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REVIEW PAPER

Nanocellulose: its applications, consequences and challenges in papermaking

Atanu Kumar Das[1] · Md. Nazrul Islam[2] · Md Ashaduzzaman[2] · Mousa M. Nazhad[3]

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Abstract
This paper has aimed to bring out the state-of-the-art information about nanocellulose, its application in papermaking process, effect on paper properties and challenges. In papermaking process, nanocellulose is used as bionanofiller and bionanocoating material. The main objective of considering nanocellulose as a bionanofiller is to retain the strength properties after using inorganic filler, such as, GCC (ground calcium carbonate) and PCC (precipitate calcium carbonate); nanocellulose also helps to increase the filler content in paper sheet without loss of paper strength. That's why, the application of nanocellulose as filler and coating material can solve the issues of using inorganic filler in papermaking industry. Though the production of nanocellulose needs higher energy, which increases the production cost in papermaking process, it can be minimized using different advanced technologies, such as, chemical and enzymatic pretreatment. Additionally, the incorporation of nanocellulose in recycled and virgin fiber increases some physical and strength properties of the paper. Furthermore, the increment of filler content in paper sheet due to incorporating nanocellulose can also reduce the amount of fiber in papermaking process, and it reduces the cost of fiber, which can mitigate the drawback of paper production cost because of adding nanocellulose partially. Therefore, nanocellulose has the potential to use for making value added paper and paper products to compensate the production cost.

Keywords Cellulose nanomaterials · Bionanofiller · Bionanocoating material · Paper properties · Value added paper

Introduction

Nanotechnology deals with the nanoscale of artificial and natural structures, which is about 1 to 100 nm. One nanometer is 10^{-9} of a meter, and in an ordinary solid, it is around the distance from one end to the other of a line of five neighboring atoms [1]. For getting idea of size of the nanoscale, 10 hydrogen atoms set in a row make 1 nm wide; a sheet of paper is about 100,000 nm thick [2].

Nanotechnology is anticipated to be an acute driver of global economic growth and development in the near future. By this time, this broad multi-disciplinary field is showing the scope of materials, devices and systems examined, engineered and fabricated at the nanoscale [3]. The main objective of nanotechnology is to modify materials to achieve a special purpose. In the contemporary years, nanotechnology as a novel tool is applied in many areas like pulp and paper [4]. Nanotechnogical applications are the utmost in the packaging sector, followed by electronics, pharmaceuticals, plastics, cosmetics and others including paper coating [5]. Packaging products need higher strength, water and fire resistance properties besides these antimicrobial property is important for packaging products especially for food packaging. These demands are creating opportunities to apply nanotechnology in the pulp and paper industry [3, 6–9]. The most plentiful natural polymer in the world is cellulose, demonstrating 1.5×10^{12} tons of total biomass production per year in the planet [10]. For advanced sustainability and renewability, high-performance materials derived from cellulose-based materials can fulfill the demand of modern society. Cellulose nanomaterials (CNs) among the cellulosic materials have confirmed the potentiality of utilization for

✉ Atanu Kumar Das
atanu03ku@yahoo.com; atanu.kumar.das@slu.se

1 Department of Forest Biomaterials and Technology, Swedish University of Agricultural Sciences, SE-90183 Umeå, Sweden

2 Forestry and Wood Technology Discipline, Khulna University, Khulna 9208, Bangladesh

3 Pulp and Paper Centre (PPC), The University of British Columbia, Vancouver, BC, Canada

value added materials over the past period. CNs are possible to use for some specific purposes, which are thought to be impossible for cellulosic materials. CNs as a novel type of cellulose-based materials that are influencing cellulose technology, science, and product development for the next generation of renewable or sustainable products [11]. Properties of nanosized particles are attributed to large surface area and ability to form more hydrogen bonds resulting in strong network. CNs possess some special properties, and these are high aspect ratios and mechanical properties, low thermal expansion and density, large specific surface area, barrier properties, biocompatibility and biodegradability. Even, additional functionalities can be obtained from modifying surface-accessible hydroxyl groups of CNs by chemical [12–14].

