

A Comprehensive Review of Nanocellulose Modification and Applications in Papermaking and Packaging: Challenges, Technical Solutions, and Perspectives

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The increasing usage of petroleum-based compounds has prompted numerous environmental concerns. Consequently, there has been a steady rise in research on the synthesis of useful materials from natural sources. Paper technologists are seeking environmentally acceptable dry end and wet end additives. Among the bio-based resources available, nanocellulose is a popular sustainable nanomaterial additive in the paper industry because of its high strength, high oxygen barrier performance, low density, great mechanical properties, and biocompatibility. NC's extensive hydroxyl groups provide a unique possibility to dramatically modify the hydrophilicity and charge of the surface in order to improve their potential applications in the paper industry. The current paper reviews two series of surface modifications, each with various subcategories, depending on why modified nanocellulose is added in the paper production: to improve barrier properties or to improve mechanical properties of packaging materials. The methods presented in this study use the minimum amount of chemically hazardous solvents to have the least impact on the environment. This review focuses on modifications of nanocellulose and their subsequent application in the papermaking. The knowledge and the discussion presented in this review will form a literature source for future use by various stakeholders and the sustainable paper manufacturers.

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INTRODUCTION

Background and Motivation

Packaging is an important component, in the view of customers, to maintain the quality and safety of the goods, provide information about the product, facilitate transportation, reduce food waste during transportation and storage, allow safe storage, prevent product damage, and reduce economic losses. To achieve the above goals, packaging systems must provide a physical barrier to microbial and physicochemical damages, environmental conditions, and external stimuli (Verghese *et al.* 2013; Mohammadian *et al.* 2020). As a result, the packaging industry currently relies upon paper, aluminum, conventional petroleum-based derivatives such as polyethylene, polyvinyl chloride, polypropylene, polystyrene, waxes, and/or fluorine-based derivatives as coating that serve a variety of functions, in order to manufacture competitive packages (Ferrer *et al.* 2017). For instance, paper is a significant packaging material in both developed and developing countries. Paper can take that role because it is a non-toxic, non-polluting, non-radiation-emitting material for versatile use that performs exceptionally well in terms of environmental friendliness (Huang 2017).

Wood fibers in paper packaging create a porous structure that allows oil, water, and gas molecules to pass through, leading to inadequate barrier properties. Additionally, paper packaging readily absorbs moisture from the environment and/or packaged food, resulting in deterioration of its mechanical strength, which is detrimental to the integrity and quality of packaged goods (Zhang *et al.* 2021a). Therefore, various materials can be used to address such issue, including aluminium foil and other metalized films, which are extensively used in food packaging. Aluminum foil provides a more complete barrier against moisture, oxygen, and other gases, volatile aromas, and impact of light than that of any plastic laminate material (Lamberti and Escher 2007). However, as reported by Wang *et al.* (2018), the packaging systems that contain an aluminum foil layer are difficult to recycle, and therefore they have a significant environmental footprint (Wang *et al.* 2018).

Another way to improve the barrier properties of the paper/paperboard would be the application of synthetic polymers derived from petrochemicals, since some of them have superior mechanical properties, heat stability, and certain barrier properties against a variety of compounds such as oxygen, carbon dioxide, water vapor, and aroma compounds (Cazon and Vázquez 2020). However, plastics production and processing are energy-intensive processes; they result in increased greenhouse gas emissions of enormous magnitude, which contribute to global warming. Moreover, plastic waste has been a pressing issue for many years due to plastics' resistance to degradation (Yogalakshmi and Singh 2020). Indeed, these fossil-based plastics are usually non-biodegradable, non-compostable, and contribute to environmental pollution (Dissanayake *et al.* 2021). Furthermore, when they are burned, fossil carbon dioxide as well as toxic pollutants such as carbon monoxide, chlorine, hydrochloric acid, dioxin, furans, amines, benzene, 1,3-butadiene, and acetaldehyde are released, posing a risk to the environment and human health (Dissanayake *et al.* 2021).

Recently, the consumption of this kind of synthetic polymer has increased massively due to the COVID-19 situation (Gorrasi *et al.* 2020). This pandemic has resulted in an increase in demand for certain paper products, as reflected in Fig. 1 (Liu *et al.* 2020).

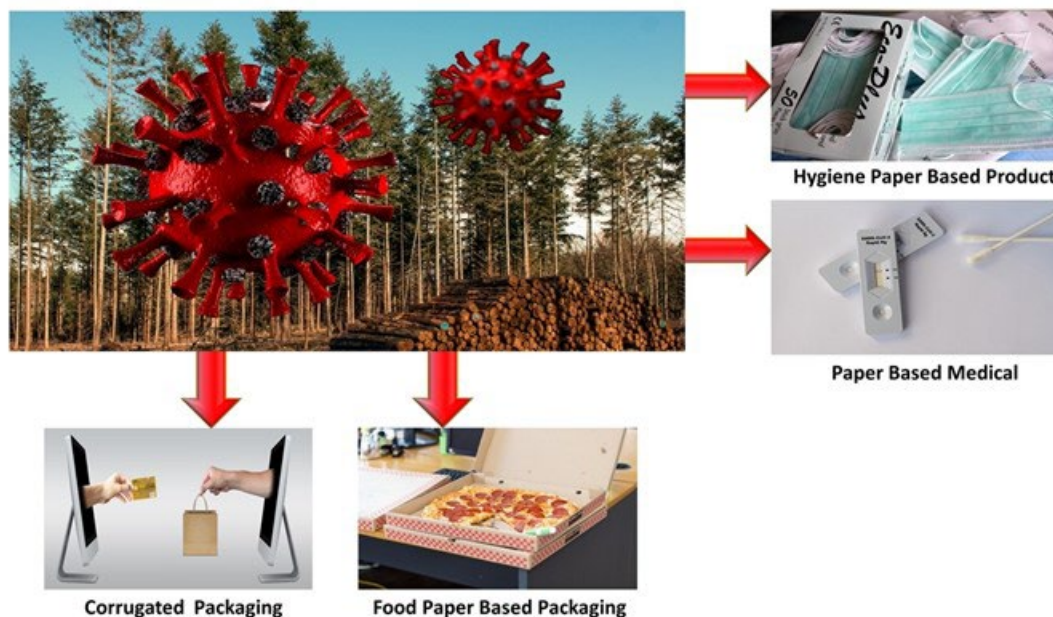


Fig. 1. Potential increasing demand for paper products amid the COVID-19 pandemic, redrawn from (Liu *et al.* 2020)

Unfortunately, the total waste generated has recently accelerated globally due to ineffective waste management and inadequate segregation systems. Only about 173 million tonnes waste is collected for recycling and landfill purposes after consumer use, with the remainder being improperly discarded (Rai *et al.* 2021). In addition, consumer preferences and health requirements are contributing to shifts in preferences toward more natural, high-quality products (Petkoska *et al.* 2021). Renewable carbon resources, such as starch, cellulose, hemicellulose, lignin, and plant oil, are derived from plant and wood biomass *via* photosynthesis from atmospheric carbon dioxide (Iwata 2015). They are being introduced into the market as a true alternative to conventional plastic formulations in packaging, reflecting additional benefits in their commercial use, including biocompatibility, barrier properties, nontoxicity, and non-polluting characteristics (Mellinas *et al.* 2016). Among the carbon-renewable sources, the properties of nanocellulose (NC) are critical to its significance: it is biodegradable and renewable, has high stiffness and reinforcing capacity, and has a large specific surface area (Skočaj 2019). NCs have a high elastic modulus (110 to 220 GPa) and tensile strength (7.5 to 7.7 GPa), as well as a low density and a high concentration of hydroxyl groups on their chains, which expand their application for a variety of desired purposes (Samadani *et al.* 2019).

Nanocellulose (NC) has significant potential to be used in papermaking process as a sustainable packaging material. The concept of sustainable packaging involves analysis and documentation to assess the design, material selection, processing, and lifecycle of the package (Abdul Khalil *et al.* 2016). The Sustainable Packaging Alliance's definition of sustainable packaging identified four principles that must be considered concurrently when evaluating or designing packaging. Packaging must meet the following criteria (Lewis *et al.* 2010): I) be fit for purpose (effective): NC due to the durability and flexibility, can maximize the product-to-packaging ratio and in turn, meet this requirement (Abdul Khalil *et al.* 2016); II) consume as little material, energy, and water as possible (efficient): cellulose-based material is expensive in terms of price, but there is a cost-effective isolation

method, which results in greater availability for developing relatively inexpensive polymer-based products. For instance, manufacturing biodegradable plastics from agricultural waste may pave the way for researchers and scientists seeking cost-effective, environmentally friendly materials (Yaradoddi *et al.* 2020); III) generate as little waste as possible (cyclic); IV) pose the fewest possible health and safety concerns to people and ecosystems (safe): The incorporation of NC into the packaging to substitute fossil fuel-based materials reduces the impact on the environment. In particular, recycled cellulose is utilized to make transparent films for pharmaceutical and food packaging, which reduces the amount of raw materials required during the manufacturing process while also degrading in the environment (Oun *et al.* 2020). Based on this information, cellulose offers promising prospects for use as a sustainable packaging material, but there are some issues that will need further investigation when utilized at higher production levels. For instance, studies have reported a reduction in dewatering rates following the addition of different types of NC to papermaking feed (Rantanen and Maloney 2013). Furthermore, in humid conditions, the barrier performance is significantly reduced because of NC's inherent hygroscopic nature (Hubbe *et al.* 2017; Koppolu *et al.* 2019). Such obstacles imply a strong need for continued research in the coming years to maximize the potential of NC in a wide variety of packaging applications. Currently, the only cellulose-based film-like material that is routinely used in packaging is cellophane. It is used as a packaging film due to its effectiveness as a gas barrier in dry conditions. However, the production of cellophane is environmentally damaging, as it produces harmful by-products and utilizes sulfur-based chemicals (Reshmy *et al.* 2020). To overcome these drawbacks, the NC needs to be somehow modified or combined with biopolymers or other environmentally friendly NC. Depending on the purpose of the addition of NC in papermaking, whether as coating material or wet end additive, it may require a particular type of modification. This review is particularly focused on research publications that shed light on well-known obstacles to successfully implementing modified NC products to enhance packaging performance. The review will build knowledge and discuss the potential for modified NC to be used in papermaking to address the aforementioned concerns regarding the development of gas, oil, and water-resistant coatings and enhancement of the mechanical strength while minimizing the dewatering problems. A better understanding of different NC modification processes and the parameters that must be considered will potentially provide solutions to improve the paper quality, optimize the runnability of the papermaking process, enhance barrier properties, and in turn, increase the use of this technology in packaging. Indeed, the effectiveness of packaging materials is directly related to mechanical and barrier properties. Therefore, whenever in the manuscript we discuss the positive effects of application modified nanocellulose on their mechanical or barrier properties, it means their effectiveness on the packaging material. With this objective in mind, the following outline for this review paper is provided.

Objectives, Scope, and Limitations

As shown in Table 1, numerous review articles and chapters have been published on NC modification and the application of NC in the papermaking processes. The main purpose of this article is to expand the knowledge on the use of modified NC in paper manufacturing, with an emphasis on more environmentally friendly modification methods.

A brief overview of sustainable cellulose-based packaging is outlined in Section 1. Section 2 summarizes NC for packaging applications and provides a comprehensive overview of the NC, types and characteristics, sources of NC, barrier and strength agents. In Section 3, a key overview of the challenges associated with the use of NC in papermaking

is provided, including I) Technical challenges, ii) Processability and runnability challenges including dewatering retention and formation challenges, iii) Economic challenges. Section 4 contains techniques for NC modification that are divided into two parts: hydrophobization and charge functionalization. Section 5 presents modified NC which have been used in papermaking and the possibilities of using modified NC in the papermaking process are highlighted together with the discussions on future directions of research and development in the studied area.

Table 1. Review Papers Focusing on the Modification of NC and Application of NC in Packaging

Review paper	Reference
A comprehensive review on production, surface modification and characterization of nanocellulose derived from biomass and its commercial applications	Kumar <i>et al.</i> 2021
Recent advances in hydrophobic modification of nanocellulose	Sun <i>et al.</i> 2021a
Recent advances in nanocellulose processing, functionalization, and applications: A review	Thakur <i>et al.</i> 2021
Plant-based nanocellulose: A review of routine and recent preparation methods with current progress in its applications as rheology modifier and 3D bioprinting	Yadav <i>et al.</i> 2020
Nanocellulose: From fundamentals to advanced applications	Trache <i>et al.</i> 2020
Nanocellulose: Its applications, consequences, and challenges in papermaking	Das <i>et al.</i> 2020
Nanocellulose production: Exploring the enzymatic route and residues of pulp and paper industry	Michelin <i>et al.</i> 2020
Recent studies on modified cellulose/nanocellulose epoxy composites: A systematic review	Neves <i>et al.</i> 2020
A review of the surface modification of cellulose and nanocellulose using aliphatic and aromatic mono- and di-isocyanates	Abushammala & Mao 2019
Bacterial nanocellulose in papermaking	Skočaj 2019
Chemical modification of nanocellulose in wood adhesive: Review	Vineeth <i>et al.</i> 2019
Industrial application of nanocelluloses in papermaking: A review of challenges, technical solutions, and market perspectives	Balea <i>et al.</i> 2020
Nanocellulose in papermaking	Lengowski <i>et al.</i> 2019
Nanocellulose-based multilayer barrier coatings for gas, oil, and grease resistance	Tyagi <i>et al.</i> 2019
Nanocellulose, a versatile green platform: From biosources to materials and their applications	Thomas <i>et al.</i> 2018
Surface functionalized nanocellulose as a veritable inclusionary material in contemporary bioinspired applications: A review	Chin <i>et al.</i> 2018
Nanocellulose in thin films, coatings, and plies for packaging applications: A review	Hubbe <i>et al.</i> 2017
The potential of nanocellulose in the packaging field: A review	Li <i>et al.</i> 2015

NANOCELLULOSE FOR PACKAGING APPLICATIONS

Nanocellulose

Biofibers are derived from biological sources and can be divided into non-wood fibers and wood fibers (Gholampour and Ozbakkaloglu 2020). Asyraf *et al.* (2020) defined natural fibers by origin (animals, minerals, plants, and microorganisms) and they are classified further in Fig. 2.



Fig. 2. Classification of the origin of different biofibers, redrawn from Campilho (2015)

Certain non-wood fibers outperform wood fibers in terms of physical and mechanical properties, cellulose content, crystallinity, and weight. As a result, they are suitable for engineering applications (Gholampour and Ozbakkaloglu 2020). The fiber's characteristics are affected by its dimensions and chemical composition. They are principally composed of cellulose, hemicellulose, starch, wax, lignin, and other substances (Vijay *et al.* 2019). Among these substances, cellulose is the most critical component discussed in this review due to its high mechanical strength properties, large surface area, intriguing optical and rheological properties, and ease of surface modification. In theory, the tensile strength of cellulose crystalline areas is 50 to 100 GPa, which makes them stronger than E-glass. Additionally, NC exhibits shear-thinning and anomalous behaviour such as time-dependent increase in viscosity, which are advantageous for rheology properties (Jasmania and Thielemans 2018). It's interesting to note that cellulose is the most abundant natural polymer on the globe, since it is created by photosynthesizing green plants, which cover 30% of the planet's land area (Oberlintner *et al.* 2021). Additionally, algae, bacteria, fungi, grasses, and aquatic plants, and animals can be sources of cellulose (Dhali *et al.* 2021). Furthermore, Amara *et al.* (2021) outlined the isolation of cellulose from available agro-industrial by-products and wastes, which offers numerous economic and environmental advantages (Amara *et al.* 2021). One has to keep in mind that the amount of cellulose, its degree of crystallinity, and crystalline size might vary between different sources of cellulose (Dhali *et al.* 2021).

Cellulose fibers can be converted into fibrils with a lower diameter that is eventually made up of organized linear cellulose molecular chains. Due to this hierarchical structure, as shown in Fig. 3, fibrillated cellulose has a great deal of morphological and fibril size variability, resulting in mechanical, optical, thermal, fluidic, and ionic capabilities that can be significantly superior to those of parent cellulose fibers (Li *et al.* 2021d).

