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Review

Recent Progress in Functional Edible Food Packaging Based on Gelatin and Chitosan

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Abstract: Nowadays, edible and eco-friendly packaging applications have been studied as an alternative to conventional/synthetic packaging due to the great interest of consumers in healthy, safe, and natural food, and of researchers in meeting the needs of consumers and producers. Various biopolymers are being extensively explored as potential materials for food packaging. The edible biopolymers utilized so far for packaging applications include proteins, lipids, and polysaccharides. Occasionally, these biopolymers have incorporated different bioactive substances to enhance the composite films' characteristics. Gelatin and chitosan are two of the most important biopolymers for the production of films. Different biopolymers or bioactive substances have been incorporated into the matrix to enhance the gelatin-based and chitosan-based films. By incorporating other biopolymers and bioactive compounds, the composite films' overall physicochemical and mechanical characteristics are improved. Additionally, by incorporating bioactive compounds (polyphenolic compounds, natural extracts, and essential oils), the composite films present important biological properties, such as antioxidant and antimicrobial activities.

Keywords: gelatin; chitosan; films; bioactive compounds; biopolymers; packaging application

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1. Introduction

The main direction of food packaging is to preserve the quality and aspect of products, and this can be obtained by reducing lipid oxidation, inhibiting microbial growth, and therefore extending the shelf life of food products. The conventional packaging is mostly made from petroleum-based plastics [1,2]. The manufacturing of plastic globally has grown in the last decades, and 40% of the plastic produced is utilized in packaging applications [3]. Even though plastic is advantageous as a packaging element because it is low-priced, it has a light weight, and its facility in form molding, excellent mechanical strength, and thermal sealing, the sizeable utilization of plastic packaging may also result in unfavorable outcomes for the environment [4–6]. These adverse environmental effects of plastic packaging are related to its low biodegradability and reduced reuse and recycling. Therefore, large quantities of plastic may cause world contamination and pollution [7,8].

Nowadays, edible and eco-friendly packaging applications have been studied as an alternative to conventional/synthetic packaging, due to the great interest of consumers in healthy, safe, and natural food, and of researchers to meet the needs of consumers and producers and to obtain biodegradable and nontoxic films/coatings for the food industry. Therefore, various biopolymers are being extensively explored as potential materials for food packaging. The edible biopolymers tested so far for packaging applications include proteins, lipids, polysaccharides, and all achievable mixtures among these. Occasionally, these biopolymers or their combinations have incorporated different additives, such as antioxidants, antimicrobials, flavors, or colors, to enhance the characteristics of the films [4,9].

Additionally, controlling the migration of components from packaging materials to foods is essential because it could result in the transfer of undesirable compounds that could reduce the safety of food for consumption or change its sensory and nutritional properties [10]. However, migration may also be desirable whenever the incorporated compounds are meant to be released gradually over time to preserve the food from any unfavorable chemical reactions, and thus extending the shelf life [11]. For this reason, it is essential to also research the components that can be passed from the packaging to the food.

Protein and polysaccharides are the most suitable and renewable biopolymers for food packaging applications [12,13]. Films/coatings based on gelatin and chitosan have been studied intensively in the last few years [14–17].

This review highlights the recent progress in food packaging based on gelatin and chitosan or on combinations of them or with other biopolymers and bioactive compounds. The overview also provides the most important physical, chemical, mechanical, and biological characteristics of the obtained composite films and their possible applications for the food industry.

2. Gelatin

Gelatin is one of the important components of protein-based packaging, and it can be obtained from collagen by its partial hydrolysis. It possesses the capacity to form adequate films for the food packaging industry [18,19]. The primary rheological properties of gelatin are bloom and viscosity, and these properties are typically the outcome of the production process utilized. The average molecular weight, amino acid content, and chain polymerization level are all connected to the viscoelastic characteristics [20]. Commonly, gelatin is obtained from certain mammals, like pork and cow, or poultry [19,21].

Nowadays, alternative gelatin sources, such as gelatin from various fish species, are being explored [19,22]. Due to its functional qualities, such as its capacity to bind water, produce gels, operate as a gas barrier, form films, create foam, and have an emulsification property, gelatin is widely utilized in the food, pharmaceutical, photographic, and cosmetics sectors [23].

Although gelatin exhibits excellent gas barrier and swelling properties, it has poor mechanical resistance and is permeable to water vapor. Gelatin's poor water vapor barrier characteristic limits its application as a packing material. The limitations of gelatin can be improved by combining it with other valuable components [4,9,14,24].

According to previous research, the performance of gelatin for food packaging applications has been improved by combining gelatin with other biopolymers, such as chitosan, starch, soy protein, pectin, and carboxymethylcellulose (CMC) [25–30], or with other natural compounds, such as polyphenols [31–35] and essential oils [36–45]. For instance, combining gelatin with chitosan, starch, and tapioca starch improved the mechanical characteristics [25,26,29]. Incorporating polyphenols in the gelatin matrix was reported to add antioxidant and antimicrobial activities for the composite films [19,31,32,35]. Also, a gelatin–essential oil composite presented low WVP in comparison with the native gelatin films [37–39,41,42]. Figure 1 contains a schematic representation of gelatin types and origination, and the compounds that can be incorporated in the gelatin matrix.

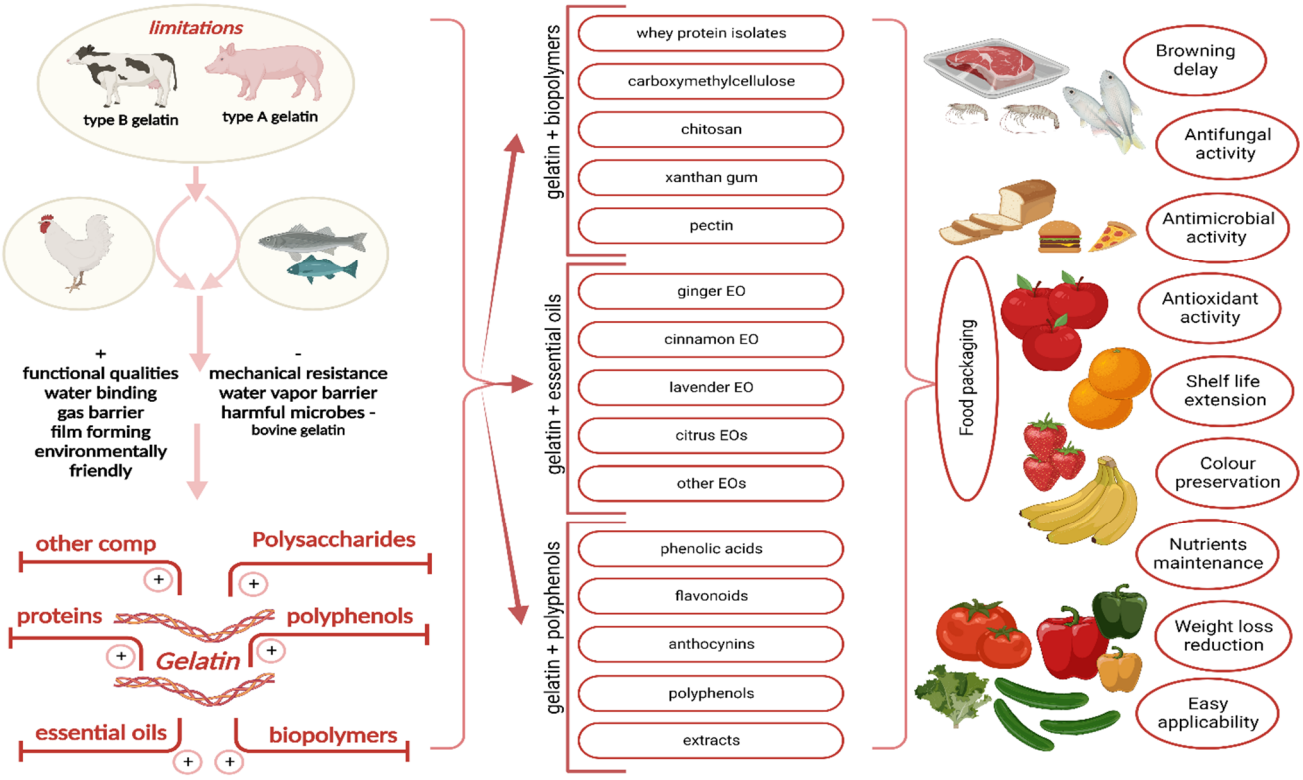

Figure 1. Schematic representation of gelatin matrix incorporated with functional materials for improving the characteristics of the composite films.

2.1. Origination of Gelatin

2.1.1. Gelatin Obtained from Mammals

Most gelatin sources originate from mammals, particularly cattle and pigs, with pig skin representing 46% of all sources, bovine hide approximately 29%, and pork and cattle bones 23% [46]. Due to their abundant availability, bovine and porcine skin gelatins are used extensively in the food sectors.

Gelatin is produced by partial hydrolysis of collagen, and is classified into two types. Gelatin from bovine skin is usually defined as type B gelatin and is prepared through an alkaline method. In contrast, gelatin from porcine skin is defined as type A gelatin and is prepared through an acidic method. Type A gelatin has an isoelectric point at pH 7–9.4, and has higher amino acid content compared to type B, which has an isoelectric point at pH 4.8–5.5. Due to its greater gel characteristics (gel strength and viscosity), and powerful film-forming properties, mammalian gelatin is more often used than other sources [24].

