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MINI REVIEW ON NANOFIBRILLATION TECHNIQUES TO OBTAIN CELLULOSE NANOFIBER FROM LIGNOCELLULOSIC BIOMASS

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ABSTRACT

Cellulose nanofiber (CNF) has been recognized as a new family of sustainable nanomaterials with significant mechanical, colloidal, low density, renewable and biodegradable properties. These properties make CNF a promising material either as an additive or primary material for the development of various applications such as for packaging, coatings, adsorbent, papermaking, biomedicine, cosmetic and automotive industries. Prior to CNF production, cellulose has to be isolated from various sources of lignocellulosic biomass such as oil palm, bamboo, flax bast, hemp, kraft pulp, and rutabagas. The isolated cellulose then can be nanofibrillated into CNF using several nanofibrillation treatments. However, the nanofibrillation treatment is one of the major factors that can influence the final characteristics of CNF. The selection of nanofibrillation treatment is important as each treatment produces CNF with different properties especially in terms of diameter sizes, lengths, thermal, crystalline and mechanical properties. The CNF properties have to comply with the requirement of the targeted application, as different applications require different specifications of CNF. Other external factors may also influence the properties of CNF. Hence, this review focused on discussing the current CNF production treatment as well as factors that may influence the nanofibrillation process of CNF production.

Introduction

In recent years, there are concerns raised by the public on environmental sustainability especially in utilising biomass for the development of bio-products. Numerous bio-products have been manufactured from biomass, especially those derived from agricultural waste such as oil palm empty fruit bunches, bamboo, kenaf, and hemp. Biomass has been used as either a primary material or additive for products ranging from household to spectacular materials such as biocomposites, biofuels, biosugars and many more[1-7]. Among them, particular attention has been given to utilise biomass for the production of value-added biomaterial known as cellulose nanofiber (CNF) [8-14]. This might be because of its commercial value, as despite derived from invaluable biomass, CNF can be sold to the market at a very high price, approximately 10 – 100x higher than the price of raw biomass.

CNF is a cellulosic fiber with a dimension of 100 nanometer (nm) or less with several micrometer lengths. It is also known as a versatile material due to its excellent properties such as having extremely high specific area, high stiffness, high porosity with excellent pore interconnectivity, low weight, high biodegradability, and renewability [15-19]. In the last 5 to 8 years, interest concerning the applications of CNF in the fields of material sciences, biomedical engineering, cosmetics, military materials, pharmaceuticals, foods and packaging has been increased worldwide [11, 20-30]. Fig. 1 shows recent applications of CNF.

