NANOCELLULOSE AND ITS POTENTIAL USE FOR SUSTAINABLE INDUSTRIAL APPLICATIONS

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***Abstract*— Nanocellulose (NC) and its wide applications have attracted high attention due to its desirable properties such as high surface area, extraordinary mechanical properties, high reactivity and easy modification of NC surface due to the presence of primary hydroxyl groups. NC also presents several environmental benefits, including high potential availability because its production is coming from natural sources, renewability and nontoxicity. This paper briefly summarizes some of the activities of the research group "Cellulose, Paper and Water Advanced Treatments" from Complutense University of Madrid that were presented in CAIQ 2019, including the main types of NC, the production processes and their characterization. Additionally, the most promising NC applications are described such as for paper and board, for wastewater treatment, food and cement-based materials. Moreover, a market perspective of NC is also presented.**

***Keywords*— Nanocellulose, applications, market perspectives, paper, water treatment, food, cement**

I. INTRODUCTION

Cellulose is a linear and natural polymer composed of β-β-D-glucopyranose units joined together by β-1-4-glycosidic bonds and it has an important structural role in animals and plants. Nanocellulose (NC) can be defined as cellulose in the form of nanostructures, which have at least one dimension between 1 and 100 nm (Blanco *et al.*, 2018). NC can be obtained using a top-down approach by defibrillation of cellulose fibers from different raw materials such as wood, plant fibers, tunicates or agricultural by-products. Chemical, mechanical and/or enzymatic pretreatments prior to mechanical defibrillation allow to facilitate the separation of fibers increasing the NC yield and reducing the cost (Mondal, 2017). Moreover, NC is also produced following a bottom-up approach in which bacteria produce glucose units that extruded out of their membrane and self-assembly forming the nanofibers (Campano *et al.*, 2016). NC has unique properties due to its combination of the nanoscale dimensions (e.g., high surface area, lightweight, stiffness, high strength) and inherent properties of the cellulose (e.g., biodegradability, renewability, sustainability, high potential availability) when NC is produced from natural sources such as wood, plants or agro-waste by products. In addition, NC has a high reactive surface of hydroxyl groups which is suitable for surface modification (Chin *et al.*, 2018).

Anselm Payen was the first to isolate cellulose from plants and various woods in the 19th century (Fisher, 1989). However, it was not until the last decade when NC has emerged as a promising material. From 2006 to 2018, the total number of scientific publications and patent documents with the search term "nanocellulose" was 3163, 2277, 1286 and 995 using Web of Science (WOS), Current Contents Connect (CCC), BIOSIS Previews or MELINE® databases, respectively (Fig. 1).

The number of publications since 2006 addressing NC, broken down by country, is shown in Fig. 2, with a clear leadership of China and the United States with 691 and 527 publications, respectively.

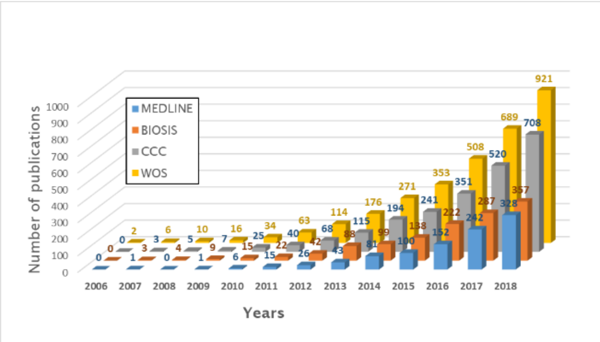

Figure 1. Number of publications with the search term "nanocellulose" using different databases (WOS=Web of Science; CCC= Current Contents Connect; BIOSIS; MEDLINE®).

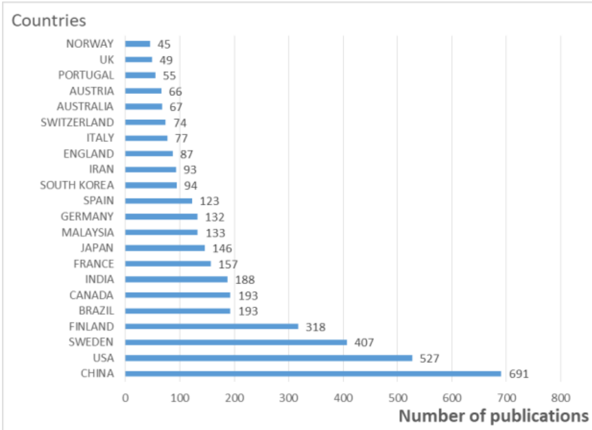

Figure 2. Number of publications related to nanocellulose per country from 2006 until 2018 (adopted from WOS database; search data: 10 September 2019).

