

Recent advances on chitosan-based films for sustainable food packaging applications



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ABSTRACT

The recent sharp increase of sensitivity towards environmental issues arising from plastic packaging has boosted interest towards alternative sustainable packaging materials. This new trend promotes the industrial exploitation of knowledge on chitosan-based films. Chitosan has been extensively investigated and used due to its unique biological and functional properties. However, inherent drawbacks including low mechanical properties and high sensitivity to humidity represent major limitations to its industrial applications, including food packaging. In the present study, the scientific literature of the last five years has been extensively reviewed (source: Web of Science) addressing chitosan-based films for their potential application in the food packaging industry. The contribution summarizes the various strategies adopted to overcome inherent drawbacks and improve the properties of chitosan-based films, with special regards for blending with natural and synthetic biopolymers.

1. Introduction

Annually, more than 350 million tons of plastics are produced in the world (Ritchie & Roser, 2018). It is expected that plastics will account for 20 % of total oil consumption by 2050 (Newell, Qian, & Raimi, 2016; Cui, Borgemans, Qin, Liu, & Li, 2019). Packaging, particularly food packaging, is one of the largest application fields for plastics (Cui, Surendhiran, Li, & Lin, 2020). Food packaging is represented as a co-ordinated system for processing, transporting, distributing, retailing, protecting and preserving food to satisfy the industry demands and consumer desires, to retain food safety and to protect food from external contamination with optimal cost (Marsh & Bugusu, 2007; Shin & Selke, 2014; Yam & Lee, 2012). However, accumulation of huge amounts of plastic waste in the environment, and also rapid depletion of fossil reserves and increases in the cost of petroleum, are pushing the food packaging industry toward the development and application of eco-friendly materials, such as bioplastics (Arikan & Ozsoy, 2015; Philip, Bartsev, Ritchie, Baucher, & Guy, 2013).

Bioplastics can be referred to as plastics obtained from renewable resources (biobased), plastics that are biodegradable and/or compostable, or materials that feature both properties (Kumar & Thakur, 2017). Hence, not all biobased materials are biodegradable and not all

biodegradable materials are produced from renewable resources (Rujnić-Sokele & Pilipović, 2017). A schematic classification of biodegradable polymers according to their source is presented in Fig. 1.

The term “biobased” refers to the derivation of material from biomass (Soroudi & Jakubowicz, 2013). The term “biodegradable” indicates materials that can disintegrate or break down naturally into CO₂, CH₄, H₂O, and inorganic compounds, or biomass in which the prevalent process is the enzymatic function of microorganisms (Peelman et al., 2013), that can be measured by standardized tests (ASTM Standard D-5488-94d). Some of these polymers can also be compostable, which means disintegration occurs in a compost site at a rate consistent with known compostable materials and without releasing toxic substances (Siracusa, Rocculi, Romani, & Dalla Rosa, 2008).

As stated by the European Bioplastic Organization, bioplastics constitute approximately 1 % of the total global plastics production annually (Rujnić-Sokele & Pilipović, 2017). Packaging, as one of the largest application fields for bioplastics, shares almost 65 % of the total bioplastics market. This number is predicted to rise continuously in the upcoming years mainly due to the increasing consumer requirements for sustainable products and growing awareness over environmental issues (van den Oever, Molenveld, van der Zee, & Bos, 2017).

