Sustainable
Food Technology

ROYAL SOCIETY
OF CHEMISTRY

REVIEW

View Article Online
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Cite this: DOI: 10.1039/d5fb00677e

The state-of-the-art on chitosan: historical perspectives and non-thermal modification technologies for sustainable packaging films

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Chitosan has garnered significant attention in the field of polymer science over the decades due to its associated characteristics, such as biodegradability, antimicrobial properties, and film-forming abilities. With increasing concerns, extensive research has been conducted to enhance the functional properties of chitosan using various modification techniques. Recently, the modification of chitosan-based materials using non-thermal technologies has drawn attention due to their ability to improve mechanical strength, barrier properties, and bioactivity without compromising environmental sustainability. This review discusses the historical discovery and development of chitin and chitosan and further emphasizes their growing significance in biomaterials science. A key focus of this study is to apply non-thermal modification technologies, such as cold plasma, irradiation, high pressure processing, ultrasonication, pulsed electric fields, and electrospinning, for the modification of chitosan-based materials for their application in active and intelligent packaging systems and shelf-life extensions, thereby aligning with the principles of green chemistry. Non-thermal modification technologies significantly enhance the structural and functional properties of chitosan films, making them viable alternatives to conventional plastic packaging. The mechanical properties, antimicrobial efficacy, oxygen barrier properties, and thermal stability of chitosan are among its key benefits, and the proposed material has been successfully used as a food packaging material for preserving fresh produce and increasing the shelf life of several types of food products. This approach not only supports the transition towards a circular economy but also contributes to a significant reduction in plastic packaging.

Received 11th October 2025
Accepted 27th March 2026

DOI: 10.1039/d5fb00677e

rsc.li/susfoodtech

Sustainability spotlight

Chitosan, a deacetylated derivative of chitin, has evolved from a waste-derived biopolymer into a promising material for sustainable packaging. Initially extracted from crustacean shells, it is now widely explored in films due to its biodegradability and antimicrobial properties. This review outlines the historical development of chitosan in materials science and highlights the recent advancements in non-thermal modification techniques, such as ultrasound, cold plasma, pulsed electric fields, and high-pressure processing. These methods improve the solubility, film-forming ability, and barrier properties without affecting its natural functionality. This study also integrates sustainability metrics, including life cycle assessment and carbon footprint reduction, linking them with structural and performance characteristics for smart, active, and future-ready food packaging systems.

Introduction

In recent years, fossil-derived polymers have posed significant environmental challenges because they are non-biodegradable in nature, which in turn increases the carbon footprint and toxic pollutants.[1] Additionally, various chemicals are used for the production of synthetic polymers, and some of them are considered carcinogenic and harmful to the environment. Therefore, there is a growing shift towards the use of natural polymers, which are biodegradable and eco-friendly.[2] According to the Institute of Bioplastics and Biocomposites (2016), global bioplastic production grew from 1.6 million tons in 2013 to 2.0 million tons in 2015, indicating a steady increase in the adoption of biodegradable and biobased plastics. In 2019, the global market for biodegradable plastics was valued at about USD 4.65 billion, and it was projected to grow by the end of 2025 at a compound annual growth rate of 17.04%, reaching a market value of up to 12.06 million. Biodegradable plastics such as PLA (polylactic acid) and PHA (polyhydroxyalkanoates) account for over 55.5% of the total global bioplastic production.[3] Bioplastic production is dominated by Asia, which contributes to 63.1%, followed by North America (13.5%), Europe (13.0%), and South America (10.0%). While designing packaging, one of the key attributes of sustainable packaging is biodegradability,

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ensuring that materials break down naturally without causing any environmental hazards. Biodegradability is not only a functional requirement but also a crucial environmental necessity. Biodegradable polymers break down 60% faster than conventional plastics, which can take over 1000 years to degrade in the environment. Biodegradable plastics can fully decompose within 180 days.[4] Non-biodegradability contributes to landfills, plastic waste, and pollution, causing an ecological imbalance. As a result, the use of natural polymers or biopolymers (substances derived inherently from living matter) has gained more attention due to their associated advantages and increasing consumer awareness regarding the use of synthetic polymers.[2]

Among the available biopolymers, chitin is considered the second most abundant biopolymer after cellulose, as first described by Henri Braconnot in 1811. This naturally occurring biopolymer is found in ordered macrofibrils within the exoskeletons of mollusks and crustaceans and in the cell walls of fungi; it is reported that 70% of chitin comes from marine species.[5] However, 'chitin' was formally named in 1823 by Odier, who isolated it from the cuticles of insects. During the 19th and 20th centuries, researchers recognised its structural similarity to cellulose. Chitosan, a deacetylated form of chitin, was first described by Charles Rouget in 1859. Despite these early discoveries, the application of chitin and chitosan remained limited until the mid-20th century. By the late 20th century, chitin and chitosan had gained much popularity due to their non-toxicity, biodegradability, and biocompatibility.

Chitosan, a naturally derived polysaccharide, has gained significant attention across various industries due to its non-toxicity, biocompatibility, excellent film-forming properties, and cost-effectiveness.[6] Chitosan is the *N*-deacetylation product of chitin and is a heteropolysaccharide composed of β-1 → 4-linked glucosamine and *N*-acetylglucosamine units, and it is the second most abundant biopolymer after cellulose.[7] Chitosan is widely used in the medicine, food, cosmetics, biomedical and packaging industries due to its biodegradability, antimicrobial, and other bioactive functions.[6] However, the use of chitosan is limited due to its low solubility, high crystallinity, and limited functional groups for any chemical reaction.[8] Its limited

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solubility in neutral and alkaline solutions poses a significant challenge for broader industrial applications. The rigid crystalline structure of native chitosan prevents it from dissolving in aqueous solvents at pH > 6.5, making functionalization and processing crucial. Moreover, chitosan is insoluble in most organic solvents, restricting its application for various purposes. Therefore, the modification of chitosan has become essential in enhancing its functional properties to expand its applications.[9] The primary methods for chitosan modification include chemical modification, where chemical modification introduces functional groups to improve solubility, reactivity, and bioactivity; however, these methods have several drawbacks. Conventional chemical modifications require harsh reagents that generate toxic by-products, and harsh chemical modification alters chitosan's biocompatibility, degrades its molecular structure, and limits its applications in various sensitive fields, such as pharmaceuticals and food packaging. Additionally, it often requires high temperature and prolonged processing times, making the process energy intensive and costly, leading to low yields.[10] Hence, to overcome these limitations, eco-friendly and sustainable modifications of biopolymers using non-thermal technologies, such as cold plasma, high pressure processing, irradiation, pulse electric field, ultrasonication, and microwaves, have gained significant attention.[11] Social consciousness as a catalyst has been increasing dramatically as the public concern related to plastic pollution, climate change, and an imbalance in the ecosystems has heightened its demand for environmentally friendly alternative solutions and a shift towards greener technologies.[12] These modifications of the natural biopolymer chitosan using non-thermal technologies in contrast to conventional modifications allow for controlled modification without compromising its structural integrity, enhancing functional properties without any degradation or harmful by-products. Additionally, these techniques do not rely on heat, but the use of mechanical-, electrical-, and radiation-based energy to alter and modify their structure and functionality is being explored.[13] Non-thermal technology often allows for targeted modifications, *e.g.* improved solubility, antimicrobial activity, and biocompatibility without any structural breakdown. Chitosan modification using non-thermal technology has been explored for a wide range of applications, such as plasma treatment, which introduces functional groups onto chitosan's surface, thereby improving its properties, such as hydrophilicity, adhesion and biocompatibility.[14] In ultrasonication-based non-thermal technology, ultrasonication time and heat treatment influence the structure and properties of chitosan/graphene oxide tailored films. Hence, sonication improved the UV and visible light barrier properties of the chitosan/graphene oxide films, making them effective for packaging applications.[15] By adopting these methods, industries can reduce their ecological footprint and provide a greener, eco-friendly alternative to conventional modification methods, thereby supporting sustainability, safety, and environmental responsibility.

