A short review: Nanocellulose for smart biodegradable packaging in the food industry

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Abstract. The role of food packaging has increased beyond by changing consumer preferences and expectations. In addition to these essential functions, the packaging extends shelf life, improves quality, and develops from environmentally friendly material. Nanocellulose is one of the renewable and natural source products that has been widely studied. Nanocellulose can develop into food packaging material due to its properties of oxygen and water vapor barriers when used as layers, fillers in composites and thin films stand cents. However, the use of nanocellulose is still limited due to its lack of physical properties. On the other hand, research in nanocellulose for smart packaging applications enhanced with several sensors or indicators accelerated in the last decade. This review mainly summarizes the fundamental properties of nanocellulose as a food packaging material, preparation nanocellulose material, their application on smart packaging, and their future perspective. The selection of nanocellulose material preparation methods and the selection of active ingredients that play a role in active and intelligent functions in packaging considerably determine the success of nanocellulose applications in food packaging main criticism appears of the difference between the results obtained in the model test and actual food storage conditions.

1. Introduction

The global production of cellulose is nearly 7.5 million tons every year [1]. Cellulose consists of crystalline regions and amorphous regions that can be utilized to obtain nanocellulose [2]. Nanocellulose has a high potential to develop into food packaging due to its properties. Nanocellulose has a very high surface area and crystallinity that can be adjusted. It gives nanocellulose materials great potential to continuously enhance the properties of oxygen and water vapor barriers when used as food packaging material [3]. There are three types of nanocellulose: bacterial nanocellulose, nanocrystalline cellulose, and cellulose nano-fibrillation. Cellulose nano-fibrillation has good permeability for oxygen, which is due to its dense network structure. Whereas in nanocrystalline, the density can be attributed to small and uniform particles.

Food packaging has four essential functions: protection, communication, comfort, and containments [4]. However, food packaging has increased beyond changing consumer preferences, and packaging also contributes to extending shelf life, improving quality, and developing from environmentally friendly material. Packaging objectives protect food quality but cannot be appreciated when causing natural resources and waste disposal [5]. Each packaging material has certain advantages, including
certain types of packaging suitable for certain types of food, such as solid, semi-solid (pasta), and liquid (beverage) food. Not all packaging materials are safe for food and health. Food packaging safety requirements are as follows: Food packaging must not be toxic and leave no residue on food, must be able to maintain the shape, taste, hygiene, and nutrition of food ingredients; Packaged toxic substances must not migrate into packaged foodstuffs; The shape, size, and type of packaging can provide effectiveness; and packaging materials do not pollute the environment.

Consumer demands regarding food packaging continue to increase along with the increasingly fierce competition in the food product market. One of the promising strategies in winning the food product market is to create interactive packaging. Interactive packaging promises several features that are not found in conventional packaging. An interactive packaging system is classified as active and intelligent packaging. Active packaging is specially designed to extend shelf life, improve product quality and convenience for consumers. Active packaging is generally equipped with an absorber or emitter that is part of the packaging or separately with packaging in the form of labels or sachets. Active components (absorbers and emitters) for active packaging will interact with food products and play an active role in extending shelf life and improving product quality [6]. Emitters in active packaging play a role in releasing antioxidant compounds and antimicrobial compounds to inhibit oxidation and the growth of microorganisms and controlled respiration rates [7]. Whereas intelligent packaging does not act directly on extending the shelf life of food products, it aims to provide real-time product information. Intelligent packaging has an indicator for monitoring changes in the internal and external environments and communicating the packaged food product [8]. Indicators on intelligent packaging can be part of the package itself (ink printed on the package, part of the packaging cover, barcode) or separate from the package (labels, sachets, tags), which monitors the product’s condition in real-time. Another aspect that draws attention to Intelligent packaging is how sustainable the intelligent packaging concept is global.