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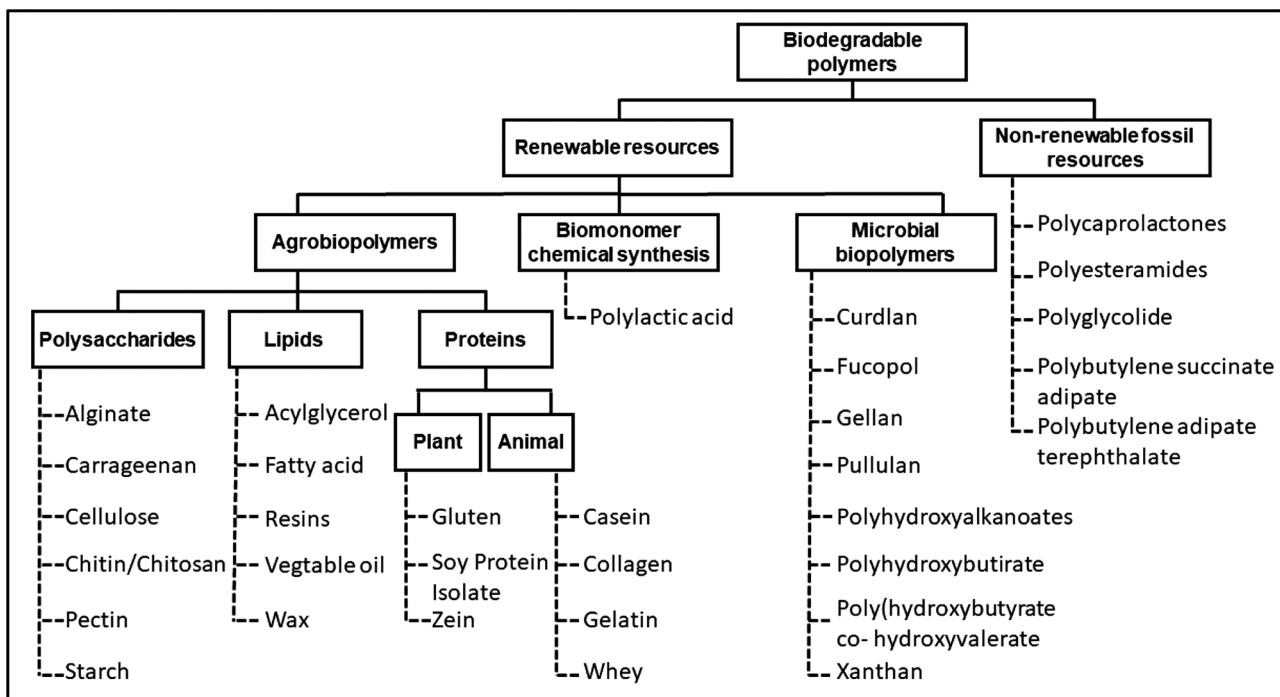


Fig. 1. Schematic classification of biodegradable polymers.

Bioplastics, whether biobased, biodegradable, or both, have unique advantages over conventional plastics to reduce reliance on fossil resources and to mitigate carbon footprint and greenhouse gas emissions. Besides, they promote resource efficiency and offer extra waste management options such as organic recovery (Arikan & Ozsoy, 2015; Kumar & Thakur, 2017).

Recently, biodegradable polymers derived from renewable resources have been proposed as the future generation of packaging materials (Lei et al., 2014). The basic material employed to form bio-based films are polysaccharides, proteins, lipids, and their derivatives (De Leo et al., 2018; Ramos, Valdés, Beltrán, & Garrigós, 2016). Proteins and polysaccharides have acceptable mechanical and gas barrier properties, but they show high moisture sensitivity (Rhim & Ng, 2007). On the contrary, lipid films exhibit acceptable water vapor barrier property and high oxygen permeability, but they have poor mechanical properties (Vodnar, Pop, Dulf, & Socaciu, 2015). Among polysaccharides, chitosan has received considerable attention from academics and industry for food packaging applications due to its particular physicochemical features, biodegradability, non-toxicity, biocompatibility, good film-forming properties, chemical stability, high reactivity (Dutta, Tripathi, & Dutta, 2012; Lago et al., 2014; Mujtaba et al., 2019). General values for the parameters of interest for food packaging applications are reported in Table 1. The reader should consider these values as merely representative since they could dramatically change upon addition of additives such as plasticizers or crosslinker. Chitosan has also intrinsic antioxidant and antimicrobial activities against fungi, molds, yeasts, and bacteria (Aider, 2010; Leceta, Guerrero, & de la Caba, 2013). However, inherent drawbacks of chitosan including low mechanical and thermal stability and high sensitivity to humidity are causing a major restriction for its industrial applications (Elsabee & Abdou, 2013). One strategy to overcome these drawbacks is blending chitosan with other biopolymers to combine their advantages as well as minimize their disadvantages. Therefore, the objective of the present paper is to provide a comprehensive literature review of the last five years addressing chitosan-based films and strategies adopted for the improvement of their performances for potential food packaging applications. To get an idea of the complexity of the theme and of the work that has been done in the last years, an

interrogation of the Web of Science database was performed searching "chitosan, film, blend, food packaging" within title, abstract and keywords in the timeframe 2015–2020. Nevertheless, among the very large number of papers available dealing with a wide range of characteristics and functionalities, only the most significant studies and achievements will be analyzed and discussed.

