Nanocellulose and Nanocellulose-Based Composites

Subjects: Nanoscience & Nanotechnology
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Nanocellulose is the most abundant material extracted from plants, animals, and bacteria. Nanocellulose is a cellulosic material with nano-scale dimensions and exists in the form of cellulose nanocrystals (CNC), bacterial nanocellulose (BNC), and nano-fibrillated cellulose (NFC). Owing to its high surface area, non-toxic nature, good mechanical properties, low thermal expansion, and high biodegradability, it is obtaining high attraction in the fields of electronics, paper making, packaging, and filtration, as well as the biomedical industry. To obtain the full potential of nanocellulose, it is chemically modified to alter the surface, resulting in improved properties.

nanocellulose biodegradable nanocomposites chemical functionalization extraction applications

1. Introduction

Polymeric cellulosic materials with high biodegradability and eco-friendliness have received a lot of attention, owing to the damage caused by petroleum base products, such as global warming, green gas emissions, and many others [1]. Several researchers are working on cellulosic fibers, from which nanocellulose can be extracted. The use of nanocellulose as a reinforcement in composites is because of their mesoscopic properties [2]. Nanocellulose is derived from plant cell walls and has extremely valuable properties, such as high surface area and strength [3][4][5]. Furthermore, the nanocellulose surface is easy to modify because of the large number of hydroxyl groups present in its structure. Nanocellulose has numerous applications in our daily lives, including filtration membranes, food packaging, biomedical, and so on [6].

2. Nanocellulose and Its Various Sources

As shown in **Figure 1**, the cell walls of most plants contain hemicellulose, cellulose, and lignin. Lignin acts as a binder between cellulose and hemicellulose, holding them together. It has high stiffness and strength and can protect the cell wall from the outside environment. The amount of lignin in the plant cell wall ranges from 10 to 25% by weight, while the amounts of hemicellulose and cellulose are 20–35% and 35–50%, respectively [1][7][8]. The main component of the cell wall is cellulose, composed of repeating units of cellobiose, linked together with β-1,4 linkages, as shown in **Figure 1**. Intermolecular or intermolecular hydrogen bonding is used to connect the repeated units. Bonding occurs between the same or different chains via open hydroxyl groups [9]. Hemicelluloses are primarily xylans and glucomannans, connected by short or branched chains. Hydrogen bonding plays a vital role in providing compactness, strength, and solvent impermeability to the networks in cellulose fibers.