Based on its existence, food packaging can be divided into primary, secondary, and tertiary packaging. The most important thing to note is food packaging as primary packaging because it is in direct contact with food, so it has tremendous potential for the transfer (migration) of substances/components from food packaging into food. In the food processing process, physical and chemical changes can occur, whether desired or not. After going through the processing process, food does not remain stable but can continue to change, so that it is necessary to choose the proper packaging so that the shelf life of food products can be increased and the nutritional value can still be maintained. Currently, many countries are increasingly paying particular attention to food packaging safety in circulation because of the potential for components from packaging to migrate into food. Some components of food packaging can cause harmful effects and endanger health. These hazardous components can come from residue packaging starting materials such as monomers, catalysts used to speed up the reaction rate, decomposition products of essential ingredients, and additives used in the food packaging manufacturing process. Therefore, bio-degradable and bio-based packaging materials have received significant attention due to food preservation. This review mainly summarizes the fundamental properties of nanocellulose as a food packaging material, preparation nanocellulose bio-composite, their application on smart packaging, and their future perspective.

2. Nanocellulose definition and classification
Nanocellulose is a cellulose material with at least one of its dimensions in length, width, or diameter on a nanometer scale [9]. The chemical and mechanical approach can be used to obtain the nanocellulose crystal or fibers from the cellulose chain or synthesize microorganisms [10]. In addition, to obtain nanocrystal cellulose, acid hydrolysis and enzymatic hydrolysis are chemical techniques that often to be used. We can use high-pressure methods and high-speed defibrillation of cellulose chains to occurs and provides cellulose fibers in the nano-dimensional range [11]. The mechanical properties of BNC tubes are comparable to values reported in the literature, which indicates excellent potential in vascular implants or functional replacements in biomedicine. Production techniques, preparation techniques, and their primary nanocellulose function depend on the source and processing conditions of extracted
cellulose's inherent specific intrinsic properties, such as crystallinity, thermostability, and morphology. Moreover, the schematic preparation method of the nanocellulose family is shown in Figure 1.

![Figure 1. Methods for preparation nanocellulose family.](image)

In general, nanocellulose is classified into three, nanofibrils cellulose (NFC), nanocrystal cellulose (NCC), and bacterial cellulose (BNC) [12]. NFC and NCC extraction and isolation methods from cellulose sources are commonly called top-down methods [9], while the synthesis of nanocellulose by bacteria (BNC) is the bottom-up method. BNC is already lignin-free and no longer requiring chemical treatment or intensive milling to achieve nanoscale dimensions.

3. Preparation of nanocellulose bio-composite

Nanocellulose (NC) based composites can act as active packaging by incorporating active agents, mainly antioxidants, antimicrobials, UV blockers, O2 absorbers, alcohol emitters. Incorporating various types of organic and inorganic nanofillers into biopolymer substrates is one of the most effective approaches for improving the performance of nanocellulose in food packaging [13]. Nanocomposite preparation films can improve their mechanical properties and heat resistance and gain barriers against water vapor and gases [22]. NC can also act as a carrier of active agents in smart packaging, such as antioxidants, UV-blocking, and antimicrobials, to enhance the shelf life of many food products [16]. Numerous methods have been used to prepare nanocellulose bio-composite, such as solvent casting, supercritical solvent impregnation (SSI), physical mixing, in situ polymerization, extrusion, and layer by layer formation.