2. History, features, and potential of chitosan

Chitosan is a unique natural biopolymer, commercially originated from the deacetylation (to varying degrees) of chitin (Verlee, Minckle, & Stevens, 2017). It is the second most abundant natural polysaccharide behind cellulose (Salari, Sowti Khiabani, Rezaei Mokarram, Ghanbarzadeh, & Samadi Kafil, 2018) and the most abundant biopolymer of animal origin (Priyadarshi & Rhim, 2020). Chitin can be obtained from terrestrial arthropods (e.g., spiders, scorpions, beetles, cockroaches, and brachiopods), marine crustaceans (e.g., crab, lobster, prawn, and krill), Mollusca (e.g., squid) and microorganisms (e.g., fungi cell walls) (Zargar, Asghari, & Dashti, 2015). The waste of marine food production (particularly exoskeleton of crabs, lobsters, and shrimps) is currently the main industrial source of biomass for chitin production (Gutiérrez, 2017).

The first report on chitin traces back to 1811 by French Professor of natural history Henri Braconnot. He found out the alkaline-insoluble fraction from mushrooms and named it "fungine". In 1823, Antoine Odier extracted this alkaline-insoluble fraction from the cuticle of insects and named it "chitine", originated from the Greek word "khiton" meaning "tunic" or "envelope". Twenty years later, Jean Louis Lassaigne proved the presence of nitrogen in chitin. In 1859, Prof. C. Rouget discovered the deacetylated form of chitin. He treated chitin with concentrated potassium hydroxide solution and heat to become soluble in dilute organic acids and named it "modified chitin" (Rouget, 1859). In 1878, Ledderhose identified that chitin was made of glucosamine and acetic acid. In 1894, Hoppe-Seyler treated the shells of crabs, scorpions, and spiders with potassium hydroxide solution at 180 °C and dissolved the product in dilute acid solution and named it "chitosan". In 1894, Gilson proved the presence of glucosamine in chitin. In 1930, Rammelberg found more chitin sources apart from insects and fungi. He

Table 1 Moisture content, transparency value, color parameters including L* (lightness), a* (redness/greenness) and b* (yellowness/blueness), ΔE^* (total color difference), water vapor permeability (WVP), oxygen permeability (OP), tensile strength (TS) and elongation at break (E) of low and high molecular weight chitosan films. Adapted from Leceta, Guerrero, & de la Caba (2013).

Molecular weight	Moisture content (%)	Transparency value (A_{600}/T)	L*	a*	b*	ΔE^*	WVP ($\times 10^{-13} \text{ g cm}^{-1} \text{ s}^{-1} \text{ Pa}^{-1}$)	OP ($\text{cm}^3 \mu\text{m}^{-2} \text{ day}^{-1} \text{ KPa}^{-1}$)	TS (MPa)	E (%)
High molecular weight chitosan	15.70	0.754	96.39	-0.23	3.15	1.68	8.07	6.65	61.82	4.59
Low molecular weight chitosan	19.43	0.760	96.54	-0.57	4.74	5.29	9.21	7.70	55.83	4.58

hydrolyzed chitin in several ways and extracted chitosan from marine arthropods (e.g., crab, shrimp, and lobster). In the 1940s, both chitin and chitosan attracted considerable attention as evidenced by about 50 patents. In 1950, the structure of chitosan was discovered using X-ray (Darmon & Rudall, 1950). The first book about chitosan was published by Albert Glenn Richards in 1951 (Richards, 1951).