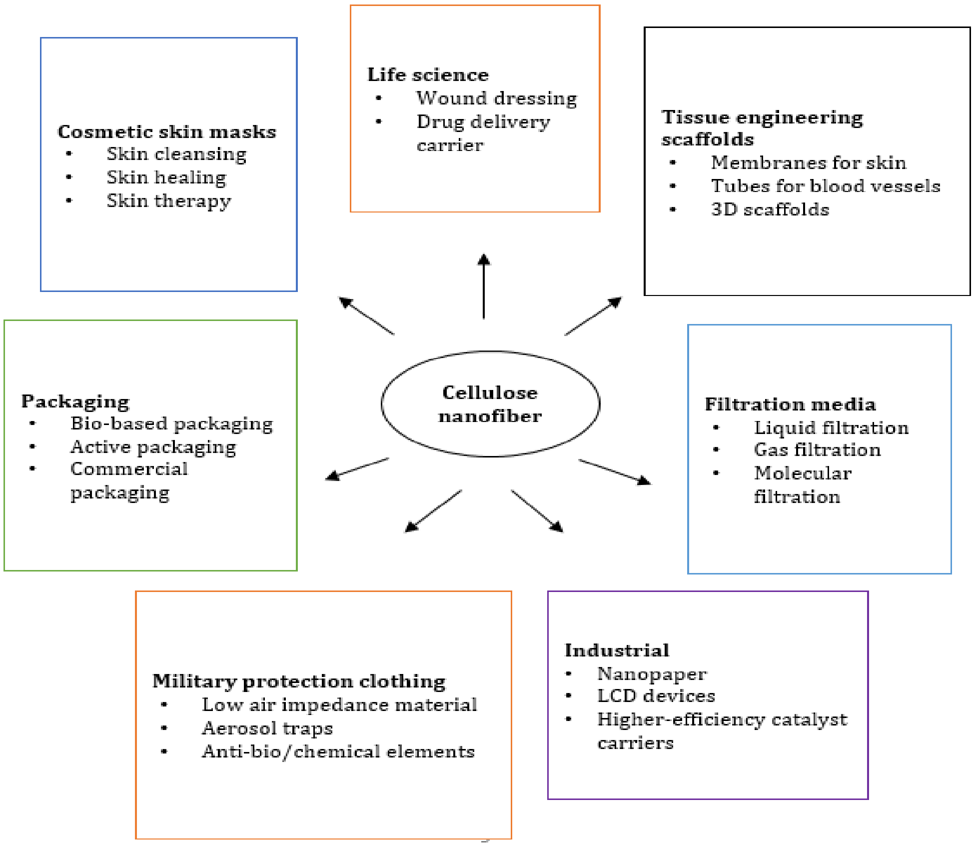


Fig. 1: Applications of nanocellulose

CNF is generally produced from extracted cellulose of wood pulp or any lignocellulosic material. Nevertheless, a limited source of wood pulp has influenced researchers and industrial players to use agricultural biomass as a raw material for CNF production, as the supply is abundant and inexpensive. In fact, the physical and chemical properties of agricultural biomass are at the same par as wood pulp. In order to produce high-quality CNF, a proper nanofibrillation treatment needs to be selected, as it needs to be able to delaminate the fiber cell walls and disintegrate the fibrils into nano-sized without affecting their properties [31].

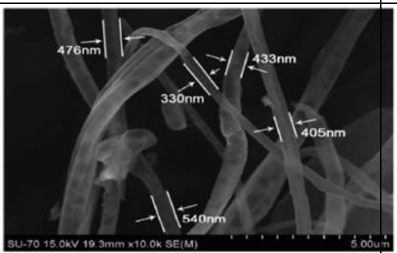
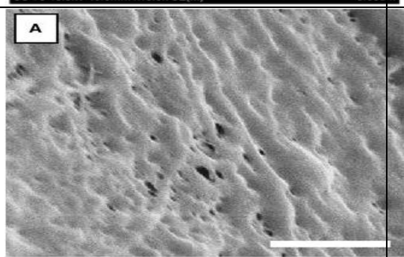
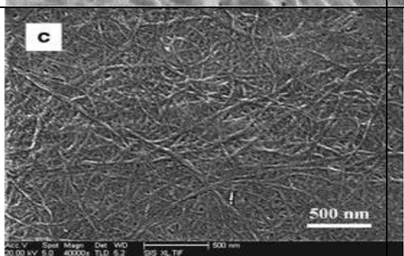
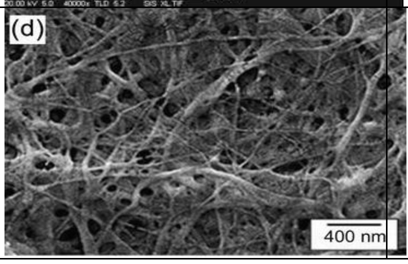
CNF can be produced from any lignocellulosic materials using several nanofibrillation treatments which include enzymatical, electrospinning, ultra-sonication, high-pressure homogenizer, ball milling, and extrusion. The selection of nanofibrillation treatment and types of lignocellulosic sources play a major role in determining the CNF's properties. For example, by grinding cotton linters, the obtained CNF has a width of approximately 10-30 nm, with a crystallinity index of around 82.3-85.7 % and thermally degraded at around 320-350 ^{O}C. Meanwhile, CNF from rice straw which was nanofibrillated by TEMPO-mediated oxidation treatment would have diameters of approximately 2-20 nm with a crystallinity index being around 60-65 % and thermally degraded at 210-310 ^{O}C [32].

