Journal of Nanostructure in Chemistry (2024) 14:113–136
https://doi.org/10.1007/s40097-023-00527-3

REVIEW

Green synthesis of nanomaterials for smart biopolymer packaging: challenges and outlooks

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Received: 20 June 2022 / Revised: 6 February 2023 / Accepted: 11 February 2023 / Published online: 26 February 2023

Abstract

There are several physical and chemical methods for synthesizing nanomaterials, while the most appropriate techniques involve using green chemistry and eco-friendly material. Recently, green synthesized materials for different applications have gained attention as a result of their environmental friendliness and cost-effectiveness. Applying green synthesized nanoparticles (NPS) in food packaging has been extensively investigated. Biopolymers require filler to enhance the optical, barrier, thermal, antimicrobial, and mechanical properties of packaging. Biopolymer packaging incorporated with green synthesized NPs is expected to simultaneously enhance performance while reducing environmental damage. The current review article focuses on biopolymer films with bio (green)-synthesized nanomaterials and their effectiveness in reducing the negative environmental implications of synthetic packaging. It also covers the general concepts of green synthesis of NPs, their production methods, their performance, and characterization, and discusses the potential, performance and recent developments of bio-nanocomposite films/coatings in biodegradable food packaging. Recent reports and trends provide more insight into the impact of green synthesized nanomaterials on food packaging.

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Graphical Abstract

Keywords Green synthesized · Eco-friendly nanomaterials · Biodegradable · Active packaging · Smart packaging · Characterisations

Introduction

In recent years, there has been a tremendous increase in the demand for new methods of controlling or combating food-borne pathogens. Consequently, packaging with enhanced features for food is crucial to ensure the safety of food and maintain food quality [1, 2]. The concept of smart food packaging was developed in response to consumer demands for products that are safer and more natural with a prolonged shelf life, lower cost and greater convenience during storage and transportation [3]. Using antimicrobial packaging, germs or microorganisms are reduced, inhibited, or delayed in their growth on food surfaces by interacting with the product or headspace inside [4]. Packaging materials incorporated with antimicrobial agents can slowly diffuse specific bactericidal or bacteriostatic chemicals into the food matrix, removing the need to directly apply large antimicrobials [5, 6].

Because of their large “surface-to-volume” ratio and outstanding (in some cases unique) chemical and physical characteristics, nanoparticles (NPs) have considerable antibacterial activity at low doses [7–9]. Furthermore, they are more resistant to harsh conditions/settings such as high pressures and temperatures, and some are harmless and even include mineral components necessary for human health [10, 11]. Although NPs are increasingly used in food packaging [12], consumer perception and acceptance are affected by concerns about toxicity and potential health risks. The migration of nanomaterials from packaging or containers into food has been demonstrated in several investigations. However, several experimental studies have also shown that the number of reports of nanomaterial movement and migration is modest compared to other migration rates [7]. As a result, researchers and manufacturers are increasingly focusing on the "green or environmentally friendly production" of nanomaterials. In green synthesis, natural resources are used such as microbes, plant extracts, bioactive molecules, etc. [13]. There are many advantages to using green synthesis methods over traditional chemical and physical methods as they are less expensive, easier to handle, safer, free of pollutants/hazardous materials, renewable, as well as being energy-free and not requiring high pressures or temperatures to do so [14]. Therefore, the increased use of green techniques to synthesize nanomaterials is extremely promising.

Chemists and chemical technologists worldwide are turning to green chemistry concepts to produce non-hazardous chemical syntheses [15]. Biosynthesis has focused on developing non-toxic compounds and clean, environmentally friendly materials. Therefore, the production of biocompatible NPs utilising biological resources such as plant extracts/gums [16], fungi, yeasts, and bacteria has received enormous attention [17].

Despite the widespread research on efficient environmentally friendly nanomaterial and their application as active ingredients in biopolymer films, there have been no comprehensive reviews investigating the potential efficiency of using biosynthesized nanomaterials/nanocomposites in active and smart food packaging. This review article focuses on biopolymer films with bio (green)-synthesized nanomaterials and their significance in reducing the negative environmental implications of synthetic packaging. This review also covers the general concepts of green synthesis of NPs, the green nanomaterial production methods, and their characterization techniques. Furthermore, the performance and recent trends of green(bio) synthesized nanocomposites in biodegradable food packaging are investigated and addressed.

Active and smart food packaging

Consumers today require a new type of packaging called active packaging [18]. Furthermore, active food packaging results from innovation that goes beyond traditional packaging functions by improving food safety and extending shelf life by working with both packaging and product. This is accomplished by stalling microbial growth, lipid oxidation, and moisture loss degradation reactions and achieving better results than traditional food packaging [9, 19–21]. Active packaging is able to absorb food-related chemicals either from food, the environment, the headspace within the packaging, or release preservatives, antioxidants, and flavorings into the food or food environment. An active substance can be directly integrated into a polymer matrix or placed in a packaging or inserted into a label [20]. Antimicrobial and antioxidant packaging, emission of carbon dioxide, and oxygen/CO_2 absorbers are some of the most well-known and widely utilized active packaging solutions [22]. Antioxidant and antimicrobial packaging seems to be the most promising applications of active food packaging technologies [18].

Smart packaging is an emerging technology that can significantly improve food traceability, safety, and quality. This technology can be defined as a set of scientific measures/actions and innovations that allow food packaging systems to track changes in the external and internal environments, and the packaged food, to inform consumers, retailers, and manufacturers about the system's status [23, 24]. Throughout the "lifecycle of the product," smart packaging system monitors product freshness and informs customers. Moreover, this type of packaging can detect and deliver information about the properties of food, thereby informing the consumer about the product’s quality and safety [25, 26]. The effectiveness of active packaging systems can be tested using the smart packaging [18, 27]. In another word, Smart packaging performs a number of functions (e.g., tracking, sensing, communicating, recording, monitoring, and using scientific logic) to extend shelf life, improve quality, and provide information about products. Contrary to active packaging, intelligent packaging contains smart elements such as detection, communication, recording, and recognition [28]. Intelligent packaging with various communication methods including radio frequency identification/sensors, time–temperature indicators, freshness indicators, etc. is promising to improve the efficiency of information transmission in the entire product distribution chain as well as in the home of consumers [18]. Schematic representations of active and intelligent packaging concepts are presented in Fig. 1.

Green synthesis of nanoparticles

Nanostructured materials were widely used in every field of applied science such as the food sector and technological and medical applications [31]. So, developing cost-effective and eco-friendly methods is essential for the rapid synthesis of nanomaterials. Among all available techniques for the synthesis of NPs, biological, physical, and chemical methods are the most common approaches [32, 33]. However, the widespread utilization of chemical reagents and significant energy consumption in physical and chemical processes have raised concerns over their negative impacts on the environment and human health. To resolve this issue, the focus of the researchers is shifted from chemical and physical methods towards the ‘green’ synthesis techniques using bio-resources available in nature that includes the use of different microbes, plant extracts, bioactive components, and alike. The ‘green’ synthesis method is simple, eco-friendly, and cost-effective and does not require hazardous stabilizing agents, chemicals, large amounts of energy, high pressure, or temperature. This method can be classified as either intra- or extra-cellular synthesis. Over the last decade, several living organisms such as plant extracts and microorganisms (bacteria, fungi, etc.) have been used in the extra- and intracellular synthesis of NPs. Green synthesis of NPs has gained increasing popularity, particularly in food and biomedical applications due to its minimal use of harmful and hazardous

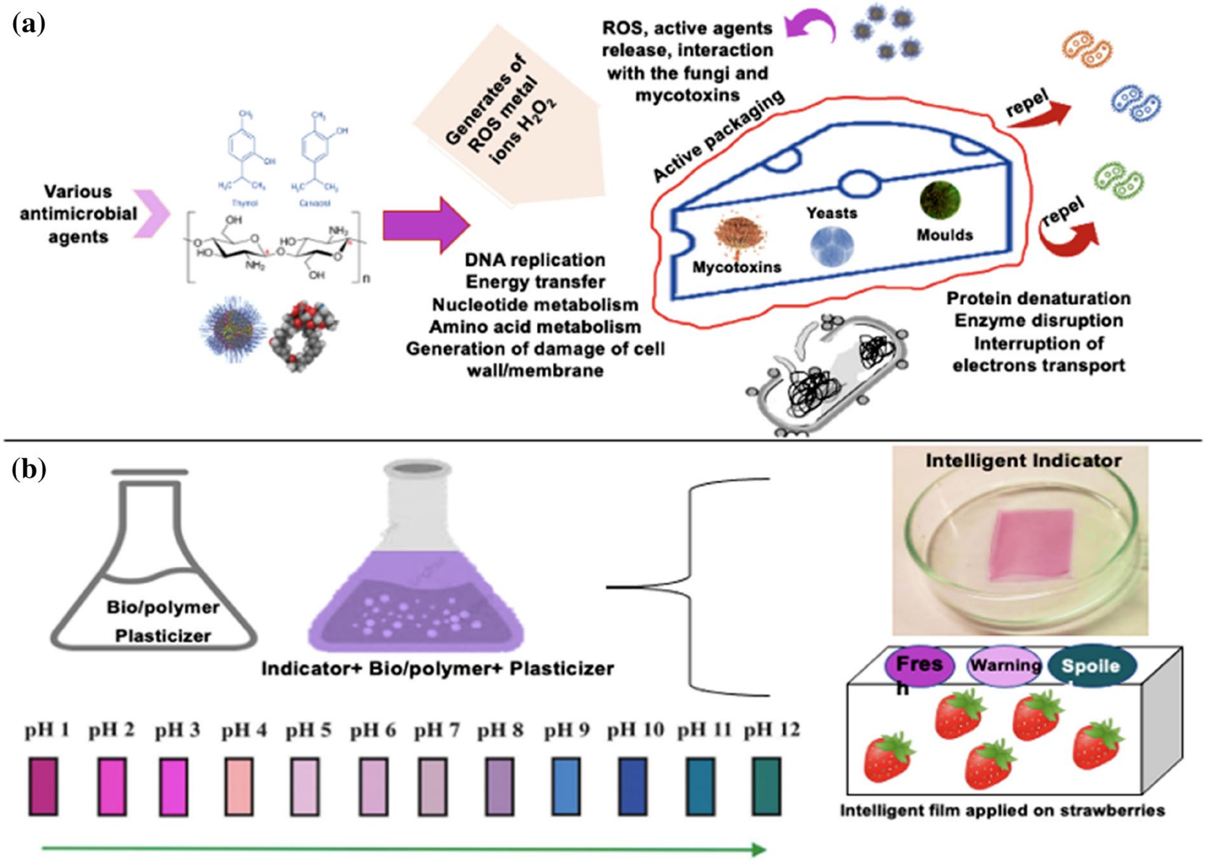


Fig. 1 Schematic diagram of **a** active and **b** intelligent packaging concepts. Adapted from [29, 30], reproduced with permissions from Elsevier

synthetic chemicals as well as their promising antibacterial activities/applications [34–36].

