

Application of Nanocellulose in Food Packaging: A review

Sh. Shahsavari

*Chemical Engineering Department, Varamin-Pishva Branch, Islamic Azad University,
Varamin, Iran*

Received: 24 September 2020; Accepted: 26 November 2020

ABSTRACT: The food business has a high demand for sophisticated and environmentally friendly packaging materials that have improved physical, mechanical, and barrier capabilities. Synthetic and non-biodegradable materials are commonly used, raising environmental issues. As a result, in recent years, research has focused on the creation of bio-based sustainable packaging materials. The potential of nanocelluloses as nanofillers or coatings for the production of bio-based nanocomposites is covered in this review, which includes: the effect of nanocellulose modification/functionalization on the final qualities, the physico-chemical interaction of nanocellulose with the neighboring polymeric phase.

Keywords: *Barrier properties, Composite, Food packaging, Nanocellulose*

INTRODUCTION

Packaging is important in the food supply chain because it protects and contains food from the processing and manufacturing phases to distribution, handling, and storage until it reaches the final consumer. Food packaging currently makes up the majority of the entire packaging industry (85 percent). Revenues from the worldwide packaging market grew from \$42.5 billion in 2014 to roughly \$48.3 billion by 2020 [1]. Plastics are the most often used packaging materials because they are lightweight, processable, have cheap production costs, and have excellent mechanical and barrier characteristics [2]. The market for plastic packaging has been growing at a pace of 20–25 percent each year [3]. Nanocellulose may be extracted from plants or made by microbial fermentation [4]. The nanocellulose

parameters, such as crystallinity, degree of polymerization (DP), fiber diameter, and length, are influenced by the extraction/production technique, which are important in defining mechanical and barrier qualities [5]. The biopolymer's chemical formula is $(C_6H_{10}O_5)_n$ DP, regardless of the cellulose source. NC has demonstrated remarkable potential to strengthen bio-based materials because to its availability, renewability, and degradability, as well as its physical-chemical and morphological characteristics [6–8]. Plant cellulose, namely paper and board, Cellophane TM, and modified cellulose derivatives such as cellulose acetate, methylcellulose (MC), hydroxypropyl cellulose (HPC), hydroxypropyl methylcellulose (HPMC), and carboxymethylcellulose (CMC), has a long history of usage in food packaging [9].

(*) Corresponding Author - e-mail: Sh.shahsavari@srbiau.ac.ir

Food Packaging Necessities

Packaging has been increasingly important in modern life, with its use increasing over time. The fundamental objective of packaging is to keep food items safe from the outside environment while being cost-effective and meeting industry and regulatory standards and customer expectations [10]. The protection provided by the package is an important element of the preservation process for the majority of food products. The requirements of a packaging system for fresh, frozen, dehydrated, thermal, or aseptic processed products are determined by: (i) intrinsic properties of the food product, such as water activity and oxidation potential, which determine perishability; and (ii) intrinsic properties of the food product, such as water activity and oxidation potential, which determine perishability; (ii) Packaging has been increasingly important in modern life, with its use increasing over time. The fundamental objective of packaging is to keep food items safe from the outside environment while being cost-effective and meeting industry and regulatory standards and customer expectations [10]. The protection provided by the package is an important element of the preservation process for the majority of food products. The requirements of a packaging system for fresh, frozen, dehydrated, thermal, or aseptic processed products are determined by: (i) intrinsic properties of the food product, such as water activity and oxidation potential, which determine perishability; and (ii) intrinsic properties of the food product, such as water activity and oxidation potential, which determine perishability.

Nanocellulose in Food Packaging

Nanocellulose may be extracted from plants or made by microbial fermentation [12]. The nanocellulose parameters, such as crystallinity, degree of polymerization (DP), fibre diameter, and length, are influenced by the extraction/production technique, which are important in defining mechanical and barrier qualities [13]. The biopolymer's chemical formula is $(C_6H_{10}O_5)_n$ DP, regardless of the cellulose source. NC has demonstrated remarkable potential to strengthen bio-based materials because to its availability, renewability, and degradability, as well as its physical-chemical and morphological characteristics [14-16]. Plant cellu-

lose, namely paper and board, Cellophane TM, and modified cellulose derivatives such as cellulose acetate, methylcellulose (MC), hydroxypropyl cellulose (HPC), hydroxypropyl methylcellulose (HPMC), and carboxymethylcellulose (CMC), has a long history of usage in food packaging [17].

