Chapter 17
Nanocellulose and Nanocellulose-Based Composites for Food Applications

Suryani Saallah, Mailin Misson, Shafiquzzaman Siddiquee, Jumardi Roslan, M. Nazli Naim, Noor Fitrah Abu Bakar, and I. Wuled Lenggoro

17.1 Introduction

Increasing global demand for energy, depletion of petroleum sources, and concern over global climate change has led to the resurgence in the development of renewable and sustainable resources as an alternative to fossil fuel or petroleum-based materials (Chen et al. 2019a, b). In response, extensive research and development in nanocellulose production, a green, bio-based and renewable bio-material have been initiated and currently a subject of immense interest as a prominent candidate to replace the petroleum-based materials (Chen et al. 2017) as reflected in the rapid growing of scientific publications related to nanocellulose over the past two decades (Fig. 17.1).

Cellulose is the most abundant and ubiquitous renewable natural biopolymer in the biosphere with an estimated annual production of 75 to 100 billion tonnes (Xie

S. Saallah (✉) · M. Misson · S. Siddiquee
Biotechnology Research Institute, Universiti Malaysia Sabah,
Kota Kinabalu, Sabah, Malaysia
e-mail: suryani@ums.edu.my

J. Roslan
Faculty of Food Science and Nutrition, Universiti Malaysia Sabah,
Kota Kinabalu, Sabah, Malaysia

M. N. Naim
Department of Process and Food Engineering, Faculty of Engineering, Universiti Putra Malaysia, Serdang, Selangor, Malaysia

N. F. A. Bakar
Faculty of Chemical Engineering, Universiti Teknologi MARA,
Shah Alam, Selangor, Malaysia

I. W. Lenggoro
Institute of Engineering, Tokyo University of Agriculture and Technology, Koganei, Japan

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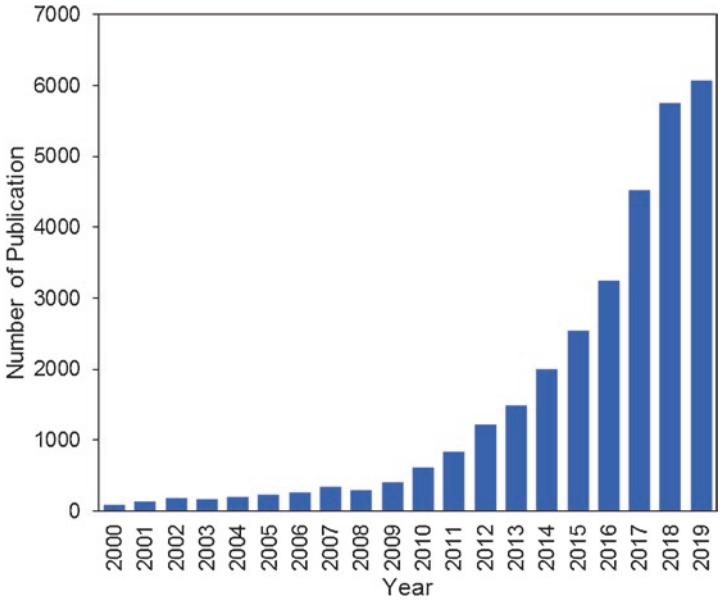

Fig. 17.1 Number of publications related to nanocellulose as analyzed by Scholar Plot[R] using data from Google Scholar (Data analysis was extracted from Scholar Plot[R] on 5 November 2019)

et al. 2016). Fibers generated from cellulosic material have long been used as lumber, textile and cordage. The extraction of nanocellulose from cellulosic materials through various chemical, biochemical, and mechanical means has enabled the emerging utilization of cellulose for much more sophisticated applications (Thomas et al. 2017). This include as synthetic reinforcement materials used in the composites and polymer matrixes, as stabilizers in food products and cosmetics, drug delivery excipient, scaffold for enzyme immobilization, tissue engineering, and biosensors, biodegradable packaging, air and water filtration, among others (Camacho et al. 2017; Tibolla et al. 2018).

According to a recent report by Global Market Insights, Inc. (2019), the global nanocellulose market in 2016 was USD 87.5 million and is forecasted to exceed USD 1 billion by 2024 by an outstanding compound annual growth rate (CAGR) of 33.8%. The growing market demand for nanocellulose is mainly attributed to its low-density, non-toxic and biodegradability in combination with its unsurpassed quintessential nano-dimensional properties including high aspect ratio and specific surface area, a high degree of crystallinity and transparency, tuneable self-assembly in aqueous media as well as its exceptional mechanical strength. Nanocellulose also holds a unique rheological behavior, good barrier properties and oxygen permeability which are desirable for food applications such as packaging, coating and as additives in food products (Kargarzadeh et al. 2017; Lee et al. 2017; Thomas et al. 2017).