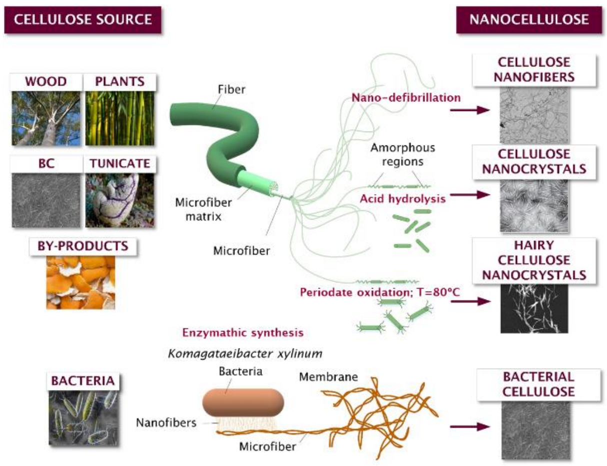

Figure 3. From cellulose to nanocellulose. Schematic diagram of possible cellulose sources to produce different types of nanocellulose. Adapted from Campano (2019).

II. NANOCELLULOSE

Cellulose nanofibers (CNF), cellulose nanocrystals (CNC), and bacterial cellulose (BC) are the main types of NC products which differ in their structure, functions, and production methods (Blanco *et al.*, 2018; Vallejos and Area, 2019). However, many terms have been used in the past to define and distinguish the different types of NC (e.g., microfibrillated cellulose, nanofibrillated cellulose, nanofibers, cellulose nanowhiskers, nanorods, microbial cellulose) (Klemm *et al.*, 2001); but, recently, the ISO/TS 20477 (ISO, 2017) has unified all NC terminology. Fig. 3 shows the origin and possible cellulose sources to produce the main types of NC.

The production of CNF generally involves a pretreatment to reduce the energy consumption of the process (Spence *et al.*, 2011; Tejado *et al.*, 2012); and a mechanical treatment, where individual nanofibers are finally isolated in a gel-like macrostructure (Dufresne, 2017). The properties of CNF are related not only to the production process but also to the raw material source and quality (Balea, 2017; Manninen *et al.*, 2011). However, it has been recently demonstrated that the quality of the CNF is not always directly related to its effects (Balea *et al.* 2017a). On the other hand, CNC are traditionally produced by acid hydrolysis (Habibi, 2010) which dissolves the amorphous domains of cellulose remaining the crystalline parts. Both phosphoric and sulfuric acids have been used for this purpose due to the presence of phosphate or sulfate groups on CNC surface that favors their water dispersion. However, other acids, such as phosphoric or hydrobromic can be also used but the dispersibility of CNC suspensions produced from these acids is lower. The production mechanism of BC starts by the extrusion of a cellulose chain by each pore of the cell wall, to produce a nanofiber with an approximate diameter of 1.5 nm (Ross *et al.*, 1991). Nanofibers are self-assembled forming microfibers and then cellulose fibers are produced with a width of around 50-80 nm (Vitta and Thiruvengadam, 2012). The strain *K. xylinus* is an acetic acid bacterium that produces cellulose in a few days using sugars such as glucose, fructose or sucrose as carbon source (Kongruang, 2008). Recently, van de Ven and Sheikhi (2016) have developed a new kind of cellulose based nanostructures called hairy NC (HNC) with a crystalline core, similar to CNC, but with different polymer chains at its ends as shown in Fig. 3. It is produced by a periodate oxidation of cellulose.

The exponential interest on NC has driven the development of many techniques to characterize NC. Recently, Foster *et al.* (2018) have reviewed the best practices, methods and techniques for characterizing CNF and CNC including dimensions, crystallinity, mechanical properties, surface chemistry, charge, purity, rheological properties, and toxicity.