CNs are extracted from sustainable and renewable raw materials. Products made from CNs have low environmental impact, less animal or human health hazardous, and low safety risks [11]. On the other hand, CNs increase the density of paper [15–17], and work as a barrier of air [15, 18–21], oxygen [20–22], and oil [21] as well. The pore is blocked closely in the matrix that causes the poor permeability [18]. CNs increase the hydrogen bond and bonding area, which are responsible for increasing the paper density [20] and strength [15, 24–33]. Even, CNs increase bending stiffness [32, 34] properties of paper. The bridging capability with neighbor fibers and formation are increased by the addition of CNs; these enhance the strength properties of paper through good fiber-to-fiber bonds [35, 36]. Considering all these special properties, CNs have been recommended for paper additives and coating material [37–40].

Scientists are working to apply CNs in paper for achieving different properties. However, this review article has been written to gather all related information about the applications and consequences of CNs in papermaking. The challenges for application in papermaking have been pointed out in this review article.

Nanocellulose and its classification

A cellulosic material having dimensions in the range of nano-scale is often named as nanocellulose. The particle diameter or width of a cellulosic material between 1 and 100 nm is called as nanocellulose [1]. Nanocellulose (NC) is classified into cellulose nanostructured materials and cellulose nanofibers; both of them are derived from either plant or non-plant [41]. Sub-groups of cellulose nanostructured materials are cellulose microcrystal (CMC) or microcrystalline cellulose (MCC) and cellulose microfibril (CMF) or microfibrillar cellulose (MFC) [41]. CMC or MCC is nanostructured cellulose material, which contains aggregated cellulose bundles having different degree of cellulose crystallinity [41]. On the other hand, CMF or MFC is cellulose nanofiber that contains remaining hemicellulose and several elementary fibrils having amorphous and crystalline regions (TAPPI WI 3021). According to Turbak et al. [37] and Herrick et al. [42], mechanical disintegration helps to produce heterogeneous pulp fiber particles called MFC. Again, subdivisions of cellulose nanofibers are nanocrystalline cellulose (NCC) or cellulose nanocrystal (CNC) or cellulose nanowhiskers and cellulose nanofibril (CNF) or nanofibrillar cellulose (NFC) [43]. NCC or CNC or cellulose nanowhisker is that type of cellulose nanofiber, which has pure structure of crystallinity [41]. CNF or NFC is one type of cellulose nanofiber, which possess amorphous regions and crystalline regions [3, 38]. Most researchers have denoted MFC or CMF as NFC or CNF [43]. The structure of MFC and NFC are like spaghetti containing both the crystalline and amorphous regions [43].

Application of nanocellulose in papermaking

Nowadays, application of nanocellulose is broadening and its uses providing different types of benefits in terms of quality [4]. Addition of CNFs can be used either in the paper making slurries in the beginning or onto the final paper as a bionanocoating material, since they are extracted from cellulosic source and present as aqueous suspension [44]. Its application in papermaking industry has shown a new dimension in the history of papermaking.

Application of nanocellulose as an additive in papermaking

Nanocellulose can be used as bionanofiller with or without retention aid during papermaking process [33]. It can be mixed earlier with a fixed furnish component, like, inorganic filler or long fiber so that it can deposit on the surface of furnish component [33]. CNF can work as dry strength additive in poorly bonded sheets made of mechanical, recycled and unbeaten chemical pulps [33]. To modify the paper properties, CNFs can be used with fibers along with different types of additives, i.e., fillers, cationic polymers [33, 44]. For instance, the addition of low concentrations of CNFs, i.e., lower than 5% of pulp increase the tensile strength [44]. Chemically modified CNF helps to improve the wettability, compatibility, hydrophobicity and possibility of interaction of nanofibers in paper matrix, and it changes the properties of cellulose [45, 46]. Missoum et al. [46] have produced CNF from enzymatic pre-treated mixed bleached hardwood (60% Spruce and 40% Pinus) by mechanical disintegration in a Masuko Grinder and then, by nanoemulsion concept of CNF, Alkyl Ketene Dimer (AKD) has been grafted in CNF to modify it. Authors have introduced modified CNF into the pulp slurry (Fig. 1), and it results the highest retention rate. AKD-modified CNF can be used to produce water repellent paper with higher mechanical properties, since CNF works as a binder and it packs the fibers [46].