Role of plant in green synthesis of nanostructures

The use of plant extracts in the reduction and stabilization of agents has sparked research interest in the synthesis of NPs, especially in the area of food safety. Plant extracts-based method is simple, cost-effective, environmentally friendly, and has less or no chances of contamination [37]. Also, plant-mediated biosynthesis is better compared to the other biological (microorganism-based) methods due to being safe to handle, fast, unique, single-step, and producible at a large scale. Numerous plant parts, such as stems, roots, leaves, flowers, fruits, seeds, bark, or the whole plants are capable of synthesising NPs in a green way [38, 39]. The names of the plants already used in the green synthesis of NPs, including the characteristics and application of the synthesized NPs are listed in Table 1. Plant extracts involved in the green synthesis of NPs can be enriched with several phytochemicals e.g., amino acids, alkaloids, aldehydes, carboxylic acids, polysaccharides, vitamins, proteins, steroids, polyphenols, terpenoids, etc. that contain many functional groups (e.g., –OH, –NH, –CHO, –COOR, and –COOH). These phytochemicals act as the dual roles as successfully control the characteristics (morphology and composition) of the synthesized NPs and participate in the NPs formation [40].

Figure 2(a, b) shows a possible route to synthesize NPs using plant extracts and microorganism. The green synthesis of metallic NPs can be classified into three categories like activation, growth, and termination stages [41]. Plant extracts operate as reducing agents in the activation stage as salt solutions are injected into plant extracts, resulting in the reduction of metal ions into nuclei. Therefore, metal nuclei are formed with the help of plant extract, which serves as a reduction agent. In the period of growth, the adjacent nuclei combine further to create the final NPs. As part of the termination process, phytochemicals of plant extracts act as a capping agent on the surface of NPs, preventing their further aggregation.

Role of bacteria and fungi in green synthesis of nanostructures

In the recent era, the utilization of microbes has experienced increased attention through various investigations

Table 1 Recent progress in the biosynthesis of nanostructured materials using plant extract and microorganisms (bacteria and fungi)

Source	NPs produced	Characterization		Applications	References
		size (nm)	Shape		
Bacteria					
Acinetobacter sp. SW30	Au	8–12	Spherical	–	[45]
	Se	100	Amorphous		
Bacillus sp.	Ag	5–15	Spherical	Anticancer effect in breast cancer MCF-7 cells	[46]
Bacillus megaterium (NCIM2326)	ZnO	45–95	Rod and cubic	Antimicrobial	[47]
Delftia sp.	Bi	20–120	Spherical	Cytotoxic and antioxidant activity	[48]
Escherichia sp.	Cu	22.33–39 nm	Spherical	Azo dye degradation	[49]
Lactobacillus johnsonii	ZnO; TiO_2	4–9	Spherical (ZnO); irregular (TiO_2)	–	[50]
MKH1 bacterium	Ag	30–60	Mainly spherical	Hydrazine electro-oxidation	[51]
Pseudoalteromonas lipolytica	Au; Ag	AuNPs 10–30 AgNPs 5–15	Spherical	Methylene and Congo red decolourisation	[52]
Shewanella loihica PV-4	Au; Pd; Pt	2–7	Spherical	Methyl orange decomposition	[53]
Streptomyces spp	Fe	65–86		Antibacterial, Biomedical application	[54]
Fungi					
Aspergillus niger	ZnO	61	Spherical	Antimicrobial	[55]
Aspergillus foetidus	Au	30–50	Spherical	Noncytotoxic against A549 cell line	[56]
Aspergillus foetidus MTCC8876	Ag	20–40	Spherical	–	[57]
Lignosus rhinocerotis	Au	49.5–82.4	Spherical	Antibacterial activity	[58]
Macrophomina phaseolina	Ag/AgCl	5–30	Spherical	Crop protection	[59]
Penicillium pimiteouiense	FeO	2–16	Spherical	Chromium removal	[60]
Penicillium oxalicum	Fe	100–140	Spherical	Penicillium oxalicum	[44]
Periconium sp.	ZnO	16–78	Quasi spherical	Antimicrobial and antioxidant applications	[61]
Phomopsis liquidambaris	Ag	18.7	Spherical	Antimicrobial and mosquitocidal agent	[62]
Trichoderma asperellum	CuO	10–190	Cubic face-centered	Effect of photothermolysis on human lung carcinoma	[63]
Plant Extract					
Artocarpus heterophyllus	Fe	33		Degradation of Fuchsin Basic dye	[64]
Calotropis gigantean	SnO_2	35	Irregular	Methyl orange degradation	[65]
Cannabis sativa	Ag	26.52 nm	Spherical	antibacterial, anti-yeast and α-amylase inhibitory activity	[66]
Cissus arnotiana	Cu	60–90	Spherical	Antibacterial and antioxidant potential	[67]
Dillenia indica L	Ag	10–23	Spherical	Biological activity	[68]
Piper betle	SnO_2	8.4	Spherical	RY 186 dye degradation	[69]
Psidium guajava	SnO_2	8–10	Spherical	RY 186 dye degradation	[70]
Polyalthia longifolia	Au, Ag and Au–Ag alloy	5–20	Spherical	Dyes Degradation	[71]
Ruellia tuberosa	FeO	20– 80	Hexagonal nanorods	Antibacterial, Water purification	[72]
Tecoma castanifolia	ZnO	70–75	Spherical	Antibacterial, Antioxidant, Anti-cancer	[73]

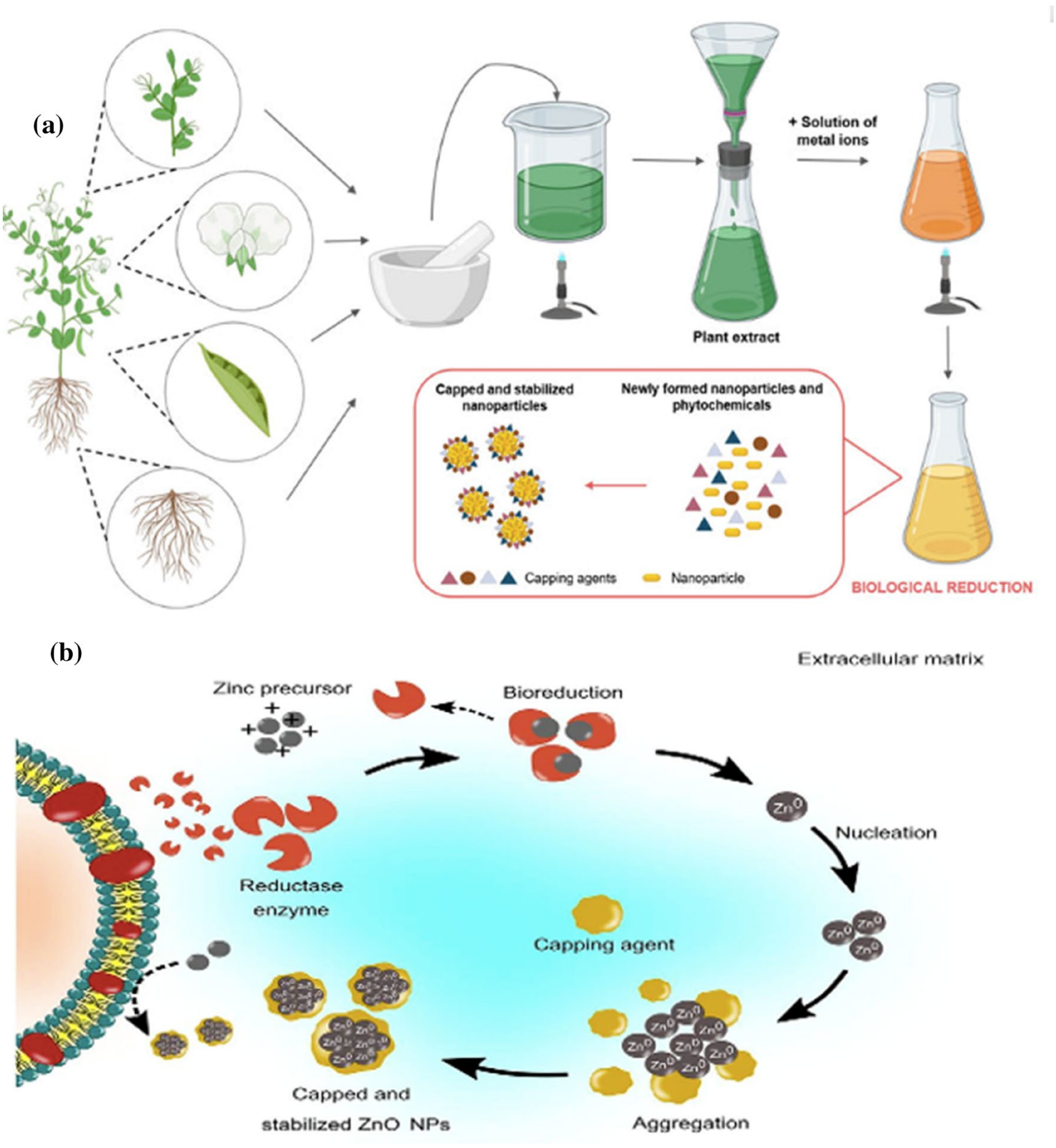


Fig. 2 Schematic illustration of the probable mechanism for extracellular biosynthesis of NPs using: **a** leaf extract and **b** microorganism (bacteria and fungi). Adapted from [42, 43], reproduced with permissions form MDPI and Springer, respectively

conducted for the green synthesis of NPs using numerous microorganism models [44]. Fungi and bacteria are frequently chosen for the green synthesis of NPs due to their rapid rates of growth, easier cultivation, and their capacity to grow under ambient physiological conditions. Green synthesis employing microbes has an advantage over plant synthesis due to microbes being more quickly replicated. Table 1 provides a summary of publications on the green synthesis of NPs mediated by bacteria and fungus including their size, shape, and potential applications.