Mammal gelatins, however, have considerable limitations and issues regarding religious concerns, as Muslims, Jews, or Hindus cannot use or ingest them for various reasons [47]. Furthermore, substitutes for porcine and bovine gelatin replacement have also been prioritized and taken into consideration due to the possible risk of transmitting harmful microbes from bovine spongiform encephalopathy, known as mad cow disease, and from foot and mouth diseases [48].

Therefore, due to these limitations of mammalian gelatin and the need to use gelatin from different sources, researchers' interest in gelatin obtained from other sources, like poultry and aquatic species, has grown considerably [47].

2.1.2. Gelatin Obtained from Poultry

Poultry skin, foot, and bone represent an alternative to mammalian gelatin. Duck, chicken, and turkey species are among the poultry sources used for this purpose. According to several studies, poultry gelatin is similar to mammalian gelatin in terms of its amino acids, secondary structure, and molecular weight (285 g/mol for poultry and 350 g/mol for mammal gelatin) [49,50].

Compared to bovine gelatin, the gel derived from chicken skin and chicken feet seems to have a much higher bloom value, according to Sarbon et al. [49] and Rahman and Jamalulail [51].

Meanwhile, according to Nik Muhammad et al. [52], commercial bovine gelatin had a bloom value of 217 g, but duck feet gelatin obtained by diverse acids treatment had a greater bloom strength (226–334 g). A higher percentage of cross-linked ß and α chain components results in high bloom strength and leads to higher melting temperatures and viscosity. Additionally, it was observed that the gelatin from chicken skin and duck feet contained amino acids like glycine, proline, hydroxyproline, and alanine, which helped increase the gel's strength and stability. Interestingly, it was found that the imino acid content (e.g., proline, hydroxyproline) of duck-feet- and chicken-skin-derived gelatins was higher than that of the bovine gelatin [37,49].

Gelatin derived from poultry products has good film-forming characteristics due to its high imino acid concentration and high bloom value [49,52].

2.1.3. Gelatin Obtained from Aquatic Species

An alternative to mammalian gelatin comes from marine sources, such as warm- and cold-water fish (skins, bones, and fins). Marine gelatin sources are not connected to the risk of bovine spongiform encephalopathy epidemics. In addition, it is suitable for usage by Muslims, Jews, or Hindus, where mammalian gelatin is prohibited [47].

Fish processing byproducts can be used as an alternative raw material for preparing high-protein ingredients, since protein makes up the majority of components of most fish. This is especially true for producing food-grade gelatin due to the significant amounts of collagen in fish [22].

Compared to mammalian gelatin's bloom values, fish gelatin often has a lower bloom value due to the variances in proline and hydroxyproline content, which depends on the fish species and environment temperature. Depending on the type of fish, the environment, and the extraction technique utilized, there may be variations in viscosity values. Proline and hydroxyproline concentrations in fish gelatins are typically lower than those in the mammalian gelatins [24].

However, several studies have reported that warm-water fish gelatins have higher imino acid levels when compared to cold-water ones [53,54].

Regarding film characteristics, fish gelatin shows potential linked to attributes such as remaining translucent, nearly colorless, water-soluble, and very extensible [55].

2.2. Gelatin-Based Composites

2.2.1. Combined Gelatin and Other Biopolymers

The variety of biopolymer combinations' physical, chemical, and textural features have been the subject of intense research in the last years to generate novel products. The formulation and characterization of combined gelatin–biopolymers films are summarized in Table 1.

In a study published by Howell, three ways were outlined that proteins might be described in terms of how they interact with other biopolymers: synergistic interactions, aggregation, and phase separation. These three characteristics may lead to fascinating and technically valuable applications. It has been noted that synergistic interactions can improve gelation qualities beyond those of the individual protein utilized alone [56], and they were observed in gelatin–whey protein isolate [57] and whey–egg albumen mixtures [58]. Proteins aggregation may occur due to electrostatic interactions, and it could be useful for improving gelation in case of β-lactoglobulin [59]. Combining two biopolymers may also occur with phase separation. In the composite obtained, the biopolymers produced separate phase networks. Phase separation has been reported in protein–protein and protein–polysaccharide mixtures [57].

Sarbon et al. [57] investigated the physical, thermal, and microstructural properties of the gelatin–whey protein isolate mixture by using a large deformation rheological test (heating gelation followed by cooling and compression). All combinations of gelatin and whey protein isolate produced gel strength values that were higher than anticipated, indicating a synergistic interaction and improvement of the gelling properties of both the gelatin and whey proteins.

Several studies also outlined the possible mixture between protein and polysaccharides. Gelatin–chitosan composite was prepared and characterization was performed of its physical and mechanical properties. The findings showed that adding chitosan significantly increased the elastic modulus (EM) and tensile strength (TS), making the films stronger than gelatin films. However, adding chitosan significantly lowered the elongation at break (EAB) characteristic. According to the structural characteristics examined, gelatin and chitosan interacted to create a novel material with improved mechanical performance [25].

Incorporating gelatin with CMC also highlighted some important modifications, such as increased TS and puncture test of the films, water vapor permeability (WVP), reduced EAB, opacity, and UV-light penetration of the films, and it increased the thermal stability. By using Fourier transforms infrared spectroscopy (FTIR) and X-ray diffraction (XRD) analyses, it has been confirmed that the functional groups of gelatin interact strongly with CMC. With the addition of CMC to gelatin-based film, crosslinking and intermolecular bonds were established and improved some of the mechanical and physical properties of the film [60].

The mechanical and physical characteristics of edible films were found to be affected by the addition of potato starch to gelatin-based film. With increasing potato starch concentrations, the TS, EM, transparency, thermal characteristics, WVP, ultraviolet, microscopy, and visible light barrier transmission improved, while the EAB lowered. This may suggest promising developments for the insertion of potato starch as a potential crosslinking agent to enhance the mechanical and physical characteristics of gelatin-based films, particularly in the context of the production of food packaging materials [61].

A formulation of three polymers was also studied. Gelatin was combined with CMC and chitosan [62,63]. Jahit et al., showed that chitosan and the CMC addition greatly impact the film's characteristics. The film's amorphous nature was minimized by making it more crystalline as the chitosan concentrations increased. Given that the formulation's gelatin/CMC/chitosan ratio of 60/30/10 exhibited the second-lowest WVP (2.250×10^{-7} g·mm·h^{-1}·cm^{-2}·Pa^{-1}) and the highest biodegradability rate, it seems ideal for prospective usage in the food packaging [62].

Table 1. The characteristics of the gelatin–biopolymers mixtures.

Formulation	Physical/Chemical/Mechanical/Biological Characteristics	References
Gelatin, whey protein isolate	synergistic interaction ↑ gelling properties, EM	[57]
Gelatin, soy protein isolate	↑ mechanical properties when the weight ratio of soy protein isolate: gelatin is 1:3	[64]
Gelatin, soy protein isolate	↑ TS, EAB, EM, flexibility	[27]
Gelatin, chitosan	↑ mechanical properties ↓ permeability good UV-light protection qualities	[25]
Gelatin, CMC	↑ TS, puncture test of film, thermal stability, WVP, ↓ EAB, opacity, and UV-light penetration of the films	[60]
Gelatin, CMC, chitosan	↓ WVP ↑ biodegradability	[62]
Gelatin, CMC, chitosan	↑ flexibility, EAB, WVP, thickness ↓ TS and puncture force	[63]
Gelatin, chitosan, xanthan gum	↑ thickness, WVP, UV-light protection, thermal stability, ↓ TS, EAB, VIS light transparency	[28]
Gelatin, starch	↑ mechanical strength, water solubility (WS), WVP, thickness ↓ opacityimproved appearance of refrigerated Red Crimson grapes	[26]
Gelatin, potato starch	↑ TS, EM, WVP, melting temperature, UV–VIS light protection ↓ WS, EAB	[61]
Gelatin, tapioca starch	↑ TS, EAB, thickness, WVP, UV-light protection, thermal stability visible light transmission, film transparency	[29]
Gelatin, pectin	↑ thickness, TS, antioxidant, and antibacterial activities ↓ WVP, EAB	[30]

↑-increased values of the tested characteristics, ↓-decreased values of the tested characteristics.

2.2.2. Combined Gelatin and Polyphenols/Extracts Rich in Polyphenols

An extensive and increasing list of bioactive substances have been or are now being integrated into films, with phenolic compounds (polyphenols, phenolic acids, flavonoids, anthocyanin) (Table 2) being the most prevalent [31,32,65–68].

Plant extracts represent an important source of polyphenols. These compounds have antioxidant and antimicrobial effects; therefore, the incorporation of polyphenols in the biopolymers matrix leads to composite films with antioxidant and antimicrobial activities. Based on that result, several studies have used these compounds in bioactive and biodegradable films [19,35,66,69–72].

The capacity of the active groups in gelatin-based films to quench radicals plays a significant role in the antioxidant activity of those films. The gelatin protein's amino acid groups provide to native gelatin films poor antioxidant activity [73,74]. However, a gelatin film's antioxidant activity is increased when it is conjugated with diverse phenolic compounds [19,31,32,67,71,74].

