International Food Research Journal 32(2): 356 - 378 (April 2025)
Journal homepage: *http://www.ifrj.upm.edu.my*

Review

Carbon-based nanofillers for biodegradable food packaging applications: A review

[1]*Terzioglu, P., [1]Gazioglu Ruzgar, D. and [2]Bakkour, Y.

[1]*Department of Polymer Materials Engineering, Faculty of Engineering and Natural Sciences, Bursa Technical University, Bursa, Türkiye*
[2]*Laboratory of NMR, Central Laboratory, College of Applied Medical Sciences, King Khalid University, Abha 61421, Saudi Arabia*

Article history

Received:
17 May 2024
Received in revised form:
6 January 2025
Accepted:
8 January 2025

Keywords

biodegradability,
bio-nanocomposites,
carbon,
food packaging,
sustainable packaging,
nanofillers

Abstract

Initially, non-degradable plastics have been employed to fulfil the demand for food packaging. However, increasing environmental concerns associated with conventional packaging materials have prompted a search for sustainable alternatives. Biodegradable polymer-based materials are emerging as significant options for packaging applications that align with the principles of sustainable development. Nevertheless, these materials frequently exhibit limitations in their properties when applied to food packaging. In response to these challenges, the development of bio-nanocomposites offers a novel approach to enhancing the properties of biodegradable materials. Incorporating nanosized fillers into biodegradable polymer matrices can facilitate the production of bio-nanocomposite food packaging. Carbon-based nanofillers have become a prominent strategy for generating nanocomposites with improved functionalities among the various methodologies. Noteworthy carbon nanomaterials, such as carbon dots, carbon nanotubes, graphene, graphene oxide, and graphitic carbon nitride have been identified as effective agents for enhancing the performance characteristics of biodegradable packaging. The present review aims to elucidate recent advancements concerning the impact of carbon-based nanomaterials on the barrier, functional, mechanical, thermal, visual, and biodegradability properties of polymers, particularly in the context of biodegradable food packaging applications, while also providing insights into future directions in this field.

DOI
https://doi.org/10.47836/ifrj.32.2.04

Introduction

Achieving food safety and security is among the important challenges faced by the global community. Food packaging is an indispensable component of the food supply chain (Trajkovska Petkoska *et al*., 2021). Food packaging is a multidisciplinary approach encompassing art, knowledge, and technology to enclose, protect, and deliver foods to end users at a reasonable price. Communication and traceability are other important functions of food packaging (Amin *et al*., 2021). As a result of practical and economic considerations, plastics are the most commonly preferred material for rigid and flexible food packaging (Terzioğlu *et al*., 2021; Priyadarshi *et al*., 2022). The global food packaging industry currently surpasses $300 billion annually and is projected to expand at a compound annual growth rate (CAGR) of approximately 5% (Priyadarshi *et al*., 2022).

The changing eating habits, market needs, and increasing concerns about minimising food waste will shape the growth of future packaging (Kumari *et al*., 2022). The most commonly used conventional food packaging materials are glass, paper, plastic, steel, aluminium, and different alloys (Trajkovska Petkoska *et al*., 2021). Plastics are favoured for their exceptional barrier and mechanical properties, widespread availability, and affordability. However, the use of petrochemical-based plastics like polyethylene, polyethylene terephthalate, polyvinyl chloride, polystyrene, and polypropylene, in this sector, is among the outstanding problems due to their inappropriate disposal and non-biodegradable nature, as well as insufficient recyclability (Asgher *et al*., 2020). Most of the packages are single-use materials

*Corresponding author.
Email: pinar.terzioglu@btu.edu.tr

with high-rate usage in a short period of time, resulting in various ecological problems (Weligama Thuppahige and Karim, 2022). Biodegradable food packaging materials are considered a promising and suitable alternative to replace traditional synthetic packaging materials and a means of reducing municipal solid waste accumulation. Generally, biopolymers are chosen to develop biodegradable packaging, while degradable synthetic counterparts are used. However, the commercialisation of biopolymer-based packaging materials is restricted depending on three main factors: (i) processing difficulty, (ii) cost of production, and (iii) performance characteristics (Kumari *et al*., 2022).

Preparing composite biodegradable packaging has been devoted to overcoming the problem of insufficient performance. Recent studies have focused on the incorporation of nanofillers into polymer matrix materials. Nanoscale fillers are used in packaging in various cases, which could be related to their efficient roles attributed to their unique properties (Mahmud *et al*., 2022). The nanoparticles incorporated in biodegradable packaging can gain new functionalities or enhance the packaging features. Nanoscale particles are used to enhance the bioactivity and chemical, electrical, optical, thermal, and mechanical characteristics of the composite materials (Jagadeesh *et al*., 2021).

However, bio-nanocomposites overall performance is mainly based on the properties of the filler (aspect ratio, particle size, surface area, compatibility with polymer matrix, *etc*.), as well as the dispersion of the filler in the polymer matrix (Wu *et al*., 2022). The type of nanofiller can be organic or inorganic. Cellulose, lignin, and chitosan are this field's most studied organic fillers. Different inorganic nanofillers can be used for this purpose, like carbon-based nanomaterials, nano clays, metals/metal oxides, and other non-metallic nanomaterials (Priyadarshi *et al*., 2022).

Among various nanofillers, the exploration of carbon-based ones to develop new functional packaging materials is of great scientific and technological interest. Carbon dots (C-dots), carbon nanotubes (CNTs), graphene, graphene oxide (GO), and graphitic carbon nitride (g-C_3N_4) are the widely used carbon-oriented nanofillers with superior functional and structural properties. In most cases, carbon-based nanofillers can perform as both an active ingredient and reinforcing agent (Figure 1). The addition of carbon nanofillers will provide intelligent/innovative characteristics to the food packaging materials as well (Moradi *et al*., 2023; Han *et al*., 2024; Riahi *et al*., 2024a).

Therefore, the present review seeks to provide a comprehensive overview of recent advancements in research concerning the evaluation of carbon-based nanofillers for the formulation of biodegradable polymer-based food packaging materials. Additionally, it identifies various factors that influence the properties and behaviours of carbon-based nanomaterials within polymeric matrices.

Applications of carbon-based nanofillers in biodegradable food packaging materials

Carbon dots

Carbon dots (C-dots) are zero-dimensional photoactive carbonaceous nanomaterials (Figure 2), including carbon quantum dots (CQ-dots), carbon nanodots (C-nanodots), carbonised polymer dots (CP-dots), graphene quantum dots (GQ-dots), and the carbon core of carbonised polymer dots (Deka *et al*., 2022). C-dots have gained tremendous interest in scientific circles due to their unique properties and easy preparation from simple, low-cost, and environmentally friendly raw materials (Riahi *et al*., 2022). The recent progress on the potential evaluation of C-dots in food packaging applications is summarised in Table 1.

The synthesis of C-dots can be achieved through two primary techniques namely top-down and bottom-up. The top-down technique involves breaking down larger carbon sources like carbon fibres, carbon nanotubes, graphite, and coal through cleavage and exfoliation. Several techniques are employed in this method, including electrochemical synthesis, hydrothermal synthesis, oxidative cleavage, solvothermal synthesis, and methods that utilise microwave or ultrasonic assistance. In contrast, the bottom-up approach focuses on constructing carbon dots from smaller organic molecules. Precursors such as saccharides (*e.g.* glucose and fructose) and organic acids (*e.g.* citric and ascorbic acids) can be subjected to pyrolysis, and these can also be combined with other molecules to introduce heteroatoms like nitrogen, boron, and sulphur for doping purposes (Ozyurt *et al*., 2023).

Carbon dots are composed of quasi-spherical nanoparticles with ≤ 10 nm particle size (Du *et al*., 2020). Carbon dots have extraordinary properties that make them promising functional nanofillers for smart

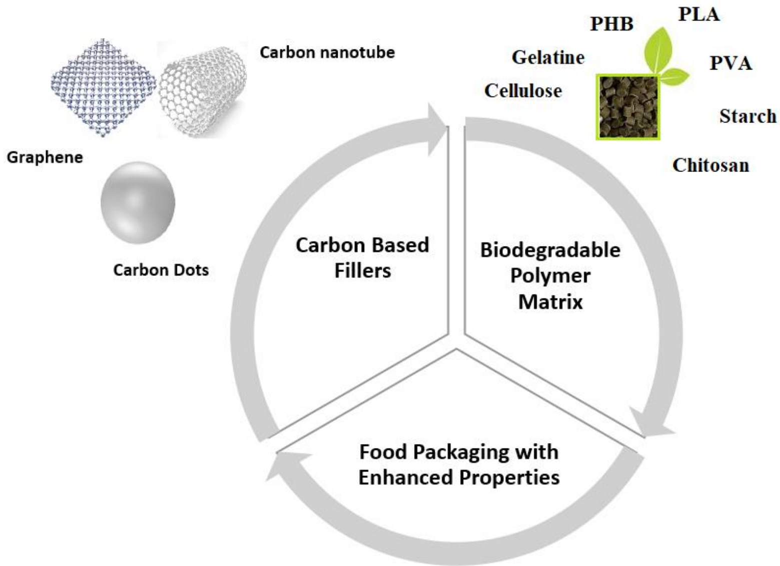

Figure 1. Schematic diagram for carbon-based-nanofiller-loaded biodegradable food packaging.

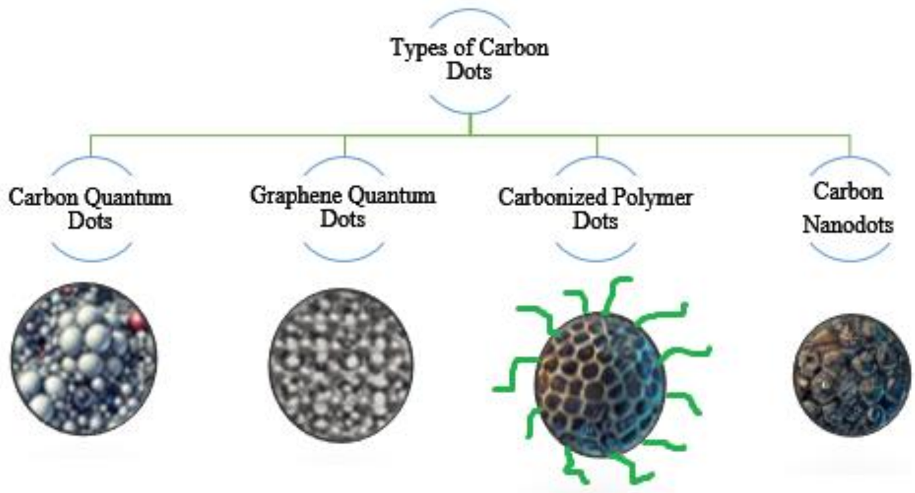

Figure 2. Classification of carbon dots (C-dots).

Table 1. Biodegradable polymer-based carbon-dot-incorporated packaging materials.