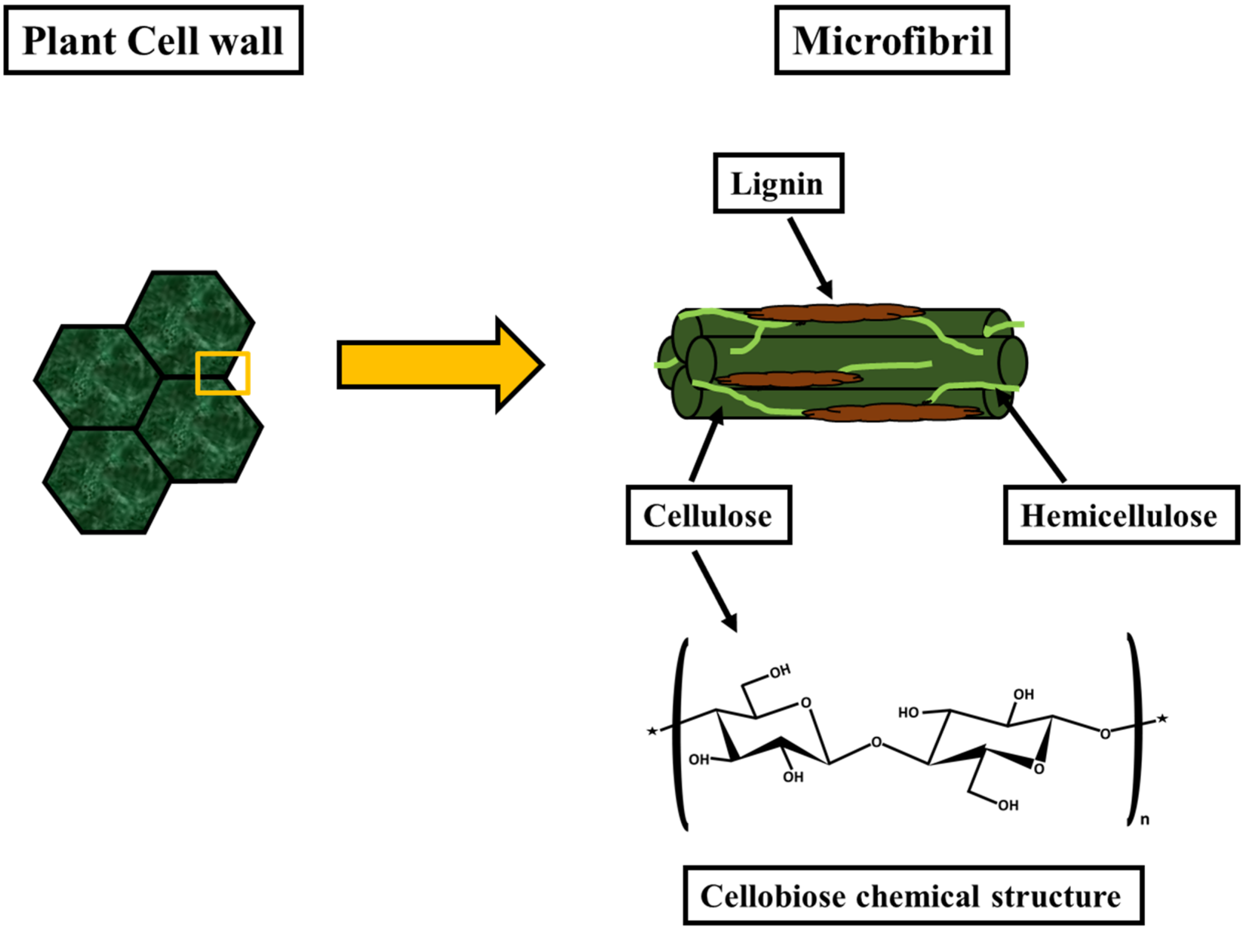


Figure 1. Plant cell wall structure and cellobiose chemical structure.

Strong hydrogen bonding networks and a variety of hydroxyl groups give cellulose fiber exceptional physical and mechanical qualities. The orderly packing of the chain molecules in the crystalline parts promotes high stiffness, whereas the amorphous parts give flexibility to the bulk material [10]. For general lignocellulosic biomass, the cellulose fibers present in between the crystalline and amorphous regions have a diameter of 3–100 μm and a length of 1–4 mm [11].

Nanocellulose fibers with a diameter of less than 100 nm and a length in the micrometers range deserve special attention. Nanocellulose fibers are transparent and rich in hydroxyl groups. These groups have a reactive surface that can be modified to obtain the desired properties [12]. Nanocellulose nanofibers have a low density of 1.6 g/cm^3 and has exceptional strength [11]. Additionally, they have a tensile strength of nearly 10 GPa and a high strength-to-weight ratio that is eight times greater than stainless steel.

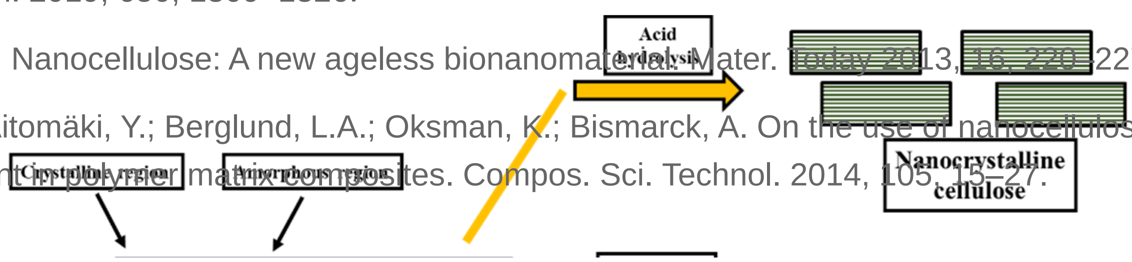
The three primary forms of nanocellulose materials are BNC, NFC, and CNC, as shown in **Table 1**. Three cellulosic forms have unique qualities, including biodegradability, tunable surface chemistry, barrier properties, non-toxicity, high mechanical strength, crystallinity, and high aspect ratio. Such a remarkable nature of nanocellulose makes it a new material for food packaging and fillers in composites [13][14]. High-strength nanocellulose, sometimes referred to as CNC, is typically recovered by the process of acid hydrolysis from cellulose fibrils [15]. It is shaped like a short rod or a whisker and has a diameter of 2–20 nm and a length of 100–500 nm. Additionally, it is entirely composed of cellulose, with high crystallinity ranging from 54 to 88%. The long and entangled nanocellulose that may be mechanically removed from cellulose fibrils is another type of nanocellulose and is known as NFC, often referred to as micro-fibrillated cellulose. Its size ranges from 500 to 2000 nm in length and 1–100 nm in diameter [16][17]. It is made from 100% cellulose, with crystalline and amorphous region parts. Another different type of nanocellulose is BNC. It is formed by bacteria, primarily *Gluconacetobacter xylinus*, over a few days to two weeks, whereas lignocellulosic biomass is the primary constituent for the extraction of CNC and NFC (top-down method). As BNC is extracted from bacteria, other amorphous compounds, such as lignin, hemicellulose, and pectin, are never present in the pure form of BNC [18][19]. The chemical makeup of BNC is identical to that of the other two types of nanocellulose.