Hubbe [47] have investigated the effect of acetylated CNF on bleached softwood pulp using retention agent C-PAM, and CNF obtained from ultra-fine friction grinder has been modified by heterogeneous acetylation process for this study. In this study, non-acetylated CNF has contributed to increase density, air resistance, burst and tensile strength of paper without considerable reduction of water absorption, while acetylated CNF has increased water repellent ability and it reduces water absorption around 23%. Surface modified CNF by lactic acid can increase the capability of drainage without compromising of mechanical and physical properties of paper [48].

Application of nanocellulose as a coating material in papermaking

As a coating material, CNF can solve the problem of dewatering, because it is added after papermaking [36]. It can be used as transparent and biodegradable packaging films having higher barrier properties [36]. The amount of coating material depends on the technique of coating [33]. There are different types of techniques like spray, bar, roll and size press coating to apply nanocellulose on the paper's surface [33]. Lavoine et al. [49] have used bar coating and size press coating to apply microfibrillated cellulose (MFC) on paper's surface. According to result, coating weight of 7 and 14 g/m^2 are for 5 and 10 layers MFC, respectively, for bar coating, but size press can produce coating weight of 3 and 4 g/m^2 by 5 and 10 layers MFC, respectively. Aulin et al. [21] and Aulin and Ström [50] have coated MFC by a rod coater for sheets and this has improved barrier properties, such as, oxygen permeability and oil resistance significantly. By this technique, the achieved coat is 1.3 g/m^2 for the unbleached paper, while it is 1.1 g/m^2 for greaseproof paper. The result shows that the reduction of air permeability for the unbleached paper is from 69,000 to 4.8 nm/Pas, whereas it is from 660 to 0.2 nm/Pa for greaseproof paper.

Syverud and Stenius [20] have studied the effect of CNF prepared by shearing disintegration coating on the unrefined softwood pulp. This coating results in higher tensile index for coated with CNF sample (40 Nm/g) than that of uncoated sample (35 Nm/g). At the same time, it improves the barrier properties of air permeability significantly from 360 to 6.5 nm/Pas for the samples coated with 8% CNF [20]. CNF reduces the surface porosity, which helps to decrease the air permeability [20]. Mixed coating of CNF and shellac on paper and paperboard can decrease the air permeability, oxygen and water vapor transmission considerably for both paper and paperboard [51].

Lavoine et al. [52] have assessed the effect of MFC coating on bending stiffness, compressive strength and barrier

