

Sustainable Food Technology

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Sustainability spotlight

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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 material science and highlights recent advancements in non-thermal modification techniques such as ultrasound, cold plasma, pulsed electric field, and high-pressure processing. These methods improve solubility, film-forming ability, and barrier properties without affecting its natural functionality. The 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.



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The state-of-the-art on Chitosan: Historical Perspectives and Non-thermal Modification Technologies for Sustainable Packaging Films

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Abstract

Chitosan has garnered a significant attention in the field of polymer science over the decades, due to the associated characteristics features such as biodegradability, antimicrobial property, and film-forming abilities. With the increasing concerns, extensive research has been conducted to enhance the functional properties of chitosan through various modification techniques. Recently, modification of chitosan-based material using non-thermal technologies has drawn attention due to its ability to improve mechanical strength, barrier properties, and bioactivity without compromising the environmental sustainability. This review discusses the historical discovery and development of chitin and chitosan and further emphasizes the growing significance in biomaterial science. A key focus of this study is directed to applying non-thermal modification technologies such as cold plasma, irradiation, high pressure processing, ultrasonication, pulsed electric field, electrospinning, and others for modification of chitosan-based materials for its application in active and intelligent packaging systems, 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 a viable alternative to conventional plastic packaging. Mechanical property, antimicrobial efficacy, oxygen barrier properties, and thermal stability of chitosan are among the key benefits and the proposed material has been successfully used as food packaging material for the preservation of fresh produce and for increasing the shelf life of several types of food products. This approach not only

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supports the transition towards a circular economy but also contributes to the significant reduction of plastic packaging.

Keywords: Chitosan; Non-thermal technologies; Biodegradable; Antimicrobial; Sustainable packaging.

Introduction

In recent years, the fossil derived polymer has posed significant environmental challenges being non-biodegradable in nature, which in turn increase the carbon footprints 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. Due to this, there is a growing shift towards the use of natural polymers, which are biodegradable and eco-friendly in nature.² According to the Institute of Bioplastics and Biocomposites (2016), global bioplastics 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 the year 2019, the global market for biodegradable plastic was valued to be about USD 4.65 billion and it was projected to grow by the end of 2025 at the compound annual growth rate of 17.04% reaching the market value of up to 12.06 million. Biodegradable plastics such as PLA (polylactic acid), PHA (polyhydroxyalkanoates) and others accounts for over 55.5 % of the total global bioplastics production.³ Bioplastic production is dominated by Asia, which contributes 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 includes biodegradability, ensuring that materials break down naturally without causing any environmental hazards. Biodegradability is not only a functional requirement, but is a crucial environmental necessity. Biodegradable polymers break down 60% faster than conventional plastics, which can take over 1,000 years to degrade in the environment. Biodegradable plastics can fully decompose within 180 days.⁴ Non-biodegradability contributes to landfill, plastic waste, and pollution causing ecological imbalance. As a result, the uses of natural or biopolymers (the substances derived inherently from living matter) are gaining more attention due to the associated shortcomings and also with the increasing consumer awareness regarding the use of synthetic polymers.²

Among available, chitin is considered as the second most abundant biopolymer after cellulose being first described by Henri Braconnot in the year 1811. This naturally occurring biopolymer is found in ordered microfibrils within the exoskeletons of mollusks, crustaceans as well as in the cell wall of fungi; it is reverberated that 70% of chitin comes from the marine species.⁵ However, 'chitin' was formally name in 1823 by Odier who isolated it from the cuticles of insects. During 19th and 20th century, researchers recognised its structural similarity with cellulose. Chitosan, the deacetylated form of chitin, was first described by Charles Rouget in the year 1859. Despite these early discoveries, the utilization as well as application of chitin and chitosan remained limited until the mid-20th century. By the late 20th century, chitin and chitosan have gained much popularity due to its 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, is a heteropolysaccharide composed of β -1 \rightarrow 4-linked glucosamine and N-acetylglucosamine units, and it is the second most abundant biopolymer after cellulose.⁷ Chitosan is widely used in medicine, food, cosmetics, biomedical as well as in the 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 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 solvent at pH > 6.5, making the functionalization and processing crucial. Moreover, chitosan is also insoluble in most organic solvents, restricting its application for various purposes. Therefore, modification of chitosan has become an outmost essential to enhance chitosan's functional properties to expand its applications.⁹ The primary methods for chitosan modification includes 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 also harsh chemical modification alters



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chitosan's biocompatibility, degrading its molecular structure, and limiting its application in various sensitive fields such as pharmaceuticals, and food packaging. Additionally, it often requires high temperature, 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 biopolymer using non-thermal technologies such as cold plasma, high pressure processing, irradiation, pulse electric field, ultrasonication, microwave, etc. are gaining 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 more shift towards greener technologies.¹² These modifications of the natural biopolymer chitosan using non-thermal technologies in contrast to conventional modifications allows controlled modification without compromising its structural integrity, enhancing functional properties without any degradation or harmful by-products. Additionally, these techniques don't rely on heat but instead the use of mechanical, electrical, and radiation-based energy to alter and modify its structure and functionality is getting explored.¹³ on-thermal technology often allows targeted modifications e.g. improved solubility, antimicrobial activity, and biocompatibility without any structural breakdown. Chitosan modification using non-thermal technology have been explored for a wide range of application such as plasma treatment 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 influences 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, the review critically discusses recent advances in non-thermal modification of chitosan, highlighting their impact

on 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.