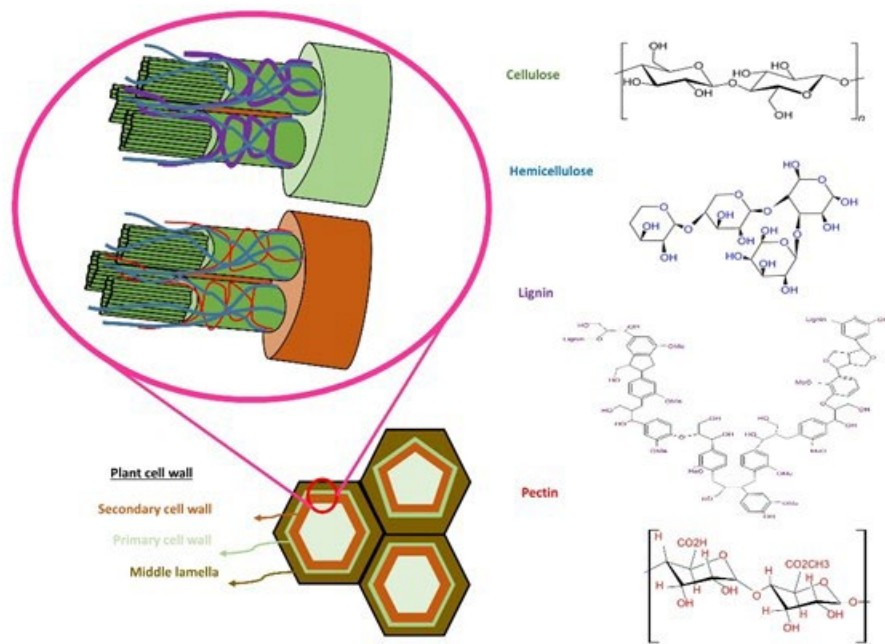


Fig. 3. Schematic illustration of the hierarchical structure of cellulose in plant-based biomass, redrawn from Rytioja *et al.* (2014); Green=Cellulose, blue=hemicellulose, purple=lignin and red=pectin

Very highly fibrillated cellulose is also known as nanofibrillar cellulose, cellulose nanofiber, and cellulose nanofibril (Abdul Khalil *et al.* 2014). Nanocellulose (NC) types can be classified based on three major factors: i) source material for cellulose (*e.g.*, trees, plants, algae, bacteria, tunicate), ii) method of isolation (*i.e.* refinement by mechanical shear or acid hydrolysis which define two classifications of NC, nanofibrillated cellulose, and cellulose nanocrystals), and iii) surface chemistry (*i.e.* surface chemistry might be as a result of a particular nanocellulose production method or the result of subsequent chemical modification) (Li *et al.* 2021c).

Nanocellulose (NC) can be isolated in a variety of ways from a wide range of lignocellulosic sources. Usually, the lignocellulosic source first undergoes some pretreatment in order to achieve superior properties for the final material (Jonoobi *et al.* 2015). Then, a variety of strategies have been used to produce NC, most notably acid hydrolysis and mechanical treatment, either alone or in combination with other methods to achieve the desired nanoparticles characteristics (Abdul Khalil *et al.* 2014). These two approaches result in obtaining particle with large differences in morphology that can be classified into two groups: nanofibrillated cellulose (NFC, also known as cellulose nanofibril) and cellulose nanocrystals (CNC). The major distinction between CNC and NFC would be the number of amorphous regions and their dimensions (Oberlintner *et al.* 2021; Sacui *et al.* 2014). CNC consists of rod-shaped nanoparticles with a diameter of 10 to 20 nm and a length as high as several hundred nanometers (Fig. 4a), depending on the biological source and isolation protocol. CNC is formed by chemically hydrolyzing the amorphous regions of cellulose (Rol *et al.* 2019a), whereas NFC consists of fibril networks with a fibril diameter of 10 to 20 nm. The nano-scale fibrils within typical NFC are long and flexible, as can be seen in Fig. 4b. They are formed through a process called fibrillation, which involves the separation of cellulose fibrils. There are multiple procedures for obtaining NFC, including

high-pressure homogenization, TEMPO-mediated oxidation, enzymatic hydrolysis, mechanical fibrillation/grinding followed by high-intensity ultrasonication, steam explosion, and cryo-crushing (Oberlintner *et al.* 2021). It is worth mentioning here that NFC is highly popular among scientists, with at least 65% of publications on the subject of NC focusing on NFC, according to Rol *et al.* (2019a).

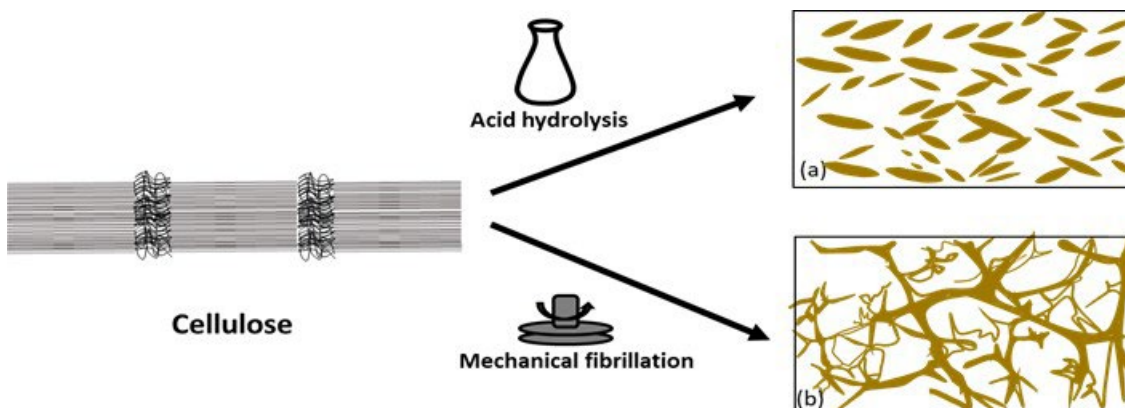


Fig. 4. (a) Distributions of CNC and (b) is NFC, redrawn from Kumar *et al.* (2021)

The two main forms of NC, namely NFC, and CNC, exhibit distinct characteristics, and these have sparked increased interest among researchers. In terms of barrier properties, as shown in Fig. 5, the web structure of NFC has a complicated diffusion path. CNC particles, on the other hand, tend to form a more organized layered structure with fewer voids and a higher density (Wang *et al.* 2020b).

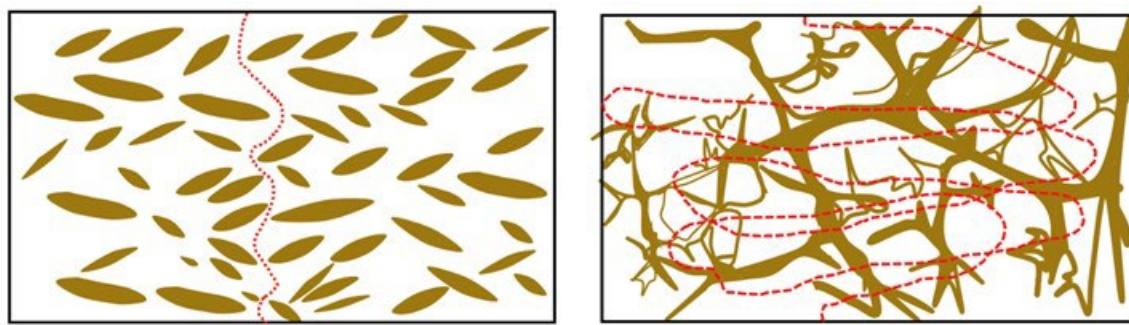


Fig. 5. Scheme of a gas passing through CNC (left) and NFC (right) films *via* cross sections, redrawn from Wang *et al.* (2020b)

The NFC suspensions can display hard solid-like viscoelastic characteristics even at relatively low concentrations, due to the densely entangled network. The viscosity, storage modulus, and loss modulus rise progressively with the increase of NFC concentration. The rheological behaviour of the CNC suspensions has been found to be strongly influenced by the concentration and aspect ratio of CNC. Elastic gel-like behaviors are observed at high concentration levels, whereas viscous liquid-like materials are observed at low concentrations (Li *et al.* 2015c). This has piqued scientists' curiosity in the rheological features of NC. In general, the percolation networks, chemical interactions, and entanglement of NC enable the achievement of highly viscous suspensions in aqueous solutions at sufficient concentrations. On the other hand, continuous shearing aligns NC with the direction of

flow, leading to decrease in viscosity as the shear rate increases. Such reductions are reversible when the systems come to rest. The particular high-low shear flow behavior of the NC make it attractive to be used as rheological modifiers in a wide variety of applications (Li *et al.* 2021c).

Both CNC and NFC, exhibit extraordinary mechanical properties in the dry state as a result of their high crystallinity and intermolecular interactions such as hydrogen bonding between cellulose chains (Saba *et al.* 2017; Hivechi *et al.* 2019). While NC has excellent mechanical properties in the dry state, its weak wet strength is a limiting factor in many applications. This issue is caused by the intrinsic hygroscopic properties of NC because their surface is covered with hydroxyl groups (Seddiqi *et al.* 2021) which also cause adsorption of moist air and liquid water by NC, resulting in considerable swelling of NC (Walther *et al.* 2020).

Nanocellulose in Packaging

As a result of the above-mentioned issues, numerous scholars have demonstrated that NC products can be used in papermaking applications, since they are advantageous in the production of strong paper and barrier-coated packaging paper products (Ehman *et al.* 2020; Hu *et al.* 2021). They also offer significant potential due to their low weight, non-toxicity, low thermal expansion, improved mechanical properties, gas barrier properties, electrical conductivity and environmental friendliness (Kumar *et al.* 2021). The use of NC to enhance barrier properties and mechanical strength of paper for packaging applications is detailed in the following sections.

Barriers (gas barrier and water vapor barrier)

Packaging with high barrier properties extends the shelf-life of foods while providing safety and protection when they are transported and stored. The industry and academia are both aiming to develop high-performance packaging. They are also concerned with having high barrier layers for biodegradable materials, which will require more research. Many strategies have been proposed for improving biodegradable polymers for use in packaging barriers such as different biopolymer blends, Chemical vapor deposition (CVD) of inorganic (like metal or metal oxide), incorporation of nanoparticles crystallization/orientation and chain architecture structures (Wu *et al.* 2021). Due to the high aspect ratio of NFCs, they are well suited for use in the aforementioned ways to enhance gas barrier capabilities (Kwon *et al.* 2020). The relatively high crystallinity of NFC could be part of the reason for these barrier qualities. It could also be due to the strong hydrogen bonds between cellulose and the matrix, as well as the longer path necessary for gas molecules to diffuse (Hubbe *et al.* 2017).

NFC as barriers can be applied to the surface of the paper in a variety of ways, including spray, bar, roll, and size press coating, resulting in a range of coat weights (Brodin *et al.* 2014). Coating paper and paperboard with CNF has no discernible effect on tensile strength but significantly improves bending stiffness. This is because bending stiffness of paper and paperboard is proportional to the third power of their thickness. Covering paper with CNF thickens the papers, resulting in a stiffer product (Mazhari Mousavi *et al.* 2018; Ham *et al.* 2020). Chemin *et al.* (2019) fabricated multi-layered hybrid thin films with more tortuous paths by using CNC and gibbsite nanoplatelets to enhance the oxygen barrier properties (Chemin *et al.* 2019). Mazhari Mousavi *et al.* (2018) also combined CNF and carboxymethyl cellulose (CMC) to coat paper, claiming that there was a significant improvement in the barrier properties of paperboard, which was directly related to the coat

weight and surface coverage (Mazhari Mousavi *et al.* 2018). Recently, nanofiller-NC composites have developed as a novel form of composite material with strong strength and gas barrier properties (Mirmehdi *et al.* 2018). Seyedmohammad Mirmehdi *et al.* (2018) proposed a coating layer of a hybrid composite of NFC (matrix) and nano-clay (mineral filler) on paper substrate. Indeed, the use of nano-clay alone lowered tensile strength as well as the rate of oxygen transmission rate (Cairns *et al.* 2019). The application of NFC, on the other hand, increased the thickness of the films, affecting their strength and barrier properties. As a result, the combination of NFC (matrix) and nano-clay can result in interesting strength and gas barrier qualities (Mirmehdi *et al.* 2018). NC can also be applied as a filler (NC-filler), contributing to improvement of the gas barrier properties of packaging material since (i) NC-filler material may act as physical obstacles and makes for a more tortuous journey (ii) the addition of NC-fillers also decreases the polymer's free volume (Rigotti *et al.* 2020). Xu *et al.* (2019a) reported the great capability of NFC and nanofillers chitin whisker (CHW) as nanofillers for improving the mechanical and barrier properties of polybutylene succinate-based films for biodegradable food packaging (Xu *et al.* 2019a). Dhar *et al.* (2015) also demonstrated the production of nano-biocomposite films based on poly(3-hydroxybutyrate)/CNC with superior gas barrier properties for food packaging applications. However, there is an issue with obtaining homogeneous NC-filler dispersion and strong NC-filler-polymer interactions (Wu *et al.* 2021). It should be added that these bio-barriers exhibit comparable properties to those of engineering polymers, with the exception of the water vapor barrier (Chowdhury *et al.* 2019). Indeed, the water vapor permeability of unmodified NC does not follow the same trends due to the hydrophilicity of cellulose (Kwon *et al.* 2020). In order to improve the water vapor barrier property of NFC-based substances, inorganic fillers or cross-linking agents, as well as chemical modification of NFC has been suggested by Yook *et al.* (2020).

Strength agents

Paper's strength is affected by three aspects: (a) fiber strength, (b) fiber bonding capability, and (c) fiber entanglement properties. The usage of strength agents mostly reflects an enhancement in fiber bonding strength due to greater hydrogen bond formation (Li *et al.* 2021a). The NCs as strength agents have the potential to enhance the mechanical strength, density, barrier properties/air resistance, filler retention, and flocculation (He *et al.* 2016). It has been demonstrated that addition modified cellulose nanofibrils to the fiber matrix improves filler retention, lead to significantly enhanced dry and wet strength of the paper (Lourenço *et al.* 2019).

The mechanical strength and modulus of NFC is particularly impressive, and the tensile strength of single fibers is nearly twice that of some metals (such as Carbon Steels ASTM A36, ASTM A529, ASTM 709 due to these 3 nm-wide fibrils (Zhao *et al.* 2018). Furthermore, NCs benefit from Young's modulus values between 20 and 50 GPa, which enables it to be used in a wide variety of fields such as nanopapers, NC films, aerogels, and hydrogels (Usov *et al.* 2015; Thomas *et al.* 2018). In papermaking, NFC could be applied in the wet section of the papermaking process to increase the strength of paper. The strength improvement is partially accomplished by filling in voids and pores at the edges of each fiber bond, thereby increasing the available bonded area (Brodin *et al.* 2014). He *et al.* (2017) demonstrated enhancement of strength properties by filling inter-fiber spaces of the sheet with higher surface area particles, and then the contact area at the fiber to fiber joint edge is increased, allowing for more hydrogen bonding sites (He *et al.* 2017). Additionally, NFC can enhance the paper's strength capabilities by establishing strong fiber-to-fiber

bonds as a result of their nanostructure and huge surface area, which leads to more hydrogen bonds formed in a strong network (Das *et al.* 2020). Lv *et al.* (2017) investigated the effect of using the MFC-ground calcium carbonate (GCC) complex on the strength and optical properties of handsheets. They used an MFC dosage of 5%, based on the GCC weight. The results indicated that the modified GCC acted like fibers and had improved retention due to the effective adsorption between GCC and MFC. Better paper strength qualities were attained at the same filler quantity (Lv *et al.* 2017). Su *et al.* (2020) also premixed MFC with GCC and discovered that the MFC-modified GCC increased the opacity of the filled paper while also improving its tensile strength. It is vital to note that high NC addition does not always result in favorable reinforcement effects for paper (Yuan *et al.* 2016) and optimization is essential in determining the ideal amount of additive.

The second parameter to optimize is the degree of fibrillation, which has a significant effect on the mechanical properties of NCs and, more specifically, on the paper sheet. This means that MFC with a broader particle size distribution shows less improvement in mechanical properties but more efficient retention in the fiber web. Even before sheet formation, NFC with a high degree of fibrillation can be distributed more effortlessly in the bulk suspension. As a result, a more homogeneous distribution occurs, which improves the nanostructure's strength (González *et al.* 2014). However, a high degree of fibrillation does not always result in a beneficial reinforced effect, owing to their shorter length and lower aspect ratio. Indeed, short fibrils are more difficult to retain on fibers, so they contribute less to the strengthening of interfibrillar bonding (Hu *et al.* 2021).