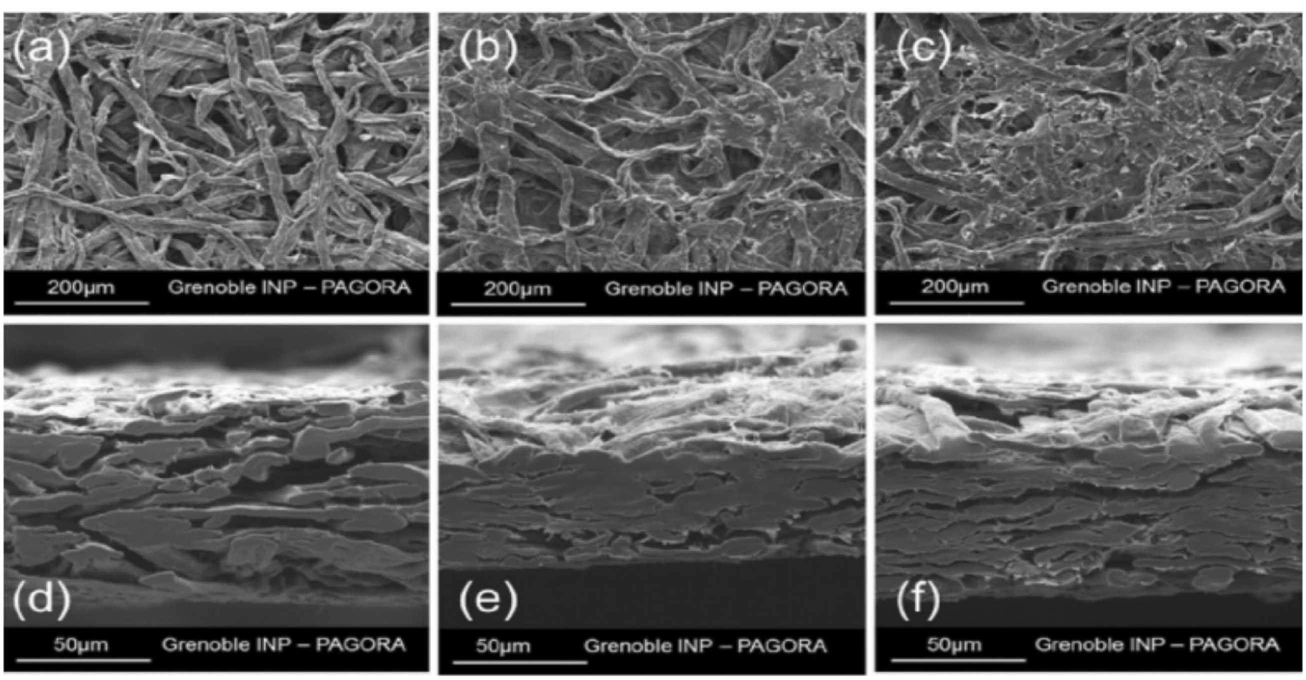

Fig. 1 SEM images of surface paper reinforced **a** without CNF, **b** with CNF, **c** with modified CNF and cross section of **d** without CNF, **e** with CNF and **f** with modified CNF [46]

properties of cardboard and used only bar coating method to apply MFC coating on cardboard. Results show that bending stiffness and compressive strength have been increased 30% in machine direction. Application of MFC increases cardboard's thickness that causes higher bending stiffness. Conversely, barrier properties of cardboard are poor and water absorption is higher for CNF-coated cardboard in this study.

CNF foam is suggested to apply on surface; it helps to obtain low coating weights with thicker coating layer [33]. Kinnunen et al. [53] have investigated the performance of CNF foam as a coating to achieve a thicker coating layer with low coating weights. Authors have mixed 2.9% CNF and an anionic surfactant together in a foam generator with compressed air to prepare foams containing 80–95% of air. This technique lowers air permeability and contact angle, but it increases smoothness [53].

Chin and Feng [54] have developed the patent US 20140050922 A1 for coating based on CNF, pigment, latex and an auxiliary additive, such as, starch, lubricants, defoamers, etc. in water suspension. This type of coat can improve the fracture resistance and smoothness of the coated paper. It also decreases the risk of heavy coat cracking and mechanical failure of the coat, like, tearing, spallation, blistering, and peeling away from the paper substrate. According to authors, this type of coated paper can be used for high-grade printing paper and wrapping paper of food and cosmetics.

Paper sheet properties

Nanocellulose has the effect on physical and mechanical properties of paper. Use of nanocellulose as bionanofiller and bionanocoating material improves the both types of properties, i.e., physical and strength properties. In this section, the effect of nanocellulose on paper properties has been discussed separately.

Effect of nanocellulose as a filler material on paper properties

Nanocellulose can be used as a filler material in papermaking industry single or with other additives [33]. This improves the physical and strength properties of paper.

