

Chapter 5

Benefits of Chitosan-Based and Cellulose-Based Nanocomposites in Food Protection and Food Packaging



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Contents

1	Introduction.....	121
2	Chitosan.....	123
2.1	Chitosan-Based Nanocomposites for Food Protection.....	125
2.2	Chitosan-Based Nanocomposite for Food Packaging.....	129
3	Nanocellulose and Bacterial Cellulose.....	136
3.1	Nanocellulose-Based Nanocomposites Used for Food Protection.....	137
3.2	Nanocellulose-Based Nanocomposites Used for Food Packaging.....	138
4	Conclusion.....	147
	References.....	148

1 Introduction

Potential risks represented by nonbiodegradable plastics are one of the serious problems which must be solved in the near future and therefore nowadays interest is growing to consume food products without chemical additives, which requires design of modern technologies to prevent food contamination and to reduce food-borne illnesses (Khan et al. 2016). Din et al. (2020) presented an overview of

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recently engineered biodegradable plastics utilized for packaging applications, use of which can contribute to reduction/elimination of waste accumulation in water reservoirs and utilizing of noxious chemical reagents and can provide cheap and biodegradable materials of natural origin as an alternative to commonly used plastics. Bio-nanocomposites (NCs) consist of two major components, in which biopolymer (continuous phase) is a matrix and the second component is reinforcement agent (dispersed phase) showing nanoscale dimensions (1–100 nm) (Sharma et al. 2020). Bio-NCs that are usually prepared by solution intercalation or melt intercalation from biodegradable resources (e.g., polysaccharides, lipids, proteins, and biodegradable synthetic polymers reinforced with nanofillers) may be utilized also for fabrication of nanofibers in order to enhance stability and improve bioavailability and controlled release of the active ingredient. Moreover, they are suitable to be used as active packaging enabling prolongation of the shelf life of food (Maftoonazad and Ramaswamy 2018; Jampílek and Kráľová 2018a). Different characteristics of biodegradable polymers used in food packaging applications were overviewed by Wroblewska-Krepsztul et al. (2018). However, biopolymers face some constraints, particularly due to their poor mechanical and barrier properties (Souza et al. 2017). For example, biodegradable packaging films and coatings fabricated from polysaccharides (e.g., such as cellulose, starch, chitosan (CS), alginate, pectin) exhibit fine barrier properties against the transport of O₂ and CO₂, while tensile strength (TS) of these films vary from each other and the values of percentage of elongation are far from reaching the values estimated for synthetic polymers (Cazon et al. 2017).

Agricultural crops, fruits, vegetables, and food products require packaging for protection from physical damage, contamination, deterioration, and to ensure increased shelf life and facilitated transport to consumers (Prasad et al. 2014, 2017a, b). Besides suitable physical and mechanical properties, the packaging must ensure that the packed products will not have any foul odor (Samanta et al. 2016; Jampílek and Kráľová 2018a). In order to suppress food deterioration, access to O₂ of food products may be suppressed via removal of O₂ from the packaging headspace and/or by addition of antioxidants to films and coatings resulting in improved antioxidant properties of food surfaces (Sahraee et al. 2019). Recent progress in the use of different edible coatings for shelf-life prolongation of fresh-cut vegetables and fruits was presented by Yousuf et al. (2018).

Traditional food packaging consists of nondegradable plastics, which is rarely recyclable and produces immense waste adversely impacting the environment. Therefore, recently increased attention is focused on the development of biodegradable bio-based materials able to extended shelf life of food products. However, as mentioned above, such bio-based materials are characterized with insufficient barrier and worse mechanical properties compared to conventional food packaging materials, which could result in a shorter shelf life of food. The physico-mechanical and functional properties of bio-based polymers can be improved and their antimicrobial activity enhanced by incorporation of different nanomaterials and such bio-NCs films can efficiently prevent food from the spoilage (Samanta et al. 2016; Al-Tayyar et al. 2020). Antimicrobial agent(s) incorporated in polymer film in antimicrobial packaging can suppress the growth and activities of targeted harmful microorganisms (Sung et al. 2013; Jampílek and Kráľová 2017). For example, many

plant essential oils (EOs) exhibit significant antimicrobial efficacy but due to their low water solubility, bioavailability, volatility, and stability in the food system, their application in food packaging in order to replace chemical preservatives harmful to health requires appropriate encapsulation into nanoscale carriers to form nanoemulsions, microemulsions, or solid lipid nanoparticles (NPs) (Prakash et al. 2018; Jampílek et al. 2019). By the incorporation of EO or their components into edible/biodegradable films reduced doses of active ingredients are necessary to obtain comparable biological effects (Sanchez-Gonzalez et al. 2011; Jampílek and Kráľová 2018a, 2018b). Nanostructured antimicrobials are characterized with higher surface area-to-volume ratio when compared with their bulk counterparts (de Azeredo 2013; Jampílek and Kráľová 2015). The biodegradability and compostability of food nanopacking materials were discussed by Gutierrez (2018).

By addition of NPs with antimicrobial properties, e.g., metal NPs to polymers antimicrobial packaging materials ensuring enhanced quality and safety of food during extended storage of food products could be fabricated (Jampílek and Kráľová 2018a; Dobrucka and Ankiel 2019; Prasad et al. 2017c). The recent findings related to antimicrobial compounds of natural origin incorporated in nanoscale structures exhibiting pronounced antimicrobial activity against pathogenic microorganisms attacking food products were summarized by Lopes and Brandelli (2018). For introduction of antimicrobial compounds into food packaging materials, direct and indirect techniques can be utilized and controlled release of antimicrobial agents from food packaging materials is desirable (Jampílek and Kráľová 2018b). However, such active packaging materials must have required barrier properties, transparency, TS, and other characteristics ensuring the necessary food protection and food safety (Khaneghah et al. 2018). Impact of various additives on functional properties of composites prepared from seaweed-derived polysaccharides (alginate, carrageenan, and agar) showing biocompatibility, availability, gelling capacity, and encapsulation efficiency (EE), which can be used for food and pharmaceutical applications, was analyzed by Khalil et al. (2017).

This chapter comprehensively summarizes the findings related to CS-based and cellulose-based NCs suitable for food protection and food packaging. Beneficial impact of nanofillers including cellulose nanofibers and nanocrystals as well as cellulose and CS nanowhiskers in NC biopolymer films on the improvement of mechanical properties and thermal stability and essential oils or metal NPs, which could pronouncedly contribute to enhanced antimicrobial activity of NC films, are highlighted. CS- and cellulose-based NC films suitable for edible coatings and extension of shelf life of food products are discussed as well.

2 Chitosan

Chitosan (CS) is deacetylated derivative of chitin, a high-molecular-weight linear polycationic heteropolysaccharide consisting of β -(1 \rightarrow 4)-linked 2-amino-2-deoxy-D-glucopyranose (β -D-glucosamine) and *N*-acetyl- β -D-glucosamine copolymers (Fig. 5.1). It can be prepared by partial alkaline *N*-deacetylation of chitin that is

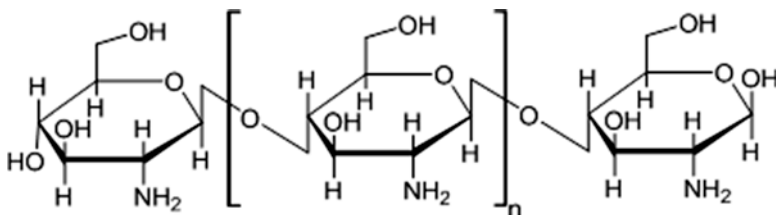
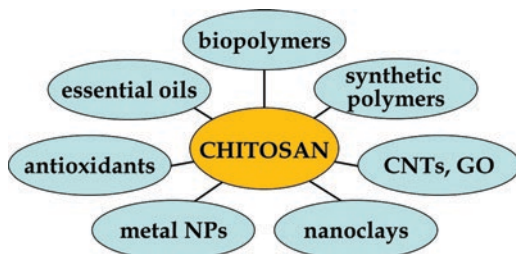


Fig. 5.1 Structure of chitosan

Fig. 5.2 Most frequent constituents on chitosan-based nanocomposites



usually extracted from shrimp and crab shells (Kumar et al. 2020). CS is biocompatible, biodegradable, and hydrophilic, can be easily modified and form films, gels, NPs, microparticles (MPs), and beads, whereby it is nontoxic, shows high bioavailability, and chemical resistance. Moreover, it has affinity to metals or proteins and shows excellent antimicrobial activity and therefore it is frequently used in pharmaceutical/medicinal applications as well as in food industry (Shariatnia 2019; Tyliaszczak et al. 2019).

CS-based films with multiple functionalities could be fabricated by different methods, including direct casting, dipping, coating, extrusion, and layer-by-layer assembly and in food packaging applications, they are frequently utilized as antibacterial, barrier, and sensing films (Wang et al. 2018a).

To the factors affecting the antimicrobial activity of CS belong the environmental conditions such as pH, neighboring components, physical form, and the structural properties of the cell wall of the target microorganisms as well as its molecular weight, degree of deacetylation and degree of substitution of CS, its concentration, and original source. The antimicrobial potential of CS can be improved by incorporating in nanoscale systems with further biologically active compounds of natural origin, metals, or drugs (Hosseinnejad and Jafari 2016; Perinelli et al. 2018). Most frequent constituents on CS-based NCs are shown in Fig. 5.2.

2.1 *Chitosan-Based Nanocomposites for Food Protection*

CS was reported to cause simultaneous permeabilization of the cell membrane to small cellular components associated with a considerable membrane depolarization of bacteria resulting in pronounced antibacterial properties (Raafat et al. 2008). As mentioned above, the biodegradability, significant antimicrobial effectiveness, and film-forming properties of CS biopolymer predestine its use in food packaging and food preservation to ameliorate food quality and safety and prolong the shelf-life of fresh meat, dairy products (e.g., cheese), and bread in the form of CS-based films or coatings (Kumar et al. 2020). An overview devoted to CS-based edible films and coatings was presented by Elsabee and Abdou (2013). Basavegowda et al. (2020) in a review paper focused on the mode of action, toxicity, and food shelf life-enhancing effects of bimetallic and trimetallic NPs exhibiting strong antimicrobial and antioxidant properties applied in food packaging.