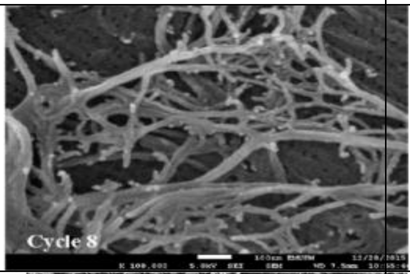
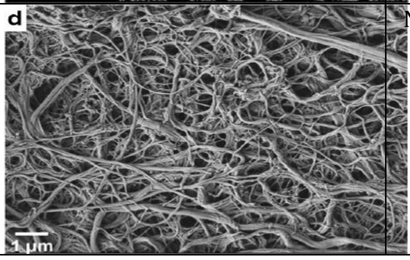
In regards to the variation of CNF properties obtained from different nanofibrillation treatments and lignocellulosic sources, this review focused on discussing those aspects as well as other factors that may affect the nanofibrillation of CNF.

Processing techniques of cellulose nanofiber

Generally, CNF can be nanofibrillated using either enzymatic or a variety of mechanical processes or by a combination thereof [31, 33]. Electrospinning, high-pressure homogenization, sonication, extrusion, wet disk milling, mechanical shearing, and cryocrushing are among the nanofibrillation treatments that have been widely used to produce CNF [15, 34-37]. Table 1 summarizes several properties of CNF obtained from various nanofibrillation treatments, and it can be observed that different treatments certainly produced different properties of CNF especially in terms of morphological properties.

Table 1: Properties of CNF produced from various nanofibrillation treatments

Nanofibrillation technique	Source of cellulose	Example of image	Crystallinity (%)	Size obtained (nm)	Reference
Electrospinning	Oil palm biomass Commercial cellulose		~45	120 - 2000	[15, 38-40]
High pressure homogenizer	Wood		Not reported	>100	[31, 41]
Ultrasonication	Wood		~80	5 ~ 20	[42]
Ball milling	Soft wood		>70	100 nm	[43-44]

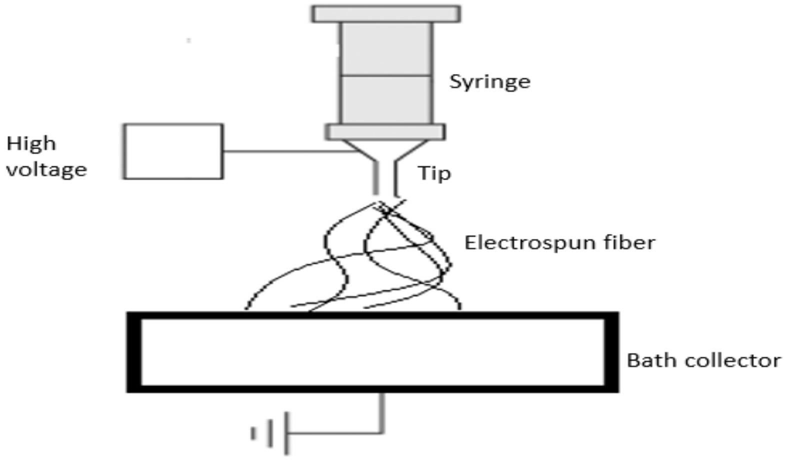
Nanofibrillation technique	Source of cellulose	Example of image	Crystallinity (%)	Size obtained (nm)	Reference
Wet disk milling	Oil palm biomass Bamboo Bagasse		~45	20 - 40	[37, 45-47]
Extrusion	Kraft pulp Oil palm biomass		Not reported	50 ~200	[9-10, 48-49]

Enzymatic nanofibrillation

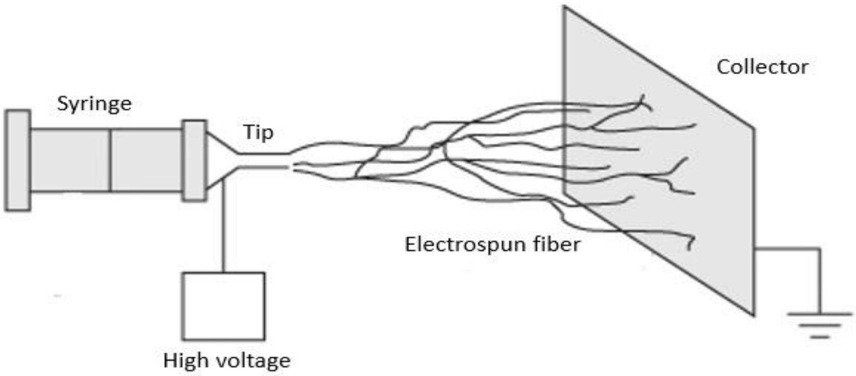
Enzymatic treatment is one of the conventional methods to produce CNF [50]. This method is usually combined with another refining technique called biorefining. In order to produce CNF, two stages of enzymatic treatments need to be conducted. At the first stage, lignocellulosic materials have to be pre-treated using enzymes that can extract only cellulose and degrade other accompanying polymers, such as pectin, hemicellulose, lignin and other impurities. For the second stage which is also known as the fibrillation stage, industrial cellulases that are responsible for hydrolysing cellulose into smaller structural elements can be used. This method however is not effective in producing CNF with a diameter size less than 100 nm, hence other mechanical treatments are often applied. For example, Paakko et al. (2007) have combined both enzymatic hydrolysis and high-pressure homogenization treatments to produce CNF in their study [34].