Nanomaterial	Other additive	Polymer matrix	Packaging application	Main findings	Tested food	Reference
Carbon nanodot	Anthocyanin extracted from *Clitoria ternatea* flower	Starch	Active smart packaging	Enhanced mechanical, barrier, thermal, and antioxidant properties	Pork	Koshy *et al*. (2021)
Carbon dots	Anthocyanins	Fish gelatine	Intelligent packaging compatible with smartphone technology	Reduced crystallinity; Enhanced water tolerance, barrier, mechanical, and thermal properties	Skinless chicken breast	Kilic *et al*. (2022)
Carbon nanodot	-	Poly(vinyl alcohol)	Active smart packaging	Fluorescence of the film was sensitive to pH and Al^{3+} ions; Enhanced antioxidant capacity	-	Zhang *et al*. (2020)
Carbon dots	-	Gelatine	Active packaging	Highly transparent; Increased UV blocking properties; High antioxidant activity; Moderate bactericidal effect on *L. monocytogenes*	-	Min *et al*. (2022)
Carbon dots	Grapefruit seed extract	Gelatine/ poly(vinyl alcohol)	Active packaging	Good UV protection; Decreased mechanical properties; High antioxidant and antibacterial activities	Pork	Min *et al*. (2023)
Carbon dots	-	Gelatine/ chitosan	Intelligent packaging	Enhanced antibacterial, antioxidant and UV shielding activities; Anti-counterfeiting and pH-responsive properties	Fish meat	Fu *et al*. (2022)
Carbon dots	-	Bacterial nanocellulose	Active and intelligent packaging	Fluorescence appearance under ultraviolet; Improved flexibility; UV protection	-	Kousheh *et al*. (2020)
Carbon dots	-	Bacterial nanocellulose	Active packaging	UV protection; Antibacterial activity	-	Salimi *et al*. (2021)
Carbon dots	-	PVA	UV protective packaging	100% UV-C and UV-B, and 20 - 60% from UV-A light protection	Grape	Patil *et al*. (2020)
Carbon quantum dots	-	Carboxymethyl cellulose	Active coating	Increased the tensile strength and elastic modulus; High antioxidant and antibacterial activities	Lemon	Riahi *et al*. (2022)
Carbon quantum dots	-	Chitosan/ gelatine	Antifungal coating and active packaging	Antioxidant and antibacterial activities	Avocado	Ezati *et al*. (2022c)
Nitrogen-functionalised carbon dots	-	Carboxymethylcellul ose/agar	Active packaging	High UV protection; antioxidant and antibacterial activities	-	Tammina and Rhim (2023)
Nitrogen-functionalised carbon dots	-	Chitosan	Active and intelligent packaging	Enhanced antioxidant, antibacterial, and UV shielding properties	Pork	Lin *et al*. (2022)
Nitrogen-functionalised carbon dots	-	Cellulose	Antifungal coating and active packaging	High UV protection; Increased water vapour permeability; High antioxidant, antibacterial, and antifungal activities	Tangerine, strawberry	Ezati *et al*. (2022b)
Sulfur-functionalised carbon dots	-	Pectin/ Gelatine	Active packaging	High UV protection; Increased mechanical properties; Strong antioxidant activity; Antimicrobial activity against *E. coli* and *L. monocytogenes*	-	Ezati *et al*. (2022d)

fluorescent sensing packaging to identify food toxins, and pH-sensitive smart packaging materials to monitor food freshness. These properties include photoluminescence, photobleaching resistance, non-toxic/low toxicity, excellent biocompatibility, easy surface modification, chemical stability, good water solubility, and pH-sensing ability (Du *et al*., 2020; Kang *et al*., 2020; Deka *et al*., 2022). Carbon dots can also be used in active packaging applications due to their antibacterial and antioxidant properties (Du *et al*., 2020). The impact of CQ-dots with a 7.8 nm size on the carboxymethyl cellulose films was recently investigated by Riahi *et al.* (2022). The hydrophilic nature and small size of CQ-dots resulted in good compatibility and homogenous dispersion of CQ-dots in the polymer matrix, which is supported by scanning electron microscopy (SEM) results. After the addition of CQ-dots, the colour of neat carboxymethyl cellulose films changed from colourless to light yellow. The colour of films varied depending on the number of CQ-dots (1 - 5 wt% of polymer). The addition of brown-coloured CQ-dots somewhat decreased the brightness (L-value) of the carboxymethyl cellulose film, while increasing the film's redness (a-value) and yellowness (b-value). The developed biocomposite film demonstrated good transparency and UV barrier, and enhanced mechanical and water barrier properties. In addition, the films showed bioactivity in terms of antioxidant, antibacterial, and antifungal activities. Then, lemon fruit was coated with the developed edible coating. The coating extended the shelf life of fresh fruit by preventing mould growth even after 21 days of storage. Moreover, CQ-dots incorporated chitosan/gelatine bioactive functional film was tested for avocado packaging, and the results exhibited that the film extended the fruit shelf life by more than 14 days with its anti-mould capacity (Ezati *et al*., 2022c). Chitosan/gelatine films containing C-dots developed by Sul *et al*. (2023) showed substantial antibacterial activity against the common foodborne pathogenic bacteria, *Listeria monocytogenes*, completely inhibiting bacterial growth within six hours of exposure. The findings of Sul *et al*. (2023) indicated that the neat chitosan/gelatine film demonstrated antibacterial activity against both *L. monocytogenes* and *Escherichia coli*, attributed to the antimicrobial properties of chitosan. In addition, Gram-positive bacteria, specifically *L. monocytogenes*, exhibited greater sensitivity to neat C-dots and chitosan/gelatine/C-dots films compared to Gram-negative bacteria like *E. coli*. This difference is primarily attributed to variations in cell wall composition and the binding affinity of C-dots to Gram-positive bacteria. Gram-positive bacteria, which lack an outer membrane, are particularly vulnerable to the harmful effects of reactive oxygen species (ROS) that can penetrate and damage their cell walls. Although the precise mechanism behind the antibacterial action of C-dots remains unclear, it was proposed that the small size of C-dots allows for rapid penetration into bacterial cells, leading to alterations in cellular metabolism, which are believed to contribute to their antibacterial properties. Therefore, the improved antibacterial activity of the chitosan/gelatine/C-dots film can be primarily attributed to the antimicrobial action of the C-dots, which is linked to their gradual release from the films. The chitosan/gelatine/C-dots active film was found to be suitable for packaging of minced beef effectively prevented the discoloration of it with antibacterial, antioxidative, and UV protection functionalities. Fluorescent C-dots were also considered as functional fillers for photodynamic inactivation technology for antibacterial packaging due to their unique properties to facilitate the generation of ROS when exposed to light (Wen *et al*., 2023). Wen *et al*. (2023) reported that when exposed to a 405 nm light source, the chitosan/CQ-dots film demonstrated the capability to generate significant amounts of ROS. Within a 40-min period, the chitosan/CQ-dots films destroyed *Staphylococcus aureus* and *E. coli*, respectively, and slow down the spoilage rate of pork.

Some of the studies revealed that the addition of C-dots significantly enhanced the UV-light blocking properties of the biocomposite films without reducing transparency (Patil *et al*., 2020; Min *et al*., 2022; Tammina and Rhim, 2023). This will bring the advantage for the use of C-dots as bioactive functional nanofillers to fabricate transparent films with UV protection that could be very helpful for food packaging applications. However, in some cases, UV-barrier food packaging materials could be developed with lower transparency with the addition of C-dots (Khan *et al*., 2023b; Khoshkalampour *et al*., 2023; Sul *et al*., 2023).

The potential use of carbon nanodots in smart packaging systems has been investigated by different researchers (Koshy *et al*., 2021). Carbon nanodots were used to reinforce anthocyanin-incorporated starch film to develop pH-sensitive intelligent packaging materials with sufficient properties (Koshy *et al.*, 2021). The incorporation of C-nanodots decreased the water sensitivity and improved the mechanical strength of the composite. As the amount of time during which pork was stored increased, the C-nanodots and anthocyanin-loaded film showed visible colour shifts from pink/purple to green. In addition, synergistic effect of C-dots and anthocyanin led to the enhancement of the antioxidant capacity, barrier, and thermal properties. Therefore, the biocomposite film was suggested as a low-cost visual indicator to indicate freshness of packed pork. In another study, it was shown that C-dots have the potential to be used in gelatine/chitosan-based multifunctional films as an indicator of fish meat freshness, due to having fluorescence brightness change under the UV light as the pH of the packaged fish varied, as well as to decrease lipid oxidation degree of fish meat during storage (Fu *et al.*, 2022).

In a recent work, C-dots were evaluated as a chemical crosslinker to fabricate colorimetric fish gelatine films including red cabbage anthocyanin extract (Kilic *et al.*, 2022). Carboxyl-functionalised carbon dots were selected as chemical crosslinker due to their surface functional groups, while UV irradiation served as the physical crosslinker. The properties of fish gelatine films were enhanced *via* incorporation of carbon dots under UV irradiation. Specifically, the crystallinity of the films decreased, indicating successful crosslinking between the carboxyl groups of carbon dots and the amino acid residues of gelatine. This can be explained by the fact that the crosslinking of carbon dots with the protein may impede the reassembly of the gelatine triple helix structure, leading to diminished intermolecular interactions among the protein chains. This interference contributes to the gradual reduction of crystallinity during the film formation process. Additionally, the water resistance, mechanical strength, thermal stability, and barrier properties of the fabricated films were all improved. The film was used as a freshness indicator for skinless chicken breast. A custom-designed smartphone application capable of image processing was developed and used to predict quantitative estimation of food spoilage in real-time monitoring. The platform for tracking food freshness should greatly reduce food waste globally and the spread of foodborne illnesses.

Modified Atmosphere Packaging (MAP) stands out as a globally renowned preservation technique employed to uphold the microbial and physicochemical integrity of food products. Fan *et al.* (2019) showed that carbon dots-incorporated chitosan nanocomposite coating could be used for MAP of fresh-cut cucumber. The findings suggested that applying a coating of C-dots and chitosan proved beneficial in restraining the growth of microorganisms and enhancing the overall storage quality of fresh-cut cucumbers. The antibacterial activity of coatings improved as the C-dots concentration increased from 0 to 4.5%. Additionally, the results of the total number of colonies, as well as mould and yeast levels on coated fresh-cut cucumber were significantly lower ($p < 0.05$) for the C-dots-containing coatings compared to the neat chitosan coating. C-dots contain abundant functional groups therefore enhanced the antimicrobial activity of the coating. MAP is also preferred for preserving fruits; however, its applicability to all fruits is limited due to substantial variations in storage conditions among different fruit types (Su *et al.*, 2023). Therefore, Su *et al.* (2023) formulated an environmentally friendly, sustainable, and cost-efficient multifunctional bio-nanocomposite coating utilising egg albumin, chitosan, and C-dots. While the incorporation of C-dots decreased the tensile strength of the coating, it concurrently enhanced the flexibility of the coating. The coating was suggested as a safe approach for preserving the quality and minimising postharvest losses of fresh lychee in terms of vitamin C content, appearance colour, and total sugar.

Functionalised carbon dots

The C-dots generally contain a large number of carboxyl, amino, hydroxyl, and other functional groups that determine their surface properties. These functional groups can be changed by surface passivating C-dots with several agents (Roy *et al.*, 2022).

Currently, the functionalisation of carbon dots with non-metallic elements is at the forefront of research (Ezati *et al.*, 2022b; 2022d; Lin *et al.*, 2022; Tammina and Rhim, 2023). Nitrogen-functionalised (Ezati *et al.*, 2022b; Lin *et al.*, 2022; Tammina and Rhim, 2023), nitrogen/phosphorus- functionalised (Khan *et al.*, 2023a), and sulphur-functionalised (Ezati *et al.*, 2022d) C-dots were evaluated in biodegradable polymer systems for packaging applications.

Using nitrogen, a plentiful and safe element to functionalise C-dots, offers significant benefits for packaging applications due to its ability to enhance the physicochemical properties of C-dots.

Bioactivities such as antibacterial and antioxidant properties, low toxicity, and high dispersibility in aqueous solutions can all be obtained through nitrogen functionalisation of C-dots (Mao *et al*., 2023). Similarly, sulphur-functionalisation provides improvement in the bioactivity behaviour of C-dots (Roy *et al*., 2022). Sulphur-functionalisation C-dots can also provide UV protection properties without affecting the transparency of films (Ezati *et al*., 2022d; Hong *et al*., 2024).

