yang et al., 2007; Morán et al., 2008).

**Table 1**

Fig. 3 shows the FTIR spectra of coconut palm petiole fibers in different stages. To reduce baseline correction effects due to the strong wavelength dependence of the background, the regions between 3680–2795 cm−1 and 1768–770 cm−1 were separately baseline corrected and area normalized. There are no significant differences between the spectra of the untreated and the dewaxed samples, which proved that no significant chemical changes of the cellulose, hemicellulose and lignin of the samples occurred during dewaxing treatment. The absorption peak at 1460 cm-1 in Fig. 3a is attributed to –CH2 deformation vibration, which is present in side chains between benzene rings. This group disappeared in the spectra of the bleached sample, indicating the degradation of a side chain of lignin (Venkataswamy et al., 1987). Meanwhile, in Fig.3c, the absorption peaks at 1636 cm-1 and 1506 cm-1, which are attributed to the stretching vibrations of –C=O and the aromatic skeletal vibration of C=C in lignin, respectively, were changed dramatically and almost disappeared, compared with those of the untreated and the dewaxed samples as shown in Fig. 3-c and 3-d. It suggested that the sodium chlorite did remove the lignin from the samples. Well-known groups such as –HClO2, –ClO2, –ClO3, and –Cl exist in the sodium chlorite water solution. The –ClO2 group as a free radical in particular, has bleaching effect by reacting with phenolic structural units of lignin (Sun et al., 2004).

**Fig. 3.**

On the other hand, the absorption peak at 1737 cm-1 in Fig. 3-c, which was ascribed to the stretching vibration of –C=O of hemicellulose, almost disappeared in Fig. 3-d and e. However, there was still a characteristic absorption peak at 1620 cm-1 with certain intensity, indicating that potassium hydroxide did remove most of the hemicellulose from the sample, but not completely. Some of the remained residual hemicellulose was insoluble in dilute alkali solution, and the rest was sucked back to the cellulose due to the decrease of alkali concentration in later the reaction. Iwamoto et al. (2008) indicated that the remained hemicellulose in the sample contributed to the formation of the nano fibrillated lignocellulose, because the inclusion of hemicellulose in the surface of purified cellulose fiber filaments could reduce the adhesion force between filaments, preventing fibril aggregation during nano-cellulose extraction with subsequent positive effect. In addition, the absorption peaks at 3328 cm-1 attributed to –OH stretching vibration, 2900 cm-1 attributed to –CH stretching vibration, 1430 cm-1 attributed to –CH2 and –OCH plane bending vibration, 1375 cm-1 attributed to –CH deformation vibration and 897 cm-1 attributed to anomeric carbon (C1) vibration, could be observed in the spectra of the bleached, alkali-treated, and the final CNFs film. All these are the characteristic peaks of cellulose, indicating highly pure cellulose obtained.

*3.2. Morphological investigation of the GU CNFs and its film*

Fig. 4 shows the SEM images of CNFs and films. The length of CNFs was greater than 100 μm, and the diameter was about 50-100 nm (as shown in Fig. 4a、c and e) with an aspect ratio of over 1,000. Research in the open literatures showed that the range of diameter distribution of cellulose microfibrils varied among the sources. Zuluaga et al. (2009) obtained loose networks of 40–60 nm-wide cellulose microfibrils from vascular bundles of banana rachis with relative mild alkaline treatments, and the length of the microfibrils were estimated to a few micrometers, resulting in a practically infinite aspect ratio; however the strong acid treatment that preferentially degrade the disordered regions along the microfibrils cut the cellulose microfibrils, resulting in shorter whisker-like nanocrystals. Alemdar et al. (2008) extracted cellulose nanofibers from wheat straw and soy hulls by a chemi-mechanical technique, and found that the wheat straw nanofibers were determined to have diameters in the range of 10–80 nm and lengths of a few microns; but the soy hull nanofibers had diameter 20–120 nm and shorter lengths than the wheat straw nanofibers. The cellulose nanofibrils derived from coconut palm petioles could be considered as an ideal reinforcing material for composites because of its high aspect ratio as compared to CNFs obtained from other agricultural residues.

Compared to G, GU and GH showed smaller diameter of nanofibrils, indicating that both ultrasonication and homogenization treatments had significant refining effects on the diameter of nanofibrils. It is also seen that the cellulose fibers were nano-fibrillated uniformly and the assembled nanofibers were identified in some parts indicating that the nanofibrils were strongly and tightly combined together to provide excellent mechanical bonding properties for the film. More crude fibers (fiber bundles) can be observed in F1 film as compared to film F2 and F3. Film F3 obtained by homogenization treatment showed most uniformly and fine structure.