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History of Chitin and chitosan

The polymers are large molecules composed of repeating structural units called monomers, where, they play a crucial role in various scientific and technological fields. Based on their source of 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 the nature's most abundant and versatile biopolymers. Trailing only in prevalence after 'cellulose'; chitin is considered as the second most surpassed polysaccharide. Over, the course of last 220 years, the study of chitin has progressively marvelled from scientific curiosity towards being one of the fundamental elements of biomaterial science, as illustrated in **Figure 1**, it traces the historical development of chitin and chitosan from their discovery to current period.

In the year 1811, Henri Braconnot, a France chemist, isolated a material from fungi using water, alcohol, dilute alkali, and named the insoluble residue as "fongine" or "fungine". This insoluble residue was later identified as chitin and making the discovery of Braconnot as the first recorded identified 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 the year 1823 during his experiment with cockchafer betel found a distinct substance, hornlike material, and named it as "Khiton, Chitine" which has the meaning of 'envelop' in Greek. Another scientist, Opperman, in 1832, envisaged that chitin was extracted from insects-similar substances as chitin can also be found in the structure of insects.¹⁸ Thereby, another conflict arose between scientist about chitin produced by arthropods and cellulose produced by plants. In the year 1859, French physiologist Charles Rouget, found that heating chitin makes it soluble in organic solvents, leading to the discovery of its derivative, chitosan.



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As reported by Crini, the era between 1894 to 1930 led to another significant controversy about the presence of chitin and cellulose to be found in the cell wall of fungi and also about its solubility in organic solvents.¹⁷ In the year 1930, Rammelburg identified more sources of chitin apart from insects and fungi and also highlighted that it can be extracted from exoskeletons of marine arthropods such as crabs, shrimps, lobsters, later in that year, he also made a remarkable contribution by identifying chitin as a polysaccharide of glucosamine. By the use of polarised radiation spectra, in the year 1950, Darmon and Rudall, identified the structure of chitin, chitosan, and cellulose.¹⁹ During the period from 1930 to 1950, chitin and chitosan has gained much attention in its extraction and isolation process, where, the first book was published in the year 1951, which has included the research conducted by various scientists on chitosan. Thereby, late in the year 1960, different polymeric forms of chitin were identified.¹⁷ Since 1960 to till date, 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 were increasing in material science for the use of sustainable processing methods for the extraction of biopolymer 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 biopolymer along with its integral mechanical properties. This era began in the late 20th and early 21st centuries as material science and environmental concerns drove its search from alternatives to traditional thermal technologies.

With due course of time, researchers have analysed that 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, making this biopolymer as an invaluable in the area of polymer science with its diverse application.²²

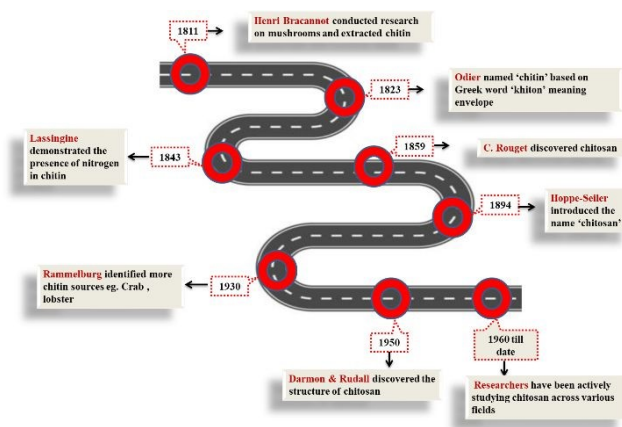


Figure 1. Historical evolution of chitin and chitosan

Chitin and chitosan: Structural and Chemical properties

Biopolymers are the units of monomers which are derived from biological sources. Their molecular structure can be linear, branched as well as cross-linked, thereby influencing the properties and their applications. Biopolymers are broadly classified into three types depending on their monomeric units such as (i) Polynucleotides: These biopolymers are composed of repeating monomeric units e.g., DNA, RNA; (ii) Polypeptides: These consist of short polymer of amino acids; (iii) Polysaccharides: These consist of linear 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-toxic nature, biodegradability as well as excellent film forming abilities. These characteristics make them a potent candidate to be used as a sustainable alternative to plastic based packaging thereby, focusing on sustainable development goals.²³