Electrospinning

Most research suggested that electrospinning is the most promising technique to produce high amounts of CNF [15], [38]. However, this technique has turned out to be difficult to perform due to the limited solubility of cellulose in volatile solvents suitable for electrospinning and the challenging chain dynamics of the cellulose molecule in solution [40]. In fact, all known solvents for cellulose appear to be nonvolatile and form strong interactions with the cellulose molecule, and this has been rendering the usual solidification mechanism in electrospinning through simple atmospheric drying. In order to proceed with this treatment, a precipitation bath as shown in Fig. 2 (a) is needed to produce CNF from lignocellulosic materials. This method nevertheless is inapplicable for other materials such as polyacrylonitrile, polyvinyl alcohol and, chitosan as shown in Fig. 2 (b). This is because the solvents used to dissolve these materials are easily evaporated after ejection from the electrospinning tip. By referring to Table 1, electrospun CNF has an average diameter ranging from 120 nm – 2000 nm which is larger than the CNF produced using other techniques [15, 38, 40, 51]. As such, extensive research to improve the electrospinning process, involving the effect of solvents and their processing parameters are still being comprehensively studied.



(a)



(b)

Fig. 2: Schematic diagram of electrospinning apparatus, (a) electrospinning with a precipitation bath, (b) electrospinning without a precipitation bath (redrawn from [52])

High-pressure homogenizing

High-pressure homogenizing is commonly used to produce CNF from various sources such as wood pulp, sugar beet, and prickly pear [41]. Fig. 2 shows the mechanism of high-pressure homogenizing. The size of the CNF produced using this technique is below than 100 nm. The process includes passing a cellulose slurry at high pressure into a vessel that is equipped with a very small nozzle. High velocity and pressure as well as impact and shear forces lead to the generation of the CNF [31]. This process requires several passes of homogenizing in order to produce the desired properties CNF. The extent of nanofibrillation improves as the number of repeated passes increases [53].