Nowadays, chitin and chitosans are simply described as copolymers of N-acetyl-D-glucosamine and D-glucosamine units linked with β -(1–4)-glycosidic bonds (Hosseinejad & Jafari, 2016). They attract considerable attention and are employed worldwide for a broad range of applications. In the food industry, they are applied as antimicrobial agents (bactericidal and fungicidal), edible films and coating (e.g., post-harvest deterioration control in fruits), additives (e.g., natural flavor extender, emulsifying agents, thickeners, stabilizing agents, and color stabilizers), integrators (e.g., dietary fiber), enzyme immobilization, encapsulation of nutraceuticals, and purification of water (e.g. removal of dyes) (Ahmed & Ikram, 2017; Dutta, Tripathi, Mehrotra, & Dutta, 2009; López-Caballero, Gómez-Guillén, Pérez-Mateos, & Montero, 2005; Zargar et al., 2015; Zhang, Li, & Liu, 2011).

Chitosan exhibits antioxidant (Ngo & Kim, 2014; Ojagh, Rezaei, Razavi, & Hosseini, 2010) and antimicrobial activity against a broad range of pathogenic and spoilage microorganisms, including fungi (yeasts and molds), Gram-positive and Gram-negative bacteria (Friedman & Juneja, 2010; Kong, Chen, Xing, & Park, 2010; van den Broek, Knoop, Kappen, & Boeriu, 2015). The antimicrobial activity of chitosan has drawn attention as a potential natural food preservative (Del Nobile et al., 2009; No, Meyers, Prinyawiwatkul, & Xu, 2007). Several hypotheses have been suggested to elucidate the mechanism of antimicrobial activity of chitosan: the most reasonable hypothesis is electro static interaction between protonated amino groups (NH_3^+) of glucosamine in the chitosan backbone and microbial negative cell membrane constituents such as phosphoryl groups of the phospholipid components, proteins, amino acids, and various lipopolysaccharides (Elsabee & Abdou, 2013; Mousavi Khanegah, Hashemi, & Limbo, 2018). This interaction affects the membrane integrity and permeability, interfering with energy metabolism, nutrient transport, provoking the permeation of proteinaceous and other intracellular components, and causing disruptions that lead to cell death of microorganisms (Goy, de Britto, & Assis, 2009). Another possible mechanism is the interaction of chitosan with cellular DNA of microorganisms, thus preventing DNA transcription, RNA translation, and protein synthesis (Raafat & Sahl, 2009; Sharif et al., 2018; Verlee et al., 2017). Moreover, chitosan acts as a chelating agent that selectively binds essential trace metals, spores, prevents the production of toxins and microbial growth (Hosseinejad & Jafari, 2016; Vodnar et al., 2015). Several researchers also suggested that microbial growth inhibition occurs by blocking the supply of essential nutrients into the cell (No et al., 2007; Raafat & Sahl, 2009).

3. Strategies for the improvement of properties of chitosan-based films

Inherent drawbacks of chitosan such as high sensitivity to water, low mechanical and thermal stability lead to a shorter food shelf life compared to the conventional food packaging material and consequently limited its applications in food packaging (Al-Tayyar, Youssef, & Al-hindi, 2020; Elsabee & Abdou, 2013). Therefore, different strategies have been proposed to tackle these issues and to improve the properties of chitosan-based materials, such as cross-linking (Jahan, Mathad, & Farheen, 2016; Khouri, Penlidis, & Moresoli, 2019; Liang, Wang, & Chen, 2019; Yeamsuksawat & Liang, 2019), enzyme treatment (Águila-Almanza, Salgado-Delgado, Vargas-Galarza, García-Hernández, & Hernández-Cocoletzi, 2019), graft copolymerization (Argüelles-Monal, Lizardi-Mendoza, Fernández-Quiroz, Recillas-Mota, & Montiel-Herrera, 2018; Wang, Yu et al., 2016; Wang et al., 2019), complexation (Wang, Wang, & Heuzey, 2016), surface coating (Khwaldia, Basta,

Table 2
Synopsis of research published between 2015 – 2020 addressing chitosan-polysaccharide blend films for food packaging applications. Improvement of the parameters investigated in each contribution is reported as follows. M: mechanical properties, WB: water barrier properties, GB: gas barrier permeability, AO: antimicrobial activity, AM: antimicrobial activity.