Thin layer formed on food surface, which can be eaten as an integral part of the food product, is known as edible packaging. Edible coating formed directly on the food surface as thin layer can improve shelf life of vegetables and fruits. On the other hand, the edible films could be formed also as thin layers separately and wrapped on food surface later. However, because the properties of such films (e.g., the mechanical properties, water vapor permeability (WVP), film color) do not achieve those of conventional packaging materials, it is desirable to improve them by incorporation of nanomaterials–nanofillers (e.g., nanostarch, nanocellulose, CS NPs/chitin NPs, nanoproteins, and nanolipids) (Jeevahan and Chandrasekaran 2019a). The quality of edible coatings and films is affected mainly with temperature, environmental humidity, interactions of polymer chains, and the hydrophilicity/lipophilicity of packaging materials (Sahraee et al. 2019). Xing et al. (2019) presented a comprehensive review related to fabrication, antimicrobial properties, and respective mechanisms of action, surface, and physical qualities of edible coatings and films with incorporated NPs showing antimicrobial activity and their application to vegetables and fruits as well. The antimicrobial activity of edible coatings and films could be connected primarily with (i) the electrostatic interaction between the cationic polymer or free metal ions and the negatively charged cell membrane, (ii) the photocatalytic reaction of NPs, (iii) the detachment of free metal ions, and (iv) partially also with the antimicrobial effectiveness of edible materials. Due to the interaction between biopolymers (e.g., CS, starch) and nanofillers (clay, inorganic, organic, or carbon nanostructures) the functionality of NC materials can be improved resulting, for example, in better barrier properties, improved mechanical strength, antioxidant and antimicrobial effectiveness or thermal stability, which can be utilized in the food industry and for food packaging (Jamroz et al. 2019). Active food packaging systems able to release antimicrobial agents into food can improve food quality and/or stability. Controlled release of antimicrobials from the packaging material and need of smaller amount of active ingredient for the same effect as well as low costs are considered as benefits and could be intensified by application of nanostructured antimicrobials. However, before practical application of such

packaging materials migration of nanostructures onto food and their possible toxicity could be estimated (Azeredo et al. 2019).

The CS and nanoclay NC, which exhibited fine antibacterial activity when introduced into a film at 3 and 1%, respectively, was able to prevent Gouda cheese against microbial contamination caused by *Escherichia coli*, coagulase-positive *Staphylococcus*, *Salmonella* spp., or mold and yeast (Mohammadzadeh-Vazifeh et al. 2020). Bio-NCs based on CS-reinforced montmorillonites (MMTs) (Cloisite® Na⁺ or Cloisite® Ca²⁺) incorporated with *Rosmarinus officinalis* EO or *Zingiber officinale* EO, which were tested as active packaging to fresh poultry meat stored 15 days at 5 ± 2 °C effectively prolonged the shelf life of tested fresh meat and it was observed that compared to neat CS film the nanoclays incorporated in biocomposite suppressed lipid oxidation by 50% and microbiological contamination by 6–16% and the EOs improved the barrier to oxidation (Pires et al. 2018). NC CS films prepared by addition of a hybrid fabricated by adsorption of thyme EO into sodium–MMT or organomodified MMT exhibited excellent antioxidant effectiveness and significant antimicrobial activity against *E. coli* and in tests performed with chicken breast fillets packaged under vacuum low lipid oxidation values were observed suggesting that such films could be used for active packaging of food products (Giannakas et al. 2019).

Polyethylene terephthalate punnets containing thyme oil and sealed with CS/boehmite NC lidding films pronouncedly decreased the incidence and severity of brown rot caused by *Monilinia laxa* in artificially inoculated peach fruits (cv. Kakawa) held at 25 °C for 5 days as well as decreased incidence of brown rot to 10% when naturally infected fruits were stored for 7 days at 0.5 °C and 90% relative humidity (RH) or for 3 days at the simulated market shelf conditions (15 °C, 75% RH) (Cindi et al. 2015).

Coating green tomatoes with a nanoscale SiO₂/CS complex film delayed weight loss and softness as well as loss of the titratable acids and total soluble solids in green tomatoes resulting in significantly extended shelf life of fruits and prevented them from oxidation stress. Favorable impact of this film on postharvest quality of tomatoes was probably caused by the lower rate of O₂/CO₂ transmission coefficient, suppressed growth of foodborne pathogenic bacteria, higher activities of antioxidants, and scavenging of reactive oxygen species (ROS) as well as anti-browning activities of related enzymes (Zhu et al. 2019).

Alginate (ALG) and ALG/CS composite films (fabricated using component ratio of ALG/CS of 65:35 and 82.5:17.5, respectively) containing natamycin inhibited the growth of *Debaromyces hansenii*, *Penicillium commune*, and *Penicillium roqueforti* already at concentration 0.01 g natamycin per gram of biopolymer, while those with fourfold greater concentration of natamycin exhibited beside very good antimicrobial also suitable functional attributes (da Silva et al. 2013). Gelatin/CS nanofibers prepared using a ratio 1:6 containing 40% of encapsulated Shirazi thyme with diameters 97–343 nm showed bactericidal effect against *Clostridium perfringens* and did not exhibit marked negative impact on color and sensory properties of sausages compared to sample containing 120 ppm nitrite suggesting that such nanoformulation would be suitable as nitrite substitute for meat products (Vafania et al.

2019). Layer-by-layer edible coating on strawberries (*Fragaria × ananassa* Duch.) using CS and carboxymethyl cellulose (CMC) or coating with 1% CS pronouncedly suppressed the loss of fruit firmness and aromatic volatile compounds of fruits, showing only minor impact on the total soluble solids and total acidity contents, considerably decreased the levels of primary metabolites involved in carbohydrate, fatty acids and amino acids metabolism, and the contents of secondary metabolites involved in terpenoid, carotenoid, phenylpropanoid, and flavonoid metabolism after 8 days of storage (Yan et al. 2019). Polylactic acid (PLA)/CS composite films fabricated by embedding nanosized CS (0.5%, 1%, and 2%) in a PLA matrix using polyethylene glycol (PEG) as cross-linking agent and polyvinyl alcohol (PVA) as plasticizer showing antimicrobial activity against aerobic microorganisms used for packing of prawn meat, were able to suppress the microbial growth and retain the biochemical quality of tested meat samples (Fathima et al. 2018).

A combination of nisin/gallic acid/CS coating with packaging ensuring atmosphere with high O₂ content (80% O₂ + 20% CO₂) was recommended by Cao et al. (2019) to preserve fresh pork, while the shelf life of minced beef meat was found to be prolonged by 2 weeks using CS film incorporating ethanolic extract of propolis (1 and 2%) separately and in combination with cellulose NPs (1 and 2%) (Shahbazi and Shavisi 2018). Genipin cross-linked antimicrobial CS-cellulose NC films with immobilized nisin and ethylenediaminetetraacetic acid (EDTA) on the surface and irradiated by low-dose γ -irradiation of 1.5 kGy inhibited the growth of *Lactobacillus* spp. as well as psychrotrophs and mesophiles in fresh pork loin meats and enhanced the microbiological shelf life of meat sample (>5 weeks) and after 35 days of storage reduced the count of *E. coli* and *Listeria monocytogenes* in tested meat by 4.4 and 5.7 log CFU/g, respectively (Khan et al. 2016). The ϵ -polylysine/CS nanofibers effectively suppressed *Salmonella typhimurium* and *Salmonella enteritidis* on chicken, maintaining the color and flavor of the meat indicating that could be used as powerful antibacterial material in food packaging and preservation systems enabling the shelf life prolongation (Lin et al. 2018).

CS-hydroxypropyl methylcellulose (HPMC) films doped with TiO₂ and neem powder, which were investigated for preservation of grape and plums, maintained unchanged polyphenol oxidase and peroxidase activity for 10 days and 3 weeks, respectively, whereby good sensory and textural qualities and extended shelf life of the grape up to 10 days were observed (Priya et al. 2014). The bio-NC materials fabricated using CS/PVA mixture with loaded TiO₂ NPs (0.5–2%) used in the coating of Ras cheese were able to reduce the weight and moisture losses, while did not exhibit impact on the normal ripening changes in the microbiological, chemical, and textural properties of the cheese and mold growth on the cheese surface was found to be completely eliminated using coating film with 2% TiO₂ NPs (Youssef et al. 2019a).

The wrapping with ZnO NPs-entrapped gelatin bio-NC film with CS nanofiber pronouncedly suppressed the growth of inoculation bacteria in cheese and chicken fillet samples, and after 12 days of storage the organoleptic characteristics of food samples were not significantly impaired and the weight loss achieved 18.91 ± 1.96 and $36.11 \pm 3.74\%$, for chicken fillet and cheese, respectively (Amjadi et al. 2019b).

Films of bio-NCs consisting of CS, guar gum, and ZnO NPs green synthesized using *Roselle calyx* extract showed ameliorated mechanical, permeability, antimicrobial, and antioxidant properties compared to those without ZnO NPs and coating of the bio-NC film containing 3% ZnO NPs on Ras cheese protected its surface for ca. 3 months from microbial growth (El-Sayed et al. 2020). CS-cellulose acetate phthalate films incorporating 5% (w/w) ZnO NPs fabricated by solvent casting method showed the most optimal TS and stiffness, low surface wettability, and high contact angle value up to 90°, better thermal stability and barrier properties and prolonged the shelf life of black grapefruits up to 9 days. The biodegradability of the NC films containing 2–7.5% (w/w) ZnO NPs ranged from 30 to 50% in 4 weeks (Indumathi et al. 2019). ZnO NPs-CS NC coatings on polyethylene (PE) films used for packaging of okra samples stored at 25 °C were found to reduce the total bacterial concentrations by 63% as compared to uncoated control, caused two times reduction in total fungal concentrations compared to this of CS-treated samples, did not affect pH, total soluble solids, moisture content, and weight of the packed okra and thus, maintained its quality (Al-Naamani et al. 2018).

NC film consisting of carboxymethyl chitosan (CMCS) and MgO NPs exhibited better thermal stability, UV shielding performance, and water-insolubility compared to pure CMCS film suggesting its suitability to be used as food packaging, especially in the case of food containing high amounts of water. Moreover, MgO NPs contributed to increased elasticity and ductility of the film already at low (1.0%) filler content and the NC showed great antimicrobial activity against *L. monocytogenes* and *Shewanella baltica* (Wang et al. 2020a).