Solvent casting is one of the simple processes to produce bio-composite. In this method, Nanocellulose can be dissolved in water or organic solvent using mechanical stirring/ultrasonication at room temperature or mixing by an autoclave at high temperatures [16]. Another approach is supercritical solvent impregnation SSI, and it uses supercritical carbon dioxide as the solvent under mild temperature to impregnate the active components into the polymeric packaging matrix [17]. The Nano Fibrillated Cellulose and Mango Leaf Extract (MLE) the biobased films prepared by SSI showed a clear advantage in antioxidant and antimicrobial properties. Developing method of novel bio-nano composites for active/intelligent packaging showed in figure 2., guest bio-compounds such as antimicrobials, dyes can be incorporated into the NC membrane with two different impregnations (ex-situ) and bottom-up built methods [24]. Clove essential oil (CEO) was incorporated by impregnating into bio-composite of chitosan and oxidized nanocellulose to enhance their antimicrobial and antioxidant activity [18]. Impregnation of 5% PVA solution in the nanocellulose bacterial matrix through the in-situ method did not cause significant changes in the arrangement of BC fibrils and could increase transparency and elongation compared to pure BC. However, it has a significant effect in reducing the strength and stiffness of the film [19].
Hybridization of nanocellulose with nanoparticles can improve the functionality of nanocellulose. Some approaches can mix nanoparticles such as nisin, MgO, and other metal nanoparticles with nanocellulose [20]. The widely used method for the manufacture of nanohybrids is the direct mixing method, and another approach is the in-situ coprecipitation method. The in-situ synthesis in BC tissue prepared MgO-bacterial cellulose (BC) nanohybrids through two methods, namely sonochemistry and wet chemistry. The ex-situ approach was done first by prepared nanocellulose and nanoparticles (NPs) separately and then mixed and interacted through various approaches followed by a light washing step to remove all unbound NPs. In this method, the nanoparticles are preloaded in the polymer matrix to serve as NP precursors first, where the ions are supposed to be uniformly distributed. The loading capacity and antibacterial activity of the ex-situ method of nanohybrid is higher than the loading capacity and antibacterial activity of the in-situ method [21].

4. Application of nanocellulose bio-composite as smart packaging

The concept of smart packaging has become an exciting topic in the food packaging sector, aiming to improve the quality and value of food products by performing one or more intelligent functions [12]. The most common active packaging system is equipped with the ability to release active compounds (such as antimicrobial agents) onto the surface of food [22]. Hybrid film CNFs with the addition of nisin used as a plastic packaging coating of LDPE were effective in inhibiting the growth of Listeria monocytogenes in ready-to-eat ham during 7 days of storage at 4 °C [23]. Another application of antimicrobial paper was from I. cylindrica, which was coated with cationic crosslinked anionic nanocellulose (H+ and Al3+) to control the growth of Gram-negative bacteria E. coli and S. typhi as well as Gram-positive S. aureus and B. subtilis [24]. The addition of nanoparticles to the nanocellulose based smart packaging can increase the ability to inhibit the growth of microorganisms and can improve physical and mechanical characteristics and smart packaging.

In contrast to active packaging, intelligent packaging aims to monitor product quality in real-time. Freshness indicator is a type of intelligent packaging that aims to inform the freshness of the product during storage. For instance, the application of hydrogen sulfide (H₂S) colorimetric sensors in meat and poultry products. Where the active compound is used in the form of Ag⁺ nanoparticles with a polymer matrix. Ag particles will react with H₂S compounds produced as a result of the deterioration process to produce an Ag₂S compound which can be seen in changes in color from yellow to colorless in chicken breast and silver carp in the packaging system [25]. Another colorimetric freshness indicator was developed from bagasse nanocellulose based hydrogel by TEMPO-mediated oxidation to monitor chicken breast spoilage. The hydrogel matrix acts as a pH-responsive with bromothymol blue/methyl red as a pH indicator. The more CO₂ produced by the deterioration process of chicken, the lower the pH.
of the product was, resulting in a color change in the pH indicator [26]. Nanocellulose-based hydrogel matrix can be responsive to stimuli, one of which is the pH stimulus. Impregnating a pH indicator dye to the hydrogel matrix can detect damage to poultry-based products through visible color changes. The application of Bromothymol blue/ methyl red as a pH indicator has several advantages: resistance to storage conditions such as high temperatures, changes in pH, and UV light. However, it also has several weaknesses, including the pH range is not exhaustive and is made of chemicals that can contaminate food. If we have to use a dye that is not a natural colorant, it should be chosen according to food-grade specifications.

