Chapter 3
Cellulose-Based Nanostructured Materials in Edible Food Packaging

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3.1 Introduction

The development of cellulose-based nanostructured materials has a great interest in the current trend of edible food packaging. The cellulose derivatives and its nanostructured materials are utilized in various food and beverage industries for obtaining tailor-made properties of food products. As shown in Fig. 3.1, the targeted food-based industries for using cellulose-based products include bakery industry, meat industry, dairy industry, cereal industry, veterinary foods, food packaging industry, and others due to its tunable physicochemical properties. In food industries, the cellulose and its derivatives are used in various formulations such as emulsifiers, bulking agents, anticaking agents, fat substitutes, and texture enhancers. Additionally, cellulose and various derivatives play a remarkable role in improving the food quality in terms of texture, color, and others with an enhanced shelf life of food products. The use of cellulose in food products and packaging (edible and non-edible) are largely applied due to increased food waste for careless handling, mechanical damages, and increase of plastic-based waste as packaging materials. Cellulose and its nanoforms are available extensively and have the characteristic attributes of biodegradability, biocompatibility, surface chemistry, improved packaging property, non-toxicity, which make them an ideal material to be used as a replacement for available conventional materials. Cellulose has a great market value for reducing the overall carbon footprint in the packaging industry and further provides improved food value. Interestingly, cellulose derivatives and nanoforms are also used in edible packaging materials to aid improved properties to other materials such as chitosan (CS), starch, and agar. Further, the use of nanocellulosic materials in edible and non-edible sectors has been increased for

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V. Katiyar and T. Ghosh, *Nanotechnology in Edible Food Packaging*,
Materials Horizons: From Nature to Nanomaterials,
https://doi.org/10.1007/978-981-33-6169-0_3

having tailored barrier, mechanical, thermal, color properties which further improved the shelf life of food products. As shown in Fig. 3.1, the properties of nanocellulosic materials can be modified via chemical, physical, mechanical methods, and developing biocomposites of nanocellulose.

Additionally, the various forms of nanocellulose are available as nanocrystals, nanofibers, nanoparticles (NPs) which can be availed by modifying the routes of synthesis. The various forms are comparable in terms of obtaining the tunable properties of packaging materials for both edible and non-edible packaging forms. The characteristic traits of nanocellulose include rheological behavior, high mechanical property, lightweight, barrier properties, nutritional properties, etc. The cellulose-based nanostructured materials are utilized in food packaging, developing starch-based foods, food stabilizers, delivery systems, etc. The nanocellulose is widely used as a stabilizing agent in food emulsions, in food packaging, and as functional food ingredients. Nanocellulose has several traits such as: (i) The rheological behavior of nanocellulose provides a route to be used as food additives; (ii) transparency, mechanical property, barrier property provide food packaging application; (iii) high surface area and surface functionality are helpful in developing food coating; and (iv) food products such as sweets, delicacies, and puddings; low-fat mayonnaise, milk ice-cream, fruit jelly, meat sauces. Further, bacterial cellulose (BC) as food packaging materials for sausage, meat casings, and others is also available.

Based on this discussion, the chapter will detail the extraction of cellulose-based materials from various available sources for the fabrication of nanoforms. Nanocellulose structures can be obtained from available sources utilizing a significant method and play a very significant role in nanotechnology-based research and development. However, the yield of nanocellulosic materials is dependent on several factors such as source, extraction process, and processing conditions.

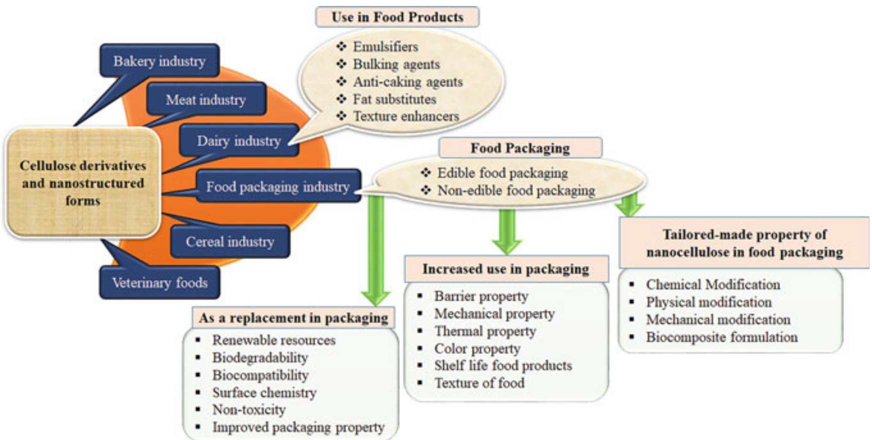

Fig. 3.1 Prospect of cellulose derivatives and nanostructured form in food sector

A brief discussion has been made on the status of using cellulose, its various forms in the field of food sectors such as bakery products, meat products, dairy products, edible food packaging, non-edible food packaging, and others. The global cellulose-based food packaging status has also been overviewed. However, the nanoforms of cellulose can also be fabricated utilizing several processes such as acid hydrolysis, mechanical methods, enzymatic hydrolysis, oxidation methods, and ionic liquid treatments. A detailed discussion about the several routes for the synthesis of nanocellulose materials has been made. Finally, the use of various forms of nanocellulose in the development of composite-based edible food packaging for enhanced shelf life of food products has also been detailed.

3.2 Outlook of Cellulose Resources to Be Used in Food Application

The wide usability of cellulose-based materials in developing packaging films is increasing such as bottles, boxes, and pouches. The plant fibers are the major sources of cellulose such as cotton, flax, hemp, and jute. Further, lignocellulosic biomass such as agricultural residues, forest residues, and energy crops is a source of cellulose. The agricultural residues include rice husk, wheat straw, corn stover, rice straw, sorghum straw, coconut husk, pineapple leaves, manure, seaweeds, barely straw, maize, barley husk, etc. The forest residues as a source for lignocellulosic biomass include woodchips, wood sawdust, wood branches, etc. Further, the energy crops as a source of lignocellulosic biomass include energy cane, Miscanthus, switch grass, etc. Cellulose is also available from plant foods such as cereals, fruits, nuts, legumes, potato with skins, seeds, and cabbage family of vegetables. In 1920, cellulose has been synthesized from delignified wood pulp, ramie, cotton, and other sources. The extraction processes for cellulose from available sources have been made in the below section. The various cellulose derivatives such as BC, carboxymethyl cellulose (CMC), enzymatically hydrolyzed CMC, cellulose acetate, ethylcellulose (EC), hydroxypropyl cellulose (HPC), hydroxypropyl methyl cellulose (HPMC), methylcellulose (MC), ethyl methyl cellulose, microcrystalline cellulose (MCC), and powdered cellulose are used as additives in food products. Further, cellulose acetate, EC, methyl cellulose, powdered cellulose are used as binders in food products. Nanocellulosic materials with versatile properties are also utilized in various food products and packaging materials for improved properties. Interestingly, the surface morphology and characteristic attributes of nanocellulosic materials are highly dependent on the pretreatment processes of lignocellulosic biomass which removes the non-cellulosic part.

3.3 Extraction of Cellulose from Available Sources

Cellulose is a homopolymeric unit consisting of β-1,4-linked anhydro-D-glucose having high molecular weight, and the repeating units are corkscrewed at 180° with their neighbor molecules. From very early days, the incorporation of cellulose fibers in composite materials has been increased tremendously in terms of effective cost, recyclability, biodegradability, and availability in comparison with the conventional reinforcing materials such as glass and aramid fibers. As discussed, the available sources of cellulose are plants such as ramie, sisal, flax, wheat straw, and potato tubers, whereas algae, bacterial source, and marine animals are also potential sources for producing cellulose [1, 2]. The cellulose and related nanoforms are a remarkable candidate to be used in edible food packaging materials for the noteworthy properties. The extraction process, sources, pretreatment process play a remarkable role in the fabrication of nanocellulose materials to be used in edible packaging materials. However, a brief discussion has been made related to the available sources of cellulose (Fig. 3.2), which are used globally to obtain cellulosic materials.