Both 2,2-azino-di-3-ethylbenzthiazoline-6-sulfonate (ABTS) and 2,2-diphenyl-1-picrylhydrazyl (DPPH) radical scavenging and reducing power analyses are used to measure the antioxidant activity of gelatin-based films. Using the ABTS and DPPH scavenging methods, Hanani et al., evaluated the antioxidant capacity of gelatin-based films. The radical scavenging ability for the control (only gelatin) film was 32% for ABTS and 53% for DPPH assay. However, the film's antioxidant activity was significantly enhanced by adding 1% pomegranate peel powder, and the ABTS and DPPH scavenging activities rose to 48% and 60%, respectively. The antiradical activity was significantly improved with the addition of pomegranate peel powder, as seen by the increase in ABTS and DPPH radical scavenging activity as pomegranate peel powder concentration increased in the gelatin-based films. The ABTS and DPPH radical scavenging activity of the gelatin film with 5% pomegranate peel powder was the highest, with 80% and 72%, respectively. Pomegranate

is well known for its high content of bioactive substances, including phenolic compounds and anthocyanins, which are strong antioxidants and may scavenge DPPH and ABTS radicals [71].

Gelatin-based films are also tested for their antioxidant properties using the reducing power assay. Wu et al., observed that the reducing power activity of gelatin film was low and gelatin film with 0.7% green tea extract exhibits a reducing power of 65% of 1.0 mg/mL vitamin C. These findings showed that the gelatin–green tea extract film's antioxidant activity was enhanced in a concentration-dependent manner in comparison with the gelatin film without the extract [69].

Similar results were obtained when gelatin films incorporated rosmarinic acid [67], chlorogenic acid [65] and grape seed extract, and gingko leaf extract [70].

Gelatin-based films with antimicrobial properties are a crucial barrier in preventing the spread of foodborne infections. Incorporating rosmarinic acid into gelatin film provides antimicrobial action with a lengthy half-life [74]. Pathogenic bacteria such as *Escherichia coli* and *Staphylococcus aureus* aggregated following treatment with gelatin-rosmarinic acid films, exhibiting a morbid morphology and afterward being fully lysed. Even after three months of storage, it was discovered that gelatin–rosmarinic acid films possessed substantial antimicrobial activity, indicating that these films offer major benefits in food packaging [74].

The antimicrobial activity of gelatin-based films containing phenolic compounds or plant extracts rich in polyphenolic content was also reported by several studies. Gelatin–protocatechuic acid [31], gelatin–epigallocatechin gallate [32], gelatin–tannic acid [68], gelatin–mangrove extract [19], gelatin–pomegranate peel extract [71], and gelatin–date by-products [72] films displayed good antimicrobial activity against Gram-negative (*E. coli*) and Gram-positive (*S. aureus*) bacteria.

Fu et al., obtained a gelatin–chlorogenic acid film with antioxidant and antimicrobial activity against *E. coli*, *Pseudomonas aeruginosa*, *Listeria monocytogenes*, and *S. aureus*, with potential applications in fresh seafood preservation [65].

Moreover, phenolic compounds can form hydrogen and hydrophobic interactions. Hydrogen and hydrophobic interactions between the hydroxyl groups present in the aromatic rings of phenolic compounds and the carboxyl groups of gelatin side chains might improve gelatin film functional characteristics [73].

One study designed a gelatin film including haskap berries extract, where the phenolic components of the extract, mainly anthocyanins and phenolic acids, generated hydrogen crosslinking between the hydroxyl groups of the phenolic compounds and the amino/hydroxyl groups of the gelatin. This crosslinking improved mechanical strength, flexibility, air, WVP, film brightness, and WS. Consequently, haskap berries extract enhanced the capacity of gelatin composite films for application in active packaging [75].

The incorporation of phenolic compounds in gelatin-based films led to the improvement of the functional properties. The protection and tamper-resistance of food packaging are significantly influenced by the TS of the packaging materials. Higher tensile strengths are typically chosen for a range of packing items as they provide a stronger seal with safe load stability and help to produce higher-quality products for the customer [24].

Several studies showed that phenolic compounds increased gelatin-based film TS yield [19,66–69,71,75]. EAB is the ratio of the modified length to the starting length when the sample is damaged. It refers to the ability of a plastic sample to withstand shape changes without developing cracks [24].

Various researchers concluded that adding natural extracts, such as phenolic compounds, to gelatin-based films increased the films' extensibility and EAB values. These compounds may have a plasticizing effect on the resulting films [19,31,67,74,75].

Good oxygen and moisture protection properties are essential for food packaging films since too much oxygen or moisture can cause lipid oxidation and microbiological degradation of food during transport or storage. Packed products' quality and storage life can be significantly enhanced when the packaging films serve as practical barriers against

oxygen or water. As a result, it is essential to keep WVP as low as achievable. Currently, gelatin film linked with phenolic compounds can produce composite films with WVP lower than simple gelatin films [19,31,35,67–70,74,75].

Table 2. The characteristics of gelatin–polyphenol mixtures.

Formulation	Physical/Chemical/Mechanical Characteristics	Biological Properties	Applications	References
Gelatin, protocatechuic acid	↑ thickness, EAB achieved fine look, ↓ light transmittance, TS, WVP	Antioxidant activity (DPPH), antimicrobial activity against *E. coli* and *S. aureus*, with high protocatechuic acid amounts.	Beef preservation	[31]
Gelatin, epigallocatechin gallate (EGCG)	↑ bloom strength	Antioxidant activity (DPPH (50%–99%), FRAP (200–662 µg Vc/g)), antimicrobial activity against *E. coli* and *S. aureus*	Active packaging	[32]
Gelatin, *Galla chinensis* extract	↑ gel strength and thermal stability, ↓ swelling of gelatin	Not determined	Packaging	[33]
Gelatin, eugenol/β-cyclodextrin emulsion	not determined	Reduced the H_2S-producing bacteria, total viable *Pseudomonas* spp. and Psychrophilic counts, total volatile basic nitrogen, K value, free fatty acids	Chinese Seabass during superchilling storage	[34]
Gelatin, mango peel	↓ WVP, solubility films more rigid and less flexible	Antioxidant activity (DPPH 70%–85%)	Active packaging	[35]
Gelatin, green tea extract grape seeds extract gingko leaf extract	↓ TS, EAB, lowest WVP lowest TS, EAB, ↓ WVP ↓ TS, EAB, WVP	All the films presented antioxidant activity (DPPH)	Active food packaging	[70]
Gelatin, *Fructus chebulae* extract	↑ gel strength, thermal stability	Not determined	Packaging	[76]
Gelatin, chlorogenic acid	not determined	Antioxidant activity (ABTS), antimicrobial activity against *E. coli*, *P. aeruginosa*, *L. monocytogenes*, and *S. aureus*	Fresh seafood preservation	[65]
Gelatin, epigallocatechin gallate	↑ TS, EM, ↓EAB	Antioxidant activity (DPPH 67%)	Reduce the oxidation of cod-liver oil	[66]
Gelatin, green tea powder	↓ TS, EM, EAB with high amounts of green tea powder	Antioxidant activity (DPPH 77%)	Reduce the oxidation of cod-liver oil	[66]
Gelatin, green tea extract	↑ TS↓ EAB, WS, WVP	Antioxidant activity (DPPH 15%–55%)	Active packaging	[69]
Gelatin, rosmarinic acid	↑ thickness, TS, EAB, light protection, ↓ WS, WVP	Antioxidant activity (DPPH 75%–90%)	Bacon preservation	[67]
Gelatin, rosmarinic acid	↑ EAB, ↓ TS, EM, WVP	Antioxidant activity (ABTS), antimicrobial activity against *E. coli* and *S. aureus*	Active packaging	[74]
Gelatin, tannic acid	↑ TS ↓ EAB, WVP, oxygen permeability	Antimicrobial activity against *E. coli* and *S. aureus*	Cherry tomatoes, grapes	[68]
Gelatin, mangrove extracts	↑ thickness, EAB, TS ↓ WVP	Antioxidant activity (DPPH 15%–60%), antimicrobial activity against *S. aureus*, *E. coli*, *Bacillus subtillis*, *Salmonella* sp.	Active packaging	[19]
Gelatin, pomegranate peel powder	↑ thickness, WVP, TS ↓ film solubility, EAB	Antioxidant activity (DPPH 59%–72%, ABTS 48%–80%), antimicrobial activity against *S. aureus*, *L. monocytogenes*, and *E. coli*	Active packaging	[71]
Gelatin, haskap berries extract	↑TS, EAB ↓WVP, WS	Antioxidant activity (DPPH)	Shrimp spoilage	[75]
Gelatin, date by-products	↓water holding capacity, WS color change	Antimicrobial activity against *E. coli* and *S. aureus*	Active packaging	[72]

↑-increased values of the tested characteristics, ↓-decreased values of the tested characteristics.

2.2.3. Combined Gelatin and Essential Oil

The antimicrobial and antioxidant capacities of the essential oils from plants and spices make them valuable food additives. Moreover, by reducing lipid oxidation, essential oils can increase the shelf life of food products. Terpenic and phenolic compounds, biologically active substances, are abundant in essential oils. Additionally, most of them are declared to be Generally Recognized as Safe. However, due to their robust flavor, their application as food preservatives is frequently restricted. Therefore, essential oils can be added to the edible film to avoid this issue [77,78]. The formulation and characterization of combined gelatin–essential oil films are summarized in Table 3.

Table 3. The characteristics of gelatin–essential oil mixtures.