ZnO-Ag NC green synthesized using *Thymus vulgaris* leaf extract entrapped into poly(3-hydroxybutyrate-co-3-hydroxyvalerate)–CS biopolymer improved the mechanical properties and antimicrobial activity of the biopolymer. The results of sensory evaluation of chicken breast refrigerated over a period of 15 days using the above-mentioned biopolymer with encapsulated ZnO–Ag NC confirmed that it has potential to replace the traditional synthetic polymers used for food packaging of poultry products (Zare et al. 2019). Degradable film consisting of CS–rice starch NC with incorporated AgNPs and ZnO NPs reduced the overall microbial contamination on the surface and prolonged the shelf life of packed peach fruits compared to unpackaged fruits as well as fruits packaged with control film (films with incorporated AgNPs being most effective in the prevention of fruit weight loss), ensured lowest microbial counts on the surface of the fruit and lowest percent (9.6%) increase in total soluble solids (Kaur et al. 2017).

Red claw crayfish-extracted CS and its NCs with AgNPs applied as coatings on fresh-cut melon reduced its respiration rate, retained a steady-state atmosphere (12.6–16.2 kPa of CO₂/2.3–3.7 kPa of O₂) within packages after 9–10 days, prevented the softening under storage, ensured better sensory properties compared to the uncoated samples, showed lower translucency and also microbicidal reduction (Ortiz-Duarte et al. 2019). Hybrid NC film consisting of CS, gelatin, PEG, and AgNPs showed improved mechanical properties and reduced light transmittance in visible light region and when used for the packaging of red grapes, their shelf life was extended for additional 14 days (Kumar et al. 2018).

CS/AuNPs NCs suitable as indicators of thermal history or frozen indicators for perishable and temperature-sensitive food products providing sharp color change by pink to dark gray upon freezing for one day, which could be potentially integrated with existing packaging sensors, for example in packaging of seafood and meats, were developed by Wang et al. (2018b).

Rosemary extract-CS NC coated on the fillets of *Huso huso*, which were inoculated with *L. monocytogenes*, delayed lipid oxidation by reducing peroxide value and thiobarbituric acid production in the samples compared to the control during 16 days of storage at 4 °C (Jafari, et al. 2017). Vanillin NPs deposited on the surface of CS films using a high-intensity ultrasonic method had no influence on the bulk properties of the CS films but increased the antimicrobial activity compared to neat CS film and pronouncedly increased antibiofilm activity, particularly against the biofilm formation of *E. coli* bacteria. Moreover, vanillin NP-coated CS films were able to completely inhibit microbial growth and markedly inhibited mold and yeast growth on fresh-cut watermelon, melon, and strawberry suggesting that such NC could be applied as biodegradable active packaging materials (Buslovich et al. 2018).

CS films incorporated with apricot kernel EO-containing oleic acid as the major fatty acid and high amount of *N*-methyl-2-pyrrolidone showing powerful antioxidant and antimicrobial activity at a ratio 1:1 enhanced water resistance as well as water vapor barrier property by 41% and TS value by 94%, while the elongation percentage value increased pronouncedly only for the film with oil:CS ratio of 0.125 and at higher ratio sharp decrease was observed. Compared to neat CS films this NC film exhibited significant antimicrobial and antioxidant properties and effectively suppressed the fungal growth on packaged bread slices (Priyadarshi et al. 2018).

2.2 Chitosan-Based Nanocomposite for Food Packaging

CS is a biocompatible, nontoxic, and biodegradable polymer. CS-based nanomaterials are characterized with notable physicochemical properties such as high surface area, porosity, TS, conductivity, photoluminescence, and better mechanical properties compared to pure CS (Shukla et al. 2013). It was reported that TS values of CS-based films are similar to those estimated in high-density PE films (Cazon et al. 2017). Flexible biodegradable food packaging as well as edible food coatings fabricated using CS can be used as an alternative for packaging materials prepared from nonbiodegradable plastics (Priyadarshi et al. 2019).

Researchers have also focused on strategies to improve the properties of CS through crosslinking agents, plasticizers, fillers (e.g., NP, whisker, or fibers) incorporated into CS or by adding natural extracts or oils blended with other polymers. Mujtaba et al. (2019) summarized the findings related to progress in the fabrication of CS-based films for food technology. The presence of $-NH_2$ and $-OH$ groups on CS enables facile crosslinking with numerous nanomaterials, which can be utilized in CS NC-based biosensors for bio-detection applications due to their high

sensitivity, selectivity, and stability enabling detection of many targets (Jiang and Wu 2019). Advances and industry challenges related to the application of metal and nonmetal-engineered NPs in food contact materials providing active and intelligent properties when used in packaging were analyzed by Hannon et al. (2015).

Plasticized hemicelluloses/CS-based edible films, which were reinforced by 5% cellulose nanofiber (CNF), showed 2.3-fold higher TS, and the films containing 20–30% glycerol as plasticizer showed TS ranging from 31.02 to 38.56 MPa and tensile strain at break (TB) in the range of 10.07–15.98%, respectively, suggesting that these films can be used for food-packaging applications (Xu et al. 2019a). Addition of CS NPs to HPMC and papaya puree films improved mechanical, thermal, as well as water vapor barrier properties of the films suggesting that their use as packaging material can prolong the shelf life of food (Lorevice et al. 2014). Cellulose/CS films with incorporated pH-responsive indicator carrot anthocyanins were designed by Tirtashi (2019) for intelligent food packaging. For the monitoring of spoilage in pasteurized milk, the color change at varying pH values (pH 2–11) was used, whereby the color of the indicator was stable during one-month storage at 20 °C. By deposition of CS/tannic acid bilayers on CNF mats with fiber diameters of 300–400 nm using layer-by-layer technology, antibacterial mats with improved hydrophilicity and mechanical properties showing >86% antibacterial activity against *E. coli* (>86%) and *S. aureus* (99%) were fabricated, which could be used for food packaging or wound dressing (Huang et al. 2019).

A ternary blend edible film fabricated using CS, gelatin, and cinnamon EO characterized with low transparency at 600 nm exhibited fine UV protection and showed higher thermal stability, contact angle, and elongation at break, but lower TS, crystallinity, and wettability than neat CS films. The powerful antimicrobial activity of this ternary blend edible film against *S. aureus* and *E. coli* was reflected in minimum inhibitory concentration (MIC) values of 52.06 µg/mL for both bacteria and minimum bactericidal concentration (MBC) values of 52.06 and 104.12 µg/mL for *S. aureus* and *E. coli*, respectively (Guo et al. 2019). Bio-NC films fabricated by incorporating CS/gallic acid (GA) NPs into a konjac glucomannan (KGM) film, in which CSGA NPs interacted with KGM through H bonds in a bio-NC matrix, showed pronouncedly ameliorated not only mechanical and barrier properties but also antimicrobial activity against *E. coli* and *S. aureus* at applied concentrations 5–10% CSGA NPs (Wu et al. 2019b). CS-KGM-cassava starch–AgNPs composite films showing resistance against moisture and antimicrobial effectiveness for food-packaging applications were designed by Nair et al. (2017).

CS nanofillers incorporated into sago starch (SS) formulations showed beneficial impact of the TS (88 MPa vs. 46 MPa for the SS film) and lower weight loss at heating than SS film (60% loss up to 390 °C vs. 67% at 375 °C) (Fauzi et al. 2019). Nanocrystalline cellulose (NCC) in transparent and biodegradable CS/guar gum (GG)/NCC film contributed to improved TS and reduced air permeability in comparison to pure CS/GG films and CS/GG composite films reinforced with NCC could be successfully used for food packaging applications (Tang et al. 2018).

CS/ε-polylysine bio-NC films fabricated using sodium tripolyphosphate (TPP) as cross-linking agent showed significant antimicrobial effectiveness against

S. aureus and *E. coli* with the increasing ratio of ϵ -polylysine, whereby sustained release of ϵ -polylysine closely related to TPP concentration was observed (Wu et al. 2019a).

Lignin NPs (1 or 3 wt%) as constituents of binary and ternary polymeric films based on PVA and CS contributed to improved TS and Young's modulus of PVA and produced a toughness effect in CS matrix as well; also pronouncedly ameliorated the thermal stability of such NCs was observed and the films were able to inhibit the bacterial growth of *Erwinia carotovora* subsp. *carotovora* and *Xanthomonas arboricola* pv. *pruni* indicating that prepared films could be applied in food packaging applications (Yang et al. 2016). New biocomposite films based on CS/PVA and MMT functionalized with thiabendazolium exhibited improved mechanical properties with Young's modulus ranging from 66.98 to 143.43 MPa and TS of 24.95–34.65 MPa, respectively and showed higher antimicrobial activities against *E. coli*, *S. aureus*, and *Pseudomonas aeruginosa* compared to the pure film (El Bourakadi et al. 2019). Koosha and Hamedi (2019) designed intelligent CS/PVA NC films containing anthocyanins from black carrot and bentonite nanoclay showing good thermal stability. While addition of bentonite reduced the TS and WVP of CS/PVA films, addition of anthocyanins had positive effect on both characteristics of CS/PVA films. Addition of anthocyanins and 5% bentonite caused 44.38%, 69.95%, and 75.20% growth inhibition of *S. aureus*, *P. aeruginosa*, and *E. coli*. Moreover, the added amounts of bentonite and anthocyanins affected the color of the NC films. Addition of sonochemically synthesized lignin NPs of 10–50 nm to PVA/CS NC hydrogel caused considerable increase in its thermal stability (Ingtipi and Moholkar 2019). Composite films from cellulose (3–5%), CS (0–1%), and PVA (0–4%) showed ameliorated mechanical properties, very good properties against UV radiations, and appropriate transparency value and were characterized with water-absorbing capacity. It was found that CS and PVA improved the mechanical properties of cellulose-based films, cellulose, and CS ameliorated the UV light-protecting effect of the films and PVA ameliorated the film transparency (Cazon et al. 2018). Eco-friendly bio-NC films based on PVA/CS reinforced with cellulose nanocrystals (CNCs; ca. 15 nm in diameter) isolated from rice straw showed comparable transparency level to that of neat PVA/CS film, but increased the TS and thermal stability compared to the PVA/CS films and exhibited good antifungal and antibacterial activity (Perumal et al. 2018).