3.3.1 Cellulose from Plant-Based Sources

The wide availability of wood pulp and cotton fibers has made potential pathways to extract cellulose. As discussed, the plant materials such as bamboo, flax, hemp, jute, ramie, sisal, rice husk, and coconut husk are also a potential source of cellulose. The plant sources are generally considered as lignocellulosic biomass, having the main hierarchical architecture consisting of cellulose, hemicelluloses, and lignin. Additionally, the grasses, water plants, some plant parts (leaf, fruit, and stem), and agricultural wastes such as sugarcane bagasse, rice straw, and wheat

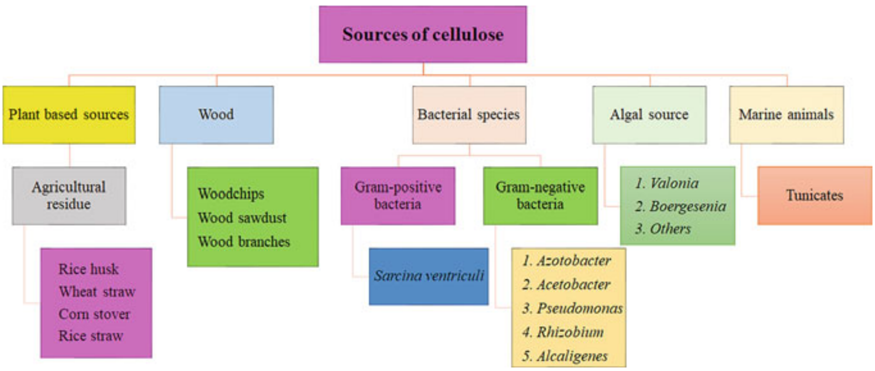

Fig. 3.2 Available sources of cellulose to be used in edible food packaging

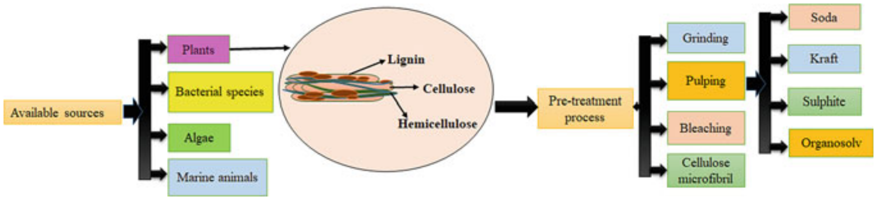

Fig. 3.3 Cellulose sources and pretreatment process for cellulose extraction

straw are available sources for cellulose. However, the percentage of cellulose from lignocellulosic biomass varies from sources to sources such as (i) cotton stalks: 58% cellulose, (ii) bagasse: 52% cellulose, (iii) sweet sorghum: 44% cellulose, (iv) corn ears: 38% cellulose, (v) pineapple leaf: 36% cellulose, (vi) corn cobs: 33.7% cellulose, and (vii) wheat straw: 32.9% cellulose. The extraction of cellulose from lignocellulosic biomass generally follows pretreatment methods such as chopping, pulping, and bleaching methods to obtain cellulose as represented in Fig. 3.3.

3.3.2 Cellulose from Bacterial Species

Gluconacetobacter xylinus (formerly known as *Acetobacter xylinum*) is the most used bacterial species for the fabrication of BC. However, BC can be obtained from both gram-positive (*Sarcina ventriculi*) and gram-negative bacteria (*Azotobacter, Acetobacter, Pseudomonas, Rhizobium, Alcaligenes, Salmonella*). The bacterial species produce cellulose microfibrils having the appearance of clear, flat, and thick gel with a large amount of water content (97–99%). The advantageous nature of BC is its high chemical purity with tunable microfibril formation and crystallization property, which can further be tailored by adjusting the culture conditions. The advantageous attributes of BC in comparison with plant cellulose include hydrophilicity, ultrafine network structure, mechanical property, water holding capacity, etc. Additionally, the BC has obtained an immense interest in edible food packaging for its characteristic attributes such as wettability property, purity, and mechanical property.

3.3.3 Cellulose from Algal Sources

The major component is the cell wall of algae with highly crystalline nature such as brown, green, yellow, gray, and brown. Various orders of algae are *Valonia*, *Boergesenia, Micrasterias denticulata, Micrasterias rotata*, *Dictyosphaeria, Siphonocladus*, *Cladophora*, *Boergesenia*, *Microdyction,*and *Rhizoclonium,* where

Valonia (I_α triclinic allomorphs type) or *Cladophora* are highly crystalline up to 95%. The algae-based materials for cellulose are recommended for their high crystallinity.

3.3.4 Cellulose from Marine Animals

Mostly focused marine invertebrate sea animal of Tunicates class is sea squirts (*Ascidiacea*), and their different species are *Metandroxarpa uedai, Halocynthiaroretzi,* and *Halocynthia papillosa*. The tunics (outer tissue) of tunicates are generally consisting of cellulose, lipids, mucopolysaccharides, sulfated glycans. The cellulose developed from tunics can be obtained through prehydrolysis, kraft cooking, bleaching process to remove the non-cellulosic parts. The fabricated cellulose can be obtained as 100% pure with highly crystalline in nature with CI_β lattice-type (monoclinic unit having two hydrogen bonding chains per unit cell) allomorph and further provide high microfibril aspect ratio, which is a useful approach to be used in packaging application. However, the cellulose from tunicates can be obtained as different morphological and chemical structures, and yields depending on the targeted sources of extraction.

Cellulose is assembled as an individual cellulose chain forming cell wall of the fiber (including primary cell wall and secondary cell wall). The secondary wall mostly consists of microfibrils comprising both the amorphous and crystalline regions. They are helically framed where the crystalline region is having inter- and intramolecular interaction networks. Even molecular orientations of crystalline regions also vary with different interchangeable cellulose polymorphs, namely I, II, IIII, IIIII, IVI, and IVII [3, 4]. The promising properties of cellulose have made researchers in increasing its usability for versatile application. Moreover, the isolation of cellulose in highly pure form is possible only by dissolution of hemicellulose, lignin, and remaining non-cellulosic components. Thereby, cellulose can be isolated from the various available sources with varied properties as mentioned [4, 5]. In the paper industry, pulping and bleaching methods are used to remove the components other than cellulose and further, the brightness of fabricated cellulose can be adjusted by mechanical processes. The alkali treatment is the most common method to remove the hemicellulose and lignin part because of their alkaline solubility (Fig. 3.3). Extraction of cellulose from agriculture waste (sugarcane bagasse) can be developed using steam explosion and xylanase as pretreatments followed by bleaching. The various resources have reported with their crystallinity percentage for cellulose such as sisal fibers (75%), wheat straw (77.8%), sugarcane bagasse (50%), sheath of coconut palm leaf (47.7%), and commercial microcrystalline cellulose (74%). Extraction of cellulose microfibrils from agricultural residue (coconut palm leaf sheath) has been done using chlorination and alkaline extraction process to obtain 10–15 μm diameter fibrils [6]. Another study has been made using agro-industrial biomasses (pomaces) to enhance its usefulness in the field of polymer. The obtained cellulose fraction for each pomace is having Segal crystallinity index (tomato: 48.97%, apple: 51.34%, cucumber: 53.61%, carrot: 68.73%) with low lignin content [7]. Cellulose fibers are extracted from rice husk with cellulose content of 96% after chemical treatment using alkali and bleaching to obtain a diameter of 7 μm with enhanced thermal stability [8]. Moreover, sugarcane bagasse has been used for isolation of cellulose by steam explosion and xylanase pretreatment [9]. The extracted cellulose materials have crystalline and amorphous regions, where the fabrication of nanocellulose mainly consists of crystalline regions with various percentages of crystallinity. However, the property of nanocellulose in terms of crystalline regions can be tailored for improved property.

3.4 Fabrication of Nanocellulosic Materials

Depending on the dimensions, functions, preparation methods, and sources, as shown in Fig. 3.4, nanocellulose has been subcategorized as cellulose nanocrystal (CNC), nanofibrillated cellulose (NFC), and bacterial nanocellulose (BNC). Among all, CNCs are mostly crystalline rod shaped having limited flexibility than NFC. Further, CNC may also be termed as nanowhiskers, rod-shaped cellulose crystals, or nanorods by various researchers. CNC is having low aspect ratio with diameter 2–20 nm and varying length between 100 nm and few several micrometers. The CNC or nanocrystalline cellulose (NCC) is highly crystalline nature with 100% pure cellulose content with crystalline percentage between 54 and 88% [10], whereas NFC (developed through chemo-mechanical method) is bundle of cellulose chains with long, flexible, and entangled cellulose nanofiber (CNF) with approximately 100 nm diameter having alternate crystalline and amorphous diameter and crystallinity index of 63.57% [11]. The bigger size of NFCs may be obtained due to aggregation of NFC molecules. Further, the NFC dimensions may be varied depending on the extraction method and sources. BNC is having superfine

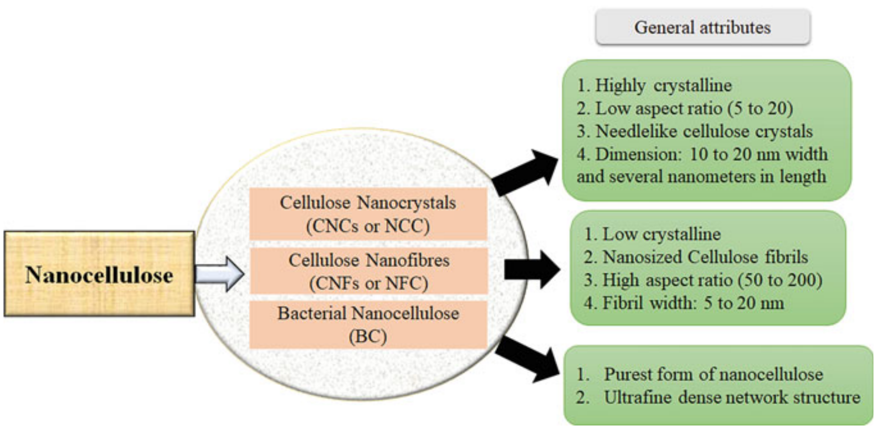

Fig. 3.4 Various forms of nanocellulose

diameter, high degree of polymerization, and high crystallinity produced by using microorganisms via two methods such as fermentation production and reactor-based production. For the fabrication of BNC, several acetic acid bacteria are used [12], where *Komagataeibacter xylinus* has received much interest due to their capability in secreting micro- or nanofibrils. An investigation has been made using high-pressure homogenizer (HPH) for preparation of BNC from pineapple peel waste by fermentation using *A. xylinum*, where the fabricated BNC (by HPH) has a crystallite size of 4.76 nm and crystallinity percentage as 86% [13]. In this regard, Fig. 3.5 details the various techniques for nanocellulose extraction.