Biopolymer	Additives	Key findings	M	WB	GB	AO	AM	Reference
Pectin (2 % w/v) ● Nano chitosan (2 % w/v)		<ul style="list-style-type: none"> Films with different ratios of pectin/nano chitosan (100:0, 75:25, 50:50, 25:75, and 0:100 w/w) were developed. Blending pectin with nano chitosan at proportions of 50:50 increased the tensile strength while water solubility, water vapor permeability, and oxygen permeability decreased. Developed films showed antimicrobial activity against <i>C. gloeosporioides</i>, <i>S. cerevisiae</i>, <i>A. niger</i>, and <i>E. coli</i>. Addition of natamycin into the biopolymers blend caused an increase in tensile strength while water vapor permeability decreased. Films containing additive showed excellent barrier to UV light. Addition of pitanga leaf extract caused an increase in antioxidant activity while a combination of pitanga leaf extract and natamycin led to the reduction of antioxidant activity. Addition of natamycin showed positive anti-fungal activity against <i>Aspergillus flavus</i> and <i>Aspergillus parasiticus</i>. Increasing concentration of chitosan in the biopolymers blend caused an increase in water vapor permeability while moisture content was reduced. Application of a purple yam starch/chitosan blend film on apple fruits for 4 weeks preserved the fruit quality compared to untreated apple samples. Films with different proportion of bacterial cellulose/chitosan 0, 1/64, 1/32, 1/16, 1/8, and 1/4 were developed. Addition of borate and tripolyphosphate into the biopolymers blend (1/32) showed an improvement in tensile strength and elasticity values mainly. Antimicrobial activity of composite film against <i>E. coli</i>, <i>B. cereus</i>, and <i>S. cerevisiae</i> reduced by addition of cross-linking agent. ● Addition of citric acid into the biopolymers blend improved tensile strength and elasticity. Active films containing citric acid showed a homogenous and compact structure. Moisture content and water solubility reduced while water vapor permeability, mechanical and antimicrobial properties improved by addition of citric acid to the biopolymers blend. Both control and films containing citric acid showed antibacterial activity against <i>E. coli</i> and <i>S. aureus</i>. Adding 5 % cellulose nanofiber to the biopolymers blend increased tensile strength. Films containing glycerol showed better mechanical properties than films containing xylitol and sorbitol. Films containing glycerol, xylitol and sorbitol showed higher water solubility, barrier properties to water vapor and oxygen, lower contact angle and opacity. ● Addition of xylan into the biopolymers blend caused an increase in elasticity and samples with 20 and 25 wt.% xylan showed higher tensile strength and elastic modulus. Thermal analysis showed that addition of xylan to chitosan moved the degradation peak to a lower temperature with a lower rate of degradation. ● Addition of carvacrol into the biopolymer blend did not show antibacterial activity against <i>E. coli</i> and <i>L. innocua</i>. 	✓	✓	✓	✓	✓	Ngo, Nguyen, Dang, Tran, and Rachtanapun (2020)
Cassava starch (2 % w/w) ● Chitosan (1 % w/w)	Pitanga leaf extract (2.25 % w/w of film forming solution) ● Natamycin (1 % w/w of film forming solution) ● Pitanga leaf extract /Natamycin mixture					✓	✓	Sirisha Nallian Chakravartula et al. (2020)
Purple yam starch (2 % w/v) ● Chitosan (0.5 and 1 % w/v)						✓		Martins da Costa, Lima Miki, da Silva Ramos, and Teixeira-Costa (2020)
Bacterial cellulose (0.5 % w/v) ● Chitosan (2 % w/v)	Borate (4 %) ● Tripolyphosphate (4 %) ● Borate/Tripolyphosphate mixture					✓		Liang et al. (2019)
Potato starch (4 % w/v) ● Chitosan (1.5 % w/v)	Citric acid (5, 10, 15, and 20 % w/w based on a dry biopolymer basis)					✓		Wu et al. (2019)
Hemicellulose (2 % w/v) ● Chitosan (2 % w/v)	Cellulose nanofiber (5, 10, 15, and 20 % w/w based on biopolymers ● Glycerol, xylitol, sorbitol (10, 20, 30, and 40 % v/w on a dry biopolymer basis)					✓		Xu, Xia, Zheng, Yuan, and Sun (2019)
Xylan (20, 30, and 40 % w/w based on chitosan) ● Chitosan (1 % w/v)	Carvacrol (10 % w/w based on dry chitosan weight)					✓		Kardem, Shen, Nabinejad, and Shu (2019)

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Table 2 (continued)