Based on the above discussion, this review critically discusses recent advances in the non-thermal modification of chitosan, highlighting its impact on the structural, physicochemical, and functional properties relevant to sustainable packaging films. Furthermore, key challenges, knowledge gaps, and future research directions are discussed to support the development of next-generation chitosan-based packaging materials.

History of chitin and chitosan

Polymers are large molecules composed of repeating structural units called monomers and play a crucial role in various scientific and technological fields. Based on their origin, polymers can be classified as synthetic and natural.[16] Among natural polymers, the discovery of chitin over two centuries ago marked the beginning of a scientific journey into one of nature's most abundant and versatile biopolymers. Trailing only in prevalence after 'cellulose', chitin is considered the second most abundant polysaccharide. Over the past 220 years, the study of chitin has progressively evolved from scientific curiosity towards being one of the fundamental elements of biomaterials science, as illustrated in Fig. 1, which traces the historical development of chitin and chitosan from their discovery to the current period.

In 1811, Henri Braconnot, a French chemist, isolated a material from fungi using water, alcohol, and dilute alkali, and named the insoluble residue "fongine" or "fungine". This insoluble residue was later identified as chitin, making Braconnot's discovery the first recorded identification of a polysaccharide from fungi. Braconnot's discovery made a remarkable footprint, highlighting his pioneering contribution towards biochemistry, after the discovery of the most abundant polymer, "cellulose," about thirty years earlier.[17] Further, August Odier, a Swiss physician, in 1823, during his experiment with cockchafer betel, found a distinct substance, hornlike material, and named it "Khiton, Chitine," which means 'envelop' in Greek. Another scientist, Opperman, in 1832, envisaged that chitin was extracted from insects; substances similar to chitin can also be found in the structure of insects.[18] Therefore, another conflict arose between scientists regarding chitin produced by arthropods and cellulose produced by plants. In 1859, French physiologist Charles Rouget discovered that heating chitin made it soluble in organic solvents, leading to the discovery of its derivative, chitosan. As reported by Crini, the era between 1894 and 1930 led to another

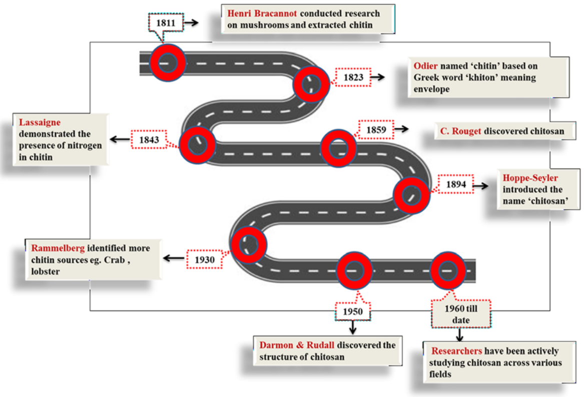

Fig. 1 Historical evolution of chitin and chitosan.

significant controversy about the presence of chitin and cellulose in the cell walls of fungi and their solubility in organic solvents.[17] In 1930, Rammelberg identified more sources of chitin apart from insects and fungi and highlighted that it can be extracted from the exoskeletons of marine arthropods such as crabs, shrimps, and lobsters. Later that year, he made a remarkable contribution by identifying chitin as a polysaccharide of glucosamine. Using polarised radiation spectra, in 1950, Darmon and Rudall identified the structures of chitin, chitosan, and cellulose.[19] From 1930 to 1950, chitin and chitosan gained much attention in terms of their extraction and isolation processes, where the first book was published in 1951, which included research conducted by various scientists on chitosan. Therefore, in the late 1960s, different polymeric forms of chitin were identified.[17] Since 1960, numerous researchers have been actively studying chitosan across various fields. This ongoing research has explored its applications in areas such as biomedicine, agriculture, food technology, and water treatment. The versatility of chitosan, due to its biocompatibility, biodegradability, and non-toxicity, has made it a subject of extensive scientific interest and innovation.[20]

With the advancement of technology and urbanization, concerns have been increasing in materials science regarding the use of sustainable processing methods for the extraction of biopolymers, such as chitosan.[21] The discovery of non-thermal technologies for the extraction of chitosan modification emerged as a response to the need for gentle and more sustainable processing methods, eventually maintaining the biopolymer's delicate structure and bioactivity in comparison to other thermal technologies that can degrade the structure of the biopolymer along with its integral mechanical properties. This era began in the late 20th and early 21st centuries, as materials science and environmental concerns drove the search for alternatives to traditional thermal technologies.

With due course of time, researchers have analysed the use of non-thermal technologies such as cold plasma, irradiation, high-pressure processing (HPP), and ozone treatment, which can effectively alter chitosan's surface properties, molecular weight, functional groups, copolymerization, hydrophilicity, and others, avoiding the potential risk of its degradation associated with heat and other parameters, which makes this biopolymer invaluable in the area of polymer science with its diverse application.[22]

Chitin and chitosan: structural and chemical properties

Biopolymers are the units of monomers derived from biological sources. Their molecular structure can be linear, branched or cross-linked, thereby influencing their properties and applications. Biopolymers are broadly classified into three types depending on their monomeric units: (i) polynucleotides: these biopolymers are composed of repeating monomeric units, *e.g.*, DNA and RNA. (ii) polypeptides: these consist of short polymers of amino acids. (iii) polysaccharides: these consist of linearly bonded polymeric sugar (saccharide) units linked together, *e.g.*, cellulose, chitosan, lignin, hemicellulose and glycogen. Polysaccharide-based polymer membranes have gained significant attention in food packaging due to their non-toxicity, biodegradability and excellent film-forming abilities. These characteristics make them a potent candidate for use as a sustainable alternative to plastic-based packaging, thereby focusing on sustainable development goals.[23]

The last decades can be considered the golden era for the most abundant biopolymer chitin (CT), which is present in nature and its *N*-deacetylated form chitosan (CS), due to its extensive and diverse applications in various sectors.[24] The second most abundant biopolymer present after cellulose, *viz.* 'Chitin', can be defined as the abundant mucopolysaccharide present in nature, which is hard and inelastic and is made up of nitrogenous compounds.[5] Further, unlike any other polysaccharide, such as glycogen or starch, being considered a 'storage polysaccharide', chitin is considered a 'structural polysaccharide'.[25] In this regard, an overview of chitosan sources, extraction, and modification techniques is illustrated in Table 1. Compared to conventional chemical routes, emerging non-thermal and green modification techniques, such as microwaves, ultrasonication, cold plasma, and pulsed electric fields, offer reduced processing times, lower chemical consumption, and improved functional efficiency. These methods also minimize environmental burden and preserve the intrinsic structure of chitosan, thereby enhancing its bioactivity and performance. However, challenges related to process scalability, equipment costs, and uniformity of treatment still limit large-scale industrial adoption. Addressing these limitations is essential for the broader implementation of sustainable chitosan processing technologies.