Formulation	Physical/Chemical/Mechanical Characteristics	Biological Properties	Applications	References
Gelatin, ginger essential oil	↑ thickness, WVP, EAB, ↓ TS	Antimicrobial activity against *E. coli* and *S. aureus*	Antimicrobial active packaging	[36]
Gelatin, cinnamon leaf oil	↓ TS, slightly decreased WVP	Antimicrobial activity against *Salmonella typhimurium*, *E. coli*, *L. monocytogenes*, and *S. aureus*	Cherry tomatoes	[37]
Gelatin, oregano essential oil	Insignificant modification	Antioxidant activity (DPPH 12%–60%, FRAP), antimicrobial activity against *E. coli* and *S. aureus*	Food active packaging	[38]
Gelatin, lavender essential oil	↓ WVP, TS	Antioxidant activity (DPPH 1%–9%, FRAP), antimicrobial activity against *E. coli* and *S. aureus*	Food active packaging	[38]
Gelatin, thyme essential oil	↑ EAB, ↓ TS, WVP	Antimicrobial activity against *L. monocytogenes* and *E. coli*	Chicken tenderloin packaging	[39]
Gelatin, citrus essential oils (bergamot, kaffir lime, lemon, lime)	↑TS, ↓EAB, WVP (glycerol 20%)↑ EAB, ↓ TS (glycerol 30%)	Antioxidant activity (DPPH, ABTS, FRAP)	Active packaging	[40]
Gelatin, root essential oils (ginger, turmeric, plai)	↑ EAB, ↓ TS and WVP	Antioxidant activity (DPPH, ABTS)plai > turmeric > ginger essential oils	Active packaging	[41]
Gelatin, *Zataria multiflora* (thyme-like plant) essential oil	↑ WVP, EAB, light barrier properties, ↓ TS	Antioxidant activity (ABTS), antimicrobial activity against *P. aeruginosa*, *E. coli*, *S. aureus*, *B. subtilis*	Antioxidant, antimicrobial active packaging	[43]
Gelatin, essential oils (bergamot, lemongrass)	↓ TS, EAB, WVP (lemongrass), solubility, transparency ↑ heat stability	Antimicrobial activity Lemongrass: *E. coli*, *L. monocytogenes*, *S. aureus*, *S. typhimurium* Bergamot: *L. monocytogenes*, *S. aureus*	Active packaging	[42]
Gelatin, essential oils (clove, garlic, origanum)	↓ thickness, WS, EABslightly decreased WVP	Antioxidant activity (DPPH 38%–72%), antimicrobial activity against *Brochothrix thermosphacta*, *Listeria innocua*, *L. monocytogenes*, *Shewanella putrefaciens*	Biodegradable food packaging systems	[44]
Gelatin, sage essential oil	↓ WVP, ↑ thickness	Antimicrobial activity against *E. coli*, *S. aureus*, *L. innocua*, *Saccharomyces cerevisiae*, *Penicillium expansum*	Fruits, vegetables, and meat packaging	[45]

↑-increased values of the tested characteristics, ↓-decreased values of the tested characteristics.

Essential oils added to edible films, in this case, gelatin films, lead to an increase in the gelatin film's biological activity and water resistance [78].

In recent years, researchers have focused on the analysis of incorporating essential oils into gelatin-based films. In a study conducted by Tongnuanchan et al., citrus essential oils were added to a gelatin-based film, which decreased WVP, and the obtained films displayed antioxidant activity [40].

Similar results, such as decreased WVP, were obtained when the gelatin-based films incorporated cinnamon leaf essential oil (0.5 %) [37], lavender essential oil (2000–6000 ppm) [38], thyme essential oil (0.5, 1, 1.5%) [39], root essential oil (ginger, turmeric, plai, different levels, 25%, 50%, and 100%, based on protein content) [41], lemongrass essential oil (5%–25% (*w*/*w* protein)) [42], garlic and clove essential oil (1 μL/cm^2 of plates) [44], and sage essential oil (2 mL/100 mL distilled water) [45].

Li et al., reported that ginger essential oil was incorporated into gelatin-based films and led to increased WVP when the ratio of oil/gelatin rose from 0% to 12.5% [36].

Kavoosi et al., reported a similar result after the WVP of a gelatin–*Zataria multiflora* composite increased when the oil/protein ratio changed from 0 to 8% [43].

Given that the WVP relies on the hydrophilic/hydrophobic ratio of the film's compounds, the specific composition of essential oils may be responsible for the observed variations in reported different WVP. Even so, adding a hydrophobic material will not necessarily diminish the WVP of the films; it also depends on how the added lipids affect the microstructure of the composite film [44]. An essential quality of food packaging materials is WVP. Loss of textural characteristics and subsequent microbiological growth in foods

could result from moisture from the atmosphere moving into food products. In light of this, a lower WVP may offer good water barrier properties in the gelatin–essential oil films [39].

The antimicrobial activity of the gelatin-based films incorporating essential oils was reported for several strains (Table 3). Li et al., prepared gelatin-based films by combining it with low-content (0%–1%) ginger essential oil. The antimicrobial activity of films that incorporated ginger essential oil was tested on *E. coli* as Gram-negative and *S. aureus* as Gram-positive bacteria. A higher log CFU/mL value indicates better antimicrobial efficiency. Low antimicrobial activity was detected on gelatin-based film for both strains. As the amount of ginger essential oil in the films grew, so did their antibacterial activity. The 1% gelatin–ginger essential oil film reported the greatest antimicrobial activity, with values of 2.65 log CFU/mL against *E. coli* and 5.63 log CFU/mL against *S. aureus* [36].

Yang et al., discovered similar results when they investigated gelatin-based films' antimicrobial activity with cinnamon leaf essential oil against *E. coli*, *S. typhimurium*, *L. monocytogenes*, and *S. aureus*. In contrast to the gelatin film, which did not prevent bacterial pathogens from growing, the inhibitory zone grew in proportion to the cinnamon leaf essential oil concentration. Additionally, the antimicrobial activities were more efficient against Gram-positive bacteria than against Gram-negative ones [37].

Similar results, where antimicrobial activities are more efficient against Gram-positive bacteria than against Gram-negative ones, were obtained when gelatin-based films incorporated other essential oils, such as thyme essential oil [39], *Zataria multiflora* essential oil [43], bergamot and lemongrass essential oils [42], clove and garlic essential oils [44], and sage essential oil [45].

The mechanism of action of oils against bacteria is attributed to cytoplasm loss due to phospholipid cellular wall degradation, or due to interactions among oils and cell enzymes. Because an external lipopolysaccharide wall or proteins protect the peptidoglycan cell wall in the outer membrane, Gram-negative bacteria are more resistant to oil attack [45,79].

3. Chitosan

Chitosan (Figure 2) is a polysaccharide-related chemical compound and a copolymer of N-acetylglucosamine and glucosamine residues linked by -1,4-glycosidic bonds. Because of its availability and low price, biocompatibility, non-toxicity, biodegradability, and film-forming properties, chitosan is regarded as the most promising replacement for conventional plastics in the production of films/coatings, with a wide range of uses in many fields, including food application, pharmaceuticals, agriculture, and beauty products [80–83].

Chitosan products are very viscous, closely resembling natural gums with antimicrobial activities due to active amino groups, and they can constitute clear films to improve the quality and shelf life of processed and fresh foods [6,84]. Due to its capacity to form a partially permeable, durable, and flexible film, chitosan can be used to create edible films that can change the internal atmosphere, reduce water loss, and postpone the spoilage of fruits and vegetables. These characteristics give chitosan advantages over other edible coatings [85,86].

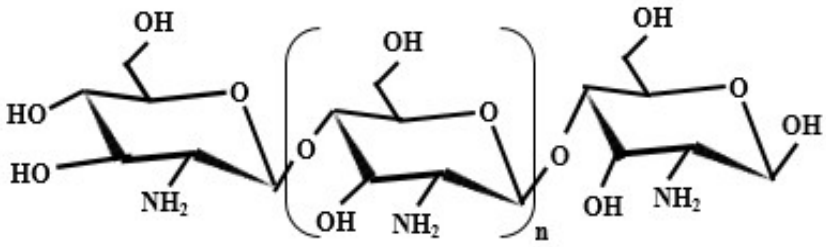

Figure 2. The chemical structure of chitosan.

In contrast to these associated advantages, chitosan-based films have drawbacks such as low UV-light barrier properties and reduced mechanical characteristics. In addition, the

hydrophilicity of chitosan films makes them extremely susceptible to moisture, which is a significant disadvantage for packaged food products with high water content. Despite chitosan's implicit antioxidant and antimicrobial properties, these might not be sufficient to avoid severe growth of microorganisms and oxidation in the ambient environment. Hence, adding natural compounds, such as phenolic compounds, plant extracts, and essential oils or other biopolymers, may provide better antioxidant and antimicrobial activities and enable the development of packaging films with improved mechanical, physical, and biological characteristics [87].

3.1. Origination of Chitosan

Chitosan is obtained from chitin. Chitin (Figure 3), after cellulose, is the second-most prevalent structural polysaccharide in nature. Because of its acetyl groups, chitin has few applications; however, the deacetylation process transforms chitin into chitosan. The acetyl group in chitin is changed into hydroxyl and amino groups in the chitosan during the deacetylation process [88,89].

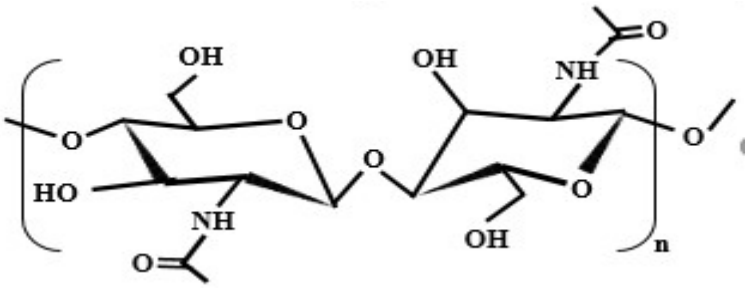

Figure 3. The chemical structure of chitin.