**Fig. 4.**

*3.3. Tensile property*

Fig. 5shows the tensile strength and modulus of CNFs films with different mechanical treatments. The density and thickness of the films are listed in Table 2. Among the three types of films, F1 showed the lowest density, tensile strength and modulus, 1.08 g/cm3, 106.83 MPa and 2.19 GPa, respectively; however, the highest density, tensile strength and modulus, 1.37 g/cm3, 160.42 MPa and 3.64 GPa, respectively, were obtained from F3. Compared to F1, F2 and F3 had 46.6% and 50.2% higher tensile strength, and 35.2% and 66.2% higher modulus, respectively, indicating that both ultrasonication and homogenization treatments had significant effects on the quality of the CNFs and the corresponding films. This result is due to the different defibrillating mechanisms of grinding, ultrasonication and homogenization. During grinding, the cell wall structure consisting of nanofibers in a multi-layered structure could be broken down into micro and/or nano-sized fibers by shearing forces generated by the grinding stones (Iwamoto et al., 2007). For the film F2, the ground fiber with grinding treatment was further refined by ultrasonification, in which the energy provided by cavitation could efficiently tear off nanofibrils from the fibril bundles (Iwamoto et al., 2005). And for F3, the ground fiber with grinding treatment was further defibrillated by a homogenizor through a large pressure drop and high shear and impact forces (Iwamoto et al., 2005). The more individual/thinner nanofibrils obtained, the denser and finer structure of films will be formed. The denser and finer structures of F3 directly contribute to the highest density and strength of the film. Wang et al. (2013) reported that the nanopaper, which derived from cellulose nanofibers of waste corrugated paper through a series of chemical treatments combined with grinding, ultrasonication and centrifugation, presented high tensile strength of 135 MPa and a tensile modulus of 6.67 GPa. Compared to the results of Wang and coworkers, the tensile strength values of films F2 and F3 were higher, but the modulus values were lower in this study. This indicated that more flexible film with higher strength could be produced from coconut palm petiole.

**Fig. 5.**

**Table 2**

*3.4. Transmittance*

Another remarkable feature of CNFs films is its high transparency (Wang et al., 2013). At 600 nm wavelength, the regular light transmittance at room temperature of sample F1 was 73.3%. Compared to F1, films from homogenization (F3) and ultrasonication (F2) increased transparency up to 76.1% and 78.1%, respectively. According to the study of Abe et al. (2007), the high transmittance of F2 may due to the CNFs are more uniform and far thinner than F1 and F3. The results of light transmittance of CNFs films obtained from this study were in agreement with those reported by others (Nogi et al., 2009; Wang et al., 2013). Consequently, CNFs film with adequate light transmittance could be achieved from coconut palm petiole.

**4. Conclusions**

In this study, cellulose nanofibrils were successfully isolated from coconut palm petiole by chemical modification followed by mechanical treatments, including grinding, grinding followed by ultrasonication and grinding followed by homogenization. FTIR spectra analysis showed that the chemical treatment removed most of hemicellulose and lignin from the palm petiole fibers, but cellulose molecular structure remained unchanged after chemical and mechanical treatments. SEM observations showed that diameter of the cellulose nanofibrils from grinding/ultrasonication was between 50-100 nm, and the aspect ratio of cellulose nanofibrils was over 1,000.

As one of the applications of this biodegradabe and renewable cellulose nanofibrils, fabrication of films without using an organic solvent has been attempted using the received cellulose nanofibrils. Compare to grinding treatment alone, the CNFs films prepared by grinding/ultrasonication and grinding/ homogenization treatments showed better tensile properties and transmittance. This work provides a new approach for more effective utilization of coconut palm petiole as potential feedstock for the production of cellulose nanofibrils.

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Table captions

**Table 1**

Functional group absorption bands of cellulose, hemicellulose and lignin

**Table 2**

Density and thickness of CNFs films

**Figure captions**

**Fig. 1.** Photographs of the samples in different stages: (a) dewaxing, (b) bleaching, (c) alkali-treatment, (d) G, (e) GU, and (f) GH

**Fig. 2.** Photographs of nanocellulose films: (a) F­1, (b) F2, and (c) F3

**Fig. 3.** FTIR spectra of : (a) the untreated, (b) the dewaxed, (c) the bleached coconut palm petiole fibers, and (d) the alkali-treated sample.

Fig. 4 SEM micrographs of freeze-dried G CNFs（a）and its film F2 (b), GU CNFs（c）and its film F2 (d), GH CNFs（e）and its film F2 (f).

**Fig.5.** Tensile properties of CNFs films