Challenges of Nanocellulose in Papermaking

Runnability challenges

In the paper manufacturing process, runnability refers to a machine's ability to produce high-quality paper at maximum capacity. Therefore, the maintenance of “good” operation plays a vital role in paper production (Bondoc 2018). Some researchers have investigated the influence of NC application on the runnability of the papermaking process (Kajanto and Kosonen 2012; Lu *et al.* 2019; Charani and Moradian 2019). Kajanto and Kosonen (2012) used a high-speed pilot paper machine to test the use of NFC as a reinforcement agent in the paper. During the trial, overall runnability was satisfactory. No web cracks are found related to the issue with paper strength or dewatering. The total retention level stayed steady and the formation remained satisfactory without the need for any further changes. Dewatering in the press and former sections was acceptable (Kajanto and Kosonen 2012). Lu *et al.* (2019) investigated the properties of base paper's wet-web strength and pressability, as well as their effect on the frequency of sheet breaks and machine runnability. They reported that after applying 5% NFC to the sample, the tensile energy absorption increased from 6.32 to 10.93 J/m². As a result, they stated that adding CNFs to the paper-making process increases wet-web strength and improves paper machine runnability (Lu *et al.* 2019).

Charani *et al.* (2019) found that combining less cationic starch (CS) (0.5%) with 3 percent NFC greatly boosted paper breaking length while lowering CS-related process challenges (Charani and Moradian 2019). Another study found that adding 3, 6, and 9 wt% NFC to handsheets increased linearly the tensile index to around 55 N m gL at 35SR, which was judged to be suitable for paper machine runnability (Vallejos *et al.* 2016). Ankerfors *et al.* (2014) used the FEX pilot paper machine to examine the impact of MFC on the runnability and paper strength of a fine-paper. During the pilot paper machine trial, no serious runnability difficulties were reported. They also demonstrated that dewatering was

affected by the MFC (NC), particularly in the wire portion (Ankerfors *et al.* 2014). As a result of the decreased drainage rate, the machine's speed must be decreased as well, and there would need to be strategies in place to compensate for the decreased drainage rate caused by the NC application. This means that, for example, by using retention and drainage aids, paper machine running speeds can be greatly boosted; also, the retention of fine and NC particles increases and contributes to the better runnability of paper machines (Jin *et al.* 2014).

Many studies have looked at the effects of NC as a coating of paper (Aulin and Ström 2013; Chowdhury *et al.* 2019; Lengowski *et al.* 2019; He *et al.* 2021). Regarding their runnability as the coating can be mentioned that resistance to dewatering into the substrate is a critical process parameter. Indeed, both wet film formation and process runnability are influenced by water retention or the rate at which water is discharged into the substrate. A rapid release of significant amounts of water from MFC suspensions into paper can induce fiber swelling and de-bonding, resulting in runnability issues such as web breaks (Lengowski *et al.* 2019). Thereby, understanding and controlling the water retention characteristics of NC suspensions is critical for a successful coating process (Kumar 2018).

Additionally, high water content in NFC coatings results in greater drying costs, which are undesirable except if outweighed by enhanced product properties (Brodin *et al.* 2014). Drying also contributes to increasing the concentration of NC and then would affect drying dynamics and assembly operations (Wang *et al.* 2019).

The next challenge with runnability is that NC suspensions are difficult to coat onto substrates using conventional coating and metering mechanisms. This is due to blockage caused by NC fiber aggregation or the NC suspensions' extraordinarily high viscosity (Kumar 2018). NFC suspensions are well-known for their high viscosity, even when the solids content is low, contributing to the limitation of the size of the operation window (Brodin *et al.* 2014). It should be added that improvement and controlling the rheological features of NFC-containing coating formulations is a significant challenge for lowering water content while maintaining a uniform coating layer (Brodin *et al.* 2014). As a result of changes in the rheology, papermakers must vary the nip load, doctor blade angle, or pressure when introducing NFC into the composition (Sharma *et al.* 2020).

Processability Challenges

Dewatering

Nanocellulose (NC) has not yet been widely applied in the papermaking industry, and this has been attributed to difficulties in using it (Balea *et al.* 2020). Nanofibrillated cellulose (NFC) usually increases the time needed for drainage dewatering, and a short draining time is needed in the papermaking process to minimize production costs and energy (Hu *et al.* 2021). Numerous causes to the delayed dewatering efficiency can be summarized as:

- Dense layers are created,
- Cellulose particles clog drainage channels in the wet web,
- The reaction to vacuum is inefficient because of flocculation,
- The healing mechanism allows for the repair of thin areas of paper,
- A thin membrane forms on the surface of the paper when it comes into contact with a wet-press felt, due to rewetting of the sheet (Hubbe *et al.* 2020).

Besides, NFC creates a highly entangled network in aqueous dispersion, exhibiting a gel-like network at 1% solid levels. NFC surface charge appears to influence the rheological and dewatering properties of furnishes through its influence on swelling and effective binding of water. In fact, swelling of the fiber walls results in fewer fiber contacts, which reduces floc size and therefore lowers frictional forces between fibers, affecting rheology and dewatering of the fiber suspension network under flow (Dimic-Misic *et al.* 2013). Additionally, dewatering is closely linked to the shear-thinning behaviour of furnish. When the shear field disrupts the suspension's microstructure, then the process can spread across the pad's flow channels. As a result, the application of repetitive shear cycles can help the dewatering process (Dimic-Misic *et al.* 2013). According to some researchers, the presence of cationic NFC (CNFC) can also aid dewatering while improving the wet-web strength of paper sheets (Lu *et al.* 2020b). Diab *et al.* (2015) reported that cationic microfibrillated cellulose (CMFC) was unable to increase the pulp's drainage. The author recommends that more trials using CMFC with a higher degree of cationization should be conducted to confirm that CMFC cannot be employed as drainage agents (Diab *et al.* 2015).

Lu *et al.* (2020b) assumed that the increased fines retention and density of paper sheets facilitated by CNFC addition are due to the factors including the following:

- (1) The CNFC nanoscale dimensions provide a large specific surface area, which helps in adsorbing and "catching" the fines to create the CNFC-fines complex in the pulp;
- (2) The high aspect ratio of CNFC would make it easier to twine and "lock" the fines together in order to create the CNFC-fines complex.
- (3) The high surface charge density of CNFC's quaternary ammonium ions enhances their integration with fines to create the CNFC-fines complex (Lu *et al.* 2020b).

Aside from the surface charge, other factors influencing the dewatering rate of NC used in paper production include the location of the NC addition and the type of retention additive used in paper manufacturing. Indeed, some researchers believe that by adding relatively small amounts of NFC directly to the pulp with appropriate retention aids, dewatering issues could be avoided (Li *et al.* 2021a). Ottesen *et al.* (2016) premixed NFC and retention chemicals with filler and predicted that the retention chemicals would enable NFC or fillers to adsorb preferentially to accessible surfaces. They reported large reductions in dewatering times through the use of retention chemicals that bind the NFC to the various components of the paper furnish. These benefits can be amplified even more for higher NFC doses by placing NFC in the filler fraction before adding it to the furnish (Ottesen *et al.* 2016). In general, the complicated interactions between cellulose pulp, NFC, mineral fillers, and regularly used additives should be taken into account to address this issue (Li *et al.* 2021a).

Retention

While a high aspect ratio of NC is critical for producing high-quality nanopapers, it is also one of the primary causes of agglomeration and flocculation problems. From the standpoint of colloid science, the most basic solution is to increase the electrostatic charge on the nanofibrils' surface, which will cause like charges to repel each other, preventing aggregation and flocculation (Li *et al.* 2015c). However, from the perspective of particle retention, hydrodynamic shear and turbulence can easily wash the comparatively small

negative charge particles off the web and reduce retention in the forming phase. The increase in retention appears to be due to the retention agent's charge neutralization and aggregation properties (Ahadian *et al.* 2021). Therefore, there would be a trade-off between retention and flocculation in this case that must be considered.

In terms of retention, polyelectrolytes are often used by researchers to improve the retention efficiency of fillers, fines, and small particles and to reduce dewatering times in the papermaking process. These polyelectrolytes have the ability to bind fines so that they remain in wet sheets. This happens due to a flocculation effect, such that the fines are electrostatically connected to fibers. These kinds of polyelectrolytes enhance the drainage rate and retention of small particles while influencing the reinforced effect of NFC (Hu *et al.* 2021). The presence of retention aids was found to affect the position and association of fibrils during adsorption, and hence it affected the final handsheet characteristics. For instance, the densities of sheets formed in the presence of polyvinylamine as retention aids are lower than those formed in the absence of a retention aid. This could be explained by the fact that when retention agents are present, fibrils are more likely to attach to the fibers surface before dewatering rather than filling the pores between the fibers during dewatering (Hollertz *et al.* 2017). NFC-retention additive interactions and appropriate dosage of retention additive must be studied in order to select the most beneficial combination (Merayo *et al.* 2017b). Taipale *et al.* (2010) demonstrated that by selecting the right materials and operating conditions, the strength qualities could be improved without compromising the drainage. First, a thin MFC (NC) layer can develop on the adsorbed CS, and this nanonet-work can then coat the fibers instead of filling the gaps between them (Taipale *et al.* 2010). Merayo *et al.* (2017) also believed that employing the proper retention additive at the right dose could help to alleviate the dewatering issue while using NFC in paper production. So, they investigated the idea of interaction between MFC (NC) and retention agents (polyvinylamine, chitosan (CH), cationic starch (CS), and two type cationic polyacrylamide (C-PAM additive formed by poly-quaternary ammonium chloride and C-PAM-B additive formed by polyamine, PAM and hydrated bentonite clay) in order to increase the strength of recycled paper while minimizing adverse effects on the drainage process. When comparing the behavior of each additive at the lowest doses tested, C-PAM and C-PAM-B came out on top, followed by CH. Polyvinylamine also showed satisfactory drainage time results at the lowest dose; however, such dose was ten times higher than C-PAM, C-PAM-B, and CH. The CS was the least effective additive tested, especially at low and moderate doses (Merayo *et al.* 2017b). This result also implies that the conformation of fibrillated material onto cellulose fibers is influenced by retention aids; the final configuration indicates the extent to which the degree of strength and drainage rate have been increased (Zambrano *et al.* 2020). Finally, a single retention additive may not be satisfactory, and the development of two-component reagents has been recommended as a way to improve performances (Lourenço *et al.* 2019). In the case of NFC as retention agents, it can be mentioned that NFC I with a high specific surface area has the ability to flocculate cationic particles mostly *via* bridging/networking. Following the application of high shear and flocs breaking, subsequent retention agents on the flocs may help to approach flocs, resulting in reflocculation *via* patch or networking mechanisms. However, chemical modification of NFC is required to enhance its electrostatic interactions with other particles in the system. (Korhonen and Laine 2014).

Formation

The term “formation” (more formally “formation uniformity”) refers to the spatial variation in the density of fibers in the paper (Na and Muhlstein 2019). It can be determined by examining the sheet's mass distribution in its plane. The goal of the fiber network formation process is to distribute the pulp as evenly as possible on the wire to ensure that the product properties remain consistent across the width of the web (Norman 2000). As shown in Fig. 6, flocculation and dispersion are two competing effects in the papermaking process.

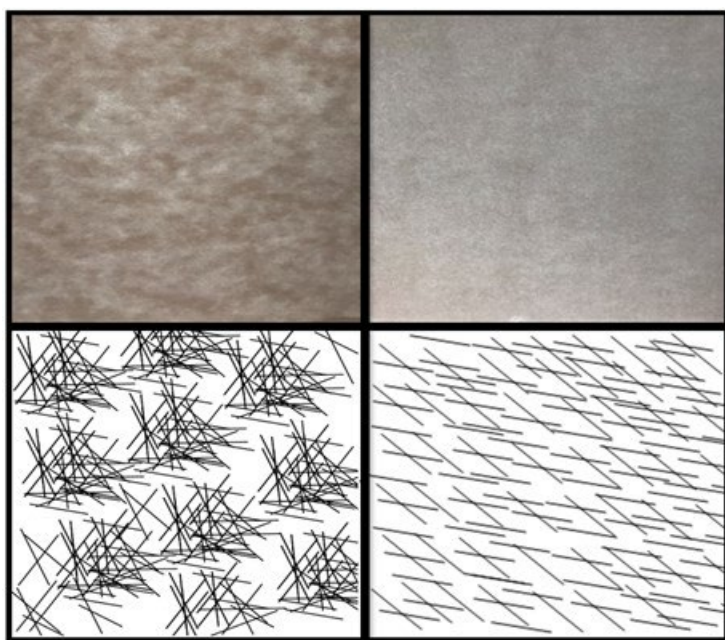


Fig. 6. Right: Random Fiber distribution; Left: Fiber Flocculation distribution, redrawn from Dodson and Serafino (1993)

The importance of paper's formation uniformity in terms of sheet appearance is widely acknowledged among papermakers, as the strength is controlled by fiber-to-fiber interactions that are developed during the process of paper formation, consolidation, and drying (Su *et al.* 2012). It has long been recognized that paper's physical properties are somewhat dependent on its formation, with the effect being significant for tensile strength but small for elastic modulus (Anson *et al.* 2007). As reported by Motamedian *et al.* (2019), the paper formation is frequently improved by shortening fibers; this explains why the refining process might have an effect on the formation (Motamedian *et al.* 2019). From another perspective, increased fiber refining increases bonding formation and can somewhat offset the negative effect of filler on strength qualities (Ankerfors *et al.* 2014). Nanofibrillated cellulose (NFC) can bridge the filler-induced voids in the fiber-to-fiber bonding area, similar to fines, and affect the fiber network (Ämmälä *et al.* 2013). Two alternative processes can be used to explain the enhancing effect of NC on base paper: (1) NC promotes fiber–fiber bonding by bridging fibers together, and (2) NCs can create network architectures between relatively coarse fibers, enhancing the load-bearing capacity of papers (Li *et al.* 2021a). As NCs are derived from natural fibers, possessing inherent characteristics of pulp fibers, the slender filaments can form a densely entangled network structure (Li *et al.* 2021a). Merayo *et al.* (2017a) also confirm that the sheet homogeneity (formation index) improves due to a reduction in floc size by using NFC.

However, their small size makes it challenging to retain NFC during sheet formation in order to make use of their bond-forming capacity. As mentioned above, various solutions have been proposed to maintain the NC in the sheet formation process. For example, the use of a retention aid not only assists in the retention of the NC in the furnish, but it also can contribute to the formation of a homogeneous formation (Diab *et al.* 2015). Ahola *et al.* (2008) combined paper NFC with a cationic poly(amideamine epichlorohydrin) to improve the wet and dry strength of paper and they showed that when PAE first adsorbed on the fiber surface, it generated a homogeneous and viscous coating of NFC. Salminen (2010) studied the effect of (C-PAM on the short fiber fraction and CS on the long fiber fraction on the formation, retention, and strength of the paper. They claimed that adding C-PAM to short fiber fractions would prevent long fibers flocculation, resulting in the improved formation and, in turn, increased wet web strength.

Economic Challenges

Nanocellulose (NC) applications have a 6.4 million metric ton yearly market potential in the United States, and a 35 million metric ton global market potential. Currently, cellulose nanoparticles have the largest volume potential in paper and packaging applications. The automobile, construction, personal care, and textile sectors are other examples of high-volume applications (Shatkin *et al.* 2014). According to Nelson *et al.* (2016), the United States Department of Agriculture's global NC market size projection will be reached around 2045. Commercialization of technologies for producing NC from kraft or dissolving pulp has occurred recently. Pulp is a costly raw material that necessitates rigorous, often energy-intensive chemical or mechanical processing (Blair and Mabee 2021). Additionally, chemical costs and investment in manufacturing equipment and paper machines, are other factors that influence the cost of NC manufacture (Li *et al.* 2021a). Due to the confidentiality issue, it is hard to provide a precise estimate of the end-use product's cost. A rough calculation based on the raw material pricing and manufacturing costs revealed that the cost of NFC will range between USD\$ 7 and 12 per kilogram of dry material (Dhali *et al.* 2021). Estimated CNC production costs differ widely across the literature; de Assis *et al.* (2017) approximated that manufacturing costs for CNC production ranged from 3632 to 4420 (dry equivalent) USD t^{-1} . Their model process was acid hydrolysis of dissolving pulp (de Assis *et al.* 2017). There are currently a few pilot projects producing these nanomaterials, such as CelluForce (Canada), USDA's Forest Products Laboratory, InnoTech Alberta (Canada), FPInnovations (Canada), and Cellulose Lab (Canada) (Rudie 2017). Numerous difficulties and obstacles remain along the path from pilot to large-scale production. For instance, CNC that is produced *via* acid hydrolysis presents challenges in terms of waste treatment, yield, drying, and redispersion. Numerous efforts have been made to overcome these types of problems such as the fabrication of NC using solid organic acids hydrolysis (Chen *et al.* 2016; Jia *et al.* 2017). The recovery of and the reuse of these kinds of acids is easily achievable through a conventional and commercially proven crystallization process at a low or ambient temperature. In fact, since they can be recovered using a conventional and commercially proven crystallization method, organic acids are uniquely excellent candidates for making sustainable and green cellulose nanomaterials (Chen *et al.* 2016; Jia *et al.* 2017).