Table 1. Types of nanocellulose materials.

Nanocellulose Types	Sources	Extraction Method and Size
CNC	Cotton, tunicin, mulberry bark, hemp, wood, wheat straw.	Acid hydrolysis 5–70 nm in diameter 100–250 nm in length
BNC	Sugars and alcohols.	Extracted from bacterial synthesis 20–100 nm in diameter
NFC	Wood, hemp, flax, potato tuber, sugar beet.	A mechanical method of breaking the cellulose 5–60 nm in diameter

3. Nanocellulose Extraction Processes

The use of agricultural leftovers for the extraction of nanocellulose is a very appealing field from the researcher's point of view. Agriculture leftovers are lignocellulosic biomass that are rich in cellulosic content. The extraction of nanocellulose from lignocellulosic biomass involves various steps that can be seen in **Figure 2**. First, the pretreatment eliminates the non-cellulosic compounds, including lignin, hemicellulose, pectin, etc., followed by nanocellulose extraction through various extraction techniques [20].



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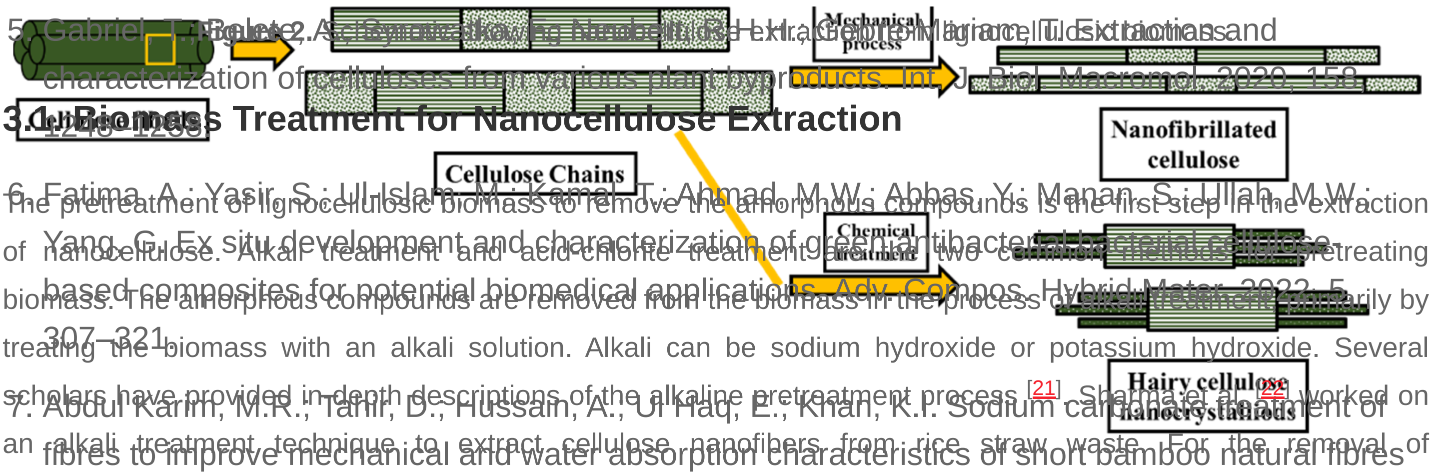

Figure 2. A Synoptic showing Nanocellulose extraction.