Biopolymer	Additives	Key findings	M	WB	GB	AO	AM	Reference
● Carboxy methyl chitosan (1 % w/v) ● Chitosan (2 % w/v)	● Nisin (1000 and 6000 IU/mL of film forming solution)	<ul style="list-style-type: none"> • Blending carboxymethyl chitosan with chitosan increased elasticity while it reduced thermal stability. • Incorporation of nisin into the biopolymers blend showed antibacterial activity against <i>L. monocytogenes</i>. 	✓	✓	✓	✓	✓	Zinnet et al. (2019)
● Starch (1.5 % w/v) ● Chitosan (1.5 % w/v)	● Clove essential oil (3, 6, 9, and 12 % w/w) ● Nano titanium dioxide (1, 3, 5, and 7 % w/w)	<ul style="list-style-type: none"> • Addition of nano titanium dioxide into the biopolymers blend caused an increase in tensile strength and antioxidant activity while water vapor permeability and elasticity decreased. • Addition of clove essential oil into the biopolymers blend reduced tensile strength, water content, and water vapor permeability while antioxidant and antibacterial activity against <i>E. coli</i> and <i>S. aureus</i> improved. 	✓	✓	✓	✓	✓	Li et al. (2019)
● Carboxymethyl cellulose (2 % w/v) ● Chitosan (1 % w/v)	● Cinnamon essential oil (1.5 % v/v) ● Oleic acid (1 % w/v) ● Glutaraldehyde (0.01 % w/v)	<ul style="list-style-type: none"> • Addition of glutaraldehyde into the biopolymers blend caused an improvement in mechanical property, water solubility, and water vapor permeability. • Addition of cinnamon essential oil into the biopolymers blend showed antioxidant and antimicrobial activities against <i>L. monocytogenes</i> and <i>P. aeruginosa</i>. • Addition of cinnamon essential oil and glutaraldehyde at the same time into the biopolymers blend increased the antioxidant and antibacterial activities. • Addition of oleic acid into the biopolymers blend reduced water solubility, tensile strength, antioxidant and antimicrobial activities while elasticity, and water vapor permeability increased. 	✓	✓	✓	✓	✓	Valizadeh et al. (2019)
● Gum arabic (1.5 % w/v) ● Chitosan (1.5 %w/v)	● Cinnamon essential oil (8 % w/w of total solid)	<ul style="list-style-type: none"> • Increasing gum arabic proportion into the biopolymers blend (1:0, 1:0.25, 1:0.5, 1:1, 1:2, 1:4) reduced thickness, water content, tensile strength, elasticity, and water vapor permeability. • Addition of cinnamon essential oil into the biopolymers blend showed antioxidant activity. Antioxidant activity enhanced when the ratio of chitosan/gum arabic changed from 1:0 to 1:2. Antioxidant activity quickly reduced by further increasing of gum arabic proportions in biopolymers blend to 1:4. 	✓	✓	Xu, Gao, Feng, Yang, Shen and Tang et al. (2019)			
● Gum arabic (1.5 % w/v) ● Chitosan (1.5 %w/v)	● Cinnamon essential oil (5, 10, and 15 % w/w based on gum arabic) ● Clove essential oil (10 % w/w based on gum arabic) ● cinnamon and clove essential oil combination (5 % w/w based on gum arabic)	<ul style="list-style-type: none"> • Addition of essential oils into the biopolymers blend decreased the ζ-potential and viscosity, while particle size increased. • Addition of essential oils (in particular cinnamon essential oil) caused an increase in elasticity while tensile strength and water barrier properties were reduced • Films containing cinnamon essential oils or a combination of cinnamon and clove showed better water barrier properties compared to films containing clove. • Films containing cinnamon and clove essential oils combinations exhibited better antimicrobial activity against <i>E. coli</i> and <i>S. aureus</i>. Besides, films containing cinnamon essential oil showed better antibacterial activity compared to the clove essential oil. • Composite films showed antibacterial activity against aerobic mesophilic bacteria. 	✓	✓	Xu, Gao, Feng, Huang, Yang et al. (2019)			
● Corn starch (3 % w/v) ● Cassava starch (3 % w/v)	● Glutaraldehyde(10 % w/v based on a dry biopolymer basis)						✓	Luchese et al. (2018)
● Chitosan (0.5 % w/v) ● Hardleaf oachestnut starch 0.5, 2, and 8 % w/v ● Chitosan (2 % w/v)	● <i>Litsea cubeba</i> oil (4, 8, 12, and 16 % w/w, based on biopolymers total weight)	<ul style="list-style-type: none"> • Blending chitosan and hardleaf oachestnut (ratio 1:1) caused an increase in tensile strength and an improvement in water vapor permeability. • The incorporation of <i>Litsea cubeba</i> oil into the biopolymers blend (ratio 1:1) decreased tensile strength, elasticity, water vapor permeability, water content, and water solubility while contact angle values increased. • Addition of <i>Litsea cubeba</i> oil into the biopolymers blend showed antimicrobial activity against <i>E. coli</i> and <i>S. aureus</i>. 	✓	✓	✓	✓	✓	Zheng et al. (2018)