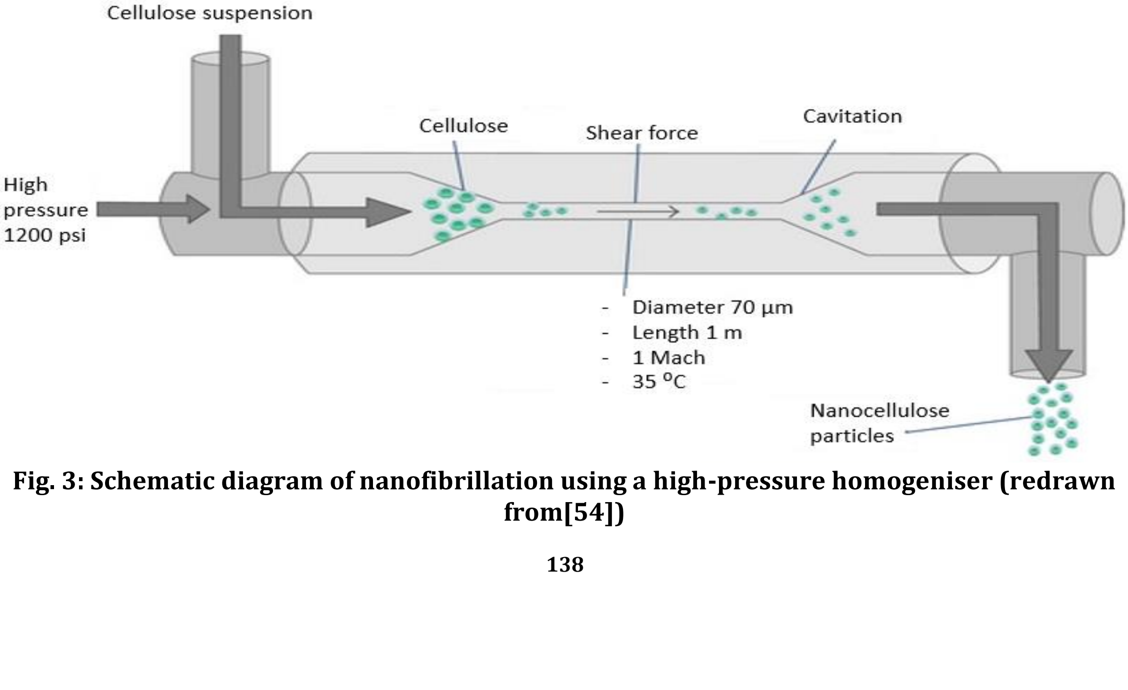


Fig. 3: Schematic diagram of nanofibrillation using a high-pressure homogeniser (redrawn from[54])

Ultra-sonication

Ultra-sonication is another technique that is frequently used to produce CNF. This method is often being used to produce CNF from wood sources [55]. Studies showed that the diameter distribution of the resulting CNF was highly dependent on the output power of the ultrasonic treatment. Fig. 4 shows a schematic diagram of the ultrasonication process. Ultrasound energy is transferred to the cellulose chains through a process called cavitation, which refers to the formation, growth, and violent collapse of cavities in water. The energy provided by cavitation is approximately 10–100 kJ/mol, which is within the hydrogen bond energy scale. Thus, this ultrasonic impact would gradually disintegrate the micron-sized cellulose into CNF [55]. The morphological image of CNF produced by ultrasonication as shown in Table 1 shows that the diameter of CNF was in a range of 5 to 20 nm. However, the diameter size of CNF obtained using this technique is diverse. The CNF can be aggregated with a wide distribution in width due to the complicated multi-layered structure of lignocellulosic materials.

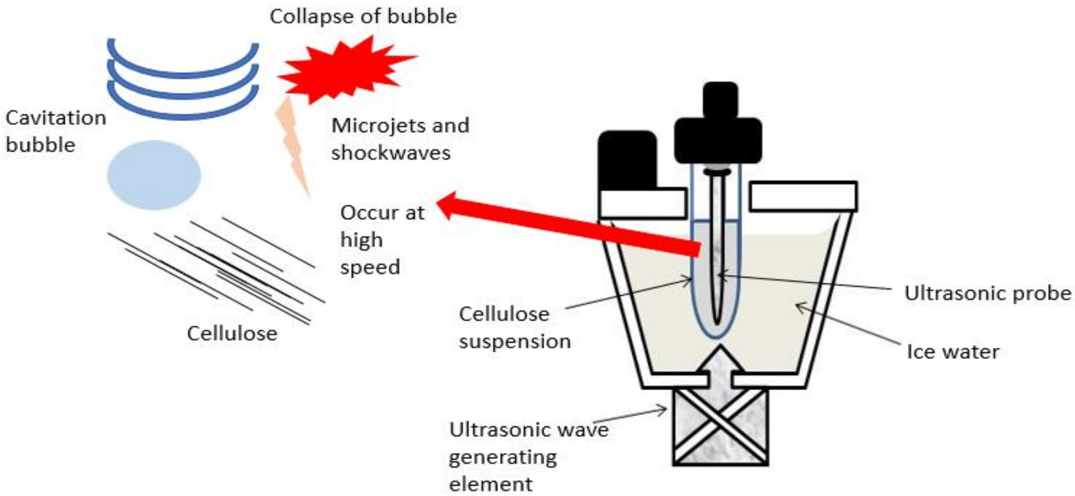


Fig. 4: Schematic diagram of nanofibrillation by ultra-sonication (redrawn from [56])

Cryocrushing

Cryocrushing is a process in which the cellulose fibers are frozen in liquid nitrogen prior to the fibrillation process. The frozen cellulose fibers are then crushed using a mortar and pestle. The application of high impact force to the frozen cellulose fibers then ruptures their cell walls due to the exerted pressure from ice crystals within the cell walls that were formed after immersion in liquid nitrogen, thus resulting in the formation of CNF [57]. Alemdar and Sain, (2008)[58] reported that the diameter size of CNF produced by cryocrushing is within a range of 10 – 80 nm and with lengths that are less than 100 nm, which can be considered as a short form of CNF. The crystallinity and thermal degradation temperatures were also found to be increased by 35 and 38 %, respectively.