A flexible, transparent, thermally stable bio-NC of CS/polyvinyl pyrrolidone (PVP) film reinforced with CNFs (9–11 nm and 100–200 nm) showing improved thermal stability and mechanical and barrier properties suitable for food packaging applications was designed by Kumar et al. (2019). Biopolymer NC films fabricated by casting film-forming emulsions containing CS/Tween 80/rosehip seed oil with dispersed MMT nanoclay were found to be flexible, with better mechanical, gas, and water vapor barrier properties. Moreover, they exhibited antioxidant activity and effective antibacterial activity against *Bacillus cereus*, *E. coli*, and *S. typhimurium* (Butnaru et al. 2019).

By addition of *N*-(2-hydroxyl)-propyl-3-trimethylammonium chitosan chloride to sodium CMC-based films, new composite materials with enhanced

physicochemical properties suitable for food and drug packaging were fabricated by Wang et al. (2018c). Chicken breast fillets packaged with CS- and polycaprolactone-based bilayer films incorporated with grape seed extract (15% w/w) and nanocellulose (2% w/w), which were maintained 15 days under refrigerated conditions, showed lower levels of thiobarbituric acid reactive substances and considerable reduction in the counts of mesophilic aerobic bacteria and coliform bacteria compared with controls suggesting that such bilayer materials represent an active packaging material for food products delaying the microbial growth and oxidation and extending the shelf life during refrigerated storage (Sogut and Seydim 2019). Bao et al. (2018) designed CS-xylan/cellulose nanowhiskers (CNW) NC films, in which reinforcement effects of CNW nanofillers were reflected in improved TS and elongation at break of the NC films; good antibacterial activity was mediated by CS, and antioxidant activity was connected with the presence of xylan.

NC film consisting of CS and spherical polyaniline NPs (45–100 nm) showing mechanical, electrical, and antimicrobial properties suitable to be used in intelligent food packaging was fabricated by Mohammadi et al. (2019a). With increasing polyaniline concentration in the film increased antifungal activity against *Aspergillus niger* and antibacterial activity against *E. coli*, enhanced TS and elastic modulus, while decreased electrical resistance of the film was observed. Oliveira et al. (2019) designed electrical conducting blends prepared from CS and a conductive polymer polyaniline (PANI) utilizing benefits of a rapid electrical response of PANI and the high magnitude of the electrical response of CS, which are suitable as smart packaging for food. In contact with food, these blends exhibiting electrical conductivity can monitor changes in its physicochemical properties.

CS-based composite consisting of CS, bentonite (BT), and poplar hot water extract (10 wt%) showed UV light transmittance at 280 nm by 99.36%, i.e. higher than the film containing only CS and BT, and also its oxidation resistance was 9.65-fold of this estimated with CS-BT film. Moreover, the composite film had a denser structure and improved mechanical and water vapor barriers properties (Sun et al. 2019a). CS/clay/glycerol NC films showed improved thermal stability due to the presence of clay and glycerol and clay component pronouncedly ameliorated also the TS and tensile modulus of CS films, whereby the best strength and stiffness as well as water resistance showed CS films containing 5 wt% of clay and 20 wt% of glycerol. The presence of both clay and glycerol strongly reduced the ductility of CS (Kusmono and Abdurrahim 2019). Habel et al. (2018) designed a high-barrier, biodegradable food packaging consisting of a PLA foil (25 μm) furnished with a glycol CS–clay NC coating (1.4 μm), in which the barrier side of the foil inhibited bacterial colonization, while the uncoated PLA side ensured biodegradability. Unmodified MMT or activated food-grade MMT/CS biocomposites containing high amounts of MMT and low amounts of CS (70:30, 75:25, 80:20, % w/w) were reported to be appropriate carriers, which can covalently immobilize the protease reaching immobilization yield of 18% and 14–17%, respectively (Cacciotti et al. 2019). NCs of CS with octadecylamine MMT supplemented by *Nigella arvensis* seed (black cumin) extract at doses 1–5% exhibited stronger antibacterial activity against *E. coli* and *Salmonella enterica* serotype *Typhimurium* SL 1344 compared

to *S. aureus* and *Streptococcus mutans* ATCC 25175 showing potential to prevent the antimicrobial formation when used as packing materials (Ilk et al. 2018).

Composite films consisting of CS and graphene oxide (GO) characterized with denser structure showed improved mechanical strength and lower UV light transmission and considerably higher WVP, compared to pure CS film and their complete degradation in soil compost was observed within 20 days. Increasing GO oxidation degree ameliorated mechanical strength and light barrier properties of composite films suitable for food packaging (Lyn et al. 2019). The antibacterial activity of CS–Ag NC against *S. aureus* and *E. coli* was reported to increase with increasing concentration of AgNPs in the NC up to 0.03 M and then remained constant (Fahimi and Ghorbani 2019). Khawaja et al. (2018) tested antibacterial activity of GO and NCs consisting of GO and AgNPs, GO, and CS as well as GO–CS–AgNPs against *S. aureus*, *S. mutans*, *E. coli*, *Klebsiella pneumoniae*, *P. aeruginosa*, and *S. typhi* and found that the activity decreased as follows: GO–CS–AgNPs > GO–Ag > GO–CS > GO, GO–CS–AgNPs NC being particularly effective showing potential to be applied as biomaterial or in food industry. Soft and tough CS-iron oxide-coated GO NC hydrogel films showing suitable thermal and mechanical properties and considerable antimicrobial activities against methicillin-resistant *S. aureus* and *E. coli*, as well as against the opportunistic dermatophyte *Candida albicans* were reported as suitable materials to be used not only in biomedical applications but also in the food industry (Konwar et al. 2016). Advantages and health risks of the use of carbon nanotubes (CNTs) composite materials in food packaging were discussed by Xu (2018).

CS-based NC films, in which AgNPs green synthesized using *Nigella sativa* extract with average particle size of 8 nm were unevenly distributed over the CS matrix, showed improved TS and elongation and lower WVP compared to CS films. Moreover, these NC films showed a sustained pH-dependent release of AgNPs and Ag⁺ ions and notable antibacterial and antioxidant activity showing promising potential to be used for food packaging (Kadam et al. 2019). The incorporation of anthocyanin-rich purple corn extract (PCE) and AgNPs into CS film pronouncedly improved the mechanical strength, water vapor and light barrier ability, antioxidant effectiveness, and antimicrobial properties of CS film, which was connected with the synergistic effect between PCE and AgNPs. Moreover, due to abundant anthocyanins in PCE, the films responded to different pH buffers with color change suggesting that they were recommended as an excellent active and intelligent food packaging material (Qin et al. 2019). CS-based active NC films with bacterial CNCs and AgNPs showing mean particle sizes of 20–30 and 35–50 nm, respectively, which were homogeneously dispersed in CS matrix showed pronouncedly improved sensibility to water, WVP, and mechanical properties compared to neat CS film as well as powerful antibacterial activity against foodborne pathogens indicating their suitability as packaging material for increasing shelf life of packaged foods (Salari et al. 2018).

By incorporation of MMT–CuO-90 NC (fabricated by heating at 550 °C for 90 min) into the CS films at concentration 3% w/w an increase of TS, and elongation at break by more than 50% and reduction of the WVP by ca. 55% and 32%,

respectively, was observed, whereby these films were found to be more effective against *B. cereus* and *S. aureus* than against *P. aeruginosa* and *E. coli*. Moreover, the presence of MMT–CuO-90 NC in the CS film could improve its mechanical properties and reduce both UV transition and water solubility showing minor impact on the transparency of the film (Nouri et al. 2018).

The gelatin-based NC containing CS nanofibers and ZnO NPs showed fine mechanical and water barrier properties due to its high dense and less permeable structure and due to the synergistic effect between CS nanofibers and ZnO NPs increased antibacterial activity of NC was observed (Amjadi et al. 2019a). Novel biodegradable composite films for food packaging based on CS biopolymer with incorporated ZnO NPs (40–80 nm; 1 and 3% (w/v)) and *Melissa officinalis* EO (0.25 and 0.5% (w/v)) showing antimicrobial activity were prepared. With the increasing concentrations of ZnO NPs and EO water solubility and WVP of films showed a decrease, while their TS and antibacterial properties increased. However, while with increasing ZnO NPs the film opacity increased, increasing amounts of EO exhibited opposite effect reflected in increased transparency (Sani et al. 2019). Biodegradable films prepared from Mahua oil-based polyurethane and CS with entrapped ZnO NPs containing 5% ZnO NPs in the composite showed improved TS and stiffness, enhanced hydrophobicity, barrier properties, and antibacterial properties of the biodegradable film, which lost 86% of weight after one month in the soil. On the other hand, the shelf life of carrot pieces wrapped with the NC film was prolonged up to 9 days and the film with ZnO NPs was able to reduce the bacterial contamination much more than the PE film (Sarojini et al. 2019). CS-ZnO NC coatings on PE films were able to completely inactivate and prevent the growth of food pathogens, while films coated with CS reduced the viable cell counts of *S. aureus*, *E. coli*, and *S. enterica* after 24-h incubation only tenfold compared to the control suggesting that PE films coated with CS-ZnO NC represent an active packaging material which can prolong the shelf life of food (Al-Naamani et al. 2016).

CS antimicrobial film containing 1% TiO₂ NPs evenly distributed in CS film matrix was characterized with improved TS, water barrier, and ethylene photocatalytic degradation properties, and showed antimicrobial activity against bacteria (*E. coli*, *S. aureus*, *P. aeruginosa*, and *S. typhimurium*) and fungi (*Penicillium* and *Aspergillus*) (Siripatrawan and Kaewklin 2018). Effective inactivation of *S. aureus* by CS-TiO₂ NC film designed for packaging, in which TiO₂ NPs (Degusa 25) were dispersed in CS matrix, was reported by Kustiningsih et al. (2019). Potential of antimicrobial CS-TiO₂ composite film damaging membranes with subsequent leakage of cellular substances for food packaging applications was highlighted also by Zhang et al. (2017). Food packaging films based on CS, TiO₂ NPs, and black plum peel extract was characterized with better mechanical strength and barrier properties against UV–vis light and water vapor, stronger free radical as well as ethylene scavenging activities and antimicrobial activity than CS film. The abundant anthocyanins were responsible for the sensitivity of the film color to pH (Zhang et al. 2019). Ethylene photodegradation activity at exposure to UV light showed also active packaging made from CS containing TiO₂ NPs, which also retarded the

ripening process and quality changes of the packaged tomatoes (Kaewklin et al. 2018).