3.4.1 Acid Hydrolysis

The first attempt of preparing successful CNC was made in 1947 using the technique of acid hydrolysis of cellulose with sulfuric acid and hydrochloric acid by Nickerson and Habrle [14]. Besides, the techniques for obtaining nanocelluloses such as enzymatic hydrolysis, organic acid hydrolysis, solid acid hydrolysis, oxidative degradation, ionic liquid, and subcritical hydrolysis have been represented in a tabulated form in below section (Table 3.1). The acid hydrolysis is the most commonly used among the other methods including sulfuric acid (H_2SO_4), hydrochloric acid (HCl), phosphoric acid (H_3PO_4), hydrobromic acid (HBr), and their mixed acids [15]. Among all, H_2SO_4 has been used mostly due to the negative surface charges and producing more stable suspension of nanocellulose. In general, a concentration of 60–65%, with reaction time and temperature of 30–60 min and 40–50 °C, respectively, is required to develop CNC (30 wt% CNC yield). The decreased yield may be related to the high acid concentration, temperature, and time, which can be tailored further for increased yield of CNC. However, the yield of CNC, its properties, characteristics such as crystallinity and aspect ratio can be monitored by varying the acid concentration. Further, CNC obtained from eucalyptus with yield of 70 wt% can also be obtained via tailoring the reaction conditions in terms of acid concentration, temperature, and reaction time [16]. The bamboo pulp has been used as the source for cellulose to obtain CNC (rod and porous network form) with yield of 32.3 wt% using sulfuric acid hydrolysis. The thermal stability for CNC presented two-step degradation temperature range at 180 ∼ 320 °C and 350 ∼ 450 °C [17]. Moreover, from oil palm empty fruit bunch pulp, CNC having spherical shape and average diameter 30–40 nm has been obtained using H_2SO_4 solution (64% w/v) at 45 °C in an ultrasound bath for almost 2 h [18], whereas to obtain increased yield of nanocellulose, acid concentration is the key parameter. Further, studies have been made on bleached kraft (eucalyptus pulp), where acid concentration between 58 and 62 wt%, moderate temperature of 50–60 °C, and reaction time between 30 and 180 min are preferred to maximize CNC yield [19]. The extraction of CNCs using hydrochloric acid hydrolysis under hydrothermal conditions (neutralization with ammonia) can provide thermally stable CNC with 93.7% yield, 88.6% crystallinity, maximum degradation

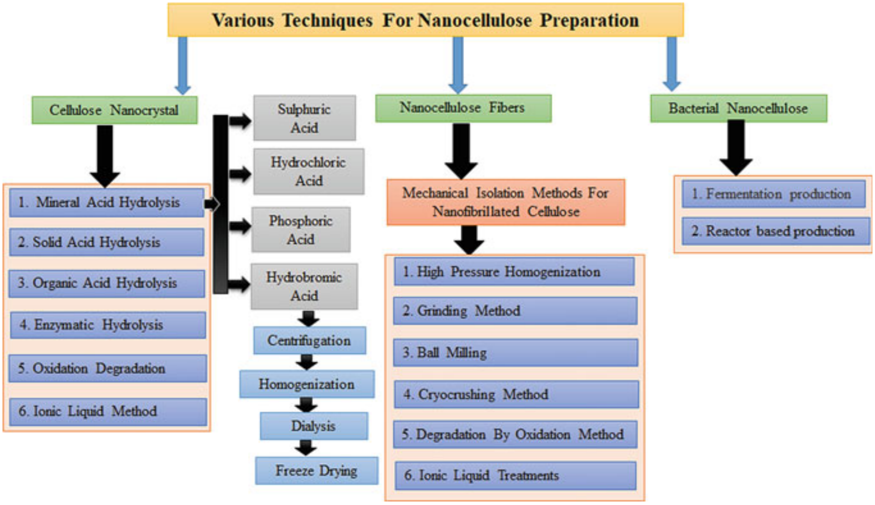

Fig. 3.5 Targeted techniques for extraction of nanocellulose

temperature of 363.9 °C [20]. Further, the presence of ammonia in the hydrochloric acid hydrolysis under hydrothermal condition can provide stability of CNC suspensions. Moreover, a further study has been reported using different mineral acids to obtain tailored morphological structure of nanocellulose, where the CNCs can be utilized to develop poly(lactic acid)-based composites with improved thermal stability as non-edible packaging materials [21]. The fabrication of acetylated CNF from sisal fiber has been done using chemical methods such as strong alkali treatment to swell fibers, bleaching treatment to remove lignin, and acetylation treatment to reduce the intermolecular hydrogen bond [22]. Further, the surface chemistry of nanocellulosic materials can be tailored for various applications. Moreover, among available, CNCs can be widely utilized as a potential nanofiller for the various characteristic attributes [23].

3.4.2 Mechanical Methods

In plant cell walls, cellulose fibers are converted to nanofibrils applying high and strong mechanical disintegration power, where the nanocellulosic materials can be obtained with dimensions of 10–100 nm. In this regard, the several mechanical techniques to obtain nanocellulose are grinding, cryocrushing, homogenization, microfluidization, ultrasonication, etc. In case of grinding method, cellulose slurry is introduced between static and rotating grindstones, and the grinding of the materials occurs due to frictional effects which deliver smaller-sized materials with higher surface areas. There are various factors which may affect the obtained

Table 3.1 Fabrication of nanocellulose using various techniques from available sources

Sl. no.	Source	Method	Procedure	Results	References
1	Rice husk	Acid hydrolysis	50 °C, 10.0 mol L^{-1} of sulfuric acid, 40 min	10–15 nm	[6]
2	Sisal fibers	Nitric acid and acetic acid	Boiling temperature 90 min, diluted to (1:5 ratio)	Diameter of 27 ± 13 nm and length of 658 ± 290 nm	[25]
3	Recycled pulp	Enzymatic hydrolysis	84 EGU of endoglucanase per 200 mg, 50 °C for 60 min	33–80-nm widths and 100 nm–1.8 μm in length	[14]
4	Sugarcane bagasse	High-pressure homogenization	Ionic liquid (1-butyl-3-methylimidazolium chloride ([Bmim]Cl))	Nanocellulose diameter: 10–20 nm	[32]
5	Sisal fibers	Enzymatic hydrolysis and mechanical shearing	Enzyme Celluclast (Cclast) (endoglucanase) or ecopulp energy (Eco) (exoglucanase) (minimum 0.1 wt% incubated at 50 °C for 2 h) Homogenized and incubated again for 50 °C for 2 h, manual 10 min mixing an 100 °C for 10 min to stop reaction	Length (Cclast = 251 ± 47 and Eco = 234 ± 63) Diameter = 7.2 ± 0.8 and 6.6 ± 1.3, respectively	[15]
6	Pulp fiber	Cellulase and xylanase	20 u/mL (cellulase: xylanase) = 9: 1	Mean particle size of about 30 nm	[29]
7	Oil palm empty fruit bunch pulp	Ultrasound-assisted acid hydrolysis	With 64% (w/v) of H_2SO_4 solution in an ultrasound bath At 45 °C for 2 h	Average diameter range of 30–40 nm	[18]
8	Banana fibers	Steam explosion in alkaline medium followed by oxalic acid treatment	Pressure 20 lb or 15 min, repeated for 8 times with pressure release	Length 200–250 nm Diameter 4–5 nm	[35]
10	Commercial MCC	Subcritical hydrolysis	120 °C, heating and cooling rate (6 and 3 °C/min), 60 min, pressure 20.3 MPa	242 ± 98 nm in length and 55 ± 20 nm in width	[37]
11	BNC	High-pressure homogenizer	Sugar 10% (w/v), ammonium sulfate 0.5% (w/v), and add acetic acid, bacteria starter 10% (v/v), and incubated for 10 days. at 150 bar and 5 repeated cycle	4.76 nm	[13]

BNC: Bacterial nanocellulose; *MCC*: microcrystalline cellulose nanocellulosic materials. The ball milling such as planetary ball mill, mixer ball mill, and vibration ball mill is utilized to obtain the grinding of cellulosic materials. Further, vibratory ball mill is also used for defibrillation of ramie fibers and others. Another fibrillation method to obtain nanocellulose is cryocrushing, where water-swollen cellulose fibers are first immersed in liquid nitrogen and further crushed using mortar and pestle to rapture the plant cell wall [24]. The cryocrushing method needs very high energy consumption; thus, it is not economic.