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Through the last decades, it can be considered to be 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 application in various sectors.²⁴ The second most biopolymer present after cellulose viz. 'chitin' can be defined as the abundant mucopolysaccharide present in nature, which is hard, inelastic, and made up of nitrogenous compounds.⁵ Further, unlike any other polysaccharide such as glycogen or starch being considered as 'storage polysaccharide', chitin is considered as the 'structural polysaccharide'.²⁵ In this regard, the overview of chitosan sources, extraction, and modification techniques has been represented in **Table 1**. Compared to conventional chemical routes, emerging non-thermal and green modification techniques such as microwave, ultrasonication, cold plasma, and pulsed electric field etc. offer reduced processing time, 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 cost, 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 of their cuticle for forming the exoskeleton and microstructural fibrils.²⁶ It is also found in crustaceans such as crabs, lobsters, shrimps, mollusks, and insects as well as in the cell wall of fungi. Chitin is composed of N-acetylglucosamine units linked by β -(1 \rightarrow 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 compound. There are three polymeric forms of chitin: α , β and γ ; in α -chitin, the chains are arranged in 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 have higher crystallinity, eventually, decreasing 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, whereas, the

γ - nature of chitin is arranged as where two strands align parallel to each other and one is arranged anti-parallel. Chitin, derived from sources like crustaceans, fungi, algae etc. as presented in **Figure 2**, undergoes a chemical extraction process involving demineralization, deprotonization, and deacetylation to produce 'chitosan'. When chitin is deacetylated, its N- acetyl groups are partially removed, which converts N-acetyl-glucosamine units into D-glucosamine units, this derivative is called as chitosan.²⁹ This structural transformation enhances its solubility and bioactivity to be applied in diverse applications. This biopolymer tends to be a valuable candidate due to its antimicrobial, antifungal, and antioxidant properties contributing 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 more removal of acetyl group, increasing the number of free amino (-NH₂) groups in the molecule. In simple terms, chitosan can be defined as chitin with degree of deacetylation value (DD) of 50% or higher thereby, making it soluble in acidic solutions. When chitosan is dissolved in acidic solution, its free amino (-NH₂) groups, becomes protonated (-NH₃⁺) in 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 the functional groups such as primary hydroxyl (-OH) at carbon- 6 (C6), secondary hydroxyl (-OH) at carbon-3 (C3), and amino (-NH₂) 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 (-NH₂) in chitosan is considered to be highly reactive allowing the molecule to be functionalized such as acetylation, grafting, crosslinking as well as carboxylation. Thus, functionalizing chitosan tailors its properties for specialized industrial uses, making it a valuable material across multiple domains.²⁴ Precisely, because of this flexible nature of chitosan, its mechanical, physical, and chemical properties can be modified accordingly, thereby, enhancing its biodegradability, biocompatibility, as well as strength. This adaptability has made chitosan to be used in myriad application in diverse fields from

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agriculture, biomedical engineering, pharmaceuticals, drug delivery, food

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Table 1. Overview of chitosan: Sources, extraction, and modification techniques.

Sl no	Parameters	Specification and details	References
1.	Sources of chitosan	<ul style="list-style-type: none"> • Exoskeletons of crustaceans: Lobsters, shrimps, krill, and crayfish. • Mollusks: Octopus, clams, oysters, squids, and snail. • Algae: Diatoms, brown algae, and green algae. • Insects: Housefly, silkworms, cockroaches, spiders, beetles, etc. 	[31]
2.	Chitin Content %	<ul style="list-style-type: none"> • Crustaceans: 20-30% • Shrimp cuticles: 30-40% • Crab cuticles: 15-30 % • Fungi cell wall: 2-44% 	[31]
3.	Extraction techniques (Chemical extraction)	<ul style="list-style-type: none"> • Demineralization- Using acid such as: HCL, HNO₃, Temperature: 25- 100 ° C, Time: 30 min – 12 h • Deproteinization- Using alkali such as: NaOH, KOH, Na₂SO₃, Na₂CO₃, Temperature: 25 – 100 ° C, Time: 30 min – 72 h • Deacetylation- Using alkali such as: NaOH/KOH (30-65%), Temperature: 80- 150 °C, Time: 1- 8 h 	[32,31]
4.	Structural Properties	<ul style="list-style-type: none"> • Degree of deacetylation (more than 50%), • Crystallinity • Molecular weight • Solubility 	[33]
5.	Modification techniques	<ul style="list-style-type: none"> • Chemical modification: Acylation, esterification, etherification, quaternization reaction, carboxylation and graft copolymerization. • Non-thermal modification: Cold plasma, irradiation, pulsed electric field, supercritical fluid extraction, microwave, high pressure processing, electrospinning and ultrasonication. 	[34]
6.	Application	<ul style="list-style-type: none"> • Drug delivery, food industry, agriculture, biomedical and pharmaceuticals, packaging, textile industry, paper industry, waste water treatment. 	[31]

