2023-12

A Comprehensive Review on Metal Oxide-Nanocellulose Composites in Sustainable Active and Intelligent Food Packaging

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Recommended Citation

Kalpani Y. Perera, Dileswar Pradhan, Aideen Rafferty, Amit K. Jaiswal, Swarna Jaiswal, A comprehensive review on metal oxide nanocellulose composites in sustainable active and intelligent food packaging, Food Chemistry Advances, Volume 3, 2023, 100436, ISSN 2772-753X, DOI: 10.1016/j.focha.2023.100436

Funder: Technological University Dublin, Dublin, Ireland

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Food Chemistry Advances 3 (2023) 100436

Contents lists available at ScienceDirect

Food Chemistry Advances

journal homepage: www.elsevier.com/locate/focha

A comprehensive review on metal oxide-nanocellulose composites in sustainable active and intelligent food packaging

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ARTICLE INFO

Keywords:
Nanocellulose
Antimicrobial properties
Barrier properties
Food packaging
Metal oxide

ABSTRACT

The aim of this article is to provide an overview of the potential advantages and drawbacks of nanocellulose and metal oxide-based composites in food packaging. These materials offer improved mechanical and barrier properties, as well as antioxidant and antimicrobial benefits that extend the shelf life of food products. Nanocomposite structures protect food from various physiological factors and immobilize enzymes, while metal oxide nanoparticles provide antibacterial effects against Gram-positive and Gram-negative bacteria. However, there are concerns regarding the safety of nanoparticles and their potential migration into packaged food during processing and storage. This article explores these issues and highlights the need for further research to ensure the safe and effective use of these materials in food packaging. The successful implementation and commercialization of nanocellulose and metal oxide-based composites in food packaging could offer significant benefits to the food and beverage industry by improving the quality and shelf life of products.

Introduction

Food packaging provides protective barriers around food products to prevent interference or contamination from physical, chemical, or biological sources. Moreover, packaging can delay the deterioration of food products, preserve the benefits of processing, extend shelf-life, and manage or enhance food quality and safety. Food packaging plays a crucial role in protecting food from external microorganisms and bacteria. Maintaining the health and welfare of consumers depends highly on food safety. Additionally, packaging keeps the food protected from extreme temperatures and physical damage (Perera et al., 2021).

Novel food packaging techniques are cited as a standard reference for product safety and quality improvement through optimal packaging use. New food packaging technologies have emerged due to consumers' desire for convenient, ready-to-eat, tasty, mild, and shelf-stable products. Food packaging techniques such as active packaging, intelligent packaging, and bioactive packaging, designed to interact with the food or its surroundings and influence the consumer's health, have been crucial advancements in the field of packaging technology (Carina et al., 2021). These new techniques help keep food products fresher for longer, extend their shelf life, enhance their quality, and provide indications. Advances in novel food packaging technologies include retarding oxidation, hindering respiratory processes, preventing microbial contamination, and inhibiting moisture entrapment, as well as the use of CO_2 scavengers or emitters, aroma emitters, biosensors, ethylene scavengers, ripeness indicators, and constant release of antioxidants during storage (Majid et al., 2018).

Active packaging aims to improve the package system's performance by intentionally including subsidiary constituents in the packaging material. In active packaging, a compound is intentionally released or absorbed from the food or the packaging headspace. The effect is to extend the shelf life of products by stalling the degradative reactions of lipid oxidation, microorganism growth, and moisture loss and gain. Oxygen scavengers, ethylene scavengers, and moisture scavengers make up the largest portion of active packaging; however, the number of active packaging emitters has recently increased (Sharma et al., 2022).

Intelligent packaging enhances communication with the external world beyond mere packaging. Diagnostic and indicator functions are added to intelligent packaging. Furthermore, they may be used to achieve automation, customer engagement, and marketing. In general,

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https://doi.org/10.1016/j.focha.2023.100436
Received 24 June 2023; Received in revised form 2 September 2023; Accepted 3 September 2023
Available online 9 September 2023

intelligent packaging uses sensor and indicator technology to provide information about the product's condition, including factors such as tightness, temperature, freshness, and more. An example of this would be a display of color change to signify a leakage or the presence of salmonella contamination. There are many different ways intelligent packaging can be used to control optimal product conditions (Soltani Firouz, Mohi-Alden & Omid, 2021).

In recent years, numerous studies have focused on the application of nanomaterials to food packaging, including zinc oxide, clay, silver, carbon nanotubes, titanium dioxide, copper, and copper oxide. In addition to improving barrier/mechanical properties of food packaging, nano-sized packaging enhances the physicochemical properties of food and reduces microbial load by decreasing the Trojan horse effect, membrane function, and reactive oxygen mechanisms (Emamhadi et al., 2020). The current review plays a significant role in contributing to the existing body of knowledge within the field of active and intelligent food packaging. It contributes by focusing on a specific combination of materials, addressing current concerns, and presenting potential practical implications. The use of nanocellulose and metal oxide-based composites in food packaging represents a cutting-edge area of research and innovation. This review likely showcases new developments and concepts that can stimulate further interest and research in this evolving field.

Nanocellulose, a plant material consisting of nanoscale cellulose fibrils, is a pseudoplastic material that exhibits the property of a specific type of fluid or gel that is very thick in normal conditions. Nanocellulose has strong marketing potential in the growing food and beverage market for packaging and bulk products, with various applications. It works in preventing food spoilage and obstructing oxygen entry into food products (Perumal et al., 2022). Nanocellulose replaces polystyrene-based foams, offering a more sustainable and eco-friendlier alternative for food packaging. This innovative material not only improves food quality and extends the shelf life of products but also exhibits remarkable barrier properties against oxygen and moisture. Furthermore, nanocellulose can act as a carrier for various active substances such as antioxidants and antimicrobials, contributing to enhanced food safety and preservation.

Nanocellulose

Nanocellulose, a pseudoplastic consisting of plant material containing nanosized cellulose fibrils, belongs to a specific family of fluids, gels, which are typically thick under normal conditions. Nanocellulose has lateral dimensions ranging from 5 to 20 nanometers and longitudinal dimensions ranging from several tens of nanometers to several microns. In general, nanocellulose can be divided into three categories: cellulose nanocrystals (CNC), cellulose nanofibers (CNF), and bacterial nanocellulose (BNC). The Transmission electron microscopy (TEM) images of these nanocellulose types are depicted in Fig. 1. Due to nanocellulose's small size, the safety of foods containing nanocellulose cannot be confirmed with certainty. Therefore, research on nanocellulose as a material for food packaging is developing more swiftly than studies involving direct food addition (Lu et al., 2021).

