REVIEW   **OPEN ACCESS**

Cellulose in Nature – Versatile Sources for Novel Applications: A Literature Review

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Abstract
Introduction: Cellulose is the most abundant organic compound and the main component of the plant cell wall. However, it can be derived from other sources: tunicates, algae, and bacteria. Different sources of cellulose were shown to produce end-products of different mechanical properties and were considered for producing cellulose in non-industrial conditions.
Methods: Procedures for the extraction of cellulose from different sources are described. They are similar for plants and algae, including bleaching and purification processes amongst others, while bacteria found in symbiotic cultures of bacteria and yeasts (SCOBYs) are capable of growing cellulose layers above their cultivation media. After extraction or bacterial cultivation, mechanical treatments are performed in order to modify new cellulose layers for specific applications.
Results: Plant sources of cellulose are various and widely available, and often used for the industrial production of cellulose. Algae-derived microcrystalline cellulose (MCC) is similar to that from plant sources, but has higher crystallinity and, often, requires simpler extraction processes. Finally, cellulose grown by bacteria found in SCOBYs is the most optimal for non-industrial conditions, owing to the simplest cultivation and extraction procedures.
Discussion: On a large scale, plant sources of cellulose are the optimal ones. The main downside of algal cellulose is that it is season-dependent, and more difficult to acquire than bacterial and plant sources. While having access to laboratory conditions for incubations and using a pure bacterial culture would be preferable, cultivation methods are simple enough to be adapted for home conditions. Also, conditions of incubation can be varied based upon the intended properties of the end-product: the efficiency of cellulose growth and its properties depend on the chosen carbon source. When bacteria produce a cellulose layer or it is extracted from another source, mechanical treatments for tuning porosity and other properties are applied.
Conclusion: Sources of cellulose are numerous, and some are more suitable than others for non-industrial production, namely, using easily obtainable SCOBYs. This allows for a wide variety of applications: from artificial skin and face masks, to sustainable batteries and different food products.

Keywords: cellulose; tunable porosity; bottom-up synthesis; SCOBY; crystallinity

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Introduction

Cellulose, being the main component of the plant cell wall, is a tough and water-insoluble polymer, which is one of the most abundantly found organic compounds. [1]. D-glucopyranose molecules linked by 1-4 glycosidic bonds (Figure 1) form chains that can either be arranged as fibrils or exhibit an amorphous structure. Between 40 and 45% of natural cellulose regions exhibit a higher degree of order, forming microcrystalline cellulose (MCC), while 55 to 60% of natural cellulose is amorphous [2]. Both growth and post-treatment conditions, along with the extraction process, affect the degree of crystallinity (DC), and therefore the derived material's properties such as strength and hydrophilicity. MCC is expected to have different chemical resistance compared to amorphous regions due to its ordered structure which inhibits penetration of chemicals [3]. MCC serves as a desirable filler which can be used for enhanced mechanical properties of silicones amongst other applications.
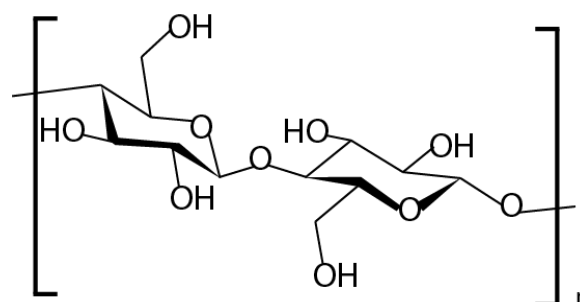

Figure 1. Chemical structure of the repeating unit of cellulose.

Types of Cellulose and Characteristics

Cellulose is found as one of four polymorphs: I, II, III (III$_I$ and III$_{II}$), and IV [4]. Cellulose I (CI) is the native material found in plants and consists of parallel polymer chains within crystallites and represented the unmodified cellulose. The remaining polymorphs are types of modified