packaging industry, and in waste water treatment .⁵

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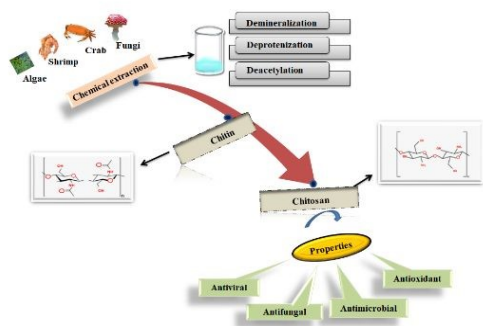


Figure 2. Extraction process, Structural transformation and properties of chitin and chitosan.

Modification of chitosan using non-thermal technologies

The technological advancement with minimum effects and thereby, maintaining the 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 alter its functional, and physiochemical properties without using excessive heat, maintaining structural integrity, and making it a versatile component and expanding 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 as being an environment 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 works on the principle of exposing the food or packaging materials³⁷ to ionised gases made up of neutral molecules, electrons and charged ions, these particles transfer energy through collisions, producing highly reactive species such as ozone, UV radiation, nitrogen oxides, etc.,³⁸ which eventually disrupts bacterial cell walls, reduces spoilage, and contamination, extends the shelf-life.³⁹ Further,

it modifies the packaging properties such as flexibility, tensile strength, and oxygen resistance, maintains food freshness without involving chemicals or

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high temperatures⁴⁰ and also stands out as a potential option for enhancing food safety without compromising the quality.⁴¹ Cold plasma interacts with the biopolymers such as polysaccharides, proteins, lipids, and also interacts with the phenolic compounds by modifying their functional groups through the addition or removal of specific chemical bonds ensuring consistent across the films without uneven effects, strengthens polymer integrity, reduces gas permeability, enhances water resistance, 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 hydrogen bonding and electrostatic interactions between chitosan and 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, minimized aflatoxin and peroxide levels. Further, the plasma treatment of 120 s along with chitosan coating of concentration 1.5%, preserved hardness and color of pistachios during the storage. This combined effect of edible coating along with plasma treatment can be a cutting-edge technology for the preservation of fresh produce ensuring microbial safety along with the 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, better water vapour barrier (**Figure 3**). ATR-FTIR (Attenuated Total Reflectance-Fourier



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Transform Infrared) analysis revealed the 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 prime. Cold plasma treatment was applied on 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 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 on their study observed the effect of cold plasma treatment on the antimicrobial chitosan films containing thymol as an active ingredient. Plasma treatment significantly increased the 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 also the thymol diffusion rate increased after the treatment. Plasma treated chitosan-thymol films offers better antimicrobial efficiency, and controlled 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 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, directly correlated with the increased chitosan deposition level at this power. Furthermore, the surface-modified films showed strong antimicrobial activity against *Staphylococcus aureus*. In contrast, higher discharge powers (≥ 100 W) adversely affected film performance due to polymer degradation.⁴⁹ A drastic increase in water repellence character was observed with the contact angle reaching 121° , when ultrafast plasma modification was applied on chitosan/nanocellulose biocomposite films, while maintaining the films mechanical strength and barrier properties. 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 contact angle, atomic composition, and mechanical properties remain unchanged over the period of 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 offers a sustainable alternative to conventional packaging, aligning with global efforts towards green and smart packaging solutions.

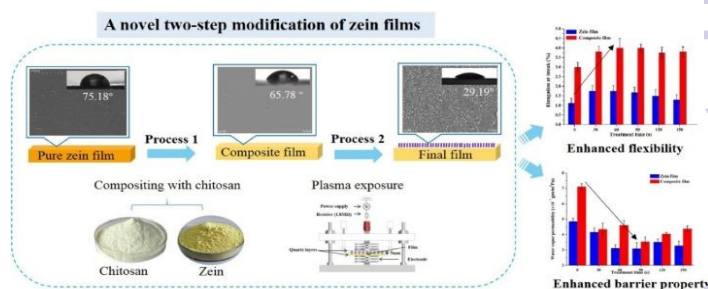


Figure 3. Development of composite film using cold plasma for packaging applications. Reproduced from Ref. [46] with permission from Elsevier, Chen et al., *Industrial Crops and Products*, 2019, 129, 318-326. Copyright 2019