Additionally, many researchers have worked on high-pressure homogenization method, where cellulose is passed through small nozzle at high pressure (50–200 MPa), high velocity to generate shear rate to reduce the particle size into nanoscale. This method is mostly used to prepare CNF. The high shear forces, large pressure drop, turbulent flow, the number of homogenization cycles, and inter-particle collisions are the main criteria to achieve reduction of cellulose fibrils. Few disadvantages of this method are homogenizer clogging, high energy consumption, and damage of crystalline structure due to excessive mechanical shear. Thereby, various other mechanical treatments have been performed like grinding, milling, cryocrushing, etc [25]. Also, ultrasonication has attracted a considerable interest to isolate CNF, which involves application of sound energy, physical as well as chemical systems. Here, cavitation occurs with introduction of intense shear forces, shock waves, and others. The extreme environment conditions with very high temperatures, pressures, and heating/cooling rates help in cleaving the strong cellulose interfibrillar hydrogen bonding into nanofibers. A work has been made on the fabrication of NCC from oil palm empty fruit bunch pulp (EFBP) using mechanical technique, i.e., ultrasound-assisted acid hydrolysis [18].

3.4.3 Enzymatic Hydrolysis

An interesting method to produce nanocellulose fibrils in contrast to mineral acid hydrolysis methods is enzyme hydrolysis, which does not lead to toxic residues, and further, very mild thermal and pressure conditions are required for value-added applications. An investigation was performed using combination of enzymatic hydrolysis and mechanical shearing to produce NPs from sisal pulp, whereas hydrolysis using enzyme endoglucanase in microwave heating allows faster NP production compared with conventional heating [26]. Further, the morphology of cellulosic NPs can further be tuned by combined processes of mechanical shearing, acid hydrolysis, and enzymatic hydrolysis [27]. The enzymatic hydrolysis is generally obtained using cellulases, which helps in cellulosic fiber hydrolysis. Further, cellulase enzymes are categorized into three types such as endoglucanases (β-1,4-endoglucanases/A-type and B-type cellulases), exoglucanases (cellobiohydrolase, also named as C- and D-type cellulases), and β-glucosidase which can convert cellobiose to glucose [28, 29]. Among all, endoglucanases are the ones with the highest interest as it hydrolyzes the amorphous region, and exoglucanase attacks the cellulose chain from reducing or non-reducing end. Further, synergistic interactions of cellulases on crystalline cellulose in various regions of cellulose microfibril provide better activities than sum of the individual activities [30]. Chen and coworkers have reported spherical-shaped cellulose NPs having mean diameter of 30 nm using enzymatic hydrolysis from pulp fibers [29].

3.4.4 Ionic Liquid Treatments

The ionic liquids for cellulose dissolution include mainly 1-butyl-3-methylimidazolium $[C_4mim]^+$, 1-hexyl-3-methylimidazolium $[C_6mim]^+$, and 1-octyl-3-methylimidazolium $[C_8mim]^+$ cations and anions such as Cl^-, $[PF_6]^-$, Br^-, $[BF_4]^-$, $[SCN]^-$. However, the normal stirring of celluloses in such liquid ions has no dissolution as reported, but heating in a microwave oven at 100–110 °C improved the dissolution rates [31]. Also, these ionic liquids are recoverable using ease methods such as ion exchange, evaporation method, and reverse osmosis for reusability. This is a green way of developing nanocellulose without any pre-treatment method with good mechanical property. Li et al. have reported a work on developing nanocelluloses (diameter range of 10–20 nm) from sugarcane bagasse using high-pressure homogenization method coupled with a homogeneous pre-treatment method using an ionic liquid [32]. Moreover, the highly crystalline nanocellulose materials can also be obtained with the aid of ionic liquids [33].

3.4.5 Steam Explosion

Steam explosion is an effective method for the development of nanofibers from biomass using high-pressure steaming followed by rapid decompression [34]. In this method, dry materials are saturated using steam at elevated pressure and temperature followed by sudden pressure release. Thus, the thermomechanical force generated by the flash water evaporation causes rapturing of the material. This method helps in reducing the non-cellulosic compounds using steam explosion and can be utilized as pretreatment methods. The CNF obtained using this technique has higher aspect ratio than other conventional methods. The principle is based on cooking in vapor phase in a temperature range of 180–210 °C and steam pressure between 1 and 3.5 MPa. The advantages of steam explosion include low energy and chemical consumption, low environmental impact, and lower capital investment. Various sources are used to obtain nanocellulose using this method such as banana fibers followed by oxalic acid treatment [35].

3.4.6 Supercritical and Subcritical Water Hydrolysis

Another method was proposed as an alternative to acid hydrolysis termed as supercritical water hydrolysis, where pure water is heated and pressurized above critical points such as to 374 °C and 22.1 MPa, where the water properties such as density, viscosity, and dielectric constant get decreased. In supercritical reactors, a pressure of 25–30 MPa is used maintaining the temperature and time. Thereby, swollen or dissolved form of the microcrystalline cellulose can be seen in near critical and supercritical water, which enables a homogeneous hydrolysis [36], whereas use of subcritical water (obtained at 120 °C and 20.3 MPa for 60 min) is another greener pathway for CNC production with high crystallinity index of 79.0% and having onset degradation temperature around 300 °C. This process provides lower corrosive nature, low and cleaner effluent, and use of low-cost reagents [37].

3.5 Overview of Cellulose and Nanocellulose in Food Sector

Cellulose has many beneficial properties such as it functions as an anticaking agent and thickening agent, and replaces fat in some food products. However, the materials should not create cytotoxicity and genotoxicity; should follow internationally standardized regulations; and should reduce the calorie value of food products. The nanocellulose forms as nanocomposites are used as food packaging and functional nanocomposites such as gas barrier coatings and food quality analysis sensors. The use of nanocellulose in various applications specifically in food sector includes food packaging (edible and non-edible), as ingredients in food functionalization, as stabilizing agents in food emulsion, as food additives, as dietary fiber, etc. [38]. Further, vegetable nanocellulose is used in salad dressings, sauces, gravies, cake frostings, whipped toppings, etc. The use of nanocellulose has been found in sauce and soy soup, retort food, bean jam, dough based products, etc. As food stabilizers, nanocellulose-based materials are used in the preparation of foams, puddings, dips, whipped toppings, etc. Further, the food applications of BC include nata-de-coco, rheology modifiers, fat replacers, artificial meat, stabilizer of Pickering emulsions, immobilizer of probiotics and enzymes, etc. BC is used as a raw material for developing dessert and artificial meat and further is used as food ingredients for gelling, water binding, emulsifying, thickening, stabilizing agents. The noteworthy health beneficial properties of cellulose and its nanoforms provide a great interest in the current food sector for versatile application. Besides food packaging, cellulose is widely utilized in the sections of (i) renewable energy such as household industry, electricity, and biogas in traffic fuel; (ii) construction materials and industrial applications such as furniture, buildings, and panels; and (iii) development of paperboard, tissue paper, musical instruments, fertilizers, etc.

3.5.1 Use of Cellulose and Nanocellulose in Food Products

Cellulose is used widely in bakery products for obtaining improved properties and healthier food products [39]. In bakery industry, cellulose and various derivatives such as methylcellulose, CMC, MCC, HPC, and others are used to improve the dough rheology and bread texture [40, 41]. Moreover, the textural properties of pasta can be improved using CMC and the solid losses in non-wheat-based pasta can also be reduced [41]. Further, cellulose derivatives such as microcrystalline cellulose, methyl cellulose, CMC, HPC, and HPMC have a great impact in obtaining the tailor-made properties of crumb of gluten-free doughs and breads [42]. The stability of frozen dough during storage can be maintained using CMC and gum arabic, where the use of the biomaterials helps in reducing the ice crystallization in frozen dough [43]. The emulsifiers such as glycerol monostearate, sodium stearoyl-2-lactylate, calcium stearoyl-2-lactylate, and diacetyl tartaric acid esters of mono- and diglycerides are used to improve the crumb properties of gluten-free doughs and breads [44]. Cellulose derivatives as emulsions are used to replace fat in biscuits [45]. In biscuit preparations, trans-fatty acid-free fat replacers such as sunflower oil–water–cellulose ether emulsions are used to reduce fat content of biscuits [44–46]. The fat content in the biscuits need to be reduced for developing healthier food products, where methyl cellulose and HPMC have good emulsifying properties and have an ability to develop stable vegetable oil-in-water-based emulsions. The sensory of rice cakes can be modified using CMC, where the cellulose derivatives help in improving the property of gluten-free cakes [47]. Thus, cellulose holds onto a great impact in the development of various bakery products for improved properties. Further, BNC has a great potential to be used as an additive for wheat bread, where the addition of BNC provides increased average pore size and softer crumb compared to control formulation [48]. In this way, cellulose and its various forms are utilized to deliver improved food attributes. In meat industry, cellulose-based casings are extensively utilized in the preparation of meat and poultry-based ready-to-eat food products such as sausages, frankfurters, and bologna [49]. Additionally, BNC is also used as an additive in tailoring the quality and stability of low-lipid low-sodium meat sausages [50].