cellulose. Cellulose II (CII) can be formed either through alkaline treatment of any of the cellulose polymorphs and through regeneration of dissolved cellulose. Ammoniacal treatment of cellulose I and II yields cellulose III (CIII$_I$) and cellulose IV (CIII$_{II}$) [5] and these allomorphs can be converted into CIV$_I$ and CIV$_{II}$, respectively [6]. As shown in Figure 2, the crystallinity of cellulose can be described in terms of crystalline and non-crystalline domains arranged as in a two-phase model, where crystalline and amorphous regions alternate [7, 8].
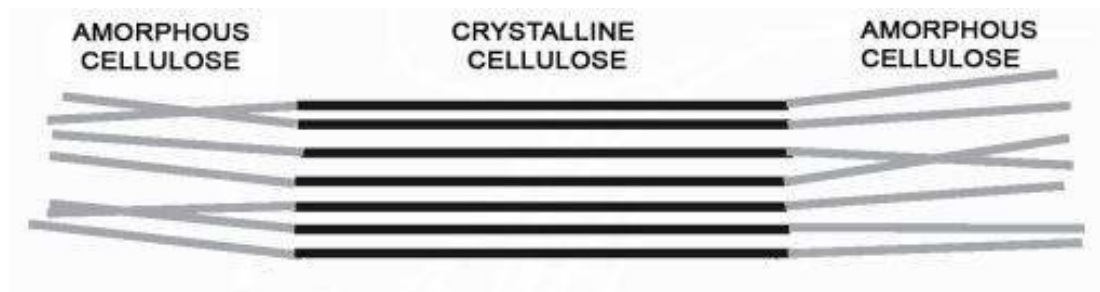

Figure 2. Two-phase model of cellulose; adapted from Quiroz Castañeda and Folch-Mallol [8].

The abundancy of cellulose crystalline regions will determine a material's properties [9]. For instance, for a higher DC hardness and stiffness (Young's modulus) increase, while flexibility decreases. Thus, degree of crystallinity is used to describe crystalline structures and influences the material's stiffness, hardness, and heat resistance [10].

Cellulose can be acquired from a variety of sources including plants, algae, bacteria, and tunicates. However, different organisms produce cellulose of various levels of DC which leads to drastically different mechanical properties and application possibilities. For example, as amorphous cellulose successfully prevents adhesion, it is effectively used as antioil-fouling material for water oil separation [11], while MCC is applied in bio-composites, owing to its chemical inactivity and hygroscopicity [12]. Because of this, finding cheaper and more efficient ways to produce cellulose would have the potential of lowering the cost of such bio-composites.

Sources of cellulose

A wide variety of plant sources can be used for cellulose extraction, with the most important being wood and cotton; however, corn, banana, rice, aloe vera, and many others can serve the same purpose [3]. Properties and structure of cellulose from plant sources depend on different parameters including climate conditions, soil type and characteristics, botanical origin amongst others. On an industrial level, it is a low-cost, high-volume, sustainable option, with a relatively simple method of extraction, but for non-industrial conditions, other sources of MCC may be easier to synthesize—the process of extracting cellulose from plant sources is complex and involves several steps suitable only for large-scale industrial operations such as physical break-down of the plant matter using processes such as grinding and homogenization [13] (Figure 3).
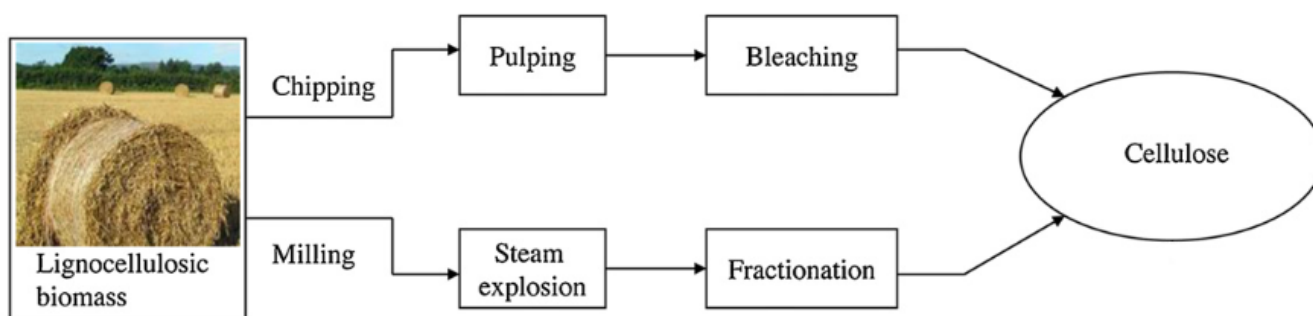

Figure 3. Industrial extraction of cellulose; adapted from Brinchi et al. [13].