Intelligent packaging equipped with a pH indicator based on bacterial nanocellulose (BC) matrix with bioindicator was extracted from black carrot anthocyanin (CA) was developed and characterized to monitor the freshness/rotteness of rainbow trout and carp fillets. The bioindicator's color represented the quality of fish fillet, dark red when fresh (indicator is dark red), charm pink in the best condition for eating, and jelly blue-khaki color when both types of fish fillets rotten [27]. Anthocyanins as bioindicator compounds have been successfully developed into intelligent packaging using chitosan and cellulose-based matrix to monitor the deterioration of milk quality. The deterioration in the quality of fresh pasteurized milk is detected through a visible color change from blue to purple after 48 hours of storage at 20 °C and can be observed with the naked eye. [28]. Anthocyanin compounds from grapes impregnated on nanocellulose bacteria by ex-situ method can detect the freshness of minced meat stored for up to 7 days where the indicator color changes when the minced meat is rotting from red when the meat is fresh and turns blue, representing the decay rate [26]. Anthocyanins act as antioxidant compounds and detect spoilage in protein-based products (milk and marine products) by detecting changes in pH. When fish-based products start to deteriorate, the pH of the headspace between the product and the packaging will increase due to the increasing number of protein-derived compounds released due to the decay process. It will be detected by the anthocyanin indicator so that it changes color from red to blue.

On the other hand, the pH of the fresh milk is close to neutral and will decrease and associated changes in milk during storage. Therefore, the anthocyanin indicator in dairy products will change color from blue to reddish blue if it decreases in quality. Bioactive compounds in nanocellulose-based smart packaging can be antioxidant, antibacterial, pH indicator dyes, and nanoparticles. Antioxidants can be added in the form of extracts from plants (tannin, pine needle extract, mango leaf extract, and black carrot extract) or in the form of essential oils such as clove oil. Bioactive compounds based on plant extracts based on polyphenols play a role in inhibiting oxidation reactions and act as pH indicators. Anthocyanins, for example, can function as bioindicators that can change color due to changes in pH. It is similar to the pH indicator's working mechanism, which can change color due to changes in pH in packaged food products. It follows the working principle of the freshness indicator and can be applied to poultry, fish and processed fish products, and meat, milk, and their products. The stability color of natural colorant solution in each storage condition of food products is different. Anthocyanin compounds have better color stability than other dyes. However, anthocyanin compounds are sensitive to high temperatures, so it is recommended to use them to monitor the quality of products stored at low temperatures.

Meanwhile, condensed tannin compounds, pine needle extract, rosemary essential oil, and clove oil have more roles in inhibiting the growth of microorganisms and inhibiting the occurrence of oxidation reactions. The addition of nanoparticles such as nisin, silver, and activated carbon in the smart packaging system can increase the antibacterial ability and improve the physical and mechanical capabilities of nanocellulose-based packaging. To conclude the application of nanocellulose in food packaging, the list of applications of nanocellulose to improve novel functional properties showed in Table 1.
Table 1. List of recent works on the development nanocellulose based Smart packaging.

| Active compound                     | Source                          | Matrix nanocellulose | Application                  | Preparation method                        | Objective                                | Ref. |
|-------------------------------------|---------------------------------|-----------------------|------------------------------|-------------------------------------------|-------------------------------------------|------|
| Condensed tannins                   | *Acacia mearnsii* bark          | Cellulose nanofibrils | Dry foods, vegetables, and meat | Physical mixing                          | Increase shelf life                       | [29] |
| Nisin                               | Sugarcane nisin                 | Cellulose nanofibrils (CNFs) | Ready-to-eat ham          | Physical mixing                          | Improve antimicrobial properties         | [30] |
| Clove oil                           | Clove, Date palm sheath fibers  | Bio-composite of chitosan (CS) and oxidized nanocellulose biopolymers | Packaged food              | Impregnating method                      | Improve antimicrobial properties and antioxidant | [18] |
| Polyphenolic-rich extract           | Mango leaf extract              | Nano-fibrillated cellulose (NFC) | Packaged food          | Super-critical solvent impregnation (SSI) and conventional solvent casting film-processing methodologies | UV-blocking, antioxidant, antimicrobial | [15] |
| *Cedrus deodara* pine needle extract| *Cedrus deodara* pine needle    | Soy protein-based films incorporated with cellulose nanocrystals (CNCs) | Packaged food              | Physical mixing                          | The suitable mechanical properties, antioxidant ability, and water vapor barrier capacity | [31] |
| Rosemary essential oil, TiO₂ nanoparticles | Rosemary essential oil, TiO₂ nanoparticles | Cellulose nanofibers | Packaged food              | Casting and evaporation method           | Improved physic-mechanical properties, antioxidant, antimicrobial | [32] |
| Soy protein isolate, Zinc oxide nanoparticle | Soy protein isolate, Zinc oxide nanoparticle | Nanocrystal          | Pork                        | Physical mixing and in situ method        | Antimicrobial especially for foodborne pathogens and reduce TVBN | [13] |
| pH-responsive dyes (bromothymol blue/methyl red) | Sugarcane bagasse               | Nanocellulose was prepared by TEMPO-mediated oxidation | Chicken                    | Immersing the nanocellulose hydrogel into the above indicator water solution | Freshness indicators, UV blocking, antioxidant | [23] |
| Anthocyanins                        | Black carrot                    | Cellulose-chitosan matrix | Pasteurized milk            | Impregnated by a sol-gel method          | Improve antimicrobial properties and antioxidant | [28] |
| Anthocyanins                        | Black carrot                    | Bacterial nanocellulose (BC) | Rainbow trout and common carp fillet | Ex-situ method                           | Improve antimicrobial properties and antioxidant | [27] |
| MgO                                 | MgO powder                      | Bacterial nanocellulose (BC) | Food packaging material | In-situ synthesis                        | Improve antimicrobial properties         | [21] |
| Silver nanoparticles (AgNPs)        | Silver nanoparticles (AgNPs)    | Bacterial cellulose nanocrystals (BCNC) | Food packaging           | Physical mixing                          | Improve antimicrobial properties and antioxidant | [33] |
5. Future perspective of nanocellulose based smart packaging