Ball milling

Zhang et al. (2015) reported the possibility of producing CNF from softwood pulp using a simple ball milling technique under ambient pressure and at room temperature [44]. The effects of milling conditions including the ball-to-cellulose mass ratio, milling time, ball size and alkaline pretreatment were investigated by them. It was found that milling-ball size should be carefully selected in order to produce fibrous morphologies instead of particulates. Milling time and ball-to-cellulose mass ratio need to be controlled as well as it might affect the fiber morphology. Meanwhile, alkali pretreatment can help in weakening the hydrogen bonds between cellulose fibrils and removing small particles, but with the risk of damaging the fibrous morphology. The swollen fibrils and the balls were mixed during milling as illustrated in Fig. 5. The CNF produced was found to have an average diameter of 100 nm and this was obtained through soft mechanical milling conditions using cerium-doped zirconia balls of 0.4–0.6 mm in diameter within 1.5 h without alkaline.

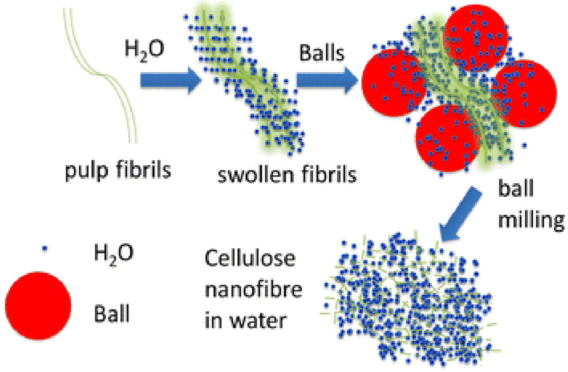


Fig. 5: Schematic diagram of cellulose nanofiber production by ball milling [44]

Wet disk milling

Nanofibrillation of cellulose by wet disk milling (Fig. 6) has been reported to produce CNF with comparable quality to the other treatments described earlier. Wet disk milling has been used to produce CNF from a variety of sources such as oil palm biomass, wood, bamboo and bagasse with a diameter size of less than 100 nm [3, 37, 45-47]. Wet disk milling is a non-chemical process and promises a low production cost [46]. This treatment allows nanofibrillation to occur at a high source solid content level as compared to ultrasonication and high-pressure homogenizing, which is an important criterion to ensure the high productivity of CNF. In regards to this matter, wet disk milling can be assumed as a suitable treatment for the commercial production of CNF. However, to the best of our knowledge, CNF production using wet disk milling is still not commonly used.

Fig. 6: Wet disk milling

Extrusion

Twin-screw extrusion allows the processing and nanofibrillation of cellulose source material with high solid content. Several studies successfully produced CNF with a small diameter size (>100 nm) using this approach [9–11, 48, 59]. Fig. 7 shows a schematic diagram of CNF production by extrusion as reported in [60]. It was found that the degree of nanofibrillation increased as the number of passes increased. This technique is also suitable to commercially produce CNF reinforced biocomposites as it is possible to combine the nanofibrillation, mixing and blending process into one production system [59]. Similar to wet disk milling, extrusion is also suitable a method to produce CNF on a large scale as compared to the other approaches described earlier.