Cellulose is also formed as a primary metabolic product of many bacteria, including *Acetobacter xylinum* and members of the *Gluconacetobacter, Sarcina*, and *Agrobacterium* genera [4]. Such cellulose is generally purer than that found in plants, does not contain lignin or hemicellulose, but rather only glucose monomers, and is formed in thin layers [14]. It is of the CI crystalline type and has a degree of polymerization (DP) between 2000 and 6000 [4]. DP in plant cellulose ranges from about 925 to 5500 and in algae it was shown to be 4300 [15]. DP has a direct impact

on cellulose application—higher values result in a tougher final paper product. Bacteria found in a symbiotic culture of bacteria and yeast (SCOBY), as seen in Figure 4, used for production of beverages as kombucha and kimchi are known for their ability to grow a cellulose layer over the medium they inhabit [16, 17].

Figure 4. SCOBY floating on kombucha it was used to ferment.

Algal cellulose also does not always contain lignin and, therefore, has an even higher degree of crystallinity than bacterial cellulose [18]. Extracting algal cellulose is a means of remediation of marine ecosystems, as excessive and unwanted booking of algae damages the marine ecosystem. The extraction of microcrystalline cellulose is relatively simple and involves three groups of algae [3]. The first group, green algae (*Chlorophyta*), contain "native cellulose" and include species from the genera *Cladophorales* and *Siphonocladales*. *Spongomorpha*, which is a member of the second group, has "mercerized-like cellulose" with polymer chains randomly oriented resulting in a low DC. Cell walls found in the members of the third group (*Spyrogira*) are not entirely made up of cellulose [3]. Using X-ray diffractometry, marine green algae cellulose was found to be more crystalline, compared to that extracted from freshwater algae [19]. Cellulose nanofiber yield and physical properties depends on the season—algae harvested too early (with underdeveloped cell walls) or towards the end of the season were observed to have low tensile strength [18], compared to the optimal samples gathered midseason [20].

Tunicates are the only known animal source of cellulose [21]. Cellulose nanofibers (CNFs) can be extracted from the cover layer located over the entire epidermis, where cellulose-protein fibrils are cemented by sulfated mucopolysaccharides or sulfated glycans or lipids. However, given that tunicates are not widely available, other sources of cellulose may be preferable for applications.

Porosity

Porosity is an important morphological parameter in cellulose application. Pore shape, volume, and size distribution are all features of porous media [22] and can be modified by chemical and mechanical treatments. In bacterial cellulose, carbon sources can affect cellulose production as metabolic pathways may be longer for non-optimal carbon sources, and, thus, pore size can vary [23]. Longer cultivation time decreases porosity due to a higher density of fibrils.

Pore size is important in the application of cellulose as it determines a material's water-holding capacity and oxygen permeability. This is a significant aspect of cellulose which makes it a fascinating candidate for applications that require films of controllable porosity, such as mask filters. The question that stems from the abundance of cellulose and a myriad of extraction methods; is it possible to grow cellulose of controllable porosity feasibly using unconventional production methods?

Methods
Extraction of cellulose from plants and algae

Before the process of extraction, to prepare for the chemical treatment, raw materials are milled or ground and purified [3].

Plant cellulose

The first step of the extraction of cellulose from plants involves removing other compounds and components, including oils, wax, pectin, lignin, and hemicellulose [3]. A method described by Samiee et al. [3] involves alkali-bleaching for purification and a treatment with $NaClO_2$ solution under acidic conditions. In the main step, amorphous regions of cellulose are removed by acid hydrolysis, with either HCl or H_2SO_4, which does not impact stronger and more resistant crystalline regions. The same process hydrolyzes the remaining hemicellulose and pectin, yielding simple sugars.

Algal cellulose

Since cellulose derived from algae and plants is similar both chemically and structurally, Samiee et al. [3] indicate that the extraction processes are similar. However, since many species of algae contain colorful pigments, the extraction process may need additional steps for pigment removal, where $NaClO_2$ or H_2O_2 are used as bleaching agents.

Tarchoun et al. [24] described a method for the extraction of cellulose from the *Posidonia oceania* brown algae, involving initial extraction with ethanol, oven drying, treatment with hot water, acidification with sodium chlorite, and a treatment with potassium hydroxide for the removal of pectin and hemicellulose. The change of color from brown to white indicates the absence of lignin, which may not be present in all algae. After treatments, samples are washed with distilled water and the extracted cellulose is left to dry

in order to prevent "self-destruction" of the material [3]. Then, mechanical treatments for controlling porosity are performed.