Cellulose nanocrystals (CNC), a naturally abundant polymer, are unique nanomaterials with a diameter of 4–55 nm and a length of 90–400 nm (Zinge & Kandasubramanian, 2020). Several properties of nanomaterials have attracted significant interest, including their chemical, mechanical, optical, and rheological characteristics. Nanocrystals of cellulose can be derived from biodegradable cellulose fibers and are therefore environmentally friendly and sustainable. The nanocrystals are essentially hydrophilic by nature but can be surface functionalized to meet a variety of demanding applications, including the development of high-performance nanocomposites with hydrophobic polymer bases (Zinge & Kandasubramanian, 2020).

Various living organisms such as amoebas, fungi, bacteria, and plants can synthesize cellulose nanocrystals. In addition, it can be made from commercially available microcrystalline cellulose (MCC) by acid hydrolysis. It is common for MCC to be obtained by mechanically processing native cellulose fibers, which consist of alternating non-crystalline and crystalline regions, and CNC to be obtained by dissolving the amorphous regions. CNCs have remarkable properties, and they have been extensively investigated as potential nanofillers in polymer nanocomposites (Fang et al., 2019). When considering the essential characteristics of CNC food packaging, it has a tensile strength of 7500–7700 MPa and an elastic modulus of 130–250 GPa. It is thermally stable up to 220 °C, has a crystallinity between 60 and 90%, and a transparency greater than 90% (Ahankari et al., 2021).

Cellulose nanofiber (CNF), an advanced biomass material, is a wood-derived fiber with a diameter of 2–20 nm and a length of a few micrometers. As it is made from plant fibers, CNF has a low environmental impact in both production and disposal. The lightweight properties of its fibers allow its modulus of elasticity to be on par with aramid fibers, which are known as high-strength fibers, and to have the same thermal expansion as glass. Oxygen cannot pass through CNF because of its excellent gas barrier properties. CNF shares many characteristics with natural cellulose, such as their low density, easy biodegradability, and reproducibility, but also possess outstanding properties such as their large surface area-to-volume ratio, and excellent structural properties (Zhang et al., 2017).

They are known to be transparent, lightweight, durable, and have a unique viscosity. The fibers are nanometer-thick (a few nanometers) and are finer than the visible light wavelength (400 nm to 700 nm), therefore they are transparent as they can easily pass through light. When considering the essential characteristics of CNF food packaging, it has an elastic modulus between 20 and 60 GPa and a tensile strength between 350 and 500 MPa. It is thermally stable up to 260 °C, has a crystallinity of 40 to 70%, and a transparency of 80 to 90% (Ahankari et al., 2021).

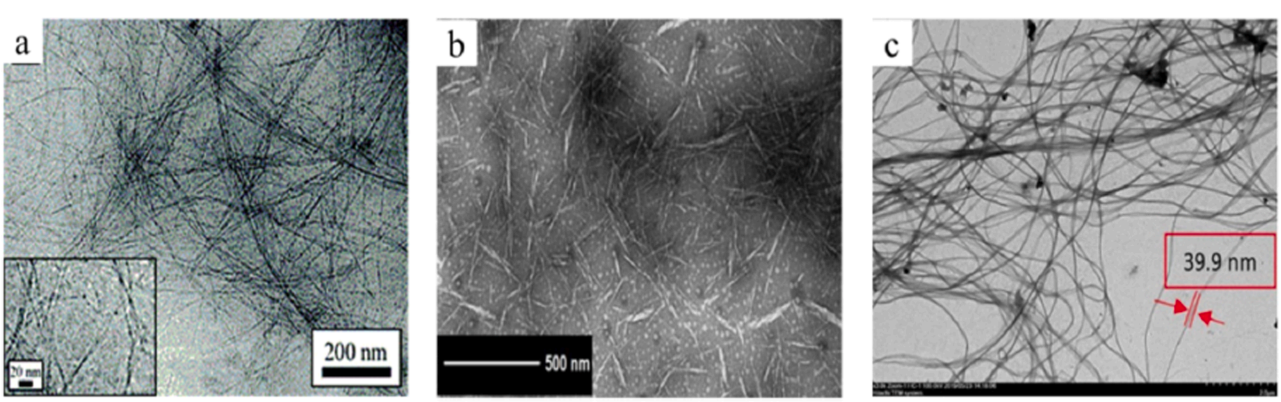

Fig. 1. Transmission electron microscopy (TEM) images of (a) cellulose nanofibers (CNF), (b) cellulose nanocrystals (CNC) and (c) bacterial nanocellulose (BNC) (Lu et al. 2021).

Bacterial nanocellulose (BNC), which shows a wide range of exceptional physical and chemical properties, has gained growing interest worldwide because of its green processing, low production costs, hydrophilic nature, and biocompatibility. The most popular BNC-producing strains come from the genus *Komagataeibacter*. There are other Gram-negative pathogenic bacteria such as *Pseudomonas* and *aerobacter* that have been previously stated to produce nanocellulose extracellularly (Abol-Fotouh et al., 2020).

Bacteria produce BNC through the simultaneous polymerization and crystallization of the polymer. Bacteria secrete glucose residues by polymerizing them into linear chains of 1,4-glucan, which are released into the extracellular space. Crystallization of the chains leads to the formation of microfibrils, and then a certain number of microfibrils entangle to form highly pure nanoribbons of 20–60 nm wide (Jabłońska et al., 2021).

In comparison to plant cellulose, BNC is a pure form of cellulose, free from lignin, pectin, and hemicellulose. In many applications, BNC's ultra-fine structure is superior to plant cellulose because it provides higher crystallinity, a greater liquid absorption capacity, a more complex polymerization, a larger specific surface area, and greater mechanical properties. In addition, the flexibility of BNC for modification makes it a superior material to plants. BNC's application sectors are always expanding, such as the bioprocessing industry, biomedical applications, and pharmaceuticals, along with wastewater treatment, electroconductive materials, packaging, and food (Abol-Fotouh et al., 2020). When discussing the essential characteristics of BNC food packaging, it has a modulus of elasticity between 15 and 138 GPa and a tensile strength of 20–2000 MPa. It is thermally stable up to 300 ℃, has a crystallinity of 50–60%, and a transparency of $>$ 90% (Ahankari et al., 2021).