Density

Researchers have observed the enhancement of density by the addition of CNF. Addition of 4% CNF in TMP sheets increases density of 4–30% [15]. Sehaqui et al. [17] have found that increasing in density for softwood kraft pulp sheets in combination with 10% homogenized CNF is 30–50%. In the same trend, density of softwood pulp sheets has been increased by 10 and 20% for the addition of 7 and 20% grinder produced CNF, respectively [16]. CNF increases density of pulp sheets in two ways [23]. Firstly, free CNF particles reduce water meniscus's radius, which increases the pressure difference between the water phase and surroundings during dewatering of the sheet, and fibers become closer to consolidate a sheet by pressing. Secondly, attached CNF as a layer on the fiber surfaces increases the contact area and the number of hydrogen bonds. Thus, fibers make contact each other by establishing bonds during wet pressing, and density is increased accordingly [23].

Permeability

Previous investigators have registered that air permeability is greatly affected by CNF. According to the report of Taipale et al. [29], air permeability has been dropped from 1450 to 450 ml/min with the increasing dosage of CNF from 0 to 30 wt%. Air permeability is reduced gradually by the addition of CNF up to 4 wt% [15]. Subramaniam [18] has found similar result and author has observed that increasing the dosage of CNF with high filler loading decreases the air permeability promptly. Author has also stated that fines can block the connectivity of the pore structure by bonding with matrix closely.

Breaking length

The addition of nanocelluse can increase the breaking length. The breaking length depends on the percentage of nanocellulose content in the paper sheet [55]. The higher percentage of nanocellulose provides the higher breaking length of the paper sheet [55].

Strength properties

CNF enhances strength properties of handsheets, which have been proved by many researchers [15, 18, 24–31]. Eriksen et al. [15] have observed that tensile index of TMP hadsheets has been increased significantly after adding 4% CNF along with filler particles to handsheets. Ahola et al. [56] have stated that deposition of CNF using a cationic polyamide-amine epichlorhydrin (PAE) by a bilayer bound technique on the fiber surface can increase both dry and wet tensile strength. Taipale et al. [29] have added 3% CNF in combination with 1.5% cationic starch to beaten softwood pulp and observed the increment of tensile strength of handsheets. Addition of fractioned CNF using gel method to kraft pulp sheet enhances tensile index [57]. Fractionating increases specific energy, which increases the tensile strength of CNF-containing paper [57]. Ahola et al. [25] have found that tensile strength can increase 100% by the addition of 6% CNF to fines free slightly beaten softwood pulp. Eriksen et al. [15] have observed that different CNF qualities increase tensile strength of TMP sheets 7 to 34%. Highly filled papers, mechanical and recycled pulps have good response to the effect of CNF on tensile strength whether well-beaten chemical pulp sheets show little response [33].

Factors affecting the tensile strength of a paper are length, shape, orientation, distribution of fiber, bonded area and bond strength [58]. The effect of CNF on tensile strength depends on amount of CNF, intensity of fibrillation, degree of pulp refining, type and strategy of addition of retention agent [15, 29, 35, 44, 56]. The increment of tensile index is proportional to the amount of CNF [36]. Similar trend has been proved by many researchers [29, 35, 56]. According to Eriksen et al. [15], fibrillation degree of CNF at a certain level also increases the tensile strength. CNF boosts specific surface area, which increases the formation of fiber-to-fiber bonds (Fig. 2) and consolidates the paper structure [36]. CNF can create bridge among neighboring fibers and forms entangled network, which can increase the bonded area. Furthermore, fibers have great affinity for CNF networks due to having similar structure for both of them. This consequence increases density, rigidity, overall tensile strength and Z-strength [35, 36]. In addition, Taipale et al. [29] have stated that CNF along with cationic starch increases fiber bonding. CNF with right retention aid can deposit on fiber surface prior to sheet formation, which softens the fiber surface and thus, increases the bonding area. In this way, CNF works as a dry strength polymer [33].