Koyama et al. [19] described a single method for the extraction of cellulose from various algae (*Valonia, Cladophora, Micrasterias, Spirogyra,* etc.) involving an alkaline treatment and acid hydrolysis, and a bleaching process similar to that described by Samiee et al. [3]. For non-industrial conditions, such method may be used, with the change in the bleaching process: $NaClO_2$ may be replaced by the easily obtainable H_2O_2.

After extraction, algal cellulose is dried to prevent "self-destruction" of the components [3].

Bacterial cellulose

Members of the *Gluconacetobacter* genus found in a SCOBY are capable of growing cellulose. According to Ashjaran et al. [17], the optimal source of carbon for these species is glycerol. However, sucrose, glucose, or another source of carbon may be used to insure a different pace of cellulose production and ultimately, different porosity levels. The growth medium is kept inside a large container and water, bags of green or black tea are added, along with vinegar, necessary for cellulose production in these bacteria. After 2-3 weeks of incubation, an impure cellulose layer containing cells and medium residue is formed [17]. Tang et al. [23] showed that *Gluconacetobacter xylinum* produced a higher yield with lower porosity after longer incubation. For purification, deionized water and 80% NaOH are used and neutralization by 1% acetic acid is performed afterwards. This results in a thin cellulose layer ready for post-extraction treatments.

Post-growth Treatments

In order to vary porosity levels, newly synthesized and purified cellulose can be dried using different methods which cause water within cellulose pores to evaporate due to capillary forces and high surface tension of water. The collapsing pressure, causing pores to shrink, is inversely proportional to the tube diameter [25]. Additionally, larger pore size affects internal liquid's pressure by increasing forces necessary for pore shrinkage, just as different temperature levels affect the way water leaves cellulose pores and, thus, the shrinkage of pores themselves. Park et al. [22] showed that water bound content drops as temperature increases. Some methods of drying more suitable for industrial use include freeze drying, drum drying, and spray drying [3]. However, there are methods that can be applied in non-laboratory conditions. Tang et al. [23] showed that freeze-drying resulted in significantly higher porosity compared to hot-air dried samples. Similar results may be replicated in home conditions by drying using accessible means: sunlight, keeping samples at room temperature or in a freezer for extended periods.

Results

Plant cellulose

The choice of a source of cellulose can be made based on the intended end-product. For instance, cellulose derived from banana peel is biodegradable and exhibits high crystallinity, while that extracted from *Eucalyptus* pulp has high retention value, strength, and high moduli of rupture and elasticity. Prakash Menon et al. [18] described various plant sources of cellulose nanofibers and properties of the fibers extracted from them including the sizes as illustrated in Table 1. It is interesting to note that a variety of different methods can be used to vary the size between a wide range of 2-260 nm.

Table 1. Sources of cellulose, methods of preparation, and properties of the material; adapted from Prakash Menon et al. (18)

Source of CNF	Method of preparation	Properties of the material developed	Reference
Rice husk from *Oryza sativa*	Hydrothermal approach, acid-alkali treatment, mechanical disruption	Size; 30–40 nm, innate fluorescence property, purity, crystallinity, thermostability	26
Fibrous residues of *Achira rhizomes*	Acid hydrolysis, high pressure homogenisation	Size; 13.8–37.2 nm, high crystallinity ($I_{cr} = 57.5\%$ and 69.8%), biodegradability, mechanical stability	27
Banana peel	Chemical and enzymatic treatment using xylanase	Size; 10.9 nm and 7.6 nm, biodegradability, high crystallinity ($I_{cr} = 49.2\%$)	28
Powder from poplar wood	Chemical pre-treatment, high intensity ultrasonication	Size; 5–20 nm, high thermostability (335 C), high crystallinity (69.34%)	29
Poplar wood, culms of moso bamboo, rice straw, corn straw	Chemical treatment, ultrasonication, high pressure homogenisation	Size; 2–5 nm, high stability, ribbon like structure, high flexibility	30