Nanocellulose has excellent potential to be developed into smart packaging materials for food products. Based on its physical and mechanical characteristics, nanocellulose has an excellent opportunity to be used as a polymer matrix and can meet food packaging requirements. To improve the physical, mechanical, moisture, and gas properties, antimicrobial, antioxidant, and UV-blocking properties, nanocellulose can be combined with other materials through several ex-situ and in-situ methods. Active ingredients can be added by making bio-composites, nanohybrids, impregnation dyes, and coatings. The active ingredients used also depend on the desired packaging properties. Active packaging can be done by adding absorbent compounds (such as oxygen absorbers, moisture absorbers) or emitting compounds such as antioxidants and antimicrobials according to the packaged product's needs. As for intelligent packaging, active ingredients must be added that can function as indicators such as pH indicators, temperature indicators, gas indicators, and even indicators of the presence of microorganisms.

Based on the results of various studies, some information has been obtained regarding the potential use of active and intelligent packaging systems that are effective with the use of nanocellulose-based nanocomposite matrices to improve and monitor the quality of various food products. Further development of nanocellulose applications in smart packaging can focus on improving safety, and research can be carried out to prevent toxicity in the films, which may be caused by storage degradation and thus result in stable packaging. It needs to be further developed related to the consistency of smart packaging during the storage and distribution process. If possible, stimuli-responsive packaging can be developed, which will only release bioactive compounds if there is a stimulus such as changes in temperature, pH, humidity, and UV light.

6. Conclusions

Nanocellulose in BNC, CNF, and CNC can be developed into smart packaging because it has reasonable physical, mechanical characteristics and barrier ability to oxygen and water vapor. Nanocellulose can be combined with active compounds to extend shelf life, improve product quality, and monitor the packaged product's actual condition. The selection of the source of nanocellulose (CNC, CNF, BNC), the method of preparation of nanocellulose materials, and the selection of active ingredients that play a role in active and intelligent functions in packaging significantly determine the success of nanocellulose applications in food packaging. Further research is needed to determine the effectiveness and stability of nanocellulose-based smart packaging during the actual food storage and distribution process. As well as how to maintain the consistency of the activity of active compounds during food storage and distribution. The application of plant extracts can be an alternative bio-based indicator and an environmentally friendly source of antioxidants and antimicrobials. Anthocyanin compounds are one type of natural colorant that can act as antioxidants and bioindicators with a wide pH range, visible color changes, and resistance to changes in pH and sunlight. However, the application of anthocyanins in smart packaging has several weaknesses, and anthocyanins are sensitive to high temperatures, so they are recommended to monitor the freshness of refrigerated or frozen food products.

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