3.5.2 Use of Cellulose, Its Derivatives, and Nanoforms in Food Packaging

Cellulose, its derivatives, and nanostructured forms are extensively utilized in the development of edible and non-edible food packaging materials as an alternative for existing packaging materials [51–54]. Cellulose in various forms such as hydrogel, composites, edible films, and edible coatings is utilized for food packaging section. The various properties of nanocellulose in food packaging involve eatable nature, biocompatibility, transparency, antimicrobial, barrier properties against oil and grease, oxygen, water vapor, liquids, mechanical properties, flexibility, etc. The packaging application of nanocellulose is utilized via solution casting, coating, layer-by-layer assembly, electrospinning, and others. The main motivation of using nanocellulose in food industry is extending the shelf life of food products, blocking gaseous elements into food materials, improving food quality, etc. The nanocelluloses have gained extreme popularity to be acted as a nanofiller material for developing bionanocomposites.

Edible Food Packaging. The cellulose delivering various health beneficial properties and further having a capability to improve packaging properties and shelf life of food products is used in the development of edible food packaging materials. The use of cellulose and its derivatives in edible food packaging includes CMC/CS bilayer-based edible coating on citrus fruit [55], methyl cellulose/sodium alginate (SA) for peaches [56], cellulose derivatives for 'Berangan' banana [57], CS/CMC/moringa leaf extract-based edible coating for avocado fruit [58], CMC/Zataria multiflora essential oil/grape seed extract-based edible coating for rainbow trout meat [59], CMC/calcium/ascorbic acid-based edible coatings for fresh-cut apples [60], etc. The application of nanocellulose in edible food packaging includes CS/CNF for cut pineapple [51], carrageenan/cellulose nanowhisker (CNW) as edible films [61], alginate–acerola puree/cellulose whisker (CW) as edible films [62], fish myofibrillar protein/bacterial cellulose nanofiber (BCNF) as edible films [63], agar/nano-BC as edible films [64], etc.

Non-edible Food Packaging. The non-edible nanocellulose-based packaging includes the use of various nanostructured forms of cellulose as a filler material in biodegradable polymers. Several researches report the development of biocomposite-based materials for food packaging applications such as CNC-reinforced poly(lactic acid) [65, 66], CNC-reinforced poly(3-hydroxybutyrate) (PHB), CNW-reinforced poly(lactic acid) [67], paper sheets coated with wheat gluten (WG)/nanocellulose/titanium dioxide nanocomposite as active food packaging [68], and paper coated with modified nanocellulose fiber (NCF)/polylactic acid (PLA) composites [69].

3.6 Market of Cellulose-Based Packaging

The segmentation of market report on cellulose film packaging has been done based on film types, sources of cellulose extraction, targeted end use, application area, and region of development (as shown in Fig. 3.6) [70]. The market is segmented based on the sources such as cotton and wood-based sources for the fabrication of films, and the films can be obtained as colored, transparent, and metalized type depending on the synthesis type. Moreover, the various end use industries for using cellulose film packaging include food and beverage, pharmaceuticals, retail and personal care, etc. The growth of global market of cellulose film packaging is related to the

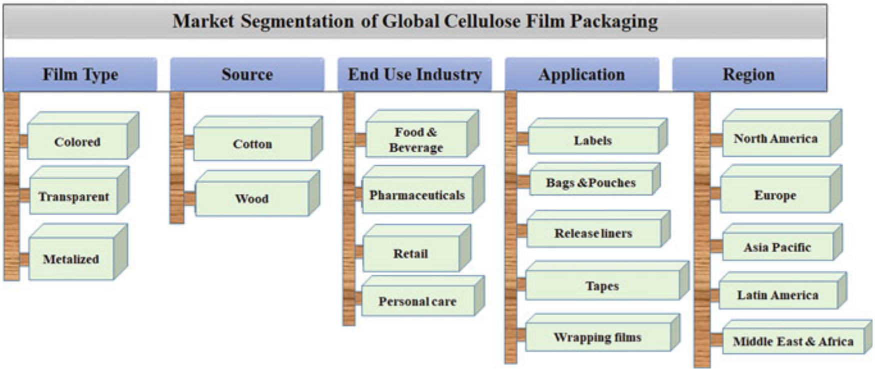

Fig. 3.6 Market segmentation of global cellulose film packaging

easy availability of raw materials and manufacturing techniques. However, the available alternatives and cost of raw materials that are required for the development of cellulose film may change the target market growth of cellulose film packaging. The region of global cellulose film packaging includes North America (USA and Canada), Europe (Germany, UK, France, Italy, Spain, Russia, and others), Asia-Pacific (China, India, Japan, Australia, South Korea, and others), Latin America (Brazil, Mexico, and others), and Middle East and Africa (GCC, South Africa, and others). The cellulose film packaging is developed by Futamura Chemical Co., Ltd., Hubei Golden Ring Co., Ltd., Celanese Corporation, Weifang Henglian Cellophane Co., Ltd., Eastman Chemical Company, Chengdu Huaming Cellophane Co., Ltd., Tembec Inc., Sappi Limited, Rotofil Srl, Rhodia Acetow GmbH, etc. In this regard, oils and fat-resistant cellulose film packaging are developed by NatureFlex^{TM}, where heat-seal resins are used for microwave applications [71].

3.7 Prospective for Edible Food Packaging Application

As discussed in the previous section, cellulose-based materials have obtained a wide usability in food sector for its noteworthy attributes. As shown in Fig. 3.7, cellulose being a non-toxic, biocompatible, widely available, renewable resource has gained an extreme enthrallment in the field of edible food packaging. Cellulose has been used for versatile application from 150 years back. The noteworthy features of nanocellulose include facile surface chemistry, promising health beneficial properties, physical and chemical properties, etc. In this regard, the section will give a brief overview about the various promising factors of nanocellulose to be used in edible food packaging materials.

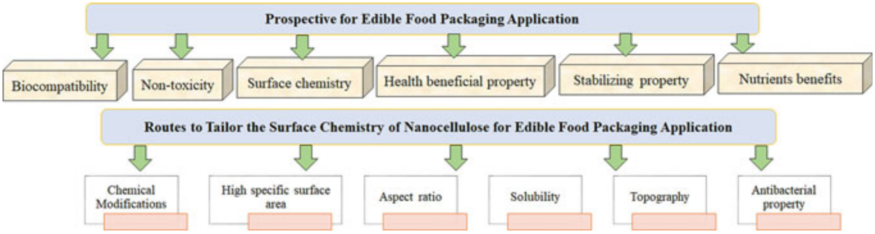

Fig. 3.7 Prospects of nanocellulose in edible food packaging

3.7.1 Biocompatibility and Non-toxicity

The biocompatibility of a material is generally defined as the compatibility of the material with the living tissues. This is a kind of biomaterial behavior, where the toxic or immunological response of the material in human body is not produced. The biocompatibility of a material does not possess toxic effect on biological systems, and the material should perform the targeted function without creating any undesirable effects in the recipient. The biocompatibility of a material is evaluated following ISO 10993 set of standards. The pure form of nanocellulosic materials is non-toxic and biocompatible, and is used widely in edible food packaging materials. The BC has the status of "generally recognized as safe" by Food and Drug Administration (FDA). Further, the compatibility and cell adhesion property of nanocellulose can be promoted by controlling the surface property of nanocellulose.

3.7.2 Surface Chemistry

Cellulose units can form intra- and interhydrogen bonding within cellulose chains and between cellulose chains. There are strong intra- and interhydrogen bonding in the crystalline region of cellulose, which makes it enable to absorb water. On the other hand, in the amorphous region, the bonding is weak, which makes them able to absorb some water. The moisture uptake of cellulose is around 8–14%, and the water uptake rate is very slow. The nanocellulose molecules can be easily functionalized due to the presence of numbers of hydroxyl groups on nanocellulose surface, which helps to provide improved dispersion in composites for acting as edible food packaging materials. The cellulose is composed of both crystalline and amorphous regions, where the CNC molecules have higher crystallinity than CNFs. The fabrication of CNC is done using strong acids, which make them more crystalline in nature ranging from 40 to 90%. The surface modification of nanocelluloses can be obtained via (i) physical modification such as utilizing surfactants (anionic, cationic, nonionic surfactants) and polymers (macromolecules and biopolymers), and (ii) chemical modification such as polymer grafting (grafting from and grafting onto) and derivatization. Further, the surface chemistry of nanocelluloses can be utilized and modified properties can be obtained using cross-linkers and compatibilizers, etc. However, the modification of nanocelluloses can be obtained via functionalization of hydroxyl groups, which increase the interaction and dispersion property, etc. However, the nanocellulosic materials have a tendency to form agglomeration due to strong hydrogen bonding; thus, the fillers need to be dispersed properly via chemical or physical modification to obtain tailored properties of polymer composites for edible food packaging materials.

3.7.3 Health Beneficial Property

The sources of cellulose include fruits with peel, green vegetables, peas, cereal brans which are directly eatable by human being. The cellulose is not considered as essential nutrients; however, the intake of cellulose is considered as health beneficial. In this regard, cellulose, its derivatives, and nanoforms have attained a great interest in the current trend of product development. Cellulose is an insoluble fiber, which has an ability to absorb water in human intestine. However, the beneficial bacteria present in large intestine can break cellulose-based materials which can be absorbed in human body. The health benefits of cellulose-based components include weight loss, lowering blood sugar, glucose level in body, lipid lowering activity, and provide a laxative effect. However, cellulose-based materials may produce excessive gas, loose stools, and allergic reactions.