Starch–CS films with incorporated TiO₂ NPs functionalized with the alkoxy silane exhibited improved TS by 33% and the insertion of Ag nanodots also ameliorated antimicrobial effectiveness of the starch–CS films (Vallejo-Montesinos et al. 2020). TiO₂ NPs added to zein/CS composite films pronouncedly ameliorated the thermal stability and mechanical properties of films and using 0.15 wt% TiO₂ NPs resulted in the TS of the zein/CS/TiO₂ films of 28.33 ± 1.53 MPa. The composite films showed higher antibacterial effect *S. aureus* compared to *E. coli* and *S. enteritidis*, whereby the antibacterial effect at exposure to UV light was better than that observed at dark conditions (Qu et al. 2019).

As biodegradable active packaging materials with good antibacterial and antioxidant activities, water vapor and oxygen barrier properties and thickness the NC based on CS and CMC and containing *Ziziphora clinopodioides* EO (1 and 2%) and methanolic *Ficus carica* extract (1%) were recommended by Shahbazi (2018).

By incorporation of 0.5% *Codium tomentosum* seaweed extract into CS-based edible films, the film solubility and elasticity increased by 50 and 18%, respectively, while the puncture strength and energy at break decreased by 27% and 39%. On the other hand, addition of this extract in the ALG films pronouncedly reduced film solubility (6%), WVP (46%), and elasticity (24%) without an impact on thermal properties (Augusto et al. 2018).

Synthetic melanin-like NPs (MNPs) reinforced CS NC films showed enhanced ultraviolet blocking and mechanical properties and high antioxidant activity. In addition, the presence of MNPs also increased the hydrophobicity and swelling ratio of the NC films, whereby the water vapor barrier property and thermal stability of the films were not impaired (Roy et al. 2020). Similarly, functional CS-based films with incorporated curcumin (1 wt%) showed improved swelling ratio, UV-blocking, water vapor barrier property, and surface hydrophobicity compared to the neat film and exhibited high antioxidant activity, whereby slower release of encapsulated curcumin was observed compared to films prepared using carrageenan or agar (Roy and Rhim 2020). Biodegradable food packaging hybrid film consisting of PVA, CS, xylan, and hydroxyapatite with incorporated curcumin was evaluated as suitable intelligent packaging enabling real-time fish freshness assessment based on the visible and sharp color changes (Vadivel et al. 2019).

Functional composite films of CS with sulfur nanoparticles (SNP), in which SNPs were dispersed evenly in the CS matrix, showed enhanced mechanical strength, water vapor barrier property, hydrophobicity, and antimicrobial activity compared to neat CS film, whereby the highest antimicrobial activity against food-borne pathogenic bacteria *L. monocytogenes* and *E. coli* resulting in complete sterilization within 12 and 6 h, respectively, was observed with SNPs capped with CS composite film (Shankar and Rhim 2018).

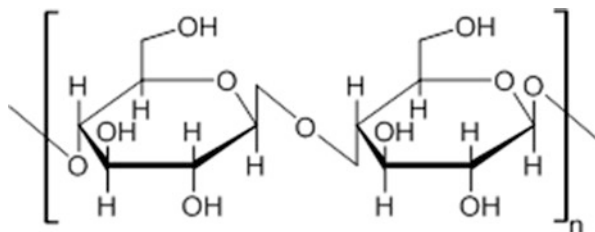
3 Nanocellulose and Bacterial Cellulose

In vascular plants, cellulose consists of β -(1 \rightarrow 4)-linked D-glucopyranose chains laterally bound by H-bonds forming microfibrils with a nanosized diameter, which are further organized in microfibril bundles (Fig. 5.3) (Klemm et al. 1998).

Using mechanical disintegration of cellulose fibers showing a width ca. 20–50 nm and length of several micrometers, which contain both crystalline and amorphous regions, cellulose nanofibers (CNFs; known also as nanofibrillated cellulose) with high length-to-diameter aspect ratios can be fabricated. On the other hand, nanocrystalline cellulose (NCC), cellulose nanocrystals (CNCs), and cellulose nanowhiskers (CNWs) are cylindrical rods of crystalline cellulose showing 5–10 nm and length: 20–1000 nm (Hutten 2016). The use CNFs, CNCs, and CNWs as reinforcement fillers in composites can considerably ameliorate their mechanical properties (Perumal et al. 2018; Ukkund et al. 2019; Jampflek and Kráľová 2020). Cellulose nanomaterials as nanoreinforcements for polymer NCs were discussed by Dufresne (2018). Electron microscope images of CNFs extracted from trees are shown in Fig. 5.4. For fabrication of CNFs, CNCs, and CNWs, various cellulose source materials, including agricultural waste can be used (Sangeetha et al. 2017; Jampflek and Kráľová 2020). CNFs were isolated for example from banana peels (Tibolla et al. 2019), wheat straw fibers (Fan et al., 2018), rice straw pulp (Hassan et al. 2018), pineapple leaf (Balakrishnan et al. 2017), sugarcane bagasse (Patil et al. 2018), or coconut palm petiole residues (Xu et al. 2015), while CNCs were fabricated from rice straw (Perumal et al., 2018), rice husks (Kargarzadeh et al. 2017), mango seed shells (Silva et al. 2019), or sugarcane bagasse (Kassab et al. 2019). It could be mentioned that the use of agricultural waste for preparation of CNFs and CNCs is favorable not only due to low costs, but using this approach woody species playing a substantial role in moderating the climate could be preserved that is particularly desirable under changing climatic conditions (Jampflek and Kráľová 2020).

Bacterial cellulose (BC) is an extracellular polymer produced by many microorganisms (the best producer, the *Komagataeibacter* genus, uses semisynthetic media and agricultural wastes). BC is characterized by the nanoporous structure and it contains high water content and numerous free –OH groups (Cacicedo et al. 2016). It could be underlined that BC is produced as naturally nanostructured membranes, capable to grow in a medium containing

Fig. 5.3 Structure of cellulose



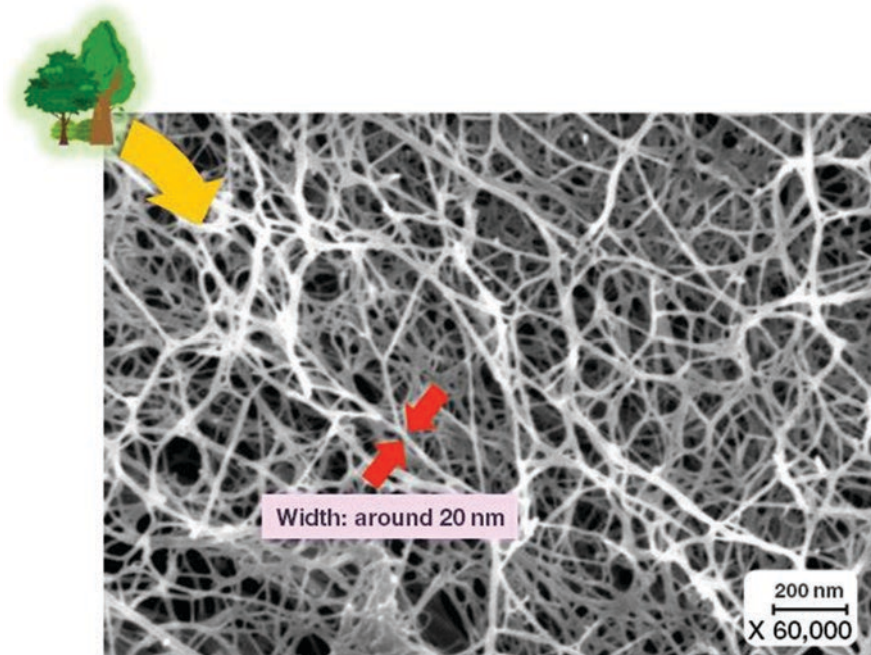


Fig. 5.4 Electron microscope image of nanofiber cellulose extracted from trees (Nippon, 2019)

other biopolymers, resulting in formation of bio-NCs. Moreover, it can be impregnated with other components or disintegrated into nanofibrils or nanocrystals and therefore the use of BC nanostructures for food packaging application is favorable (Azeredo et al. 2017).

3.1 Nanocellulose-Based Nanocomposites Used for Food Protection

New sustainable eco-friendly antimicrobial and antioxidant active ingredients could be introduced in food packaging to achieve improved safety and quality of food products and reduce adverse impact on the environment. Introduction of such active compounds in edible packaging could lower and/or limit the environmental impact, whereby edible and eco-friendly food packaging can also reduce the waste (Fortunati, et al. 2019). Dhar et al. (2019) in a review paper evaluated the properties of nanocellulose and its strategic modifications to design active food packages enabling real-time monitoring of food quality, while Hemavathi and Siddaramaiah (2019) highlighted the crucial role of polymers in food supply chain as a packaging material able to store, protect, and preserve food from spoiling and damage.

Nsor-Atindana et al. (2017) analyzed the functionality and nutritional aspects of microcrystalline cellulose (MCC) providing beneficial impact on gastrointestinal physiology, and hypolipidemic effects and affecting the expression of several enzymes involved in lipid metabolism; MCC was investigated as a functional ingredient in various food products as discussed as well.

Incorporation of MMT and ϵ -poly-(L-lysine) (PL) into CMC matrix resulted in improved UV and water vapor barrier properties, TS and hydrophobicity of CMC film, and effective antimicrobial activities against bacteria (*S. aureus* and *E. coli*) as well as phytopathogenic fungi (*Botrytis cinerea* and *Rhizopus oligosporus*) reflected in >90% inhibition rate of microbial growth at 7.5 wt% of PL. When strawberries were coated with a film-forming solution of the composite CMC/MMT/PL with 7.5 wt% PL, their shelf life was prolonged for 2 days (He et al. 2019).