Optical properties

Researcher have also observed the effect of CNF on optical properties of paper. Eriksen et al. [15] have observed that addition of 4% CNF in TMP sheets reduces light scattering coefficient 2–5 m^2/kg. CNFs increase sheet density and bonded area, which cause reduction of specific surface area and light scattering coefficient. For this consequence, brightness and opacity of sheet are reduced [33].

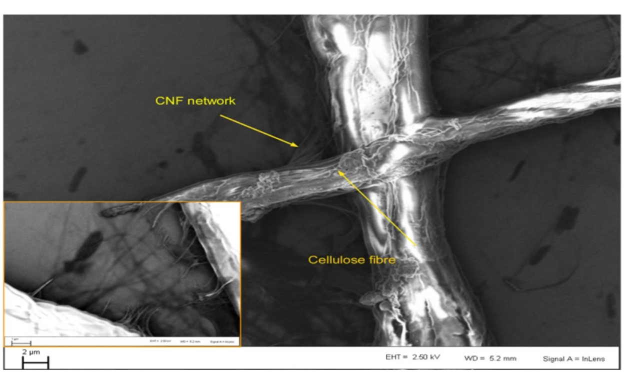

Fig. 2 FE-SEM image shows the function of CNFs is bounded to larger cellulose fibers. The smaller picture represents a zoom in of two adjacent fibers with a CNF network partially filling up the gap between the fibers [36]

Effect of nanocellulose as a coating material on paper properties

The scope of the feasibility of CNF as paper coatings has been studied as well [33]. A thin layer of CNF is enough to modify the surface properties, but a thicker layer is required for barrier application [33]. CNF can work as a barrier material during using as a material of film or paper coating [33]. There are some strength and barrier properties obtained after using CNF as follows:

Strength properties

The applied forces crack the paper and break the paper gradually. Using CNF as a coating material on the paper's surface can increase the tensile index and strength properties as well [33]. Syverud and Stenius [20] have noticed that tensile index can increase 35 to 40 Nm/g after coating with CNF layer. Brodin et al. [33] have stated that nanofiber makes huge number of inter-fibrils bonds, because it has more fibrils in comparison to pulp fibers. These bonds enhance the strength properties of paper.

Bending stiffness

CNF as a coating has an effect on the bending stiffness of paper. CNF coated cardboard or paperboard shows higher bending stiffness [52]. The paper coated with five layers MFC by bar coating can improve 50% bending stiffness [49]. Ridgway and Gane [32] have mentioned similar result for the bending stiffness, and it has been increased slightly for the CNF coated base paper. The increased number of bonding [33] may gear up the bending strength of paper.

Smoothness

CNF has ability to increase the surface smoothness, and the application of CNF coating after pre-coating improves the surface smoothness of CNF coated paper [32]. Pores created by fiber-to-fiber crossing are filled by CNF and thus, it increases the smoothness of paper [33].

Air barrier

CNF used as a film or coating has effect on air barrier property and it can reduce the air permeability. The air barrier properties can be improved by 90% when the paper is coated with five layers MFC by bar coating [49]. Investigation of Syverud and Stenius [20] informs the effect of coat weight on air permeability. Authors have used coat weight 2 to 8 g/m^2 on the base paper, and the air permeability has been decreased from 65,000 to 33,000 nm/Pas for the base paper. CNF films can reduce the air permeability up to 10 nm/Pas [33]. Aulin et al. [21] have applied a single layer of carboxymethylated CNF coating on wrapping paper and greaseproof paper. For the wrapping paper, the air permeability has been decreased 69,000 to 4.8 nm/Pas for the coat weight 1.3 g/m^2, while the coat weight 1.8 g/m^2 decreases air permeability up to 0.3 nm/Pas. On the other hand, the air permeability has been decreased from 660 to 0.2 nm/Pas with the coat weight of 1.1 g/m^2 for the greaseproof base paper. The pores of paper are field by CNF [33], which works as a barrier for air permeability.