3.1. Biomass Treatment for Nanocellulose Extraction

The pretreatment of lignocellulosic biomass to remove the amorphous compounds is the first step in the extraction of nanocellulose. Alkali treatment and acid-chlorite treatment are the two common methods for pretreating biomass. The amorphous compounds are removed from the biomass in the process of alkali treatment primarily by treating the biomass with an alkali solution. Alkali can be sodium hydroxide or potassium hydroxide. Several scholars have provided in-depth descriptions of the alkaline pretreatment process [21]. Sharma et al. [22] worked on an alkali treatment technique to extract cellulose nanofibers from rice straw waste. For the removal of hemicelluloses and lignin, the rice straw was heated to 90–160 °C for 1–2 h after being soaked in various NaOH concentrations (8–16%).

Acid-chlorite treatment is another technique used for the removal of amorphous compounds. The simultaneous treatment of sodium chlorite, along with glacial acetic acid as an acidifying agent, removes the majority of the lignin from the lignocellulosic biomass. This procedure is also referred to as a bleaching or delignification technique. It is carried out when a mixture of lignocellulosic biomass and distilled water is added to the sodium chlorite and acetic acid solution at 70–80 °C for 4–12 h [23]. After completion of the treatment, the mixture is stirred continuously for an entire night before being washed with distilled water to bring the pH level to a neutral state. This leads to a collection of white residue, which is then dried at 50 °C in an oven to obtain lignin-free holocellulose [24].

3.2. Nanocellulose Isolation

Acid hydrolysis, mechanical treatment, processes, and enzymatic hydrolysis are some of the techniques used for nanocellulose isolation. **Table 2** shows three cellulose isolation processes. The most commonly used technique for extracting nanocellulose is acid hydrolysis [25]. It involves the use of strong acids, such as sulfuric acid, that can easily hydrolyze the amorphous area of cellulose fibrils by esterifying the hydroxyl groups with sulfate ions [26][27]. Maiti et al. [28] suggested the use of 47% of sulfuric acid to recover nanocellulose from waste tissue papers, China cotton, and south African cotton. The results show that the types of the precursors and the hydrolysis conditions largely determine the shape and size of the nanocellulose. It was noted that esterification creates a colloidal dispersion of crystalline nanocellulose inside the reaction mixture. Acid hydrolysis can be performed by other mild acids, such as formic acid, phosphoric acid, etc. Bond cleavage is the mechanism behind the acid hydrolysis of cellulose. The glycosidic bonds present between anhydroglucose units are subjected to hydrolytic cleavage, resulting in the rearrangement of tangling chains and strain release. Acid hydrolysis thereby dissolves the amorphous portion, leaving the crystalline sections intact. Formed crystalline regions are then projected to mechanical treatment, which transforms them into fine cellulose particles. The use of a 30–50% concentration of sulphuric acid in acid hydrolysis gives very fine cellulose particles. Reinforcing fine cellulose particles in biocomposites facilitates high plastic deformation in the composite. Acid hydrolysis reactions and the properties of nanocellulose can be controlled by varying the concentration, time, and temperature provided [29]. The size of cellulose particles and their distribution depend highly on the temperature, time, and concentration of the acid. A

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high temperature and short reaction times are usually recommended for dilute acid hydrolysis, using dilute acid eliminates the need for acid recovery. Iranmanesh et al. [30] performed dilute acid hydrolysis and found that, to achieve a high yield from acid hydrolysis, high temperature, less time, and high pressure are required. Similarly, Hamelinck et al. [31] performed hydrolysis using concentrated acid and concluded that hydrolysis through concentrated acids requires moderate temperatures and longer reaction times. The use of concentrated acid enhances the difficulty of recovering acid from the mixture. Later stages of acid hydrolysis also involve the washing of cellulose, which is usually achieved by centrifuging the mixture of cellulose and cold water [32]. The main disadvantage of this procedure is the need to treat acid-containing wastewater before releasing it into the environment.

Table 2. Nanocellulose isolation methods.