Metal oxides for food packaging material

In recent years, the use of nanoparticles in the food packaging industry has significantly increased. The high reactivity, adhesion, enhanced bioavailability, bioactivity, and surface effect of the nanoparticles (NPs) aid in employing NPs in food packaging. The use of nanosized materials for packing food substances results in a longer shelf life of food products and enhanced performance (Perera, Jaiswal, & Jaiswal, 2022). Metal oxides such as Zinc oxide (ZnO), Titanium oxide (TiO_2), Copper oxide (CuO), Magnesium oxide (MgO), and Graphene oxide (GO) are generally used for food packaging materials (Perera, Jaiswal et al., 2022).

The inorganic compound ZnO finds application in several industries, such as pharmaceuticals, cosmetics, food, rubber, commodity chemicals, painting, ceramic, and glass. A potential application in food preservation involves ZnO nanoparticles with antimicrobial properties. A polymeric matrix has been coated with nanoparticles of ZnO to provide antimicrobial activity to packaging materials and improve certain packaging properties (Oun, Shankar & Rhim, 2020). Nanomaterials are widely used in the industry, of which ZnO nanoparticles are considered multifunctional because they are antimicrobially active, exhibit near UV emission, show piezoelectricity, exhibit optical transparency, and exhibit electrical conductivity. For ZnO nanoparticles to have antibacterial activity, it is crucial to understand the phase, the crystallite size, the lattice constants, the crystal orientation, and the surface defects (Roy et al., 2021).

TiO_2 NPs have been gaining increasing attention due to their importance as a food additive, food safety, and allergy prevention. In addition to being hazard-free, these nanoparticles are also cost-effective to produce. In packaging and films for active foods, they have antimicrobial and photocatalytic activity that makes them desirable. These nanoparticles have been found to have antibacterial properties in several ways. Their main mechanism is the destruction of microorganisms by hydroxyl radicals and reactive oxygen species (ROS) produced by light reactions in water (Sharma et al., 2023). These materials are a prime candidate for active food packaging due to their ultraviolet blocking and high oxidizing properties, which are a result of their photocatalytic activity. Food science is one of many fields in which metal and metal oxide nanoparticles, notably TiO_2 NPs, can play a key role (Perera, Sharma et al., 2022).

MgO NPs are NPs that are highly reactive, extremely stable, with a large surface area, are less hazardous, and are inexpensive. As a result, it primarily serves as a catalyst and an antibacterial agent. Reactive oxygen species (ROS) generation underlies the antibacterial activity of MgO NPs' mechanism of action (Zhang et al., 2020). Graphene oxide is a promising packaging material because of its distinctive qualities, which include high barrier properties, good mechanical, electric, thermal properties, a large specific surface area, high electrical conductivity, carrier mobility, antibacterial, antifungal, antioxidant, and biocompatibility (Rossa et al., 2022).

Thus, the usage of metal oxide in food packaging is able to enhance the functional properties of the packaging material by providing antimicrobial efficiency, UV barrier properties, and ethylene scavenging activity in addition to the other beneficial qualities.

Characteristics of nanocellulose – metal oxide based composite

Characterisation of nanocomposites is critical during the development stage in order to obtain a material with desirable properties for food packaging applications. Some important characteristics of nanocomposite materials are morphology, thermal properties, functional groups, mechanical properties, UV blocking efficiency, and barrier properties. The source of nanocellulose, metal oxide nanoparticles, and other substances used in the formulation of nanocomposites have the greatest influence on the properties of the developed material.

CNC-metal oxide blend

ZnO NPs have been utilized in combination with CNCs by various researchers to form nanocomposites for food packaging applications as shown in Table 1. In a research, Azizi et al. (2014) showed that the blend of CNCs and ZnO were compatible with PVA/Chitosan and diffused uniformly in the matrix of the polymer mixture. With an increase in nano-sized filler quantity from 0 to 5.0 wt%, CNCs/ZnO increased the modulus and tensile strength of the films from 395 to 932 MPa and from 55.0 to 153.2 MPa, respectively. When 1.0 wt% CNCs/ZnO was utilized for formulation, the thermal stability of the nanocomposite improved. Besides, the film demonstrated antibacterial efficacy against *Staphylococcus aureus* and *Salmonella choleraesuis*. In another research, Yu et al. (2021) utilized acetylated CNC (ACNC) with PLA and ZnO to develop a ternary nanocomposite intended to be used as active food packaging material. When ZnO content of $\leq$5 wt% was utilized for formulation, a uniform distribution of ZnO and ACNC in the PLA matrix was observed, which resulted in a homogenous film. The concentrations of Zn^{2+} that migrated from the composite material to the food-simulants were less than the established migration limit (5 mg/kg). Besides, the film demonstrated remarkable antibacterial efficiency against *Escherichia coli* and *Staphylococcus aureus*. In addition, the mechanical strength, UV blocking, water and oxygen barrier properties of the film improved with the addition of ZnO and CNC.

TiO_2 in combination with CNC for development of nanocomposite material have been reported in several research studies. In a research, El-Wakil et al. (2015) developed a nanocomposite material by combining wheat gluten with CNC and TiO_2. The water resistance property and the tensile strength improved when 0.6% of TiO_2 and 7.5% of CNC was utilized for the formulation of nanocomposite. In addition, after 2 h of exposure to UVA light illumination, the paper coated with composite film (3 layers) showed remarkable antibacterial properties, with efficacy of around 98.5%, 100%, and 100% against *Staphylococcus aureus, Escherichia coli*, and *Saccharomyces cervisiae*, respectively. In another study, Nguyen and Lee (2023) utilized apple peel extract (APE)

Table 1
CNC-metal oxide-based nanocomposites for food packaging application.