In the green synthesis of NPs, microbe-derived proteins, enzymes, and other organic components play a crucial role. These biomolecules, released to the suspensions or growth medium, are responsible for the production of NPs of different sizes and morphologies with mono- and polydisperse NPs. Additionally, proteins released by microbes may serve as stabilizing agents that deliberates the stability of NPs [45, 48]. Figure 2b illustrates the plausible mechanism for green synthesis (extracellular) of NPs using microbes. The mechanism involves either the release of the enzyme into the growth medium or enzyme-mediated synthesis on the cell membrane [74]. Bio-reduction was started by the electron exchange of NADPH through the NADPH-dependent reductase, serving as an electron carrier. For instance, the positively charged metal acquired electrons can be reduced to the zero state, resulting in the synthesis of NPs. However, the exact mechanism for this has not yet been fully elucidated.

In summary, in this section, we have reviewed previous research on 'green' nanoparticle synthesis, methods and their advantages. Various natural extracts such as plants, fungi, bacteria, yeast, and plant extract are being used to produce nanomaterials. Among them, the plant extract is the most effective in reducing agents and stabilizing for synthesising controlled materials (i.e., control structures, shapes, and other features) [75]. A green nanoparticle is eco-friendly, sustainable, chemical-free, less expensive, and can be mass produced. Hence, the application of 'green' materials/nanoparticles derived from biocomponents is likely to be widespread in many areas, including pharmaceutical, environmental, food, and cosmetic industries [76].

Bio-based and biodegradable nanocomposite films

The problem of environmental waste generated by petrochemical-based non-recyclable plastic packaging materials combined with demands for high-quality food products have resulted in a growing market for biodegradable packaging made from renewable natural biopolymers such as proteins, carbohydrate polymers/polysaccharides [5, 77–79]. The term “biodegradable” is generally employed to explain those materials that can be degraded enzymatically by living organisms (e.g., bacteria and fungi) to H_2O, CO_2, and biomass under aerobic conditions and to biomass, methane, and hydrocarbons under anaerobic conditions [80]. Therefore, in many applications, there is significant interest in replacing synthetic plastics with biodegradable materials [5, 81, 82]. Some of the natural polymers and aliphatic polyesters such as polyhydroxybutyrate, polycaprolactone, and polylactic acid, and their copolymers are biodegradable, but their commercial applications are limited due to their higher cost compared to petroleum-based plastics that makes them find usage in niche sectors only [83]. The natural polymer-based packaging materials have however some inherent shortcomings, such as low water resistance and mechanical properties. Nanocomposite technology has proven to be a promising option for overcoming these limitations [84]. Figure 3 illustrates a modified biopolymer with improved properties.

Cellulose-based films

Regarding biodegradable and renewable polymers, one of the most abundant substances is cellulose. It is made from various sources such as crop, wood, cotton, some bacteria and cellulosic agriculture/food waste [86]. Cellulose is a linear polymer comprising of D-glucose units linked together via β (1 → 4) linkages containing highly ordered crystalline regions in conjugation with more disoriented amorphous parts [87]. Due to its unique properties, such as environmental friendliness, excellent mechanical properties, and non-toxicity, it is becoming increasingly important as

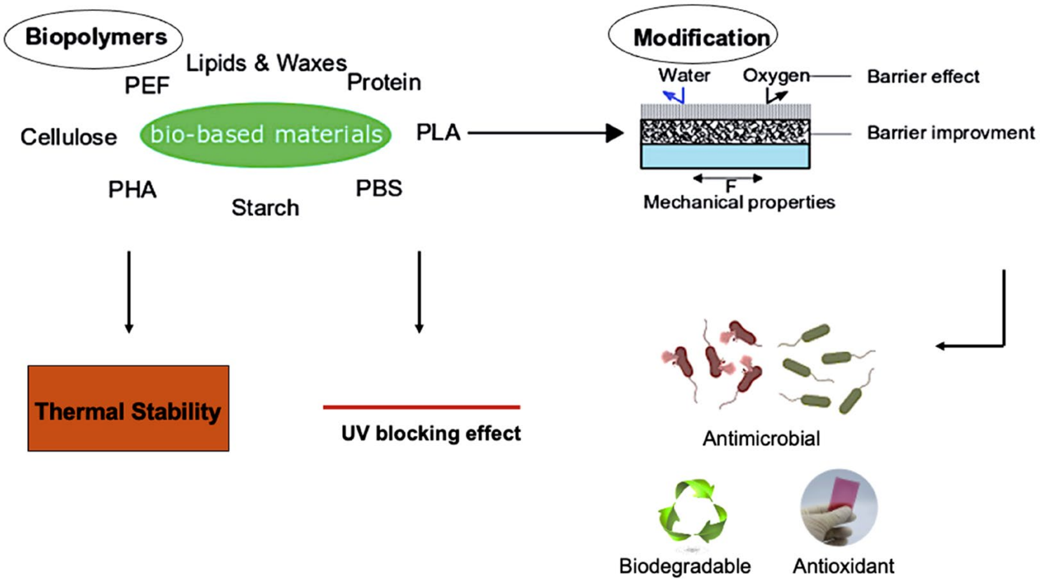


Fig. 3 Biodegradable nanocomposite film and its properties (*PHA* Polyhydrodyalkanoate, *PBS* Polybutylene succinate, *PEF* Polyethylene furanoate, *PLA* Polylactic acid). Adapted from [20, 85], reproduced with permission form Springer and MDPI, respectively

a renewable resource to replace petroleum-based materials [88]. Using cellulose, synthetic packaging material and waste can be reduced. Due to its lightweight, it also has the potential to lower the weight of packaging material. More importantly, cellulose could easily incorporate or encapsulate versatile natural antioxidant and antimicrobial agents. Additionally, micro- or nanoscale cellulose forms such as microfibrillated cellulose, cellulose NPs and microcrystalline cellulose could be used as reinforcements for tuning the mechanical and functional properties of nanocomposite films [89].

Zhao et al. [86] obtained high-purity cellulose from durian shell, and it was subsequently used to fabricate cellulose films. The durian-rind cellulose fiber was like cotton linter-derived commercial cellulose, according to the XRD and FTIR results. Cellulose film had a smooth surface and a good appearance and outstanding transparency. Moreover, the film had a high tensile strength and rigidity (TS). In four weeks, the durian rind film could be completely degraded under soil conditions, which indicates that its biodegradation rate is higher than that of cellophane. Therefore, durian could be used as a promising cellulosic resource to fabricate biodegradable packaging. In a similar study, glycerol and cellulose sulfate were combined to form novel edible films. [90]. In addition to being dense, flexible, homogeneous, transparent, and water-soluble, these films were also resistant to fats and oils. Also, adding glycerol to the films decreased their TS and "elongation at break" (EB), while increasing the molecular weight of the cellulose sulfate improved them. Moreover, it was found that the "water vapor permeability" (WVP) could be reduced by the addition of glycerol and increasing the molecular weight of the cellulose sulfate. Furthermore, the cellulose sulfate-based coating could improve the bananas' shelf life [90]. The shelf life of various foods is extended by the use of edible films made from cellulose-based materials (Table 2).

The cellulose and its derivatives have been blended with proteins, enzymes, phenolic compounds, metal particles, and synthetic and natural polymers to modulate their film properties [91–98]. For example, Gao et al. [99] developed microfibrillated cellulose/chitosan/polypyrrole composite films that show improved oxygen barrier, antioxidant, and antibacterial. The composite films had bactericidal activity against *Escherichia coli* (*E. coli*) and possessed good thermal stability and free radical scavenging activity.

Cellulose-based ZnO nanocomposite films were prepared and characterized by Fu et al. 2015 [108]. In addition to good mechanical properties, the nanocomposite films demonstrated excellent UV-blocking properties, high thermal stability as well as antibacterial effects against *E. coli* and *Staphylococcus aureus (S. aureus)*. The viable bacteria were reduced dramatically after 3 h of exposure, and all of them were killed within 6 h. Oun and Rhim [109] have reported that the cellulose nanocrystals/carboxymethyl cellulose nanocomposite films resulted in improved TS and reduced WVP. A study has also shown that adding microfibrillated cellulose, cellulose nanoparticles, and microcrystalline cellulose to a starch biopolymer matrix can improve its properties as mechanical and water vapor barriers [110]. The nano- and micro-structures of cellulose have also been used in alginate, agar, chitosan, carrageenan, and protein films to modulate their water vapor and mechanical barrier properties [111–115].