Nanocellulose Sources

The nanosize of the fibres (less than 100 nm) in at least one dimension distinguishes nanocellulose, nanofiber, or nano-structured cellulose. High crystallinity, high degree of polymerization, high mechanical strength, low density, biocompatibility, non-toxicity, and biodegradability are all characteristics of NC [18]. Its chemical surface modification is enabled by the high number of hydroxyl groups. NC production may be classified into two types: bottom-up and top-down. Steam explosion, enzyme-assisted, and acid hydrolysis (using sulfuric and hydrochloric acids) are examples of top-down techniques, which are followed by mechanical treatments (high pressure homogenization, microfluidization, and cryocrushing) [19]. These techniques produce two types of NCs: (i) nanofibrillated cellulose (NFC), which has a diameter of 5 to 20 nm and a length of 2 to 10 micrometers, and (ii) cellulose nanocrystals (CNC), which are the most crystalline structures produced by hydrolysis [20,21]. The differences between NFC and CNC are significant. NFC filaments are longer and more flexible, with alternating crystalline and amorphous areas, whereas CNC filaments are more crystalline and have a rod-like form. NFC crystalline domains are similar in size to CNC crystalline domains. The synthesis of bacterial nanocellulose (BNC) and cellulose from tunicates are two examples of bottom-up techniques.

Nanocellulose Based Composites

Nanocellulose (NC) has been utilized to make nanocomposites for food packaging as both a coating and filler. To improve the composites overall performance, it's important to understand each component's contribution and how they interact. Coating is the process of applying a thin film on the surface of a substrate to create a multilayered material [22]. Fast food, pet food, and bakery items, for example, all require strong oxygen and grease barriers in their packaging [23]. It

has been reported that NC-based coatings are used on paper and paperboard [24]. In general, covering paperboard with NC-based layers reduced oxygen permeability and increased grease resistance, although there was still significant water vapour permeability [25]. Combinations with polypyrrole and PLA, for example, have been found to decrease NC's high water vapour permeability [26]. Before coating the paperboard, polypyrrole particles were added to the NFC suspension. After coating, there was a decrease in oxygen permeability and an increase in mechanical properties [27]. In a different method, a multilayer coating was applied to paperboard with PLA as the outer layer and CNC as the intermediate layer, resulting in reduced oxygen and water vapour permeability [28]. NFC thin layers were deposited on amorphous PLA and semi-crystalline PLA substrates to create transparent multilayer films. A high adhesion was achieved between the layers, resulting in increased mechanical performance [29]. The coating of NFC onto multilayer bio high-density polyethylene (HDPE) film is another intriguing example. The NFC layer on bio-HDPE reduced oxygen permeability (at both 0 and 80 percent relative humidity) and enhanced grease resistance. The water vapour barrier of plain bio-HDPE was not compromised by the NFC layer loading [30].

Nanocellulose Based Composites Processing

Regardless of the type of NC, the circumstances under which the components are processed and the composite manufacturing processes employed are crucial in achieving fiber dispersion in the main matrix, which is critical for the final material's performance. The literature [31] mentions several ways for manufacturing nanocellulosic composites, including:

- Solvent casting
- Melt processing
- Impregnation
- Layer-by-layer assembly
- Coating
- All-cellulose composites

Safety of Nanocellulose Based Composites

All materials in contact with food (FCMs) must fulfill the criteria of the framework Regulation (EC) No 1935/2004, according to current European legislation.