Chitin is naturally found as organized crystalline microfibrils that serve as structural elements for funguses or yeast cell walls and the external skeletons of arthropods. Crab and shrimp shells are currently the primary commercial sources of chitin, where it is present in the α-chitin form [90]. Another significant source of chitin is squid, found in the β-form, which has been reported to be more susceptible to deacetylation. Due to the substantially weaker intermolecular hydrogen link caused by the parallel configuration of the major chains, this chitin also exhibits increased properties such as higher solubility and reactivity, and a better affinity for solvents and swelling than the α-chitin [91]. Chitin can be found in the γ-form, mainly in fungi and yeast, as a mixture of the α- and β-forms instead of a distinct polymorph [92]. Algae, fungi, bacteria, and some species of insects can also serve as substitute sources of chitin and chitosan [88,89,92].

To extract chitin from the shell, protein and minerals must be removed by deproteinization and demineralization. Additionally, a discoloration step is added [88,92].

Typically, "chitosan" is a group of polymers created following the variable degrees of chitin deacetylation [90]. In reality, chitin and chitosan are distinguished by the degree of deacetylation, which represents the equilibrium of the two types of residues. Chitosan is a product that has a deacetylation degree greater than 50% [92]. Deacetylation also results in a depolymerization process, as shown by modifications in chitosan's molecular weight. Using an enzymatic or a chemical procedure, chitin can be transformed into chitosan [88]. Because of their low costs and efficiency for mass production, chemical techniques are frequently utilized to produce chitosan for commercial usage [90].

3.2. Chitosan-Based Composites

3.2.1. Combined Chitosan and Other Biopolymers

Films and coatings produced from chitosan have some disadvantages, including low water resistance, low UV-light barrier properties, and reduced mechanical characteristics,

when compared to films formed by mixing two or more biopolymers, rendering them unsuitable for use in films/coatings production in the food industry [17,87].

Several naturally occurring biopolymers, such as polysaccharides (e.g., cellulose, pectin, starch, or alginate) and proteins (e.g., protein isolate, gelatin, or collagen), can be combined with chitosan to create films (Figure 4). As the produced films are affordable, stable, and display improved properties (water and thermal stability, mechanical or biological properties), polysaccharide blends generally provide several advantages over other biopolymer blends [17]. The physical, mechanical, and biological properties of chitosan films that have incorporated other biopolymers for packaging materials have been investigated (Table 4) [93–99].

Table 4. The characteristics of the chitosan–biopolymers mixtures.

Formulation	Physical/Chemical/Mechanical Characteristics	Biological Properties	Applications	References
Chitosan, corn starch	↑ WS, TS, EAB, ↓ WVP by comparison with corn starch film color change	Not determined	Active packaging	[93]
Chitosan, starch	↑ thickness and WS, ↓ WVP	Antimicrobial activity against *L. innocua*	Active packaging	[94]
Chitosan, sporopollenin	↓ thickness, light transmittance, ↑ TS, EAB, Young's modulus successfully incorporate sporopollenin into chitosan, enhanced hydrophobicity of films	Antifungal activity against *Aspergillus niger*, antioxidant activity	Active packaging	[95]
Chitosan, pectin	↑ thickness, WVP, WS, TS, EAB, Young's modulus ↓ density and opacity	Not determined	Packaging	[96]
Chitosan, nanocellulose	↑ thermal stability, oxygen barrier properties, thickness, WVP, TS, Young's modulus, ↓ film's transparency	Antimicrobial activity against *S. aureus*, *E. coli*, and *Candida albicans*	Chicken meat	[97]
Chitosan, Sardinella protein isolate	↑ thickness, moisture content, opacity, UV–VIS light barrier, WS, ↓ WVP, TS, and EAB, color change	Antioxidant activity (DPPH), antimicrobial activity against *S. aureus*, *Micrococcus luteus*, *L. monocytogenes*, *Bacillus cereus*, *Salmonella enterica*, *P. aeruginosa*, *E. coli*, *Klebsiella pneumoniae*	Shrimp packaging	[98]
Chitosan, CMC, sodium alginate	The optimal contents of the chitosan, CMC, and sodium alginate for the preparation of this composite film were 1.5%, 0.5%, and 1.5%. ↑ TS, EAB, WVP	Antimicrobial activity against *E. coli* and *S. aureus*	Packaging	[99]

↑-increased values of the tested characteristics, ↓-decreased values of the tested characteristics.

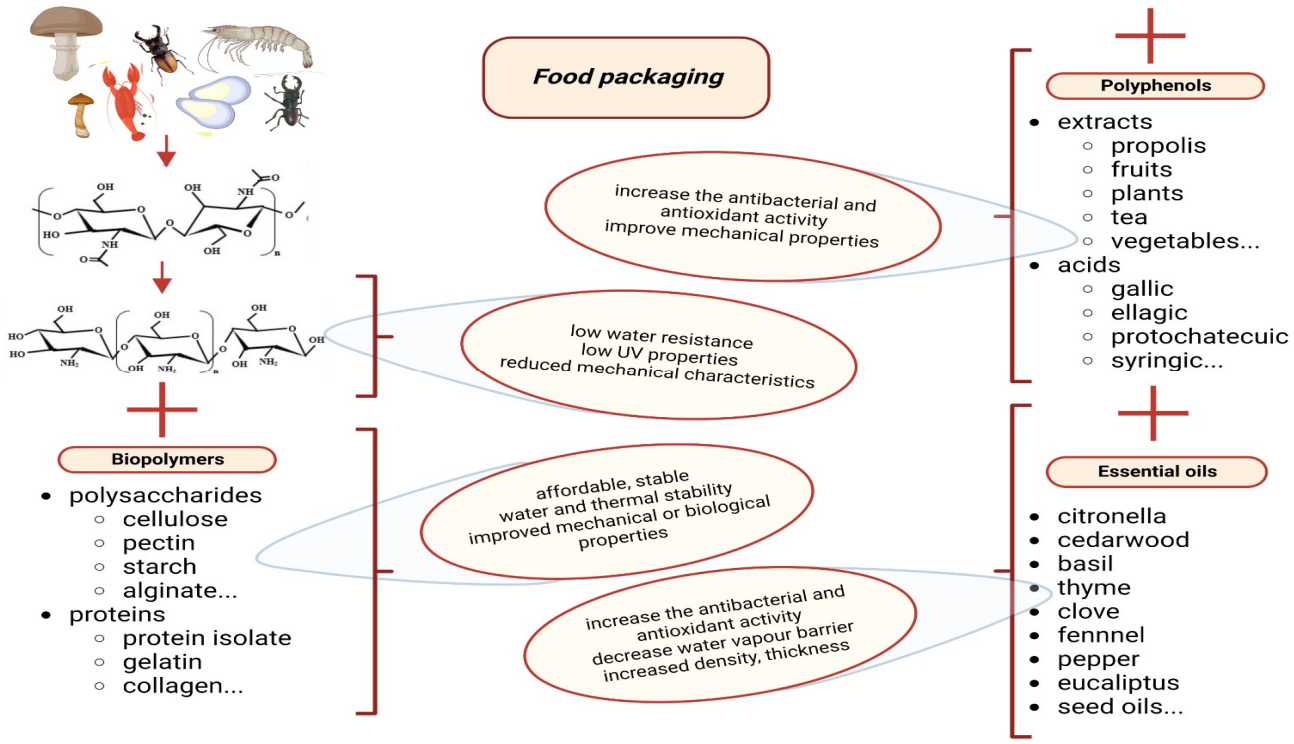

Figure 4. Schematic representation of chitosan matrix incorporating functional materials for improving the characteristics of the composite films.

For example, chitosan was combined with corn starch. The cross sections of films exhibited a continuous surface devoid of a blurry and porous structure, devoid of phase separation among the two polymeric materials, and a compact design in the polymer composite [93]. The study's findings demonstrated that chitosan could interact with corn starch to establish hydrogen bonds between the $NH3^{+}$ of the chitosan and the OH^{-} of the corn starch, which enhanced the composite film's mechanical characteristics, TS and EAB, while lowering its WVP, properties needed for films used in the food industry packaging [93].

Similar results were obtained in the study published by Escamilla-García et al., where chitosan was combined with starch to enhance the composite film's physical, mechanical, and biological properties. Additionally, the chitosan–starch films presented antimicrobial activity against *L. innocua*, which indicates that these composites could be used to ensure the safety of food products in the packaging industry [94].

Films produced from the sporopollenin–chitosan blend were developed and characterized for the first time in a study published by Kaya et al. [95]. Sporopollenin is a biopolymer obtained from plant pollens; in that study, pollens of *Betula pendula* were used, that possess outstanding properties, including biocompatibility with other materials, nontoxicity, and biodegradability, as well as good thermal and strong acid and basic solutions resistance. To take advantage of these essential benefits, and the fact that this biopolymer is easy to collect and available in nature, sporopollenin samples were mixed into chitosan film to obtain a composite with improved characteristics. The incorporation of sporopollenin into the chitosan matrix has been confirmed by several analyses. The incorporation of growing quantities of sporopollenin into chitosan-based films was favorable, considering that physical, chemical, and mechanical characteristics were improved, and the films' hydrophobicity and biological (antioxidant and antifungal) properties were enhanced. These results indicate that sporopollenin could be recommended as a material for manufacturing chitosan-based composites [95].

Younis et al. [96] have developed a biodegradable packaging material by combining chitosan with pectin. While chitosan and pectin may be used separately due to their capacity to produce films, the current research found that mixing chitosan and pectin could develop a composite film with better characteristics than either of its parts. Combining chitosan with pectin may have synergistic effects that enhance several film properties, notably mechanical characteristics. The chitosan and pectin intermolecular interactions (hydrogen bonds, hydrophobic interactions, and ionic complexation) significantly enhanced the network of polymers in the film matrix. They allowed the TS, Young's modulus, and EAB to increase, which were likely responsible for these synergistic effects [96].