Cellulose and its derivatives-based films have been also utilized as carriers for antioxidant and antimicrobial agents. In addition, cellulose is used to fabricate multilayer packaging material that slows the release of bioactive compounds, provides a water vapour barrier and selective gas barrier, increases the shelf life of food products, and regulates the ripening process [116].

Green nanocomposites from bacterial sources

Bacterial cellulose-based composites

Bacterial cellulose is a natural polymer with extraordinary and differential attributes which is receiving particular interest in the food sector. Several factors determine its properties, such as the applied bacterial strain, the culture medium composition, and the post-incubation conditions. The physical and morphology properties of the bacterial cellulose are mainly determined by the bacteria used and the composition of the culture medium [117]. In contrast to plant cellulose, bacterial cellulose is synthesized purely, and it contains no pectin, hemicellulose, or lignin. Moreover, it doesn't require any expensive extraction or purification methods, nor does it use any chemicals that are hazardous to the environment [118].

Films made of disassembled bacterial cellulose are the most widely used and studied applications of bacterial cellulose. The polymer has the potential to be easily processed into nanocrystals, nanofibrils, and microfibrils. As reinforcements, these structures can physically be incorporated into different polymeric matrixes for fabricating polymeric composites [119]. The modified bacterial cellulose has a high surface area which could reinforce polymers' structure and promote powerful physical interactions (particularly hydrogen bonds), leading to superior mechanical, thermal, and barrier properties [120].

In research performed by Liu et al. [121], Gluconacetobacter xylinum was used as a versatile bio factory to synthesize bacterial cellulose and induce the formation of Ag/AgCl NPs, and in turn bacterial cellulose–Ag/AgCl nanocomposite. The nanocomposite fabrication process was conducted in room condition without using toxic agents or creating hazardous products. The nanocomposite exhibited desirable

Table 2 Preservation of various foods by cellulose and its derivatives-based films

Films	Process conditions	Food application	Results	References
Carboxymethyl cellulose/sodium caseinate	Carboxymethyl cellulose (0.5%) Sodium caseinate (0.75%) Glycerol (2.25%) Casting method	Fresh trout fillets	↓ Microbial growth ↓ Chemical changes ↑ Shelf-life	[100]
Hydroxypropyl methylcellulose/polyethylene oxide/gallic acid nanofibers (Nanofibers fixed to the inner surface of polyamide/polyethylene packages)	Hydroxypropyl methylcellulose (4%) Polyethylene oxide (1%) Tween 80 (2%) Gallic acid (2, 5, and 10%) Electrospinning process	Walnuts	↓ Oxidation	[101]
Nanocellulose/chitosan	Chitosan (1–1.3%w/0.6v) Nanocellulose (0–2%w/w chitosan) Glycerol (30–90%v/w chitosan) Casting method	Grounded meat	↓ Lactic acid bacteria Unchanged pH	[102]
Carboxymethyl cellulose/zinc oxide/chitosan	Chitosan (0.5%) Carboxymethyl cellulose (0.5%) Zinc oxide (2, 4, 8%) Casting method	Soft Cheese	↓ Microbial growth ↑ Shelf-life	[103]
Ethylene-based cellulose (bacterial cellulose nanofiber film doped with $KMnO_4$)	Bacterial cellulose gel membrane $KMnO_4$ (8, 0.8, 0.08 mM) Drying at room temperature	Banana	A portable optical film sensor can be used for determining the temperature and time of fruit storage in the packaging	[104]
Cellulose nanocrystals /polyvinyl alcohol	Polyvinyl alcohol (3–5%) Cellulose nanocrystals (1–4.5%) Casting method	Mangoes	↑ Antifungal activity ↓Weight loss	[105]
Cellulose/polylactide (NatureFlex™ E946; truncated cone-shaped compostable bioplastic tray (Polylactide-PLA Ingeo) packed into compostable film bags)	–	Fresh-cut tomatoes	↑ Antioxidant activity Retained total solids, total soluble solids, sugars during storage	[106]
Oxidized cellulose (dialdehyde cellophane)	Cellophane (1.5%) $NaIO_4$ (75 mM) pH 6 Reaction time (4 h)	Strawberries Tofu	↓ Bacterial count ↑ Acceptance	[107]

PLA polylactide

activity in inhibiting bacterial growth of Gram-positive *S. aureus* and Gram-negative *E. coli*, indicating the feasibility of utilizing microorganisms to produce nanocomposites for biomedical materials and food. In another research, a new nanocomposite film based on chitosan and bacterial cellulose nanofibrils was fabricated by a fully green casting method [122]. Renewable nanocomposite materials showed low oxygen permeability, reasonable thermal stability, and better mechanical properties. Similarly, "carbon quantum dots-titanium dioxide" (CQD-TiO_2) NPs were added to bacterial cellulose to fabricate antibacterial nanocomposites. The bacterial cellulose/CQD-TiO_2 nanostructure displayed antibacterial effects against *S. aureus* [123].

The bacterial cellulose/ε-polylysine composite has been developed by Zhu et al. [124]. The composite showed no significant loss of antimicrobial effect after autoclaving (30 min at 121 °C) and its oxygen permeability considerably lower than that of polyvinyl alcohol and polyethylene membranes. Moreover, the bacterial cellulose/ε-polylysine composite showed bacteriostatic and bactericidal effects against a variety of Gram-negative and Gram-positive bacteria, and it was able to extend the shelf-life of sausages. Nguyen et al. [125] used nisin-enriched bacterial cellulose films as antimicrobial packaging to inhibit the growth of total aerobic bacteria and *Listeria monocytogenes* on the surface of vacuum-packaged frankfurters. In another study conducted by Yordshahi et al. [117], bacterial nanocellulose nanocomposite film enriched with postbiotics of lactic acid bacteria was able to prevent the growth of *L. monocytogenes* in fresh ground beef. Furthermore, bacterial cellulose/guar gum/polyvinylpyrrolidone/carboxymethyl cellulose films have been developed for the packaging of fresh berries. Due to their restricted oxygen and WVP, the film maintained the texture and color of the berries for 15 days [126]. Bacterial cellulose-based smart packaging systems have been also successfully used as pH indicators for freshness/spoilage monitoring of various foods [127].

It has been also reported that the incorporation of bacterial crystalline nanocellulose and silver NPs into chitosan dispersion provided nanocomposite films with enhanced barrier, tensile, and antibacterial properties [120]. The tensile and barrier attributes of corn starch films have been also increased by the addition of bacterial crystalline nanocellulose [128]. Additionally, the polyvinyl alcohol nanocomposite films incorporated with bacterial crystalline nanocellulose experienced an improved TS, elastic modulus, and thermal stability in comparison to the polyvinyl alcohol-based films [119]. The different forms of bacterial cellulose could be therefore used to develop biodegradable packaging materials for use in the food industry.

Generally, a combination of bacterial cellulose and other components could be used in food packaging (Fig. 4). It is possible to immerse bacterial cellulose membranes in dispersions of other components (Fig. 4A). This method avoids membrane breakdown, which is its main advantage leading to a simplified procedure. It is also possible to disassemble bacteria's cellulose membranes (using physical or chemical methods) to use them in formulations with other components. Films are produced using this approach (Fig. 4B). The formation of "bacteria cellulose nanocrystals" (BCNC) is usually accomplished by acid hydrolysis, which eliminates most of the amorphous cellulose parts while keeping needle-like crystals of nanosized diameters. These can be used to reinforce films and coatings (Fig. 4C). Lastly, a composite can be formed in situ by incorporating other components (usually a polymer) into the culture medium (Fig. 4D) [129].

Gellan gum-based composites

Gellan gum is an anionic exopolysaccharide made by the bacteria *Sphingomonas elodea* and has many properties in common with alginate, starch, carrageenan, and xanthan gum [131]. It is composed of a complex tetra-saccharide repeating unit of D-glucose, D-glucuronic acid, and rhamnose [132]. The low cytotoxicity and biocompatibility of gellan gum make it a popular food additive used as an stabilizer, gelling agent, and thickening agent [133, 134].