"Materials shall be manufactured in accordance with good manufacturing practice so that they do not transfer their constituents to food in quantities that could endanger human health; or cause an unacceptable change in the composition of the food; or cause a deterioration in the organoleptic characteristics thereof," according to the regulation. Although this regulation does not address specific rules for nanoparticles used in food contact materials, these standards also apply to nanocellulose-based composites. Regulation (EU) No. 10/201 on plastic materials, as well as Regulation (EC) No. 450/2009 on active and intelligent packaging materials, both provide that chemicals in nano-form must be evaluated separately. The mechanisms of mass transfer and interaction between nanoparticles and their host materials and food may differ from those understood at the typical particle size scale. As a result, nanoparticles may cause varying levels of exposure and hazardous characteristics. As a result, pre-market authorizations based on a risk assessment of a drug with a conventional particle size do not cover the use of the same substance in its nano-dimension, which may only be used if expressly authorized and listed in the above-mentioned rules' positive lists [32].

Cellulose Nanofibrils

When utilized in its unaltered state, the barrier characteristics of NC films are good for gases but poor for WV. Water vapor barrier characteristics are being improved on a regular basis. Sharma *et al.* heated the CNF films at 175°C for 3 hours and found that the WV permeability was reduced by 50%. The crystallinity and hydrophobicity of CNF increased after heating, but the porosity decreased, preventing water molecules from diffusing. The ability to interact with the network is provided by a modified CNF with a high specific area and aspect ratio. However, even though its performance as a gas barrier is equivalent to synthetic polymers, TEMPO oxidized CNF (TOCN) falls short when it comes to water barrier characteristics (235 g/m²/day for 50% RH). Bideau, et al used two methods to make TOCN, PVA, and Polypyrrole (PPy) nanocomposite films. I) via chemical polymerization in situ, and II) by dispersing pyrrole over the TOCN/PVA film and allowing it to polymerize. The film's performance was evaluated against gas and water va-

por barrier characteristics in the second processing condition. The outer layer of PPy, which is hydrophobic, protects the film against water penetration (18g/m²/day). This film's performance was similar to the commercially available petro-based polymers. (16.8 g/m²/day).

Final Remark

The goal of this study was to see how useful nanocellulose may be in the food packaging business. A significant amount of research has been published on the creation of composites using various kinds of nanocellulose (NFC, CNC and BNC). Regardless of the matrix phase, nanocellulose was shown to have a beneficial impact, notably enhanced mechanical and oxygen characteristics, especially at low humidity. On the other side, there is a lack of water resistance (both vapour and liquid), which is a crucial quality for food packing. Several methods for modifying nanocellulose to improve water resistance, promote better dispersion in hydrophobic matrices, and offer functionality, specifically for use in active and intelligent packaging, have been documented. Solvent casting, melt processing, impregnation, coating, layer by layer deposition, electrospinning, and all-cellulose composites are some of the methods that may be used to make NC composites, allowing nanocellulose to be used as a reinforcing agent, a barrier agent, and for smart packaging. Before using nanocellulose in food contact materials, studies on its toxicity and the influence of nanocellulose on total composite migration should be examined, with existing data suggesting no or minimal toxicity. However, because nanocellulose is not yet approved for food contact applications, more study is needed to show its safety. Although NC is manufactured industrially and commercially, its primary uses are in the composites and automotive sectors. Due to present technological limitations in terms of rheological behavior, scalability, and overall characteristics of the final material in relation to the needs of today's packaging systems, using NC-based materials to replace petroleum-based plastics remains a big issue. The impact of using reusable and recyclable, but non-renewable materials, as well as food safety and quality requirements, must be considered and balanced with the sustainability of the global process of

handling biomatter and turning into packaging, such as the energy input and use of solvents, for example. Nonetheless, the potential for specific uses of functionalized nanocellulose for specific and tailor-made high-value applications is generally recognized, and this is likely to drive research activities in the near future. To close the legal, economic, and technological gap between sustainable and conventional packaging in the food sector, more research and investment in nanocellulose-based materials should be done.