Recently, Costa et al. [97] evaluated the characteristics of chitosan–nanocellulosebased films. Combining nanocellulose into other biopolymeric matrices makes possible the development of greater composites while maintaining their biodegradability and improving their mechanical properties and barrier characteristics [100]. Therefore, in this study, the addition of nanocellulose enhanced the thermal stability and oxygen barrier properties and slightly increased WVP. The improvement of the mechanical characteristics of the chitosan/nanocellulose-based films was noticed by the rise in TS and Young's modulus of the composite. Additionally, these composite films exhibited antimicrobial activity against *E. coli*, *S. aureus*, and *Candida albicans*. The reduction of total volatile basic nitrogen on the surface of the chicken meat by chitosan/nanocellulose-based films suggests their potential application as packaging for retarding beef deterioration [97].

Moreover, Sardinella protein isolate, obtained from blue crab and *Sardinella aurita* by-products, was used as a biopolymer in a study conducted by Azaza et al. [98]. The protein isolate was incorporated into the chitosan matrix to obtain an active packaging composite film with better characteristics. Incorporating the protein isolate into the chitosan matrix improved the UV–VIS light barrier due to the formation of links between the two polymers, and it decreased WVP due to the strengthening of the cross-linking in the composite films and the limitation of the mobility of the polymer matrix. Although the protein isolate incorporation into the chitosan matrix led to a slight decrease in mechanical properties, the results showed that the composite films had better biological characteristics than the control film [98].

In a recent study, chitosan was combined with two other biopolymers, sodium alginate and carboxymethyl cellulose, and the characteristics of the new composite films were assessed [99]. These three biopolymers were used due to the potential antibacterial activity of the chitosan films, the high strength of the CMC, and the flexibility and film-forming capacity of sodium alginate. The composite film presented improved mechanical properties and also good antibacterial activity, with a 96% antibacterial rate against *E. coli* and a 93% antibacterial rate against *S. aureus*; therefore, the composite film has potential for use as an active packaging [99].

3.2.2. Combined Chitosan with Polyphenols/Extracts Rich in Polyphenols

In an attempt to increase the biological and functional properties of chitosan-based films, several studies have been performed and assessed on the effects of the incorporation of various kinds of natural extracts rich in polyphenols or phenolic compounds into the chitosan matrix (Table 5). Plants react to stress by producing polyphenolic compounds as secondary metabolites. Polyphenolic compounds present in plants are phenolic acids, flavonoids, and anthocyanins. These compounds minimize oxidation and cell damage and act as powerful antibacterial and antioxidant agents. When they are utilized in film or coating formulations, the polyphenolic compounds display synergy and increase the composite films' total antibacterial and antioxidant activity [17]. Several studies have found that incorporating phenolic compounds or plant extracts rich in polyphenolic content into chitosan films improves their mechanical properties (Table 5).

The mechanical characteristics of a film are highly reliant on intra-molecular bonding, the type of chitosan matrix, the microstructure of the chitosan network, and the presence of crystalline phase inside the film [101,102]. The literature indicates that modifications

of the mechanical characteristics of chitosan films by the incorporation of various phenols/extracts rich in polyphenols are not similar. Some compounds increase the mechanical strength, whereas others reduce it.

The incorporation of propolis extract (5%–20%) increases the TS and EAB in chitosan–propolis composite film [103,104]. Additionally, it has been described in several studies that the incorporation of phenolic compounds, such as gallic acid [105], epigallocatechin gallate nanocapsules (2.5%, 4.5%, 6.0% (*w*/*v*)) [106], ellagic acid (0.5%, 1.0%, 2.5%, 5.0% *w*/*w*) [107], protocatechuic acid (0.8, 1.6, 2.4, 3.2 g) [108], proanthocyanidins (5, 10, 15, 20 wt%) [109], syringic acid (0.25%, 0.5%) [110], phenolic acids (ferulic, caffeic, tannic, gallic, 1%) [81], or curcumin (1%) [111], into chitosan-based films, reinforces the mechanical strength of the chitosan composite. In chitosan-based films, the incorporation of olive leaves extract (10%, 20%, 30% *w*/*w*) [112], or purple rice and black rice extracts ((1, 3, 5 wt%) [113], achieved an increase in the film's TS and EAB, whereas the addition of pomegranate peel extract (10 g/L) [114], thyme extract (0.15% *w*/*w*) [115], turmeric extract [116], mango leaf extract (1%–5%) [117], or purple-fleshed sweet potato extract (5, 10, 15 wt%) [118] improved only the TS, and the addition of pomegranate peel extract (1%, 2%, 3%) [119], grapefruit seed extract (0.5, 1.0, 1.5% *v*/*v*) [120], *Berberis crataegina* fruit extract (1 g) [121], and *Nigella sativa* seed-cake extract (2.5%, 5%, 7.5%) [122] improved only the EAB. On the other hand, several studies have reported that the incorporation of polyphenols/extracts rich in polyphenols may decrease the mechanical characteristics [84,101,102,114,123–126].

Usually, an increase in the strength of chitosan–polyphenols films is associated with a strong bond between the phenolic compounds and the chitosan matrix that explains the enhanced stiffness. On the contrary, a decline in strength is related to the reduction of intermolecular interactions between chitosan chains in the presence of polyphenolic compounds [116]. Riaz et al. [102] reported that the decrease in mechanical characteristics is due to the reduction of intermolecular interactions between chitosan chains and the loss of crystalline phase inside the film.

The barrier properties of chitosan-based films are helpful in maintaining the preservation and nutritional value, and in prolonging the shelf life, of food products [127].

A recent study indicated that chitosan–propolis films' water vapor and oxygen permeability decreased with increased amounts of propolis incorporation into the chitosan matrix [103]. In one study, blueberry, parsley, and red grape extracts [128] were incorporated into the chitosan matrix. Oxygen permeability was reduced by an average of 21% for films containing 5% blueberry extract, by 16% for films containing 5% parsley extract, and by 14% percent for films containing 5% red grapes extract. The decrease in oxygen permeability of the chitosan–extract films is caused by the potential of cross-linking between the polyphenolic content of the extracts and the chitosan matrix [128].

Furthermore, the incorporation of epigallocatechin gallate nanocapsules [106], protocatechuic acid [108], turmeric extract [116], and *Sonneratia caseolaris* leaf extract [84] into chitosan-based films reported UV–VIS light barrier properties for the chitosan composite films. The UV–VIS light barrier property of a film is a significant aspect, because the packaged food's resistance to oxidative degradation might be enhanced by the UV–VIS light barrier characteristics, which could prevent nutritional loss, color changes in the food, and off flavors [108]. The UV–VIS barrier property could be related to the incorporation of polyphenolic compounds into the inter-molecular pores of the chitosan matrix, which might block UV–VIS light transmission, and to the aromatic groups present in polyphenolic compounds, which might absorb the UV–VIS radiations [106,116].

Since one of the main purposes of a film is to prevent moisture transfer between the food and the environment in order to avoid or postpone deterioration, WVP should be reduced as low as necessary to keep products fresh [123]. Decreased WVP was reported in several studies for chitosan-based films incorporating various natural extracts abundant in polyphenols, such as pomegranate peel extract [119], tea extract [124], *Lycium barbarum* fruit extract [125], honeysuckle flower extract [126], *Nigella sativa* seedcake extract [122], mango leaf extract [117], *Herba Lophatheri* extract [129], Chinese chive (*Allium tuberosum*) root

extract [102], and olive leaves extract [112]. Similar results were reported when phenolic compounds were incorporated into the chitosan matrix. For instance, the incorporation of syringic acid in the chitosan matrix improves WVP [110]. The results show that chitosan–syringic acid films significantly decreased WVP in comparison to control films, and WVP decreased as the syringic acid amounts increased [110].

Chitosan-based films may be used as active packaging in order to inhibit food oxidation. Antioxidant activity of the chitosan-based films was improved by the addition of propolis extract [103,104]. With an increased amount of propolis being integrated into the chitosan matrix, the DPPH radical scavenging capacity improved. This result might be explained by the presence of phenolic compounds in the propolis extract [104]. Chitosan-based films incorporating different phenolic compounds have higher antioxidant activity than simple chitosan films [81,108]. The enhancement of the antioxidant activity of chitosan-based films was noticed with the addition of blueberry, parsley, and red grapes containing polyphenols extracts [128]. Recently, Rambabu et al. [117] reported that chitosan-based film incorporating mango leaf extract presented higher antioxidant activity compared with control films. The antioxidant activity of mango leaf extract is due to the presence of polyphenolic content and some compounds with antioxidant potential, such as mangiferin [117].

It is well known that polyphenols are natural compounds with a variety of biological properties. There are various phenolic compounds that possess antimicrobial activity; therefore, antimicrobial properties of chitosan films are expected to increase with the addition of polyphenols to their composition [130]. Several researchers evaluated the biological properties of chitosan-based films enriched with polyphenols/extracts rich in polyphenols (Table 5). The combination of chitosan-based films with polyphenols/extracts rich in polyphenols augmented the antimicrobial activity against Gram-negative and Gram-positive bacteria [84,101,102,107,114,116,121,129,131]. Recently, Sun et al. [123] reported that polyphenol compounds extracted from thinned young apple increased the antimicrobial activity of chitosan-based films against three molds (*Colletotrichum fructicola*, *Botryosphaerial dothidea*, and *Alternaria tenuissima*). Moreover, the composite films did not have activity against yeasts (*S. cerevisiae*, *Baker's yeast*, and *Tropical candida*) [123].