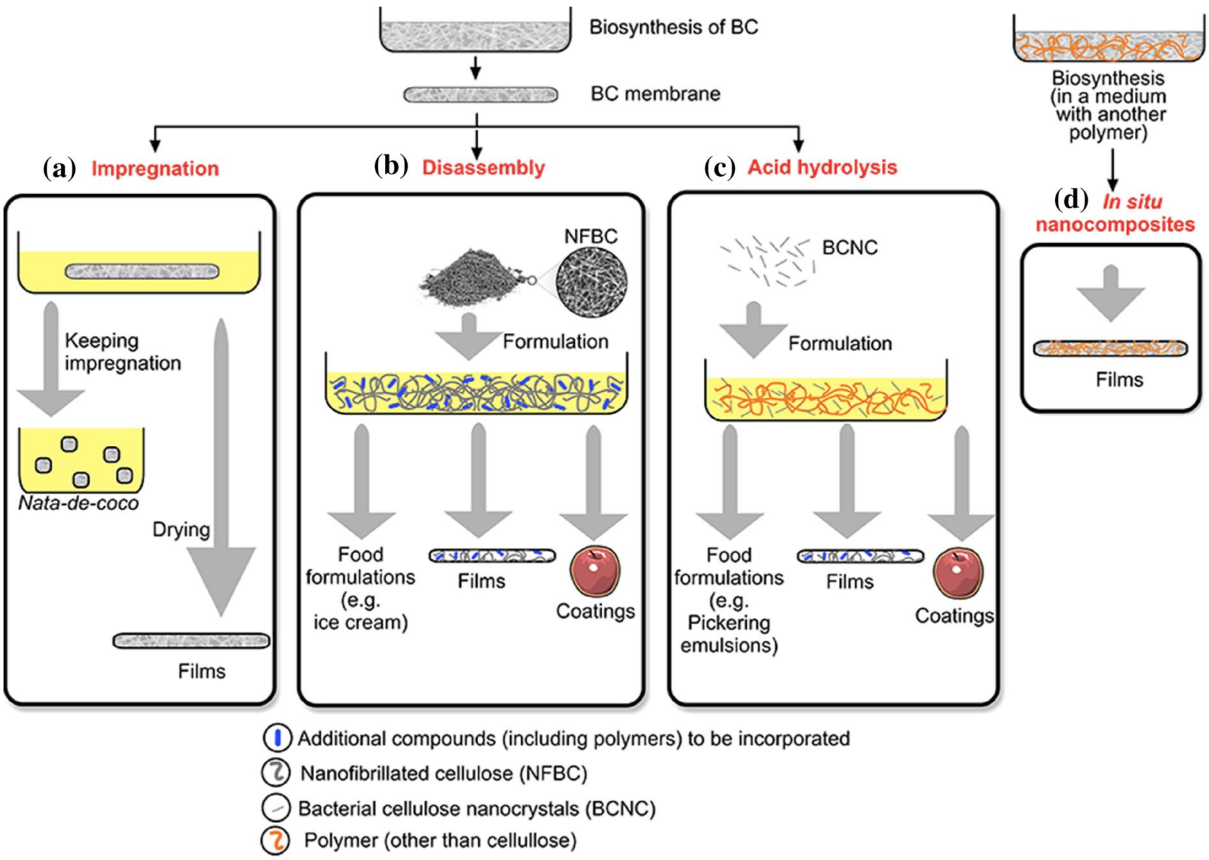


Fig. 4 Techniques for mixing bacterial cellulose with other components for use in food packaging, **A** Impregnation; **B** Disassembly; **C** Acid hydrolysis and **D** In situ nanocomposites. Adapted from [130], reproduced with the permission from Elsevier

The k-Carrageenan/xanthan gum/gellan gum nanocomposite films containing TiO_2 NPs were developed as new packaging materials and showed enhanced TS, thermal stability, tensile modulus, glass transition temperature, and low vapor permeability. Moreover, the nanocomposites effectively shielded the UV light, and showed partial antimicrobial property against *S. aureus*. Thus, these nanocomposites packaging materials can provide UV protection in the food and non-food sectors [135]. Razali et al. [136] developed gellan gum-TiO_2 nanotubes-based bio-nanocomposite films with high TS, Young modulus, and antibacterial activity against *Streptococcus, E. coli, Pseudomonas aeruginosa*, and *S. aureus*. Similarly, zinc oxide NPs were added to the gellan gum/xanthan gum nanocomposite, increased the glass transition temperature, thermal stability, and TS and decreased the WVP of the nanocomposite films, which can be used in food and pharmaceutical packaging [137]. Similar findings have been reported by Balasubramanian et al. [138], who studied the barrier, rheological, and antimicrobial properties of gellan gum-based ternary nanocomposite films including zinc oxide NPs and polyacrylamide. The gellan gum-based nanocomposite films could be utilized for biodegradable packaging purposes.

Starch-based films

As a semi-crystalline polymer, starch consists of 1,4-α-D-glucopyranosyl repeating units: amylose and amylopectin. The availability, biodegradability, and low cost of starch-based materials have made them increasingly popular in food packaging. However, starch has some drawbacks, including poor mechanical features and moisture barrier (strong hydrophilic behaviour) to traditional non-recyclable plastic films commonly used in food packaging [83, 139]. Nanocomposite concepts have recently emerged as a viable approach for improving packaging films' mechanical and barrier properties (Table 3).

On this point, the waxy starch nanocrystals in cassava starch were used to reinforce nanocomposites of cassava starch and characterized by Nancy et al. [140]. The rubbery storage modulus of the nanocomposites was increased by 380%, and the WVP was decreased by 40%, due to the strong filler/matrix interactions based on hydrogen bonding. Furthermore, it has been reported that reinforcement agents such as cellulosic fibers could improve the mechanical resistance of starch-based films [141, 142] or clays [83, 143, 144]. In this way, kaolin (a hydrated aluminosilicate) served as a booster in a thermoplastic starch matrix by De Carvalho et al. [143]. Incorporating kaolin into thermoplastic starch led to an around 130% and 50% increase in the TS and elastic modulus of the film, respectively. In addition, kaolin resulted in a significant decrease in moisture uptake in comparison with the pure starch matrix. In another study, it has been reported that the mechanical and physical properties of conventional composites or pure polymer can be remarkably improved by the nanometer-size dispersion of polymer–clay nanocomposites [145]. There have been similar results reported by Tang et al. [146] who worked on the mechanical resistance and biodegradability of starch/poly (vinyl alcohol) biodegradable film embedded with silicon dioxide NPs. Nanoparticles are also able to decrease the WVP of starch-based films. In this context, the addition of ZnO NPs to the pea starch composite has been shown to significantly reduce the WVP of the final nanocomposite [147].

It is also worth noting that biodegradable thermoplastic materials in the food packaging industry need the possibility to add antioxidant and/or antimicrobial agents to enhance the shelf-life of food products. Starch films are outstanding intermediates for transporting these preservatives. Even though incorporating organic compounds such as bacteriocins, lysozyme, essential oils, and organic acids is effective in ameliorating a product's safety and quality, the mechanical and barrier features of the films are typically depleted. Accordingly, bioactive nanofillers with antioxidant and antimicrobial features have been applied to solve this problem [154–156]. On this point, Chang et al. [157] reinforced the glycerol plasticized starch matrix films with chitosan NPs, and the nanofiller increased the TS, storage modulus, and thermal stability of the film, mainly due to the nanofiller/matrix interactions induced by the uniform distribution of the nanoparticles. Similarly, Shapi'i et al. [158] reported that the addition of chitosan nanoparticles to the starch matrix resulted in nanocomposites with enhanced tensile strength (7.96-fold) and EB (0.35-fold) and decreased WVP (fourfold) and oxygen permeability (0.51-fold). The starch/chitosan nanoparticle nanocomposites could be therefore employed as food packaging materials.

In this regard, it has been shown that the addition of silver NPs to the gelatinized starch suspension led to nanocomposite films with increased thickness and opacity, and lower WVP, while maintaining the UV-barrier and heat-sealing capacity of the matrix. The silver nanoparticle loaded nanocomposite films prevented the growth of *Salmonella spp.* and *E. coli* and were able to prolong the fresh cheese samples' shelf-life for 21 days [151]. Therefore, starch-based nanocomposites can be promising candidates to use in food packaging to improve the shelf-life of various food products.

In conclusion, the substantial environmental damage caused by plastics has made environmental studies and methods to eliminate and control plastics a priority. Recyclable biopolymeric materials from natural sources are the best solution for synthetic plastic waste [159]. However, pure biopolymer packaging has poor properties, and blending it with nanofillers can improve its barrier, thermal, antimicrobial, and mechanical properties. Besides,

Table 3 Starch-based nanocomposite films

Films	Process conditions	Properties	Food application	Results	References
Thermoplastic starch (TPS)/carbamide/ ethanolamine/organic activated montmorillonite (OMMT)	TPS (62–70% w/w) OMMT (0–8% w/w) Carbamide (15% w/w) Ethanolamine (15% w/w) Twin-screw extruder at 130 °C	↑ Tensile strength ↓ Elongation at break	–	–	[148]
Starch (rice starch and waxy rice starch)/chitosan/silver nanoparticles	Chitosan (1.2%) Starch (2%) Glycerol (20%) Silver nanoparticles (1.806%) Casting method	↑ Tensile strength ↑ Oxygen gas barrier ↑ Antimicrobial effect ↓ Water vapor barrier	–	–	[149]
Starch/poly(vinyl alcohol)/graphene oxide/silver	Poly(vinyl alcohol (4%) Graphene oxide (0.5%) Starch (50%) $AgNO_3$ (0.1%) Casting method	↑ Tensile strength ↑ Thermal stability ↑Antimicrobial activity	–	–	[150]
Starch/silver nanoparticles	Corn starch (3% w/v) Silver nanoparticles (5, 10, 25 and 50 μM) Glycerol (30% w/w starch) Casting method	↑ Thickness ↑ Opacity ↑Antimicrobial activity ↓Water vapor permeability	Cheese	↓ Microbial count ↑ Shelf-life for 21 days	[151]
Starch/ZnO-chitosan nanoparticles	Starch (3% w/v) Glycerol (30% w/w starch) Citric acid (10% w/w starch) ZnO-chitosan nanoparticles (1–5% w/w starch) Casting method	↑ Tensile strength ↑Antimicrobial activity ↓Water vapor permeability	–	–	[152]
Starch/salicylic acid/waxy maize starch nanoparticles/κ-carrageenan	Hydroxypropyl cassava starch (5% w/v) Salicylic acid (0–6% w/w of hydroxypropyl cassava starch) Glycerol (2% w/v) waxy maize starch nanoparticles/κ-carrageenan (0, 1, 3, 5, 7, 9% of hydroxypropyl cassava starch) Casting method	↑ Tensile strength ↑ Thermal stability ↑ Water vapor barrier ↑Antimicrobial activity ↓ Transparency ↓ Elongation at break	Yogurt	↓ Microbial growth	[153]
Starch/chitosan nanoparticles	Corn starch (5% w/v) Chitosan nanoparticles (1–4% w/v) Glycerol (1.5% w/v) Casting method	↑ Tensile strength ↑ Young's modulus ↑Antimicrobial activity ↓ Elongation at break ↓Water vapor permeability ↓Water vapor transmission rate	–	–	[4]

the migration of nanomaterials from packaging into food and their toxicity are also concerns that have been demonstrated. Therefore, incorporating green synthesized environmentally friendly nanomaterials into biopolymer is extremely promising. The following section explains how green synthesized nanocomposites can be applied to biodegradable food packaging.