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Table 2 (continued)

Biopolymer	Additives	Key findings	M	WB	GB	AO	AM	Reference
● Cassava starch ● Chitosan (0, 25, 50, 75, 100, and 150 mg chitosan/g starch ratios)	● Gallic acid	<ul style="list-style-type: none"> Increasing chitosan proportion in biopolymers blend decreased moisture content, water activity, water vapor permeability, total phenolic contents, and antioxidant activities. Blending chitosan and cassava starch reduced the growth of spoilage microorganisms and prolonged the shelf life of cooked ham. 	✓	✓	✓	✓	✓	Zhao, Teixeira, Gänzle, and Saidana (2018)
● Burdock root inulin (4 % w/v) ● Chitosan (2.5 % w/v)		<ul style="list-style-type: none"> Water vapor permeability, water solubility, water content, tensile strength, and lightness value of inulin-chitosan films were reduced with increasing oregano-thyme essential oils blend. Addition of oregano-thyme essential oils blend into the biopolymers blend increased elasticity, opacity, a^*, and b^* values. Active films containing oregano-thyme essential oils blend showed antioxidant and antibacterial activity against <i>E. coli</i>, <i>L. monocytogenes</i>, <i>S. aureus</i>, and <i>S. typhimurium</i>. Increasing proportion of chitosan in biopolymers blend showed a significant reduction in the growth of <i>S. enterica</i>, <i>E. coli</i> O157:H7, and <i>L. monocytogenes</i> and also had a significant increase in the antioxidant activity. Blending chitosan and corn starch showed an increase in water solubility, total color differences, tensile strength and elasticity, and a reduction in crystallinity, elastic modulus, and water vapor permeability. Increasing concentration of chitosan in biopolymers blend caused an increase in water vapor permeability and water content values. Addition of plant extracts into the biopolymers blend improved UV-Vis light barrier properties. Active films containing beetroot, cranberry, and blueberry extracts showed higher antibacterial activity against <i>E. coli</i>, aerobic mesophilic bacteria, and fungi (<i>P. notatum</i>, <i>A. niger</i>, and <i>A. fumigatus</i>). 	✓	✓	✓	✓	✓	Soni, Mahmoud, Chang, El-Giar, and Hassan (2018)
● TEMPO cellulose nanofiber (0, 15, 25 and 100 wt%) ● Chitosan (0, 75, 85 and 100 wt%)		<ul style="list-style-type: none"> Corn starch (5 % w/v) ● Chitosan (1, 2, 3 and 4 % w/v) 	✓	✓	✓	✓	✓	Ren et al. (2017)
● Rice starch (2 % w/v) ● Chitosan (2 % w/v)	<ul style="list-style-type: none"> Cranberry, blueberry, beetroot, pomegranate, oregano, pitaya, and resveratrol extract (0.5, 2, and 5 % w/w based on dry biopolymers weight) 	<ul style="list-style-type: none"> Increasing pectin proportion in biopolymers blend caused an increase in water solubility, water content, and swelling index. Increasing chitosan proportion in biopolymers blend increased tensile strength and reduced elasticity values. Addition of xanthan gum into the biopolymers blend did not affect the water vapor permeability, solubility, and moisture content. Increasing xanthan gum proportion in biopolymers blend caused an increase in tensile strength while it reduced elasticity values. Increasing chitosan proportion in biopolymers blend up to 60 % (w/w) caused an increase in tensile strength and elastic modulus while elasticity values reduced. Incorporation of zinc oxide nanoparticles into the biopolymers blend showed antimicrobial activity against <i>S. aureus</i>, <i>P. aeruginosa</i>, <i>E. coli</i>, <i>C. albicans</i> and prolonged the shelf life of white soft cheese. 	✓	✓	✓	✓	✓	Baron et al. (2017)
● Xanthan gum (1.5 % w/v) ● Chitosan (1.5 % w/v)		<ul style="list-style-type: none"> Tapioca starch (3 % w/w) ● Chitosan (20, 40, 60, 80 % w/w of dry starch solid weight) ● Carboxymethyl cellulose (1 % w/v) ● Chitosan (2 % w/v) 	✓	✓	✓	✓	✓	de Moraes Lima et al. (2017)
		<ul style="list-style-type: none"> Zinc oxide nanoparticles (2, 4, and 8 % w/w) 						Shapi'i and Othman (2016)
		<ul style="list-style-type: none"> Chitosan (2 % w/v) 						Youssef El-Sayed, El-Sayed, Salama, and Dufresne (2016)