This natural polymer is the principal component in the exoskeleton of arthropods, as the arthropods use chitin in their cuticle to form exoskeletons and microstructural fibrils.[26] It is also found in crustaceans, such as crabs, lobsters, shrimps, mollusks, and insects, as well as in the cell walls of fungi. Chitin is composed of *N*-acetylglucosamine units linked by β-(1 → 4) glycosidic bonds due to its higher degree of crystallinity and strong intermolecular hydrogen bonding; chitin is insoluble in water and in most other organic solvents.[27] Chitin usually contains 6–7% of nitrogen compounds. There are three polymeric forms of chitin: α, β and γ; in α-chitin, the chains are arranged in an anti-parallel fashion; because of this anti-parallel nature of α-chitin, it enhances the intermolecular hydrogen bonding, making the structure more compact and stable; because of this strong hydrogen bonding, chitin has higher crystallinity, which eventually decreases its solubility in water and most of the organic solvents, thereby making it durable and rigid.[28] The β-form is generally found in mollusks and is aligned parallelly; however, the γ-nature of chitin is arranged so that two strands align parallel to each other and one is arranged anti-parallel. Chitin, derived from sources like crustaceans, fungi, and algae, as presented in Fig. 2, undergoes a chemical extraction process involving demineralization, deproteinization, and deacetylation to produce 'chitosan'. When chitin is deacetylated, its *N*-acetyl groups are partially removed, which converts *N*-acetyl-glucosamine units into D-

Table 1 Overview of chitosan: sources, extraction, and modification techniques

Sl. No.	Parameters	Specification and details	References
1	Sources of chitosan	• Exoskeletons of crustaceans: lobsters, shrimps, krill, and crayfish. • Mollusks: octopus, clams, oysters, squids, and snails. • Algae: diatoms, brown algae, and green algae. • Insects: houseflies, silkworms, cockroaches, spiders, beetles*etc.*	31
2	Chitin content (%)	• Crustaceans: 20–30% • Shrimp cuticles: 30–40% • Crab cuticles: 15–30% • Fungal cell wall: 2–44%	31
3	Extraction techniques (chemical extraction)	• Demineralization: using acids, such as HCl and HNO_3; temperature: 25 °C–100 °C; and time: 30 min–12 h. • Deproteinization: using alkalis, such as NaOH, KOH, Na_2SO_3, and Na_2CO_3; temperature: 25 °C–100 °C; and time: 30 min–72 h. • Deacetylation: using alkalis, such as NaOH/KOH (30–65%); temperature: 80 °C–150 °C; and time: 1–8 h.	31,32
4	Structural properties	• Degree of deacetylation (more than 50%) • Crystallinity • Molecular weight • Solubility	33
5	Modification techniques	• Chemical modification: Acylation, esterification, etherification, quaternization, carboxylation and graft copolymerization. • Non-thermal modification: cold plasma, irradiation, pulsed electric fields, supercritical fluid extraction, microwave, high pressure processing, electrospinning and ultrasonication.	34
6	Applications	• Drug delivery, food industry, agriculture, biomedical and pharmaceuticals, packaging, textile industry, paper industry, and wastewater treatment.	31

glucosamine units; this derivative is called chitosan.[29] This structural transformation enables its solubility and bioactivity to be applied in diverse applications. This biopolymer is a valuable candidate due to its antimicrobial, antifungal, and antioxidant properties, contributing to its widespread use in various fields. The degree of deacetylation (DD) in chitosan is represented as the proportion of D-glucosamine units relative to *N*-acetyl-D-glucosamine units in the polymer. A higher degree of deacetylation of chitin indicates greater removal of the acetyl group, increasing the number of free amino ($-NH_2$) groups in the molecule. In simple terms, chitosan can be defined as chitin with a degree of deacetylation (DD) value of 50% or higher, thereby making it soluble in acidic solutions. When chitosan is dissolved in an acidic solution, its free amino ($-NH_2$) groups become protonated ($-NH_3^+$) in an acidic environment, signifying its unique characteristic as the only naturally occurring cationic (positively charged) polymer. Due to this cationic nature, chitosan gives it distinct and functional properties.[30] However, chitosan is more reactive than chitin because of the presence of functional groups such as primary hydroxyl (–OH) at carbon-6 (C6), secondary hydroxyl (–OH) at carbon-3 (C3), and amino ($-NH_2$) group at carbon 2 (C2), where the presence of these groups makes chitosan highly diverse and versatile for chemical modifications. Basically, the free amino site ($-NH_2$) in chitosan is considered highly reactive, allowing the molecule to be functionalized *via* acetylation, grafting, crosslinking and carboxylation. Thus, functionalizing chitosan tailors its properties for specialized industrial uses, making it a valuable material across multiple domains.[24] Precisely, because of the flexible nature of chitosan, its mechanical, physical, and chemical properties can be modified accordingly, thereby enhancing its biodegradability, biocompatibility, and strength. This adaptability has made chitosan suitable for myriad applications in diverse fields, including agriculture, biomedical engineering, pharmaceuticals, drug delivery, food packaging, and wastewater treatment.[5]

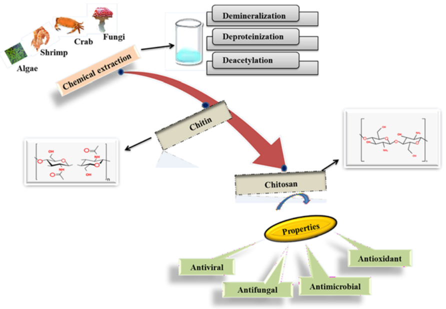

Fig. 2 Extraction process, structural transformation and properties of chitin and chitosan.

Modification of chitosan using non-thermal technologies

Technological advancements with minimal effects, maintaining quality and safety, have drawn a lot of interest.[35] As stated, the modification of chitosan using non-thermal technologies, such as plasma treatment, irradiation, high-pressure processing, pulse electric field, and ultrasonication, alters its functional and physiochemical properties without using excessive heat, maintaining structural integrity, making it a versatile component and expanding the application of chitosan in various industries by aligning with all the principles of green chemistry.

Modification of chitosan using cold plasma. In the current century, cold plasma, the fourth state of matter, which is different from solid, liquid, and gas, has gained much attention as a non-thermal technology in the food processing industries because it is an environmentally friendly and sustainable technique; the distinctiveness of this technology lies in its non-thermal, cost-effective, versatile, and environmentally friendly characteristics.[36] This non-thermal technology functions on the principle of exposing food or packaging materials[37] to ionised gases composed of neutral molecules, electrons and charged ions. These particles transfer energy through collisions, producing highly reactive species, such as ozone, UV radiation, and nitrogen oxides,[38] which eventually disrupt bacterial cell walls, reduce spoilage and contamination, and extend the shelf-life.[39] Further, it modifies packaging properties, such as flexibility, tensile strength, and oxygen resistance, maintains food freshness without involving chemicals or high temperatures[40] and stands out as a potential option for enhancing food safety without compromising quality.[41] Cold plasma interacts with biopolymers, such as polysaccharides, proteins, and lipids, interacts with phenolic compounds by modifying their functional groups through the addition or removal of specific chemical bonds, ensuring consistency across the films without uneven effects, strengthens polymer integrity, reduces gas permeability, enhances water resistance, and improves mechanical properties for better durability.[42,43] At the molecular level, cold plasma treatment generates reactive oxygen and nitrogen species (RONS) that induce surface activation of chitosan by introducing or exposing polar functional groups, such as –OH, –COOH, and $-NH_2$. These newly formed functional groups enhance the hydrogen bonding and electrostatic interactions between chitosan and the incorporated additives, such as essential oils and nanoparticles. As a result, improved interfacial adhesion, uniform dispersion, and controlled release of bioactive compounds are achieved, leading to enhanced antimicrobial and barrier performance.[44]