In this chapter, we present the applications of nanocellulose and its composites in the food sector, one of the major growth enablers for the nanocellulose market.

Particularly, we highlight the function of nanocellulose as additives and ingredients in food products and the applications of nanocellulose-based composites in the development of active food packaging and edible coatings. Beforehand, sources, chemical and structural compositions of cellulose followed by nanocellulose and its classification are described.

17.2 Cellulose

17.2.1 Sources of Cellulose

As the most abundant biopolymer on the earth, cellulose has been used for centuries in highly diverse applications, even before its polymeric nature was recognized and well understood. It is a major component of lignocellulosic biomass, which mostly refers to plants or plant-based materials and widely distributed in wood of higher plants, forest and agricultural residues (Torabi et al. 2016).

Based on Fig. 17.2, conventional sources such as wood and cotton are ranked as the primary source of cellulose followed by forestry residues which consist of branches, leaves, bark, and other portions of wood and agricultural residues including palm oil empty fruit bunch, rice straw, rice husk, wheat straw, corn cob, banana rachis, and coconut husk which are mostly left on the fields as post-harvests residue. Industrial waste is the processed waste produced mainly by the food industries such as sugar cane bagasse, vegetable peels (i.e potato, carrot, tomato) and fruit peels (orange, apple, pear, banana), which is another source of cellulose emerging recently

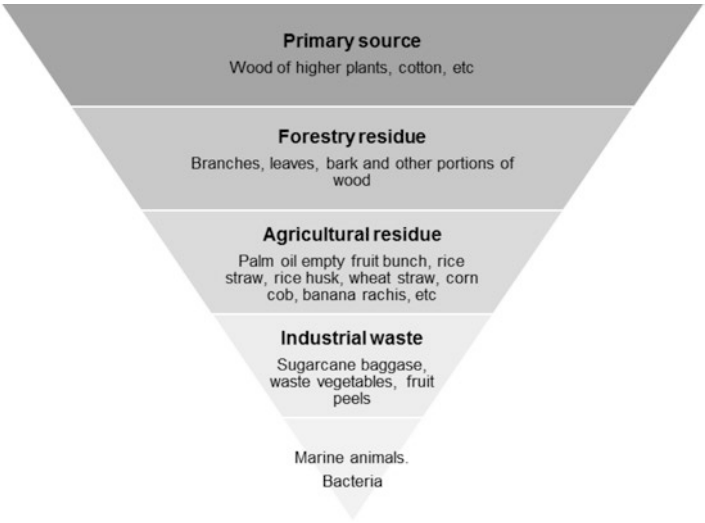

Fig. 17.2 Hierarchy of cellulose from different sources (Source: Torabi et al. 2016)

Table 17.1 Composition of lignocellulosic biomass from different sources (g/100 g of dry matter)

Source	Composition (%)			References
	Cellulose	Hemicellulose	Lignin	
Sugarcane bagasse	51.52	19.07	29.03	Huang et al. (2017)
Banana peel	35.27	19.51	6.68	Tiwari et al. (2019)
Banana peel	12.10	10.20	2.90	Tibolla et al. (2018)
Sugarcane bagasse	47.35	17.86	24.41	Moreno et al. (2018)
Banana rachis	49.12	18.57	20.23	Moreno et al. (2018)
Pineapple leaf	45.32	17.43	25.63	Moreno et al. (2018)
Bamboo	41.72	22.86	20.91	Xie et al. (2016)
Pear peel	38.50	23.60	28.10	Chen et al. (2019a, b)
Rice straw	36.50	38.00	22.00	Oun and Rhim (2018)
Orange bagasse	11.85	15.58	1.67	Mariño et al. (2018)
Unwoven cotton waste	58.72	6.20	15.99	Maciel et al. (2019)

(Rajinipriya et al. 2018). These vary in cellulose content and composition as described in Table 17.1. In addition to its plant origins, cellulose can also be obtained from several marine animals such as tunicate and algae. It is also secreted extracellularly by numerous bacterial species such as Acetobacter, Agrobacterium, Alcaligenes, Pseudomonas, Rhizobium and Sarcina.

17.2.2 Plant Cellulose

Plant fibers are the main natural sources of cellulose. Their hierarchical structure is essential not only for most living plants but also for several industries that take advantage of the outstanding features of natural fibers. Plant fiber wall structure is composed of cellulose molecules, elementary fibril, microfibril, macrofibril and lamellar membrane (Fig. 17.3). It was reported that the elementary fibrils are having a width of around 1.53.5 nm. These elementary fibrils were bundled to form microfibrils, which form the core structural units of the plant cell wall as a mechanism to reduce the free energy of the surface. Each microfibril might contain up to 40 cellulose chains and are 10–30 nm in width. The bundling of microfibrils formed a macrofibril, larger than 100 nm in width (Lee et al. 2017).