3.7.4 As Emulsifying and Stabilizing Agents

The nanocellulosic materials have an ability in forming stable emulsions, which helps in improving the texture and suspension of emulsion-based coatings. The nanocellulose can be used for developing water/oil-based emulsions, and it is considered as natural emulsifier and stabilizers. The nanoemulsion-based edible coatings can also be developed using nanocellulosic materials. The functionalized nanocellulose and BNC are also used in stabilizing the oil-in-water-based emulsions.

3.7.5 Nanocellulose-Based Antibacterial Materials and Other Properties

The nanocellulose materials have very less antimicrobial and antifungal activity, which can be modified using various modes of action. The surface of nanocellulosic materials can be modified utilizing different molecules, and conjugation of antimicrobial agents with nanocellulosic materials can also be obtained using different cross-linkers. The general antimicrobial agents that are conjugated with nanocellulosic materials include organic compounds, metal NPs, antibiotics, metal oxide NPs, etc. Further, the nanocellulose-based materials can be provided with antioxidant property, when polyphenolic and active agents are added to it via surface modifications. Further, various bioactive molecules are added to nanocellulosic materials including BC for improved packaging properties.

3.8 Traits of Nanocellulose Biocomposites for Edible Food Packaging

The well dispersion of nanocellulose in various matrix materials helps to improve the characteristic attributes of packaging materials such as barrier, thermal, mechanical, wettability, and other properties to be acted as ideal packaging materials for improved shelf life of food products. Since the past decades, the nanocellulose-based polymer composites are considered as emerging materials to be used in various forms of food packaging materials. The high mechanical strength, water resistance property, stability, barrier properties make it a superior candidate for edible food packaging materials. In this regard, nanocellulose-based materials are widely utilized as a potential alternative for available petrochemical-based packaging materials. In Table 3.2, a brief discussion about the available edible packaging materials has been made.

3.8.1 Barrier Property

The nanocelluloses such as CNC, CNF, CNW, and BCN have a capability in improving the barrier properties of edible food packaging. As discussed earlier, the cellulose-based nanomaterials can form strong network via hydrogen bonding, which make gaseous molecules hard to pass through the composites by enhancing the barrier properties. The improved barrier properties of nanocellulose at its nanodimension and its composites can be related to the formation of dense and percolated network formed by nanocellulose with various dimensions. The formed dense network creates a tortuous path for the gaseous molecules and thus decreases the permeability of the films. Further, it has been observed that the oxygen barrier properties of CNF-based films are better than CNC-based films, as CNF-based films can form a high entanglement within the matrix materials. However, nanocellulose being hydrophilic in nature may provide low water barrier properties. The nanocellulose materials at high relative humidity may have a problem of structural disintegration which decrease the oxygen and water vapor barrier properties [75]. However, the nanocellulose materials can be modified to reduce its hydrophilic

Table 3.2 Packaging properties of some available edible packaging materials in terms of edible films and coatings

Sl. no.	Edible packaging composition	Packaging properties
1	CNF and mgCNF-reinforced CS [51]	**Mechanical properties:** Tensile properties: CS: ~6.27 ± 0.7 MPa; CS/1.5mgCNF: ~57.86 ± 14 MPa **Young's modulus:** CS: ~462.36 ± 64 MPa; CS/1.5mgCNF: ~2348.52 ± 276 MPa **Composition of iron:** **CS-mgCNF1.5: 6.3** ± 0.06 ppm **Thermal properties:** CS: 10% loss in weight: 80 °C; CS: 50% loss in weight: 334 °C CS-CNF0.5: 10% loss in weight: 116 °C; 50% loss in weight: 342 °C CS-mgCNF0.5: 10% loss in weight: 95 °C; 50% loss in weight: 351 °C **Transparency in UV region:** CS: 85%, CS-CNF0.5:71%, CS-CNF1:69%, CS-CNF1.5:64%, CS-mgCNF0.5:42%, CS-mgCNF1:33%, CS-mgCNF1.5:18%
2	CNF-reinforced CS [72]	**3% CNF/CS (1%)** Color: L:86.3, a:−0.64, b:3.02; WVP: 14.15 g.mm/kPa.d.m^2 **3% CNF/CS (2%)** Color: L:71.7, a:−0.60, b:6.08; WVP: 13.62 g.mm/kPa.d.m^2
3	Nanocellulose fibers-reinforced sodium caseinate [73]	**1 wt% nanocellulose fibers-reinforced sodium caseinate** Opacity: 2137.7 ± 434.0 AU nm/mm; contact angle: 32.8 ± 4.2°; Average porosity: 0.116% **3 wt% nanocellulose fibers-reinforced sodium caseinate** Opacity: 3215.2 ± 214.9 AU nm/mm; contact angle: 29.7 ± 1.5°; Average porosity: 0.194%
4	CNC-reinforced pectin [74]	**2% CNC-reinforced pectin films** Glass transition temperature (Tg): 57.9 ± 0.66 °C; melting temperature (Tm): 167.79 ± 5.56 °C; melting enthalpy (ΔH): 128.65 ± 1.27 J g^{-1} **7% CNC-reinforced pectin films** Tg: 57.39 ± 0.39 °C; Tm: 165.36 ± 0.94 °C; melting enthalpy (ΔH): 140.5 ± 7.5 J g^{-1}
5	Bacterial CNF (BCNF)-reinforced fish myofibrillar protein [63]	**2 wt% BCNF-reinforced fish myofibrillar protein** TS: 6.84 ± 0.09 MPa; elongation at break: 90.49 ± 2.93%; WVP : ~2.95 × 10^{-10} g/ms Pa; contact angle: ~82.76° **6 wt% BCNF-reinforced fish myofibrillar protein** TS: 8.94 ± 0.33 MPa; elongation at break : 87.63 ± 2.52%; WVP: ~2.28 × 10^{-10} g/ms Pa; contact angle: ~87.70°

(continued)

Table 3.2 (continued)

Sl. no.	Edible packaging composition	Packaging properties
6	Nano-BC-reinforced agar [64]	**3 wt% nano-BC-reinforced agar films** Moisture content: ~19.50%; WS: ~21.56% TS: ~27.95 MPa WVP: ~8.69×10^{-11} gm/m^2 Pa s **10 wt% nano-BC-reinforced agar films** Moisture content: ~13.79%; WS: ~18.55% TS: ~44.51 MPa WVP: ~6.88×10^{-11} gm/m^2 Pa s

WVP: Water vapor permeability; *TS*: tensile strength; *WS*: water solubility; *UV*: ultraviolet; *mgCNF*: magnetic cellulose nanofibers

property at high-moisture environments via chemical modifications. In this regard, the metal oxides can be used to modify properties of nanocellulose for improved barrier properties. As discussed, CNFs can be developed by mechanical fibrillation method, where the pure films of these kind of CNFs have very good oxygen barrier properties. Interestingly, CNF provides tailor-made barrier properties, where partially acetylated CNF-based films are also utilized for modified atmospheric packaging. The barrier properties of nanocellulose-based composites are better than the available synthetic polymer that are used in the food packaging industry. The CNF-based films are more complex and compact in comparison with the cellulose fiber-based films, which make them a potential candidate for water barrier properties. The ordered structure of nanocellulose and crystallinity behavior of nanocellulose create a percolation network in composites which slower the diffusion of gaseous molecules, thus reducing water vapor permeability (WVP).

3.8.2 Mechanical Property

Crossbonding in cellulose-based nanomaterials provides the maximum strength in composites. CNFs have an exceptional mechanical property and thus are used as a potential candidate in food packaging materials for transportation purpose. The fruits and vegetables are very much prone to get effected by mechanical damages such as abrasion during transportation. The mechanical properties of nanocellulosic materials are better than the available lignocellulosic biomass materials for having uniform morphological structures. The mechanical properties of nanocellulose also depend on the percent crystallinity, which in turn depends on the source. The nanocellulosic materials are available in its various polymorphic forms which further differ the composite properties.

3.8.3 Thermal Property

The nanostructured forms of cellulose are thermally stable than the cellulosic materials. The thermal properties are dependent on the source materials, extraction procedure, condition of processing, etc. The nanocellulosic materials provide higher crystallinity (may get varied for nanofibers, nanocrystals, whiskers, etc.), flexible orientation, and removal of non-cellulosic materials, which in turn improves the thermal stability of nanocellulosic materials. The composites of nanocellulose are influenced by the intermolecular bonding between filler and matrix materials. The surface chemistry of nanocellulose helps to improve the thermal properties due to intermolecular interaction forming percolation network. However, nanocellulose extracted using sulfuric acids contains sulfate groups, which reduces the thermal stability of nanocellulose materials. However, the sulfate groups on nanocellulose can be functionalized to obtain improved thermal stability.

3.8.4 Optical Property

The optical property of nanocellulosic composites is influenced by various types of cellulosic nanomaterials such as CNC, CNF, CNW, and others. The transparency and color coordinates (L, a*, b*, hue, and chroma) are found to influence by the inclusion of functionalized nanocellulosic materials in different matrix materials.

3.8.5 Others

The other properties of nanocellulosic composites such as moisture content, water solubility (WS), wettability, and thickness are affected greatly by the different types of matrix materials.