CMC film coated with ZnO NPs, which was used as the packaging material for pork meat for 2 weeks of cold storage at 4 °C, was able pronouncedly suppress the rising in total volatile basic nitrogen and pH levels, limited decline of lightness and redness, and maintained the water-holding capacity compared to the control meat samples and under cold conditions caused increased occurrence of cell membrane rupture in *S. aureus* (Suo et al. 2017). Chicken breast meat packed with NC films based on CMC, okra mucilage (OM), and ZnO NPs and stored at 4 °C pronouncedly suppressed the growth of *S. aureus* counts and lactic acid bacteria and considerably suppressed lipid oxidation and total volatile nitrogen due to the presence of okra mucilage and ZnO NPs resulting in extended shelf life of meat products. The best antimicrobial effect and suppression of undesirable changes in the meat samples and higher sensorial score under chilling storage was observed with the film consisting of CMC/OM50%/ZnO NPs (Mohammadi et al. 2019b).

NC prepared via Schiff base reaction by anchoring nisin to the cellulose beads mixed with γ -Fe₂O₃ NPs exhibited susceptible magnetic response and good thermal stability and showed long-term antimicrobial effectiveness against *Alicyclobacillus acidoterrestris* DSM 3922 suggesting its suitability to be used in food industry (Wu et al. 2019c).

Active packaging based on cellulose–CS–Ag/TiO₂ NC tested for storage of clarified butter showed peroxide value of 2.72 mEq O₂/kg and lower counts of molds and yeasts (5.8 CFU/g) and *E. coli* (6.12 CFU/g) after 6 months of storage (Apjok et al. 2019).

3.2 Nanocellulose-Based Nanocomposites Used for Food Packaging

Nanocellulose (NCell) materials are derived from cellulose, the most abundant biopolymer on our planet. NC hydrogels based on NCell or reinforced with NCell are characterized with superior mechanical properties, biocompatibility, and biodegradability and can provide a slow and controlled release profile of incorporated active

ingredients (Nascimento et al. 2018). Polymers obtained from celluloses, CS, and other native or modified carbohydrate polymers (e.g., starches, glucans, pectins, and gums), could be used for fabrication of self-assembled polymer composite materials, including films, hydrogels, micelles, and particles and can be used as encapsulating agents of food additives, food coatings, or as edible films. Considering their mechanical properties and sensitivity to temperature, pH, individual ions, ionic strength, and enzymes, they can provide a base for the design and fabrication of advanced materials for food industry (Valencia et al. 2019). Recent finding related to hydrogels and aerogels incorporating N Cel, including physical and chemical cross-linking strategies, postmodifications, or gel structure control were discussed by De France et al. (2017).

A review paper of Khalil et al. (2019) discussed matrix filler combinations suitable for fabrication of neutral or negatively charged polysaccharides-based composite films with improved mechanical and barrier properties suitable not only for food packaging but applicable also in agriculture, biomedicine as well as constructions sector. Fabrication of eco-friendly biopolymer composites used for green packaging, novel processing techniques used to produce high-performance lignocellulosic reinforced materials with better properties, green modification of organoclay by antibacterial natural compounds, and the use of green-modified organoclays as compatibilizing and reinforcing material for different incompatible biopolymers (e.g., CS, CMC, and PLA) was discussed by Moustafa et al. (2019). Idumah et al. (2019) summarized recent findings related to actual trends in the fabrication and characterization of polymer NCs as biodegradable and environment-friendly packaging materials. A review paper summarizing recent findings related to the progress in the design of new nanomaterials for food packaging and discussing the possible mechanisms of antibacterial activity of certain biologically active nanomaterials and their impact on health was presented by Huang et al. (2018). Polymeric NCs and nanoscale coatings suitable for food packaging were overviewed also by Vasile (2018). Youssef and El-Sayed (2018) in a review paper focused on the degradable packaging materials showing notable performance, excellent mechanical properties, thermal stability, and antibacterial activity as well as on the use of bio-NCs in various packaging applications. Golmohammadi et al. (2017) discussed the feasibility of NC applications in (bio)sensing technology in order to obtain analytical information in various fields (e.g., in medical diagnostics, monitoring of the environment, for ensuring high level of food safety, physical/mechanical sensing and labeling, or in bioimaging applications). The researchers emphasized that in the near future the NC (bio)sensing platforms could replace currently used (bio)sensors based on plastic, glass, or conventional paper platforms. A critical review focused on the biological impact of N Cel was presented by Endes et al. (2016), drawing the attention to certain forms of N Cel, which due to their specific physical characteristics could represent some hazard to living organisms. Gomez et al. (2016) in their review paper analyzed applications of NC as a stabilizing agent, functional food ingredient, and in food packaging suggesting potential use of N Cel as a stabilizing agent in food emulsions, as dietary fibers, and as materials able to reduce the caloric value of food.

Biomedical, food, and nutraceutical applications of NCs containing green cellulose nanomaterials fabricated from renewable sources and characterized with excellent mechanical strength and biocompatibility were overviewed by Amalraj et al. (2018). The recent progress in the utilization of cellulose-based nanomaterials in food and nutraceutical applications was overviewed by Khan et al. (2018). The use of NCell in green food packaging with emphasis on the several types of biopolymers with NCell fillers forming bio-NCs was analyzed by Vilarinho et al. (2018).

A composite cassava starch films fabricated with stearic acid-modified MCC or NCC as strength agent using the casting method showed reduced thermal stability but enhanced mechanical and hydrophobic properties compared to neat starch films. Use of 0.5% MCC and 1.5% NCC resulted in the TS increase of cassava starch film by 484.5 and 327.7%, respectively, while addition of 2% MCC and 0.5% NCC increased film hydrophobicity by 65.0 and 30.3%, respectively, suggesting more favorable enhancement effect of MCC (Chen et al. 2020). CMC nanocrystals showing diameters of 30–50 nm incorporated in cassava starch film resulted in improved mechanical and water barrier properties and better water solubility compared to incorporation of CNCs suggesting that hydrophilic modification of CNCs used for reinforcement of films resulted in improved physicochemical properties of these composite films (Ma et al. 2017). CNF–bengkoang starch bio-NC film prepared with ultrasonication containing 2 wt% of CNF showed TS of 9.8 ± 0.8 MPa, which was by 160% higher than that of neat starch film and exhibited also lowest moisture absorption and WVP, highest thermal resistance, and high transparency compared to the bio-NC films containing lower CNF amounts (0.1–1.5%) (Mahardika et al. 2019). Corn nanostarch-based NC film loaded with 8.0 wt% CNCs modified by crosslinking with citric acid, and subsequently amidated with CS exhibited a 230.0% increase in TS compared to pure corn nanostarch film and showed a decrease of the moisture absorption ability and WVP by 25.6 and 87.4%, respectively, with simultaneous increase of the water contact angle value by 18.1% as well as higher antimicrobial activities against *S. aureus* and *E. coli* (Chen et al. 2019). Corn distarch phosphate/NCC films with incorporated nisin and ϵ -polylysine showed synergistic antimicrobial activity against *E. coli* and *S. aureus*, ameliorated mechanical and barrier properties, higher thermal stability and thus, they could be recommended as active food packaging material (Sun et al. 2019b). Rice starch-based edible films, which were fabricated via solution casting with addition of NCell extracted from banana pseudostems (2–10%), were characterized with improved mechanical and barrier properties suggesting their suitability to be applied for food packaging (Jeevahan and Chandrasekaran 2019b).

PLA/starch NC films reinforced with CNFs isolated from MCC exhibited improved TS, Young's modulus, and reduced air permeability compared to neat PLA/starch films (Mao et al. 2019).

In starch/CNWs composite films fabricated by casting of the mixtures of starch/CNWs homogenous aqueous suspensions and subsequent drying, the layers of CNWs were located within starch matrices in a parallel direction to NC film surfaces, whereby increasing amount of CNWs resulted in increased layer thickness. In such self-assembled multilayer structures, both the interaction and evaporation rate

of the solvent affect the NPs dispersion and they can serve as an appropriate material for food packaging ensuring favorable mechanical and gas barrier properties (Liu et al. 2017).

A multilayer structure, in which layers of cellulosic nanomaterials providing O₂ resistance are coated with other layers of polymers that provide moisture resistance and tightness, may be an alternative to packaging materials dominated by synthetic plastics (Wang et al. 2018d). By incorporation of cellulose microfibrils into cellulose agar bio-NC films the TS increased from 38.8 ± 3.2 to 49.4 ± 4.3 , the elongation at break showed ca. 6% increase and films showed reduced swelling; increased degradation rate in soil suggested their suitability for food packaging applications (Raj et al. 2019). Biodegradable biocomposites consisting of high-pressure microfluidized cellulose fibers and carrot minimal processing waste (33 wt%) showed TS of ca. 30 MPa, ca. 3% elongation at break, and Young's modulus of ca. 2 GPa indicating its suitability to be applied for food packaging (Otoni et al. 2018).

CNFs composite films containing lignin particles of colloidal dimensions (CLPs) designed by Farooq et al. (2019) were found to be strong, ductile, and waterproof and provided antioxidant activity and UV-shielding with improved visible light transmittance; approx. double toughness of films was achieved at application of an optimal CLPs content (10 wt%) when compared to a neat CNF film without added lignin particles.

Soy protein-based films incorporating CNCs and pine needle extract exhibited improved mechanical property, antioxidant ability, and water vapor barrier capacity suggesting their suitability to be used as an active food packaging material (Yu et al. 2018). Investigation of the impact of CNFs on morphological, mechanical, optical, and barrier properties of biopolymers isolated from whey protein showed that in the films containing up to 4% CNFs these were well dispersed in the whey protein matrix and acted as reinforcing agent making the bio-NC films more resistant and less flexible (Carvalho et al. 2018).