Table 5. The characteristics of chitosan–polyphenol mixtures.

Formulation	Physical/Chemical/Mechanical Characteristics	Biological Properties	Applications	References
Chitosan, propolis extract	↑ TS, EAB, ↓ WVP, oxygen permeability color changes of the films	Antioxidant activity (DPPH), antimicrobial activity against *S. aureus*, *Salmonella Enteritidis*, *E. coli*, and *P. aeruginosa*	Active packaging	[103]
Chitosan, propolis extract	↑ thickness, thermal stability, TS ↓ transparency, EAB, WS color change	Antioxidant activity (DPPH (49.8%–94.5%), ABTS (20.3%–83.6%)), antimicrobial activity against *Staphylococcus hominis*, *Pantoea* sp., *Arthrobacter* sp., *Erwinia* sp., *B. cereus*, *E. coli*, *S. aureus*, *Metschnikowia rancensis*, *Cladosporium* sp., *Penicillium brevicompactum*, *Botrytis cinerea*, and *Alternaria* sp.	Active packaging	[104]
Chitosan, gallic acid	↑ TS (for chitosan:gallic acid ratio 1:0.1, 1:0.5) ↓ EAB and WVP (for chitosan:gallic acid ratio 1:0.1)	Antioxidant activity (DPPH, ABTS), antimicrobial activity against *E. coli* and *L. monocytogenes*	Active food packaging	[105]
Chitosan, epigallocatechin gallate nanocapsules (with zein)	↑ TS, EAB, VIS-light protection	Antioxidant activity (DPPH)	Active packaging	[106]
Chitosan, ellagic acid	↑ EAB, WVP ↓ TS, Young's modulus, UV-light protection good thermal stability	Antioxidant activity (DPPH), antimicrobial activity against *P. aeruginosa* and *S. aureus*, prevent photo-oxidation of light-sensitive foods	Active packaging	[107]

Table 5. *Cont.*

Formulation	Physical/Chemical/Mechanical Characteristics	Biological Properties	Applications	References
Chitosan, protocatechuic acid	↑ thickness, opacity, WS, UV-light barrier ↓ moisture content, WVP, EAB, color change TS increased up to 1% acid incorporation, afterwards decreased	Antioxidant activity (DPPH)	Active packaging	[108]
Chitosan, thinned young apple polyphenols	↑ thickness, density, WS ↓ WVP, TS, EAB, water content	Antioxidant activity (DPPH 68%–92%), antimicrobial activity against *E. coli*, *S aureus*, *L. monocytogenes*, *Colletotrichum fructicola*, *Botryosphaerial dothidea*, and *Alternaria tenuissima*	Active packaging	[123]
Chitosan, apple peel polyphenols	↑ thickness, density, WS, ↓ thermal stability, WVP, TS, EAB, moisture content, transparency color change	Antioxidant activity (DPPH 30%–67%, ABTS 70%–90%), antimicrobial activity against *E. coli*, *B. cereus*, *S. aureus*, and *S. typhimurium*	Active packaging	[101]
Chitosan, proanthocyanidins	↓ thermal stability	Antioxidant activity (DPPH, ABTS), antimicrobial activity against *M. luteus*, *B. subtilis*, *E. coli*, *S. aureus*, *Proteus vulgaris*, and *P. aeruginosa*	Active packaging	[132]
Chitosan, proanthocyanidins	↑ thickness, opacity, thermal stability, WS, WVP, TS, UV–VIS light barrier ↓ moisture content, EAB, oxygen permeability color change	Antioxidant activity (DPPH), antimicrobial activity against *E. coli*, *Salmonella*, *S. aureus*, and *L. monocytogenes*	Active packaging	[109]
Chitosan, syringic acid	↑ thickness, density, WS, opacity, TS when the amount of syringic acid was under 0.5% and EAB when the amount of syringic acid was 0.25%, ↓ moisture content, thermal stability and WVP color change	Antimicrobial activity against *S. aureus* and *E. coli*	Preservation of quail egg Active packaging	[110]
Chitosan, phenolic acids (ferulic acid, caffeic acid, tannic acid, gallic acid)	↑ TS, EAB, Young's modulus, thermal stability, WVP color change	Antioxidant activity (DPPH 17%–89%)	Active packaging	[81]
Chitosan, curcumin	↑ TS ↓ EAB, WVP,	Antimicrobial activity against *S. aureus* and *Rhizoctonia solani*	Active packaging	[111]
Chitosan, carvacrol	↓ WVP, TS, EAB, thickness and transparency, change color to yellow	Antioxidant activity (FRAP), antimicrobial activity against *E. coli* and *S. aureus*	Active packaging	[114]
Chitosan, pomegranate peel extract	↑ thickness, TS ↓ EAB and transparency change color	Antioxidant activity (FRAP), antimicrobial activity against *S. aureus*	Active packaging	[114]
Chitosan, pomegranate peel extract	↑ EAB ↓ TS, WVP	Antioxidant activity (DPPH 21%–57%)	Active packaging	[119]
Chitosan, thyme extract	↑ TS, EM, opacity decreased: EAB, color change	Antioxidant activity (DPPH)	Active packaging	[115]
Chitosan, turmeric extract	↑ TS, Young's modulus, WVP, UV–VIS barrier property	Antimicrobial activity against *S. aureus* and *Salmonella*	Active packaging	[116]
Chitosan, tea extract	↑ thickness, WS ↓ water content, WVP, TS, EAB	Antioxidant activity (DPPH)	Active packaging	[124]
Chitosan, grapefruit seed extract	↑ thickness, EAB, ↓TS	Antifungal activity	Bread preservation	[120]
Chitosan, maqui berry extract (*Aristotelia chilensis*)	not determined	Antioxidant activity (DPPH, FRAP), antimicrobial activity against *Serratia marcescens*, *Alcaligenes faecalis*, *Aeromonas hydrophila*, *Pseudomonas fluorescens*, *Citrobacter freundii*, *Achromobacter denitrifican*, *S. putrefaciens*	Active packaging	[131]
Chitosan, *Lycium barbarum* fruit extract	↑ density ↓ TS, EAB, WVP, WS, moisture content	Antioxidant activity (DPPH)	Active packaging	[125]
Chitosan, honeysuckle flower extract (*Lonicera japonica* Thunb)	↑ WS, density ↓ WVP, TS, EAB, moisture content	Antioxidant activity (DPPH), antimicrobial activity against *E. coli*	Active packaging	[126]

Table 5. *Cont.*

Formulation	Physical/Chemical/Mechanical Characteristics	Biological Properties	Applications	References
Chitosan, *Berberis crataegina* fruit extract	↑ thickness, EAB ↓ transparency, TS, WS, Young's modulus	Antioxidant activity (DPPH 86%), antimicrobial activity against *E. coli*, *S. thypmurium*, *Proteus microbilis*, *Proteus vulgaris*, *P. aeruginosa*, *Enterobacter aerogenes*, *S. aureus*, *Streptococcus mutans*, *Bacillus thuringiensis*	Active packaging	[121]
Chitosan, *Nigella sativa* seedcake extract	↑ thickness, EAB, ↓ moisture content, WVP, TS color change	Antioxidant activity (DPPH, FRAP)	Active packaging	[122]
Chitosan, mango leaf extract	↑ thickness, TS, EM, ↓ moisture content, WS, WVP, EAB	Antioxidant activity (DPPH, FRAP, ABTS)	Cashew nuts preservation	[117]
Chitosan, *Herba Lophatheri* extract from dried leaves of *Lophatherum gracile* Brongn	↑ opacity, density, ↓ WS, WVP, moisture content color change, higher oil resistance	Antioxidant activity (DPPH), antimicrobial activity against *E. coli* and *S. aureus*	Active packaging	[129]
Chitosan, Chinese chive (*Allium tuberosum)* root extract	↑ thickness, thermal stability ↓ TS, EAB, WS, WVP, moisture content	Antioxidant activity (DPPH 20%–47%, ABTS 28%–57%), antimicrobial activity against *B. cereus*, *S. aureus*, *E. coli*, and *S. typhimurium*	Soybean oil packaging	[102]
Chitosan, *Sonneratia caseolaris* (L.) Engl. leaf extract	↑ light barrier property, WS, WVP ↓ TS, EAB, moisture content change color	Antimicrobial activity against *S. aureus* and *P. aeruginosa*	Vietnamese banana preservation	[84]
Chitosan, olive leaves extract	↑ WS, TS, and EAB, ↓ WVP	Antioxidant activity (ABTS), antimicrobial activity against *L. monocytogenes* and *Campylobacter jejuni*	Active packaging	[112]
Chitosan, blueberry extract by-products	↑ thickness, WVP ↓ oxygen permeability, water content	Antioxidant activity (DPPH, ABTS, FRAP)	Active packaging	[128]
Chitosan, parsley extract by-products	↑ thickness, WVP ↓ oxygen permeability, water content	Antioxidant activity (DPPH, ABTS, FRAP)	Active packaging	[128]
Chitosan, red grapes extract by-products	↑ thickness, WVP ↓ oxygen permeability, water content	Antioxidant activity (DPPH, ABTS, FRAP), antimicrobial activity against *E. coli*	Active packaging	[128]
Chitosan, purple-fleshed sweet potato extract	↑ thickness, WS, WVP when the extract exceeded 5 wt% and TS when the extract was 5 wt% ↓ EAB, WVP when the extract was 5 wt%, TS when the extract exceeded 5 wt%, moisture content and light transmittance	Antioxidant activity (DPPH), color variations of films to pH, pink-red (pH 3.0–6.0), purple-brown (pH 7.0–8.0), and greenish–green (pH 9.0–10.0)	Monitoring food spoilage	[118]
Chitosan, purple rice extract	↑ thickness, EAB, TS, light barrier property, and WVP when the extract exceeded 1 wt% ↓ moisture content, change color	Antioxidant activity (DPPH), pH-sensitive in different buffer solutions	Monitor pork spoilage	[113]
Chitosan, black rice extract	↑ thickness, EAB, light barrier property ↓ moisture content, TS when the extract exceeded 1 wt% change color	Antioxidant activity (DPPH)	Active packaging	[113]

↑-increased values of the tested characteristics, ↓-decreased values of the tested characteristics.