However, the industrial scale application of NC is just starting and it is hindered by the risks of a new process and the costs of the NC. Therefore, further research and development is needed to address various cost related issues associated with the production, characterization and the variables that influence on the application of the NC products (de Assis *et al.*, 2018a, b).

III. NANOCELLULOSE APPLICATIONS

There are many potential markets for NC that include both high-volume (e.g., paper, board, construction-based materials, textiles, plastics, automobile parts, environmental treatments) and low-volume (e.g., aerospace materials, medical implants, tissue engineering, drug carriers, wound dressings, cosmetics, hygiene products, pharmaceuticals, food additives/stabilizers, paint additives) applications (Kim *et al.*, 2015). Moreover, several researchers have studied different strategies to facilitate the use of NC at industrial scale. In this paper, we present a few of the most promising studies and strategies to implement the use of NC in industry.

A. Paper and board industry

Paper and board industry is a sustainable sector, leader in recycling. The State of the Global Paper Industry (Kinsella, 2018) reports that 57% of the paper and board produced was recovered and recycled, with a potential increase up to 64% by 2028. In 2015, the World Business Council for Sustainable Development (WBCSD, 2015) reported that in developed countries the recovery of paper and board is approaching the maximum that can be achieved, noting that it is near 70%, over 70% and almost 80%, in the USA, Europe and Japan, respectively. However, the increase of recycling produces a decrease of the recycled paper quality due to the hornification process, which deteriorates the fibers each time they are recycled, reducing the inter-fiber bonding and the swelling ability of the fibers (Blanco *et al.*, 2013; Hubbe *et al.*, 2007).

In this context, many researchers and papermaking industries have studied the potential use of NC as an alternative strengthening agent for both virgin and recycled pulps (Balea *et al.*, 2016a; Boufi *et al.*, 2016; Osong *et al.*, 2016) instead of the traditional refining process and the use of strength additives to counterbalance the low quality of secondary fibers by increasing the tensile strength up to 50%. However, there are many factors that

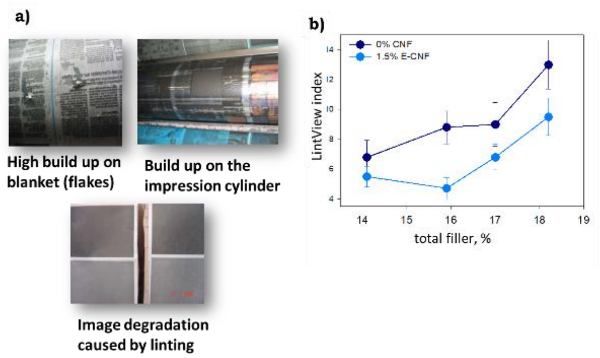

Figure 4. (a) Linting problems in offset printing and (b) effect of total filler and CNF on linting propensity. Adapted from Balea *et al.* (2016b).

influence on the mechanical properties of the final product when NC is used such us the type of NC, the cellulose source use as raw material to produce NC, the production process to obtain NC, the application method of NC into the pulp, the dispersion of NC before and after application, the type of pulp where NC is added, the retention system used, among others (Balea 2017).

Flocculation, drainage and retention studies with different retention systems have been recently studied to better understand the function of NC in the wet-end of the paper machine to facilitate its industrial implementation. For example it has been shown that 0.5 wt.% CNF is enough to counterbalance the negative effect of traditional polyacrylamide-based retention systems that improve drainage but decrease mechanical properties (Merayo *et al.*, 2017a, b).

However, the relationship between NC quality and its effects on mechanical paper enhancement is still a key point to reduce the cost of NC producing the minimum quality required for certain industrial application. Balea *et al.* (2017a) have demonstrated that the quality of the CNF, in terms of nanofibrillation degree, is not always directly related to its effects on the mechanical paper properties. However, the optimal minimum properties of NC have not been fully defined yet.

On the other hand, the removal of particles from the paper surface and its accumulation on the printing blanket, or linting, is an important problem for recycled paper mills. Recent researches have demonstrated that 1.5 wt.% of 2,2,6,6-tetramethylpiperidine-1-oxyl (TEMPO)-oxidized CNF reduced the linting propensity of high-filler-loaded recycled papers between 19 and 47%. Therefore, this reduction in linting offers the opportunity to increase the current filler contents up to 3%, without deteriorating the quality of printing (Balea *et al.*, 2016b). Moreover, CNF produced from pine sawdust, using a low-cost production process based on soda and antraquinone cooking followed by delignification before mechanical homogeneization, reduced linting propensity up to near 50% and improved the tensile index by 32.4% at the same time (Balea *et al.*, 2018) (Fig. 4).