Plant cell wall is a complex, heterogeneous network of several components, mainly cellulose, hemicellulose and lignin, and a trace amount of proteins and extractives. Cellulose represents about 35–50% of lignocellulosic biomass dry weight while the remaining 20–35% and 10–25% were accounted for hemicellulose and lignin, respectively (Phanthong et al. 2018). The composition of these three major components is species-, types- and sources-dependent. Lignin keeps the water in fibers which confers the stiffness, strength, and protection to the cell wall and acts as a binder that holds the cellulose and hemicellulose complex. Hemicellulose, which adheres to the cellulose fibrils through hydrogen bonds and

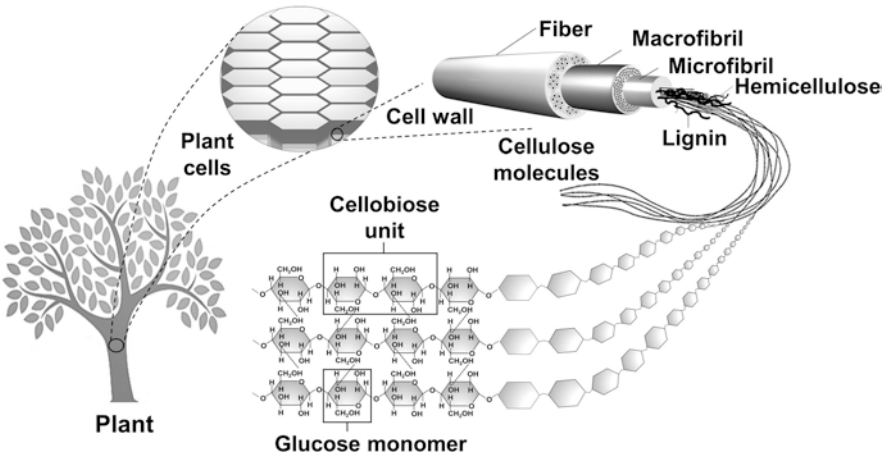

Fig. 17.3 Hierarchical structure of plant fibre. This figure was originally drawn by the author

Van der Waal's interactions and cross-linked with lignin, serves as a compatibilizer between cellulose and lignin (Halib et al. 2017; Kargarzadeh et al. 2017).

17.3 Chemical and Structural Composition of Cellulose

Since its discovery and isolation by a French chemist, Anselme Payen in 1838, the structure and properties of cellulose have been widely studied and highlighted in the literature. As the production of nanocellulose involves a breakdown of cellulose into smaller polymer branches in the nanoscale, it is necessary to comprehend the cellulose chemical and structural composition to give better insight on nanocellulose production (Chen et al. 2019a, b).

Cellulose is a structural polysaccharide, composed of repeating units of two anhydroglucose linked by a β-1,4-glycosidic bond, known as cellobiose. The anhydroglucose unit (AGU) consists of three hydroxyl groups in the equatorial position, responsible for the formation intra- and inter-molecular hydrogen bonding with the adjacent glucose unit in the same chain as well as with the different chains. These strong and tightly packed hydrogen bonding networks in the crystalline part of cellulose fibrils form a stable three-dimensional structure which makes them highly ordered and rigid, insoluble in water and highly resistant to most organic solvents (Kargarzadeh et al. 2017; Phanthong et al. 2018). The internal hydrogen bonds are also responsible for cellulose stiffness (Lee et al. 2017). On the other hand, very weak hydrogen bonds in non-crystalline amorphous domains contribute to the increased hydrophilicity and accessibility of cellulose materials (Kargarzadeh et al.

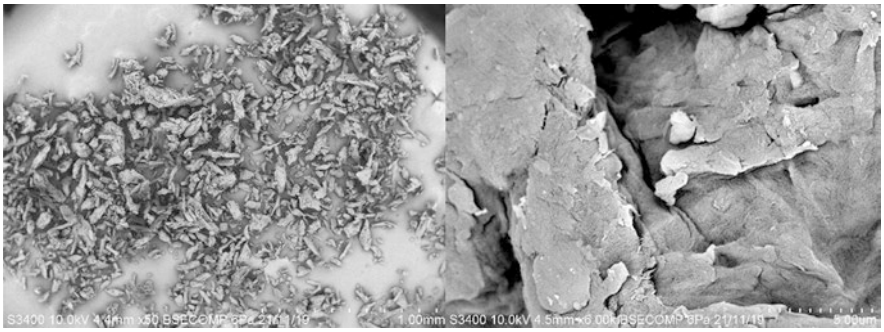

Fig. 17.4 Scanning Electron Microscope (SEM) images of microcrystalline cellulose with different magnifications. The strong and tightly packed hydrogen bonding networks of cellulose fibrils form a stable three-dimensional structure which makes them highly ordered and rigid. This figure is the author's own figure and has not been previously used or published

2017). The morphological structure of microcrystalline cellulose viewed under an electron microscope is shown in Fig. 17.4.