Source of CNF	Method of preparation	Properties of the material developed	Reference
Tomato peels	Acidified sodium chlorite, chlorine free alkaline peroxide	Size; 260 ± 79 nm, high crystallinity ($I_{cr} = 69\%$)	31
Posidonia oceanica balls and leaves	Chemical treatment, fibrillation	Size; 5–21 nm and 2–15 nm	32
Cotton stalks	Chemical treatment, ultrasonication, mechanical treatment	Size; 3–15 nm, cost effective, biodegradable	33
Waste pulp residues from paper industry	Etherification of pulp, mechanical disintegration	Size; 10–100 nm, high fibrillation, high thermostability (320° C), high nitrate adsorption capacity (0.7 mmol g^{-1})	34
Culinary banana peel	Chemical treatment, high intensity ultrasonication	High crystallinity ($I_{cr} = 63.64\%$), high thermal stability (295.33° C)	35
Eucalyptus pulp	TEMPO mediated oxidation	High water retention value (WRV = 8.3 g g^{-1}), high modulus of rupture and modulus of elasticity (MOR = 35 MPa, MOE ¼ 5160 MPa), high strength	36
Canola straw	Nanowelding	Size; 53 ± 16 nm, high tensile strength (208 MPa), Young's modulus (20 GPa), superior transparency (76%), biodegradability	37
Oil palm trunk, oil palm frond, okara	Alkaline treatment, electrospinning	Size; <500 nm, high fiber content (107.9%, 67.2%, 25.1%), high anti-oxidant activity (377.2%, 367.8%), superior mineral (Fe, Zn, Cu, Ca) binding activity, high emulsion activity ($66.3 \pm 0.6\%$, $6.6 \pm 0.1\%$, $4.0 \pm 0.1\%$)	38

Algal cellulose

While algal cellulose is structurally similar to plant cellulose, the extraction process is generally simpler and cheaper, especially as algae that do not have lignin in their fiber result in a higher quality final product [3]. Algal cellulose also exhibits a higher rate of growth compared to plants which gives them an advantage in industrial applications. Algal cellulose can grow in a wider range of conditions such as oceans, lakes, ponds, and wastewaters [3]. Koyama et al. [19] used a single method for the extraction of cellulose from various algae (*Valonia, Cladophora, Micrasterias, Spirogyra,* etc.) involving an alkaline treatment and acid hydrolysis, and a bleaching process similar to the one used in the extraction of cellulose from plant sources. For non-industrial, conditions such methods may be used, with a modified bleaching process—$NaClO_2$ may be replaced with the easily obtainable H_2O_2. Also, for ease of extraction, algae with no lignin should be chosen, in which case a step can be skipped.

Bacterial cellulose

Cellulose layers formed over media containing SCOBYs will have different properties based on the cultivation conditions [17]. Longer cultivation time decreases porosity due to a higher density of fibrils produced. Therefore, cultivation time and type of carbon source used form critical parameters that can be tuned for a controllable porosity. As for other sources of cellulose, post-growth mechanical treatments are required for this control over porosity.

Discussion
Plant cellulose

Different end-product properties of plant-derived cellulose allow for a variety of applications: food-packaging, cationic and anionic electrodes, and as drug carrying vehicles for scaffold substrate in tissue engineering [39]. However, methods for extraction are complex and are often adapted for industrial or laboratory conditions, while other types of cellulose may be adapted for the same purposes. Additionally, conventional hydrolysis treatments are not considered environment-friendly, unlike other industrial processes such as enzymatic hydrolysis [3]. As a candidate for a small-scale production, plant cellulose is perhaps not the best candidate and algae and bacteria may be preferable sources of cellulose.

Algal cellulose

Due to their strength, CNFs extracted from *Cladophora* have a good potential for applications [21] such as food packaging, wound dressings, and pharmaceutical applications such as hydrogels. They can also be used for the fabrication of conductive paper-based energy storage devices [40]. Here, nanocellulose fibers would serve as the porous interlayer and flexible substrate for lithium metal batteries [41].