In the case of NFC, the economic problem is related to the usage of mechanical methods, which involve large amounts of energy consumption (Wang and Cha 2019). Eriksen *et al.* (2008) calculated that a production NC by homogenizer can consume up to 70,000 kWh/t of energy, which is nearly 100 times more than the energy required to make

one ton of paper (Eriksen *et al.* 2008; Chauve and Bras 2014). Therefore, various pre-treatment methods have been developed to significantly reduce the energy consumption associated with NC production. Even with pre-treatments, matching the cost of NC production to specific benefits such as mechanical strength performance remains a considerable difficulty (Ang *et al.* 2019). The economy of producing NFC materials depends largely on the pre-treatment method (*e.g.* enzymatic, carboxymethylation, TEMPO-modified NFC, *etc.*). The least costly process is likely to be the enzymatic pre-treatment process, which produces NFC from pulp integrated with a pulp mill at a cost of 0.4 €/kg and is currently used in large-scale paper manufacturing applications. The cost of non-integrated NFC in papermaking applications should be less than 2.5€/kg (Klemm *et al.* 2018). Indeed, integrating the production of fibrillated cellulose products with the current forest and paper industries appears to be a synergistic technique for cutting large-scale production costs (Li *et al.* 2021d). With the advent of low-cost commercial NC sources, there is still space for new applications and improvements to existing ones in a variety of industries that require sophisticated materials (Trache *et al.* 2020). In general, the economic challenges that remain in the field of NC manufacturing that need to be overcome in order to ensure rapid utilization and commercialization include the following: 1) Cost-effectively drying of NC suspensions while preserving distinct NC particle shape for optimal re-dispersion during end-use 2) costs related to the creation of international standards across the supply chain, and 3) development of low-cost, quick characterization methods for detecting product quality (Nelson *et al.* 2016). Additionally, in the paper industry, unmodified cellulose has few applications in packaging films. When cellulose is modified chemically, mechanically, or enzymatically, it can be used in a variety of applications (Fotie *et al.* 2020). Therefore, in addition to the preparation of NC, the cost of treatment should be considered, which is currently an expensive process (Nelson *et al.* 2016; Blair and Mabee 2021).

There is an effort underway to develop processes that will enable NC to be sold at a lower cost, for instance with increased scale production costs could be significantly reduced. Furthermore, capital costs could be reduced through government investment or infrastructure repurposing. Increased process efficiency or internal energy generation could also help to reduce energy costs (Nelson *et al.* 2016). Along with the successful extraction of NC *via* pre-treatments (Ang *et al.* 2019), some practices such as developing new, environmentally friendly, and inexpensive solvents as pre-treating agents are generating increased interest in academia and industry (Liu *et al.* 2019b).

Following the decrease in the cost of production, additional research must be conducted to ensure the final materials' feasibility and marketability, in particular (1) life-cycle evaluations and degradability studies, as required to demonstrate the environmental impact; (2) continuous cost-cutting optimization of the fibrillated cellulose production process; (3) a careful balance between usability and biodegradability when discarded; and (4) a recycling technique for composites incorporating fibrillated cellulose (Li *et al.* 2021d).

MODIFICATION TECHNIQUES OF NANOCELLULOSE

Modifying NC can help address some of the above limitations, since the hydroxyl groups in the NC chain are critical in enabling NC to be used in a variety of applications. They can be adjusted in a variety of ways, depending on the application, and NC can be

impacted based on the chemical and condition utilized (Rol *et al.* 2019a). These modifications result in the acquisition of desirable properties, which improve their effectiveness for a particular application (Trache *et al.* 2020).

In this review paper, modification techniques are further classified into two types based on the reason for adding modified NC to the paper, as shown in Fig. 7. The first category is associated with the goal of improving the barrier properties of paper, with a particular emphasis on NC hydrophobization and its usage as a dry end additive. The next category is involved with the modification to impart stable positive or negative electrostatic charges on the surface to improve retention and mechanical properties. The factor that could determine the effectiveness of modification is the degree of substitution (DS), with the maximum value being 3, which occurs when all hydroxyl groups on anhydroglucose units react (Oberlintner *et al.* 2021).

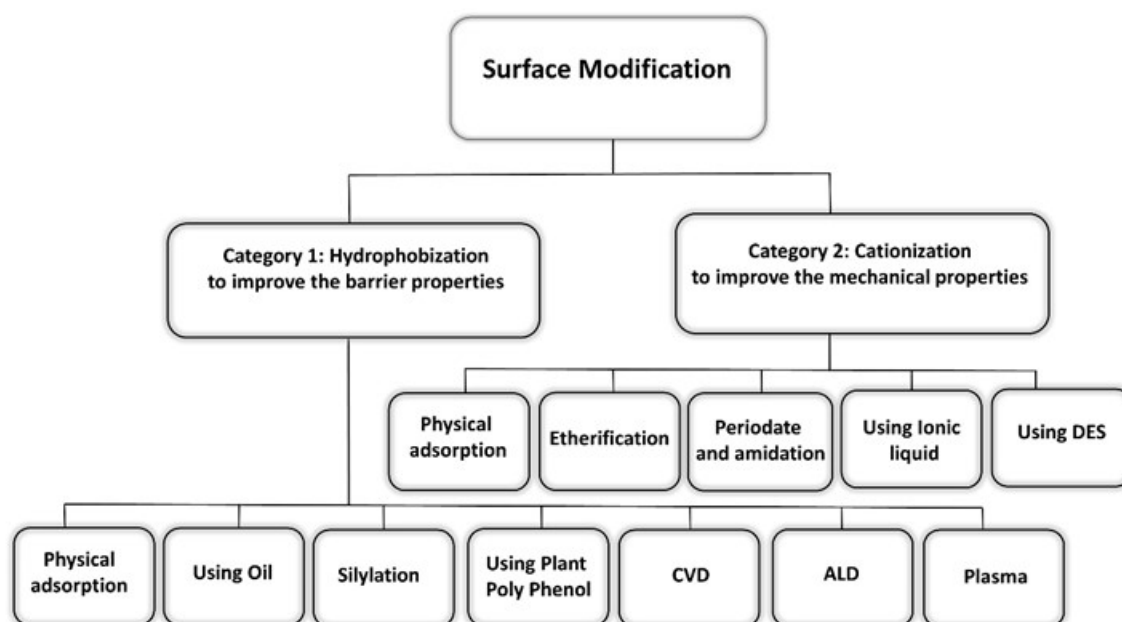


Fig. 7. Various modification methods that will be discussed in this review article

Hydrophobization of Nanocellulose

The presence of a high hydroxyl group concentration on the surface of NC confers on them inherent hydrophilicity. Self-agglomeration and hydrophilicity have limited their applicability, prompting development of surface functionalization and hydrophobization in order to effectively utilize their potential (Chin *et al.* 2018). In general, functionalization with chemical grafting is preferred over physical grafting because the covalent bonding of grafted chains to the substrate surface prevents desorption and promotes longer chemical stability (Sun *et al.* 2020). However, the need for solvent-based or toxic-based systems have limited their application to industrial scale-up. As a result, several European projects have been started, such as the SUNPAP, to discover alternatives to these solvents (Missoum *et al.* 2013). As a consequence, researchers are increasingly concentrating their efforts on developing more environmentally friendly strategies for chemical surface modification, which can be unquestionably the most important aspect of NFC functionalization (Missoum *et al.* 2013). With this in mind, in the following text summarizes some of the

hydrophobization methods (not totally green but a bit more “environment-friendly” methods) that use the fewest amount of solvent and toxic materials possible.

Hydrophobization: Physical adsorption of surfactants

Surfactant adsorption is a green, industrially scalable, and environmentally friendly method for engineering the NC surface (Chi and Catchmark 2017; Szlek *et al.* 2022). In fact, the amphiphilic nature of surfactants is ideal for modifying the particles' surface tension and contact angle. Surfactants can reduce the interfacial tension between air and water, as well as the forces that keep water in the filter cake capillaries (Patra *et al.* 2016). When they are adsorbed onto the less charged regions of NC surfaces, the net Gibbs free energy is decreased, as shown in Fig. 8 (Tardy *et al.* 2017).

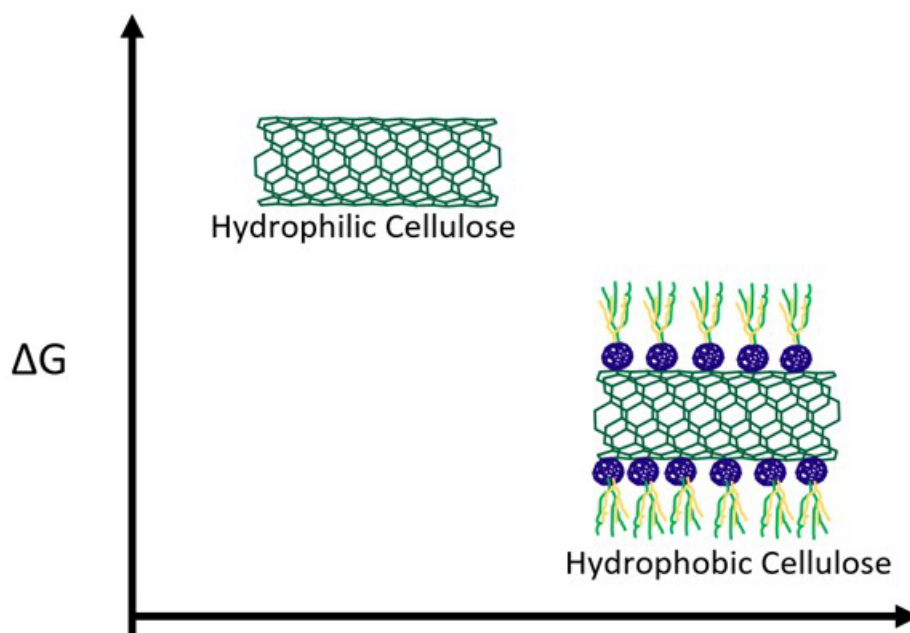


Fig. 8. Schematic illustration effect of adsorption surfactant on the Gibbs energy. Note that the details of the “hydrophilic cellulose” are not intended to represent a specific chemical structure or tubular nature.

Surfactants are made up of two parts: a hydrophilic head and a hydrophobic tail (Vaziri Hassas *et al.* 2014). According to the charge of their hydrophilic moiety, there are four types of surfactants: anionic, cationic, non-ionic, and amphoteric (Kurpiers *et al.* 2021). Two key aspects influence surfactant adsorption: the surfactant's interaction with an electrostatic force and the surfactant's hydrophobicity, which will cause it to interact with the surface if it is less polar than water (Aulin *et al.* 2008).

An anionic surfactant's affinity for NC surfaces can be explained by hydrophobic interactions between the surfactant's hydrophobic groups and the intrinsic hydrophobic domains found on cellulosic materials (Tardy *et al.* 2017). Cationic surfactants are predicted to have a high affinity for NC surfaces due to the different charges. These electrostatic interactions are enhanced by the presence of functional groups on NC. The reason for this is that electrostatic interactions are an order of magnitude more powerful than secondary interactions (Tardy *et al.* 2017). Ly *et al.* (2020) demonstrated that the nature of the

surfactants used in the CNC modification can be attributed to the increasing of water contact angle (WCA) in the modified samples. All three surfactants considered (cetyltetramethylammoniumbromide with C16 single chain, dimethyldidodecylammonium bromide with C12 double chains, and dimethyldihexadecylammonium with C12 double chains), have the ability to arrange their hydrophobic tails around CNC surfaces, resulting in hydrophobicity, although the longer the surfactant's aliphatic chain, the greater the hydrophobicity of the CNC (Ly and Mekonnen 2020). It should be added the modification of cellulose is dependent not only on the type of surfactant, but also on the amount of adsorption (Patra *et al.* 2016). For instance, de Lima *et al.* (2019) found that the surfactant had no effect on free hydroxyls. They reported that (1) the CNC has a strong physical interaction and steric stabilization, which can be seen as an increase in particle size, and (2) the surfactant does not serve as a stabilizing agent, resulting in no charge differences (de Lima *et al.* 2019).

It is important to notice that the use of surfactant chains makes the crystalline ordering decrease, and the spacing between the planes consequently will increase. Souza *et al.* (2020) modified NC during isolation with an anionic surfactant. According to their study, this *in situ* modification led to a decrease in the NC dimensions because of the electrostatic interactions with the surfactant which may improve the grinding process, making it more productive (Souza *et al.* 2020).

Hydrophobization: using oil

Vegetable oils can be used to modify the surface of cellulose nanomaterials through an esterification process or by grafting vegetable oil, as shown in Fig. 9. The majority of vegetable oils are triglycerides, which are made up of glycerol molecules and three long-chain fatty acids joined by ester bonds at the hydroxyl groups. The long-chain fatty acids are more hydrophobic than shorter fatty acids and thus more stable at high temperatures (Le and Nguyen 2020; Hashemzahi *et al.* 2022).

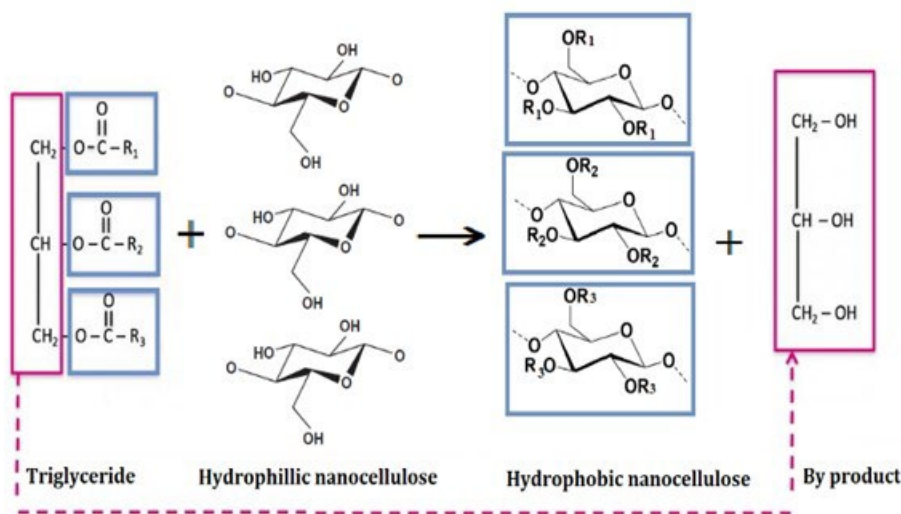


Fig. 9. Transesterification reaction between cellulose and triglycerides

Shang *et al.* (2013) modified CNC by using castor oil and diisocyanate as a linking agent. They believed that two of the three hydroxyl groups in castor oil should be terminated to leave only one active hydroxyl group available for covalent grafting onto CNC. Following a successful graft, the surface energy decreased and the contact angle increased significantly (Shang *et al.* 2013). Pourmoazzen *et al.* (2020) anchored successfully covalently cholesterol to CNC surfaces. It was clear that hydrophobic CNC had been achieved, because the contact angle was increased to above 115 (Pourmoazzen *et al.* 2020).

Transesterification results in a little increase in the crystalline index, which has been related to hydrolysis or crystallization of the amorphous portion of NC under acidic conditions and high temperatures during esterification reaction (Yoo and Youngblood 2016). Wei *et al.* (2017) used a transesterification reaction to study the chemical modification of CNC using canola oil fatty acid methyl ester. As compared to unmodified CNC, the transesterified CNC had greater thermal stability and hydrophobicity. Besides, glycerol, a by-product of the canola oil fatty acid methyl ester manufacturing process, can be used as emulsions and polymer processing aids (Wei *et al.* 2017). Gorade *et al.* (2019) developed the hydrophobic modification of microcrystalline cellulose (MCC) using rice bran oil (RBO). The WCA of AUMCC was 92.2°, with the surface energy dropping to 35.56 mN/m and water absorption of 0.9 mL/mg. AUMCC's thermal stability and crystallinity deteriorated as treatment time progressed (Gorade *et al.* 2019). Dhuiege *et al.* (2018) present a new technique for surface functionalization of CNC in water, by transesterification of vinyl acetate. They believed that developing a water-based technique would considerably reduce the treatment's environmental impact. As reported by Dhuiege *et al.* (2018), the increasing hydrophobicity contributes to the progressive change of the dispersive characteristics in various solvents (Dhuiege *et al.* 2018). Onwukamike *et al.* (2018) have done research on sustainable cellulose transesterification using the CO₂-DBU solvent. They were able to achieve a DS of 1.59 and good thermal stability (95% at 360 °C) as well as high elastic moduli (up to 478 MPa), maximum stress of about 22 MPa, and the maximum elongation (strain) of about 35% (Onwukamike *et al.* 2018). Mokhena and John (2020) used canola vegetable oil to chemically modify NFC with and without catalyst. When NFC was esterified in the presence of a catalyst, a considerable extent of derivitization occurred, leading to relatively high DS) Modification without the addition of a catalyst resulted in a less significant change in the crystallinity index (Mokhena and John 2020). Concerning the disadvantages of this method, it should be noted that this method had a significant effect on the crystallinity, mechanical, light transmittance, and thermal properties (Mokhena and John 2020).