The wide orientation of glucose molecules and hydrogen bonding networks in cellulose producing different allomorphs which can be categorized into cellulose types I, II, III, and IV. Cellulose derived from plant, referred to as native cellulose is mainly found in two crystalline forms, known as cellulose type I and II. For cellulose type I, the hydrogen-bond network is packed in parallel. Chemical regeneration of cellulose type I by acid or alkaline treatment resulted in different arrangements of antiparallel packing of the hydrogen-bond network, known as cellulose type II. Treatment of cellulose type I or II with ammonia resulted in cellulose type III and heating of cellulose type III generates cellulose type IV. Among the four allomorphs, cellulose type I is thermodynamically less stable while cellulose type II has the most stable structure (Lavanya et al. 2011; Phanthong et al. 2018).

Functional groups of cellulose can be identified by using Fourier Transform Infrared (FTIR) spectroscopy. As lignocellulosic materials consist of a combination of cellulose, lignin, hemicellulose, and other constituents, this information is particularly important to determine the cellulose purity prior to nanocellulose isolation. FTIR band assignments for the main constituents in lignocellulosic materials are listed in Table 17.2 (Hospodarova et al. 2018; Sulaiman et al. 2015).

For pure cellulose, the characteristic peaks were located at 3325 cm^{-1}, 2891 cm^{-1} and 1159 cm^{-1}, corresponding to -OH stretching, -CH stretching and -COC stretching absorptions, respectively (Fig. 17.5). The broad -OH stretching vibration band in the 3700–3000 cm^{-1} region gives considerable information regarding inter- and intra-molecular hydrogen bond vibrations. The peaks located at ~1650 cm^{-1} correspond to the vibration of water molecules absorbed in cellulose. The absorption bands at 1426, 1364, 1159, 1027 and 896 cm^{-1} are associated with stretching and bending vibrations of -CH_2 and -CH, -CH and -CO, -COC, and glycosidic bond, respectively. The band at around 1420–1430 cm^{-1} is related to the cellulose crystalline structure, while the band at 897 cm^{-1} is assigned to the amorphous region in cellulose (Hospodarova et al. 2018).

Table 17.2 FTIR band assignments of main components in lignocellulosic materials

Wavenumber (cm^{-1})	Vibration	Component
3400–3300	-OH stretching	Cellulose
2900–2800	-CH symmetrical stretching	Lignin, cellulose
1800–1700	-C=O stretching	Lignin, hemicellulose
1650–1630	Adsorbed water	Water
1505	-C=C aromatic symmetrical stretching	Lignin
1435–1425	-CH_2 symmetrical bending -C=C stretching in aromatic groups	Pectin, Lignin, Hemicellulose
1380–1320	-CH, -CO, aromatic ring	Polysaccharides and cellulose
1240–1230	-CO stretching	Lignin
1162–1159	-COC asymmetrical stretching	Cellulose, hemicellulose
899–895	Glycosidic bond	Polysaccharide
670	-CH out of plane bending	Cellulose

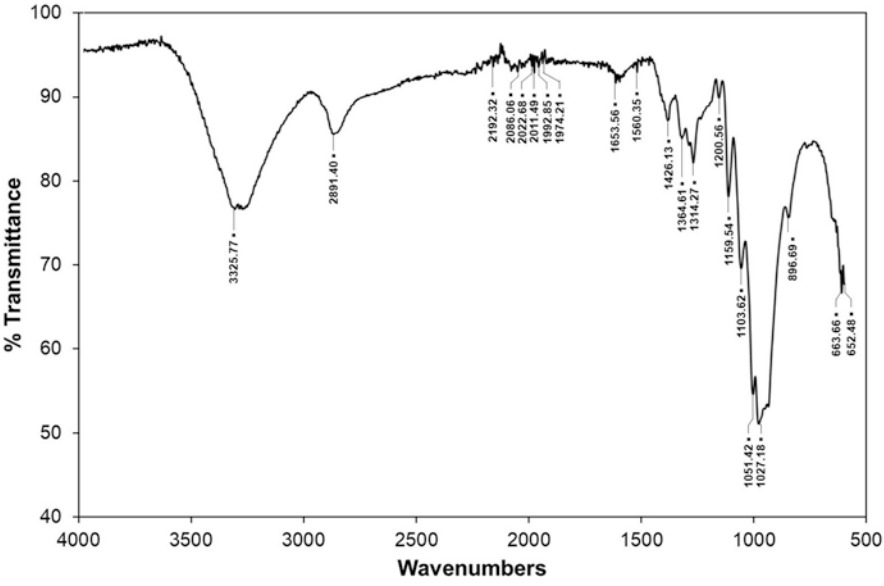

Fig. 17.5 FTIR spectra of pure cellulose (Avicel PH-101)