Nanocomposite formulation	Source of CNC	Application	Properties	References
Soy protein isolate - CNC - ZnO	Wheat bran	Antibacterial packaging film for pork preservation	Improvement in tensile strength, oxygen and water vapor barrier properties, water resistance ability, and thermal stability Reduction in elongation at break, surface hydrophobicity, and transmittance Inhibition growth of *Escherichia coli* and *Staphylococcus aureus* in pork samples Reduction in the total viable counts and the total volatile basic nitrogen values in a pork sample.	(Xiao et al. 2020)
Sodium alginate - CNW -CuO NPs	MCC	Antibacterial packaging film	Enhanced mechanical and barrier properties Exhibited promising antibacterial action against *Staphylococcus aureus, Escherichia coli, Salmonella* spp., *Candida albicans*, and *Trichoderma spp.* Films exhibited antioxidant activity Film prevents the microbial contamination in fresh cut pepper up to seven days.	(Saravanakumar et al. 2020)

in combination with TiO_2 and CNC to modify PVA for food packaging applications. The water vapor barrier properties and mechanical strength of PVA/CNC/TiO_2/APE improved by 36.6% and 49.9% over PVA. Besides, the composite material demonstrated exceptional UV blocking performance as well as strong antibacterial efficacy and antioxidant properties. These studies indicated that the source of CNC, the type of metal oxide, as well as the other ingredients such as different polymer matrices, active and intelligent substances determine the characteristics of the composite film.

CNF-metal oxide blend

ZnO has been utilized in combination with bacterial cellulose or bacterial nanocellulose to develop composite materials for food packaging as depicted in the studies of Table 2. In a study, Shahmohammadi Jebel and Almasi (2016) used BC and ZnO to develop a composite material for controlled release of antimicrobial substances. The water vapor permeability and moisture absorption of the composite film reduced, while the mechanical properties improved with the addition of ZnO nanoparticles. The film showed higher antibacterial efficacy against Staphylococcus aureus as compared to *Escherichia coli*. However, an improvement in antibacterial efficacy was observed with the use of ultrasound irradiation. Similarly, Wahid et al. (2019) used ZnO and BC to develop a composite film that showed photocatalytic activity. The film was able to degrade 91% of methyl orange in 2 h when exposed to UV light. Besides, the film displayed excellent UV-blocking characteristics

Table 2
CNF-metal oxide-based nanocomposites for food packaging application.

Nanocomposite formulation	Source of CNF	Application	Properties	References
Gelatine - CNF - ZnO - Selenium	Unknown	Multifunctional biodegradable packaging films	Improved physio mechanical and water resistance of CNF films Improvement in tensile strength, water vapor barrier properties, and thermal stability Inhibition growth of *Escherichia coli, Staphylococcus aureus, Listeria monocytogenes* and *Pseudomonas fluorescens* in gelatine based edible films Antioxidant effect on films	(Ahmadi et al. 2021)
CNF - TiO_2 - polyvinyl alcohol (PVA)	Wood pulp	PVA based packaging	Addition of nanocomposites enhanced the mechanical properties of PVA based films Filling the nanocomposites enabled the light barrier capacity of PVA based films No significant toxicity to cancerous and normal colon cells	(Yu et al. 2020)
CNF- sodium montmorillonite-TiO_2NPs- cinnamon essential oil-gelatin	Unknown		Water barrier properties of films significantly increased Highest antioxidant activity	(Amjadi et al. 2021)
CNF-ZnO- grapefruit seed extract	Hardwood	Active food packaging	Films are highly transparent Enhanced ultraviolet blocking and vapor barrier properties Effective antimicrobial and antioxidant activity	(Roy et al. 2021)
CNF-gelatin- ZnO	Unknown	Active packaging films	Increased young's modulus and tensile strength Decreased flexibility, water vapor permeability, and moisture absorption. High antimicrobial activity in Gram-positive bacteria than Gram-negative bacteria	(Ahmadi et al. 2020)
CNF-TiO_2 nanotubes	Unknown	active packaging application	Enhanced the UV-shielding properties. Decreasing the transparency Improved mechanical and water resistance properties. Enhanced antibacterial activity Scavenged ethylene gas produced in the headspace during tomato storage	(Riahi et al. 2022)
CNF-whey protein – TiO_2- rosemary essential oil	Unknown	Active packaging application	Reduced microbial growth, lipid oxidation, and lipolysis of the lamb meat during storage, which led to an increase in shelf life from around 6 to 15 days	(Alizadeh-Sani et al. 2020)
CNF- ZnO	Unknown	Active packaging applications	Excellent UV-light barrier properties and high transparency Improves the mechanical strength Slightly increased (~10%) the hydrophobicity while the water vapor barrier properties remain unaltered Introduced antibacterial activity toward foodborne pathogens.	(Roy, Biswas & Rhim 2022)

and antibacterial activities.

Several researchers have reported the use of TiO_2 with BC or BNC to develop nanocomposite materials. In a research, Yang et al. (2020) developed a composite material with TiO_2, polydopamine (PDA), and BC. Just within 30 min after irradiation, the film exhibited improvement in adsorption capability for methylene blue, Rhodamine B, and methyl orange. Besides, the results revealed that BC/PDA and BC/PDA/TiO_2 had a higher initial decomposition temperature as compared to BC/TiO_2, suggesting that PDA played a role in protecting BC. However, BC/PDA/TiO_2 composite had a significantly lower tensile strength as compared to BC/TiO_2 and BC/PDA film. In another study, Liu et al. (2017) utilized GO, TiO_2, and BC to develop a composite film intended for containing photocatalytic antibacterial characteristics. GO−TiO_2/BC had a significant influence on their antibacterial activities under near-UV irradiation, which was dependent on irradiation time and dose.

In several studies, the combination of CuO with BC or BNC for the development of composite films has been reported in recent times. Xie et al. (2020) developed a composite film by utilizing bacterial cellulose nanofiber and GO—CuO nanohybrid. The GO—CuO nanohybrids were evenly dispersed on the surface of the cellulose fibers, according to the SEM analysis. BC/GO—CuO films were more efficient against Gram-positive bacteria in comparison to Gram-negative bacteria. Besides, the BC/GO—CuO films exhibited better antibacterial efficacy in comparison to BC/CuO composites. In another study, Phutanon et al. (2019) used CuO and BC to develop a nanocomposite paper with antibacterial and photocatalyst characteristics. The composite showed superior antibacterial efficacy against *Staphylococcus aureus* and *Escherichia coli*. The composite was also thermally stable, showing resistance of up to 200 °C.