Biological Methods	Mechanical Methods	Chemical Methods
Fungi treatment	Steam explosion	Ionic treatment
Bacteria treatment	Ball milling	Alkaline treatment
Enzymatic hydrolysis	Disintegration	Acid hydrolysis
	Grinding	Oxidation
	Electrospinning	Solvent extraction
	Ultrasonication	
	Homogenization	

Enzymatic hydrolysis comes under the list of biological processes that use enzymes to break down the cellulose fibers into washed cellulose [33]. The literature reveals that cellulase, cellobiohydrolase, endoglucanase, etc., are frequently used enzymes for this process. Although the mechanism is intricate, the enzyme's activity is dependent on catalyzing the breakage of the connecting H-bonds between the cellulosic fibers [34]. Enzymatic activity involves the removal of hemicellulose, protection of cellulose from hydrolysis, and production of monosaccharides from hemicellulose for subsequent fermentation to produce bioethanol [35]. The cellulases and hemicellulases present in the process are closely connected to effectively hydrolyze a variety of lignocellulosic biomasses. When compared to acid hydrolysis, it is normally conducted under milder conditions and takes significantly longer to operate. To reduce the processing time, enzymatic hydrolysis can be used in combination with other techniques. Mohiruzzaman et al. [36] used a novel method to separate nanocellulose from wood by pre-treating the cellulose with an ionic solution, followed by laccase-enhanced enzymatic hydrolysis. A comparison of the produced nanocellulose to the existing methods of cellulose preparation shows that the produced cellulose has a high surface area and higher crystallinity with enhanced thermal properties.

Different mechanical techniques, including ball milling, ultrasonication, and high-pressure homogenization (HPH), can be used to mechanically prepare nanocellulosic fibers [14]. However, these methods require a lot of energy, which is why some pre-treatment is always suggested to save energy. HPH involves the treatment of a cellulose

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mixture at high pressure and velocity [33]. High pressure and velocity induce the cellulose micro fibrils into nanometer-sized fragments, based on the fluid shear and impact forces created. Figure 3 shows the schematic of the homogenizer used for cellulose isolation. Li et al. [37] used HPH to separate nanocellulose from sugarcane bagasse. To dissolve the bagasse cellulose, the material was initially pre-treated with an ionic liquid (1-butyl-3-methylimidazolium chloride ([Bmim]Cl)), followed by passing the homogenized solution at high pressure, without becoming clogged. The produced nanocellulose had a diameter of 10–20 nm, with a crystallinity lesser than the original cellulose. By using HPH, Wang et al. [38] separated nanocellulose from cotton cellulose. The final product obtained (20 nm in diameter) was less thermally stable and had a low crystallinity than the pre-treated cotton cellulose. A decrease in crystallinity is because of the high pressure, which disrupts cellulose intermolecular and intramolecular hydrogen bonding.