However, despite both high crystallinity and advantageous properties of algal CNFs, growing bacterial cellulose at a small scale may still be preferable for applications as it has greater reproducibility and more room for tuning of parameters for favorable properties. Figure 5 demonstrates differences in structures between the three sources of cellulose—bacterial microcrystalline fibrils are smaller than algal and plant ones (around 100 times) [42].
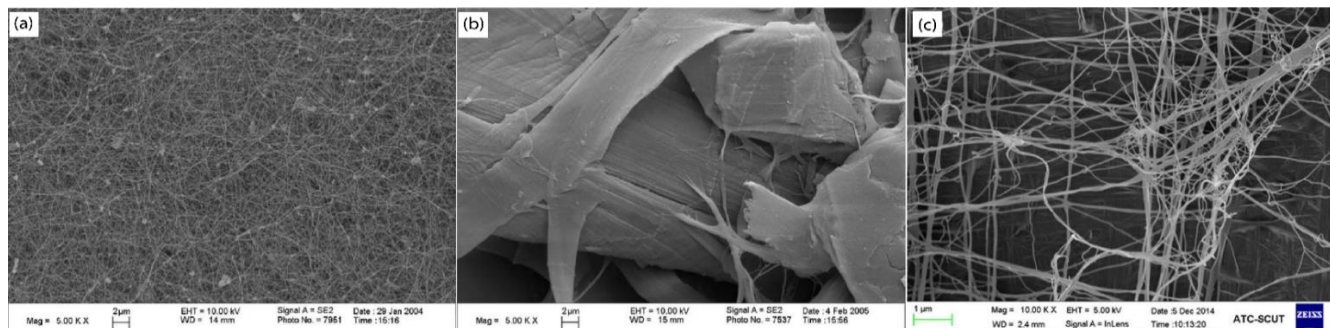

Figure 5. Scanning electron microscope images of (a) bacterial cellulose, (b) wood pulp (middle), and (c) algae cellulose; adapted from Czaja et al. [39] and Xiang et al. [42].

Bacterial cellulose

Growing bacterial cellulose at home and on a small scale is certainly possible and can be adjusted for varying porosity levels. In fact, cellulose is the main component of a fermented coconut water gel, 'Nata de Coco' [43]. Similar methods in controlled laboratory conditions are still preferable and more efficient. A single, pure bacterial culture can be grown under sterile and more controlled conditions. And the biggest challenge is the isolation of the desirable cellulose producing strain of bacteria from others that may reduce the quality of cellulose produced or in some cases, bacteria that produce toxins. However, since growing cellulose using SCOBY bacteria requires easily obtainable ingredients and no expensive equipment, it gives an opportunity to individuals to produce substantial amounts of cellulosic material where such may not be easily obtainable.

The COVID-19 pandemic has raised demand for face masks and various materials are used for their production. Pure cellulose can be used for this purpose [44], and the ability to grow cellulose layers at non-industrial/home conditions would provide an accessible alternative to commercial masks. Maximum protection is achieved by adjusting porosity during bacterial incubation and after growth. Another advantage of pure cellulose face masks is that they are often more biodegradable compared to the commercial alternatives.

Other uses of bacterial cellulose include skin transplantation—both in the case of donors and acceptors and was proven to be capable of substituting the dura mater in the dog brain [44]. In fact, bacterial cellulose has been used in a variety of medical applications from hydrogel bandages [45] to drug delivery [46]. Out of all the various sources of cellulose, bacterial cellulose allows for the easiest controllable processing of cellulose and has a high enough degree of crystallinity. Cellulose pore size determines a material's water-holding capacity and oxygen permeability and is important in tissue engineering. Artificial skin made from cellulose allows antibiotics and medication to enter wounds, while preventing outside pathogens from entering the host's body [47].

Conclusions

The study outlines differences between cellulose fibers derived from various sources, their properties, and advantages and disadvantages of each source. Overall, bacterial cultures are believed to be the most optimal one for home growth—all the required materials are widely available, and cultivation conditions can be altered for the intended end-product. After cultivation, porosity can be tuned by mechanical treatments, and the variety of final products allows for vast application: from face masks (smaller pores) to sustainable lithium batteries (larger pores). Further research is needed for improving culturing conditions and mechanical treatments for varying porosity. Also, simpler extraction processes should be explored for other sources of cellulose.

List of Abbreviations
MCC: microcrystalline cellulose
DC: degree of crystallinity
CI: cellulose I
CII: cellulose II
CIII$_I$: cellulose III$_I$
CIII$_{II}$: cellulose III$_{II}$
CIV$_I$: cellulose IV$_I$
CIV$_{II}$: cellulose IV$_{II}$
SCOBY: symbiotic culture of bacteria and yeast
CNFs: cellulose nanofibers

Conflicts of Interest
The author declares that they have no conflict of interests.

Ethics Approval and/or Participant Consent
As a literature review, the study did not require ethics approval or participant consent.

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