Modification of chitosan using Irradiation

In the field of polymer science research and applications, ionic radiations are an innocuous for 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 within the biopolymer.⁵¹ Irradiation treatment is indeed an advantageous than any other treatments 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 not only improves the surface of chitosan films, but also modifies its internal structure, making it easier to use, more effective, and applicable in more diverse fields.^{52,53} The study by



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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. High-intensity excimer laser irradiation further altered surface morphology, producing micro-foam structures above the ablation threshold at 5 J/cm². 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 hydrophilicity, surface energy, and functional performance of chitosan films for packaging applications.⁵⁴ The combined use of chitosan/essential oils/AgNPs 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 –NH₂ 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 structural integrity, barrier properties, and bioactive performance of chitosan-based films.⁵⁶

Irradiation-Induced Structural, Barrier, and Antimicrobial Modifications of Chitosan Films

Polycationic polymer 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 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. Bionanocomposites film of chitosan/bacterial cellulose nanocrystals was found to have exceptional strength, biodegradability, and high surface area. Barrier properties, which tends to be one most fundamental factor for the preservation of fresh produce as it can accelerate the respiration and transpiration leading to post-harvest losses in fresh produce therefore, chitosan/bacterial cellulose nanocrystals composite films were modified using different ionization energy (0 KGy, 5 KGy, 10 KGy, 15 KGy), which enhanced the thermal and barrier properties and also a tightly packed structure was observed with increase in crystallinity after the irradiation treatment.^{58,59} Gamma –irradiation (60°C) with doses of 5, 10, 20 and 50 KGy was applied on chitosan with deacetylation value of 74.74%. The gamma irradiated chitosan films were prepared using solution casting method. The treatment increased the tensile strength, reduction in water vapour permeability as well with increase in *b values leading to brightness of the film (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 with the dose of 2.5 KGy. It was found that the lipid oxidation decreased by the treatment of irradiation and chitosan coating. The microbial activity (aerobic mesophilic and psychrotrophic counts) decreased with a minimum of 14 days shelf-life extension of the 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 the fresh produce with no difference in sensory attributes after the 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 rate, offering sustainable solution for various packaging and biomedical industries driving innovations towards sustainable material designs and also by comforting the market needs by contributing towards sustainable development goals.

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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 liquid medium. The bubbles disrupt vigorously, creating rise in temperature and high shear force leading to efficient dispersion and separation.⁶² Ultrasound generates strong cavitation effects, which results in localised high temperatures and pressures, 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, fruits 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 done 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 higher aspect ratio as evidenced from 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 upto 35%. Hence, from this study, it is apparent that ultrasonication is an efficient method for preparing of 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 also suitable for biodegradable edible coating and food packaging applications.⁶⁵ In today's fast paced and globalized food industry, smart packaging has deepened its roots as an integral part in extending shelf life, food safety and 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 concentration (2, 2.5 or 3 wt. %)

exhibits good physical and antimicrobial properties; were sonicated using water bath (ultrasound time, 25 min); sonication attributed in effective

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reduction of viscosity for the CS/PVA film by ensuring better dispersion and uniformity with enhanced mechanical strength, and water retention property, low oxygen permeability in the range of $0.16 \pm 0.08 \text{ cm}^2 \text{ m}^{-2} \text{ day}^{-1}$. The results also demonstrates that the film increases in firmness and decreases the weight loss, thereby, extending the shelf-life of strawberries upto 21 days and these films can serve as smart packaging and sustainable solutions, improving the storage and commercialization of strawberry and its preservation.⁶⁷ Rice protein is an excellent polymer to be used in edible coating, as they have excellent resistance to water vapour permeability but due to their low solubility, there are less protein interactions thereby reducing films flexibility, strength and durability. In this study, a novel composite of rice protein hydrolysates with chitosan has been used as an edible coating and treated using ultrasound. This combined effect resulted in reduced particle size of the film forming solutions as well as 200 W of ultrasound treatment was efficient enough for smoother film surface and better functional properties such as oxygen barrier, elongation at break which increased by 125%.⁶⁸ Composite film of collagen/chitosan was prepared and treated using novel technology sweep frequency pulsed ultrasound. It was observed that optimum ultrasonication time of 10 min, sweep frequency cycle of 100 s and pulse duty ratio of 77% significantly enhanced the tensile strength, elongation at break along with reduction in water vapour permeability rate of the composite film. X-ray diffraction (XRD) and thermal stability indices revealed a more crystalline order and higher thermo 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 modification of chitosan and its physical properties by bridging the gap to be the next generation dominant biopolymer.

