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Nanocellulose production from natural and recyclable sources: A review

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Abstract. Cellulose is the most abundant biopolymer on earth and is the chain of glucose residues that can be obtained easily from nature. Having unique material properties, nanocellulose has gained interest of researchers for various applications. Cellulose is generally known to exist in cell wall of a plant. However, this paper reviews the isolation of nanocellulose not only from plants, wood, and agroforestry residues, but also from recyclable sources paper waste and animals. With appropriate treatment and process (chemical, mechanical, and biological), reduction in diameter and length of cellulose up to nanoscale is possible. Nanocellulose may appear in its three main types namely cellulose nanofibers (CNFs), cellulose nanocrystals (CNCs) and bacterial nanocelluloses. Transformation of waste to wealth by adding value to waste and natural sources has become a meaningful and interesting work.

1. Introduction

The efficient exploitation of cellulose as the most abundant biopolymer and is the primary reinforcement of cell wall in plants [1] was encouraged by social concerns for sustainable green products. Cellulose hierarchical structure and material properties comparison of cellulose as extracted from work of [2] is shown in Figure 1 and Table 1 respectively. Cellulose is a linear natural polymer that comprises of D-anhydroglucose units joined by (1→4)-β-glycosidic linkages. Not only limited to plants, fungi, and algae; animals can also produce cellulose [3].



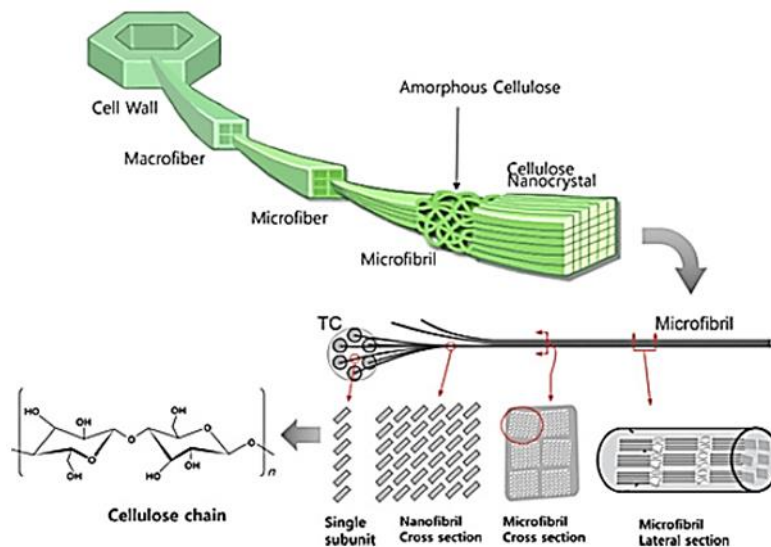


Figure 1. Hierarchical structure of cellulose [2].

Table 1. Material properties comparison of cellulose [2].

Material	Tensile strength (GPa)	Young's modulus (GPa)	Density (g/m^3)	Tensile /density	Modulus /density	Thermal expansion coefficient (ppm/K)
CNC	7.5	145	1.6	4.7	90.6	3-22
Glass fiber	4.8	86	2.5	1.9	34.4	13
Steel wire	4.1	207	7.8	0.5	26.5	15
Kevlar	3.8	130	1.4	2.7	92.9	-4
Graphite	21	410	2.2	9.5	186	2-6
CNT	11-73	270-970	1.0	11-73	270-970	-

Recent advances in nanotechnology has enabled fabrication of materials at the nanoscale level and motivation for the miniaturization process is because nanosized materials possess superior mechanical properties compared with bulk material. Nanocellulose can be isolated from its various sources through mechanical, chemical and enzymatic/biological treatments. Main types of nanocelluloses are generally refers to CNFs, CNCs, and bacterial cellulose.

Tissue engineering, reinforcement, and filter media are among the wide range of CNFs' applications. CNCs are products with high added value that can be used in many applications and portray many advantages such as high strength, lightweight, unique optical properties, stiffness, and high surface area when compared to CNFs [4]. Nanocellulose extracted recyclable material may be cheaper than commercial nanocellulose and hence very important to be studied.

2. Nanocellulose from Wood

Generally, there are two types of wood namely softwood and hardwood. Nanocellulose was successfully produced from both sources.