The studies showed that the addition of functionalised C-dots enhanced the bioactivity and photoluminescence properties of composites, and therefore they are likely to be used for UV-protective (Lin *et al*., 2022; Chen *et al*., 2023; Hong *et al*., 2024) and intelligent packaging applications (Lin *et al*., 2022). The functionalised C-dots-incorporated biofilms have been suggested as coating for fresh fruit (Ezati *et al*., 2022b), anti-browning package for fresh-cut apple (Hong *et al*., 2024), and antibacterial package for pork (Lin *et al*., 2022; Hong *et al*., 2024) and meat products (Khan *et al*., 2023a). Additionally, nitrogen-doped C-dots containing chitosan films are potential candidates as freshness indicators of meat based on pH-mediated fluorescent sensing (Lin *et al*., 2022). However, the mechanical properties of the composite films were decreased more significantly with the addition of nitrogen-functionalised C-dots compared to pure C-dots (Tammina and Rhim, 2023). The reduction of tensile strength with the addition of nitrogen-functionalised C-dots can be explained by the fact that the repulsive force between particles increased and the intermolecular interaction decreased.

On the other hand, recent studies have shown that doping C-dots with metal and metal oxide nanoparticles could be a good strategy to develop stable C-dots by making hydrogen bonds with the surface functional groups, such as COOH and OH groups, and preventing aggregation (Khan *et al*., 2024; Riahi *et al*., 2024b). Silver (Ag), zinc (Zn), titanium dioxide (TiO_2), and zinc oxide (ZnO) have been investigated to improve the properties of C-dots for multifunctional packaging applications. Khan *et al*. (2024) investigated the role of ZnO-doped C-dots with a natural colorant (anthocyanin) in carrageenan to obtain pH-responsive, intelligent, and active films for monitoring condition and extending the shelf life of shrimps. ZnO-doped C-dots made it possible to obtain films that have excellent antioxidant activity, antibacterial activity against *L. monocytogenes and E. coli*, and UV-blocking ability. The developed films extended the shelf life of shrimps. The findings of Riahi *et al*. (2024b) also supported these results, demonstrating that Zn-doped-C-dot-incorporated cellulose nanofibre/pullulan films were effective when applied to chicken breast and tofu. According to Riahi *et al*. (2024b), the incorporation of Zn-doped C-dots enhanced the interfacial compatibility of composite films. In addition, Ananthi *et al*. (2023) developed Ag-doped C-dots/agar-based sustainable and biodegradable packaging films with good antibacterial activity against both Gram-positive and Gram-negative foodborne pathogens. In another recent investigation (Riahi *et al*., 2024a), TiO_2 was selected as a doping agent for C-dots. Incorporating TiO_2 into the C-dots core chemical structure can introduce new functions and expand their applications. The TiO_2-doped C-dots with anthocyanin were used to prepare a carrageenan-based active and intelligent packaging films for preservation and on-site monitoring of shrimp freshness. The TiO_2-doped C-dots exhibited antioxidant activities against DPPH and ABTS radicals, and antibacterial activity against four different bacteria including *E.coli* and *Salmonella enterica* (Gram-negative), and *L. monocytogenes* and *S. aureus* (Gram-positive). The active films with UV-barrier, antioxidant, and antibacterial properties turned from pink to yellow/brown for fresh shrimp and inedible shrimp, respectively. Overall, the studies demonstrated that metal- and metal oxide-doped C-dots produced multifunctional biodegradable films to detect and preserve seafood and meat quality in real-time.

Carbon nanotube

Carbon nanotubes (CNTs) are tube-like nanomaterials that can be classified as single-walled carbon nanotubes (SWCNTs), double-walled carbon nanotubes (DWCNTs), and multi-walled carbon nanotubes (MWCNTs) based on the number of carbon layers (Figure 3) (Anzar *et al*., 2020). CNTs with excellent properties can improve the bioactivity, barrier, mechanical, and thermal features, as well as the functionality of food packaging materials (Wen *et al*., 2022). However, the performance and functional characteristics of CNTs incorporated biocomposite films are significantly influenced by the interfacial compatibility between the polymer matrix and CNTs (Shahbazi *et al*., 2017). It has been established that the utilisation of CNTs comes with certain challenges

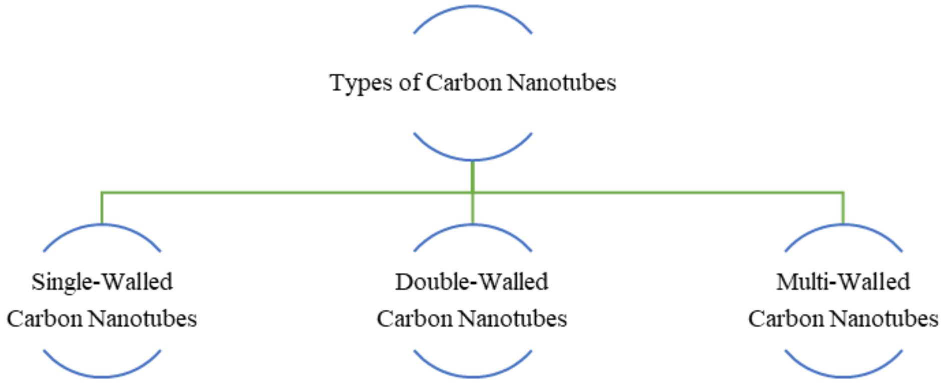

Figure 3. Classification of carbon nanotubes based on number of carbon layers.

related to their processibility. This difficulty arises from their high surface energy and van der Waals interactions, making them challenging to disperse effectively in conventional solvents (Shahbazi *et al.*, 2017). Therefore, some of the studies focused on the functionalisation of CNTs (Shahbazi *et al.*, 2017; Antolin-Ceron *et al.*, 2023). The incorporation of dendritic structures onto CNTs has the potential to enhance interactions and physical properties within blends (Antolin-Ceron *et al.*, 2023). Natural products such as tannic and citric acids can serve as effective crosslinking agents in MWCNTs/chitosan blends, facilitating various tunable interactions including hydrogen bonding, hydrophobic interactions, and charge interactions (Lu *et al.*, 2020). These interactions are crucial components in the formation of supramolecular self-assembled materials.

There is a scarcity of research in the evaluation of CNTs in biocomposite packaging development, but there have been some advancements. There is research (Cui *et al.*, 2020; Pattanshetti *et al.*, 2020; Wen *et al.*, 2022; Ibrahim *et al.*, 2023) about the effect of CNTs on the properties of biodegradable polymer-based packaging films. Owing to their hollow structure and high specific surface area, CNTs can carry a variety of compounds with diverse molecular weight sizes (Cui *et al.*, 2020). For instance, controlled-release biodegradable polymer-based nanocomposites were developed by Cui *et al.* (2020) using poly (ε-caprolactone) (PCL), polylactic acid (PLA), cinnamaldehyde, and CNTs by solvent evaporation method for active food packaging applications. It was reported that the incorporation of CNT and cinnamaldehyde would not remarkably influence the barrier properties of PLA-based film. However, it would significantly improve the anti-UV ability of the film. Compared with the neat PLA film, PLA-based film incorporated with CNTs showed higher tensile strength and elastic modulus. This situation could be due to the large surface area and high aspect ratio of CNTs, the good dispersion of CNTs in the PLA matrix, and the good compatibility between CNTs and polymer matrix. The CNTs and cinnamaldehyde-containing PLA film exhibited effectiveness against both *E. coli* and *S. aureus* for 21 days. As CNTs are rather flexible, they can interact with cell membranes, penetrate into various pathogens and foodborne microorganisms, leading to cell death (Pattanshetti *et al.*, 2020; Ibrahim *et al.*, 2023). The authors suggested the developed nanocomposite films be evaluated to enhance microbial safety and the quality of perishable foods (Cui *et al.*, 2020).

An interesting study was carried out by coating garlic microparticles on the gelatine/MWCNT nanocomposite films to prevent the interaction of food with the CNTs (Pattanshetti *et al.*, 2020). Since the migration of materials from packaging including chemicals, fillers, plasticisers, or stabilisers into food is a significant concern for food safety and quality, coating the nanocomposite film could be a solution to address the toxicity issues of MWCNTs caused by migration. Results demonstrated that the incorporation of MWCNTs to gelatine enhanced the resistance of films against oil and water. Further, bio-nanocomposite films showed antibacterial activity due to the presence of garlic particles and MWCNTs, suggesting that these developed materials could be assessed for potential applications in food packaging. Additionally, coating treatment of composites with garlic particles not only improved their bioactivity, but also brought a solution to the problem based on the toxicity of MWCNTs related to migration. In another study, Wen *et al*. (2022) reported the evaluation of MWCNTs for active food packaging applications. The researchers prepared zinc oxide-doped MWCNTs loaded poly(vinyl alcohol)(PVA)-based films and applied them to food matrices, including fresh vegetables and chicken meat. The tensile strength of composite films was found to be better than neat PVA film, due to the reinforcing effect of MWCNTs on polymer matrix, and also formation of a strong bond between the hydroxyl groups on the ZnO surface and those in PVA. The antibacterial activity, hydrophobicity, thermal stability, and water vapour transmission rate of the prepared nanocomposite films were found to be better than neat PVA film due to synergistic effect of zinc oxide and MWCNTs. The films were suggested to be used in fruits, vegetables, meat preservation, and frozen foods packaging applications. Similarly, Ibrahim *et al*. (2023) enhanced the multifunctionality of chitosan/polyethylene oxide (PEO)-based blend films with the incorporation of MWCNTs/graphene oxide. After graphene oxide doping and after co-doping with 0.4% MWCNTs, the transmittance of the blend decreased from 76 - 91% to 21 - 49% and 42 - 72%, respectively. The incorporation of fillers resulted in increased thermal stability as well as significant antimicrobial activity against foodborne pathogenic and spoilage microorganisms, which positively reflects the antimicrobial effect of both graphene oxide and MWCNTs. Dadfar and Kavoosi (2015) also reported that MWCNTs improved the antimicrobial activity and tensile strength of carboxymethyl cellulose films, as well as decreased the water solubility, water swelling, and water uptake of the packaging. In another study, Liu *et al*. (2019) utilized CNTs to improve properties of PLA/chitosan-based electrospun fibres to show their ability for strawberry preservation. These antimicrobial fibres had the capability to slow down the physiological changes in strawberries, thereby extending their shelf life. The results showed that composite fibres consisting of PLA/chitosan/CNTs hold significant potential for applications in the preservation of fruits and vegetables.

The enhancement of properties of starch film through the incorporation of MWCNTs and hydroxylated MWCNTs was investigated by Shahbazi *et al*. (2017). The study demonstrated that nanofillers can effectively address the shortcomings associated with the suboptimal physico-mechanical and structural characteristics of starch. The modified starch film exhibited significant improvements in hydrophobicity, water resistance, water barrier, and mechanical properties. Overall, CNTs seem to be potential fillers for food packaging and food preservation applications.

Graphene, graphene oxide, and reduced graphene oxide

Graphene, graphene oxide (GO), and reduced graphene oxide (rGO) have their own unique advantages that can be evaluated in various fields. A single sheet of carbon atoms organised in a hexagonal pattern is called graphene, which is considered as the thinnest as well as strongest two-dimensional (2D) nanomaterial in the world (Urade *et al*., 2023; Kiranakumar *et al*., 2024). Graphene is a valuable reinforcing filler in composites due to its 2D structure, which also gives it excellent Young's modulus, high thermal conductivity, and chemical stability (Yang *et al*., 2024).