Modification of chitosan using Pulsed electric fields



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With the growing demand for more sustainable food processing technologies, pulsed electric field has emerged as an innovative and a 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 milliseconds) with intensities 80 kV/cm, this dynamic technology is an effective way for modification of biopolymers also lower the electric field intensities are used for inactivation of microorganism, enhancement of food properties hence, by maintaining its nutritional and sensory properties.⁷¹

Biopolymer zein and chitosan were tailored with synthetic polymer PVA and pre-treated using pulsed electric field at different pulse energy. This study underscores the unique potential of pulsed electric field as a sustainable and pre-treatment method for the modification of biodegradable films, thereby, improving its mechanical, 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), pulse frequencies (50 – 300 Hz) and specific energies (80 – 650 KJ/kg) positively influenced the tensile properties and water stability of the tailored films by enhancing its 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, 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 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 field has emerged as a non-thermal technique for chitosan degradation, offering as an environmentally friendly alternative. Pulse electric filed was applied upto the strength of 25 KV cm⁻¹; with the increasing use of pulsed electric field the molecular weight of chitosan was lowered making it suitable applications requiring for low viscosity solution for drug delivery or film formation. Lower molecular weight chitosan produced by pulsed electric field shows enhanced bioactivity including antioxidant and antimicrobial properties.⁷³ Pulsed electric field provides a sustainable and controlled condition for reconstruction of

biopolymers, where, studies dictates that it is a powerful tool for modification of protein- polysaccharide (zein/chitosan) interactions, enhancing colloidal dispersions properties, promoting bioconjugation. Its ability to modify structural integrity and molecular interactions makes it a promising alternative to conventional processing techniques, fostering advancement in biomaterial science as well as in sustainable packaging industries. Treating zein-chitosan/polyvinyl alcohol film by pulse electric fields induced conformational changes in the structure of the biopolymeric films. It was observed that low intensity pulse electric field enhanced the stability of the edible film against enzyme degradation and also no toxicity was found. Pulse electric field modified edible film can be a potent technology for value addition for the application in edible food packaging.⁷⁴ Reinforcing microcrystalline cellulose with starch/chitosan and applying moderate electric field on the biopolymeric film improved the physiological properties of the film. The composite polysaccharide-based film showed excellent thermal properties, mechanical strength as well as the barrier properties. Surface morphology can be assessed through scanning electron microscopy (SEM), X-ray diffraction (XRD), which revealed that different concentration of microcrystalline cellulose along with moderate electric field are responsible for changes of properties of the films. It was observed that the application of moderate electric filed was efficient enough in decreasing the water vapour permeability in chitosan-based films. Therefore, this reinforced biopolymeric film along with moderate electric field can be a potent application for changing and modification of the properties of the film.⁷⁵ The use of pulse electric fields in application for the modification of biopolymer presents a promising technique for enhancing material properties such as conductivity, mechanical strength, and surface modifications. This method enables precise control over polymer structure, facilitating applications in diverse fields.

Modification chitosan using High pressure processing (HPP)

High pressure processing or high hydrostatic pressure (HPP) processing is a non–thermal technique for preservation of food, has gained much attention since late 20's. The first commercial use of high-pressure processing started in the land of rising sun (Japan) in the year 1980. Many food products such as yogurt, and jam has been produced by this procedure eventually showcasing



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the potential use of high-pressure processing as a non-thermal technology with minimum heat damage.⁷⁶ High pressure processing works on the principle of usage of high pressure around 300-800 MPa over a period of 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 the nutritional and sensory attributes, inactivation of enzymes, and modification of 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 physicochemical characteristics of biopolymers, leading to improved performance in packaging applications. This technique enhances the 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

Edible coating of chitosan/tea polyphenol along with high pressure processing as a synergistic effect has been applied on shrimp to increase the antioxidant and antimicrobial properties. This combined treatment of 400 MPa of high pressure along with chitosan /tea polyphenol coating; inhibited microbial growth, delayed spoilage, showed better oxidative stability and also 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 done 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/g decrease. This hurdle approach can be an effective way, making it a promising strategy for improving food safety in dry cured meats.⁸⁰ High hydrostatic pressure and nano-TiO₂ can also be applied in chitosan/polyvinyl alcohol-based films for enhancing the surface property. It was observed that high hydrostatic pressure enhanced the molecular interactions between the 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 200-400 MPa maintains the integrity of the film which suggests, resistant to degradation or dissolution and also reduce the migration of TiO₂ nanoparticles from the films; making them a potent candidate for sustainable and functional food packaging.⁸¹ Using 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. The sardine muscles when coated with the 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 for enhancing the shelf life of fresh produce at low cost without any energy consumption. High pressure processing is a promising sustainable technology enhancing the functional, preservation, modification of biopolymer by aligning with the environmental goals. Its ability to reduce energy efficiency thereby 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 the different non-thermal technologies such as cold plasma, irradiation, microwave, ultrasonication, ozone treatment, electrospinning, 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 physicochemical properties of chitosan without any thermal degradation. Microwave processing as a non-thermal technology, can be used to modify biopolymer by influencing its 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, the rapid and localised heating results in non-