BNC-metal oxide blend

ZnO has been utilized in combination with bacterial cellulose (BC) or BNC to develop composite materials for food packaging as shown in Table 3. In a study, Shahmohammadi Jebel and Almasi (2016) used BC and ZnO to develop a composite material for controlled release of antimicrobial substances. The water vapor permeability and moisture absorption of the composite film reduced, while the mechanical properties improved with the addition of ZnO NPs. The film showed higher antibacterial efficacy against *Staphylococcus aureus* as compared to *Escherichia coli*. However, an improvement in antibacterial efficacy was observed with the use of ultrasound irradiation. Similarly, Wahid et al. (2019) used ZnO and BC to develop a composite film that showed photocatalytic activity. The film was able to degrade 91% of methyl orange in 2 h when exposed to UV light. Besides, the film displayed excellent UV-blocking characteristics and antibacterial activities.

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Application of nanocellulose – metal oxide as innovative packaging materials

Active packaging

In the past two decades, the food industry has extensively studied metal-based nanoparticles and their specialized derivatives for their promising antimicrobial properties and their potential to enhance food quality. An example of this would be, several metal-based nanoparticles such as ZnO, MgO, CuO, TiO_2, silicon, calcium oxide, silver oxide and gold have previously been examined for demonstrating the antimicrobial activity and also how they potentially reduce biofouling. Despite their high specificity and high surface area to volume ratio, metal oxide and metallic nanoparticles show strong antimicrobial activity, making them a promising antimicrobial food packaging material. There has been a great deal of interest in metallic nanoparticles for their antibacterial properties, which are used in food packaging, pharmaceutical production, and biotechnology applications (Galstyan et al., 2018). Nanocellulose have excellent mechanical properties however lack the antimicrobial properties to make them efficient active packaging films. Thus, metal oxide nanoparticles with antimicrobial/ and antioxidant activity has been added to nanocellulose to incorporate antimicrobial properties to the packaging materials (Oun et al., 2020). Nanocellulose

Table 3
BNC (bacterial nanocellulose)-metal oxide-based nanocomposites for food packaging application.

Nanocomposite formulation	Source of BNC	Application	Properties	References
Chitosan, BNC, and ZnO-paper sheets		Multifunctional mixture for coatings in food packaging applications	Increased antibacterial against *Escherichia coli* Enhanced mechanical properties	(Jabłońska et al. 2021)
BC - PPy - ZnO		Antioxidative food active packaging and smart packaging	Decreased the water vapor permittivity and total soluble matter percentage. Enhanced Antioxidant activity	(Pirsa et al. 2018)
BC - PPy-ZnO		Intelligent and active packaging	Decrease the growth of microbial load in chicken thigh and could control the pH increasing. Increase the shelf life and stabilize rheological properties of chicken thigh by increasing of Antioxidant and antimicrobial activity as active packaging	(Pirsa & Shamusi 2019)
BC- ZnO NPs and propolis extracts	*Gluconacetobacter xylinum*	Active packaging	*Escherichia coli* has proved to be highly resistant when compared to *Bacillus subtilis*, and *Candida albicans*.	(Mocanu et al. 2019)

acts as a reinforcement matrix that absorbs or releases active agents in order to extend the shelf life of food products (Silva et al., 2020). Additionally, nanocellulose functions as a matrix to help with the regulated release of metal oxides, which aids the prolonged antimicrobial activity of the food packaging films.

To extend the shelf life of fruits, ethylene-scavenging antimicrobial films were created by Riahi et al. (2022) using CNF and combining TiO_2, TiO_2 nanotubes (TNT), and Cu_2O-modified TNT as shown in Fig. 1. The pure CNF showed no antimicrobial activity and the addition of TiO_2 NPs enhanced the antimicrobial activity of the packaging films immensely. The greatest antibacterial activity was seen in the TNT-Cu_2O-added CNF film, which were followed by CNF/TNT and CNF/TiO_2 films. Where *Listeria monocytogenes* and *Escherichia coli* growth were completely inhibited by the CNF/TNT-Cu_2O film after 6 and 12 h of incubation. To test the films' ability to scavenge ethylene, tomato packing made of CNF-based films was used. The tomatoes' discolouration, softening, and weight loss were postponed by the CNF/TNT-Cu_2O film because it scavenged ethylene gas produced in the headspace during storage. Under visible light, the TNT-Cu_2O film demonstrated effective photocatalytic activity, leading to a potent bactericidal effect and ethylene scavenging activity (Riahi et al., 2022).

Additionally, gelatin-based nanocomposite films containing varying concentrations of CNF (2.5, 5%, and 7.5% w/w) and ZnO NPs as an antibacterial agent (1%, 3%, 5%, and 7% w/w) were produced using the casting process by Ahmadi et al. (2020). According to the disk diffusion assay, the 5% ZnO NPs films exhibited the strongest antibacterial activity, with significantly higher inhibition in Gram-positive bacteria (*Staphylococcus aureus* inhibition zone 10.44 $\pm$ 0.44 mm) than Gram-negative bacteria (*Pseudomonas fluorescens* inhibition zone 9.75 $\pm$ 0.11 mm). *Staphylococcus aureus* and *Pseudomonas fluorescens* inoculations on chicken fillets were used to measure the antibacterial effectiveness of the film as a food packaging material. The chicken fillets' bacterial load was significantly reduced as a result of the active packaging film, especially the Gram-positive strain Ahmadi et al. (2020).

Further, active packaging films has also been developed with antimicrobial and antioxidant actions by using both antimicrobial and antioxidant agents; ZnO nanorods and grapefruit seed extract into a CNF matrix (Roy et al., 2021). When the film's antimicrobial activity was detected, the ZnO-blended film displayed delayed antibacterial activity and inhibited *Escherichia coli*'s complete growth. Low antibacterial activity was shown against *Listeria monocytogenes* by the ZnO-blended film. Due to the difference in the cell wall structures of *Escherichia coli* and *Listeria monocytogenes*, ZnO had a stronger antibacterial effect on *Escherichia coli*. Although they were more efficient against *Listeria monocytogenes* than *Escherichia coli*, both the grapefruit seed extract-incorporated film and the grapefruit seed extract/ZnO-added films shown strong antibacterial activity against both test pathogens. According to the findings, grapefruit seed extract is more effective than Gram-negative bacteria at inhibiting Gram-positive bacteria. As a result, the films that were made with both ZnO and grape seed extract displayed strong antibacterial properties. When considering the antioxidant activities of all of the films using the 2,2-diphenyl-1-picrylhydrazyl (DPPH) radical scavenging efficiency, the films containing grape seed extract displayed higher activity than the films with ZnO. Grape seed extract is added to the nanocomposite film to give it antioxidant capabilities, which should protect foods that are susceptible to oxidation and increase their shelf life.