Recently, another novel application of NC in paper industry has been developed based on the use of cationic HCN as retention additive (Campano *et al.*, 2019). The addition of 20 mg/g of cationic HCN reduced the recycled pulp drainage time by 78% and increased the total retention by 77% without an adverse effect on mechanical properties of the final paper.

Figure 5. Application of NC in food industry.

B. Water treatment

Several strategies have been conducted for environmental application of NC such as flocculation, adsorption, membrane filtration, disinfection and catalytic degradation, (Mohammed *et al.*, 2018). In most cases, NC surface is functionalized to increase the binding efficiency of certain pollutants. For instance, the presence of amine groups onto the surface of CNC increased the removal of anionic chromate forms containing Cr(VI) up to 98% (Carpenter *et al.*, 2015).

From research works, TEMPO-oxidized CNF have been commonly used for the adsorption of several metals (e.g., lead, calcium, silver…) from aqueous solution. Recently, it has demonstrated that 0.01 wt.% CNF removed water-based (or flexographic) inks by 100% for copper phthalocyanine blue ink, 87.5% carbon black ink and 83.3% for diarlyide yellow pigments in combination with cationic polyacrylamide (Balea *et al.*, 2017b). Additionally, TEMPO-oxidized CNF and chitosan, added separately (Balea *et al.*, 2019a) or grafted together (Sanchez-Salvador *et al.*, 2018), are also able to remove flexographic inks up to 100% eliminating the environmental impact of residual acrylamide monomers. Moreover, Balea *et al.* (2019a) have demonstrated that the sludge from these wastewater treatments which contain the water-based inks and CNF, could be recycled to produce fluting. Therefore, the addition of CNF as water-based ink removal agent may allow the utilization of papers printed with flexographic inks as raw materials in paper production. Then, it is possible to increase the amount of recycled paper which can be used, leading to a more sustainable paper chain.

C. Food industry

Many researchers have demonstrated the potential of NC in the food industry as food stabilizer, functional food ingredient, replacer fats, rheology modifier, food packaging, among others (Fig. 5).

Nowadays, the industry of food packaging manufactures many non-biodegradable plastics that after being used, are wasted, causing a high environmental impacts.

Because of that, many recent researches have studied the development of biodegradable coatings and films based on NC to be used in this field (Azeredo *et al*., 2017). NC films with thickness around 2-3 μm improve both mechanical and barrier properties such as oxygen permeability (< 0.01 cm3μm m-2 day-1 kPa-1) and water vapor transmission rate (<250 g·m-2·day-1) (Li *et al*., 2015; Nair *et al*., 2014). However, the oxygen permeability of the NC films increases with higher relative humidity (HR) values from 0.0006 to 0.85 cm3·μm·m-2·day-1·kPa-1, for HR=0 and 50%, respectively, because of the plasticizing and swelling properties of NC (Aulin *et al*., 2010). Moreover, several chemical treatments have been used to increase barrier properties against water, such as acetylation, through the formation of a hydrophobic surface (Gómez *et al*., 2016). NC has also shown to be promising as active materials for food packaging including new functionalities such as antimicrobial and antioxidant properties, moisture absorbers, oxygen and ethylene scavengers and carbon dioxide emitters (Souza *et al*., 2019).

NC could be incorporated into the food, as thickener, stabilizer, binder or fat replacer, to provide stabilization for foams (e.g., whippable cream and toppings, ice cream, mousses) or emulsions (e.g., salad dressings, sauces, creams), to retain moisture in meats during cooking, to improve texture and increase bulk of the baked products or to enrich appearance and mouth feel in foods (Gómez *et al*., 2016). However, NC has not been accepted yet as safe or approved food ingredient (DeLoid *et al*., 2019). Sanchez-Sálvador *et al.* (2018) have recently studied the production of oil-in water (O/W) Pickering emulsions using chemically-unmodified cellulose microfibers (CMF) instead of CNF, for food applications. These Pickering emulsions with 0.50 wt.% CMF increased up to 90 times the apparent viscosity of the emulsion phase compared to the sunflower oil without CMF, and maintained the stability over the time and towards shear forces. DeLoid *et al*. (2019) have demonstrated in physiologically relevant *in vitro* and *in vivo* systems that NC has little acute toxicity which is an important advance for acceptance of ingested NC as safe ingredient in the food industry.