REFERENCES

- [1] Lee, K.Y.; Aitomäki, Y.; Berglund, L.A.; Oksman, K.; Bismarck, A. (2014). On the use of nanocellulose as reinforcement in polymer matrix composites. *Compos. Sci. Technol.*, 105: 15–27.
- [2] Fang, Z.; Hou, G.; Chen, C.; Hu, L. (2019). Nanocellulose-based films and their emerging applications. *Curr. Opin. Solid State Mater. Sci.*, 23: 100764.
- [3] Khan, A.; Huq, T.; Khan, R.A.; Riedl, B.; Lacroix, M. (2014). Nanocellulose-Based Composites and Bioactive Agents for Food Packaging. *Crit. Rev. Food Sci. Nutr.*, 54: 163–174.
- [4] Soykeabkaew, N.; Tawichai, N.; Thanomsilp, C.; Suwanton, O. (2017). Nanocellulose-reinforced 'green' composite materials. *Walailak J. Sci. Technol.*, 14: 353–368.
- [5] Hubbe, M.A.; Ferrer, A.; Tyagi, P.; Yin, Y.; Salas, C.; Pal, L.; Rojas, O.J. (2017). Nanocellulose in thin films, coatings, and plies for packaging applications: A review. *Bio Resources*, 12: 2143–2233.
- [6] Kargarzadeh, H.; Mariano, M.; Huang, J.; Lin, N.; Ahmad, I.; Dufresne, A.; Thomas, S. (2017). Recent developments on nanocellulose reinforced polymer nanocomposites: A review. *Polymer*, 132: 368–393.
- [7] Torres, F.G.; Arroyo, J.J.; Troncoso, O.P. (2019). Bacterial cellulose nanocomposites: An all-nano type of material. *Mater Sci. Eng. C*, 98: 1277–1293.
- [8] Lee, K.Y.; Ho, K.K.C.; Schlufter, K.; Bismarck, A. (2012). Hierarchical composites reinforced with robust short sisal fibre preforms utilising bac-