3.9 Tailored Nanocellulose-Based Materials for Edible Food Packaging

As discussed earlier, edible coating is an environmental-friendly technique for providing a protective coating layer to maximize the quality and shelf life of fresh fruits and vegetables. Focusing on different biopolymers such as polysaccharides, alginates, CS, proteins, gelatin (fish/pork), and lipids has been gaining an increased attention regarding its excellent cost-effectiveness nature and film-forming function [76–80]. The biodegradable, non-toxic, high surface area with high crystallinity nanocellulose including NFC, CNC, and BNC with their unique barrier layer properties such as enhanced oxygen and water vapor properties is used for food packaging. The percolated structure of CNF and its long, flexible fiber structure with good mechanical properties provide proper dispersion in the matrix. Various investigations have been put forward to utilize nanocellulose-derived biopolymers as edible films in food packaging. An article was reported on edible films from pectin, a natural polymer reinforced with crystalline nanocellulose (2, 5, and 7 w/w %) developed using solution casting evaporation method [74]. The most important functional properties in case of edible films and coatings are their barrier properties for water vapor and oxygen, carbon dioxide gases, migration of compound, appearance such as color and gloss, physical and mechanical protection for proper handling of food products. The high surface area feature of nanoscale cellulose demonstrates its application as reinforcements for polymer matrix giving a new research area. However, its poor dispersion is a critical concern, which affects the mechanical properties of the composite materials leading to a challenging task. Thereby, modification of the nanofiller is needed for good dispersion in the polymer materials via physical and chemical methods [81, 82].

3.9.1 Chemical Modification

The chemical modification of nanocellulose can be obtained via utilizing the available hydroxyl groups on nanocellulose. The chemical agents such as compatibilizing agent, coupling agent, acetylating agent, polymer grafting agent, copolymerization, non-covalent surface modification, sulfonation, TEMPO (2,2,6,6- tetramethylpiperidine-1-oxyl)-mediated oxidation, esterification, etherification, and silation are used to improve the properties of nanocellulose for food packaging application. The chemical modifications further help to improve the interfacial compatibility with the various polymer matrices by changing and controlling the molecular interactions. However, in case of surface compatibilization of nanocelluloses, one reactive group should be present. On the other hand, for the copolymerization-aided modifications, at least two functional groups are required for improved interactions. In this case, the functional group in cellulose can react with polymer matrix via grafting, radical reactions, and organometallics for improved properties [83, 84].

3.9.2 Physical Modification

The physical treatment of cellulose is a challenging method to alter the cellulose polymorphs, thereby improving the matrix mechanical bonding and other properties [85]. The various physical methods such as cold plasma (electric discharge), dielectric-barrier discharge (creation of ions, free radicals, and other species generated by high energy electrons), ultrasonification (include no use of organic solvent), and irradiation by gamma rays are used to enhance the CNC's adhesion properties, and further, mechanochemical treatment is applied through shearing and compressing forces, etc [86–89].

3.9.3 Development of Bionanocomposites

The alternative of petroleum-based plastics such as the active-biobased food packaging has received a great attention for improved hydrophobicity, mechanical properties, and antimicrobial activity of individual biopolymers. Li et al. have reported that developing CW with CS matrix displays increase in tensile strength (TS) up to 120 MPa by incorporating 20% CNW along with excellent thermal stability and water resistance [90]. A work has been reported on the preparation of bionanocomposites using casting/evaporation method based on WG, CNC, and titanium NPs (TiO_2), which provide improved breaking length of 56% and burst index of 53% by incorporating 7.5% CNC and 0.6% TiO_2 for 3 coating layers over kraft paper. The bionanocomposite also showed excellent antimicrobial activities almost 100% against yeast *Saccharomyces cerevisiae*, gram-negative bacteria *Escherichia coli*, and gram-positive bacteria *Staphylococcus aureus* [68]. Seoane and coworkers prepared PHB/CNC bionanocomposite that has attained improved mechanical and barrier properties, when optimized concentrations of CNC (6 wt%) have been incorporated, which further act as an alternative for polypropylene in packaging applications [91]. Dehnad et al. studied the shelf life of meat that can extend meat shelf life and provide maintained quality for 6 days when compared with nylon packaging [92]. Jung et al. have investigated developing coating using Fe^{3+}-anthocyanin complexation with CNF/SA using layer-by-layer (LBL) coating method on blueberries and found that leakage of anthocyanin pigments can be eliminated [93]. Another work has been reported demonstrating the tailor-made properties of optical, mechanical, thermal, and texture properties using CS and magnetic CNF as edible nanocoating. The investigation shows improvement in storage quality of the coated cut pineapple using texture and gravimetric analysis. Another study on acerola puree and alginate reinforcing with CW provides improved water vapor barrier property in the form of nanocomposite edible film [62].

3.10 Case Studies on Nanocellulose-Based Edible Food Packaging

In this section, a brief discussion has been made about the use of various nanoforms of cellulose such as CNC, CNF, BNC, and CNW, which are utilized with other biopolymers as edible food packaging materials. However, the characteristic attributes of fabricating composite-based edible packaging can be modified using plasticizers, flavoring agents, and others as shown in Table 3.3. Different types of cellulose-based materials are utilized as primary, secondary packaging, flexible packaging, developing containers, etc. However, in this section, the use of nanocellulose as a component of edible packaging materials has been made.

Table 3.3 Application of cellulose nanostructured materials in edible food packaging

Sl. no.	Filler type	Matrix type	Targeted food product and obtained property	References
1	Nano-BC	Agar	Edible films (targeted food packaging application) **Property:** Tailor-made properties of dispersibility of nanofiller, thermal properties, mechanical properties, moisture content, WS, WVP	[64]
2	BCNC	Gelatin	Edible films (low-cost food packaging materials) **Property:** Improve properties of gelatin and BCNC-based edible films, improve mechanical and thermal properties, reduced moisture affinity of neat films	[102]
3	CNF	Mango puree	Edible films **Property:** Improve mechanical property Improve barrier property	[103]
4	MCC NPs	HPMC	Edible Films **Property:** Better moisture barrier properties, reduced water barrier properties	[98]
5	CNC	Gelatin	Edible coating on fresh strawberry **Property:** Antimicrobial effect Reduce weight loss Prolong shelf life Retention of ascorbic acid	[94]
6	CNF and magnetic CNF	CS	Edible coating on cut pineapple **Property:** Improve mechanical property Improved thermal property Improved optical property Maintained quality of cut pineapple during storage	[51]
7	BCNF	Fish myofibrillar protein	Edible films **Property:** Improved thermal property Reduce WVP Reduced swelling index	[63]
8	CW	Alginate–acerola puree Plasticizers: Corn syrup	Edible films **Property:** Improved TS Improved Young's modulus Improved water vapor barrier	[62]

(continued)

Table 3.3 (continued)

Sl. no.	Filler type	Matrix type	Targeted food product and obtained property	References
9	Crystalline nanocellulose	Pectin	Edible films **Property:** Improved TS Improved elongation at break Improved water barrier properties	[74]
10	CNF	Gelatin Glycerol (A plasticizer)	Edible films **Property:** Tunable mechanical property Tunable optical property Tunable barrier property	[104]
11	CNF Titanium dioxide REO	WPI	Edible films Food products: Lamb meat **Property:** TiO_2 and REO improve the organoleptic property of lamb meat Increase shelf life of lamb meat Inhibit the growth of spoilage and pathogenic bacteria in meat	[101]
12	Nanocellulose	CS Glycerol (plasticizers)	Edible films **Property:** Good dispersion Tunable properties in terms of TS, elongation at break, Young's modulus, barrier properties, thermal properties	[99]
13	CNF	CS	Edible films and active packaging **Property**: Antimicrobial property Barrier property	[100]
14	NCF	Sodium caseinate Glycerol as plasticizer	Edible films **Property:** Less transparency for composite films More hydrophilic than neat sodium caseinate films Improve mechanical property with the aid of nanofiller material	[73]

CNF: Cellulose nanofiber; *CS*: chitosan; *BCNC*: bacterial cellulose nanocrystal; *BC*: bacterial cellulose; *MCC*: microcrystalline cellulose; *TS*: tensile strength; *HPMC*: hydroxypropyl methylcellulose; *CNC*: cellulose nanocrystal; *REO*: rosemary essential oil; *WPI*: whey protein isolate; *CW*: cellulose whisker; *NPs*: nanoparticles; *NCF*: nanocellulose fiber; *WS*: water solubility

3.10.1 Storage Study of Edible Packaged Food Products

Nanocellulose has been widely utilized for the fabrication of composite materials to be used as edible coating materials for enhanced shelf life of food products as given below:

- Storage study of fresh strawberry has been done using edible coating based on CNC-reinforced gelatin plasticized with glycerol (storage condition: under refrigeration for 8 days) [94].

- Storage study of strawberries has been done using apple pectin, CNCs, essential oils of lemongrass (storage condition: 8 days and 10 °C ± 1 °C) [95].
- Storage study of pears has been done using CNC-reinforced CS-based edible coating (storage study: 3 weeks and ambient storage: 20 ± 2 °C and 30 ± 2% RH) [96].
- Storage study of cut pineapple has been done using magnetic CNF (mgCNF)-reinforced CS-based edible coating (storage condition: 12 days at room temperature) [51].
- Storage study of grape fruits has been done using CNF, carrageenan, glycerol, and HPMC (storage condition: refrigerated storage for 41 days) [97].
- Storage study of strawberry has been done using CS and cellulose nanofibril-based composites (storage condition: cold storage for 21 days) [72].