The next-generation materials containing oil, alkyd resins, and epoxy resin can play a key role as a coating binder because of their great flexibility in terms of construction, property changes, and their low cost (Taylor 2001). They could also be very interesting candidates for commercial application and up-scaling when combined with NC due to their hydrophobicity, biodegradability, and renewability (Aulin and Ström 2013). Their adhesive properties and excellent mechanical performance make them particularly well-suited for use in coating applications (Aziz *et al.* 2021). Aulin and Ström (2013) evaluated the effects of the different types of alkyd resin oil and varying the length of the oil on the unmodified NC-coated papers. Alkyd resins having longer fatty acid chains allow for the efficient formation of a hydrophobic polymer layer. Alkyd resins based on tall oil and linseed oil at the same oil length were compared. The alkyd resin based on linseed oil was shown to be the most effective in terms of lowering water vapor transmission rate (WVTR) due to the fatty acid composition and the degree of unsaturation (Aulin and Ström 2013).

Xu *et al.* (2019b) also used epoxidized soybean oil (ESO) as a low-cost, sustainable, and environmentally friendly raw material for forming modified cellulose. The WCA of modified cellulose aerogels was 132.6° when the mass fraction of ESO was 30 wt.% (Xu *et al.* 2019b). Huang *et al.* (2017) investigated the effect of grafting ESO with varying weight percentages onto properties of the cellulose. When the ESO concentration was raised, the WCA increased to 145.1° . The modified cellulose surface became rougher, which further increased its hydrophobic properties (Huang *et al.* 2017). Lu *et al.* (2015) developed a biodegradable agent to coat the NC film with a low WVTR. To reduce the WVTR, a rod coater was used to apply a coating agent composed of AESO and 3-aminopropyltriethoxysilane (APTS). The resulting film has great potential as a green packaging material (Lu *et al.* 2015).

Auclair *et al.* (2020) reported the use of coating systems with acrylated epoxidized soybean oil and CNC. They reported that CNC should be hydrophobized to make it more compatible with the hydrophobic AESO matrix due to a polarity mismatch between AESO and CNC. Therefore, the negative or positive electrostatic charges were applied to the CNC surface, resulting in improved polymer matrix dispersion. The enhancement compatibility was also achieved by adjusting the surface energy properties of nonpolar or hydrophobic polymer matrices (Auclair *et al.* 2020). This means that in order to improve the adhesion between MFC and the epoxy resin polymer matrix, the surface of the MFC should be modified, changing its character from hydrophilic to hydrophobic while keeping the crystalline structure of the MFC intact (Siró and Plackett 2010). Thus, two more steps are required to modify NFC with this type of biomaterial in order to achieve the desired results, which drives up the cost of coating materials. Moreover, these materials have a relatively long cure time and require high temperatures to cure (Tambe *et al.* 2016). They also typically have low tensile strengths, which limits their use as structural materials. The incorporation of fillers or reinforcements has been demonstrated to be a *viable* strategy for increasing the mechanical strength of shape-memory polymers (Sain *et al.* 2020).

Hydrophobization: Silylation

The silylation method is less environmentally damaging than the esterification methods, and it can be carried out *via* a solid-gas reaction without the use of a solvent (Jarrah *et al.* 2018). The surface hydroxyl groups on hydrophilic fibers are converted to alkyl silyl ethers *via* silylation, resulting in hydrophobic surfaces as shown in Fig. 10 (Jarrah *et al.* 2018). A silylation reaction produces ammonia as a by-product, which is a significant advantage of this method (Jankauskaitė *et al.* 2020).

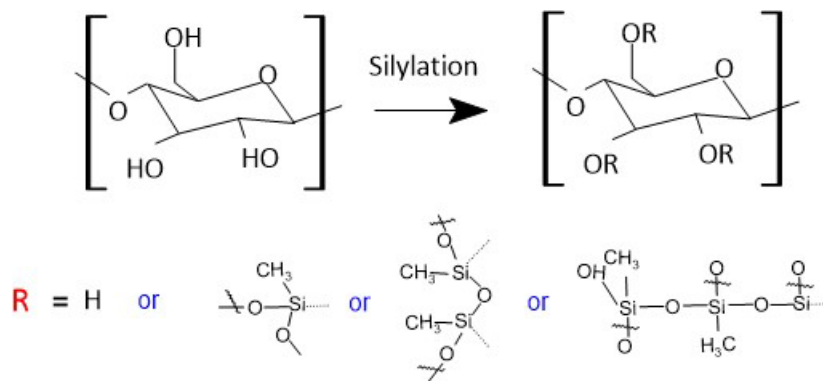


Fig. 10. Schematic diagram of the possible products of CH₃Si-modified cellulose, redrawn from Jarrah *et al.* (2018)

Khanjanzadeh *et al.* (2018) modified NC with 3-aminopropyltriethoxysilane, without using hazardous solvents. The surface morphology of nanocrystals was not considerably affected by functionalization. Nie *et al.* (2021) also modified the hydroxyl groups of the NFC surface with triethoxy-1H,1H,2H,2H-tridecafluoro-n-octylsilane, which enhanced their triboelectric charge density and hydrophobicity. The modified NFC showed growth in contact angle from 52° to 128° (Nie *et al.* 2021). Jarrah *et al.* (2018) used a gas-solid silylation reaction with silyl chlorides (RSiCl₃) to attach hydrophobic alkyl groups on the cellulose surface of cotton fibers, thereby converting them into hydrophobic materials and achieving a DS in the range of 0.10 to 0.29 per glucose unit (Jarrah *et al.* 2018). Yook *et al.* (2020) prepared various types of NFC, such as cellulose nanofibril (NFC), lignocellulose nanofibril, carboxymethylated cellulose nanofibril, silylated cellulose nanofibril (S-NFC), alkyl ketene dimer-added NFC (AKD-NFC), and coated on linerboard and wood-free paper to investigate the air, oxygen, water and grease barrier properties of these papers. The S-NFC and AKD-NFC were more water-resistant than NFC. The S-NFC had WCAs over 90 degrees. However, because of the short length of the hydrophobic alkyl groups in S-NFC, they may not inhibit effective water vapor transfer (Yook *et al.* 2020). It should also be emphasized that silane coupling agents have a high affinity for hydroxyl groups, even at room temperature; the NC core chains become silylated to the point of being soluble in the reaction medium, resulting in crystal disintegration and loss of the cellulose's original morphology (Ghasemlou *et al.* 2021)

Hydrophobization: Using plant polyphenols

The application of plant polyphenols has recently been introduced as an approach for developing hydrophobic nanoparticles in an environmentally friendly manner, in which low-cost plant polyphenols serve as precursors for the development of multifunctional coatings (Hu *et al.* 2017). Tannic acid (TA) is a naturally occurring polyphenolic compound with a wide range of bonding abilities. It can complex or cross-link macromolecules at several binding sites using a variety of interactions, including hydrogen and ionic bonding as well as hydrophobic interactions (Fan *et al.* 2017). Hu *et al.* (2017) proposed a simple method for producing CNC-TA-DA (decylamine) *via* covalent binding to increase the water contact angle and the ability to be redispersed in organic solvents. Furthermore, they believed that this simple method is less time-consuming than polymer grafting and produces particles with minimal aggregation and morphological changes (Hu *et al.* 2017). Missio *et al.* (2018) used NFC and condensed tannins to create a durable, long-lasting packaging material. The physical and mechanical requirements were met by NFC, while the tannin was added for its antioxidant qualities. This film had a higher density and better surface hydrophobicity, resulting in a 6-fold improvement in air-barrier qualities (Missio *et al.* 2018). Huang *et al.* (2019) used layer-by-layer assembly to layer CH and TA on NFC, since TA has adhesive properties as a result of the aromatic components in its composition, allowing it to form coatings on a variety of nonporous and porous substrates. The key force behind the formation of multilayers is the electrostatic interaction between positively charged CH and negatively charged TA. Improved mechanical properties and strong antibacterial properties were found in this layer by layer structured film (Huang *et al.* 2019). Missio *et al.* (2020) used a mixture of NFC and condensed TA to fabricate functional film. They also used a non-ionic surfactant to give them even more control over the tannin-cellulose interface and, as a result, over the film properties. These tannin-containing films had a higher hydrophobic character, as shown by increased WCA (Missio *et al.* 2020).

Hydrophobization: Chemical vapor deposition (CVD)

Solution-based methods are commonly used to modify materials; however, in these procedures, finding an adequate solvent can be a limiting factor due to the fact that it must be compatible with both the substrate and the coating. Conformal coating of micro- or nanoscale structures is also limited by surface tension effects. As a result, methods involving the vapor phase, such as CVD, are preferred for coating complex surfaces (Cheng and Gupta 2018). Chemical vapor deposition (CVD) methods are capable of producing homogeneous and conformal organic thin films without using solvents, catalysts, or separation agents (Hilt *et al.* 2014). CVD benefits from the excellent interface and surface morphology, growth of complex heterostructures with numerous layers, growth on patterned substrates, multiple wafer scale-up, and high layer purity (Pessoa *et al.* 2015). CVD procedures also are well-controlled techniques that allow the operator to quickly control the amount of material by changing the coating thickness (Hilt *et al.* 2014). Fluorocarbons, silicones, and organic or inorganic compounds are employed in these tactics to apply to cellulose and reduce surface energy (Leal *et al.* 2020). Yu *et al.* (2019) applied a one-step gas-solid reaction for the production of hydrophobic NC and bacterial cellulose (BC) with CVD of perfluorooctyltriethoxysilane. The results showed that perfluorooctyltriethoxysilane was successfully incorporated into the CNC and bacterial cellulose. They likewise believed that the vapor-based technique can be used to produce hydrophobic cellulosic materials on a large scale for industrial purposes (Yu *et al.* 2019). Rafieian *et al.* (2018) modified NFC via CVD of hexadecyltrimethoxysilane (HDTMS). They believed that HDTMS with the longer the alkyl chain of hexadecyl (16 carbon atoms) compared to methyl (one carbon atom) contribute to making the polymer more hydrophobic, allowing the WCA of NFC to increase to 139. They found that untreated cellulose had a higher crystallinity than those treated with CVD (Rafieian *et al.* 2018). Cunha *et al.* (2018) also studied the impacts of CVD post modification on NFC filaments properties. Two organosilanes with varied numbers of methyl substituents were chosen for this purpose. Various surface structures including continuous, uniform coating layers or three-dimensional, hairy-like structures were observed, which served to decrease the surface energy and had a big impact on water interactions (Cunha *et al.* 2018). The decrease in surface energy was not expected and explained by the influence of the polar component in the organosilanes. A drawback of CVD is that it frequently involves extremely high temperatures and vacuum conditions. In addition, *in situ* monitoring of the process is restricted to specific analysis. However, Yang *et al.* (2020b) developed low-temperature-CVD with the goal of modifying cellulose-based to make them hydrophobic. Vacuuming has been used to allow hydrophobic gas to enter and react with the wood (Yang *et al.* 2020b).

Hydrophobization: Atomic layer deposition (ALD)

Atomic layer deposition (ALD) is a sub-set of the CVD process that uses a series of self-limiting surface adsorption reactions (chemisorption) carried out in a vacuum to control composition and the thickness of the deposited film with a thickness resolution of Angstroms (Wooding *et al.* 2020). ALD allows for higher uniformity and conformance on complex substrates as compared to standard CVD procedures, since the precursor molecules have a longer lifetime to diffuse to the cavities in complicated three-dimensional substrates (Wooding *et al.* 2020). Rare-earth oxide ceramics have been recognized as having strong hydrophobic surfaces, so they should find the widespread application (Azimi *et al.* 2013). A schematic description of the ALD process is shown in Fig. 11 (Oviroh *et al.* 2019).

1. The first precursor is exposed in the reactor chamber to form a layer on the substrate.
2. Remove any remaining initial precursors and by-products.
3. The second precursor is exposed.
4. Excess second precursor and by-products are purged

The procedure is repeated until the desired film thickness is obtained.

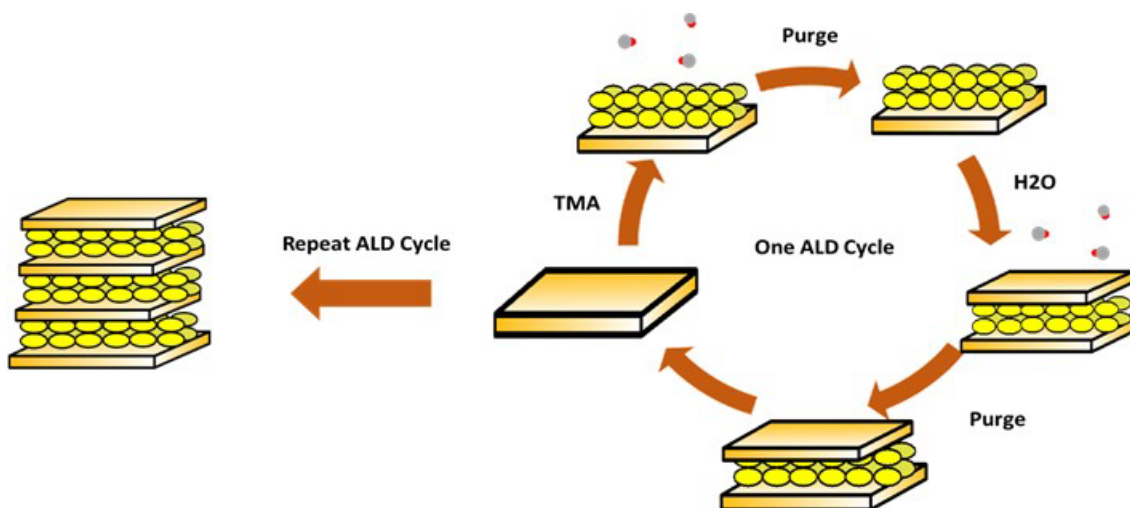


Fig. 11. A model ALD process for depositing trimethylaluminum on hydroxyl groups functionalized NC. Reproduced from Oviroh *et al.* (2019)

It should be added that increasing the number of cycles contributed to a rough structure, trapping more air, resulting in a more hydrophobic substrate, but the WCA decreased significantly after specific cycles of ALD coating. This may be because as the number of ALD cycles increased, more hydrophilic Al_2O_3 was accumulated on the surface (Lu *et al.* 2020a). Li *et al.* (2020) discovered that an increase in contact angle would be due to the hydrocarbons being drawn to the ALD-deposited metal oxide surface. The presence of hydrocarbons on the nanopaper surface was detected, but it is unclear whether this was due to accidental sources or chemical degradation of the cellulose caused by exposure to ALD precursor molecules (Li *et al.* 2020). Omerzu *et al.* (2018) evaluated the characteristics of Al_2O_3 coating layers formed on the surfaces of cellulose fibrous materials by using ATD. They state that a thin Al_2O_3 barrier coating on the surface of fibrous cellulose material effectively inhibits oxygen transport and spontaneous combustion of cellulose, but they made no mention of a water vapor barrier (Omerzu *et al.* 2018). Li and Losego (2021) modified NC paper (nanopapers) chemically and physically with simple gas-phase processing. They reported that longer trimethylaluminum exposure durations considerably increased the nanopaper's tensile strength. They also showed that the NC paper became more hydrophobic after being treated with 1cy-ALD (Li and Losego 2021). It should be noted that the WVTR results obtained were not consistent with one another and that the fragile and degradation characteristics of Al_2O_3 films influenced the barrier properties' stability. More importantly, the precise reasons for these disparate WVTR results remain unknown. Numerous attempts have been made to identify a critical innovation capable of significantly improving the alumina film's protective and service characteristics. One strategy is to enhance the ALD technique by incorporating plasma pre-treatment (Fang *et al.* 2017).

It appears that plasma-enhanced ALD requires half the number of deposition cycles as thermal ALD to achieve the same thickness of film. Additionally, no structural or protective variation in Al_2O_3 coatings was identified between the thermal ALD and plasma-enhanced ALD films produced (Omerzu *et al.* 2018).