Decontamination is a crucial step for food products, such as nuts, due to microbial resistance, particularly from spore forming bacteria. Akhavan-Mahdavi *et al.* 2023 in their study found that when plasma treatment was given to chitosan-based coated pistachios (0.5 and 1.5% w/v), the treatment extended the shelf life and minimized aflatoxin and peroxide levels. Further, the plasma treatment of 120 s along with chitosan coating of concentration 1.5% preserved the hardness and color of pistachios during storage. This combined effect of edible coating along with plasma treatment can be a cutting-edge technology for the preservation of fresh products, ensuring microbial safety along with high-quality standards for commercial application and exports.[45] A two-step modification was applied to zein films, involving an initial enhancement with chitosan, followed by cold plasma technology to improve their functional properties. It was also observed that by the combination of treatment, the elongation at break increased from 1.13% to 4.13%, and after the application of 60 s of plasma treatment, the composite film showed higher tensile strength and a better water vapour barrier (Fig. 3). ATR-FTIR (Attenuated Total Reflectance-Fourier Transform Infrared) analysis revealed a molecular interaction between zein and chitosan after plasma treatment. As a non-thermal, chemical-free process, it maintains the film's biodegradability while boosting its suitability for food packaging.[46] With the nexus of consumer preferences, replacing synthetic materials with sustainable alternatives has reached its peak. Cold plasma treatment was applied to a chitosan-based packaging film along with cinnamon oil. The study revealed that cold plasma treatment enhances the surface roughness, wettability, and antimicrobial activity of chitosan-cinnamon oil films by generating reactive species that damage bacterial cell walls and facilitate controlled oil release. Additionally, plasma-induced crosslinking improves the oxygen barrier properties. Thus, cold plasma-treated chitosan–cinnamon oil films show strong potential for active food packaging, supporting the circular economy.[47] Pankaj *et al.* 2017 in their study observed the effect of cold plasma treatment on antimicrobial chitosan films containing thymol as an active ingredient. Plasma treatment significantly increased surface roughness due to etching effects by improving film adhesion. In terms of thermal properties, there were no significant changes, no evidence of thermal decomposition, vaporization or volatile loss after plasma exposure and the thymol diffusion rate increased after the treatment. Plasma-treated chitosan–thymol films offer better antimicrobial efficiency and control the release of active ingredients.[48] Atomic force microscopy (AFM), surface free energy, and Fourier transform infrared spectroscopy (FTIR) analyses revealed that cold plasma treatment (optimal at 75 W) increased the surface roughness and introduced polar functional groups, thereby improving chitosan coating adhesion onto PLA films. The plasma-treated films exhibited enhanced water resistance and improved mechanical strength, both of which were directly correlated with the increased chitosan deposition level at this power. Furthermore, the surface-modified films exhibited strong antimicrobial activity against *Staphylococcus aureus*. In contrast, higher

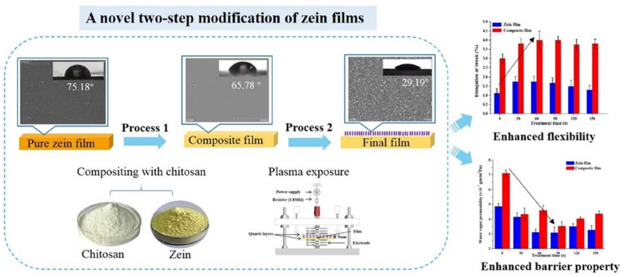

Fig. 3 Development of the composite film using cold plasma for packaging applications. Reproduced from ref. 46 with permission from Elsevier, Chen *et al.*, Industrial Crops and Products, 2019, 129, 318–326. Copyright, 2019.

discharge powers (≥100 W) adversely affected film performance due to polymer degradation.[49] A drastic increase in water repellence was observed with the contact angle reaching 121° when ultrafast plasma modification was applied to chitosan/nanocellulose biocomposite films while maintaining the mechanical strength and barrier properties of the films. Further, X-ray photoelectron spectroscopy (XPS) showed irreversible fluorine-related bonds inducing surface fluorination. This modification remained stable for over 31 days under storage conditions. It was observed that the contact angle, atomic composition, and mechanical properties remained unchanged over time. This cold plasma treatment makes it a promising, eco-friendly solution for water-resistant food packaging applications.[50] This non-thermal technology strengthens adhesion, modifies surface chemistry, and extends the shelf life without compromising its biodegradability. Cold plasma-treated films offer a sustainable alternative to conventional packaging, aligning with global efforts towards green and smart packaging solutions.

Modification of chitosan using irradiation. In the field of polymer science research and applications, ionic radiation is innocuous to the environment. In terms of green chemistry principles, irradiation is a non-thermal and ionic process, where materials are exposed to ionizing radiations, such as gamma-rays, electron beams or X-rays, eventually inducing structural and chemical modifications in the biopolymer.[51] Irradiation treatment is more advantageous than any other treatment due to its ability to penetrate deeply and uniformly within the polymer matrix at the speed of light by avoiding any thermal degradation, hence facilitating itself as a versatile treatment. Chitosan has gained significant attention in pharmaceuticals and packaging applications due to its versatile structure, such as porous scaffolds and thin films. The advantage of UV-irradiation treatment on chitosan is that it improves the surface of chitosan films and modifies its internal structure, making it easier to use, more effective, and applicable in more diverse fields.[52,53] The study by Sionkowska *et al.* (2006) demonstrated that UV irradiation effectively modifies the surface properties of chitosan films, with low-intensity UV (2–8 h) reducing the contact angle of glycerol from 84.4° to 58.8° and nearly doubling the surface free energy from 21 to ∼38–40 mN m^{-1}. High-intensity excimer laser irradiation further altered surface morphology, producing micro-foam structures above the ablation threshold at 5 J cm^{-2}. Both treatments slightly reduced the degree of deacetylation by 5–9%, without significant changes in FTIR band positions. These quantitative changes confirm that controlled UV irradiation can enhance the hydrophilicity, surface energy, and functional performance of chitosan films for packaging applications.[54] The combined use of chitosan/essential oil/AgNP composite films and gamma irradiation (1 kGy) demonstrated a synergistic effect in extending strawberry shelf life. The antimicrobial activity of the packaging along with the sterilizing effect of irradiation effectively reduced decay, minimized weight loss, maintained firmness, and preserved quality over 12 days of storage at 4 °C.[55] At the molecular level, essential oils and nanoparticles are stabilized within chitosan matrices through intermolecular interactions. The $-NH_2$ and $-OH$ groups of chitosan form hydrogen bonds with polar components of essential oils and with surface groups of nanoparticles, improving compatibility and dispersion. Electrostatic interactions between cationic chitosan and negatively charged nanoparticles help prevent aggregation, while hydrophobic interactions aid in retention and controlled release. Together, these interactions enhance the structural integrity, barrier properties, and bioactive performance of chitosan-based films.[56]