Active food packaging materials has also been produced in films with BNC which has no antimicrobial efficiency by the addition of metal oxide NPs. For instance (Mocanu et al., 2019) developed a antimicrobial food packaging film with the incorporation of ZnO NPs or/and propolis extracts into a BC matrix. In the films with only ZnO NPs, and BC, the antimicrobial effect was noticed only in the case of *Bacillus subtilis* when inhibition zones were observed. In contrast to Gram-negative bacteria (*Escherichia coli*), which are not responsive to this synergistic impact, propolis extracts and ZnO had a synergistic effect on yeast (*Candida albicans*) and Gram-positive bacteria (*Bacillus subtilis*). According to the experimental findings, Gram-negative and eukaryotic cells were unaffected by BC-ZnO. Thus, as per this study it is concluded that by only utilizing ZnO NPs and BC antimicrobial films cannot be developed which is effective to both Gram-positive and Gram-negative bacteria but rather other antimicrobial agents are needed (Mocanu et al., 2019). The research on nanocellulose and metal oxide nanoparticles in food packaging that was previously mentioned indicates that CNC, CNF, or BNC do not generally have antibacterial activity or other useful active properties. The addition of nanocellulose improved the mechanical and barrier properties of the packaging matrix, and the additional metal oxide nanoparticles demonstrated antibacterial properties against both Gram-positive and Gram-negative microorganisms.

Intelligent packaging

The state of packed food or its surrounding environment is monitored through intelligent packaging materials and products. It monitors a product's condition, safety, and location during shipping, storage, retail sales, and usage. The future of food packaging, according to experts, will be the next generation of intelligent packaging. In general, the three core technologies used in intelligent packaging systems are data carriers, indicators, and sensors (Perera et al., 2021). As a result, materials for food packaging based on metal oxide and nanocellulose have recently been developed.

Pirsa et al. (2018) developed an intelligent food packaging material by synthesising novel films based on BC, BC modified by polypyrrole (PPy), and a PPy–zinc oxide nanocomposite (BC–PPy–ZnO) by using a soft polymerization method. As per the findings, BC-PPy and BC-PPy-ZnO films might be utilised in food packaging to estimate food spoilage and the period of storage through the analysis of the film conductivity. These parameters included food odor, temperature, pressure, the presence of volatile compounds, and storage time (Pirsa, Shamusi & Kia, 2018).

In an additional previous study, BNC was assembled by chemical synthesis method. The active and smart packaging of fresh chop chicken meat was developed with bacterial cellulose containing zinc oxide and PPy. This study was designed to investigate the antimicrobial, antioxidant, and electrical conductivity characteristics of film when it is packaged. It is possible to estimate the time and temperature of the chicken thigh meat holding by scanning and observing changes in the electrical resistance of the packing film. This allows us to reasonably accurately predict the food's expiration date using this intelligent package (Pirsa & Shamusi, 2019).

Further studies were conducted on polypyrrole based intelligent packaging material, where BNC/PPy/TiO_2-Ag (BC/PPy/TiO_2-Ag) nanocomposite film was developed to detect and measure the growth of 5 pathogenic bacteria, which increase growth with food spoilage (Ghasemi et al., 2020). Changes in electrical resistance were observed in response to bacteria including *Escherichia coli, Staphylococcus aureus*, *Staphylococcus epidermidis*, and *Aeromonas hydrophyla*. High electrical conductivity and sensitivity were observed in sensors with a high concentration of Pyrrole, as indicated by the results. The sensors responded better to Gram-negative than Gram-positive microorganisms. Thus, intelligent packaging material can detect the microbial growth during food spoilage and inform the consumers.

Since neither nanocellulose nor metal oxides are sensitive to pH changes, temperature changes, or other atmospheric changes, which are essential in intelligent food packaging, it is imperative to include other bioactive compounds when designing an intelligent packaging material using nanocellulose and metal oxides. When it comes to the aforementioned studies, the well-known conducting polymer PPy has a number of intriguing characteristics, such as strong conductivity, quick and easy polymerization, environmental degradability, and heat stability. Thus, it is used in analytical chemistry as a sensor and biosensor. As a result, it's the ideal distinguishing feature for smart food packaging.

Further, studies on BNC and metal oxide based food packaging materials have been conducted by Sukhavattanakul and Manuspiya (2020). Where they developed a H_2S gas sensor using BNC-based surface-loaded silver nanoparticles (AgNPs) and alginate-molybdenum trioxide nanoparticles (MoO_3NPs). The hybrid film of BNC–AgNPs/alginate–MoO_3NPs detected H_2S gas due to a change in Mo oxidation state and reduction by atomic hydrogen intercalating on the film. The film color changed from transparent light greyish-white to opaque dark brown black due to the dissociation of hydrogen molecule from the reaction of AgNPs and H_2S gas on oxide surfaces, which decomposes into $MoO_2^{+1/2}$ and H_2O by the fast decrease of Mo oxidation state from Mo^{6+} to Mo^{5+}, slowly forms Mo^{4+} states, and forms a very low-intensity MoS_2^{4+} at 1 h and 24 h. The exposure duration increasingly intensifies color. Thus, this intelligent packaging film can sense the increase of H_2S gas during food spoilage. However, since both of these topics are still being researched, the state of the art for intelligent food packaging materials based on nanocellulose and metal oxide is quite limited.

Application of nanocellulose – metal oxide in packaging of food products

Meat products

Many various considerations must be made in order to design packaging materials and procedures that are appropriate for meat products. The technique used to package meat has an impact on both the product's safety and shelf life. Not just for health reasons, but also to ensure higher sustainability of contemporary agriculture and to increase food availability for a growing population, meat packing materials and methods must be optimised (Schumann & Schmid, 2018). Due to microbial growth and chemical degradation, notably through oxidation, fresh meat is extremely vulnerable to spoiling during storage. Innovative packing materials are therefore required to slow down these processes. To extend the shelf life and safety of meat products while lowering food waste, there has been a significant of interest in the development of active and intelligent food packaging materials (Alizadeh-Sani, Mohammadian & McClements, 2020). Therefore, there is a lot of interest in the use of sustainable, active, or intelligent food packaging materials, such as food based on nanocellulose and metal oxides.