GO is a material composed of single atomic layers that combine carbon, hydrogen, and oxygen molecules (Kiranakumar *et al*., 2024). The overall size of the unit cell in GO is comparable to that of graphene, which is similar to the honeycomb lattice. Graphene is similarly conserved in GO; however GO is more complex structure due to the presence of oxygen atoms, leading to the creation of a number of active oxygen-containing groups for GO, such as hydroxyl and epoxy groups on the plane, and carbonyl and carboxyl groups at the edges. Its high negative charge density and hydrophilicity, which have attracted a lot of attention, are attributed to these functional groups. Compared to graphene, GO has better dispersibility which makes its use advantageous to develop polymer composites (Yang *et al*., 2024).

To restore graphene-like characteristics, rGO is utilised to deoxidise GO and fix its conjugated structure. Regarding its comparatively advantageous conductivity, rGO is one of the graphene derivatives which has similar characteristics to both graphene and GO (Xiong *et al*., 2024). Due to its structural resemblance to graphene, rGO is thought to possess several of graphene's most notable qualities, including excellent electrical conductivity, lofty mechanical characteristics, great thermal stability, and transparency (Nguyen *et al*., 2023).

In recent years, graphene-based materials (GBMs) are very important fillers for the bioplastic reinforcement area to improve some final properties of nanocomposite (Barra *et al*., 2020; Kaur *et al*., 2021; Omerović *et al*., 2021; Vasseghian *et al*., 2022). GO and graphene have similar properties, but the production of GO is easier and cheaper. Also, GO allows a wide variety of functionalisation with the help of high oxygen content (Ryu *et al*., 2020; Kaur *et al*., 2021). Strong and flexible GBMs have some characteristics such as large specific surface area, high Young's modulus, high thermal and electrical conductivity, biocompatibility, better mechanical and gas barrier properties, and they can easily be dissolvable in the biopolymer matrix (Carvalho and Conte Junior, 2020; Vasseghian *et al*., 2022). Biodegradable polymer-incorporated GBMs have great application area in the active packaging sector as antimicrobials and antioxidants (Rossa *et al*., 2022; Vieira *et al*., 2022).

A recent work showed that low quantity of graphene nanoplatelets addition to poly(3-hydroxybutyrate) had high impact on mechanical, thermal, UV-VIS, conductivity, gas, and vapour barrier properties without changing the inherent polymeric matrix properties (Papadopoulou *et al*., 2019). Mechanical improvements of the composite structure are not being limited by the nanostructure morphology (Mergen *et al*., 2020). However, in some studies, a slight reduction in elongation at break was noticed in the composites based on chitosan/rGO (Barra *et al*., 2019) and chitosan/GO (Terzioglu *et al*., 2020). This drawback can be solved by mixing GO with other nanofillers (*e.g.* ZnO) (Terzioglu *et al*., 2020). The use of graphene derivatives such as GO can help increase the structure's thermal stability. The degradation temperature of cellulose nanocrystal/GO nanohybrids-incorporated poly(3-hydroxybutyrate-*co*-3-hydroxyvalerate) was postponed up to 20°C compared to bare biofilm (Li *et al*., 2019). The reason behind this situation is the creation of effective heat and gas barrier properties (Wang *et al*., 2019). Moreover, GO-incorporated poly(3-hydroxybutyrate-*co*-3-hydroxyvalerate) (Li *et al*., 2019) and alginate (Weng *et al*., 2019) composites showed better gas, water vapour, and UV light barrier properties. All these improvements in the barrier characteristics are the result of the high surface area of the graphene-based nanofillers (Vasseghian *et al*., 2022).

The interaction of bio-based polymers and their composites with water is a crucial parameter that affects their suitability for packaging applications, especially for foods. Literature investigations have shown that surface hydrophobicity of biocomposites was improved by adding graphene derivatives to the polymer composites. For example, by combining PLA/starch matrix with increasing concentrations up to 0.6 wt% of GO-grafted maleic anhydride, the surface contact angle was changed from 67.10° to 81.15° (Wang *et al*., 2019). Therefore, improved water resistance can expand their use in packaging for items that require moisture barriers. Other research work about chitosan/GO composites investigated the influence of GO oxidation degree on the water solubility of films. GO samples with varying oxidation levels were prepared using a modified Hummers' method by adjusting the graphite-to-potassium permanganate ratios to 1:2, 1:4, 1:6, and 1:8. The neat chitosan film exhibited the highest water solubility (37.75%). The solubility of oxidised-GO-loaded films was changed from 22.41% to 27.00% when the ratio of graphite to potassium permanganate was increased from 1:2 to 1:8 (Han Lyn *et al*., 2019).

To improve the water vapour and oxygen barrier properties, UV-blocking, and aging performance, Li *et al*. (2021) used the aligned rGO nanosheets for biodegradable polybutylene adipate terephthalate (PBAT) films. The results of this study showed that the addition of 0.48 vol% graphene content to the composite films provided 80% decrease in water vapour permeability and 99% decrease in oxygen permeability. That is why aligned graphene nanosheets-combined biodegradable polymers are important for sustainable packaging materials (Li *et al*., 2021). Another study about the synthesis and characterisation of GO nanoplates and GO-polyethylene glycol (PEG) hybrid structures showed that GO-PEG-based nanocomposite structures had the highest thermal, mechanical, and antibacterial properties. Further, the MTT assay showed that the GO-PEG nanohybrid improved cell biocompatibility compared to GO and decreased the toxicity of GO at high concentrations. Due to their more desirable properties for the packaging industry, modified GO-containing nanocomposites (PVA/CS/GO-PEG) may thus be ideal options (Mohammadi and Babaei, 2022).

Charoensri *et al*. (2021) focused on the incorporation of ZnO/rGO into poly(butylene adipate-co-terephthalate) biodegradable film to enhance antimicrobial and physical properties. According to the functional and antibacterial activity characterisations, nanocomposite structure can improve the mechanical and thermal properties. Also, nanocomposite biodegradable films exhibited superior antibacterial activity by preventing the growth of bacteria (Charoensri *et al*., 2021). The antibacterial activity of composites films can be explained by the fact that the surface of ZnO induces bacterial cell damage by generating ROS, such as superoxide anions (O_2^-) and hydroxyl radicals (OH•). Additionally, the addition of rGO onto the surface of ZnO lowers the resistance to electron transfer from light generation. This can be attributed to the reduced charge potential. The more negative zeta potential of ZnO observed with the increased rGO concentration highlights another key factor contributing to the enhanced antimicrobial activity of the nanocomposite films. In another study, the synthesis of polyhydroxyalkanoate (PHA) and polylactic acid (PLA)-graphene nanocomposite films were achieved by the addition of different graphene content to improve mechanical properties and the conductivity (Gürler and Torğut, 2022).

Furthermore, starch/chitosan matrix-based bio-nanocomposites enriched with GO nanoparticles showed high barrier properties against various microorganisms for food industry applications. The study also confirms that both chitosan and GO exhibit bactericidal activity against Gram-negative and Gram-positive bacteria. Having good optical properties makes these composite structures suitable to use as active elements of packaging (Krystyjan *et al*., 2022).

Gu *et al*. (2021) conducted research on a novel chitosan-based nanocomposite film resembling a sandwich structure which was loaded with rGO-immobilised nanosilvers. In this work, authors used corn stalk as green reductant and GO as a template, and a simple sandwich-like chitosan nanocomposite film was produced for the controlled release of AgNPs. The results showed that the nanocomposite film continued AgNPs release for up to 14 days. The films demonstrated effective inhibition of bacteria, including *E. coli* and *S. aureus*, with no observed toxicity to cells. Therefore, the nanocomposite film can be used as a safe packaging material to improve the shelf life of products (Gu *et al*., 2021). Graphene-reinforced potato starch composite films were studied by simple addition of graphene to glycerol-plasticised potato starch films. When graphene concentration was increased up to 1%, potato starch/graphene composite films showed better electrical conductivity. Also, improved tensile strength was achieved when the light transmittance decreased, resulting in more opaque films (Gürler and Torğut, 2021). In another research, biodegradable membranes was produced for food packaging by using poly(vinyl alcohol) (PVA), starch, GO, and rGO to enhance the antibacterial and mechanical properties of the materials. For that reason, different concentrations of GO and rGO were incorporated into the membranes. Both GO- and rGO-incorporated membranes exhibited antibacterial activity. Among the PVA/starch membranes, the membrane incorporated with 20 mg of rGO showed an excellent tensile strength and great antibacterial activity against *E. coli* and methicillin-resistant *S. aureus* (Iqbal *et al*., 2022). The recent progress on the potential evaluation of graphene-based materials in food packaging applications is summarised in Table 2.

Graphitic carbon nitride

A newly discovered polymeric nanoparticle that is a metal-free semiconductor is called graphitic carbon nitride (g-C_3N_4) (Mousavi *et al*., 2021; Zhang *et al*., 2021). It is known as the most stable allotrope of carbon nitrides under ambient conditions (Gaddam *et al*., 2020). It has unique stability in terms of resistance to chemicals and heat endurance owing to its 2D aromatic tri-s-triazine structure (Dong *et al*., 2014; Gaddam *et al*., 2020). Moreover, it is easily synthesised, cost-effective, and non-toxic (Mousavi *et al*., 2021). It has strong visible-light absorption. Therefore, when g-C_3N_4 is irradiated with visible light, it generates electron-hole pairs, which produce ROS that lead to bacterial cell death (Sun *et al*., 2017; Yu *et al*., 2021). Hence, g-C_3N_4 could be a good candidate for antibacterial applications due to its photocatalytic activity. Furthermore, due to its high mechanical strength and the said properties, g-C_3N_4 is preferred as a nanofiller to improve the features of polymer-based materials (Gaddam *et al*., 2020).

Recently, researchers focused on the evaluation of g-C_3N_4 as a filler for food packaging materials (Mousavi *et al*., 2021; Ni *et al*., 2021a; 2021b; 2022). The novel antimicrobial bio-nanocomposite films were prepared by the incorporation of g-C_3N_4 to chitosan by solution casting method to apply in the preservation of tangerine fruits (Ni *et al*., 2021a). The addition of g-C_3N_4 greatly enhanced the hydrophobic, mechanical,

Table 2. Biodegradable polymer-based graphene-based materials incorporated packaging materials.

Nanomaterial	Other additive	Polymer matrix	Packaging application	Main findings	Tested food	Reference
Graphene oxide	-	Chitosan	Active packaging	Good electrical and mechanical properties; Great antioxidant activity	-	Barra *et al.* (2019)
Graphene oxide	Zinc oxide	Chitosan	Potential food packaging	Enhanced mechanical properties and thermal stability	-	Terzioglu *et al.* (2020)
Graphene oxide	Cellulose nanocrystals	Poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV)	Active packaging	Highest thermal stability and mechanical properties; Excellent barrier properties; Good antibacterial activity	-	Li *et al.* (2019)
Graphene oxide	-	Alginate	Robust packaging materials	Excellent mechanical properties, thermal stability, and moisture barrier performance	-	Weng *et al.* (2019)
Graphene nanoplatelets	-	Polyhydroxybutyrate (PHB)	UV protective food packaging	Improved thermal stability, tensile strength, reduction in oxygen, and water vapour permeability	Potato chips, milk product	Manikandan *et al.* (2020)
Graphene oxide grafted with maleic anhydride and subsequently modified by dodecyl amine	-	Poly (lactide)-starch	UV protective food packaging	Improvements in thermal stability; Enhanced surface hydrophobicity, UV-shielding capacity, and aging resistance properties	-	Wang *et al.* (2019)
Graphene oxide	-	Chitosan	UV protective food packaging	Improvement in mechanical strength; Lower UV light transmission	-	Han Lyn *et al.* (2019)
Graphene oxide and glycol-decorated graphene oxide	Glycol	Polyvinyl alcohol/ chitosan	food-drug packaging	Nanohybrid structure enhanced biocompatibility, and improved thermal, mechanical and antibacterial properties	-	Mohammadi and Babaei (2022)
Graphene oxide	-	Starch/ chitosan	Active packaging	High barrier properties against many microorganisms and water vapour; High biodegradability; Good optical properties	Meat	Krystyjan *et al.* (2022)
Graphene oxide	Silver nanoparticles	Chitosan	Active packaging	Durable antibacterial effect and good antibacterial activity against *E. coli* and *S. aureus*; No toxicity to cells	-	Gu *et al.* (2021)
Graphene oxide and reduced graphene oxide	-	Poly (vinyl alcohol)/starch	Active packaging	More stable structure and excellent barrier against water vapour and oxygen transmission	Gooseberries	Iqbal *et al.* (2022)