Irradiation-induced structural, barrier, and antimicrobial modifications of chitosan films. Cationic biopolymer chitosan when tailored with carvacrol essential oil as an bioactive coating on the green beans and irradiated using gamma-irradiation with 0.25 KGy for decontamination of two types of pathogens *E.coli* and *S. typhimurium,* Severino *et al.*, 2015, in their study found that these combined effect inhibited the growth of both *E.coli* and *S. typhimurium* during the entire storage period, thereby, enhancing the shelf life of green beans and minimising post-harvest losses and consumption of energy. This process of decontamination provides a promising solution to address the growing demand for food safety and preservation without compromising the quality.[57] The use of gamma-irradiation for the modification of bacterial cellulose nanocrystals tailored with chitosan nanocomposite films is a cutting-edge approach for various applications of the packaging film. The bionanocomposite film of chitosan/bacterial cellulose nanocrystals was found to have exceptional strength, biodegradability, and high surface area. Barrier properties are one of the most fundamental factors for the preservation of fresh produce, as they can accelerate respiration and transpiration, leading to post-harvest losses in fresh produce. Therefore, chitosan/bacterial cellulose nanocrystal composite films were modified using different ionization energies (0 KGy, 5 KGy, 10 KGy, and 15 KGy), which enhanced the thermal and barrier properties, and a tightly packed structure was observed with an increase in crystallinity after the irradiation treatment.[58,59] Gamma-irradiation (^{60}C) with doses of 5, 10, 20 and 50 KGy was applied to chitosan with a deacetylation value of 74.74%. The gamma-irradiated chitosan films were prepared using the solution casting method. The treatment increased the tensile strength, reduced water vapour permeability and increased *b values, leading to brightness of the films (opacity); these films can be a potent candidate for preserving food from light degradation.[60] The keeping quality of perishable items such as chicken meat can be enhanced by the synergistic effect of edible coating (chitosan/grape seed oil) treated with irradiation at a dose of 2.5 KGy. It was found that lipid oxidation decreased with the treatment of irradiation and chitosan coating. Microbial activity (aerobic mesophilic and psychrotrophic counts) decreased with a minimum of 14 days of shelf-life extension of chicken meat. The results indicated that the combined effect of irradiation along with chitosan coating can be an effective method for preserving perishable food or fresh produce with no difference in sensory attributes after irradiation treatment.[61] Biodegradable films modified with irradiation can be an eco-friendly solution to reduce the usage of petroleum-based plastics with better mechanical, tensile, oxygen, and water vapour transmission rates, offering a sustainable solution for various packaging and biomedical industries, driving innovations towards sustainable material designs and meeting the market needs by contributing towards sustainable development goals.

Modification of chitosan using ultrasonication. Ultrasonication is an eco-friendly and energy-efficient technique that utilizes the principle of acoustic sound waves to disrupt the hydrogen bonding between the molecules, for better dispersion and individualization. Acoustic sound waves occur when high frequency sound waves are generated, leading to the formation of microscopic bubbles in a liquid medium. The bubbles disrupt vigorously, which creates a rise in temperature and high shear force, leading to efficient dispersion and separation.[62] Ultrasound generates strong cavitation effects, which result in localised high temperatures and pressures, and intense shear force. The ability of ultrasound to enhance food processing efficiency while maintaining product quality has led to its increasing adoption in various applications, including dairy processing, meat tenderization, fruit and vegetable preservation, and beverage stabilization.[63] Ultrasonication rearranges polymer structures and improves inter-chain bonding, reducing absorption and enhancing moisture resistance.[64] In the study performed by Wang *et al.* 2024, they found that raw chitin nanofibres were obtained by 8 min of ultrasonication, which effectively broke down chitin into well-preserved nanofibers. The nanofibers also had a higher aspect ratio, as evidenced by atomic force microscopy (AFM), making them highly adaptable for film forming ability. The chitin nanofiber (CNF) was functionalized with curcumin, which eventually decreased the oxygen transmission rate along with the increase in the antimicrobial activity; it also enhanced the UV-blocking ability and tensile strength, which was increased up to 35%. Hence, from this study, it is apparent that ultrasonication is an efficient method for preparing raw chitin nanofibers, which serves as an excellent reinforcement in chitosan-based films, making it a potent candidate exhibiting excellent barrier properties, mechanical strength and bioactive properties, and suitable for biodegradable edible coating and food packaging applications.[65] In today's fast-paced and globalized food industry, smart packaging is an integral part of extending shelf life, food safety, reducing food waste and enhancing supply chain efficiency.[66] Strawberry is a highly valued fruit, but its post-harvest losses are maximum. Chitosan (CS) when tailored with polyvinyl alcohol (PVA) with different concentrations (2, 2.5 or 3 wt%) exhibited good physical and antimicrobial properties, the solutions were sonicated in water bath for 25 min. Sonication contributed to the effective reduction of viscosity for the CS/PVA film by ensuring better dispersion and uniformity, with enhanced mechanical strength, water retention property, and low oxygen permeability in the range of 0.16 ± 0.08 cm^2 m^2 day^{-1}. The results also demonstrate that the film increases in firmness and decreases the weight loss, thereby extending the shelf-life of strawberries up to 21 days, and these films can serve as smart packaging and sustainable solutions, improving the storage and commercialization of strawberries and their preservation.[67] Rice protein is an excellent polymer to be used in edible coating because it has excellent resistance to water vapour permeability, but due to its low solubility, there are fewer protein interactions, thereby reducing the film's flexibility, strength and durability. In this study, a novel composite of rice protein hydrolysates with chitosan was used as an edible coating and treated using ultrasound. This combined effect resulted in a reduced particle size of the film-forming solutions as well as 200 W of ultrasound treatment, which was efficient enough for a smoother film surface and better functional properties, such as oxygen barrier and elongation at break, increasing by 125%.[68] The composite film of collagen/chitosan was prepared and treated using a novel technology, sweep frequency-pulsed ultrasound. It was observed that the optimum ultrasonication time of 10 min, sweep frequency cycle of 100 s and pulse duty ratio of 77% significantly enhanced the tensile strength and elongation at break, along with a reduction in the water vapour permeability rate of the composite film. X-ray diffraction (XRD) and thermal stability indices revealed a more crystalline order and a higher thermal stability of the film after sonication. Moreover, the combined effect exhibited good antibacterial and antioxidant properties.[69] Thus, it can be noted that ultrasound treatment can be an effective method for improving and modifying the functional properties of edible films and paving the way for more effective and sustainable food packaging materials. Ultrasonication is a powerful tool for the modification of chitosan and its physical properties by bridging the gap to become the next-generation dominant biopolymer.

Modification of chitosan using pulsed electric fields. With the growing demand for more sustainable food processing technologies, pulsed electric field has emerged as an innovative and non-thermal technology that enhances food preservation, improves extraction efficiency and reduces energy consumption.[70] Pulsed electric field uses short bursts of high-voltage electric pulses (nano-to milli-seconds) with intensities of 80 kV cm^{-1}. This dynamic technology is an effective way to modify biopolymers and lower the electric field intensities used for the inactivation of microorganisms, enhancing food properties by maintaining its nutritional and sensory properties.[71]

Biopolymer zein and chitosan were tailored with synthetic polymer PVA and pre-treated using a pulsed electric field at different pulse energies. This study underscores the unique potential of the pulsed electric field as a sustainable and pre-treatment method for the modification of biodegradable films, thereby improving their mechanical and thermal properties without the use of any type of chemical cross-linkers. It was evidenced that the pulsed electric field strengths (0.9–3.5 kV cm^{-1}), pulse frequencies (50–300 Hz) and specific energies (80–650 kJ kg^{-1}) positively influenced the tensile properties and water stability of the tailored films by enhancing their stability and durability; pulsed electric field also influenced the film morphology from amorphous to a semi-crystalline structure. By reducing the reliance on chemical cross linkers and additives, the pulsed electric field supports the development of environmentally friendly materials.[72] Molecular weight (MW) significantly affects the film forming ability of chitosan owing to its application. Chitosan with a higher molecular weight often limits its application due to its viscosity and film forming abilities. Traditional degradation methods, such as enzyme hydrolysis and acid hydrolysis, tend to be time consuming and environmentally harmful. Pulsed electric fields have emerged as a non-thermal technique for chitosan degradation, offering an environmentally friendly alternative. A pulsed electric field was applied up to a strength of 25 kV cm^{-1}; with the increasing use of a pulsed electric field, the molecular weight of chitosan was lowered, making it suitable for applications requiring a low viscosity solution for drug delivery or film formation. Lower molecular weight chitosan produced by a pulsed electric field shows enhanced bioactivity, including antioxidant and antimicrobial properties.[73] Pulsed electric field provides a sustainable and controlled condition for the reconstruction of biopolymers, where studies dictate that it is a powerful tool for modification of protein-polysaccharide (zein/chitosan) interactions, enhancing colloidal dispersion properties and promoting bioconjugation. Its ability to modify structural integrity and molecular interactions makes it a promising alternative to conventional processing techniques, fostering advancements in biomaterials science and sustainable packaging industries. Treating zein-chitosan/polyvinyl alcohol film with pulse electric fields induced conformational changes in the structure of the biopolymeric films. It was observed that a low intensity pulse electric field enhanced the stability of the edible film against enzyme degradation, and no toxicity was found. Pulse electric field-modified edible film can be a potent technology for value addition in edible food packaging.[74] Reinforcing microcrystalline cellulose with starch/chitosan and applying a moderate electric field to the biopolymeric film improved the physiological properties of the film. The composite polysaccharide-based film showed excellent thermal properties, mechanical strength and barrier properties. Surface morphology can be assessed by scanning electron microscopy (SEM) and X-ray diffraction (XRD), which revealed that different concentrations of microcrystalline cellulose along with a moderate electric field are responsible for changes in the properties of the films. It was observed that the application of a moderate electric field was sufficiently efficient in decreasing water vapour permeability in chitosan-based films. Therefore, this reinforced biopolymeric film along with a moderate electric field can be a potent application for changing and modifying the properties of the film.[75] The use of pulse electric fields in the application for the modification of biopolymers presents a promising technique for enhancing material properties, such as conductivity, mechanical strength, and surface modifications. This method enables precise control over the polymer structure and facilitates applications in diverse fields.