- terial cellulose as binder. *Compos Sci. Technol.*, 72: 1479–1486.
- [9] Vilarinho, F.; Silva, A.S.; Vaz, M.F.; Farinha, J.P. (2018). Nanocellulose in green food packaging. *Crit. Rev. Food Sci. Nutr.*, 58: 1526–1537.
- [10] Ferrer, A.; Pal, L.; Hubbe, M. (2017). Nanocellulose in packaging: Advances in barrier layer technologies. *Ind. Crops Prod.*, 95: 574–582.
- [11] Bonwick, G.; Bradley, E.; Lock, I.; Romero, R. (2019). Bio-based materials for use in food contact applications. In Report to the Food Standards Agency; Fera Science Ltd.: York, UK.
- [12] Fuertes, G.; Soto, I.; Carrasco, R.; Vargas, M.; Sabattin, J.; Lagos, C. (2016). Intelligent packaging systems: Sensors and nanosensors to monitor food quality and safety. *J. Sens.*, 2016: 9.
- [13] Biji, K.B.; Ravishankar, C.N.; Mohan, C.O.; Srinivasa Gopal, T.K. (2015). Smart packaging systems for food applications: A review. *J. Food Sci. Technol.*, 52: 6125–6135.
- [14] Vilela, C.; Moreirinha, C.; Domingues, E.M.; Figueiredo, F.M.L.; Almeida, A.; Freire, C.S.R. (2019). Antimicrobial and conductive nanocellulose-based films for active and intelligent food packaging. *Nanomaterials*, 9: 980.
- [15] Wang, J.; Gardner, D.J.; Stark, N.M.; Bousfield, D.W.; Tajvidi, M.; Cai, Z. (2018). Moisture and oxygen barrier properties of cellulose nanomaterial-based films. *Acs Sustain. Chem. Eng.*, 6: 49–70.
- [16] Langowski, H.C. (2017). Shelf life & quality of packaged foods. In *Food Packaging Materials-Testing & Quality Assurance*; Singh, O., Wani, A.A., Langowski, H.C., Eds.; CRC Press: Boca Raton, FL, USA, 11–66.
- [17] Paunonen, S. (2013). Strength and barrier enhancements of composites and packaging boards by nanocelluloses - A literature review. *Nord Pulp Pap Res. J.*, 28: 165–181.
- [18] Sharma, A.; Thakur, M.; Bhattacharya, M.; Mandal, T.; Goswami, S. (2019). Commercial application of cellulose nano-composites - A review. *Biotechnol. Rep.*, 21: e00316.
- [19] Garrison, T.F.; Murawski, A.; Quirino, R.L. (2016). Bio-based polymers with potential for biodegradability. *Polymers*, 8: 262.
- [20] Fabra, M.J.; Lopez-Rubio, A.; Lagaron, J.M. (2014). Nanostructured interlayers of zein to improve the barrier properties of high barrier polyhydroxyalkanoates and other polyesters. *J. Food Eng.*, 127: 1–9.
- [21] Pelissari, F.M.; Ferreira, D.C.; Louzada, L.B.; Santos, F.d.; Corrêa, A.C.; Moreira, F.K.V.; Mattoso, L.H. (2018). Starch-based edible films and coatings: An eco-friendly alternative for food packaging. In *Starches for Food Application*, 1st ed.; Clerici, M.T.P.S., Schmiele, M., Eds.; Elsevier Inc.: Amsterdam, the Netherlands.
- [22] Mangaraj, S.; Goswami, T.K.; Mahajan, P.V. (2009). Applications of plastic films for modified atmosphere packaging of fruits and vegetables: A review. *Food Eng. Rev.*, 1: 133–158.
- [23] Plackett, D. (2011). *Biopolymers: New Materials for Sustainable Films and Coatings*; John Wiley & Sons: Hoboken, NJ, USA.
- [24] Teo, P.S.; Chow, W.S. (2014). Water vapour permeability of Poly(lactic acid)/Chitosan binary and ternary blends. *KMUTNB Int. J. Appl. Sci. Technol.*, 7: 23–27.
- [25] Espino-Pérez, E.; Bras, J.; Ducruet, V.; Guinault, A.; Dufresne, A.; Domenek, S. (2013). Influence of chemical surface modification of cellulose nanowhiskers on thermal, mechanical, and barrier properties of poly(lactic) based bionanocomposites. *Eur. Polym. J.*, 49: 3144–3154.
- [26] Ruka, D.R.; Simon, G.P.; Dean, K.M. (2013). In situ modifications to bacterial cellulose with the water insoluble polymer poly-3-hydroxybutyrate. *Carbohydr. Polym.*, 92: 1717–1723.
- [27] Lee, H.; You, J.; Jin, H.J.; Kwak, H.W. (2020). Chemical and physical reinforcement behavior of dialdehyde nanocellulose in PVA composite film: A comparison of nanofiber and nanocrystal. *Carbohydr. Polym.*, 232: 115771.
- [28] Vartiainen, J.; Lahtinen, P.; Kaljunen, T.; Kunnari, V.; Peresin, M.S.; Tammelin, T. (2015). Comparison of properties between cellulose nanofibrils made from banana, sugar beet, hemp, softwood and hardwood pulps. *O Pap*, 76: 57–60.
- [29] Piringer, O.G. (2000). Permeation of gases, water vapor and volatile organic compounds. In *Plastic Packaging Material food barrier Function*,

- Mass Transport, Quality Assurance and Legislation; Piringer, O.G., Baner, A.L., Eds.; Wiley-vch: Weinheim, Germany.
- [30] Mohsen, A.H.; Ali, N.A. (2018). Mechanical, color and barrier, properties of biodegradable nanocomposites poly(lactic acid)/nanoclay. *J. Bioremediat. Biodegrad.*, 9: 6.
- [31] Díez-Pascual, A.M.; Díez-Vicente, A.L. (2014). Poly(3-hydroxybutyrate)/ZnO bionanocomposites with improved mechanical, barrier and antibacterial properties. *Int. J. Mol. Sci.*, 15: 10950–10973.
- [32] Visanko, M.; Liimatainen, H.; Sirvio, J.A.; Mikkonen, K.S.; Tenkanen, M.; Sliz, R.; Hormi, O.; Niinimäki, J. (2015). Butylamino-functionalized cellulose nanocrystal films: Barrier properties and mechanical strength. *RSC Adv.*, 5: 15140–15146.

AUTHOR (S) BIOSKETCHES

Shadab Shahsavari, Assistant Professor, Chemical Engineering Department, Varamin-Pishva Branch, Islamic Azad University, Varamin, Iran, *Email: Sh.shahsavari@srbiau.ac.ir*