Fibrillation of cellulose fibers from Eucalyptus and Pine pulp into nanosized fibrils was done using homogenization process where a dilute fiber concentration lower than 1-2 wt% was pass through mechanical homogenizer and fibers were oxidized previously under neutral conditions to facilitate the fibrillation process and to cut down the number of passes [5,6]. The morphology of CNFs isolated from the Eucalyptus and Pine were observed using field emission scanning electron microscope (FESEM) shows size ranging from 5- 20 nm in diameter. Isolation of CNFs from wood using one-time grinding treatment was enabled in an undried state after discarding matrix substances with diameter ranging from 12-20 nm [7]. Apart from homogenization process, CNFs are able to be isolated from Eucalyptus kraft

pulp using other process which includes one of the most studied CNFs extraction method acid hydrolysis, refining and sonication [8]. CNFs with size if 10-60 nm were attained from sulfonated celluloses using a high-pressure homogenizer and bleached birch chemical wood pulp as raw material [9]. CNFs were individualizes from poplar wood through high-intensity ultrasonication together with chemical pretreatment gives CNFs with diameter ranging from 5-20 nm [10]. The results obtained from fourier-transform infrared spectroscopy (FTIR) and x-ray powder diffraction (XRD) in this study indicates that hemicellulose and lignin were extensively removed in the CNFs and gives crystallinity of approximately 69%.

Biological treatment of softwood kraft pulp using genetically modified fungus isolated from fungus infected Dutch elm tree was done by [11] and they found that pre-refining natural fibers before biological treatment process improved the CNFs percentage yield in less than 50 nm diameter range.

3. Nanocellulose from Nonwoody Plants and Crops

CNCs were effectively isolated from tomato peels and was demonstrated by either chlorine-free sodium hydroxide/hydrogen peroxide (NaOH/H₂O₂) or acidified sodium chlorite/potassium hydroxide (NaClO₂/KOH) processes. CNCs with significantly improved specific surface area, pore volume, mesoporosity and more uniformly nanofibers with 42 nm width average could be assembled from CNCs in 1:1 v/v *tert*-butanol/water mixture [12]. Steam explosion techniques was used to treat *Hibiscus sabdariffa* fibers to produce CNFs in higher cellulose percentage and lower lignin percentage [13]. Figure 2 below shows the scanning electron microscope (SEM) images of untreated and treated *Hibiscus sabdariffa* fibers.

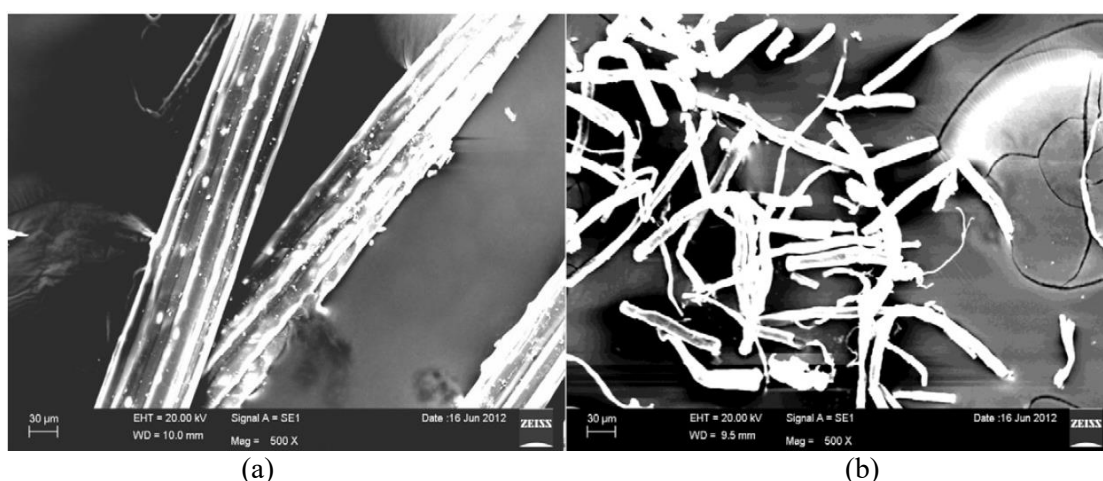


Figure 2. SEM images (a) untreated fibers (b) treated fibers. Work of [13].