Modification of chitosan using high-pressure processing (HPP). High pressure processing or high hydrostatic pressure (HPP) processing is a non-thermal technique for the preservation of food, which has gained much attention since the late 20s. The first commercial use of high-pressure processing started in the land of the rising sun (Japan) in 1980. Many food products, such as yogurt and jam, have been produced using this procedure, eventually showcasing the potential use of high-pressure processing as a non-thermal technology with minimum heat damage.[76] High pressure processing functions on the principle of the use of high pressure around 300–800 MPa over time. Since high pressure processing does not use heat, it preserves the food's natural colour, flavour, and texture, which eventually helps in reducing microbial load, maintaining nutritional and sensory attributes, inactivating enzymes, and modifying biopolymers.[77,78] In the biopolymer industry, high pressure processing enhances the functional properties of biopolymer-based packaging materials. The high-pressure processing technique alters the structural and physiochemical characteristics of biopolymers, leading to improved performance in packaging applications. This technique enhances molecular interactions, such as hydrogen bonding and hydrophobic interactions, which strengthen the integrity of the biopolymer matrix. This results in improved resistance to mechanical stress and environmental conditions.[78]

An edible coating of chitosan/tea polyphenol along with high pressure processing as a synergistic effect has been applied to shrimp to increase its antioxidant and antimicrobial properties. This combined treatment of 400 MPa of high pressure along with chitosan/tea polyphenol coating inhibited microbial growth, delayed spoilage, and showed better oxidative stability and the shrimp retained hardness and colour stability during the entire storage period. This method can be an effective way to extend the shelf life of shrimp while maintaining its quality.[79] The study performed by Martillanes *et al.* 2021 focuses on evaluating the effectiveness of high-pressure processing combined with chitosan-based films in reducing *L. monocytogenes* contamination on sliced dry-cured Liberian ham. The combination of high-pressure processing with nisin and oryzanol-enriched chitosan film resulted in the greatest reduction of *L. monocytogenes*, achieving a 6 log cfu per g decrease. This hurdle approach is effective, making it a promising strategy for improving food safety in dry-cured meats.[80] High hydrostatic pressure and nano-TiO_2 can also be applied in chitosan/polyvinyl alcohol-based films to enhance surface properties. It was observed that high hydrostatic pressure enhanced the molecular interactions between chitosan and alcohol films. This combined treatment improved the mechanical and barrier properties of the film, leading to better durability and stability, with higher antimicrobial properties. The applied hydrostatic pressure in the range of 200–400 MPa maintains the integrity of the film, which suggests resistance to degradation or dissolution and reduces the migration of TiO_2 nanoparticles from the films, making them a potent candidate for sustainable and functional food packaging.[81] The use of high pressure processing and antioxidant-enriched gelatin-chitosan edible films with oregano and rosemary extract improved the chemical stability (preventing oxidation) and microbial safety of cold-smoked sardines (*Sardina pilchardus*), making them long lasting. When the sardine muscles were coated with edible films, the phenol content and antioxidant power increased. This effect was even stronger when high pressure was applied likely because the pressure helped transfer antioxidant compounds from the film to the fish. This combined effect was more effective at reducing microbial counts, particularly sulphide-reducing bacteria; no luminescent bacteria or *Enterobacteriaceae* were detected in any samples after the treatment.[82] This can be an effective way to enhance the shelf life of fresh produce at a low cost without any energy consumption. High pressure processing is a promising sustainable technology that enhances the functional, preservation, and modification of biopolymers while aligning with environmental goals. Its ability to reduce energy efficiency makes it an eco-friendly solution for various industries, such as food, medicine and biomedical applications.

Modification of chitosan using other non-thermal techniques. As shown in Table 2, it represents different non-thermal technologies, such as cold plasma, irradiation, microwave, ultrasonication, ozone treatment, electrospinning, and pulsed electric field for modifying chitosan-based films, demonstrating their impact on mechanical, barrier, and functional properties. These techniques tend to be effective processes in altering and enhancing the physiochemical properties of chitosan without any thermal degradation. Microwave processing, a non-thermal technology, can be used to modify biopolymers by influencing their chemical and physical properties without the need for direct heat. Microwaves generate heat by causing polar molecules (such as water) to oscillate. This results in internal heating unlike conventional methods that rely on external heat sources.[83] Although microwaves do generate heat, rapid and localised heating results in non-thermal effects. These effects include changes in the conformation of the polymer chains, which do not involve a significant increase in temperature. The microwave field induces mechanical stress, breaking or altering the molecular structure of biopolymers and potentially improving their solubility, viscosity or reactivity.[84] A study explores the surface modifications of microcrystalline cellulose using urea under microwave radiation to enhance the reinforcement of chitosan films. The concept combines the use of microwave processing, which can rapidly and efficiently modify the cellulose surface and urea and eventually introduce functional groups between microcrystalline cellulose and chitosan. Further, the reinforcement effect was analysed using scanning electron microscopy (SEM), Fourier transform infrared spectroscopy (FTIR) and X-ray diffraction (XRD), where it has been revealed that it enhances mechanical properties, such as tensile strength, Young's modulus and fracture energy, which are 2.0, 2.4 and 6.0, respectively, in comparison to pure chitosan. This is basically due to the strong interactions between microcrystalline cellulose and chitosan polymer chains in the film matrix. This composite film also acts as a better barrier against moisture; this improved barrier property is the combined effect of microwave treatment along with strong interactions between microcrystalline and chitosan, which reduces the pathways for water molecules to pass through the film.[85] Researchers demonstrated that microwave-assisted modification of chitosan using a pulsed power (SPS) mode significantly enhances reaction efficiency compared to conventional heating. SPS irradiation enabled rapid heating to target temperatures (40 °C-80 °C within 100 s at 30–100 W *versus* 600 s under water-bath heating), resulting in up to 35% higher alkylation yields without inducing chain degradation or deacetylation. Microwave irradiation primarily improved reaction kinetics during the initial minutes while reducing overall energy consumption. The resulting alkylated chitosan exhibited amphiphilic behaviour, shear-thinning rheology, and aggregation dependent on the degree of alkylation, highlighting microwave processing as an energy-efficient and scalable route for functional chitosan derivatives.[86]