Hydrophobization: Plasma

Plasma is usually referred to as a gas that's been electrically charged with freely moving electrons in both the negative and positive states. Plasma treatment technology offers numerous advantages for surface modification because it is less toxic, safe, and developable technology compared to wet technologies (such as acetylation, alkylation, and carbamoylation) (Yao *et al.* 2021). Plasma can insert specific moieties throughout different mechanisms, such as cross-linking, deposition, etching, and functionalization, without modifying the bulk properties, as shown in Fig. 12 (Laureano-Anzaldo *et al.* 2020).

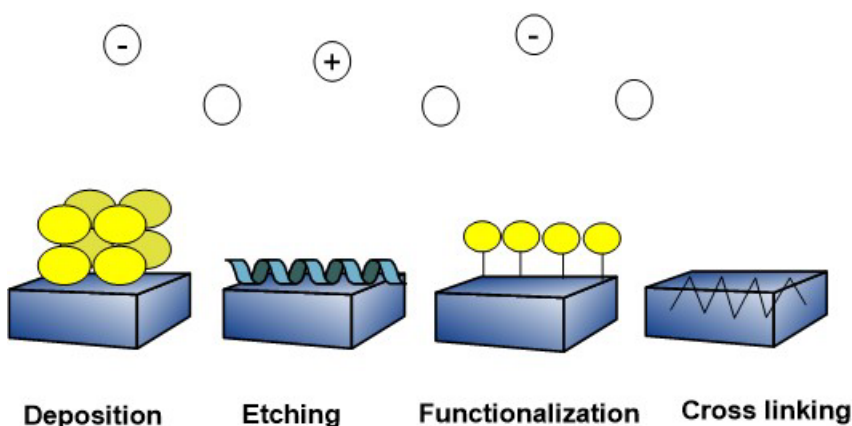


Fig. 12. Mechanisms of surface modification achieved with plasma treatment, redrawn from Laureano-Anzaldo *et al.* (2020)

Several distinct plasma approaches have been employed to hydrophobized cellulose nanomaterials: Atmospheric pressure, submerged liquid plasma, and low-pressure plasma (Oberlintner *et al.* 2021). Samanta *et al.* (2012) treated cellulosic (viscose rayon) fabric with He/1,3-butadiene (BD) plasma in a parallel-plate DBD reactor, achieving hydrophobicity with WCA of 142° by incorporation of CH_x fragments on cellulose –OH sites (Samanta *et al.* 2012). Samanta *et al.* (2016) treated hydrophilic cotton (cellulosic) textile with helium-fluorocarbon (He- CF_4) gases in an indigenously developed atmospheric pressure plasma reactor to impart hydrophobic functionality (Samanta *et al.* 2016). Due to the reaction of different CF_4 species with cellulose, hydrophilic textile became hydrophobic after plasma treatment (Hubbe *et al.* 2015; Samanta *et al.* 2020). Yao *et al.* (2021) used oxygen plasma treatment in combination with a condensation reaction with TriSilanoll-sobutyl-Polyhedral oligomeric silsesquioxane to create a new hydrophobic cellulose-based organic/inorganic nanomaterial (Cellulose/TriSilanollsobutyl (TS)- Polyhedral oligomeric silsesquioxane (POSS)). The hydroxyl groups on the cellulose surface were not only activated, but the surface energy was also lowered and the surface roughness was raised, thanks to the plasma etching (TS-POSS modification) (Yao *et al.* 2021). Panaitescu *et al.* (2018) used submerged liquid plasma to surface functionalize NC. They were able to observe the effect of plasma on the morphology and surface chemistry of NC (Panaitescu *et al.* 2018). Matouk *et al.* (2020) functionalized CNC using cylindrical atmospheric pressure dielectric

barrier discharge in order to minimize surface hydrophilicity and increase dispersion in polar organic solvents. They showed that using plasma with the right chemistry, such as Ar/CH₄, Ar/NH₃, and Ar/SiH₄, can encourage hydrophobicity or hydrophilicity in CNC films by altering its elemental chemical composition and roughness (Matouk *et al.* 2020). Kartick *et al.* (2021) studied the hydrophobic functionalization of cellulosic fabric using helium/tetrafluoroethane plasma, a commercially available fluorocarbon gas (CF₄), at atmospheric pressure. The highly hydrophilic cellulosic fabric became superhydrophobic after plasma treatment, with a water absorbency time of >60 minutes, a WCA of 153, and a water rolling angle of 5 (Samanta *et al.* 2021). As highlighted by Poodt *et al.* (2012) the intriguing potential of plasma has been demonstrated. However, before implementing this method on a commercial scale, further safety and risk evaluations should be conducted on cold plasma interactions (Bahrami *et al.* 2020).

It is worth mentioning that CF₄ gases, which are commonly used for hydrophobization using plasma, are one of the most persistent greenhouse gases and should not be released into the atmosphere. Furthermore, the precise mechanisms and kinetics of the reactions that occur on the surface of cellulose nanomaterials remain unknown (Oberlintner *et al.* 2021).

Functionalization by Cationization

The capability of nanocellulose to adsorb on fiber surfaces is important for its use in high-strength applications (Li *et al.* 2021a). Accordingly, functionalized NC appears to be promising in terms of promoting bonding and subsequent mechanical strength of paper. Surface functionalization of nanoparticles is the process of joining various organic and inorganic materials together at the nanoscale *via* covalent or noncovalent bonds such as hydrogen bonds, the electrostatic force, or the van der Waals force (Pattnaik *et al.* 2020). Numerous methods of surface covalent functionalization of NC have been described, including the following: (TEMPO) oxidation (Lourenço *et al.* 2017), periodate oxidation (Sirviö *et al.* 2011), esterification (Wang *et al.* 2018), amidation (Pettignano *et al.* 2019), silylation (Nie *et al.* 2021), etherification (Li *et al.* 2021b), and other chemical modifications. Based on recent research, cationic nanocellulose has garnered considerable attention because it has the potential to provide electrostatic binding with anionic particles, which could open up more opportunities for functional NC (Sunasee and Hemraz 2018) to be used as flocculating agents and strength additions in the paper-making process. The idea of cationic fiber was established in pulp and paper production to act as a replacement for the typical cationic polymer used in the papermaking industry (Gao *et al.* 2016). In the pulp and paper industry, polyelectrolytes with cationic groups, such as CS and C-PAM, are used in papermaking to increase mechanical properties, promote load retention, facilitate drainage rate, and lower the biological oxygen requirement of pulp mill effluents (Li *et al.* 2018). Therefore, the addition of cationic fiber also can lead to improvements in filler retention, drainage, and the ability to absorb anion particles (Gao *et al.* 2016). Figure 13 shows the basic mechanisms through which cationic NC interacts with fibers (Abbott *et al.* 2006).

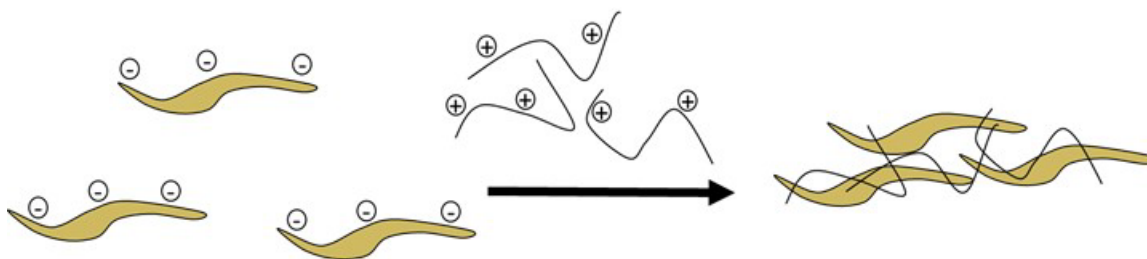


Fig. 13. Fibers and CNFC: electrostatic interactions upon the addition of cationic nanocellulose, redrawn from Sharma *et al.* (2020)

Lu *et al.* (2020b) demonstrated that the addition of cationic cellulose nanofiber (CNFC) slowly increased the basis weight, fines retention, and density of paper sheets since the addition of CNFC can enhance fines retention in paper sheets. Indeed, CNFC would adsorb and flocculate fines in the pulp slurry to produce the CNFC-Fines complex, and then it would attached in the wet web structure *via* a “bridging” effect and can pave the way for the dewatering process (Lu *et al.* 2020b). There are three common methods for producing cationic modified fibers. The first technique is to directly cationize the fibers in order to obtain the cellulose fiber amine compounds. The second method involves coupling a cationic prepolymer with a short chain to the fiber surface in order to achieve a fiber with a high surface charge density. Thirdly, cationic polymers are grafted onto the fiber using free-radical copolymerization; cationic monomers and neutral monomers are linked to the fiber surface *via* free radical copolymerization (Gao *et al.* 2016). In the following sections, we will compile a list of cationization processes in order to determine the one that has minimized environmental impact while still producing high-performance CNFC.

Cationization: Physical adsorption by cationic surfactant

NC consists of negatively charged particles that are expected to interact strongly with cationic surfactants electrostatically (Brinatti *et al.* 2016). Certain surfactants such as for example quarternary aluminium salt can change the surface on NC, and it has been shown that their zeta potential reverse from negative to positive (Xiang *et al.* 2019). The adsorption of cationic surfactants on cellulose exhibited a complex behaviour that varied according to the charge of the cellulose, the molecular structure of the surfactant, and the ionic strength of the solution (Alila *et al.* 2005). Dai and Fan (2013) demonstrated the green nano-modification in two stages: (i) cationization of natural fibers with dodecyltrimethylammonium bromide and (ii) NC modification. The research used dodecyltrime-thylammonium bromide to pretreat fibers by adding cations to their surface, and then NC to change the pretreated fibers (Dai and Fan 2013). Mariano *et al.* (2018) modified NFC by using cationic surfactants (*e.g.*, dodecyltrimethylammonium bromide (C12TAB), tetradecyltrimethylammonium bromide (C14TAB), and hexadecyltrimethylammonium bromide (C16TAB)) as modifying agents. Surfactants were added to NFC suspensions, causing the surface of the nanoparticles to be covered by these molecules, and lowering the ζ -potential (Mariano *et al.* 2018). Xiang *et al.* (2019) also modified CNC by electrostatic interaction of hexadecyl trimethyl ammonium bromide. The hexadecyl trimethyl ammonium bromide layer contributed a huge amount of positive charges to the CNC (Xiang *et al.* 2019). Kushan *et al.* (2020) modified CNC solutions by using cationic [C10 mim][Cl] and [C10 mim][FeCl₄] surfactants as well. They showed that CNC network development is predom-

inantly driven by hydrophobic contact of surfactant tails due to high electrostatic interaction between anionic CNC and cationic [C10 mim][Cl] surfactants. Hu *et al.* (2015) also modified CNC with cationic alkyl ammonium surfactants dodecyl-dimethylammonium bromide and cetyltrimethyl ammonium bromide. Both surfactants were adsorbed onto CNC with concentration-dependent morphology. Surfactant molecules adsorbed with alkyl tails pointing outward at low concentrations, resulting in hydrophobic CNC. At greater concentrations, surfactant aggregation morphologies on CNC were detected, and the hydrophobicity of CNC was diminished. Besides, CNC becomes cationic at higher surfactant concentrations since surfactant aggregates on CNC are thought to be responsible for this surface charge reversal (Hu *et al.* 2015).

Cationization by etherification

Etherification is a chemical process that involves grafting bifunctional monomers onto the fiber surface to facilitate the reaction between polymer chains and fibers. Currently, two commercially viable reagents, 3-chloro-2-hydroxypropyl trimethyl ammonium chloride (CHPTAC) and 2,3-epoxypropyl trimethyl ammonium chloride (EPTAC), are used to cationize NC in an alkaline medium (mostly using NaOH) (Sharma *et al.* 2020).

The presence of many cellulosic -OH groups on NFC allows for etherification reactions to modify NFC (Sethi *et al.* 2018). Depending on the substitution groups employed, a variety of features can be introduced or adjusted by selectively modifying these hydroxyl groups. The solubility and viscosity of cationized cellulose are affected by the degree of molar substitution, the distribution and type of substituents, and the molecular weight of cellulose and cellulose derivatives (O'Brien *et al.* 2021). Liu *et al.* (2019a) used cationic NFC (CNFC) as an additive in the preparation of reconstituted tobacco sheets. CNFC was created by homogenizing bleached hardwood kraft pulp that had been cationic etherized under high pressure (Liu *et al.* 2019a). Gu *et al.* (2020) stated that glycidyltrimethylammonium chloride (GTMAC)-involved etherification could be a potential method for producing well-dispersed nanoparticles, especially from parenchyma cellulose. With the help of ultrasound, quaternary ammonium groups were successfully grafted onto cellulose chains on the surface of NC (Gu *et al.* 2020). Luo *et al.* (2021) created cationic tobacco fiber by etherifying activated hydroxyl groups under alkaline circumstances using the etherifying compound EPTAC. The effect of alkali dosage on the tobacco cellulose nanofiber charge density was studied, and it was concluded that the charge density of tobacco cellulose nanofiber increased when the alkali dosage was raised since the degree of etherification of tobacco fibers was substantially increased as a result of the increased swelling and activation ability of hydroxyl groups (Luo *et al.* 2021). Chakrabarty and Teramoto (2021) used an alkali-activated etherification reaction with GTAC to modify the surface of NC in order to improve the ability of emulsion stabilization. It should be noted that these methods necessitate the use of specialized organic solvents, which break up the H-bonding network in cellulose. In general, the dissolution can transform cellulose I into cellulose II. These two materials differ in their crystal structure by the hydrogen-bond pattern, with cellulose I having parallel macromolecular chains while cellulose II has an antiparallel arrangement. The next difference is recognized by density functional theory optimizations, which showed that the cellulose I chain sheet models twisted to the right, while the cellulose II model twisted to the left. In general, materials made from cellulose I exhibit better mechanical properties than materials made from cellulose II. Indeed, the elastic moduli of

cellulose I are about twice those of cellulose II. While industrial cellulose production technology has existed for more than 100 years, in the vast majority of cases, commercial cellulose products are still made with cellulose II (Tu *et al.* 2021).

Since cellulose is poorly soluble in water and most organic solvents, a substantial amount of organic solvents is required for this purpose (Tang *et al.* 2017). Odabas *et al.* (2016) showed the use of solvents such as dimethylsulfoxide (DMSO), tetrahydrofuran (THF), or isopropanol to increase the cellulose modification (Odabas *et al.* 2016). Chaker and Boufi (2015) also reported DS values between 0.05 and 0.19 by using dimethylacetamide (DMAC) as solvent and potassium hydroxide (KOH) as a catalyst (Chaker and Boufi 2015). High DS is difficult to achieve and has a significant impact on energy consumption as well (Rol *et al.* 2019b). Therefore, the high temperature and the production of waste products severely limit the application of this method (Li *et al.* 2021b). The next drawback related to the etherification reaction worth mentioning would be the formation of diol and other toxic by-products. Additionally, sequential steps such as dispersing, reacting with the reagents in an alkaline medium, neutralizing the reaction mixture, separating, washing, and drying the final product are frequently required (Sharma *et al.* 2020). Furthermore, the traditional method for cellulose etherification reaction takes a long time, typically 8 to 80 hours (Zhang *et al.* 2021c), and it has low efficiency as well. Ultrasonication is widely regarded as a green mechanical method for helping modification of the surface of cellulosic materials and increasing reaction rates through a process known as cavitation. As a result, after etherification in more than 10% cellulose consistency, a higher DS could be achieved by ultrasonic processing for sonication at the frequency of 10 min per 0.5 h, indicating that a longer duration of sonication facilitated the increase in the number of cationic substituents on the PC surface (Gu *et al.* 2020). Microwave use is another way to aid etherification. Zhang *et al.* (2021c) synthesized quaternary cellulose samples with different DS ranging from 0.16 to 0.51 by etherification with the assistance of microwaves (Zhang *et al.* 2021c).

Cationization: Periodate oxidation and reductive amination

As previously stated, cellulose's insolubility in most common solvents is a fundamental impediment to its use in different processes (Marcus 2019). It has been noted that the unique interactions of cellulose with solvent are largely dependent on the cellulose's substitution sites. Due to the absence of hydrogen bonding between cellulose chains, 6-O-methylcellulose proved highly soluble in a wide range of organic solvents, but 2,3-di-O-methylcellulose is just soluble in polar aprotic solvents (Fox *et al.* 2011). Periodate oxidation is a highly selective oxidative fragmentation of the C2-C3 glycol bond on the glucose ring, yielding a dialdehyde NC (DAC) product (Li and Xu 2014). This DAC can be used as a reactive intermediate for further derivatization, as well as a starting point for a variety of other chemical processes (Jin *et al.* 2015). Due to its biocompatibility, solubility in hot water, and potential for further modification *via* the aldehyde groups, DAC has received considerable attention in bio-based products (Esen and Meier 2020). So, pretreatment of C2 and C3 anhydroglucose units position by periodate oxidation could be a good suggestion as shown in Fig. 14. Zhang *et al.* (2019) claim that it is easily modified *via* interfacial Schiff base coupling owing to the existence of highly reactive aldehyde groups. They also stated that the Schiff base reaction between DAC and aniline is a straightforward and easy scale-up approach of interfacial covalent assembly for producing solvent-responsive nanoparticles (Zhang *et al.* 2019).