The outcome of steam explosion in alkaline medium are the hydrolysis of hemicelluloses within the fiber, and the produced sugars may later be washed out in water, leaving a residue of cellulose and lignin then leads to the cleavage of hemicelluloses–lignin bonds also. The reaction leads in an increased hemicellulose water solubilisation and in an increased lignin solubility in alkaline solvent, leaving the solid residue cellulose with a reduced degree of polymerization. CNCs extraction from garlic straw using acid hydrolysis gives an average diameter of 6 nm and average length of 480 nm of CNCs produced and was considered as promising results that added value to garlic straw [14]. Microwave liquefaction of bamboo, combined with chemical treatment and ultrasonic nanofibrillation process was proven to successfully extracted CNFs with diameter ranging from 2-30 nm and this method was found to remove non-cellulosic compounds from the bamboo efficiently [15]. Due to lignification degree and relatively loose cell wall structure in the parenchymal cells in bamboo, they became an attractive source of materials for CNFs [16]. Acid hydrolysis of naturally colored and white cotton fibers results in cotton nanofibers with diameter of 6-18 nm and length of 82-225 nm and white cotton nanofiber gives higher

yield [17]. High pressure defibrillation and chemical purification of hemp fiber individualized CNFs that have width of 30-100 nm and length of several micrometers [18]

4. Nanocellulose from Agroforestry Residues

Non-cellulosic components such as lignin, hemicellulose and pectic substances were eliminated through a combination of chemical and mechanical treatments to prepare CNFs [19,20]. CNFs with width ranging from 8-10 nm was prepared by alkaline treatment followed by TEMPO-mediated oxidation and subsequent homogenization of an agricultural by product, corn husk [21]. 5-60 nm width of CNFs were successfully isolated from pineapple leaf fibers by employing steam explosion process [22]. A unique interconnected web-like structure of nanofibers was contributed by the high-pressure defibrillation. A combination of chemical and mechanical treatment was used by [23] to treat wheat straw and soy hulls in order to extract CNFs. Wheat straw nanofibers have diameters ranging from 10 to 80 nm with length of a few thousand nanometers, while soy hulls nanofibers have diameter ranging from 20 to 120 nm with length shorter than that of wheat straw nanofibers. Using thermal gravimetric analysis of thermogravimetric analysis (TGA), the thermal properties of the nanofibers was found to rise dramatically after the mechanical treatment of cyrocrushing, disintegration and defibrillation. Nanocellulose resulting in 10-20 nm in diameter was isolated from sugarcane bagasse using high pressure homogenization in a homogeneous media with pre-treatment with ionic liquid [24]. By using high pressure homogenizer and high speed blender, triticale crop residue was used as source for CNFs production with lateral size of 20-30 nm [25]. The researcher of this project concluded that conversion of triticale pulps into CNFs with low energy demand using a high-speed blender is possible.

5. Nanocellulose from Paper Waste

Tremendous amount of paper waste was discarded annually, hence creating environmental pollution. New approaches to transform this paper from waste to wealth has become a meaningful and challenging work.

Preparation of CNCs from old newspapers and recycled newsprints used direct acid hydrolysis with alkali and bleaching pre-treatment [4] and CNCs produced present similar properties. Combination of acid hydrolysis and mechanical ultrasonic treatments enables extraction of rod-like CNFs from waste newspaper [26]. Potential of municipal paper waste for production of CNFs was studied by [27] using three different techniques; pulping, flotation, and washing, and was then subjected to ultrafine grinding. Ultralong cellulose nanofibers with extremely high aspect ratio were manufactured successfully from waste corrugated paper pulp through a combination of a series of chemical treatment, grinding, ultrasonication and centrifugation results in CNFs with diameter ranging from 30-100 nm [28].

6. Nanocellulose from Animals

Study on cellulose sourced from animals was not as advanced as on plants, fungi, and bacteria. However, researchers have proved that cellulose, can also be extracted from animals such as tunicate, prawns and crabs.

CNFs with cross-sectional dimensions of 8×20 nm measured by atomic force microscopy (AFM) was obtained from tunicate (*Halocynthia papillosa*) [3], an invertebrate marine animal shown in figure 3 below. Mantles from the tunicate were separated and were cut into small pieces. To purify the cellulose, sodium chlorite and sodium hydroxide were used according to delignification method. The disintegration of purified cellulose into microfibrils used two processes; TEMPO-mediated oxidation and sulfuric acid hydrolysis, and then subsequent mechanical treatment in water.