Biodegradable bio-NCs based on plasticized sugar palm starch and 1.0 wt% nanofibrillated celluloses obtained from sugar palm fibers exhibited improved mechanical and thermal stability and water barrier of the starch polymer and this bio-NC material prepared from natural sources could be applied for plastic packaging and food containers (Ilyas et al. 2020). Study of the impact of polyol-based plasticizers on physicochemical properties of CNCs-filled ALG bio-NC films showed that although the presence of plasticizers markedly impaired the TS and the elastic modulus of the neat ALG films and pronouncedly increased the elongation at break, these characteristics were improved by incorporation of CNCs into the films. Moreover, the presence of both CNC and plasticizers in ALG bio-NC films decreased WVP and moisture uptake of these films and the films exhibited good optical transparency suggesting their potential to be used for packaging applications (El Miri et al. 2018). Bio-NC film consisting of cellulose NPs prepared from the nonedible part of jackfruit (*Artocarpus heterophyllus*), PVA, and fennel seed EO exhibited sevenfold increase in TS and sixfold increase in elongation at break compared to neat PVA film due to incorporation of cellulose NPs and can represent a convenient alternative for traditional food packaging materials (Jancy et al. 2020).

Addition of 10% nanoscale BC to the agar-based edible films considerably reduced moisture content, water solubility, and WVP and increased the TS of films to 44.51 MPa, while the elongation at break increased with increasing BC concentrations only in the range 0–5%, and then decreased in the presence of 8–10% BC (Wang et al. 2018e).

Bio-NCs consisting of ionically cross-linked CS and CMC serving as matrix for CNC nanoreinforcement agent achieved TS of 60.6 MPa, Young's modulus of 4.7 GPa, and water vapor transportation rate of 7982 g μm^2 per day when the applied CNC concentration was 10 wt%. On the other hand, composites with <5 wt% CNC were found to be a significant barrier coating against penetration of liquids (water, oil) on the paperboard substrate (Chi and Catchmark 2018).

Due to blending of ALG with needle-shaped CNWs (length: 200–400 nm, diameter: 15–30 nm), which were isolated from the mulberry pulp, the elastic modulus and TS of the composite ALG-based film increased by 35 and 25%, respectively, compared to pure ALG film, while no change in WVP was observed when for the blending 4 wt% CNW was applied (Wang et al. 2017).

Antimicrobial composite films fabricated from xylan and hydroxyethylcellulose using citric acid and PEG-400 as crosslinker and plasticizer with incorporated β -cyclodextrin/sodium benzoate and cured for 40 min achieved TS up to 62.3 MPa and very low oxygen permeability calculated through the oxygen gas transmission rate multiplied by the average film thickness ($1.0 \text{ cm}^3 \mu\text{m m}^{-2} \text{ d}^{-1} \text{ kPa}^{-1}$) and showed high antimicrobial activity against *S. aureus* indicating its great potential in the field of sustainable food packing materials (Yang et al. 2019).

A mixture of the κ -carrageenan, CNC, and glycerol was used to prepare bio-NC films by solution casting, which showed improved mechanical and barrier (water and UV) properties and thermal stability compared to pure κ -carrageenan films, particularly when 7–9% CNC was used (Yadav and Chiu 2019). Ameliorated mechanical properties of κ -carrageenan-based NC films reinforced with CNCs were also reported by Kassab et al. (2019).

Strong antibacterial properties against both *E. coli* and *S. aureus* exceeding 99% showed also PVA films containing BC and ϵ -polylysine exhibiting strong antibacterial activity also after reusing twice, whereby the biocomposite films had improved thermal stability and mechanical properties compared to pure PVA films (Wahid et al. 2019). By oxidation of CNFs and CNCs using periodate oxidation the introduced dialdehyde groups can act as a crosslinking agent. Introduction of these fillers containing dialdehyde groups (DCNF and DCNC, respectively) in PVA films enhanced their water resistance. However, while DCNC exhibited only chemical network reinforcement effect, DCNF induced physicochemical networking effect to PVA (Lee et al. 2020). Biodegradable conductive NC fiber membrane fabricated by in situ polymerization of indole in the presence of electrospun BC nanofibers activated in acidic medium showed good antibacterial activity against *S. aureus* and *E. coli* and could be used in food packaging as well as in biomedical preparations, electronic devices, and biosensors (Cai et al. 2018). Addition of nanofillers, NCC, and chitin whiskers (CHW) to biodegradable poly(butylene succinate) (PBS) restricted the mobility of polymer chains, while nucleation and

recrystallization of polymer were stimulated resulting in the increased crystallinity degree from 65.9 to 75.6%. An increase of TS of the neat PBS-based films (23.2 MPa) was observed following CHW and NCC addition to 43.6 and 32.9 MPa, respectively. Using a compatibilizer, methylene diphenyl diisocyanate, at a dose of 4% in PBS films containing 3% NCC a decrease in O₂ transmission rate from 737.7 to 23.8 cc/m²/day was observed, while water vapor transmission rate declined from 83.8 to 30.8 g/m²/day (Xu et al. 2019b). Poly(propylene carbonate)/poly(3-hydroxybutyrate)-based bio-NCs reinforced with CNCs showed improved oxygen barrier properties, which at application of 1 wt% CNCs increased ca. 18-fold, suggesting the promising potential of such bio-NCs to be used as degradable material for food packaging (Jiang et al. 2020). PVP-CMC hydrogel films with incorporated BC and guar gum (GG) showed improved elastic and load-bearing capacity as well as barrier and hydrophobic properties and were found to be 80% biodegradable in 28 days in vermicompost (Bandyopadhyay et al. 2019). In the NCs of poly(butylene adipate-co-terephthalate) reinforced with CNCs fabricated by solution casting method and then covered with Ag thin film by magnetron sputtering, the surface modification caused higher degree of crystallinity of CNCs and decreased their length and diameter. CNCs addition enhanced the storage modulus of the polymer by >200%, contributed to better thermal and mechanical properties and the NC inhibited *E. coli* biofilm formation (Ferreira et al. 2019). By incorporation of polydopamine-functionalized MCC into a konjac glucomannan films were prepared showing improved mechanical properties, pronouncedly reduced WVP, and high thermal stability, which could be recommended as prospective food packaging material (Wang et al. 2019).

A composite of the CMC-based film with functionalized halloysite nanotubes fabricated via adsorbing metal ions on uniformly charged acid-treated halloysite nanotubes showed a considerably improved mechanical and water vapor barrier properties and thermal stability as well as significant antimicrobial activity against both *E. coli* and *L. monocytogenes* (Wang and Rhim 2017). Bio-NC films prepared by incorporation of two types of bentonite with the platelet aspect ratios 300–500 (PGN) and 150–200 (PGV), respectively, into CNF matrix showed lower CNF degradation temperature and strength compared to neat CNF films. The PGN-containing samples were more hydrophilic and at 15% PGN the water vapor transmission rate decreased from 425 to 375 g/m² per day, while higher PGN amounts exhibited adverse impact; PGN also intensively suppressed the O₂ passage in dry state and to a lower extent at increased RH (Zheng et al. 2019). Cellulose NC foams containing low amounts of surface-modified MMT exhibited pronouncedly better thermal, mechanical, and barrier properties than the pure cellulose foam and could represent a good alternative to expanded polystyrene foam trays for dry food packaging (Ahmadzadeh et al. 2015). Corn starch/MMT/CNF composite films prepared using one-dimensional (1D) CNFs and two-dimensional MMT plates showed increased TS and Young's modulus and transparency, while reduced moisture susceptibility compared to neat starch film suggesting that using binary fillers with different geometric shapes and aspect ratio biodegradable starch-based NCs suitable for food packing and preservation can be designed (Li et al. 2019a). NC with PEG and

Na-MMT nanofillers strongly improved the tensile mechanical properties of CMC film resulting in up to 260% increase of modulus, up to 250% increase in the strength and up to 300% increase in elongation and a ca. fivefold decrease in WVP compared to neat CMC film suggesting its applicability in edible food packaging (Fiori et al. 2019).

In cellulose-based composite foams prepared using nanoclay the presence of nanoscale clay in the cellulose matrix contributed to better uniformity of foam structure as well as to higher density, compressive strength, and Young's modulus suggesting that such composite foams could replace the commercial synthetic foams applied for packing of food products (Ahmadzadeh et al. 2016).

By integrating hydroxyapatite (HAp) NPs into a CNF matrix using the one-directional freeze-drying technique NC films were prepared, which can function as ammonia sensors and could be used as indicators for freshness in smart and biodegradable food packaging materials. The gas-sensing performance was affected mainly by the amount and morphology of HAp, its distribution within the microporous CNF matrix, and doping on its surface; at application 5 wt% of HAp the detection limit of NH_3 was 5 ppm, the sensitivity reached up to 575%, and response/recovery was achieved in 210/30s (Narwade et al. 2019).

Idumah et al. (2020) in a review paper devoted attention to surface treatment approaches of some nanomaterials (e.g., cellulose, nanoclay, halloysite nanotubes, graphene, and CNTs) and their impact on properties of composite films applied for packaging, highlighting the innovations in polymeric NC packaging materials and electrical sensors for food and agriculture. In ternary NC of poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV)/CNC/GO fabricated via a simple solution casting method containing 1 wt% covalently bound CNC-GO NCs prepared by chemical grafting, the TS and elongation to break increased by 170.2 and 52.1%, respectively, and maximum degradation temperature (T_{max}) increment showed an increase by 26.3 °C compared to neat PHBV; the ternary NC also showed very good barrier and antibacterial activity as well as lower migration level for food stimulants than the neat PHBV or binary noncovalent NCs of CNC and GO (1:0.5 wt%) (Li et al. 2019b).

Addition of thyme EO (20–40% w/w) to the bio-NCs of whey protein isolate (WPI) and CNF resulted in reduced WVP, increased crystallinity index, and glass transition temperature. The formed films were less rigid and elastic and showed a decrease in TS, elongation at break, puncture strength and deformation, and elastic modulus. The color of bio-NC films containing EO was less yellow, showing a tendency to green, less saturated, and less transparent than the WPI-CNF films (Carvalho et al. 2020). Methylcellulose (MC)/CNC-based NC films containing encapsulated nanoemulsion of oregano and thyme EOs with the size 100 nm and showing a slow release (35%) of volatile components over 3 months of storage, improved TS and decreased water barrier properties, were able to reduce the fungal growth in infected *Oryza sativa* plants during 2 months of storage at 28 °C, whereby these favorable properties could be enhanced by irradiation of bioactive films at 750 Gy (Hossain et al. 2019). Active NC films based on soy protein isolates, clove EO, and microfibrillated cellulose (MFC) with diameters of 50–60 nm, a length of $485 \pm 2 \mu\text{m}$, a high aspect ratio of 8800, and 35.5% of crystallinity, in which MFC

reinforced the protein matrix, showed increased mechanical strength and Young's modulus of the films, enhanced barrier properties to O₂ and water vapor; increased antioxidant properties and antimicrobial activity against bacteria with increasing MFC content were observed. On the other hand, clove EO increased the O₂ permeability and decreased WVP (Ortiz et al. 2018).