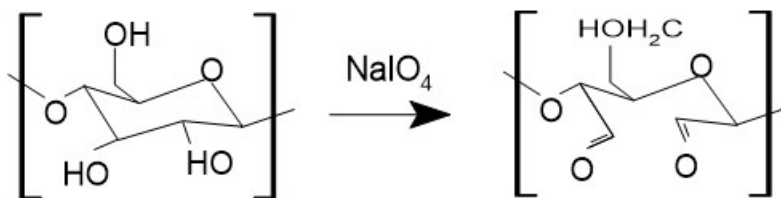


Fig. 14. Periodate oxidation and reductive amination of NFC, redrawn from Xu *et al.* (2020)

Sirviö *et al.* (2011) cationized highly oxidized DAC and reported that the concentration of cationic groups increased from 2.85 to 4.27 mmol/g as the reaction time increased from 1 to 72 hours. Under these conditions, 87 percent of the aldehyde groups reacted with (2-hydrazinyl-2-oxoethyl)-trimethylazanium chloride Girard's reagent T (GT). Huang *et al.* (2020) also synthesized cationic dialdehyde NFC (CDAC) with three different degrees of oxidation by oxidizing cellulose with metaperiodate and then cationizing it with (carboxymethyl) trimethylammonium chloride hydrazide as GT. They select GT, a typical grafting reagent in the paper industry that is relatively inexpensive and common, in order to introduce positive charges onto the NFC surface and make it more capable of interacting with negatively charged molecules (Huang *et al.* 2020). Kokol *et al.* (2021) also employed the two-step technique described below to functionalize NFC: sodium periodate oxidation of NFC to DAC and subsequent reaction with hexamethylene-diamine (HMDA) *via* a Schiff-based reaction to obtain NFC-ald-HMDA viscose yarn. They introduced this modified NFC to a ring-spun yarn as an antibacterial ingredient (Kokol *et al.* 2021).

Yang and van de Ven (2016) synthesized hairy cationic CNC (CNCC) by oxidizing cellulose with periodate and cationizing it with GT. They assert that this is a simple and environmentally friendly procedure that can be carried out in mild aqueous conditions. Wang *et al.* (2020a) synthesized CNCC *via* periodate oxidation and the Schiff base reaction. Periodate oxidation and disintegration were used to prepare di-aldehyde CNC from kraft pulp. Through a Schiff base reaction followed by sonication, the obtained DAC was further modified to include quaternary ammonium groups, yielding CNCC with an average length of 400 nm and a width of 15 nm. The combination of this CNCC and anionic alginate was used to make hydrogel beads (Wang *et al.* 2020a). Tian *et al.* (2017) also used this type of modification to obtain CDAC in order to be used in composite production (Tian *et al.* 2017). Still, these methods have a number of drawbacks including the use of large amounts of organic solvents (Yang *et al.* 2019b). Furthermore, the loss of crystallinity is thought to be caused by the opening of rings and the destruction of their ordered packing (Li *et al.* 2011).

Cationization: Using ionic liquid

The desire for more environmentally friendly functionalization procedures has resulted in significant research efforts aimed at identifying acceptable solvents. Ionic liquids (ILs) are a new class of organic molten salts with melting points less than 100 °C. They exhibit a number of desirable properties, including low vapor pressure, high thermal and chemical stability, high solvating capacities, structure tunability, ease of recycling, and nonflammability, and thus they offer the possibility for widespread industrial use. The development of ILs has created a new and versatile foundation for the widespread use of cellulose resources and the development of novel functional materials (Zhang *et al.* 2017). This kind of solvent (ILs) with strong hydrogen bond acceptors has demonstrated a high level of efficacy in dissolving and treating cellulose (Zhang *et al.* 2014).

The cellulose present in wood and plant fibers is mainly cellulose I β , a crystal structure with a certain shape. This shape can be changed by various treatments, such as dissolution, which weakens the hydrogen bonds between the hydroxyl groups, resulting in decreased crystallinity (Kovalenko 2010). Therefore, the solvent system that is chosen for modification needs to be carefully selected. Solvent systems have been studied for many years, and some researchers, such as Lin and Tsuchii (2022), have classified them into four groups based on their solubility. In Type A, cellulose was dissolved in large amounts, whereas Type B dissolved just slightly and Type C swelled the cellulose, but could not dissolve it at all. Type D was neither able to dissolve nor swell cellulose (Lin and Tsuchii 2022).

In general, ILs as solvent are thermally stable across a wide temperature range and can be used in a variety of reaction media for homogenous cellulose modification. The considerable intermolecular and intramolecular hydrogen bonding of cellulose is anticipated to be disrupted by ILs (Zhang *et al.* 2014).

Abbott *et al.* (2006) illustrated the efficient cationic functionalization of cellulose using an IL analog composed of a mixture of a choline chloride derivative and urea that serves as both solvent and reagent. It was discovered that all of the cellulose's available hydroxyl groups had been modified by ILs (Abbott *et al.* 2006). Qian *et al.* (2019) formulated novel poly ILs with 1-vinyl-3-aminopropyl imidazolium cations to modify cellulose. They first obtained dialdehyde cellulose aerogel *via* a simple oxidation process and then used sodium cyanoborohydride to prepare polyionic liquid (PIL) modified cellulose. The modified cellulose had a highly porous structure that was well-connected (Qian *et al.* 2019). Yang *et al.* (2019a) immobilized polyoxometalate (POM) supported PIL on the surface of CNC *via* in-situ Cu-mediated reversible deactivation radical polymerization (RDRP). CNC@PIL@POM demonstrated a nanorod-type core-shell structure with a high degree of crystallinity due to the CNC core and a PIL/POM composite shell (Yang *et al.* 2019a). Bernard *et al.* (2018) created cationic cellulose *via* PIL. The process began with the chemical modification of rice husk cellulose with thionyl chloride to form chlorinated cellulose (6-chloro-6-deoxycellulose, CDC), followed by surface grafting with 1-methylimidazole (Bernard *et al.* 2018).

The recovery of ILs is still in its early stages and is a frequent topic of discussion in the cellulose processing industry (Druel *et al.* 2018). Moreover, the industrial manufacturing and expense of ILs currently preclude their use in a wide range of fields. If the cost of ILs can be significantly reduced in the future, it will encourage its continued use in the pulp and papermaking industries (Li *et al.* 2019).

Cationization: Using deep eutectic solvent

Deep eutectic solvents (DESs) have been recognized as the most impressive environmentally friendly substitution solvents and reagents for some of the chemical and industrial processes. They are created by combining two or more chemical reagents into one solution with a lower melting point than the original reagents. It ought to be added that DESs can serve as both a reagent and a reaction medium in the fabrication of functionalized NFC and CNC (Selkälä *et al.* 2020). Unfortunately, the desired attention has not been received in the literature related to the use of DESs as alternative solvents for cellulose functionalization (Yang *et al.* 2019b).

DES should be able to disrupt the initial hydrogen-bonding networks in cellulose chains and DESs, then construct new hydrogen-bonding networks between cellulose molecules and DESs (Tang *et al.* 2017). Abbott *et al.* (2006) describe the first effective use of

a DES in organic synthesis processes. Their DES containing a choline derivative and urea as both a reagent and a solvent is used in the cationic functionalization of cellulose (Abbott *et al.* 2006). Ren *et al.* (2016) propose a simple and effective DES functionalization approach for better cellulose dissolution. A novel allyl-functionalized deep eutectic solvent (DES) was created *via* allylic replacement of the –OH group in choline cation. As a result, substituting –OH groups in the choline cation may increase the dissolving capacity of ChCl-based DESs (Ren *et al.* 2016). Li *et al.* (2018) created a DES by combining aminoguanidine hydrochloride and glycerol (AhG) and using it as a reaction medium and reagent (aminoguanidine hydrochloride) to cationize DAC. To make cationic NC, the cationized celluloses were mechanically disintegrated. The structure of NC could be altered to produce highly cationic NFC (CNFC) or cationic CNC (CCNC) (Li *et al.* 2018).

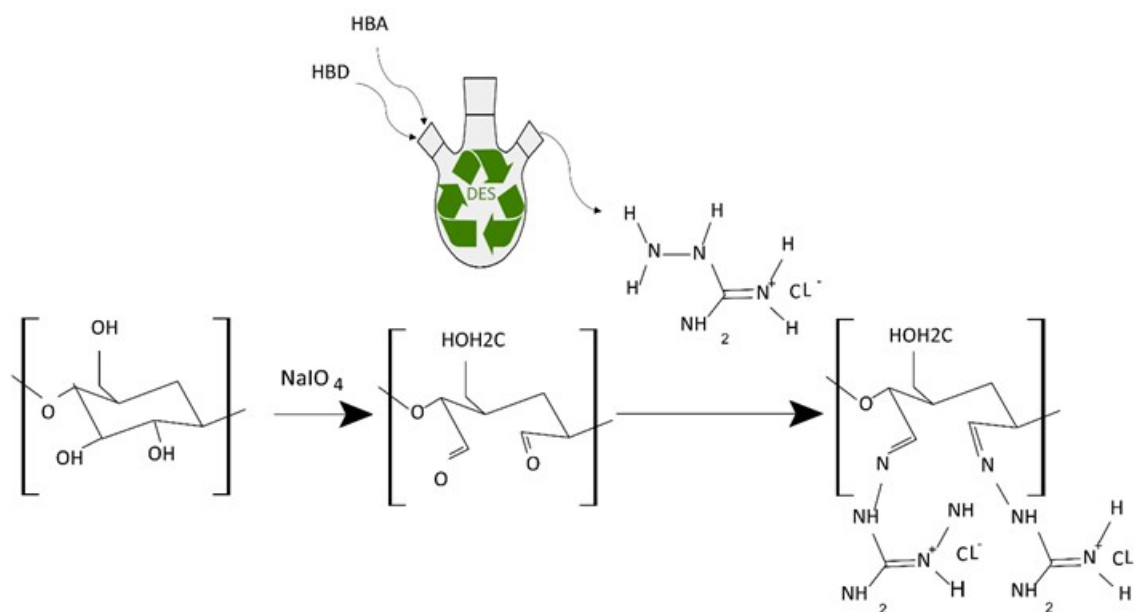


Fig. 15. Cationization of oxidized cellulose pulp in aminoguanidine hydrochloride–glycerol deep eutectic solvent, redrawn from Li *et al.* (2018)

Deep eutectic solvents (DES) represent an economically appealing alternative, as the recycled version performs identically to the original. It has been shown that after five repetitions, the recycled DES had a similar reaction performance to the original DES (Li *et al.* 2018). Selkala *et al.* (2020) formulated CCNC by oxidizing pulp with periodate and then cationizing it in a DES in order to be used in dye removal (Selkälä *et al.* 2020). Jaekel *et al.* (2021) also used a DES to establish a sustainable approach for preparing CCNC. They formulated reactive eutectic media containing ammonium formate as the hydrogen acceptor component in combination with various organic acids, namely glycolic, lactic, and levulinic acid, which served as hydrogen bond donors. Based on the use of reactive eutectic media, they introduced a new approach for extracting CNC and functionalizing them with positively charged functional groups in a one-step procedure (Jaekel *et al.* 2021). Asante *et al.* (2020) also produced CCNC in a two-step process involving subsequent periodate oxidation and cationization in a DES and the modified cellulose was disintegrated mechanically to release CCNC. Cationization of DAC was accomplished in DES, which was

formed by mixing aminoguanadine hydrochloride and glycerol in a 1:2 molar ratio (Asante *et al.* 2020).

Thus, the implementation of a low-cost DES opens up new possibilities for sustainable cellulose functionalization, as the reaction could be more environmentally friendly by further reducing the reaction time and recycling the DES (Selkälä *et al.* 2016). However, DESs have low ionic conductivity and a high viscosity. These shortcomings can be addressed by developing a novel hybrid DES with extraordinary properties (Lien *et al.* 2021).

APPLICATION OF MODIFIED NANOCELLULOSE IN PAPERMAKING

In general, papermakers appreciate techniques that enhance the paper's strength and barrier characteristics, which can be accomplished through the use of NC in a variety of doses and forms (Hubbe 2014). However, as already mentioned, their usefulness as a dry end additive declines when the relative humidity surpasses 70%, because of the moisture sensitivity of NC. Additionally, NC as a wet end additive reduces drainability by blocking the pores of the wet paper web (Sharma *et al.* 2020). To avoid the limitations of NC in the papermaking process, a feasible approach is to modify NC and then apply it in the papermaking process. This section considers modified NC that has been used in the paper industry as dry end additive or wet end additive. Finally, the potential for using these approaches for NC modification in paper making is discussed. The goal is to be able to select certain types of environment-friendly additives/modifiers, having in mind low cost and ease of handling.

Modified NC-additive Used in Coating of Paper

The silylation of cellulose fibers has been investigated, with the goal of imparting hydrophobic characteristics of paper (Rol *et al.* 2019a). Yook *et al.* (2020) used silylation in an aqueous system to create hydrophobic NFC and coated them on linerboard and wood-free paper to evaluate their barrier properties. The introduction of hydrophobic functional groups to NFC through silylation transformed the surface chemical characteristics of NFC from hydrophilic to hydrophobic. The modified NFC coating exhibited better barrier properties than unmodified NFC coating, but the difference in functional groups induced a discrepancy in the barrier properties against water vapor and oxygen (Yook *et al.* 2020).

Moderate hydrophobicity properties can be created by adsorbing surfactants to the NC surface (Rol *et al.* 2019a). Tiitu *et al.* (2006) reported on the application of surfactant to modified NC, which was then coated on paper as dry end additive. They prepared complexes of carboxymethyl cellulose and alkyltrimethylammonium surfactant to coat the paper using spray coating techniques (Tiitu *et al.* 2006). Xhanari *et al.* (2011) used cetyltrimethylammonium bromide as a surfactant to modify NFC. The mixtures were then deposited on top of filter paper to investigate the barrier properties. They observed that the wettability of filter paper coated by surfactant-NFC was slightly reduced (Xhanari *et al.* 2011). To promote wettability, Abou-Zeid *et al.* (2018) identified another task for surfactants. They prepared nano-biocomposite films of polylactic acid (PLA)/CNC to coat onto paper. The cationic and anionic surfactants hexadecyltrimethylammonium bromide and sodium lauryl sulfate were responsible for avoiding CNC re-aggregation during drying and promoting CNC dispersion in PLA (Abou-Zeid *et al.* 2018).

In terms of bio-polyphenol application, the majority of research on bio-polyphenol incorporation into composites for packaging has been focused on combining tannin, NC,

and cellulose film. As tannin is easily washed away due to its water solubility, so it is unsuitable for direct incorporation into the papermaking process (Ji *et al.* 2020). It seems that it would be more appropriate to use it in the coating process. Missio *et al.* (2018) developed a strong, sustainable packaging material using NFC and condensed tannins. The tannin-infused cellulose films exhibited a high density and increased surface hydrophobicity, resulting in a six-fold increase in their air-barrier properties. They concluded that NFC-tannin films are well suited for a variety of applications in packaging technology (Missio *et al.* 2018). Ji *et al.* (2020) created a novel cellulosic paper by incorporating tannin into kraft pulp for potential use in active food packaging. It was discovered that after tannin incorporation, paper became surface hydrophobic with contact angles greater than 90°, which was most likely due to covalent bonds between tannin and cellulose (Ji *et al.* 2020).

Gas techniques such as CVD also have shown good efficiency in nanopaper coating. Phanthong *et al.* (2016) used the CVD method to create amphiphobic NC-modified paper with high durability using a simple two-step method. NC-coated paper was chemically vapor redeposited with trichloro-(1H,1H,2H,2H-tridecafluoro-n-octyl)silane (FOTS). The paper obtained exhibited superhydrophobicity and oleophobicity, repelling both polar and non-polar liquids, and the contact angles of water and n-hexadecane were 156° and 144°, respectively (Phanthong *et al.* 2016). Another gas technique used to hydrophobise nanopaper is ALD (Shim *et al.* 2013; Nam *et al.* 2020). Li *et al.* (2020) modified the surface chemistry, wetting properties, and wet strength of NFC nanopaper using a few cycles of metal oxide ALD to increase the hydrophobicity of the NFC nanopaper, presumably by increasing the amount of hydrocarbons on the surface of the nanopaper (Li *et al.* 2020).