Figure 3. Tunicate [29].

Chitin nanofibers were prepared from exoskeleton of prawns and crabs and squid pens by [30] through a series of purification steps and followed by simple mechanical treatment. Results from this research gives fine nanofiber networks with approximately 10 nm uniform width.

7. Nanocellulose Extraction Method

To extract nanocellulose from cellulose, several methods have been developed and used by previous researchers. Primarily, hydrolysis method that involves breakdown of amorphous region of cellulose fibers [31]. Owing to its short reaction time, acid hydrolysis is commonly performed to extract nanocellulose with sulphuric acid as the preferred acid used. Researchers commonly agree that acid hydrolysis method involves acid concentration that is relatively high (50-70 wt%), approximate temperature around 40-50°C and retention time between 30-90 minutes [32]. Typical CNC production using acid hydrolysis procedure is as presented in Figure 4 below.

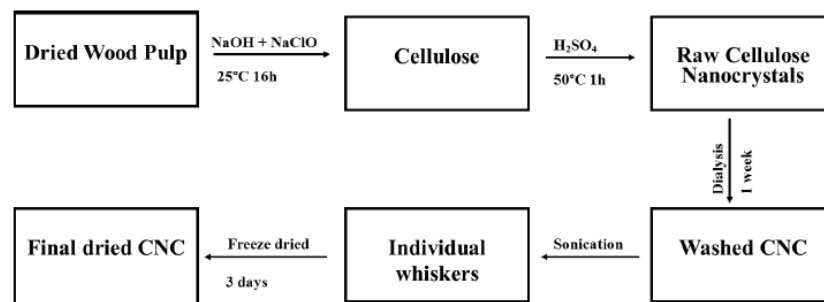


Figure 4. Typical procedure for CNC production using acid hydrolysis.

Enzymatic hydrolysis is an alternative hydrolysis method from acid hydrolysis that uses enzymes to breakdown cellulose to sugars and these enzymes are commonly found in fungi and bacteria [33]. [34] study the CNC production from wheat straw using cellulase and combined with ultrasonication treatment and this work reported that this method requires a significant amount of time and though the obtained yield is rather similar to acid hydrolysis treatment yield, time consumption makes it at a disadvantage site. Another form of nanocellulose, bacterial nanocellulose was produced by *Acetobacter* species possess unique properties such as high water absorbance and biocompatibility makes it great for wound treatment [35,36].

While acid hydrolysis has the disadvantages of harsh conditions and enzymatic solution seems to require a significant amount of time, subcritical water (SCW) treatment stand out as no harsh conditions is needed in this treatment and it is relatively quick process unlike enzymatic hydrolysis. CNCs production from cellulose powder was produced using SCW at temperature 120-200°C with pressure being maintained then followed by filtration, dialysis and sonication, leaving CNCs suspension [37].

Other method that has been actively explored by researchers to produce nanocellulose is mechanical processing. Typical method used are pressure homogenization or ultrasonic [5,6]. Combination of sonication and hydrolysis also have gained more interest to improved aspect ratio of CNC [38].

8. Conclusion

Nanocellulose are finding newfound interest in many applications due to its unique characteristics and properties. Despite common knowledge that cellulose present in cellulose of plant cell wall, researchers and scientists have proved that nanocellulose can also be extracted from animals' exoskeleton and animals such as tunicate. With suitable method used to treat these cellulose sources, nanocellulose can be isolated and exist in CNCs, CNFs and bacterial nanocellulose form.

With saturated study on nanocellulose extraction from natural sources, there are emerging interest in adding value to waste to transform waste into valuable material or product. Processing nanocellulose from waste such as paper waste can add value to waste instead of just disposing these wastes and can be used for many applications such as super water absorbent material, hydrogel to absorb oil in the ocean and many more. Despite the nanocellulose produced may portray inferior properties compared to commercial nanocellulose, transformation of waste has become eloquent work.

Many methods have been studied to produce nanocellulose with hydrolysis becomes preferred method, more enhanced study on subcritical water hydrolysis that is more advantageous than acid hydrolysis and enzymatic hydrolysis in terms of cleaner, safer, and shorter time treatment should be done in treating waste into added value material or product.

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