3.10.2 Tunable Packaging Property as Edible Packaging Materials

The nanocomposite based on CNF-reinforced mango puree provides tunable packaging properties for developing edible films. The incorporation of CNF provides improved mechanical properties in terms of TS and Young's modulus for forming a fibrillary network within mango puree. The development of edible films based on MCC and lipid-coated MCC NP (LC-MCC) incorporated HPMC has a promising attribute in the field of food packaging for being flexible and transparent materials. The TS of MCC and LC-MCC-incorporated HPMC films provide increased TS up to 53 and 48%, respectively, in comparison with neat HPMC films [98]. Additionally, the fabrication of CNF and magnetic CNF (mgCNF)-dispersed CS-based edible coating materials can adequately improve the packaging quality and shelf life of cut pineapple in terms of firmness and weight loss [51]. As shown in Fig. 3.8, the tailored surface morphology of CNF (Fig. 3.8a) can be obtained via developing mgCNF (Fig. 3.8b), where iron particles get adsorbed onto CNF via single-step coprecipitation method. Further, the use of CNF, CS, and curcumin can provide uniform edible films (Fig. 3.8c). Further, the appearance of stored uncoated (e″) and dipped coated (e′) cut pineapple using curcumin-loaded CNF-dispersed CS solution at refrigerated storage for 1 month time duration has been shown. In this regard, several types of nanocellulosic materials are used to improve the appearance and quality of various food products during storage time. Further, CW from cotton fibers is used as a filler material in developing composites of alginate–acerola puree with plasticizer which can be used as edible food packaging material with a tailor-made property of overall tensile property and water vapor barrier property [62]. The fabrication of pectin/crystalline nanocellulose-based biocomposite edible films delivers an improved film property in terms of mechanical property, and barrier property [74]. Further, this kind of pectin-based packaging materials can be considered as fully biodegradable and renewable packaging materials for food products. The properties of CS-based edible films can be tailor-made with the aid of nanofiller nanocellulose and glycerol as a plasticizer. With the aid of central composite design considering two variables, CNF concentration (0–20 g/100 g) and glycerol (0–30 g/100 g) are used for optimizing the development of CS-based edible films, where with 15 g/100 g CNF concentration and 18 g/100 g glycerol concentration are considered as optimum condition [99]. The CNF-dispersed CS films have antimicrobial property, with an efficacy in improving barrier property [100]. Further, Pereda and others have developed sodium caseinate/nanocellulose-based edible films, where the TS and tensile modulus have been found to be improved with increased nanocellulose [73]. The application of CNF, TiO_2, rosemary essential oil (REO), and whey protein isolate (WPI)-based edible nanocomposite films helps in improving the sensory quality, organoleptic properties, and shelf life of lamb meat [101]. The synthesis of biocomposite films has a great interest in delivering the controlled release of antimicrobials. The fabrication of WPI/CNF composite films containing TiO_2 and REO is very efficient in preserving quality of meat for maintaining food properties, where the films with TiO_2 and REO have an ability to inhibit pathogenic bacteria in meat, and are considered economic. The nanocellulose and CS-based edible films in packaging ground meat have an ability to decrease the lactic acid bacteria, which helps in improving the shelf life of ground meat [92]. Further, the storage study of grape fruits with edible coating application using CNFs, HPMC, Ꝁ-carrageenan has been reported, where application of spray system is utilized for coating fruits [97].

BC is obtained in its pure form and provides superior mechanical, thermal, and water holding property, which make it a suitable candidate for edible food packaging in comparison with plant-based cellulose. The biocomposite edible films based on BCNF-reinforced fish myofibrillar protein have a positive impact in improving the film properties such as physical property, thermal property, and others [63]. The addition of 6 wt% of BCNF in the preparation of composites improves the TS about 49% than the neat films. The composite films further improve the physical properties and thermal properties, and reduce WVP and solubility indexes. The TS of the composite edible films for BCNF-reinforced fish myofibrillar protein is ~ 6, ~ 6.84, ~ 7.19, and ~ 8.94 MPa for 0, 2, 4, and 6 wt% nanofiller, respectively. Moreover, the WVP of the composite edible films for BCNF-reinforced fish myofibrillar protein is $\sim 3.41 \times 10^{-10}$ g/ms Pa, $\sim 2.95 \times 10^{-10}$ g/ms Pa, $\sim 2.73 \times 10^{-10}$ g/ms Pa, and $\sim 2.28 \times 10^{-10}$ g/ms Pa for 0, 2, 4, and 6 wt% nanofiller, respectively. Interestingly, the swelling index of the developed edible films is also improved ~ 215, ~ 212, ~ 199, and ~ 172% for 0, 2, 4, and 6 wt% nanofiller, respectively. The use of nanofiller nano-BC in the development of agar-based edible films has an improved thermal stability and mechanical properties [64]. The various contents of nano-BC have different effectivenesses in agar-based edible film preparation such as: (i) 3–5% nano-BC has a good dispersion in agar-based films; (ii) 10% nano-BC decreases moisture content by 60.4% compared with neat agar films; (iii) 10% nano-BC decreases WS by 13.3% compared with neat agar films; (iv) 10% nano-BC decreases WVP by 25.7% compared with neat agar films; (v) 10% nano-BC-dispersed agar-based edible films provide TS and elongation at break of 44.51 MPa and 13.02%, respectively; (vi) maximum mass loss

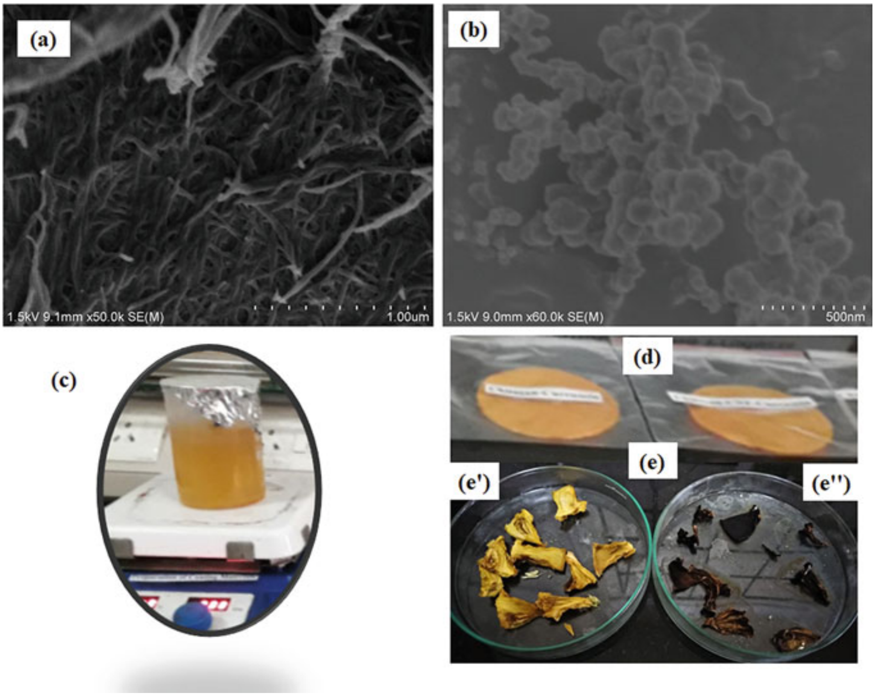

Fig. 3.8 Surface morphology of (a) CNF, (b) functionalized CNF (mgCNF), (c) Preparation of edible coating solution, (d) Edible Films and (e) storage of cutpineapple using CNF-dispersed CS-based edible coating at refrigerated storage for 1 month time duration with edible coating (e') and without edible coating (e")

temperatures for neat agar and 10% BC-reinforced agar-based edible films are 303.9 and 315.6 °C, respectively [64]. As discussed, BC as long fiber form can be obtained from *G. xylinus* [102]. The BCNC is obtained following acid hydrolysis process at various processing conditions. The use of bacterial cellulose nanocrystal (BCNC) as a nanofiller material in developing gelatin-based nanocomposite forms a percolation network, which resulted in improved mechanical property such as incorporation of 3 and 4 wt% BCNC in gelatin-based matrix having TS of 103.1 and 108.6 MPa, respectively. Additionally, the reinforcements of BCNC improve the thermal stability, degradation temperature, and dynamic mechanical property of gelatin.

3.11 Conclusion

The noteworthy properties of nanocelluloses such as abundancy, renewable resource, reduced carbon footprint, biocompatible, non-toxic, high strength, light-weight, dimensional stability, thermal stability, optical transparency, and reduced oxygen permeability make it a potential candidate to be used in edible food packaging. Moreover, the nanocellulose-based edible coating has been utilized with other applied packaging technology for obtaining improved shelf life of perishable food products. In this regard, the synergistic effect of edible nanocoating and non-edible food products as secondary packaging materials can be utilized for transportation of food items. Further, research is going on to apply various packaging technologies such as reduced oxygen storage, passive and active modified atmospheric storage, controlled atmospheric storage for improved shelf life of food products.

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