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thermal effects. These effects include changes in the conformation of the polymer chains, which does not involve significant temperature increases. The microwave field induces mechanical stress, breaking or altering the molecular structure of biopolymers and potentially improving its solubility, viscosity or reactivity.⁸⁴ A study explores the surface modifications of microcrystalline cellulose using urea under the 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 can 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 the mechanical properties such as tensile strength, young's modulus and the fracture energy which is 2.0, 2.4 and 6.0 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 (40–80 °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, porosity with enhanced mechanical properties, making them useful in 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 transforms infrared spectroscopy (FTIR), X-ray diffraction (XRD), and thermal analysis revealed the reduction in crystallinity due to electrospinning which has made the film more amorphous through dissolution. By the addition of chitosan, the thermal stability increased as well water solubility test showed that the composite film can 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 showed profound antibacterial property. These fibres have large surface area making them more effective against *E. coli* and additionally due to the special surface texture of electrospun fibres, it hinders the cell wall of bacterial by disrupting their adhesion into the biopolymeric film and 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 potent candidate for antimicrobial packaging solution for perishable food products.

Additionally, the allotrope of oxygen is ozone (O₃) with higher oxidizing power. Ozone is a strong oxidizing agent that reacts with organic molecules by introducing functional groups such as hydroxyl, carbonyl and carboxyl group. Ozone treatment is considered safe to be used in food processing as approved by Food and Drug Administration (FDA). Ozone treatment alters the surface chemistry of biopolymer nanofibers or films by introducing oxygen-containing functional groups. This enhances hydrophilicity, wettability, mechanical as well as the barrier properties of the film.⁹⁰ Additionally, PHA is the class of biodegradable biopolymers produced by microorganisms, they possess excellent biodegradability and biocompatibility but generally lack strong antimicrobial properties; so as to enhance its antimicrobial properties they are tailored with other antimicrobial agents such as chitosan and chitooligosaccharides using ozone treatment. This combined treatment



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enhances the antibacterial and biodegradable properties. Researcher claims 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 also 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 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 also increases moisture absorption, making them particularly effective for fruit preservation and antimicrobial food wraps. This ozone treated composite food coating wraps can be a potent source for extending the shelf life and minimizing the post-harvest losses.⁹² Across different non-thermal technologies 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. Such 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 chitosan's functionality without degrading its structure as illustrated in **Figure 4**. These methods improve its biodegradability, making it eco-friendlier and more suitable for sustainable applications. Further, the enhanced biodegradation supports waste reduction and aligns with circular economy principles by promoting resource efficiency. This approach enables the development of biodegradable materials for agriculture, food packaging industries thereby, reducing environmental impact.

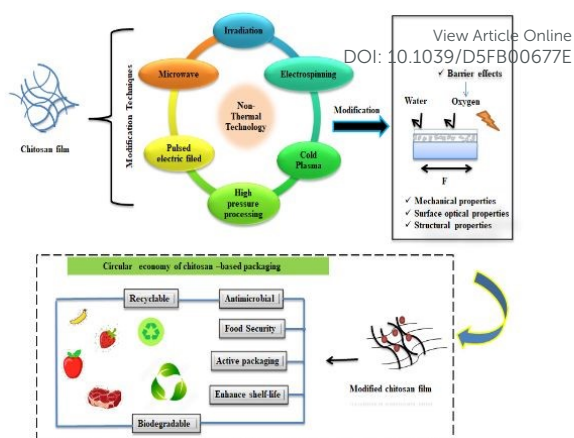


Figure 4. Non-thermal modification of Chitosan: Enhancing biodegradation for circular economy.



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DOI: 10.1039/D5FB00677E**Table 2. Modification of chitosan-based films using non-thermal technologies.**

SI no	Material	Non-thermal technologies	Outcome	References
1.	•Chitosan/wampee seed essential oil	• Cold plasma treatment	<ul style="list-style-type: none"> • Provides synergistic effects due to the combined use of edible coating of chitosan along with cold plasma treatment on golden pompano fillets. • The treated fillets showed the lowest bacterial count, peroxide value, 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)	<ul style="list-style-type: none"> •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 highest stability during the 31 days of storage period. 	[50]
3.	•Chitosan	•Pulsed electric field/Ozone treatment	<ul style="list-style-type: none"> •Combined effects resulted into lower the molecular weight of chitosan ($M_w < 2,500$ Da) and lower the crystallinity. 	[94]
4.	•Chitosan/Grape	•Gamma-irradiation	<ul style="list-style-type: none"> •Used as an edible coating on chicken breast meat. 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 retained during the entire 21 days of storage period. 	[61]
5.	•Chitosan/essential oil/silver nanoparticles	•Gamma-irradiation	<ul style="list-style-type: none"> •Provides 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 (%) retained during the 12 day of storage period. 	[55]
6.	Quinoa protein/chitosan/transglutaminase (TG)	• Ultrasonication	<ul style="list-style-type: none"> •The thermal stability of the composite film increased after ultrasonication treatment. •The treatment enhanced the structural and mechanical properties of the composite films. 	[95]