Soy protein-based packaging materials with the incorporated CNFs, pine needle extract (PNE), and lactic acid showed enhanced TS due to the presence of CNFs and significantly improved light barrier property mediated by CNF and PNE. Films containing PNE also showed pronounced antioxidant activity and antimicrobial effectiveness against foodborne pathogens (*S. aureus*, *E. coli*, *L. monocytogenes*, and *S. typhimurium*), although CNFs mitigated the release of both active compounds from the film matrix (Yu et al. 2019a).

Yang et al. (2020) designed multi-nanofibers composite film based on hybridization of bacterial CNF/CS, in which curcumin (Cur) MPs/NPs were dispersed. This composite film exhibited better mechanical strength and barrier property due to the presence of bacterial CNF as well as notable antioxidant capacity and antibacterial effectiveness and was able to detect pH change and trace amount of H₃BO₃ suggesting its suitability as a smart and active food packaging material.

Multifunctional NC/metal or metal oxide (e.g., AgNP, ZnO NP, CuO NP, and Fe₃O₄ NP) hybrid nanoscale materials characterized with high antibacterial efficiency, ultraviolet barrier, and mechanical properties could be recommended not only for food packaging, but also for biopharmaceutical, biomedical, and cosmetics formulations (Oun et al. 2020). Antibacterial CMC-based nanobiocomposite films containing three types of metallic NPs (AgNPs, CuO NPs, and ZnO NPs) at concentrations 2 wt% showed improved mechanical properties and lower WVP values than the pure CMC film and films containing individual NPs in the same concentration. Moreover, the CMC films with incorporated metallic NPs exhibited antibacterial properties against *S. aureus* and *E. coli* (Ebrahimi et al. 2019).

In CNF/AgNPs NCs fabricated using UV irradiation the AgNPs with mean size ca. 28 nm coated on CNFs interfered with the formation of intra-chain and inter-chain H-bonds of cellulose and the NC showed significant antimicrobial activities against *S. aureus* and *E. coli* (Yu et al. 2019b). As a suitable material for food packaging applications, cellulose NC films with in situ generated AgNPs using tamarind nut powder as a reducing agent showing good antibacterial activity and improved mechanical properties were recommended by Mamatha et al. (2019). NC films fabricated from a mixture of cellulose acetate and nearly spherical AgNPs with sizes 5–18 nm, green synthesized using different polyphenolics, showed significant antibacterial activity against *E. coli*, *S. aureus*, *S. typhi*, *B. cereus*, and *K. pneumoniae* but low activity against *Pseudomonas* spp. and released less AgNPs than the allowed limit indicating that they could be applied as a food packaging system (Marrez et al. 2019). Composites of CNC–AgNPs with sizes up to 30 nm prepared using dialdehyde cellulose nanocrystal as both reducing and stabilizing agent showed notable antibacterial activities against *S. aureus* and *E. coli* and their addition into the pulp resulted in handsheets exhibiting pronounced antibacterial activities, higher mechanical properties, and a reduced air permeability; such composites could be

applied in food packaging materials (Xu et al. 2019c). BC–AgNPs composites containing <2% (w/w) AgNPs showed better antimicrobial activity than colloid Ag with the same concentration and were found to protect foodstuff from microbial spoilage for 30 days, while in foodstuff stored in regular polythene bag spoilage was detected within 15 days. Consequently, the BC–AgNPs composites with Ag concentration <2% are suitable as a lining of regular food packaging material in order to prolong shelf life till one month (Adepu and Khandelwal 2018). TS of CNC/Ag/ALG bio-NC films designed by Yadav et al. (2019) increased up to 57%, while WVP decreased up to 36% compared with neat ALG films and the bio-NC films also showed improved UV and water barrier properties than the pure ALG films. PVA added to the antimicrobial films fabricated by incorporating AgNPs into NC at concentration 3 wt% contributed to 20-fold rise in the elongation at break of the films compared to pure NC film, improved oxygen barrier capacity, and a moderate improvement of WVP was observed as well. Moreover, such NC films exhibited significant antimicrobial activities against *E. coli*, showing growth inhibition of naturally present bacteria for at least 10 days at 4 °C (Wang et al. 2020b). A biodegradable packaging film based on CMC/PVA-zeolite doped with Au³⁺ possessed TS up to 8.69 kgf/mm² compared to 2.05 kgf/mm² of the neat CMC/PVA film; NC film showed enhanced water vapor transmission and gas transmission rate due to the presence of zeolite doped with Ag⁺ or Au³⁺ and exhibited improved antimicrobial properties (Youssef et al. 2019b).

Cellulose-based papers modified with Ag–TiO₂ NCs tested as packaging were found to be most effective in the preservation of bread nutritional compounds, while those modified with Ag–TiO₂–zeolite NC were able to prolong the bread microbiological safety for 10 days at 20 °C and for 12 days at 4 °C, i.e. 2 days longer than the cellulose-based papers modified with Ag–TiO₂ NC (Mihaly-Cozmuta et al. 2017). A blend suspension of wheat gluten containing 7.5% CNCs and 0.6% TiO₂ NPs applied to coat commercial packaging unbleached kraft paper with three coating layers resulted in an increase in breaking length and burst by 56 and 53%, respectively and papers coated with suspension containing TiO₂ NPs showed >98.5% decrease in number of viable bacteria compared to TiO₂ NPs-free coated paper (El-Wakil et al. 2015).

The NC of agar and CMC with MMT modified by Ag showed strong antibacterial activity against *E. coli* and *Bacillus subtilis* indicating its suitability to preserve food by controlling foodborne pathogens and spoilage bacteria when used as packaging material (Makwana et al. 2020). Cellulose acetate-based nanobiocomposite films containing green-synthesized AgNPs/gelatin-modified MMT nanofiller and thymol showing not only improved TS and oxygen barrier, antioxidant properties and antimicrobial activities against bacteria and fungi suitable for active food packaging use were designed by Dairi et al. (2019).

Addition of increasing concentrations of CuO NPs (1, 1.5, and 2%) to bio-NCs based on the biodegradable branched polysaccharide kefiran and CMC increased contact angle and ultimate TS and decreased WVP and elongation at break; improved color parameters and percentage of light transmission with increasing CuO NPs concentration and inhibitory effects against *E. coli* and *S. aureus* were

estimated as well indicating that such NCs can be utilized in food packaging (Hasheminya et al. 2018). In bio-NCs films based on kefiran-CMC carbohydrates with incorporated CuO NPs and *Satureja khuzestanica* EO the simultaneous application of CuO NPs and EO pronouncedly enhanced the physical and mechanical properties of the films, caused synergistic effect on antimicrobial properties against *E. coli* and *S. aureus* and considerably changed the color parameters and percentage of UV and visible light transmission (Hasheminya et al. 2019).

Films based on BC modified by polypyrrole-ZnO NPs NC, which can be applied in antioxidative food active packaging and smart packaging, were designed by Pirsá et al. (2018). Zhao et al. (2017) developed hierarchically structured cellulose@ZnO NCs based on the formation of hierarchical three-level structures induced by self-assembly, namely cellulose/ZnO nanofibers, layers, and microfibers, and in the final step, ZnO MPs were deposited onto the surface of cellulose/ZnO microfibers. As the crucial driving force for the formation of cellulose/ZnO nanofibers, the electrostatic attraction between cellulose and ZnO was estimated. Such cellulose@ZnO NC showed increased antibacterial activities against *E. coli* and *S. aureus* and could be applied in food packaging materials. Poly(3-hydroxybutyrate) (PHB) modified by BC nanofibers using plasma treatment showed an increase of stiffness and strength as well as improved antibacterial activity reflected in growth inhibition of *E. coli* (by 63%) and *S. aureus* (44%), respectively. Application of the ZnO plasma coating manifested as continuous layer of self-aggregated ZnO NPs on PHB surface strongly affected the mechanical and thermal properties of PHB-BC NC and completely inhibited *S. aureus* growth (Panaitescu et al. 2018).

4 Conclusion

The population of our planet is growing rapidly, so it is necessary to ensure the production of the necessary amount of healthy crops, but also to focus on reducing food waste by extending the shelf life of food. Therefore, the use of suitable packaging that has suitable mechanical properties, thermal stability, gas barrier properties, and electrical conductivity and exhibits notable antimicrobial activity able to reduce spoilage as well as eliminate negative effects on human health is necessary. Currently, mass-used plastic packaging is not biodegradable and causes enormous environmental pollution. Moreover, various toxic agents are released during its combustion. For example, during the combustion of polyethylene, polypropylene, and polystyrene, toxic monomers of these plastics are formed, which have a negative effect on the nervous system and blood circulation. Although some plastic packagings can be recycled, a significant part accumulates in landfills, but also reaches the seas and oceans, where they endanger marine animals in the ingestion of plastic waste. The solution is an effective transition to the use of biodegradable biopolymers—materials from natural renewable sources (very often residues from other products, i.e. reuse of existing waste), which include cellulose and chitosan. The rapid development of nanotechnologies has enabled the production of efficient

nanocomposites, based on the incorporation of so-called “nanofillers” (e.g., essential oils, metal NPs, cellulose nanofibers, cellulose whiskers, etc.) into a matrix of biopolymers, thereby improving the mechanical, barrier, and antimicrobial properties of nanocomposite films that can be used as biodegradable packaging or edible coatings. Instead application of synthetic preservatives, prolonging the shelf life of foods can be achieved by incorporating compounds with antimicrobial activity in their bulk, but mainly in nanosized form into biodegradable films from natural sources used as packaging. By appropriate selection of the nanocomposite components, the prepared film can act as an active/intelligent coating that is able to monitor changes in temperature, pH, various decomposition products, or the presence of bacteria in a real time. Let us therefore believe that these more environmentally friendly and at the same time more food- and consumer-protective packaging will, after thorough toxicological and environmental tests, become a common packaging in which food and other products intended for consumption will be packed.

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