3.2.3. Combined Chitosan and Essential Oil

Essential oils are secondary plant metabolites with strong fragrance and great antioxidant and antimicrobial properties. The main content is represented by bioactive compounds, such as polyphenolic compounds, alkaloids, aldehydes, carotenoids, and monoterpenes [133]. In order to decrease their volatility, and enhance antioxidant and antimicrobial activities, essential oils may be incorporated in polymer matrices such as chitosan [17]. A unique advantage of essential oils utilization seems to be the synergistic effects of their constituents, as contrasted to the sum of the activities of the separate bioactive compounds [134]. Several studies have reported that chitosan films incorporating essential oils have improved characteristics (Table 6).

Shen and Kamden [135] reported that the incorporation of citronella and cedarwood essential oils into the chitosan matrix affected the mechanical characteristics. The TS of composite films was reduced when the amounts of the essential oils increased. They observed that incorporating low amounts of essential oils led to increased EAB. However,

they noticed that by incorporating increased amounts of essential oils, a decrease in EAB was obtained. This result may be described as the substitution of stronger polymer–polymer bonds with weaker polymer–oil bonds in the film network [135].

Similar results were reported by other researchers. Priyardashi et al. [136] observed an increase in EAB for chitosan–apricot kernel essential oil films with the incorporation of low amounts of the essential oil; however, when the ratio of chitosan: essential oil exceeded 1: 0.5, the EAB decreased. Moreover, they observed an increase in the TS with an increase in the essential oil amount being incorporated into the film [136].

A similar pattern for the EAB of chitosan–*piper betle* Linn oil films was reported in the study conducted by Nguyen et al. [137].

The addition of basil essential oil and thyme essential oil to chitosan-based films improved the mechanical characteristics of the composite film. The TS and EM were increased, whereas the EAB decreased [138].

An important property of the composite films is water vapor permeability. Decreased WVP was reported in several studies for chitosan-based films incorporating various essential oils, such as citronella (10%, 20%, 30% w/w) [135], cedarwood (10%, 20%, 30% w/w) [135], basil (0.5 g, 1 g/100 g) [138], thyme (0.5 g, 1 g/100 g) [138], and apricot kernel (0.125, 0.25, 0.5, 1%) [136]. Hydrogen and covalent bonds between the chitosan matrix and the bioactive compounds may result in reduced WVP of chitosan–essential oil films. These interactions might minimize the capacity of hydrophilic groups to establish hydrophilic linkages and, therefore, minimize the interactions with water, resulting in a composite film with better moisture resistance. Additionally, even at low concentrations, the existence of a hydrophobic dispersion causes discontinuity in the hydrophilic phase that leads to decreasing WVP [135,136].

Several studies reported that by incorporating essential oils into a chitosan matrix, the biological activities may be improved. For instance, Liu et al. [139] prepared chitosan–peppermint essential oil and chitosan–fennel essential oil films and evaluated the antioxidant activity of the composite films using DPPH scavenging method. The chitosan-based film had the lowest antioxidant activity among all the films (55%). The chitosan-based film's antioxidant activity could be associated with the NH_2 units in the chitosan matrix, units that interacted with DPPH and generated stable molecules. The incorporation of essential oils into chitosan-based films enhanced their ability to scavenge DPPH. The chitosan–peppermint essential oil film had higher antioxidant activity (67%) than the chitosan-based film, due to the peppermint composition with antioxidant properties. Moreover, the trans-anethole molecule in fennel essential oil could be responsible for the greater antioxidant activity of the chitosan–fennel essential oil film (68%) compared to the chitosan–peppermint essential oil film [139].

In another study, Hafsa et al. [140] prepared chitosan–*Eucalyptus globulus* essential oil films with different essential oil content and evaluated its antioxidant and antimicrobial activities. The authors reported that the DPPH scavenging ability of the composite increased with increasing essential oil content. The highest antioxidant activity was 44% (chitosan incorporating 4% (v/v) essential oil), which was substantially higher than that of chitosan-based film (only 10%). Furthermore, the antimicrobial activity of the composite films was tested against three bacteria, *E. coli*, *S. aureus*, and *P. aeruginosa*, and two fungi, *C. albicans* and *Candida parapsilosis*. The results of the study showed that all composite films showed antimicrobial activity against all strains tested and the antimicrobial activity increased with increasing essential oil content [140].

Similar results were obtained in a study conducted by Priyadarshi et al. [136]. They incorporated different amounts of Apricot kernel essential oil into chitosan-based films, and they tested the biological activities for all the composite films and also for the chitosan-based film. It was observed that the antioxidant and antimicrobial activities increased with increasing essential oil content. Moreover, the chitosan–apricot kernel essential oil films were tested for antifungal activity. The authors evaluated the potential of the composite films for the inhibition the of growth of *Rhizopus stolonifer* on bread slices. The films were

observed to successfully limit the growth of fungi on bread, hence extending its shelf life [136].

Table 6. The characteristics of chitosan–essential oil mixtures.

Formulation	Physical/Chemical/Mechanical Characteristics	Biological Properties	Applications	References
Chitosan, citronella essential oil	↑ EAB (low essential oil content), thermal stability ↓ WVP, TS, moisture content	Not determined	Packaging	[135]
Chitosan, cedarwood essential oil	↑ EAB (low essential oil content), thermal stability ↓ WVP, TS, moisture content	Not determined	Packaging	[135]
Chitosan, basil essential oil	↑ thickness, TS, EM ↓ WVP, EAB	Tested for antifungal activity, but the film did not inhibit the growth of *A. niger*, *Botrytis cinerea*, and *R. stolonifer*	Packaging	[138]
Chitosan, thyme essential oil	↑ thickness, TS, EM ↓ WVP, EAB	Tested for antifungal activity, but the film did not inhibit the growth of *A. niger*, *B. cinerea*, and *R. stolonifer*	Packaging	[138]
Chitosan, fennel essential oil	↑ density, thermal stability, and opacity ↓ WS, water swelling, thickness, and moisture content color change	Antioxidant activity (DPPH 68%)	Active packaging	[139]
Chitosan, peppermint essential oil	↑ density, thermal stability, and opacity ↓ WS, water swelling, and thickness color change	Antioxidant activity (DPPH 66%)	Active packaging	[139]
Chitosan, *Eucalyptus globulus* essential oil	↑ opacity ↓ moisture content, WS	Antioxidant activity (DPPH 23%–43%), antimicrobial activity against *E. coli*, *S. aureus*, *P. aeruginosa*, *C. albicans*, *C. parapsilosis*	Active packaging	[140]
Chitosan, apricot kernel essential oil	↑ opacity, TS ↓ moisture content, WS, WVP EAB first increased, and then when the ratio of chitosan:essential oil exceeded 1: 0.5 decreased	Antioxidant activity (DPPH 26%–35%), antimicrobial activity against *S. aureus* and *B. subtillis*, antifungal activity against *R. stolonifer*	Inhibited the growth of fungi on bread, active food packaging	[136]
Chitosan, *piper betle* Linn oil	↑ UV-light barrier, EAB (at 0.4 and 1% oil incorporation), ↓ thermal stability, TS, EM, and EAB (at 1.2% oil incorporated)	Antioxidant activity (DPPH), antimicrobial activity against *S. aureus*, *E. coli*, *P. aeruginosa*, and *S. typhimurium*	King orange preservation	[137]

↑-increased values of the tested characteristics, ↓-decreased values of the tested characteristics.

4. Conclusions

Gelatin and chitosan are two of the most important biopolymers for the production of films and coatings. Although gelatin exhibits excellent gas barrier and swelling properties, it has poor mechanical resistance and is permeable to water vapor molecules. The chitosan-based films have some disadvantages, including low water resistance, low UV–VIS light barrier properties, and reduced mechanical characteristics.

These limitations could be improved by combining gelatin and chitosan with other biopolymers or with bioactive compounds, such as polyphenols, natural extracts, and essential oils. Combining gelatin and chitosan with other biopolymers leads to improved mechanical properties, water vapor, and UV–VIS light barriers, and greater thermal stability of the obtained films. Additionally, incorporating different polyphenolic compounds, natural extracts rich in phenolic content, and essential oils into gelatin-based and chitosan-based films leads to increased physicochemical and mechanical properties and, even more relevant for the food industry, improved biological properties, and antioxidant and antimicrobial activities.

All these improved characteristics of composite films help in maintaining the quality, reducing lipid oxidation, avoiding microbial growth, and extending the shelf life of the packed products.

On the other hand, the implementation of alternative biodegradable materials in the existing infrastructure, and the technological transfer of the findings from a laboratory scale to industrial levels, represents a serious economic effort and a great challenge for both stakeholders and the scientific community.

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