In the study of Alizadeh-Sani et al. (2020), a biopolymer based packaging material that contained cellulose nanofiber- whey protein matrix, TiO_2 NPs, and rosemary essential oil was created utilizing a casting approach. The microbiological count, chemical stability (pH, lipid oxidation, lipolysis), and optical features of this packaging were examined to determine its capacity to prevent lamb meat from chemical and microbial decomposition after 15 days of chilled storage (4 °C). The shelf life of the lamb meat was extended from about 6 to 15 days thanks to the active packaging, which dramatically decreased microbial growth, lipid oxidation, and lipolysis during storage. In this case, the integration of TiO_2 into cellulose nanofiber/whey protein matrix has a number of possible benefits, including higher heat resistance, decreased permeability, greater tensile strength, and enhanced antibacterial activity (Alizadeh-Sani et al., 2020).

Another study investigated the preservation of microbial and sensory quality of lamb meat during the storage at 4 - 1 °C using a whey protein isolate, CNF nanocomposite film containing 1.0% (w/w) titanium dioxide and 2.0% (w/v) rosemary essential oil. Over the course of a 15-day period, lamb meat was evaluated for microbial and sensory properties in two groups (control and treatment). A significant reduction in the bacterial counts of the treatment group was observed following the use of nanocomposite films. Furthermore, microbial and sensory tests confirmed that nanocomposite films significantly increased the shelf life of treated meat (15 days vs. 6 days) over control meat. It was concluded from the results of this study that the edible nanocomposite films effectively preserved the microbial and sensory qualities of lamb meat. Thus, this application is recommended in red meat particularly (Alizadeh Sani, Ehsani & Hashemi, 2017).

According to the afore mentioned research, nanocellulose and metal oxide-based packaging materials can be utilised to extend the shelf life of packaged meat. This occurs as a result of the increased antibacterial and barrier capabilities of packing films. However, additional research is required in this area to evaluate the migration of NPs into food products and the associated safety concerns.

Fruits and vegetables

Fruits and vegetables, which are rich in vitamins, minerals, dietary fiber, polyphenols, and other nutrients, play a crucial role in offering fresh, wholesome, and nutritious food to people worldwide. An estimated 40–50% of fruits and vegetables are lost annually due to post-harvest physiological metabolism (e.g., respiration and transpiration), unsuitable storage conditions (e.g., wrong gas environment, temperature, and humidity), and deterioration caused by microbial proliferation. Effective food packaging is vital for protecting food from becoming contaminated by chemicals, physical damage, shock, dust, temperature, light, humidity, and bacteria, thereby extending its shelf life and decreasing food waste (He et al., 2023). Thus, a successful food packaging material containing nanocellulose and metal oxides is advantageous for the fruit and vegetable packaging industry.

Saravanakumar et al. (2020) designed an antibacterial polymeric film with sodium alginate and cellulose nano whisker (CNW) embedded with CuO NPs for fresh cut pepper as shown in Fig. 2. Here, CNW improves the barrier qualities of the film to prevent food from absorbing moisture. CuO NPs protect food against microbial infestation. The film constructed of CNW (0.5%), SA (3%)-CuNPs (5 mM) displayed excellent antibacterial action against a variety of pathogens, as evidenced by a larger zone of inhibition against *Staphylococcus aureus* (27.49 0.91 mm), *Escherichia coli* (12.12 0.58 mm), *Salmonella* sp. (25.21 1.05 mm), *Candida albicans* (23.35 0.45 mm). In terms of DPPH and ABTS scavenging activity, the film also demonstrated remarkable antioxidant activity. As indicated in Fig. 2, the optimal composition of the film prevents microbiological contamination in freshly cut pepper for up to 7 days. The results indicated that the colonization of microbiological pollutants, including total bacteria, total fungus, total *Listeria spp.*, and total *Salmonella*, was considerably inhibited in fresh-cut pepper coated with CNW (0.5%) -SA (3%)- CuO NPs (5 mM) films as compared to the untreated group (Saravanakumar et al., 2020).

Riahi et al. (2022) developed CNF-based ethylene-scavenging antimicrobial films including several types of TiO_2 NPs to increase the shelf life of fruits and vegetables. CNF-based films were made by combining TiO_2, TNTs, and Cu_2O-modified TNT (TNT-Cu_2O). The addition of nanoparticles considerably improved the UV-blocking characteristics of the film at the expense of a minor reduction in transparency. Depending on the nanoparticle type, the incorporation of nanofillers increased the film's mechanical and water resistance capabilities. CNF/TNTCu_2O exhibited the most potent antibacterial action against *Escherichia coli* and *Listeria monocytogenes*, followed by CNF/TNT and CNF/TiO_2 films. To increase shelf life and postpone post-harvest fruit quality loss, such as color change, weight loss, and firmness, CNF-based films were utilised to package tomatoes. Particularly, the film containing TNT-Cu_2O exhibited effective photocatalytic activity under visible light, resulting in a potent bactericidal impact and ethylene scavenging activity. Thus, CNF-based films with enhanced characteristics and effective antibacterial and ethylene-scavenging activities can be utilised to extend the shelf life of harvested vegetables and fruits (Riahi et al., 2022) .