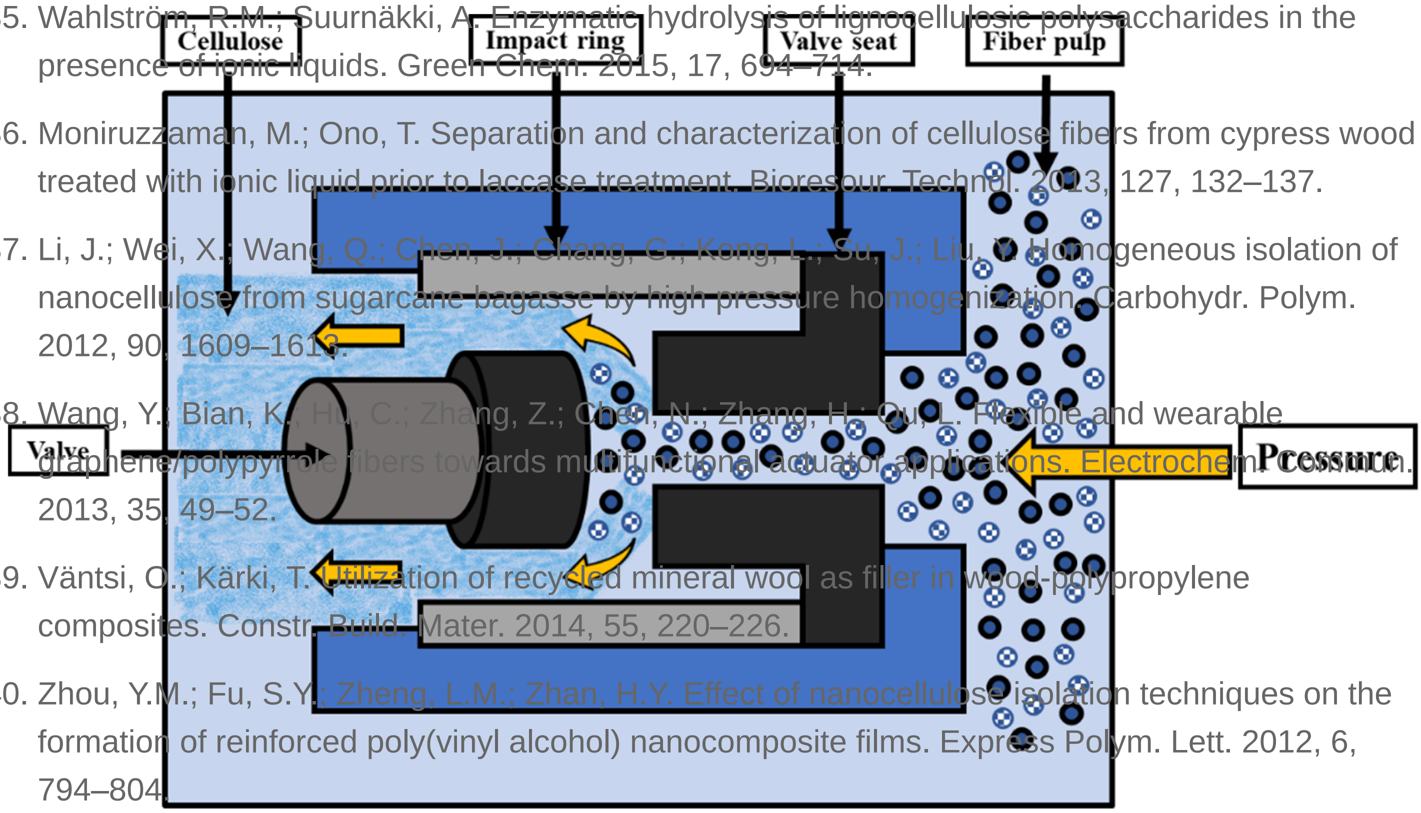

Figure 3. Schematic for HPH.

Cellulose fiber can also be defibrillated, using the ultrasound's hydrodynamic forces, through the process of ultrasonication [39]. In this technique, ultrasonic energy is generated by mechanically oscillating the power and implosion of gas bubbles. The generated energy is then absorbed by the liquid molecules [40][41]. Tang et al. [42] used the process of ultrasonication to extract the nanocellulose from the wood pulp. The results showed that cellulose obtained without ultrasonication has a yield of 48.16%, whereas ultrasonication of the sample gives the cellulose sample a yield of 85.38%. The obtained nanocellulose had widths of 10–100 nm and a yield of 85.38%.

Cellulose fibers are defibrillated using another mechanical technique, which involves the use of ball milling. Since the 1990s, researchers have used ball milling for grinding and improving particle size. The milling jar contains

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milling balls of various sizes, as one of the primary components of the ball milling machine. Planetary ball mills and vibration ball mills are some of the common types of ball mills used in industry and laboratories today [43]. The planetary ball mill is the one that defibrillates cellulose and biomass the most frequently. In the planetary ball, the mill balls collide with each other and with the wall of the milling jar, thus creating friction, which helps in size reduction [44][45]. The created friction grinds the large-size materials into smaller particles with large surface areas. The friction is linked with the shear forces that are produced between the balls and the surface of the rotating jar as a result of the centrifugal force [46][47]. The application of shear force onto the cellulose breaks them into nano-size particles. The number and size of the balls, time, weight ratio between the balls and material, and speed are a few of the variable elements on which the characteristics of ball-milled products depend [48]. The planetary ball mill is also shown in **Figure 4**. Ago et al. [49] investigated the properties of ball-milled, cotton-derived cellulose at 400 rpm for two hours by varying the water content in the mixture. It was discovered that the presence of a small amount of water (10 wt.%) in a dry state changes the cellulose type I to amorphous. However, when water is up to 30 wt.% cellulose type I, it transformed into the stable form of cellulose type II. This suggests that the crystalline structure of the cellulose is greatly influenced by the amount of water in the milling jar [49].