Further, electrospinning is a versatile fabrication method that is used to create nanofibrous materials from biopolymers. It works on the principle of applying high voltage to a polymer solution and causing the material to form ultrafine fibres. The resultant electrospun fibres possess unique properties, such as high surface area and porosity with enhanced mechanical properties, making them useful in a wide range of applications.[87] Fat dissolving oral films (FDOFs) were developed using chitosan and pullulan through electrospinning technology. These edible films are precisely designed to dissolve quickly in the mouth, making drug delivery more efficient. Fourier transform infrared spectroscopy (FTIR), X-ray diffraction (XRD), and thermal analysis revealed a reduction in crystallinity due to electrospinning, which made the film more amorphous through dissolution. With the addition of chitosan, the thermal stability also increased; the water solubility test showed that the composite film could dissolve within 60 s in water. This composite-modified film using electrospinning can be a promising solution and value-added edible coating.[88] PVA/chitosan blend films were prepared using electrospinning under specific conditions at 21 kV and a 15 cm distance. The study reveals that the films have tiny fibre-like structures, which show profound antibacterial properties. These fibres have a large surface area, making them more effective against *E. coli*. Additionally, the special surface texture of electrospun fibres hinders the cell wall of bacteria by disrupting their adhesion to the biopolymeric film, thereby decreasing the growth of the bacteria and projecting its antibacterial activity.[89] Overall, the combination of electrospinning and chitosan-based films/edible coating can be used as a potent candidate for an antimicrobial packaging solution for perishable food products.

Additionally, the allotrope of oxygen is ozone (O_3) with a higher oxidizing power. Ozone is a strong oxidizing agent that reacts with organic molecules by introducing functional groups, such as hydroxyl, carbonyl and carboxyl groups. Ozone treatment is considered safe for use in food processing, as approved by the Food and Drug Administration (FDA). Ozone treatment alters the surface chemistry of biopolymer nanofibers or films by introducing oxygen-containing functional groups. This enhances the hydrophilicity, wettability, mechanical and barrier properties of the film.[90] Additionally, PHA is a class of biodegradable biopolymers produced by microorganisms, and they possess excellent biodegradability and biocompatibility but generally lack strong antimicrobial properties; to enhance their antimicrobial properties, they are tailored with other antimicrobial agents, such as chitosan and chitooligosaccharides, using ozone treatment. This combined treatment enhances the antibacterial and biodegradable properties. Researchers claim that the antibacterial effects are particularly strong against Gram-negative bacteria due to their negatively charged cell walls, which attract the positively charged chitosan molecules, and ozone treatment introduces functional groups onto the composite film, thereby improving reactivity.[91] In this study, lignocellulose films were blended with 2% chitosan to develop edible films. Short ozone

Table 2 Modification of the chitosan-based films using non-thermal technologies

Sl. No.	Material	Non-thermal technologies	Outcomes	References
1.	• Chitosan/wampee seed essential oil	• Cold plasma treatment	• Provides synergistic effects due to the combined use of the edible coating of chitosan along with cold plasma treatment on golden pompano fillets. • The treated fillets showed the lowest bacterial count and peroxide values and inhibited lipid lysis with enhanced antimicrobial activity, thereby extending the shelf life throughout the storage period.	93
2.	• Chitosan/cellulose nanocrystals	• Cold plasma treatment (low temperature generated)	• Improved packaging film property through surface modification, such as improved water repellence, significantly increasing the contact angle while boosting mechanical strength and reducing water vapour transmission. • The treated packaging film showed the highest stability during the 31 days storage period.	50
3.	• Chitosan	• Pulsed electric fields/ozone treatment	• Combined effects resulted in a lower molecular weight of chitosan (M_w < 2500 Da) and lower crystallinity.	94
4.	• Chitosan/grape	• Gamma-irradiation	• Used as an edible coating on chicken breast meat. The combined effects of irradiation (2.5 kGy) and chitosan/GSE coating enhanced the antimicrobial activity. • All the coated samples showed lower TBA (thiobarbituric acid) and pH values, and the sensory quality was retained during the entire 21 days storage period.	61
5.	• Chitosan/essential oil/silver nanoparticles	• Gamma-irradiation	• Provides a synergistic effect due to the combined use of irradiation and edible coating on fresh strawberries. • The composite film exhibited strong antimicrobial activity against pathogenic bacteria. • Weight loss, firmness and decay (%) were retained during the 12 days storage period.	55
6.	• Quinoa protein/chitosan/transglutaminase (TG)	• Ultrasonication	• The thermal stability of the composite film increased after ultrasonication treatment. • The treatment enhanced the structural and mechanical properties of the composite films.	95
7	• Chitosan/*Sargassum pallidum* polysaccharides	• Ultrasonication	• Ultrasonication enhanced the transparency, elongation, and tensile strength of the film along with the antioxidant activity, which has also increased. • The treated films extended the shelf life of the strawberry, inhibiting deterioration at room temperature (25 °C) during the 7 days storage period.	96
8	• Chitosan/Mandarin essential oil	• High pressure processing (HPP)/pulsed light processing	• The combination of edible coating along with HPP on green beans resulted in a significant reduction of *L. innocua,* thereby increasing the shelf life. • The treatment also retained the firmness of green beans.	97
9	• Chitosan/starch/microcrystalline cellulose	• Pulsed electric fields	• The treated composite film presented good thermal and mechanical properties, generating more rigid films.	75
10	• Chitosan/polyvinyl alcohol/essential oils	• Electrospinning	• Antioxidant activity showed the highest value for the composite film. • The bilayer lamination of the composite film increased the thermal resistance with an increase in antimicrobial activity.	98
11	• Chitosan	• High hydrostatic pressure (HHP)	• The shelf life of rainbow trout fillets increased due to the combined effect of chitosan-based coatings and HHP. • The combined treatment also resulted in a protective effect both chemically and microbiologically.	99
12	• Chitosan/cinnamon oil	• Cold plasma treatment	• The strength and the antioxidant property of the films increase after the treatment. • Cold plasma-induced composite film showed higher antimicrobial activity along with improved oxygen barrier properties.	47

treatment enhanced the strength and hydrophobicity, while prolonged treatment of 120 min improved the film uniformity, swelling and antibacterial effects. Chitosan incorporation further strengthens the film and increases moisture absorption, making it particularly effective for fruit preservation and antimicrobial food wraps. This ozone-treated composite food coating wrap can be a potent source for extending the shelf life and minimizing post-harvest losses.[92] Across different non-thermal technologies,

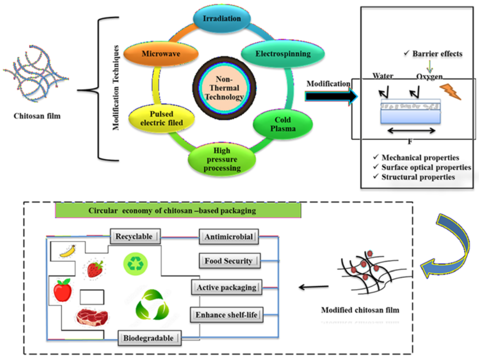

Fig. 4 Non-thermal modification of chitosan: enhancing the biodegradation for circular economy.

such as microwave processing, electrospinning, ozone treatment, irradiation, ultrasonication, and cold plasma, the improvement in chitosan-based films can be largely attributed to molecular-level interactions. These techniques modify the surface chemistry and microstructure of chitosan by introducing or activating functional groups, thereby promoting hydrogen bonding, electrostatic interactions, and physical entanglement between chitosan chains and incorporated additives or reinforcing agents. These interactions enhance interfacial adhesion, uniform dispersion, and controlled release behaviour, ultimately leading to improved mechanical strength, barrier performance, and functional properties of the modified films.[44,56]

Non-thermal technologies enhance the functionality of chitosan without degrading its structure, as illustrated in Fig. 4. These methods improve its biodegradability, making it eco-friendlier and more suitable for sustainable applications. Further, enhanced biodegradation supports waste reduction and aligns with circular economy principles by promoting resource efficiency. This approach enables the development of biodegradable materials for the agriculture and food packaging industries, thereby reducing the environmental impact.