17.3.1 Nanocellulose

Nanocelluloses refer to cellulosic materials with at least one dimension in the nanoscale. A combination of inherent desirable cellulose properties and fascinating features of nanomaterials have made nanocellulose a promising alternative to replace the conventional materials made from non-renewable resources. Thanks to its biodegradability, lightweight, low density (1.6 g/cm^3), and outstanding mechanical

strength, nanocellulose is currently in high demand worldwide for a wide range of applications. Specifically, nanocellulose has an elastic modulus of up to 220 GPa, greater than Kevlar, tensile strength of up to 10 GPa comparable to cast iron and eight times higher weight to strength ratio than stainless steel (Phanthong et al. 2018). Additionally, nanocellulose offers unprecedented opportunities for surface functionalization due to their abundance reactive surface of hydroxyl groups.

Extraction of nanocellulose from lignocellulosic matters can be performed through chemical, mechanical, biological or combination of two (or more) approaches. Details description of these approaches can be found in Thomas et al. (2017) and Tan et al. (2019). Depending on the sources and extraction methods, nanocellulose can be categorized into three main groups; cellulose nanofibrils (CNFs), cellulose nanocrystal (CNCs) and bacterial nanocellulose (BC) (Table 17.3) (Lee et al. 2017). Although all types of nanocellulose are having similar chemical composition, their morphology, dimension, crystallinity and some other properties are different those largely determine their role in later applications.

17.3.2 Cellulose Nanofibrils (CNF)

Currently the nanocellulose markets are dominated by cellulose nanofibrils (CNF), accounted for more than 50% of market share in 2016 (Transparency Market Research). Several pilot facilities have been developed for CNFs production especially in Japan including Nippon Paper, Oji Paper, and Daicel as well as in other countries such as Finland (VTT), Sweden (Innventia), Canada (Kruger), among

Table 17.3 Terminology and description of different type of nanocellulose (Source: Lee et al. 2017)

Type of Nanocellulose	Other terminology	Description
Cellulose nanofibrils (CNFs)	– Cellulose microfibrils – Microfibrillated cellulose – Nanofibrillated cellulose – Cellulose nanofibers	Diameter: <100 nm Length: up to several microns Isolation process: Top-down (mechanical disintegration, enzymatic hydrolysis)
Cellulose nanocrystals (CNCs)	– Nanocrystalline cellulose – Cellulose nanowhiskers	Diameter: 5–70 nm Length: 100–250 nm (plant); 100 nm to several microns (tunicates, algae, bacteria) Isolation process: Top-down (acid hydrolysis)
Bacterial nanocellulose (BNC)	– Bacterial cellulose – Microbial cellulose – Biocellulose	Diameter: 20–100 nm Length: up to several microns Isolation process Bottom-up approach by bacterial synthesis

others. CNFs are viewed as important advanced biomaterials solutions in the packaging and composites market (Gómez et al. 2016; Tan et al. 2019).

Cellulose nanofibrils are a flexible nano-scale fiber consisting of bundles of elementary nanofibrils that are constructed from alternating crystalline and amorphous domains. As indicated in Table 17.3, CNFs are sometimes referred to as cellulose microfibrils (CMFs), microfibrillated cellulose, nanofibrillated cellulose or cellulose nanofibers. The width of CNFs can vary between 20 to 50 nm and length from 500 nm up to several microns. Isolation of CNFs from lignocellulosic materials can be performed through top-down approach (Fig. 17.6) employing mechanical disintegrations including high-pressure homogenization, cryo-crushing, ball-milling, microfluidization and high-intensity ultrasonication (Thomas et al. 2017).

Large-scale production of CNFs is hampered by the high energy required for the mechanical disintegration of lignocellulosic materials into CNFs. Hence, chemical pre-treatment with acid and/or alkali, or biological pre-treatment with enzymes, are often required to loosen the lignocellulosic structure (Tibolla et al. 2017). It is worth to highlight that CNFs have certain negative properties such as poor compatibility with hydrophobic polymers, which limit their application, especially in the development of composite materials. In this case, chemical modification is necessary to reduce the CNFs hydrophilicity (Kargarzadeh et al. 2017).