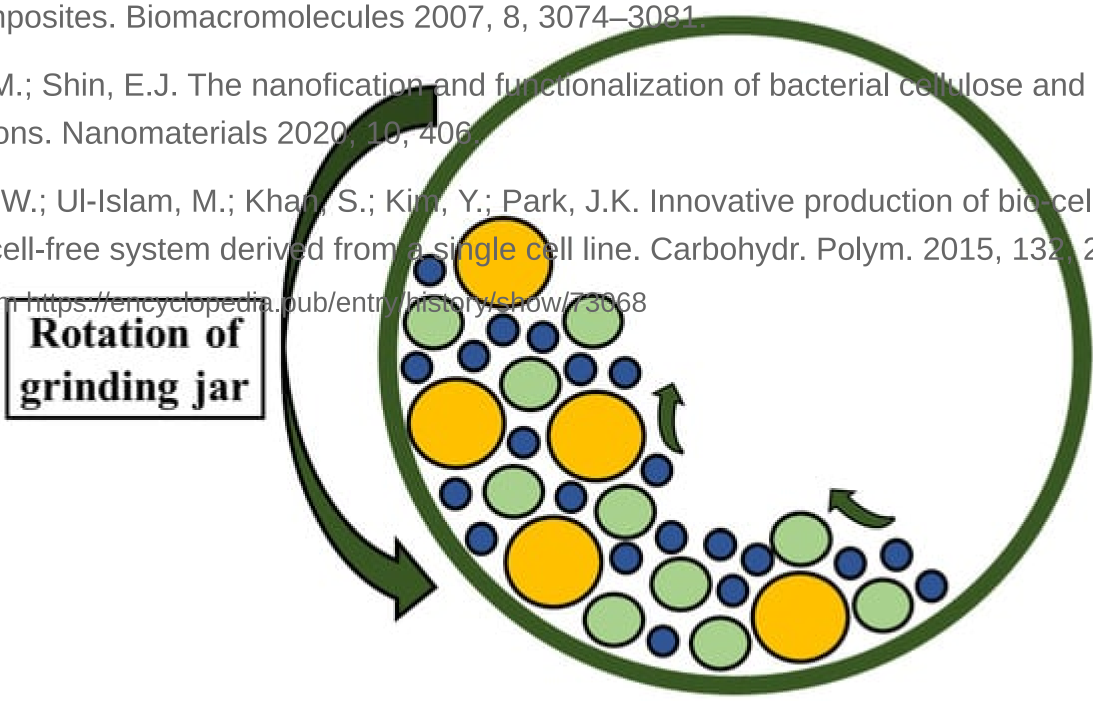

Figure 4. Schematic for planetary ball mill.

3.3. BNC Extraction

Besides plants, bacteria can be used to produce cellulose. Bacterial cellulose can be used as a primary source for the production of CNC and cellulose nanowhiskers because of their high purity and crystallinity. It is generally acknowledged that the source of the bacterial cellulose and the isolation techniques utilized affect the shape of BNC. Some commonly used bacteria are *Pseudomonas, Rhizobium, Sarcina, genera Acetobacter, Azotobacter,* and *Alcaligenes. Acetobacter xylinum*, a species of bacteria that produces acetic acid, is the most effective generator of BNC. Cellulose biosynthesis is the method of extraction of BNC. Extracted BNC has a width of less

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than 100 nm and is 100–1000 nm long [50][51]. BNC isolation from bacterial cellulose can be achieved by using acid hydrolysis, enzymatic hydrolysis, and ionic liquids. By acid hydrolysis, CNC and BNC can likewise convert into bacterial nanocrystals. However, acid hydrolysis also has some disadvantages, as it decreases the degree of polymerization (DP) and reduces the number of sulphate-containing nanocrystals. Reduction in DP and nanocrystals brings down the mechanical properties of cellulose nanocomposites. Therefore, the enzymatic system is suggested to retain the actual properties of bacterial cellulose. Ullah et al. [52] developed a cell-free enzyme system for producing bio-cellulose. The system was developed using a single-cell line and contained all the enzymes needed to run a successful biosynthesis process. The prepared bio-cellulose were scattered and had extracellularly produced glucose chains. The results revealed that a better yield can be obtained by following the produced cell-free system.