Circular economy and life cycle assessment of chitosan-based packaging

The circular economy approach to the non-thermal modifications of chitosan focuses on sustainable processing methods that minimize energy consumption and waste generation. By utilizing non-thermal modification techniques, such as cold plasma, irradiation, pulsed electric field, high pressure processing, and electrospinning, chitosan can be tailored for various applications in food, biomedical, and bioplastics while maintaining its biodegradability. These eco-friendly techniques align with the circular economy principles, thereby extending the shelf-life, minimizing post-harvest losses, reducing reliance on harsh chemicals, and potentially utilizing waste from marine and agricultural sources.

Life-cycle assessment (LCA) of chitosan-based films modified using non-thermal techniques evaluates their environmental impact from raw material sourcing to end-of-life disposal. In a study, the LCA of cellulose nanofibril (CNF)-reinforced chitosan composite films evaluates the environmental impact across their production, usage and disposal phases. This assessment considers raw material extraction (*e.g.* chitosan from seafood waste and CNF from plant fibres), energy consumption in non-thermal modification techniques and the environmental footprint of film fabrication processes, like casting or extrusion. The results indicate that CNF-reinforced chitosan films have a lower carbon footprint, enhanced mechanical properties and improved biodegradability, supporting circular economy principles. The end-of-life scenario, such as composting or recycling, further enhances their sustainability, making them a viable eco-friendly alternative to synthetic plastic packaging.[100] LCA of chitosan-based films confirms their eco-friendly potential, thereby lowering environmental impact and biodegradability compared to conventional plastics, which supports sustainable packaging solutions.

Challenges to industrial scale-up and cost barriers

Despite significant advances in non-thermal modification technologies and the demonstrated potential of chitosan-based films for sustainable packaging, the translation of these laboratory-scale innovations into industrial-scale production remains challenging. Critical barriers related to process scalability, economic feasibility, energy demand, and equipment costs continue to limit large-scale adoption, necessitating a focused discussion on industrial implementation constraints.

Chitosan requires additional chemical or physical modifications often to improve mechanical strength, barrier performance, and pH stability, further increasing production costs and complicating food-contact regulatory approval. Intrinsic limitations such as moisture sensitivity (starch-based films) and pH-dependent solubility of chitosan restrict industrial applicability, and although oxygen barrier improvements of 40–60% have been reported at the laboratory scale, these are difficult to reproduce under industrial processing conditions. Moreover, fewer than 20% of studies address scalability, cost reduction, and regulatory compliance, highlighting a persistent gap between academic research and industrial translation. Nevertheless, commercial examples including Mater-Bi® and fungal chitosan-based coatings demonstrate that industrial adoption is feasible when economic, scalability, and regulatory challenges are addressed concurrently.[44] Among polysaccharides, chitosan has attracted considerable interest for edible coatings of pre-processed fish products due to its biodegradability, film-forming ability, and inherent antimicrobial activity. However, the large-scale industrial implementation of chitosan-based coatings remains challenging because of the variability in raw material quality, pH-dependent solubility, and difficulties in achieving uniform coating thickness under high-throughput processing conditions. To enhance mechanical strength, barrier properties, and stability, chitosan often requires chemical, physical, or composite modifications, which can increase formulation complexity, production costs, and regulatory burdens. Additionally, the incorporation of active compounds may affect coating reproducibility and storage stability. Addressing these scale-up and modification challenges is essential for the successful industrial adoption of chitosan-based edible coatings in fishery products.[101] Overall, the chitosan-based coatings show strong potential for food preservation; their industrial adoption is limited by scale-up challenges related to raw material variability, pH-dependent solubility, and formulation sensitivity. Moreover, modification strategies required to improve mechanical and barrier properties increase costs and regulatory complexity. Overcoming these issues through cost-effective processing, scalable modifications, and standardized protocols is essential for successful commercial implementation.

Conclusions

The biopolymer chitosan has recently drawn researchers' attention due to its excellent antimicrobial and antioxidant properties and film forming abilities. It is developed from naturally available sources with historical significance as a highly promising material for sustainable food packaging. Moreover, its non-toxicity and biocompatibility features provide an eco-friendly alternative to conventional synthetic plastic materials. Owing to these attractive properties, chitosan has been used in a number of fields, such as medicine, agriculture, biomedical, food packaging, wastewater treatment, biosensors, and textile industries. However, the need to improve chitosan's mechanical properties, water resistance, and oxygen barrier properties has driven research into various modification techniques. Among these, non-thermal technologies have emerged as potential candidates for enhancing chitosan's functional and structural properties without compromising its bioactivity, thereby aligning with the principles of 'green chemistry'. The current paper reviewed applications of chitosan-based films modified using non-thermal technologies, such as cold plasma, irradiation, pulsed electric field, ultrasonication, high pressure processing, microwave, electrospinning and ozone treatment. These advancements not only enhance the structural and barrier characteristics of chitosan films but also expand their application in active and intelligent packaging systems. The non-thermal modification of chitosan plays a pivotal role in sustainable development, enhancing its properties in energy efficiency, environmental friendliness, and economic viability. These modifications enable the widespread adoption of biodegradable high-performance chitosan-based films, contributing to waste reduction, food preservation and reduction in fossil fuel-based plastic usage. With increasing market demand, industries seek greener alternatives for packaging; non-thermal modification can be a resilient pathway for achieving global sustainable development goals, thereby maintaining material efficiency and functionality.

According to researchers, future studies should focus on optimizing non-thermal modification strategies for industrial-scale applications and cost-effectiveness, and on developing hybrid biopolymer composites to enhance performance and meet the growing demand for sustainable materials in additive manufacturing. Modified chitosan shows great potential as a bio-ink for 3D printing applications in food packaging, biomedical scaffolds and tissue engineering. With continued innovation, chitosan holds immense promise as a next-generation material for eco-friendly and functional food packaging systems.

Future outlook and industrial implications

Non-thermal modification technologies (*e.g.*, cold plasma, ultrasonication, irradiation, microwave processing, and electrospinning) offer scalable and energy-efficient routes for enhancing the functionality of chitosan-based films, with reduced thermal degradation and improved retention of bioactive additives, making them highly attractive for industrial food packaging applications. Modified chitosan films with tailored surface properties and enhanced barrier and antimicrobial performance can significantly reduce post-harvest losses and extend the shelf life of perishable foods, supporting the circular economy and sustainability goals by replacing petroleum-based plastics. However, comprehensive investigations into scale-up feasibility, cost-benefit analysis, and regulatory compliance are required to facilitate the industrial adoption of these non-thermal technologies. Environmental impact assessments, including life cycle analysis (LCA) and evaluation of surface wettability under real storage conditions, are essential to quantitatively assess sustainability benefits and to optimise material design. Recent advances in surface wettability and sustainable coating technologies highlight the critical role of surface functionality in governing the barrier performance of packaging materials.[102]

Future studies should also prioritise the development of standardised processing protocols, long-term performance evaluation, and real-world application trials, supported by strong industry-academia collaborations to bridge the gap between laboratory-scale innovations and commercial implementation.

Author contributions

Susmita Bora: original draft writing, review and editing. Tabli Ghosh: conceptualization, supervision, validation, final review and editing.

Conflicts of interest

The authors declare no conflicts of interest.

Data availability

No datasets were generated or analysed in the study.

Acknowledgements

The authors are thankful to the Sustainable Food Lab at the Department of Food Engineering and Technology, School of Engineering, Tezpur University, Assam, India, for the support to carry out the work.

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