and thermal properties of neat chitosan films. It was determined that the antibacterial activity of g-C_3N_4-incorporated chitosan bio-nanocomposite films against *E. coli* and *S. aureus* was remarkably improved under visible light irradiation for 15 min. The addition of 30% g-C_3N_4 of the chitosan weight was found to be optimal for antibacterial activity, hydrophobicity and mechanical properties in terms of tensile force. Furthermore, the g-C_3N_4-incorporated chitosan films exhibited better performance than commercial PE and neat chitosan film groups for the storage of tangerines. Ni *et al*. (2022) developed chitosan/hollow g-C_3N_4/curcumin biocomposite films. The nanosized hollow g-C_3N_4 sphere was used to improve thermal stability and achieve slow release for curcumin. The antibacterial activity of chitosan/hollow g-C_3N_4/curcumin films was found to be lower than the chitosan/curcumin films, which may be related to slow release of curcumin from the chitosan/hollow g-C_3N_4/curcumin films. The results demonstrated that the prepared chitosan/hollow g-C_3N_4/curcumin film could effectively preserve bananas for ten days with the smallest weight loss. The latest studies focused on the synergistic effect of g-C_3N_4 with active agents like copper (Cu) (Mousavi *et al*., 2021) and molybdenum disulphide (MoS_2) (Ni *et al*., 2021b). Mousavi *et al*. (2021) initially deposited Cu nanoparticles on g-C_3N_4 and then used it as an antibacterial filler in a starch/alginate biopolymer matrix for active packaging applications. The results showed that the incorporation of Cu5%-g-C_3N_4 could significantly improve the antibacterial, water vapour permeability, and mechanical properties of starch/NaAlg film. Graphite carbon nitride nanosheets/MoS_2 nanodots were introduced to konjac glucomannan films as a photosensitiser and a photothermal agent, respectively by Ni *et al*. (2021b), to enhance the antibacterial activity of neat films. The developed nanocomposite films were applied to cherry tomatoes. The graphite carbon nitride nanosheets/MoS_2 nanodots-loaded films showed better mechanical properties, hydrophobicity, and thermal stability compared to neat konjac glucomannan films. The antibacterial activity of nanocomposite films was determined after 5 min of dual light irradiation. The inhibition zone size of the composite film increased with increasing content of graphite carbon nitride nanosheets/MoS_2 nanodots up to 10%. Under dual light irradiation, the temperature of the neat film increased from 28 ± 1 to 38 ± 1°C, while nanocomposite film reached 55 ± 4°C for 5 min. Hence, short-term dual light irradiation and appropriate temperature increase resulted in the enhancement of the antibacterial activity of the film against *E. coli* and *S. aureus*. The cherry tomatoes in 10% graphite carbon nitride nanosheets/MoS_2 nanodots-loaded nanocomposite films also exhibited the largest stiffness, reduced spoilage, and thus significantly prolonged shelf life. The studies revealed the potential of g-C_3N_4 nanofiller for active food packaging films with excellent performances.

Other carbon-based nanomaterials

Biochar emerges as a non-toxic, sustainable, and renewable substance crafted through the pyrolysis process of biomass (She *et al*., 2019). Biochar is one of the most important carbon-based fillers that has desirable properties such as wide availability, low cost, high porosity, and chemical stability (Parin *et al*., 2020). Biochar stands out as a promising alternative to petroleum-derived fillers like carbon black used in polymeric materials (Kane and Ryan, 2022). Kane and Ryan (2022) reported the challenges associated with utilising biochar as a filler material in PLA. It was revealed that higher biochar content led to a reduction in the onset of thermal degradation and melt viscosity in PLA. The presence of inorganic compounds within biochar was believed to catalyse the thermal decomposition of PLA, offering insights into the complex interactions between biochar and PLA matrices. A bioplastic–biochar composite packaging was developed by Diaz *et al*. (2020). The effect of biochar content (10, 20, and 30 wt%) on the properties of fully biodegradable thermoformed containers based on starch/PCL biocomposites was investigated. It was reported that the thermoforming ability did not change with the increased biochar loading amount. The incorporation of biochar reduced the elongation at break; however, it did not remarkably affect the elasticity modulus or tensile strength. The developed biocomposite coffee cup lids were proposed as suitable candidates for future sustainable packaging products. According to Botta *et al*. (2021), biochar was an efficient filler for poly(butylene adipate-co-terephthalate) (PBAT) biocomposite blown films. The uniform distribution of biochar and strong adhesion within the polymeric matrix led to a notable rise in the elastic modulus. The 5 and 10% biochar-loaded PBAT films demonstrated favourable characteristics suitable for packaging and compost bag applications.

Hydrochar is a carbon-rich, solid material produced through hydrothermal carbonisation of biomass in the presence of water at relatively low temperatures (typically 180 - 250°C) and high pressures (Rodríguez-Narvaez *et al*., 2022). Zhang *et al*. (2022) constructed biochar and hydrochar microsphere incorporated zein films using solution casting method. Using the biochar which had superior thermal stability compared to hydrochar resulted in development of composite films with improved thermal properties. The mechanical tests revealed that the incorporation of biochar enhanced both the stiffness and toughness of biopolymeric matrix, which was related to their excellent compatibility and hydrogen bonding interactions.

Biochar-incorporated films were also evaluated for active packaging applications. In a recent study by Feng *et al*. (2024), a novel approach was introduced for preserving blueberry fruit using a film composed of PVA/chitosan embedded with silver-loaded biochar nanoparticles. The research revealed that PVA/chitosan films containing 3 wt.% of silver-loaded biochar nanoparticles demonstrated robust antibacterial properties, exceptional antioxidant activity, excellent thermal stability, and notable hydrophobic characteristics. Moreover, the PVA/chitosan/silver-loaded biochar-based film effectively shielded against light, thus minimising lipid peroxidation and discoloration. This feature is highly beneficial for fruit preservation purposes. Alves *et al*. (2024) surveyed the impact of varying quantities (10 - 50 wt.%) of ZnO/biochar nanoparticles on the characteristics of thermoplastic starch-based films produced using melt-mixing and hot-pressing. Results indicated that incorporating 50 wt.% of ZnO/biochar led to a 30% decrease in the film's water solubility, accompanied by a 1.5-fold increase in stiffness. Furthermore, the incorporation of ZnO/biochar endowed the starch matrix with functional attributes, including antioxidant activity, antimicrobial efficacy, and electrical conductivity.

Another fascinating carbon-based nanomaterial is activated carbon due to its high specific surface area, well-ordered pore structure, low toxicity, good electrical conductivity, and surface functionality (Sobhan *et al*., 2019). Sobhan *et al*. (2019) investigated the addition of activated carbon to nanocellulose biodegradable film for smart packaging. It was determined that the films had thermal stability up to 270°C. However, the films were not found to be suitable for smart food packaging owing to their poor electrical properties for the biosensing function. Another smart packaging film was designed using activated carbon/silver/cellulose nanofibres (Sobhan *et al*., 2020). The study was a novel approach to fabricate antimicrobial and conductive nanocomposite films. These nanocomposites hold promise for diverse applications in sensors and antimicrobial materials, hence presenting a sophisticated solution for smart food packaging films. dos Santos *et al*. (2022) reported beneficial outcomes of utilising chitosan coatings infused with palmitic acid and activated carbon on surfaces of paperboard. This sustainable approach suggested as a natural alternative to enhance the moisture and fat barrier characteristics of cellulosic packaging materials, while preserving biodegradability and recyclability.

Overall evaluation

The development of innovative packaging materials using carbon-based nanofillers incorporated polymeric bio-nanocomposites opens up a number of opportunities for the food packaging sector. Their remarkable mechanical, electrical, antibacterial, antioxidant, and thermal qualities have made their way into practically various applications of packaging. However, it is important to select the most suitable filler type as well as biodegradable polymer matrix for the optimum design. The general evaluation for the most featured properties of carbon-based fillers is given in Table 3.

For biodegradable packaging, graphitic carbon nitride (g-C_3N_4) and graphene oxide (GO) are generally more compatible due to their biodegradability and environmental friendliness. Carbon dots (CDs) can also be beneficial for their biocompatibility, antioxidant, and antimicrobial properties (Zhao *et al*., 2023). While carbon nanotubes and graphene offer strength and barrier benefits, they may pose environmental and toxicity concerns.

Graphene is a hydrophobic and non-polar material; thus, it interacts weakly with polar polymers, which may lead to insufficient dispersion. Due to its functional groups and good dispersibility in water, GO can be used as nanofiller in hydrophilic or polar polymers (Eslami *et al*., 2023). The sheets are sufficiently stabilised by the oxygen-containing functional groups on GO to enable better

Table 3. Featured properties of carbon-based fillers for biodegradable packaging.

Material	Advantage	Disadvantage	Cost
Carbon dots	- Biocompatible, - Bioactive, - UV resistance, - Easy integration - Environmentally friendly, - Sustainable (Ezati *et al*., 2022a; Manikandan and Min, 2023; Zhao *et al*., 2023)	- Limited strength	Low (Zhao *et al*., 2023)
Carbon nanotubes	- High strength, - Good barrier properties	- Toxicity concerns - Environmental persistence (Lajeunesse *et al*., 2013)	Moderate to high
Graphene	- Biocompatible, - High strength, - High chemical stability, - Excellent barrier properties (Farjadian *et al*., 2020; Rossa *et al*., 2022; Yang *et al*., 2022)	- Hydrophobicity - Low compatibility with most polymers (Ashok Kumar *et al*., 2022; Belay, 2023)	High (Yang *et al*., 2022)
Graphene oxide	- Water dispersible and hydrophilic, - Good barrier properties, - High surface area (aspect ratio), - Good antimicrobial activity, -Surface functionalization capability (Malhotra *et al*., 2020; Mohammadi and Babaei, 2022; Rossa *et al*., 2022)	-Not dispersive in organic solvents -Highly susceptible to aggregate in the polymer matrix (Malhotra *et al*., 2020; Mohammadi and Babaei, 2022)	Low to moderate (Yang *et al*., 2022)
Reduced graphene oxide	- High surface area, - Excellent electrical conductivity, - Good chemical resistance - Bioactive (Joshi *et al*., 2023)	- Hydrophobicity (Joshi *et al*., 2023)	Moderate
Graphitic carbon nitride	- Non-toxic, - Good chemical and thermal stability, - Bioactive, - Hydrophilicity, - Optical transparency (Liu *et al*., 2023; 2024; Tari *et al*., 2024; Zhang *et al*., 2024)	- Low surface area in the bulk form (Kyriakos *et al*., 2024)	Low to moderate (Liu *et al*., 2024)

incorporation and even dispersion within these matrixes when compared to graphene (Agarwal and Zetterlund, 2021). Since rGO also has a hydrophobic surface, it disperses efficiently in non-polar solvents or matrix polymers (Nguyen *et al*., 2023).

Price is a key parameter in the choice of filler. Notwithstanding its enormous potential, graphene has several drawbacks that prevent widespread use and large-scale commercialisation, such as high production costs, issues with quality control, scalability problems, uncertainty about the effects on the environment and human health, and challenges with application integration (Yang *et al*., 2024). Also, the high price of CNTs in food packaging is a significant limitation to their widespread adoption, despite their advantageous properties such as strength, lightweight, and electrical conductivity. The cost factors are influenced by the production methods, material sources, and market demand. However, high production costs of CNTs still remain a challenge that researchers are actively addressing through innovative synthesis techniques (Liu *et al*., 2022).