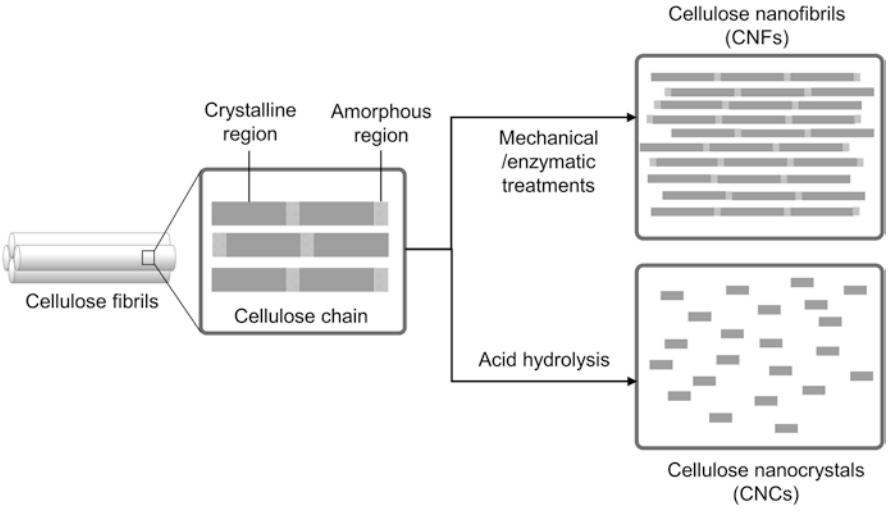

Fig. 17.6 Isolation of nanocellulose by top-down approaches. This figure was originally drawn by the author

17.3.3 Cellulose Nanocrystals (CNCs)

Needle-like nanocellulose, or also known as cellulose nanocrystals (CNCs), nanocrystalline cellulose and cellulose nanowhiskers have a width ranging from 5 to 70 nm. Depending on the sources, the length can vary between 100 to 250 nm for plants, and 100 nm up to several microns for tunicate, algae, and bacteria (Börjesson and Westman 2016).

Isolation of CNCs from cellulosic materials is commonly performed via hydrolysis with strong acid. With simpler procedures and shorter reaction time, sulfuric acid is usually the reagent of choice in comparison with other types of acids such as hydrochloric (HCl), hydrobromic (HBr), and phosphoric (H_3PO_4). CNCs have produced through this process consists of functionalized surface, high crystallinity and good colloidal stability in water. Unfortunately, acid hydrolysis is corrosive, the high tendency of cellulose over-degradation, low yield, requires a large amount of water and generates a huge amount of waste which eventually cause environmental pollution (Chen et al. 2017; Harini et al. 2018; Chen et al. 2019a, b). Given these drawbacks, diluted or organic acids have been suggested to provide milder reaction conditions, but with compromised efficiency. This drawback could be overcome by the addition of metal salt catalysts such as Chromium (III) Nitrate $Cr(NO_3)_3$ as reported by Chen et al. (2019a, b). This co-catalyst promotes complete solubilization of cellulose, hemicellulose and lignocellulosic into water-soluble chemicals and fuels.

17.3.4 Bacterial Nanocellulose (BNC)

Bacterial nanocellulose (BNC) is another kind of nanocellulose. Unlike CNCs and CNFs where the nanocelluloses were isolated through a top-down approach, BNC is synthesized by bacteria through the bottom-up process. Synthesis of BNC involves building up of low molecular weight of sugars by bacteria mainly by *Gluconacetobacter* for a few days up to two weeks. Nanocellulose synthesized by bacteria is free from other typical components in lignocellulosic biomass such as lignin and hemicellulose. BNC has extensive network structure due to the random motion of bacteria, with width ranging from 20 to 100 nm and length from 500 nm up to several microns. (Corral et al. 2017; Kargarzadeh et al. 2017; Sharma et al. 2019)

17.4 Applications

History of cellulose in food application can be traced back to the 1980s when Turbak and his group developed a series of food products using nanocellulose as the stabilizer owing to its exceptional wettability to water over oil, or as an additive in whipped cream, salad dressing and sauces. However, the progress has been stagnant, and this product was not commercially exploited mainly due to the high production costs (Tan et al. 2019). In the 2000s, with the rapid development of nanotechnology, the production and utilization of nanocellulose have received considerable attention, driven by the development of more cost-effective extraction technology and the availability of advanced analytical instruments for nanocellulose characterization. Today, the application of nanocellulose in food industries has grown tremendously especially as food additives, functional food ingredients, active packaging, and edible coating.

17.4.1 Food Additives and Stabilizers

Nowadays, a new generation of food products which combined the health benefits and excellent sensory quality is in high demand. In the development of food products, additives such as enzymes, emulsifiers, oxidants, and hydrocolloids are usually used to facilitate processing, improving microstructure for elevated mouth-feel and preserve the freshness of food products (Tan et al. 2019). With the distinctive rheological behavior in combination with the multifaceted advantages of nano-scale materials, nanocellulose-based additives are of great interest to be used as an emulsifying and stabilizing agent of food products such as salad dressings, foams, puddings, among others (Camacho et al. 2017; Gómez et al. 2016).