Application of green synthesized nanocomposites on biodegradable food packaging

Compared to pure biopolymer, the nanoparticles incorporated into the biopolymer matrix significantly improve nanocomposites' functions in terms of physicochemical, thermal, and optical properties [160]. The reason is that nanoparticles have high surface energy and area and can improve interfacial interaction and nano-scale dispersions in biopolymer matrices. For example, the polarities and structural characteristics of polymeric side chains, their polydispersity, hydrogen bonding, molecular weight, crosslinking, and crystalline nature can affect biopolymer barrier properties. In general, the number of free volume holes in a polymer film, the degree of polymer movement, the interactions between individual polymers, and the interactions with gases all contribute to its permeability. The barrier features of the homogeneous polymer film are altered/influenced by adding nanosized fillers to the polymer matrix [161]. Due to the impermeability of nanomaterials, which prevents gas molecules from finding a straight passage perpendicular to the surface, they are forced to move around filler materials. Green chemistry has been confirmed to be an energy-saving path for nanoparticle synthesis. The chemical synthesis methods use hazardous and toxic substances (e.g., sodium borohydride, hypophosphite, and hydrazine hydrate) intensively. So, developing a laboursaving/economical and sustainable green method for the synthesis of NPs is a continuous effort. Researchers have recently focused on incorporating green synthesized nanomaterials into biopolymers to enhance their mechanical, barrier, optical, thermal, and antimicrobial properties. In the following sections, we will address in more detail the recent application of green synthesized nanomaterials to biopolymers.

Green synthesized metal/metal oxide nanoparticles incorporated in food packaging

Metal/metal oxides such as Ag_2O, CuO, ZnO, CaO, TiO_2 and MgO are among the most widely utilized nanomaterials in various industries. At the nanoscale size, the particles present unique properties that are different from their properties in bulk quantity [162]. The unique properties of metal/metal oxide nanomaterials are their catalytic properties, UV absorption, selective activity, physicochemical properties, high surface area, great flexibility at high temperatures, sensitivity, and high mechanical and biological properties [11]. Several methods and strategies have been used to synthesize metal oxide nanomaterials, including physical, chemical, and biological processes [163]. Though the chemical and physical techniques use less time to manufacture bulk quantities of metal/metal oxide NPs, they have many disadvantages, including low production yield, high energy loss, high heat generation, and expensive equipment. The core issue with these approaches is the use of toxic chemicals, such as protective reagents to maintain stability, which limits their biocompatibility and imposes many environmental problems. Hence, recently, the "green synthesis" of metal/metal oxide NPs has gained extensive attention from researchers and manufacturers. The advantages of green synthesis processes over conventional physical and chemical processes include their affordability, simplicity, safety, lack of harmful ingredients, accessibility, and renewable nature [14]. Therefore, the increase in the synthesis of metal/metal oxide/bimetallic NPs using green approaches is incredibly promising [164]. Green synthesized metal/metal oxide NPs have diverse applications such as antimicrobial, target recognition, biomedical, targeted drug delivery, biosensor and food packaging applications [165]. This part reviews recent studies to better understand green synthesized metal/metal oxide nanomaterials-based incorporated food packaging. The main attraction of the food packaging industry has always been the use of appropriate packaging materials and techniques to reduce food losses and provide healthy and safe food products [166]. Nanocomposite technology has been one of the most interesting areas in food packaging fabrication due to the tunable properties of nanocomposite packaging materials. Kumar et al. [167] developed a nanocomposite film-based chitosan and gelatin containing green synthesized zinc oxide (ZnO) NPs. Green synthesized ZnO NPs (range of 20–40 nm) were obtained from fruit extract of Cassia fistula. According to the results, nanocomposite films loaded with green synthesized ZnO NPs showed improved thermal stability, EB, and compactness properties.

Compared to the control (chitosan–gelatin hybrid) films, the developed films with 2% and 4% ZnO NPs showed a heterogeneous, compact, and smooth surface morphology. Nanocomposite film indicated remarkable antimicrobial activity against *E. coli*. In another work reported by Marrez et al. [168] an environmentally safe nanocomposite film based on cellulose acetate incorporated with green synthesized silver NPs was developed for using it as an antibacterial food packaging for some foodborne pathogenic bacteria. Green synthesized silver NPs (Ag NPs) were synthesized from polyphenols such as gallic and pyrogallic acid, rutin, and quercetin. All prepared AgNPs had activity against the examined pathogenic bacteria and the nanocomposite film showed high antimicrobial activity. The authors suggested that the nanocomposite film had an intense antibacterial activity with a nonhazard effect, increasing the chance of using it as an active food packaging system. Basumatary et al. [169] developed an agar-based nanocomposite film loaded AgNPs as filler. The aqueous fruit extract of *Lagerstroemia speciosa*, a medicinal plant widely used in northeastern India, was used to synthesize AgNPs. The results showed that the incorporation of AgNPs improved the EB, antibacterial properties, thermal stability, and semblance of nanocomposite films, while TS was reduced. In addition, the orange-brown hue of the composite films can protect the packaged food from UV rays. Gudimalla et al. [170] developed sodium alginate films doped by green synthesized Ag NPs using *Nymphae odorata* plant extract and examined the antimicrobial activity of the green synthesized Ag NPs and the doped films. According to the result, a very low concentration of Ag NPs was found to inhibit the entire bacterial activity of *Staphylococcus aureus* and *Escherichia coli*, and films also depicted a similar effect. The results confirmed the efficacy of the prepared Ag NPs and films as antibacterial agents. In another study, the effect of green synthesis Ag NPs with oregano essential oil (OEO) on polyvinyl alcohol PVA/starch film was investigated by Srikhao et al. [171]. Their results showed that the films containing NPs and 5 wt.% OEO had the lowest concentration of inhibitory against *E. coli* and *S. aureus*. Moreover, the mechanical properties and the water resistance of films were enhanced with the addition of Ag NPs with oregano essential oil. This study proved that the loading of green synthesized Ag NPs and OEO enhanced the efficiency of bioactive nanocomposite film for food packaging applications. Cheviron et al. [172], prepared the Ag NPs by an ex-situ method via a green chemistry process. The green synthesized NPs were then incorporated into the potato starch matrix. The authors claimed that the prepared nanocomposite films could be considered promising candidates for many applications in antimicrobial, biomedicines, and sensors. Figure 5 shows the development of a nanocomposite film loaded with green synthesized Ag NPs as fillers for food packaging applications.

Green synthesized carbon dots incorporated in food packaging

Carbon quantum dots (CDs), one of the newest class of carbonaceous nanomaterials with a diameter of less than 10 nm, have been reported to have several unique/desirable properties, such as low cytotoxicity, excellent photostability, outstanding biocompatibility, ease of functionalization [173], supreme antibacterial and antioxidant activities [174], water solubility, in addition to the usual advantages/features of nanomaterials. Therefore, CDs are widely applied in various scientific and technological fields [175, 176], such as sensors/biosensors [177], cell imaging [178, 179], energy sector [180], anti-counterfeiting applications [181], nanomedicine [182] and so on. The aforementioned characteristics of CDs and their promising applications have opened new avenues for their applicability in the food packaging/preservation industry. Accordingly, there are several systematically-conducted pieces of research/studies in the literature [174, 183] showing the great potential of these unique nanostructures as ideal nanofillers/additives to enhance the thermal, mechanical, physical, UV barrier, and water barrier properties of food packaging, as well as their potential to fabricate active packaging [174, 184], with desired antibacterial/antioxidant properties [185], and smart packaging with the ability to detect food freshness/spoilage/quality [186]. A detailed review of the literature clearly shows that, in recent years, there has been a significant increase in the application of green/bio-synthesized CDs for the fabrication of bio-nano composite films/coatings with multiple functions (pH sensing, UV-shielding, antioxidant, antibacterial, etc.) [187–189]. The application of green(bio) synthesized CDs is developing rapidly, but it remains a challenge to ensure the consistency of their physicochemical properties (e.g., fluorescence quantum yield) compared to conventional manufacturing processes. This challenge has impacted their use in food packaging but has also enabled the development of more economical and biocompatible nanocomposite films/coatings that can be used in the smart food packaging industry. For example, Zhao et al. [190] recently studied the effect of green synthesized CDs (derived from banana) in combination with aqueous chitosan (CH) solution on the shelf life and stability of soy milk. In this study, soy milk samples were prepared with different concentrations of chitosan solution (“0.00%, 0.08%, 0.12%, 0.16%, and 0.20%”) and