The choice of carbon-based nanofiller is critical as it directly impacts several properties (*e.g*., mechanical, thermal, and barrier) of the biodegradable polymer matrix. The key considerations have been presented in the present review. However, selecting an appropriate filler for biodegradable polymer matrices involves a multifaceted approach that considers the types of polymer, properties of potential filler, processing methods, desired performance characteristics, and environmental impacts. By carefully evaluating these factors, researchers can develop effective biodegradable nanocomposites tailored for packaging industries.

Conclusion

Food packaging materials are fundamental components of everyday life. The food industry predominantly employs synthetic petroleum-based materials for packaging purposes. However, there has been a notable shift in consumer preference towards sustainable and biodegradable food packaging solutions, driven by increasing environmental concerns regarding the disposal of synthetic polymer products. In order to effectively compete with conventional plastics, biodegradable polymers must be enhanced to improve specific properties, including barrier functions against light, gas, and water vapour, as well as mechanical, thermal, and other functional attributes. The integration of nanotechnology presents a promising avenue for enhancing packaging functionalities. Recent research has highlighted the viability of incorporating biodegradable polymers with carbon-based nanofillers for active and intelligent food packaging applications. Such nanocomposites are capable of improving food safety, preserving quality, and extending the shelf life of food products. However, comprehensive *in vitro* and *in vivo* toxicity assessments and migration studies are necessary to establish the safety of biodegradable packaging materials that utilise carbon-based nanofillers. Furthermore, scientific data regarding the synergistic effects of carbon-based nanofillers in conjunction with other organic fillers is scarce. Future investigations should concentrate on the interactions between these carbon-based nanofillers and bioactive agents to achieve superior outcomes. Additionally, it is vital to address the recycling processes related to bio-nanocomposites, as innovative strategies are needed to recycle composite packaging materials that incorporate diverse fillers effectively.

Acknowledgement

The authors extend their appreciation to the Deanship of Research and Graduate Studies at King Khalid University for financially supporting the present work through the Large Research Grant Project scheme (grant no.: RGP2/224/45). The authors gratefully thank Dr. Sukhwinder Kaur Bhullar for proofreading the manuscript.

References

Agarwal, V. and Zetterlund, P. B. 2021. Strategies for reduction of graphene oxide - A comprehensive review. Chemical Engineering Journal 405: 127018.

Alves, Z., Brites, P., Ferreira, N. M., Figueiredo, G., Otero-Irurueta, G., Gonçalves, I., … and Nunes, C. 2024. Thermoplastic starch-based films loaded with biochar-ZnO particles for active food packaging. Journal of Food Engineering 361: 111741.

Amin, U., Khan, M. U., Majeed, Y., Rebezov, M., Khayrullin, M., Bobkova, E., … and Thiruvengadam, M. 2021. Potentials of polysaccharides, lipids and proteins in biodegradable food packaging applications. International Journal of Biological Macromolecules 183: 2184-2198.

Ananthi, P., Hemkumar, K., Subasini, S. and Pius, A. 2023. Development of biodegradable films reinforced with silver functionalized cow milk carbon dots for active food packaging applications. Materials Today Sustainability 24: 100609.

Antolin-Ceron, V. H., Gonzalez-Jauregui, D., Astudillo-Sanchez, P. D., Cabrera-Chavarria, J., Andrade-Melecio, H. A., Barrera-Rivera, K. A. and Martinez-Richa, A. 2023. Influence of carbon nanotube functionalization on the physical properties of PCL diol/chitosan blends. Journal of Chemical Technology and Biotechnology 98(7): 1673-1689.

Anzar, N., Hasan, R., Tyagi, M., Yadav, N. and Narang, J. 2020. Carbon nanotube - A review on synthesis, properties and plethora of applications in the field of biomedical science. Sensors International 1: 100003.

Asgher, M., Qamar, S. A., Bilal, M. and Iqbal, H. M. N. 2020. Bio-based active food packaging materials: Sustainable alternative to conventional petrochemical-based packaging materials. Food Research International 137: 109625.

Ashok Kumar, S. S., Bashir, S., Ramesh, K. and Ramesh, S. 2022. A comprehensive review - Super hydrophobic graphene nanocomposite coatings for underwater and wet applications to enhance corrosion resistance. FlatChem 31: 100326.

Barra, A., Ferreira, N. M., Martins, M. A., Lazar, O., Pantazi, A., Jderu, A. A., … and Nunes, C. 2019. Eco-friendly preparation of electrically conductive chitosan - Reduced graphene oxide flexible bionanocomposites for food packaging and biological applications. Composites Science and Technology 173: 53-60.

Barra, A., Santos, J. D. C., Silva, M. R. F., Nunes, C., Ruiz-Hitzky, E., Gonçalves, I., … and Marques, P. A. A. P. 2020. Graphene derivatives in biopolymer-based composites for food packaging applications. Nanomaterials 10(10): 2077.

Belay, M. 2023. Preparation of biodegradable reduced graphene oxide/agar composites by *in situ* reduction of graphene oxide. International Journal of Polymer Science 2023: 583522.

Botta, L., Teresi, R., Titone, V., Salvaggio, G., La Mantia, F. P. and Lopresti, F. 2021. Use of biochar as filler for biocomposite blown films: Structure-processing-properties relationships. Polymers 13(22): 3953.

Carvalho, A. P. A. and Conte Junior, C. A. 2020. Green strategies for active food packagings - A systematic review on active properties of graphene-based nanomaterials and biodegradable polymers. Trends in Food Science and Technology 103: 130-143.

Charoensri, K., Rodwihok, C., Ko, S. H., Wongratanaphisan, D. and Park, H. J. 2021. Enhanced antimicrobial and physical properties of poly (butylene adipate-co-terephthalate)/zinc oxide/reduced graphene oxide ternary nanocomposite films. Materials Today Communications 28: 102586.

Chen, S., Zeng, Q., Tan, X., Ye, M., Zhang, Y., Zou, L., … and Hu, K. 2023. Photodynamic antibacterial chitosan/nitrogen-doped carbon dots composite packaging film for food preservation applications. Carbohydrate Polymers 314: 120938.

Cui, R., Jiang, K., Yuan, M., Cao, J., Li, L., Tang, Z. and Qin, Y. 2020. Antimicrobial film based on polylactic acid and carbon nanotube for controlled cinnamaldehyde release. Journal of Materials Research and Technology 9(5): 10130-10138.

Dadfar, S. M. M. and Kavoosi, G. 2015. Mechanical and water binding properties of carboxymethyl cellulose/multiwalled carbon nanotube nanocomposites. Polymer Composites 36(1): 145-152.

Deka, M. J., Chowdhury, D. and Nath, B. K. 2022. Recent development of modified fluorescent carbon quantum dots-based fluorescence sensors for food quality assessment. Carbon Letters 32(5): 1131-1149.

Diaz, C. A., Shah, R. K., Evans, T., Trabold, T. A. and Draper, K. 2020. Thermoformed containers based on starch and starch/coffee waste biochar composites. Energies 13(22): 6034.

Dong, G., Zhang, Y., Pan, Q. and Qiu, J. 2014. A fantastic graphitic carbon nitride (g-C_3N_4) material - Electronic structure, photocatalytic and photoelectronic properties. Journal of Photochemistry and Photobiology C - Photochemistry Reviews 20: 33-50.

dos Santos, J. W. S., Garcia, V. A. S., Venturini, A. C., Carvalho, R. A., da Silva, C. F. and Yoshida, C. M. P. 2022. Sustainable coating paperboard packaging material based on chitosan, palmitic acid, and activated carbon - Water vapor and fat barrier performance. Foods 11(24): 4037.

Du, F., Shuang, S., Guo, Z., Gong, X., Dong, C., Xian, M. and Yang, Z. 2020. Rapid synthesis of multifunctional carbon nanodots as effective antioxidants, antibacterial agents, and quercetin nanoprobes. Talanta 206: 120243.

Eslami, B., Ghasemi, I. and Esfandeh, M. 2023. Using pegylated graphene oxide to achieve high performance solid polymer electrolyte based on poly(ethylene oxide)/polyvinyl alcohol blend (PEO/PVA). Polymers 15(14): 3063.

Ezati, P., Priyadarshi, R. and Rhim, J.-W. 2022a. Prospects of sustainable and renewable source-based carbon quantum dots for food packaging applications. Sustainable Materials and Technologies 33: e00494.

Ezati, P., Rhim, J.-W., Molaei, R. and Rezaei, Z. 2022c. Carbon quantum dots-based antifungal coating film for active packaging application of avocado. Food Packaging and Shelf Life 33: 100878.

Ezati, P., Rhim, J.-W., Molaei, R., Priyadarshi, R. and Han, S. 2022b. Cellulose nanofiber-based coating film integrated with nitrogen-functionalized carbon dots for active packaging applications of fresh fruit. Postharvest Biology and Technology 186: 111845.

Ezati, P., Roy, S. and Rhim, J.-W. 2022d. Pectin/gelatine-based bioactive composite films reinforced with sulfur functionalized carbon dots. Colloids and Surfaces A - Physicochemical and Engineering Aspects 636: 128123.

Fan, K., Zhang, M., Fan, D. and Jiang, F. 2019. Effect of carbon dots with chitosan coating on microorganisms and storage quality of modified-atmosphere-packaged fresh-cut cucumber. Journal of the Science of Food and Agriculture 99(13): 6032-6041.

Farjadian, F., Abbaspour, S., Sadatlu, M. A. A., Mirkiani, S., Ghasemi, A., Hoseini-Ghahfarokhi, M., … and Hamblin, M. R. 2020. Recent developments in graphene and graphene oxide: Properties, synthesis, and modifications - A review. ChemistrySelect 5(33): 10200-10219.

Feng, Q., Fan, B., He, Y.-C. and Ma, C. 2024. Antibacterial, antioxidant and fruit packaging ability of biochar-based silver nanoparticles-polyvinyl alcohol-chitosan composite film. International Journal of Biological Macromolecules 256: 128297.

Fu, B., Liu, Q., Liu, M., Chen, X., Lin, H., Zheng, Z., … and Yang, D.-P. 2022. Carbon dots enhanced gelatine/chitosan bio-nanocomposite packaging film for perishable foods. Chinese Chemical Letters 33(10): 4577-4582.

Gaddam, S. K., Pothu, R. and Boddula, R. 2020. Graphitic carbon nitride (g-C_3N_4) reinforced polymer nanocomposite systems—A review. Polymer Composites 41(2): 430-442.

Gu, B., Jiang, Q., Luo, B., Liu, C., Ren, J., Wang, X. and Wang, X. 2021. A sandwich-like chitosan-based antibacterial nanocomposite film with reduced graphene oxide immobilized silver nanoparticles. Carbohydrate Polymers 260: 117835.

Gürler, N. and Torğut, G. 2021. Graphene-reinforced potato starch composite films: Improvement of mechanical, barrier and electrical properties. Polymer Composites 42(1): 173-180.

Gürler, N. and Torğut, G. 2022. Dielectric biodegradable biopolymer-based graphene nanocomposites for use in the packaging industry and capacitor application. ChemistrySelect 7(32): e202201975.

Han Lyn, F., Chin Peng, T., Ruzniza, M. Z. and Nur Hanani, Z. A. 2019. Effect of oxidation degrees of graphene oxide (GO) on the structure and physical properties of chitosan/GO composite films. Food Packaging and Shelf Life 21: 100373.