Oxygen barrier

Researchers have reported the effect of CNF films, laminates and coatings on the reduction of oxygen transmission rate (OTR) or oxygen permeability [19–21]. Syverud and Stenius [20] have measured OTR values 17–18 ml/m^{2}day for mechanical treated CNF films with the basis weight of 17 and 29 g/m^2 at 0% relative humidity (RH) on top side and 50% RH on bottom side. Chinga-Carrasco and Syverud [22] have found lower OTR values 3.0 ml/m^{2}day for TEMPO-oxidized CNF films at 50% RH. Fukuzumi et al. [19] have also observed lower oxygen permeability for TEMPO-oxidized CNF than that of polylactic acid (PLA) coating.

Aulin et al. [21] have explained the higher OTR value, the higher RH. At higher RH, CNF–CNF hydrogen bonds are replaced by CNF-water hydrogen bonds, and fibrillar network is swelled. These cause higher oxygen transmission rate. CNF films can be used as an alternative of synthetic films, e.g., ethylene vinyl alcohol (EVOH) [19, 50]. AKD or alkyd resins can reduce the hydrophilicity of CNF film and coatings [19, 50].

Oil barrier

Oil resistance for some food packaging materials is obligatory. Aulin et al. [21] have penetrated castor oil and turpentine into CNF and non-CNF coated base papers. Authors have found that CNF coated base paper shows higher oil resistance property in comparison to non-CNF coated base paper. Even the higher coated paper has the higher oil resistance with less air permeability [21]. As pores are blocked by fine CNF [33], it reduces the oil transmission capacity of the paper.

Challenges of application of nanocellulose in papermaking

The main problem of the implementation of CNFs in paper making process is the reduction of dewatering [44]. The main reasons of dewatering problem are plugging of interfiber pores, increasing of capillaries required for water flow and reduction of sheet's permeability [29, 59]. Especially carboxymethylated nanofibers contribute more to reduce the drainability [36]. Drainage can be improved by selecting appropriate type and correct dosing of retention agents so that nanofibers can be adsorbed onto the fiber's surface [29, 56, 60]. However, present research is showing to overcome the problem [44, 48, 59, 61]. Huge energy requirement is the key constraint for the commercial production of cellulose nanofibers [62]. Enzymatic or chemical pretreatment prior to mechanical treatment has been proposed by researchers to reduce the energy consumption during processing [63–65].

Conclusions

Nanocellulose has been established as an effective additive to enhance the overall properties of paper. CNFs enhance the density and strength properties of paper, but these reduce the opacity and porosity of paper. These alternations can be adjusted by controlling the dosage and degree of fibrillation of added CNFs. CNFs also help to recover the strength properties of paper, which are lost due to using different types of minerals as fillers. CNFs are used as strength enhancer mainly for hardwood and softwood pulps. Incorporation of CNFs for enhancing strength properties is becoming popular for TMP, agricultural wastage pulp and recycled pulp. CNFs are more effective as strength enhancer for chemical pulp in comparison to TMP owing to having more fines in TMP. Another potential use of CNFs as coating material is showing new dimension in papermaking. It improves strength and barrier properties of paper.

The main challenge for application of CNFs is reduction of production cost as paper is lower cost product with high volume. Furthermore, runnability is challenging in terms of CNF production and for the paper machine. Drainage rate is reduced due to adding CNFs in paper suspensions. Drying cost is an important issue for papermaking and coating as CNF coating contains higher water. Researchers are working on the aforementioned issues. However, many researches are needed to reduce CNFs production cost and improve drying performance and drainage rate of paper obtained from using CNF additive as either filler or coating material. Controlling the rheological properties of CNF containing coating formulations should be taken under consideration as well.

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Compliance with ethical standards

Conflict of interest On behalf of all authors, the corresponding author states that there is no conflict of interest.

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