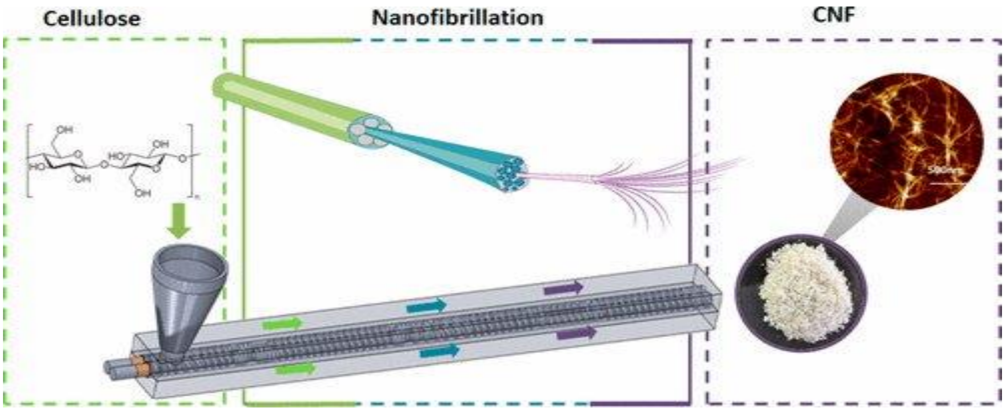


Fig. 7: Schematic diagram of nanofibrillation by extrusion [60]

Factors influencing nanofibrillation of cellulose

Several factors that may give effects on the nanofibrillation process include nanofibrillation techniques, fiber chemical composition and drying history during pretreatment. Without considering these factors, the nanofibrillation process would be difficult to complete and require a longer duration of time, higher cost and might as well requires the usage of hazardous chemicals. A summary of findings focusing on factors that may influence CNF formation has been summarised in Table 2.

Table 2: Factors influencing the CNF formation

Factors	Findings	References
Chemical composition of extracted material	a. The presence of high amount of lignin and hemicellulose from the extracted material influences nanofibrillation. However, a small amount of hemicellulose is known to facilitate the beating of pulp which is related to the phenomenon of "hornification". b. The dissolution of lignocellulosic material in ionic liquids is also influenced by the chemical composition of the lignocellulose. The presence of lignin and hemicellulose lowers the dissolution of the fibers for CNF production through electrospinning.	[3, 37, 40, 46, 61-62]
Drying history	a. The drying history during pretreatment also influences the degree of hornification. Drying reduces fiber swelling capacity and conformability which affects the hornification phenomenon. b. For example, the never-dried refined bleached pulp has a high degree of nanofibrillation using a twin-screw extruder and through wet disk milling, respectively.	[46, 48]
Nanofibrillation treatments	a. Nanofibrillation technique will influence the size of CNF produced. b. For example, electrospinning produces larger-sized of oil palm biomass-CNF (150-2000 nm) while other treatments such as wet disk milling, high-pressure homogenizer and ultra-high sonication produce smaller oil palm biomass-CNF (<100 nm).	[31, 37, 40, 63-64]

Conclusion and future recommendation

In this review, several nanofibrillation treatments for CNF production were reviewed. Different nanofibrillation treatments and lignocellulosic materials result in different properties of the produced CNF. All the advantages and disadvantages of the fibrillation techniques were summarized in Table 3. It is also interesting to note that CNF formation was not solely influenced by the processing treatments, but other factors as well; chemical composition of extracted material and drying process. Looking at the current trend of study, scientists recently focussing on discovering nanofibrillation treatments that can process high solid content source material, and free from chemical processing. From this review, it was revealed that both wet disk milling and extrusion are the most convenient treatments that can be applied for the commercial production of CNF, as both treatments are capable of producing a high capacity of CNF with low production cost.

Table 3: Advantages and disadvantages for each nanofibrillation technique

Nanofibrillation technique	Advantages	Disadvantages
Enzymatic nanofibrillation	- No harsh chemicals required.	- Time consuming. - Complex process (require combination of techniques). - High production cost because enzymes are usually expensive.
Electrospinning	- Large amount of CNF can be processed. - Can be done in a continues production system. - Solvent can be reused.	- High energy required. - Large size of CNF obtained (>100 nm).
High pressure homogenizer	- Small size of CNF could be obtained. - No chemicals required.	- Low concentration of CNF is usually produced. - Not suitable for large production scale.
Ultrasonication	- No chemicals required - Small size of CNF could be obtained.	- Low concentration of CNF is usually produced. - Not suitable for large production scale.
Ball milling	- No chemicals required. - Can be done under ambient pressure and temperature.	- Low concentration of CNF is usually produced. - Time and energy consuming.
Wet disk milling	- High concentration of CNF can be produced. - No chemicals required. - Can be done under ambient pressure and temperature. - Low-production cost. -Suitable for large production scale.	- Time consuming.
Extrusion	- High concentration of CNF can be produced. - Suitable for large production scale.	- Usually only suitable for polymer composite production (combination of several processes).

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