Han, Z., Zhu, H. and Cheng, J.-H. 2024. Constructing a novel humidity sensor using acrylic acid/bagasse cellulose porous hydrogel combining graphene oxide and citral for antibacterial and intelligent fruit preservation. Carbohydrate Polymers 326: 121639.

Hong, S. J., Riahi, Z., Shin, G. H. and Kim, J. T. 2024. *Pseudomonas aeruginosa*-derived carbon dots doped with sulfur as active packaging materials for fresh food preservation. Food Bioscience 57: 103506.

Ibrahim, A. M. M., Abou Elfadl, A., El Sayed, A. M. and Ibrahim, I. M. 2023. Improving the optical, dielectric properties and antimicrobial activity of chitosan-PEO by GO/MWCNTs - Nanocomposites for energy storage and food packaging applications. Polymer 267: 125650.

Iqbal, M., Niazi, M. B. K., Jahan, Z., Ahmad, T., Hussain, Z. and Sher, F. 2022. Fabrication and characterization of carbon-based nanocomposite membranes for packaging application. Polymer Bulletin 79(7): 5019-5040.

Jagadeesh, P., Puttegowda, M., Mavinkere Rangappa, S. and Siengchin, S. 2021. Influence of nanofillers on biodegradable composites - A comprehensive review. Polymer Composites 42(11): 5691-5711.

Joshi, S., Bobade, H., Sharma, R. and Sharma, S. 2023. Graphene derivatives - Properties and potential food applications. Journal of Industrial and Engineering Chemistry 123: 1-18.

Kane, S. and Ryan, C. 2022. Biochar from food waste as a sustainable replacement for carbon black in upcycled or compostable composites. Composites Part C - Open Access 8: 100274.

Kang, C., Huang, Y., Yang, H., Yan, X. F. and Chen, Z. P. 2020. A review of carbon dots produced from biomass wastes. Nanomaterials 10(11): 2316.

Kaur, G., Sharma, S., Mir, S. A. and Dar, B. N. 2021. Nanobiocomposite films: A "greener alternate" for food packaging. Food and Bioprocess Technology 14(6): 1013-1027.

Khan, A., Ezati, P. and Rhim, J.-W. 2023a. Chitosan/starch-based active packaging film with N, P-doped carbon dots for meat packaging. ACS Applied Bio Materials 6(3): 1294-1305.

Khan, A., Priyadarshi, R., Bhattacharya, T. and Rhim, J.-W. 2023b. Carrageenan/alginate-based functional films incorporated with *Allium sativum* carbon dots for UV-barrier food packaging. Food and Bioprocess Technology 16(9): 2001-2015.

Khan, A., Riahi, Z., Tae Kim, J. and Rhim, J.-W. 2024. Carrageenan-based multifunctional packaging films containing Zn-carbon dots/anthocyanin derived from Kohlrabi peel for monitoring quality and extending the shelf life of shrimps. Food Chemistry 432: 137215.

Khoshkalampour, A., Ghorbani, M. and Ghasempour, Z. 2023. Cross-linked gelatine film enriched with green carbon quantum dots for bioactive food packaging. Food Chemistry 404: 134742.

Kilic, B., Dogan, V., Kilic, V. and Kahyaoglu, L. N. 2022. Colorimetric food spoilage monitoring with carbon dot and UV light reinforced fish gelatine films using a smartphone application. International Journal of Biological Macromolecules 209: 1562-1572.

Kiranakumar, H. V., Thejas, R., Naveen, C. S., Khan, M. I., Prasanna, G. D., Reddy, S., … and Jameel, M. 2024. A review on electrical and gas-sensing properties of reduced graphene oxide-metal oxide nanocomposites. Biomass Conversion and Biorefinery 14(12): 12625-12635.

Koshy, R. R., Koshy, J. T., Mary, S. K., Sadanandan, S., Jisha, S. and Pothan, L. A. 2021. Preparation of pH sensitive film based on starch/carbon nano dots incorporating anthocyanin for monitoring spoilage of pork. Food Control 126: 108039.

Kousheh, S. A., Moradi, M., Tajik, H. and Molaei, R. 2020. Preparation of antimicrobial/ultraviolet protective bacterial nanocellulose film with carbon dots synthesized from lactic acid bacteria. International Journal of Biological Macromolecules 155: 216-225.

Krystyjan, M., Khachatryan, G., Khachatryan, K., Konieczna-Molenda, A., Grzesiakowska, A., Kuchta-Gładysz, M., … and Nowak, N. 2022. The functional and application possibilities of starch/chitosan polymer composites modified by graphene oxide. International Journal of Molecular Sciences 23(11): 5956.

Kumari, S. V. G., Pakshirajan, K. and Pugazhenthi, G. 2022. Recent advances and future prospects of cellulose, starch, chitosan, polylactic acid and polyhydroxyalkanoates for sustainable food packaging applications. International Journal of Biological Macromolecules 221: 163-182.

Kyriakos, P., Hristoforou, E. and Belessiotis, G. V. 2024. Graphitic carbon nitride (g-C_3N_4) in photocatalytic hydrogen production: Critical overview and recent advances. Energies 17(13): 3159.

Lajeunesse, A., Blais, M., Barbeau, B., Sauvé, S. and Gagnon, C. 2013. Ozone oxidation of antidepressants in wastewater - Treatment evaluation and characterization of new by-products by LC-QToFMS. Chemistry Central Journal 7(1): 15.

Li, F., Yu, H.-Y., Wang, Y.-Y., Zhou, Y., Zhang, H., Yao, J.-M., … and Tam, K. C. 2019. Natural biodegradable poly(3-hydroxybutyrate-*co*-3-hydroxyvalerate) nanocomposites with multifunctional cellulose nanocrystals/graphene oxide hybrids for high-performance food packaging. Journal of Agricultural and Food Chemistry 67(39): 10954-10967.

Li, J., Wang, S., Lai, L., Liu, P., Wu, H., Xu, J., … and Wang, W.-J. 2021. Synergistic enhancement of gas barrier and aging resistance for biodegradable films with aligned graphene nanosheets. Carbon 172: 31-40.

Lin, W., Huang, G., Yang, W., Zeng, S., Luo, X., Huang, J. and Li, Z. 2022. A dual-function chitosan packaging film for simultaneously monitoring and maintaining pork freshness. Food Chemistry 392: 133242.

Liu, F., Wang, Q., Zhai, G., Xiang, H., Zhou, J., Jia, C., … and Zhu, M. 2022. Continuously processing waste lignin into high-value carbon nanotube fibers. Nature Communications 13(1): 5755.

Liu, S., Gao, X., Fan, H., Zhang, M., Waterhouse, G. I. N. and Zhu, S. 2023. Green and recyclable graphitic carbon nitride/chitosan/polyvinyl alcohol photocatalytic films with efficient antibacterial activity for fruit packaging. International Journal of Biological Macromolecules 236: 123974.

Liu, S., Jiang, X., Zhang, M., Gao, X., Jiang, R., Waterhouse, G. I. N. and Fan, H. 2024. Porous carbon nitride-chitosan/zein bilayer functional films with efficient photocatalytic antibacterial activity and tunable water barrier performance for food packaging. Food Bioscience 61: 104748.

Liu, Y., Wang, S., Lan, W. and Qin, W. 2019. Fabrication of polylactic acid/carbon nanotubes/chitosan composite fibers by electrospinning for strawberry preservation. International Journal of Biological Macromolecules 121: 1329-1336.

Lu, R., Zhang, X., Cheng, X., Zhang, Y., Zan, X. and Zhang, L. 2020. Medical applications based on supramolecular self-assembled materials from tannic acid. Frontiers in Chemistry 8: 583484.

Mahmud, J., Sarmast, E., Shankar, S. and Lacroix, M. 2022. Advantages of nanotechnology developments in active food packaging. Food Research International 154: 111023.

Malhotra, N., Villaflores, O. B., Audira, G., Siregar, P., Lee, J.-S., Ger, T.-R. and Hsiao, C.-D. 2020a. Toxicity studies on graphene-based nanomaterials in aquatic organisms: Current understanding. Molecules 25(16): 3618.

Manikandan, N. A., Pakshirajan, K. and Pugazhenthi, G. 2020. Preparation and characterization of environmentally safe and highly biodegradable microbial polyhydroxybutyrate (PHB) based graphene nanocomposites for potential food packaging applications. International Journal of Biological Macromolecules 154: 866-877.

Manikandan, V. and Min, S. C. 2023. Biofabrication of carbon quantum dots and their food packaging applications - A review. Food Science and Biotechnology 32(9): 1159-1171.

Mao, L., Zuo, J., Liu, Y., Zheng, B., Dai, X., Bai, Z., … and Yao, J. 2023. Alginate based films integrated with nitrogen-functionalized carbon dots and layered clay for active food packaging applications. International Journal of Biological Macromolecules 253: 126653.

Mergen, Ö. B., Arda, E. and Evingür, G. A. 2020. Electrical, optical and mechanical properties of chitosan biocomposites. Journal of Composite Materials 54(11): 1497-1510.

Min, S., Ezati, P. and Rhim, J.-W. 2022. Gelatine-based packaging material incorporated with potato skins carbon dots as functional filler. Industrial Crops and Products 181: 114820.

Min, S., Ezati, P., Yoon, K. S. and Rhim, J.-W. 2023. Gelatine/poly(vinyl alcohol)-based functional films integrated with spent coffee ground-derived carbon dots and grapefruit seed extract for active packaging application. International Journal of Biological Macromolecules 231: 123493.

Mohammadi, S. and Babaei, A. 2022. Poly (vinyl alcohol)/chitosan/polyethylene glycol-assembled graphene oxide bio-nanocomposites as a prosperous candidate for biomedical applications and drug/food packaging industry. International Journal of Biological Macromolecules 201: 528-538.

Moradi, M., Molaei, R., Kousheh, S. A., Guimarães, J. T. and McClements, D. J. 2023. Carbon dots synthesized from microorganisms and food by-products - Active and smart food packaging applications. Critical Reviews in Food Science and Nutrition 63(14): 1943-1959.

Mousavi, S. N., Daneshvar, H., Seyed Dorraji, M. S., Ghasempour, Z., Panahi-Azar, V. and Ehsani, A. 2021. Starch/alginate/ Cu-g-C_3N_4 nanocomposite film for food packaging. Materials Chemistry and Physics 267: 124583.

Nguyen, V. T., Tran, N. T., Huynh, T. L., Le, D. V. and Hoang, D. 2023. Reduced graphene oxide/cellulose microfiber hybrid from the Vietnamese nipa palm tree - Synthesis, properties, and applications for preparation of poly(methyl methacrylate) composite. Journal of Thermoplastic Composite Materials 36(1): 253-273.

Ni, Y., Nie, H., Wang, J., Lin, J., Wang, Q., Sun, J., … and Wang, J. 2022. Enhanced functional properties of chitosan films incorporated with curcumin-loaded hollow graphitic carbon nitride nanoparticles for bananas preservation. Food Chemistry 366: 130539.

Ni, Y., Shi, S., Li, M., Zhang, L., Yang, C., Du, T., … and Wang, J. 2021a. Visible light responsive, self-activated bionanocomposite films with sustained antimicrobial activity for food packaging. Food Chemistry 362: 130201.

Ni, Y., Sun, J. and Wang, J. 2021b. Enhanced antimicrobial activity of konjac glucomannan nanocomposite films for food packaging. Carbohydrate Polymers 267: 118215.

Omerović, N., Djisalov, M., Živojević, K., Mladenović, M., Vunduk, J., Milenković, I., … and Vidić, J. 2021. Antimicrobial nanoparticles and biodegradable polymer composites for active food packaging applications. Comprehensive Reviews in Food Science and Food Safety 20(3): 2428-2454.