D. Cement-based materials

Engineered cementitious composites, such as fiber-reinforced cement composites (FCCs), have a conglomerate structure formed with different materials in macro, micro and nano scales in terms of size. In this scenario, the addition of NC allows the production of more resistant cement composites improving mechanical properties (modulus of elasticity (MOE) and rupture (MOR), internal bonding strength) or composites with special properties, replacing inorganic fibers or synthetic polymeric materials. da Costa *et al.* (2018) have demonstrated that the combination of CNF (less than 1 wt.%) and cellulose fibers (8 wt.%) is adequate to form bridges in nano and microcracks, which improve the mechanical properties of the hybrid composites increasing MOR (19.9 MPa) with respect to FCCs without CNF (14.8 MPa) and increasing the fracture energy (0.422 *versus* 0.395 kJ/m2). Moreover, in fiber cement production NC could be used as additive to modify the rheology of the slurry, reducing the porosity, and inducing interactions with other components of the mixture (Balea *et al*., 2019c).

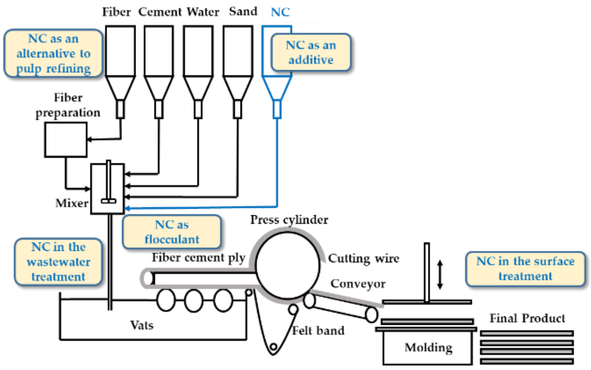

Figure 6. Potential application of NC in fiber reinforced cement composites

Balea *et al.* (2019c) have reported several unexplored fields in the fiber-reinforced cement composites in which NC may have a great potential, including its use as anionic flocculant, as surface treatment agent, or as additive for wastewater treatment (Fig. 6).

III. MARKET PERSPECTIVES

Based on Nano Cellulose Market Research Reports (Nanocellulose, 2019), the NC market is anticipated to register a compound annual growth rate (CAGR) higher than 18% during the period 2019-2024, reaching €576.5 million by 2023, although Biobased Markets estimate a higher growth up to 30% (Miller, 2019). Nowadays, key producers in the NC market include CelluForce Inc., Paperlogic, American Process Inc., CTP/FCBA, Melodea Ltd, American Process Inc., Oji Paper, Chuetsu Pulp and Paper, Nippon Paper Industries Co. Ltd, and Borregaard, among others.

The main drawback of the commercial progress of NC is its cost-competitiveness and the availability of producing high volumes for large-scale industrial use, such as in paper, packaging and construction products at low cost.

Different strategies have been studied to reduce the production cost of CNF and to facilitate the implementation of NC at industrial scale. During the last few years, several treatments such as enzymatic hydrolysis, carboxymetlylation, mechanical refining, and TEMPO-mediated oxidation have been developed as pretreatments before the defibrillation process to decrease the energy requirements from values higher than 100 to 2–4 Kwh/Kg (Klemm *et al.*, 2018; Osong *et al*., 2016).

Recently it has been demonstrated the viability of producing NC in situ using different recycling cellulose streams of the papermaking process as cellulose source which eliminate transportation costs of NC and reduce industrial wastes (Balea *et al*., 2019b; Campano *et al*., 2017, 2018). The addition of 3 wt.% of CNF produced in situ from recycled fibers improved tensile index of old newsprint (ONP) and old corrugated container (OCC) by 30% and 60%, respectively (Balea *et al*., 2019b). The in situ production of NC and the knowledge of the minimum NC quality required for a certain application, will favor the online control of the NC properties and its tuning according to the production requirements.

However, the use of NC at large-scale in other industrial applications remains a challenge due to the need of developing value proposition throught the entire supply chain (Miller, 2019).

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