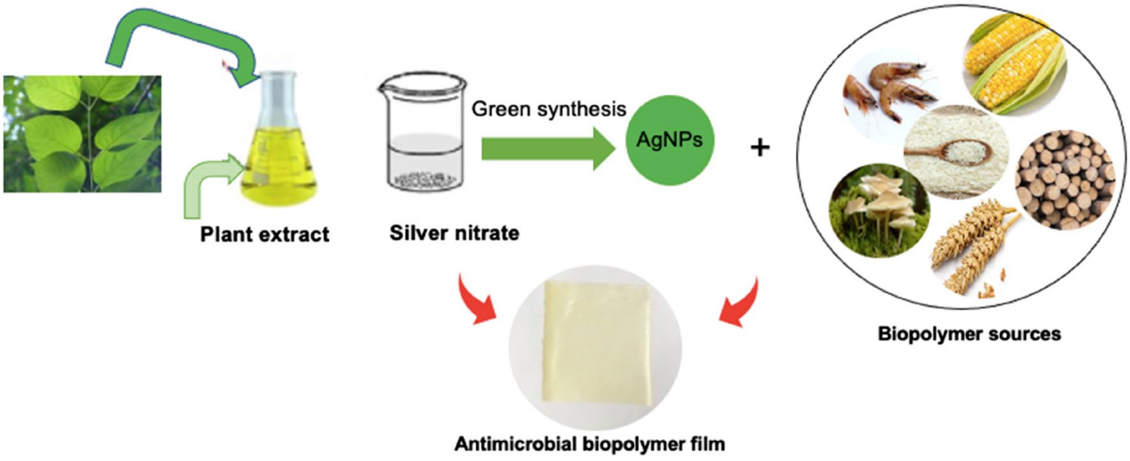


Fig. 5 Procedure green synthesis of AgNPs for incorporating into biopolymers

banana-based CDs ("4%, 6%, and 8%") to evaluate the shelf life at room temperature. According to the reported results, the optimized amount of the green synthesized CDs could effectively inhibit the growth of *Bacillus subtilis, Staphylococcus aureus,* and *Escherichia coli*. After being stored at room temperature for up to 4 days, soy milk samples containing 0.16% chitosan and 8% CDs displayed a longer shelf life and a noticeably lower total bacterial count. They found that bacterial growth decreased from 100 to 0% when the content of CD increased from 0 to 10%. Similar to liquid media, *B. subtilis, S. aureus*, and, *E. coli* also grow on solid media; colony counts gradually decline as CD content rises. (Fig. 6a and b). Only a small number of bacteria were able to proliferate on the solid plates containing 8% CDs (61, 11, 8 for *B. subtilis, S. aureus*, and, *E. coli,* respectively). At a 10% CD content, there were almost no colonies of any type of bacteria on the plates (Fig. 6b). Figure 6c shows the changes in the appearance of the tested soymilk samples stored at room temperature. As can be seen, precipitation was observed in the control sample after two days of storage, while the soy milk samples showed better storage stabilities. The color of the control sample and the sample containing 0.16% CH altered after four days of storage, however, no color change or precipitation was observed in the sample containing 0.16% CH + 8% CDs. Overall, the results suggest that CDs combined with an aqueous chitosan solution can effectively extend the shelf life and improve the stability of soymilk and can be considered as a potential composite combination and an alternative to conventional heat treatment to extend the shelf life of foodstuff. In another study, Koshy et al. [191] reported the successful green synthesis of nitrogen-doped CDs using soy protein isolate (SPI, as a natural source) and incorporated them in a starch matrix containing anthocyanin extracted from *Clitoria ternatea* flower (CTE) to produce an intelligent and cost-effective biopolymer matrix that was used to monitor the freshness of packed pork samples at ambient conditions. According to the authors, the addition of CDs reduced the water sensitivity and increased the mechanical strength of the film. The addition of green synthesized CDs also increased the thermal stability of the starch film. As a result of the intermolecular hydrogen bonding that occurs among the functional groups in CD and the matrix, thermal stability increases. The produced films (starch/CD/CTE film) under the optimized conditions were used to check the freshness of stored pork due to the ability of the developed pH indicator films to change color with pH variations.

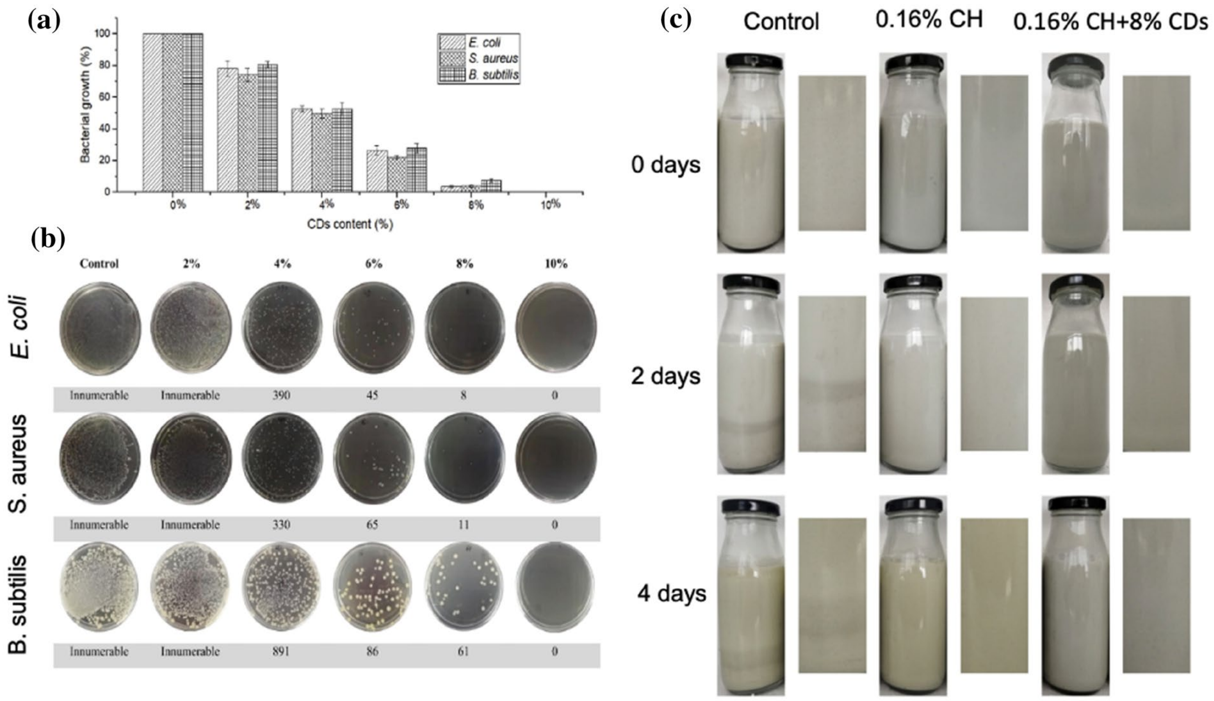


Fig. 6 The results of antibacterial properties of green synthesized CDs and alteration observations in the quality of soy milk samples kept at ambient temperature upon treatment with chitosan and chitosan + CD bio-nanocomposites: **a** Histogram of bacterial growth as a function of CD concentration after 24 h incubation in the liquid medium, **b** Photographs of *B. subtilis, S. aureus*, and *E. coli* colonies on solid medium (with varied CD contents) after 24-h incubation (Bacterial colony counts are represented by the numbers below the photographs), and **c** Variation of different samples stored at room temperature for 0, 2, and 4 days. Adapted from [190], reproduced with permission from Elsevier

In a recent report, Fan and co-workers [192] investigated the effect of green synthesized CDs incorporated into a CH matrix to produce coatings on microorganisms and their impact on the storage quality of fresh-cut cucumbers packed in a modified atmosphere. In this study, the inhibition zone diameter approach has been used to investigate the antibacterial efficacy of the CH and CDs nanocomposite against *S. aureus* and *E. coli*. The results showed that at 4.5% CDs and CH, the inhibition zone diameters against *S. aureus* and *E. coli* were much larger than those at 0% CDs and CH. This observation indicates that antibacterial activities increased with increasing CDs/CH coating with the concentrations of CDs ranging from 0 to 4.5%. It was also found that the diameter of the inhibition zone of the CD/CH coating at a concentration of 4.5% CD was larger against *S. aureus* than against *E. coli*. Weight loss study results showed that the diminish in weight loss for the samples coated with CDs/CH was significant compared with the control samples. As a result of storage, the 4.5% CD/CH coating lost the least weight, primarily because adding abundant functional groups to the CH solution resulted in increased surface crystallization of the CD/CH coating, creating good barrier properties to reduce WVP. In addition, in this study, the coating with 4.5% CDs/CH was found to reduce firmness well, maintaining the content of TSS and ascorbic acid, inhibiting the activity of POD, and retaining flavor. Coating with the optimized CDs/CH can also reduce water mobility. Overall, this study proved that the CDs/CH nano-biocomposites can extend the preservation time (up to 15 days) of fresh cucumbers packed under a modified atmosphere. These promising results could be applied to other vegetables.

In another interesting study, Patil et al. [193] prepared a highly flexible (Fig. 7a–f), transparent, and re-emissive composite film from polyvinyl alcohol and tea waste-derived CDs (synthesized from tea waste residue powder as a sustainable material, PVA@WTR-CDs). They found that the higher the concentration of WTR-CDs in PVA films, the higher the UV blocking ability of the composite films (absorption of UV light and reemission in the visible range). The PVA@WTR-CDs composite films with the optimized concentration of CDs can block the 100% of UV-C and UV-B regions and 20–60% of UV-A region. According to the observations, PVA@WTR-CDs composite films' maximal UV blocking was achieved by adjusting the films' transparency and thickness. Also, by gradually adding the UV blocker, no drastic changes in the inherent mechanical characteristics and tensile strength of the PVA films were noticed. The practicality of the composite film was also investigated on a model fruit (grape). As shown in Fig. 7(g), the applicability experiments in this study were performed in two groups for 30 h under the UV lamp. Cups A, B, and C contain equal numbers of grapes, and cup A was taken without wrapping the film, while cups B and C were wrapped with the untreated PVA film and PVA@WTR-CDs composite thin film, respectively.

Figure 7g reveals that significant, eye-perceptible changes in grape color and shape occur only after fifteen hours of UV light exposure. Grapes in cups A and B were slightly brownish compared to grapes in cup C. The grapes in cups A and B were slightly brownish compared to the grapes in cup C. With continued UV irradiation for up to 30 h, the grapes in cups A and B became browner and more shriveled than those in cup C. All finding reveal that the grapes in cup C, wrapped with composite film (PVA@WTR-CDs), were not damaged by the UV light. These observations demonstrate that the composite film can be considered the next generation UV-blocking composite materials and can be practically used for food packaging, other UV-blocking applications, pharmaceutical storage, wrapping, coating, etc.