The potential use of bacterial nanocellulose as an additive in bread-making has been studied through thermo-rheological and dynamic oscillatory experiments by Corral et al. (2017). The addition of BNC has resulted in improved bread quality, reducing browning index, increasing the specific volume, porosity, luminosity, and moisture content. Moreover, the bread produced was more tenders and have less firm crumb than the control formulation without BNC, which make it more acceptable to the consumer. Based on these promising findings, in 2017, Marchetti and co-workers have employed BNC as an additive for the development of low-lipid and low-sodium meat sausages. The sausages show improvement in water-binding properties, hardness, cohesiveness, and chewiness with the addition of up to 0.267 g of dry BNC/100 g of batter was added. Besides act as fat mimetic and at the same time, the BNC could maintain the quality of the sausage with shelf-life of 45 days under vacuum refrigerated storage. Very recently, Marchetti et al. (2019) have investigated the rheological behavior of BNC-formulated gluten-free muffin. Low concentration of dry BNC (0.12 to 0.18 g/100 g) in the raw batter could produce batter with high elasticity, low specific gravity, and low flow indexes. The high viscosity

of the batter due to the addition of BNC could entrap more air and ultimately improve the volume of the baked product.

Besides improving the food products characteristic, many efforts have been devoted to the development of additives as a low-calorie fat replacer. This is due to the growing concern over the adverse health effect of foods with a high content of fat (Franco 2018). Removing fat from a food product is a possible and easiest solution, but the desirable properties such as flavor, aroma, and texture are compromised. Therefore, the addition of the so-called ''fat replacer' is a promising alternative. In general, fat replacers can be obtained from protein and carbohydrates. In most cases, a combination of several fat replacers is necessary to obtain fat-mimicking properties (Aaen et al. 2019).

Nanocellulose-based materials have recently been proposed as a low-calorie fat replacer for food products due to their availability and excellent viscosifying and gel-forming abilities. However, the interaction between the charged nanocellulose and other food components such as electrolytes and additives might affect their rheological behavior. With this view in mind, Aaen and co-workers (2019) have studied the rheological interactions of TEMPO-oxidized CNFs with pure water, electrolyte solutions and xantham gum, the commonly used food additives. Their findings suggest the applicability of CNFs as fat replacers in food products, even at low concentrations. The presence of salt in food could reduce the amount of CNFs required to obtain the desired storage modulus even further. It is worth noted that the interaction of highly charged CNFs with high salt content may promote aggregation of the CNFs component.

Nanocellulose has also been used as a food stabilizer owing to its well-dispersibility in water and self-assembled properties. The electrostatic interaction among the hydroxyl groups leading to the formation of a sol-gel structure and promoting stabilization of oil in water emulsion by the formation of a steric barrier at the oil and water interface. The addition of nanocellulose in the range of 0.10 to 0.30 wt% has shown to improve the stability of ice cream based on the melting and falling time. Besides significant improvement in stability, the addition of a small amount of nanocellulose could eliminate the need to change the composition of the ice-cream mix, thus the good flavor and texture of the frozen dessert could be maintained (Gómez et al. 2016).

17.4.2 Functional Food Ingredients

According to Functional Food Centre (FFC), functional food is defined as "Natural or processed foods or food ingredients that contains known or unknown biologically-active compounds; which in defined amounts provide a clinically proven and documented health benefit for the prevention, management, or treatment of chronic disease" (Gómez et al. 2016). Based on this definition, nanocellulose, especially in the form of nanofibrils (CNFs) could be considered as a potential functional food

ingredient owing to their dietary fiber characteristics that able to give positive health effects especially gastrointestinal related diseases.

As mentioned previously, CNFs have distinctive properties when compared to other sources of food fibers. For example, CNFs of approximately 18 nm in width and high aspect ratio derived from Parenchyma cells of pears and apples by one-time fibrillation using grinding apparatus is expected to improve the nutritional and technological properties of foods. Moreover, with their high viscosity and good dispersibility in water, the CNFs produced could be a good candidate in the formulation of functional foods (Ifuku et al. (2011).

While various studies have proved the potential of nanocellulose to be used as food ingredients based on the quality attributes, limited information is available on their effectiveness with regard to the improvement of human health, safety aspects, and possible side effects. Thus, biological, biochemical and histological tests on mice supplemented with an increasing amount of CNFs have been conducted by Andrade et al. (2015) to find the effect of the addition of CNFs on their health condition. Accordingly, no significant changes in the blood sugar level, lipid profile, and mineral nutrients were observed within the range of CNFs concentration studied. More importantly, the CNFs did not cause an adverse effect on the animals' hepatic system and metabolism.