Regulatory aspects of nanomaterials used in food packaging

Numerous inorganic and bio-based nanomaterials, possessing unique chemical and physical attributes, have been investigated as a potential

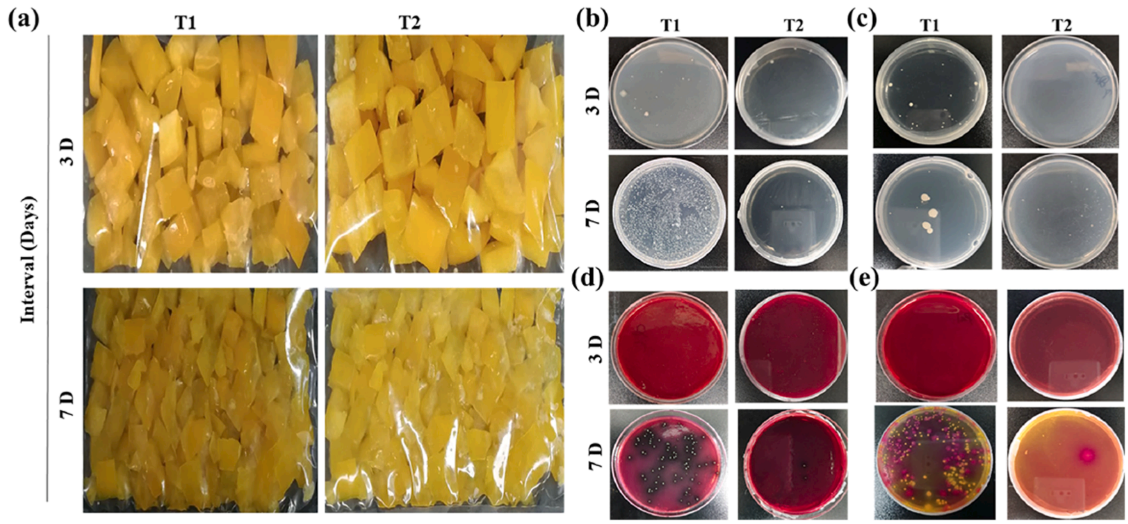

Fig. 2. CNW (0.5%) -SA (3%)- CuO NPs antimicrobial packaging film for fresh cur pepper applications. (a) Visual observations, T1 is the uncoated control group, and T2 is the coated group. (b) total bacterial counts (c) total fungal counts, (d) total *Listeria* spp. Counts, and (e) total Salmonella spp. (Saravanakumar et al. 2020).

additives to polymers for improving their performance (Huang, Li & Zhou, 2015). However, the synthesis and application of nanomaterials might pose a number of risks that could have an impact on both human health and the environment (Ungureanu et al., 2022). Therefore, the safety aspects of nanomaterials, which are a critical element of nanocomposites, has raised public concern when they are utilized in food packaging given the likelihood of their migration into the packaged food during processing and storage (Jafarzadeh et al., 2021; Pradhan, Jaiswal, & Jaiswal, 2022). Hence, it is crucial to thoroughly investigate the migration activities, toxicity and various others safety aspects of nanomaterials intended to be used in the development of food packaging material. Besides, setting up the safety standards for the specific nanomaterials will help the regulatory agencies around the world to form relevant legislation which would in turn play a key role in the commercialization of innovative nanomaterial based composite material in food industry.

A number of federal entities in the USA are in charge of products based on nanotechnology. The Food and Drug Administration (FDA) established the Nanotechnology Task Force in 2006 with the goal of creating regulatory frameworks for nanotechnology-based products that will assure their efficacy and safety while simultaneously promoting favorable technical innovation (Huang et al., 2015; Silvestre, Duraccio & Cimmino, 2011). Nano-enabled food products are not subject to any specific rules in Canada, although they are nonetheless governed by the Public Health Agency of Canada (PHAC) and the Canadian Food Inspection Agency (CFIA). South African Nanotechnology Initiative (SANi) in South Africa regulates the safety and risk assessment of nanoparticles (Basavegowda, Mandal & Baek, 2020).

European Food Safety Authority (EFSA) approval is necessary in the European Union before an ingredient can be used in food contact materials (FCM) and is included to a positive list (EFSA Panel on Food Contact Materials et al., 2021). This process has been documented in "Regulation (EC) No 1935/2004 of the European Parliament and of the Council of 27 October 2004 on materials and articles intended to come into contact with food and repea ling Directives 80/590/EEC and 89/109/EEC" (Borrell Fontelles & Nicolai, 2004). For instance, if employed as an additive at up to 0.025% w/w in polymers such polyolefins, polyesters, and styrenics that don't swell when in contact with aqueous foods and food simulants, silver nanoparticles do not pose a safety risk to consumers (EFSA Panel on Food Contact Materials et al., 2021). The utilization of active and intelligent substances are covered under "Commission Regulation (EC) No 450/2009 of 29 May 2009 on active and intelligent materials and articles intended to come into contact with food" (Communities,2009). The Commission Regulation (EU) No 10/2011 of 14 January 2011 is focused on plastic materials and articles intended to come into contact with food (Commission, 2011).

A formal definition of industrial nanomaterials and recommendations for evaluating their effects on human health have been developed by Australia's National Industrial Chemicals Notification and Assessment Scheme (NICNAS). In addition, the Food Standards Australia New Zealand (FSANZ) has investigated the competency of the food regulatory framework in Australia and New Zealand on risks to human health owing to nanotechnologies under the current regulations (Sothornvit, 2019).

Conclusion and future perspectives

The use of nanocellulose and metal-oxide nanoparticles in food packaging offers improved mechanical, barrier, and antibacterial capabilities and is an eco-friendly, biodegradable alternative to plastic food packaging materials. Several studies have been conducted on active and intelligent food packaging materials based on nanocellulose and metal oxide nanoparticles, applicable to various food categories. These materials enhance the physical, chemical, and biological properties of food packaging materials. However, there is a need for more studies on CNC, intelligent packaging, migration, and toxicity assessment of these packaging materials. To fully understand the impact of these materials on the food packaging industry, the environment, and human health and safety, further research is required. Future perspectives in this field include the development of advanced nanocomposite materials with multifunctional properties, such as self-healing or self-cleaning capabilities. The integration of nanomaterials with sensor technologies may also enable the creation of smart packaging systems that can monitor food quality and safety throughout the supply chain. Collaboration between academia, industry, and regulatory bodies will be crucial in addressing safety concerns and establishing standardized protocols for the evaluation and commercialization of these novel packaging materials. Additionally, the development of cost-effective and scalable production methods for nanocellulose and metal-oxide nanoparticles will help facilitate their widespread adoption in the food packaging sector.

CRediT authorship contribution statement

Kalpani Y. Perera: Investigation, Writing – original draft. **Dileswar Pradhan:** Investigation, Writing – original draft. **Aideen Rafferty:** Investigation, Writing – original draft. **Amit K. Jaiswal:**

Conceptualization, Supervision, Writing – review & editing. **Swarna Jaiswal:** Conceptualization, Supervision, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

Acknowledgment

The authors would like to acknowledge the funding from Technological University Dublin, Dublin, Ireland under the Research Scholar Award-2021.

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