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Sl no	Material	Non-thermal technologies	Outcome	References
7.	• Chitosan/ <i>Sargassum palladium</i> polysaccharides	• Ultrasonication	<ul style="list-style-type: none"> •Ultrasonication enhanced the transparency, elongation, and tensile strength of the film along with the antioxidant activity has also increased. •The treated films extended the shelf life of strawberry, inhibiting deterioration at room temperature (25° C) during 7 days of storage. 	[96]
8.	•Chitosan/ mandarin essential oil	• High pressure processing (HPP)/ pulsed light processing	<ul style="list-style-type: none"> •The combination of edible coating along with HPP on green beans resulted in significant reduction of <i>L. innocua</i>, thereby, increasing the shelf life. •The treatment also retained the firmness of green beans. 	[97]
9.	•Chitosan/Starch/ microcrystalline cellulose	•Pulsed electric field	<ul style="list-style-type: none"> •The treated composite film presented good thermal properties as well as mechanical properties, generating more rigid films. 	[75]
10.	•Chitosan / Polyvinyl alcohol/essential oils	• Electrospinning	<ul style="list-style-type: none"> •Antioxidant activity showed highest for the composite film. •The bilayer lamination of the composite film increased the thermal resistance with increase in antimicrobial activity. 	[98]
11.	•Chitosan	•High hydrostatic pressure (HHP)	<ul style="list-style-type: none"> •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 protective effect both chemically and microbiologically. 	[99]
12.	•Chitosan/ cinnamon oil	•Cold plasma treatment	<ul style="list-style-type: none"> • The strength as well as the antioxidant property of the films increases after the treatment. •Cold plasma induced composite film showed higher antimicrobial activity along with improved oxygen barrier properties. 	[47]



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Circular economy and Life cycle assessment of chitosan-based packaging

The circular economy approach to the non-thermal modifications of chitosan focuses on the sustainable processing methods that minimize energy consumption and waste generation. By utilizing the non-thermal modification techniques such as cold plasma, irradiation, pulsed electric field, high pressure processing, electrospinning etc. chitosan can be tailored for various applications in food, biomedical, developing 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, LCA of cellulose nanofibrils (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 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, supporting sustainable packaging solutions.

Challenges in 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 to 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, large-scale industrial implementation of chitosan-based coatings remains challenging because of 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 burden. 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



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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 protocol is essential for successful commercial implementation.

Conclusions

The biopolymer chitosan has recently drawn researcher's attention due to its excellent antimicrobial, antioxidant properties, and film forming abilities. It is being developed from a naturally available sources with historical significance to a highly promising material for sustainable food packaging. Moreover, its non-toxicity and biocompatibility feature provides an eco-friendly alternative to conventional synthetic plastics materials. Owing to these attractive properties, chitosan has found usage in number of fields such as medicine, agriculture, biomedical, food packaging, waste water treatment, biosensors, textile industries, etc. 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 a potential candidate for enhancing chitosan's functional and structural properties without compromising its bioactivity thereby, aligning to 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. Non thermal modification of chitosan plays a pivotal role in sustainable development enhancing its properties in energy- efficiency, environmentally friendly, and economically viable matter. These modifications enable the widespread adoption of biodegradable high-performance chitosan-based films, contributing to waste reduction, food preservation as well as reduction in fossil fuel- based plastic usage. With the increasing market demand, industries seek greener

alternatives for packaging; non-thermal modification can be a resilient pathway for achieving the global sustainable development goals thereby maintains material efficiency and functionality.

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According to researcher's, future research should focus on optimizing non-thermal modification strategies for industrial scale application, cost-effectiveness, developing hybrid biopolymer composites for enhancing performance as well as with 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 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 optimise material design. Recent advances in surface wettability and sustainable coating technologies highlight the critical role of surface functionality in governing barrier performance of packaging materials.¹⁰²

Future research should also prioritise the development of standardised processing protocols, long-term performance evaluation, and real-world



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application trials, supported by strong industry–academia collaborations to bridge the gap between laboratory-scale innovations and commercial implementation.

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Author contributions

Nurin Afzia: Original draft writing, review and editing.

Tabli Ghosh: Conceptualization, supervision, validation, final review and editing.

Data availability

No datasets were generated or analysed during the study.

Declarations

Ethical approval

Compliance with ethical approval.

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Declaration of competing interest

The authors declare no conflict of interest.

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Data availability

No datasets were generated or analysed during the study.