Ozyurt, D., Al Kobaisi, M., Hocking, R. K. and Fox, B. 2023. Properties, synthesis, and applications of carbon dots - A review. Carbon Trends 12: 100276.

Papadopoulou, E. L., Basnett, P., Paul, U. C., Marras, S., Ceseracciu, L., Roy, I. and Athanassiou, A. 2019. Green composites of poly(3-hydroxybutyrate) containing graphene nanoplatelets with desirable electrical conductivity and oxygen barrier properties. ACS Omega 4(22): 19746-19755.

Parin, F. N., Yildirim, K. And Terzioğlu, P. 2020. Biochar loaded chitosan/gelatine/poly(ethylene glycol) biocomposite beads: Morphological, thermal and swelling properties. Journal of Innovative Science and Engineering 4(2): 56-68.

Patil, A. S., Waghmare, R. D., Pawar, S. P., Salunkhe, S. T., Kolekar, G. B., Sohn, D. and Gore, A. H. 2020. Photophysical insights of highly transparent, flexible and re-emissive PVA @ WTR-CDs composite thin films: A next generation food packaging material for UV blocking applications. Journal of Photochemistry and Photobiology A - Chemistry 400: 112647.

Pattanshetti, A., Pradeep, N., Chaitra, V. and Uma, V. 2020. Synthesis of multi-walled carbon nanotubes (MWCNTs) from plastic waste and analysis of garlic coated Gelatine/MWCNTs nanocomposite films as food packaging material. SN Applied Sciences 2(4): 730.

Priyadarshi, R., Roy, S., Ghosh, T., Biswas, D. and Rhim, J.-W. 2022. Antimicrobial nanofillers reinforced biopolymer composite films for active food packaging applications - A review. Sustainable Materials and Technologies 32: e00353.

Riahi, Z., Khan, A., Rhim, J.-W., Shin, G. H. and Kim, J. T. 2024a. Carrageenan-based active and intelligent packaging films integrated with anthocyanin and TiO_2-doped carbon dots derived from sweet potato peels. International Journal of Biological Macromolecules 259: 129371.

Riahi, Z., Khan, A., Rhim, J.-W., Shin, G. H. and Kim, J. T. 2024b. Sustainable packaging film based on cellulose nanofibres/pullulan impregnated with zinc-doped carbon dots derived from avocado peel to extend the shelf life of chicken and tofu. International Journal of Biological Macromolecules 258: 129302.

Riahi, Z., Rhim, J.-W., Bagheri, R., Pircheraghi, G. and Lotfali, E. 2022. Carboxymethyl cellulose-based functional film integrated with chitosan-based carbon quantum dots for active food packaging applications. Progress in Organic Coatings 166: 106794.

Rodríguez-Narvaez, O. M., Medina-Orendain, D. A. and Mendez-Alvarado, L. N. 2022. Functionalized green carbon-based nanomaterial for environmental application. In Koduru, J. R., Karri, R. R., Mubarak, N. M. and Bandala, E. R. (eds). Sustainable Nanotechnology for Environmental Remediation, p. 347-382. United States: Elsevier.

Rossa, V., Monteiro Ferreira, L. E., da Costa Vasconcelos, S., Tai Shimabukuro, E. T., Gomes da Costa Madriaga, V., Carvalho, A. P., … and de Melo Lima, T. 2022. Nanocomposites based on the graphene family for food packaging: historical perspective, preparation methods, and properties. RSC Advances 12(22): 14084-14111.

Roy, S., Ezati, P., Rhim, J.-W. and Molaei, R. 2022. Preparation of turmeric-derived sulfur-functionalized carbon dots - Antibacterial and antioxidant activity. Journal of Materials Science 57(4): 2941-2952.

Ryu, B. D., Han, M., Ko, K. B., Cuong, T. V., Lim, C.-H., Lee, G. H. and Hong, C.-H. 2020. Gallium dopant-induced tunable electrical properties of reduced graphene oxide using metal organic chemical vapor deposition. Applied Surface Science 504: 144500.

Salimi, F., Moradi, M., Tajik, H. and Molaei, R. 2021. Optimization and characterization of eco-friendly antimicrobial nanocellulose sheet prepared using carbon dots of white mulberry (*Morus alba* L.). Journal of the Science of Food and Agriculture 101(8): 3439-3447.

Shahbazi, M., Rajabzadeh, G. and Sotoodeh, S. 2017. Functional characteristics, wettability properties and cytotoxic effect of starch film incorporated with multi-walled and hydroxylated multi-walled carbon nanotubes. International Journal of Biological Macromolecules 104: 597-605.

She, D., Dong, J., Zhang, J., Liu, L., Sun, Q., Geng, Z. and Peng, P. 2019. Development of black and biodegradable biochar/gutta percha composite films with high stretchability and barrier properties. Composites Science and Technology 175: 1-5.

Sobhan, A., Muthukumarappan, K., Cen, Z. and Wei, L. 2019. Characterization of nanocellulose and activated carbon nanocomposite films' biosensing properties for smart packaging. Carbohydrate Polymers 225: 115189.

Sobhan, A., Muthukumarappan, K., Wei, L., Van Den Top, T. and Zhou, R. 2020. Development of an activated carbon-based nanocomposite film with antibacterial property for smart food packaging. Materials Today Communications 23: 101124.

Su, X., Lin, H., Fu, B., Mei, S., Lin, M., Chen, H., … and Lin, Y. 2023. Egg-yolk-derived carbon dots@albumin bio-nanocomposite as multifunctional coating and its application in quality maintenance of fresh litchi fruit during storage. Food Chemistry 405: 134813.

Sul, Y., Ezati, P. and Rhim, J.-W. 2023. Preparation of chitosan/gelatine-based functional films integrated with carbon dots from banana peel for active packaging application. International Journal of Biological Macromolecules 246: 125600.

Sun, L., Du, T., Hu, C., Chen, J., Lu, J., Lu, Z. and Han, H. 2017. Antibacterial activity of graphene oxide/g-C_3N_4 composite through photocatalytic disinfection under visible light. ACS Sustainable Chemistry and Engineering 5(10): 8693-8701.

Tammina, S. K. and Rhim, J.-W. 2023. Carboxymethylcellulose/agar-based functional film incorporated with nitrogen-doped polyethylene glycol-derived carbon dots for active packaging applications. Chemosphere 313: 137627.

Tari, E., Ugraskan, V. and Yazici, O. 2024. Enhanced mechanical, thermal and optical properties of poly(vinyl alcohol)/functionalized-graphitic carbon nitride composites. Fullerenes, Nanotubes and Carbon Nanostructures 32(5): 464-470.

Terzioglu, P., Altin, Y., Kalemtas, A. and Celik Bedeloglu, A. 2020. Graphene oxide and zinc oxide decorated chitosan nanocomposite biofilms for packaging applications. Journal of Polymer Engineering 40(2): 152-157.

Terzioğlu, P., Güney, F., Parın, F. N., Şen, İ. and Tuna, S. 2021. Biowaste orange peel incorporated chitosan/polyvinyl alcohol composite films for food packaging applications. Food Packaging and Shelf Life 30: 100742.

Trajkovska Petkoska, A., Daniloski, D., D'Cunha, N. M., Naumovski, N. and Broach, A. T. 2021. Edible packaging: Sustainable solutions and novel trends in food packaging. Food Research International 140: 109981.

Urade, A. R., Lahiri, I. and Suresh, K. S. 2023. Graphene properties, synthesis and applications - A review. JOM 75(3): 614-630.

Vasseghian, Y., Dragoi, E.-N., Almomani, F. and Le, V. T. 2022. Graphene derivatives in bioplastic: A comprehensive review of properties and future perspectives. Chemosphere 286: 131892.

Vieira, I. R. S., de Carvalho, A. P. A. and Conte-Junior, C. A. 2022. Recent advances in biobased and biodegradable polymer nanocomposites, nanoparticles, and natural antioxidants for antibacterial and antioxidant food packaging applications. Comprehensive Reviews in Food Science and Food Safety 21(4): 3673-3716.

Wang, P., Tang, S., Sheng, F., Cai, J., Fei, P., Nawaz, A., … and Xiong, H. 2019. Crystallization, thermal stability, barrier property, and aging resistance application of multi-functionalized graphene oxide/poly(lactide)/starch nanocomposites. International Journal of Biological Macromolecules 132: 1208-1220.

Weligama Thuppahige, V. T. and Karim, M. A. 2022. A comprehensive review on the properties and functionalities of biodegradable and semibiodegradable food packaging materials. Comprehensive Reviews in Food Science and Food Safety 21(1): 689-718.

Wen, F., Li, P., Yan, H. and Su, W. 2023. Turmeric carbon quantum dots enhanced chitosan nanocomposite films based on photodynamic inactivation technology for antibacterial food packaging. Carbohydrate Polymers 311: 120784.

Wen, Y.-H., Tsou, C.-H., de Guzman, M. R., Huang, D., Yu, Y.-Q., Gao, C., … and Wang, Z.-H. 2022. Antibacterial nanocomposite films of poly(vinyl alcohol) modified with zinc oxide-doped multiwalled carbon nanotubes as food packaging. Polymer Bulletin 79(6): 3847-3866.

Weng, S. X., Yousefi, N. and Tufenkji, N. 2019. Self-assembly of ultralarge graphene oxide nanosheets and alginate into layered nanocomposites for robust packaging materials. ACS Applied Nano Materials 2(3): 1431-1444.

Wu, W., Liu, L., Goksen, G., Demir, D. and Shao, P. 2022. Multidimensional (0D-3D) nanofillers: Fascinating materials in the field of bio-based food active packaging. Food Research International 157: 111446.

Xiong, H., Liu, H., Feng, X., Sun, Y., Huang, Q. and Xiao, C. 2024. A review of two-dimensional porous graphene with in-plane pores: Pore construction and membrane applications. Carbon 229: 119547.

Yang, K., Wu, C. and Zhang, G. 2024. A state of review for graphene-based materials in preparation methods, characterization, and properties. Materials Science and Engineering B 310: 117698.

Yang, Y., Wei, Y., Guo, Z., Hou, W., Liu, Y., Tian, H. and Ren, T. 2022. From materials to devices: Graphene toward practical applications. Small Methods 6(10): 2200671.

Yu, Y., Zheng, J., Li, J., Lu, L., Yan, J., Zhang, L. and Wang, L. 2021. Applications of two-dimensional materials in food packaging. Trends in Food Science and Technology 110: 443-457.

Zhang, F., Zhang, J., Wang, H., Li, J., Liu, H., Jin, X., … and Zhang, G. 2021. Single tungsten atom steered band-gap engineering for graphitic carbon nitride ultrathin nanosheets boosts visible-light photocatalytic H_2 evolution. Chemical Engineering Journal 424: 130004.

Zhang, M., Liu, S., Gao, X., Jiang, X., Zhang, E., Fan, H. and Zhu, S. 2024. Highly flexible carbon nitride-polyethylene glycol-cellulose acetate film with photocatalytic antibacterial activity for fruit preservation. International Journal of Biological Macromolecules 266: 131161.

Zhang, Q., Yang, X., Guo, Z., Fang, Y., Li, K. and Sheng, K. 2022. Zein composite film with excellent toughness - Effects of pyrolysis biochar and hydrochar microspheres. Journal of Cleaner Production 367: 133039.

Zhang, X., Wang, H., Niu, N., Chen, Z., Li, S., Liu, S.-X. and Li, J. 2020. Fluorescent poly(vinyl alcohol) films containing chlorogenic acid carbon nanodots for food monitoring. ACS Applied Nano Materials 3(8): 7611-7620.

Zhao, L., Zhang, M., Mujumdar, A. S. and Wang, H. 2023. Application of carbon dots in food preservation: A critical review for packaging enhancers and food preservatives. Critical Reviews in Food Science and Nutrition 63(24): 6738-6756.