A continuous effort is being made to develop a sustainable green and cost-effective method for the synthesis of nanoparticles that replaces chemical synthesis methods. A green-synthesised nanomaterial can simultaneously improve biopolymer properties while reducing toxicity from chemical-synthesised nanomaterials. Last but not least, the potential use of by-products, waste and environmentally friendly (green/biological) sources as feedstock/starting material for CDs synthesis could increase sustainability and reduce costs in the food supply chain, and perhaps provide various new functions/applications for smart food packaging industry in the near future.

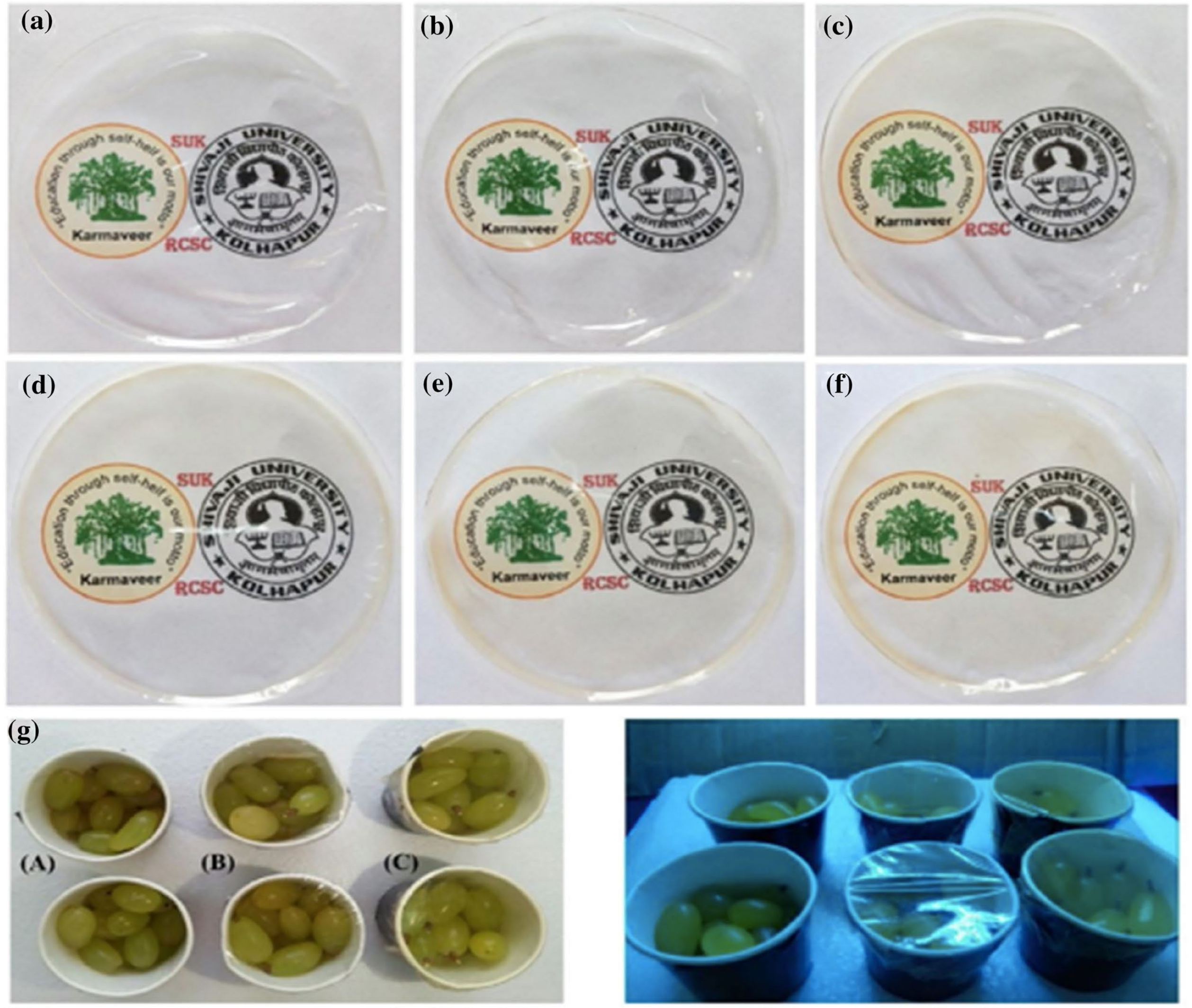
Current challenges and future perspectives

The unique characteristics of nanomaterials, such as their high surface area, make them ideal for higher process efficiency. Nevertheless, there are some serious drawbacks to consider.

A material functionalized with nanoparticles incorporated or deposited on its surface, for instance, has the potential to cause a health risk. Nanomaterials can release into food products, which is necessary to assess potential exposure to nanomaterials in food packaging, which is a major challenge due to the lack of analytical tools and methods. Several regulations and laws are being established at the national and international levels to minimize health risks.

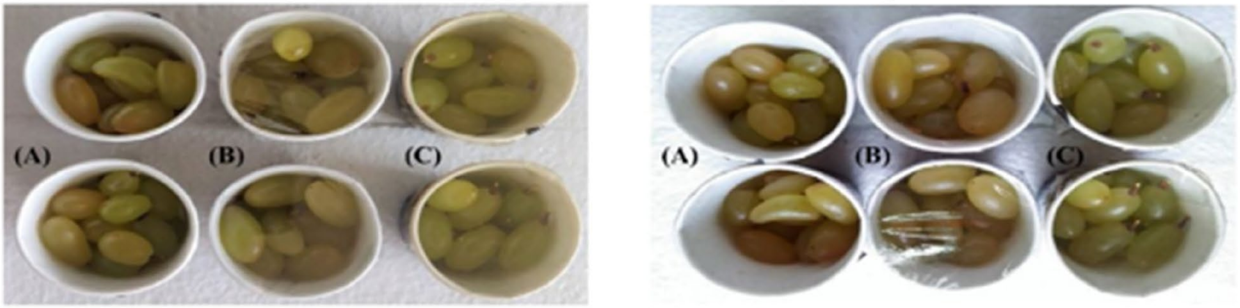
Since hazardous chemicals and materials are being employed to produce nano-materials, chemical companies have been pressured to substitute toxic reagents and harmful solvents.

The development of safer and earth-abundant nanoengineered materials is expected soon. It is very promising to develop biogenic nanoparticles due to their excellent efficiency in reducing environmental contaminants and the inherent greenness in their production methods. Due to the



(1) Image before exposure to UV at 0 hr

(2) Image under UV



(3) Image before exposure to UV at 15 hr

(4) Image under UV exposure after 30 hr

Fig. 7 Photographic images demonstrating the transparency of PVA films without any additive: **a** PVA@WTR-CDs composite films, **b–f** as the concentration of WTR-CDs rises (0.5, 1.0, 2.0, 3.0, 5.0 mg), and **g** Digital images showing the efficacy of produced composite green synthesized CD-based films in food packaging applications when exposed to UV light at different time intervals. Cups **A** without films, **B** wrapped with untreated PVA films, and **C** wrapped with PVA@WTR-CDs composite films. Adapted from [193], reproduced with permission from Elsevier bio-renewable nature of biomaterials, they are sustainable, relatively inexpensive, energy-efficient, and ecologically reliable, and they could substantially improve bio-packaging properties.

Several critical future perspectives should be considered regarding the application of biogenic nanomaterials into biopolymers. There must be further study on sustainability and toxicity issues before these green-synthesised nanomaterials can be applied on an industrial and commercial scale. In addition, since nanomaterials' behaviour, morphology, and properties can be altered by reaction parameters and stability issues, it is crucial to analyze and optimize some critical and challenging aspects of nanomaterial production. Besides, green-synthesised nanomaterials should be compared to conventionally prepared nanoparticles based on cost-effectiveness studies.

Conclusions

Despite the valuable properties of NPs, including antimicrobial, thermal, and mechanical stability, due to their toxicity, their use is limited at high concentrations. Therefore, the green biosynthesis of NPs by plants and microorganisms is considered environmentally acceptable and could help reduce the associated toxicity. New packaging materials that perform better can be produced by incorporating nanotechnology into food packaging. According to the results, nanocomposite materials have considerable potential for enhancing food packaging quality and safety. That can be achieved through low concentration incorporation of nanocomposites affecting the packaging morphological, barrier, optical, mechanical, physical, and thermal properties. However, due to the potential migration of NPs from the packaging material into food, the most critical issue is NPs' safety and toxicological effects. Green synthesized NPs could be a promising option to overcome this issue. The present article reviews the general concepts of green synthesis of NPs, their production methods, their performance, and their characterization. It also covers the role and influence of green synthesized NPs in modifying the properties of food packaging (biopolymer) regarding their performance. It is important for future research and development of 'green' materials and nanoparticles to extend from laboratory-based work to the industrial scale while continuing to address traditional and present concerns, particularly in health and environmental protection. While 'green' material and nanoparticle synthesis are likely to be applied extensively in essential areas such as pharmaceuticals, food, and cosmetics, it is also likely to be used extensively in other critical areas, including environmental remediation. Furthermore, marine algae and marine plants are mainly unexplored for their ability to biosynthesise metals and their oxide materials and nanoparticles. Thus, biogenic synthesis gives rise to several possibilities for green preparation strategies.

Acknowledgements Funding from the Australian Research Council (ARC DECRA G1005399) is gratefully acknowledged.

Funding Open Access funding enabled and organized by CAUL and its Member Institutions.

Data availability No research data were shared.

Declarations

Conflict of interest The authors declare no conflict of interest.

Ethical approval There was no need for ethics approval for the present research.

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