The health benefits of nanocellulose-supplemented food products such as milk, soy protein isolate, whey protein isolate, and starch have been reported by Liu and co-workers in a series of studies utilizing three types of nanocellulose; CNFs, CNCs, and TEMPO-CNFs. Overall, the presence of the nano-scale dietary fiber could improve gastrointestinal health, glucose tolerance, insulin response, promotes satiation, thus benefiting weight loss. The positive correlation between nanocellulose and gastrointestinal health is further supported by a recent study conducted by Chen et al. (2019a, b). Oral consumption of CNFs by Western diets (WD)-fed mice at a sub-chronic concentration of 30 mg/kg body weight could decrease fat absorption in the jejunum and protected the liver from a high-fat burden.

17.4.3 Active Packaging

The importance of packaging in the protection, communication, convenience and containment of goods is undisputable. However, these basic functions are no longer sufficient as society demand for a new generation of packaging with improved functionalities to prolong the shelf-life and reduce the growth rate of microorganism in food products. Hence, research and development in the field of 'active' packaging have increased tremendously in the last few years (Gómez et al. 2016).

Active packaging is the new generation of packaging materials designed to deliver protective agents such as antimicrobial agents, preservatives, antioxidants, and oxygen and moisture barrier. Polycarbonate- and polyethylene-based packaging with antimicrobial agents such as copper, zinc and phenolic substances have been widely reported (Lee et al. 2017). With the increasing concern over the

environmental impact of petroleum-based packaging, trends have now shifted towards the use of green, sustainable and biodegradable polymers from natural resources particularly cellulose-based materials (Rajinipriya et al. 2018; Tan et al. 2019).

While cellulose can form strong and stiff packages, the barrier properties are not comparable to those of the synthetic polymers. The breakings down of cellulosic materials have produced nanocellulose with promising properties for active packaging applications. Films made from nanocellulose have a high air and oxygen barrier. It has been reported that the oxygen permeability of nanocellulose films with thickness of 21 ± 1 mm was 17 ± 1 ml m^{-2} day^{-1}, competitive with those made from synthetic polymers such as ethylene-vinyl alcohol (EVOH) (3–5 ml m^{-2} day^{-1}) and polyvinylidene chloride (PVdC) films (9–15 ml m^{-2} day^{-1}) of roughly the same thickness. The excellent barrier properties of nanocellulose film are mainly contributed by its high crystallinity, tight fiber network; entangle structure and dense fiber packing (Gómez et al. 2016).

Despite this outstanding feature, the oxygen permeability of nanocellulose films is strongly influenced by relative humidity. At high humidity, the interaction of nanocellulose with water molecules caused the film to swell and increasing the oxygen permeability. Several modifications have been implemented to overcome this drawback. One such example is by developing a nanocellulose composite film. Active packaging made from nanocellulose-based composite such as CNFs/PPy/PVA (Bideau et al. 2017), poly(sulfobetaine methacrylate) (PSBMA)/BNC (Vilela et al. 2019) and clove oil/chitosan/β-cyclodextrin citrate/CNFs (Adel et al. 2019), have shown improved hydrophobicity and mechanical properties as well as combines the biodegradability, antibacterial and antioxidant properties for active packaging.

17.4.4 Edible Coating

The edible coating is usually made from starch due to its high transparency, odorless, tasteless and semi-permeability. However, starch-based films exhibit poor water resistance and mechanical strength. Incorporation of nanomaterials in starch films could be an effective strategy to overcome these limitations. Jeevahan and coworkers (2019) have shown that nanocellulose obtained from banana pseudostems has improved film properties of rice starch-based edible films. This work was further extended to other types of starch including wheat, maize, and potato, and similar findings were observed. Besides improving the mechanical and barrier properties, nanocellulose can also act as an antimicrobial agent and oxygen scavenger.

Another example of the application of nanocellulose in edible food coating in the development of agar-based edible film (Wang et al. 2018). Crystallinity, thermal stability and tensile strength of the edible film were improved with the addition of bacterial nanocellulose with concentration ranging between 3 to 10 wt%. Significantly decreased moisture content, water solubility, and water permeability

was also observed. This indicates that nanocellulose has high potential to be used as reinforcing material in edible food coating.

17.5 Conclusion and Outlook

Nanocellulose is an inexhaustible and sustainable material derived from lignocellulosic biomass with ground-breaking applications in the food industry. The fundamental characteristics that make nanocellulose a fascinating material are the fact that it combines the inherent properties of cellulose (low-density, biodegradability, non-toxicity) and nano-dimensional characteristics including high surface area, high aspect ratio, and extensive hydrogen bonding ability. These features contribute to high crystallinity and excellent mechanical strength of nanocellulose. For food applications, its unique rheological behavior, good barrier properties, and oxygen permeability are particularly attractive. It should be noted that while the role of nanocellulose in improving food quality attributes is indubitable, information on the safety aspects and possible side effects of nanocellulose is still limited, which calls for more and in-depth investigation. Moreover, as the current extraction processes mainly rely on acid hydrolysis, more environmentally friendly approaches are highly sought.

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