Modified Nanocellulose-additive Used as Wet End Additive in Papermaking

Regarding cationic additives in the paper industry, cationic cellulose can be attached to long fibers and fines *via* hydrogen bonding and electrostatic attraction, hence increasing wet web strength and structure tightness (Lu *et al.* 2020b). Aguado *et al.* (2017) studied the application of cationic cellulose to flocculate fillers in papermaking. It has been demonstrated that the water-soluble cationic cellulose compounds can help in the flocculation of papermaking fillers and the flocculation capacity was highly correlated with the DS of cationic cellulose. Their best results came from the flocculant with the highest degree of polymerization and charge (Aguado *et al.* 2017). Li *et al.* (2015b) carried out cellulose etherification with CHPTAC while in NaOH/urea aqueous solution. The performance of this cationic cellulose was assessed as a modifier of GCCate, as well as based on the influence of the modified fillers on paper characteristics. Their results demonstrated that surface modification of GCC fillers with cellulose-CHPTAC increased filler retention and decreased the harmful effect of filler addition on paper strength (Li *et al.* 2015b). Gao *et al.* (2016) synthesized cationic NFC with 2,3-epoxypropyl trimethyl ammonium chloride (EPTMAC) for use as an additive in the papermaking process. They demonstrated that the addition of CMCF had a positive effect on the bulk of paper sheets. This was due to the fact that fibrils can serve as binders between fibers and fines, acting as a physical filler. They also suggested that it has the potential to be used as a strength additive to increase the tensile index of paper sheets (Gao *et al.* 2016). The effectiveness of an oppositely charged ion can increase dramatically as the number of valence charges increases (Hubbe 2014). Lua *et al.* (2020) discussed this issue by comparing varying cationic charge densities of CNFC as excellent additives in the papermaking process to achieve a wet web with improved performance. They concluded that CNFC, particularly those with a higher charge density, act as effective fines flocculants and create a "bridging" effect between pulp fibers

and the CNFC-Fines complex, resulting in increased wet-web strength (Lu *et al.* 2020b). The fact that high-charge cationic polymers typically exhibit high-affinity adsorption characteristics is one of the reasons why they are highly regarded by papermakers for a variety of applications (Wågberg 2000).

The DS is the next important aspect to consider, since recent research has shown that the DS of cellulose is substantially related to the resulting properties. It has been found that if the DS increases in the NC cationization process, the mechanical strength of fiber networks created as a result of applying high DS modified NC additives will improve (Zhang *et al.* 2021c). Since periodate oxidation produces reactive intermediates that are highly soluble in a wide range of solvents, this intermediate helps to boost the DS. Then, these high DS products can be used as a wet-end additive to improve mechanical strength qualities. Sirviö *et al.* (2011) synthesized cationic cellulose compounds by cationizing DAC obtained by periodate oxidation, since the aldehyde functionalities have high reactivity with respect to subsequent derivatization. This cationized cellulose has been used to conduct research using water-soluble CDAC as a biopolymeric flocculation agent in papermaking. Their findings indicate that it has a high potential for use as new, environmentally friendly flocculation aids (Sirviö *et al.* 2011). Campano *et al.* (2019) also synthesized CNCC through periodate oxidation and cationization. Then they investigated the ability of CNCC to flocculate the filler (kaolinite). They believed that CNCC has the potential to replace traditional wet-end retention aids, not only because it avoids the negative effects of traditional polymers on paper mechanical strength, but also because it allows for fine-tuning the characteristics of the flocs formed to meet industry needs by carefully varying the amount of CNCC added over a large number of effective dosage intervals (Campano *et al.* 2019).

The next suggested strategy for increasing the DS is assisted DES, which has been employed in paper manufacturing. It is critical to note that a simple and inexpensive route to cationic NC *via* DES can significantly expand their application range. Suopajarvi *et al.* (2017) reported that choline chloride-urea DES pretreatment significantly improved the nanofibrillation of cellulose board pulp grades. The properties of samples were investigated as paperboard strength-enhancing additives. Improved board handsheet qualities indicated that DES-treated NC may be used as a board strength enhancer (Suopajarvi *et al.* 2017). Zhang *et al.* (2021b) created the CCNC in two steps, starting with periodate oxidation and ending with cationization with DES, which was synthesized using aminoguanidine hydrochloride and glycerol. They achieved enhanced water resistance in self-assembling nanopaper by complexing oppositely charged CNC and NFC. They also noticed that not only did electrostatic complexation facilitate the fabrication of NC nanopapers by significantly reducing draining time, but it also improved the mechanical properties of NC sheets (Zhang *et al.* 2021b).

Benefits and Consequences of Various Approaches for NC in Papermaking

Reflecting on the current global environmental situation, it is critical to look for sustainable sources of raw materials to help us break our reliance on fossil fuels. Those serving the paper industry are responsible for developing substitutes for non-ecofriendly materials such as styrene-butadiene latex and C-PAM, as well as other conventional petroleum-based synthetic polymers that are commonly utilized in paper mills. As it has been already discussed, bio-based polymers including cellulose have potential to reduce environmental burdens, but substantial capital requirements and the paper industry's inability to price above marginal cost limit their potential (Sharma *et al.* 2020).

On the other side, the runnability also is difficult to achieve both during cellulose manufacture and during application as a wet end or dry end additive (Yang *et al.* 2020a). So, it has been suggested to minimize the runnability issues by using modified NC as an additive. There are already a variety of methods for functionalizing cellulose at the lab scale, but additional research is needed to validate these technologies for real-world applications and eventual commercialization. Thus, it is critical to choose the technique of modification with an inquisitive attitude; if the method can benefit from low cost, ease of operation, absence of toxicity, and ease of access, and can be done continuously, then application and commercialization of these types of additives will be more appealing. To meet these objectives, a variety of methods have been outlined in this paper. Apart from the information provided here in earlier sections, each treatment exhibits some advantages and disadvantages that must be considered when determining the most appropriate methods to be used in the paper manufacturing process.

The following summarizes the benefit and drawbacks of the above-mentioned approaches for obtaining modified NC as an additive suitable to be use in the paper production process:

- Surfactant adsorption is a relatively environmentally friendly and cost-effective method for making cellulose more hydrophobic. While this type of NC can be used in papermaking, there are several considerations to keep in mind. To modify the NC, a large amount of surfactant is required because detectable amounts of surfactants are not absorbable (Kaboarani and Riedl 2015). As a result, if some of the surfactant goes to the furnish, it makes the foam stable and hinders inter-fiber bonding, and it reverses the effects of hydrophobic sizing agents. There is also a minor decrease in film wettability due to the process of adsorption (Hubbe 2014).
- The silylation process creates an environmentally friendly paper packaging material that is easy to manage and has a high barrier performance. However, there is a trade-off between cellulose crystallinity and silylation extent in order to get a higher degree of hydrophobization (Ghasemlou *et al.* 2021). Moreover, hydrophobic groups in silylation may not be effective to obstruct water vapor transport due to the short length of the hydrophobic alkyl groups. On the other hand, such a system could serve as an excellent coating for water barrier qualities (Yook *et al.* 2020).
- Plant oils are abundant, inexpensive, and non-toxic basic ingredients. Onwukamike *et al.* (2018) suggested that modified NC could be coated with trans-esterified oil or bio-epoxy oil and then used as a paper coating material over kraft paper because trans-esterified oil and bio-epoxy oil have a lower viscosity and a lower surface tension than their corresponding source oils, which results in improved wettability and the formation of a uniform film on the surface (Onwukamike *et al.* 2018). Linseed oil has typically been used in a wide range of industrial coating formulations; nonetheless, linseed oil-based coatings are fragile and turn yellow over time (Tambe *et al.* 2016). As a result, if linseed oil is used as a paper coating substance, the color of the oil-coated paper will change. The high viscosity of oil requires the use of organic diluents to be used as coating substances, which extends the curing time needed to eliminate excess solvent from the system. The use of catalysts is also a part of this method. Because of these constraints, the use of linseed oil and other highly unsaturated natural oils in the paper coating industry

has been restricted. The use of bio-epoxidized oil as a coating for paper might then be recommended. As a result of the several steps required, the cost of the coating goes up significantly (Tambe *et al.* 2016). It is worth noting that the usage of certain epoxy derivatives (such as bisphenol-A diglycidyl ether, bisphenol-F diglycidyl ether, and novolac glycidyl ether) has been restricted in materials and objects intended to come into food by EU regulation. The epoxy derivatives mentioned above are synthetic epoxy resins, and efforts are being made to replace them with bio-based epoxy resins (Altuna *et al.* 2014; Islam *et al.* 2022).

- The modification by bio-polyphenol (tannin) and then coating of paper increase the antioxidative and UV-blocking properties of the handsheets (Ji *et al.* 2020). However, there are certain drawbacks to this strategy. For example, the extraction methods to obtain polyphenols necessitate complex analytical techniques and expensive equipment (Missio *et al.* 2017); thus, a simpler approach to fractionation must be considered to reduce the cost of cellulose modification using this kind of material.
- Chemical vapor deposit (CVD) as the next more environmentally friendly method benefits from the excellent interface and surface morphology, growth of complex heterostructures with numerous layers, growth on patterned substrates, multiple wafer scale-up, and high layer purity (Pessoa *et al.* 2015). Vapor deposition benefits from excellent interface and surface morphology, multiple wafer scale-up, and high layer purity. However, the major limitation of CVD is that it frequently requires a high temperature to convert precursors to reactive products capable of forming thin films (Sun *et al.* 2021b). So, this method is not suitable for heat-sensitive materials such as paper. In terms of ALD, it should be noted that today's batch reactors with vacuum chambers are mostly used for paper coating. However, roll-to-roll reactors and atmospheric processes are being developed by different research groups and equipment suppliers. The drawback of this kind of reactor is that the depositions take place at high temperatures. This had a considerable impact on the topography and morphology of the extrusion coating polymers (Kääriäinen *et al.* 2011).
- Plasma treatment is the next low-cost, environmental-friendly and simple method to modify cellulose (Vizireanu *et al.* 2018). Plasma treatment of cellulose has been thoroughly investigated for a variety of applications in the textile and paper industries, and also other sectors, including cleaning, sterilization, activation, and enhancement of hydrophilicity or hydrophobicity. The "corona" method is insufficiently feasible and effective for high-speed processing of porous materials such as textiles and paper. The Diffuse Coplanar Surface Barrier Discharge plasma system has been successfully demonstrated for surface activation of paper. Due to the very high plasma power density and ambient air as working gas, the Diffuse Coplanar Surface Barrier Discharge plasma system might be applicable for in-line paper coating technology with a standardly high manufacturing speed (Černák *et al.* 2009). However, despite a good potential using it within papermaking, it's worth noting that there are difficulties among researchers in interpreting and comparing the experimental results obtained through the use of plasma (Bahrami *et al.* 2020).
- Cationic NC derivatives were put through a variety of tests to see whether they were suitable for usage as additives. Etherification, as a common method for

cationization of a surface of NC, has disadvantages including taking a long time for the reaction and difficulties to achieve a high DS (Li *et al.* 2021b; Rol *et al.* 2019a). The mechanical strength of paper and the DS of modified NC used as additives in papermaking are highly correlated. Cationic additives perform better by increasing the bonded area, which results in an increase in the sheet's density (Lu *et al.* 2020b). Thus, the effect of cationic NC as a dry strength additive is enhanced when the additive contains a greater number of positions capable of bonding with the fiber in furnish.

- Cationic cellulose compounds with a high cationic charge have been found to perform better (Vuoti *et al.* 2018). So, periodate oxidation treatment can activate the C2-C3 position and then act as an intermediate for the further cationization to improve the charge density and DS, and when used as an additive in papermaking, it contributes more bonding, resulting in paper with superior mechanical properties. However, this method is defined on a case-by-case basis. This means that it is a combination of the starting hierarchy and oxidation scenarios, and as a result, it cannot be generalized; nonetheless, research into generalizing this method will continue (Nypelö *et al.* 2021).
- The next strategy for improving the performance of cationic cellulose in papermaking is to dissolve it in an ionic liquid before modification in order to activate the hydroxyl group. Ionic liquid as green solvent has a variety of appealing properties, including chemical and thermal stability, non-flammability, and immeasurable low vapor pressure. However, the industrial manufacturing and expense of ILs currently preclude their use in the paper industry (Zhu *et al.* 2006).
- Deep eutectic solvents (DES) is the next green solvent, and it can be used to improve the performance of cationic cellulose as a paper additive. The previously mentioned modification methods had a number of limitations, including a high cost and the need for special grade pulps. So, use of DES modified cationic cellulose promotes possibility to achieve a cost-effective modification of high consistency mixing while using industrial-grade cellulose pulp that does not require the use of harmful solvents, expensive cellulose grades, or cellulose solubilization. Many of these issues are overcome when deep eutectic solvents are used for the modification (Vuoti *et al.* 2018). The use of DESs could be a promising method for inducing the swelling of cellulose, which would enhance the chemical reactivity of that material. More innovation and in-depth research are unquestionably required.

CONCLUSIONS AND FUTURE PERSPECTIVES

Green chemistry involves application of processes that remove or greatly reduce harmful compound intake and output. This approach is becoming popular as a strategy for environmental sustainability. In this instance, there is a growing tendency toward developing techniques for nanocellulose (NC) modification using green technologies. These modifications allow for the introduction of novel capabilities not found in native NC, enabling them to be applied in a broader range of industries. In the paper industry, as NCs demonstrate advantageous properties such as high surface area, tailorable surface chemistry, anisotropic shape, and superior mechanical and gas barrier properties, researchers are enticed to use them to improve the mechanical and barrier properties of paper. However, the manufacture of NC-based paper has encountered a number of difficulties such as a decrease in dewatering rate in the wire section, which raises the cost of operation. There are also concerns with the wettability and water vapor barrier in a coating due to the hydrophilicity of NC. The first section of this review paper discussed surface modification of NC with the goal of improving the water barrier properties of NC using green eco-friendly methods. The procedures outlined in the section “Modification Techniques of Nanocellulose” have the advantage of requiring the least amount of chemically hazardous solvents and allowing for the following scenario: In the first scenario, the formation of a chemical bond reduced the surface hydroxyl groups and hydrophilicity. In the second case, non-covalent surface modification or surface deposition aids in improving surface hydrophobicity. In the final scenario, changing the morphology of the surface, such as creating an etched surface through plasma treatment, can help to change the hydrophilicity behavior of the surface. Thus, modifying the morphology, functional group, and surface with the least amount of hazardous solvents can improve the water barrier qualities of NC.

Another series of modification techniques have been summarized in this work with the aim of creating cationic sites on the surface of NC in order to overcome the dewatering rate limitation and make them more useful in the paper industry. If NC is used as a wet-end additive to improve the mechanical properties of paper, an issue with a reduction in dewatering rate will emerge. One of the major causes of this problem is the plugging of drainage channels in the wet web by NC. To address this issue, the addition of more active binding sites, particularly those with cationic charges, has been proposed. The addition of a cationically charged group to the hydrophilic backbone of NC can result in the production of amphiphilic NC, which has the potential to increase the wet-web strength through adsorption-flocculation effects on fines and fiber pulps. As the number of valence charges increases, the effectiveness of an oppositely charged ion increases dramatically. Therefore, the methods produce a greater charge density, which works as an effective fines flocculant and bridge between pulp fibers and fines, leading to increased wet web strength with a low dewatering rate impact. It should be emphasized that the majority of the treatments discussed and their effects on the paper application were carried out on a small scale by the various cited researchers. As a result, despite the benefits of these modified NC in the paper sector, scaling up is still necessary. This transition from laboratory to upper scale needs to be facilitated in order to be able to use the aforementioned approaches to track the evolution of the paper industry and identify the best answer for the technical, processability, formation, and economic issues that the paper industry faces. To our mind, future research work on surface chemical modification and application in the paper industry should be focused on five primary objectives: i) research and development of new, low-cost, and environmentally friendly methods of NC modification; ii) scale-up research from batch to

continuous procedures to evaluate laboratory results and their potential commercial applications; iii) a study on the efficiency of hydrophobized/functionalized NC in the paper industry; iv) analysis of the environmental impact of NC hydrophobization/functionalization and their application in paper production; and v) A study on cost estimation (e.g., energy consumption and building the facility on an industrial scale) and technical feasibility. The authors hope that this study will inspire researchers' interest in improving NC characteristics, thereby expanding their industrial applications and supporting the sustainable utilization of renewable resources.

Competing Interests Declaration

There are no conflicts of interest in the submission of this manuscript, and all authors have given their approval for publication.

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