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Biopolymer	Additives	Key findings	M	WB	GB	AO	AM	Reference
● Carboxymethyl cellulose (2 % w/v) ● Quaternized chitosan (5 % w/v)		<ul style="list-style-type: none"> ● Increasing carboxymethyl cellulose proportion in biopolymers blend caused an improvement in tensile strength, thermostability, and water vapor permeability values while oxygen permeability and opacity values increased. ● Increasing carboxymethyl cellulose proportion in biopolymers blend caused a reduction in antibacterial activity against <i>E. coli</i> and <i>S. aureus</i>. ● Higher proportion of quaternized chitosan in biopolymers blend delayed the deterioration of banana fruit. ● Quaternary ammonium chitosan was water soluble over a wide range of pH values. The antibacterial activity of quaternized chitosan films was better than that of chitosan itself, but these films showed poor mechanical properties. 	✓	✓	✓	✓	✓	Hu, Wang, and Wang (2016)

Aloui, & El-Saeid, 2014), fillers incorporation (Abdelrazeq, Elashmawi, & Labeeb, 2010), high-energy irradiation (Shahbazi, Rajabzadeh, & Ahmadi, 2017) and blending with other biopolymers (Muxika, Etxabide, Uranga, Guerrero, & de la Caba, 2017; Wang, Qian, & Ding, 2018). Blending chitosan with other polymers to form a composite film could combine the advantages of the base polymers into a film with higher performances compared with those of each constituent. A series of natural and synthetic polymers have been reported to blend with chitosan, such as pectin (Baron, Pérez, Salcedo, Córdoba, & do A. Sobral, 2017; Younis & Zhao, 2019), cellulose and its derivatives (Noshirvani et al., 2017; Valizadeh, Naseri, Babaei, Hosseini, & Imani, 2019), starch (Ren, Yan, Zhou, Tong, & Su, 2017; Suriyatem, Auras, & Rachtanapun, 2018), gelatin (Bonilla, Poloni, Lourenço, & Sobral, 2018; Guo et al., 2019), soy protein isolate (Li et al., 2017), polyvinyl alcohol (Do Yoon, Kim, Kim, & Je, 2017), polylactic acid (Liu, Wang, Zhang, Lan, & Qin, 2017), etc.

4. Chitosan blends

A polymer blend is a compatible or phase-separated mixture of at least two polymers or copolymers, that is produced to enhance the physical properties of each component (Cazón & Vázquez, 2020; Khan, Mansha, & Mazumder, 2018). The objective of polymer blending is to develop composite materials in a simple and cost-effective route which would combine the features of components, possibly enhancing their useful attributes, and minimizing their drawbacks (Parameswaranpillai, Thomas, & Grohens, 2015; Unger, Sedlmair, Siesler, & Hirschmugl, 2014).

The success of polymer blending as a strategy to improve packaging materials relies on the wide range of resulting physical, thermal, mechanical, barrier, and optical properties. Therefore, studying these properties plays a key factor in the suitable formulation of blends addressed to specific applications.

The growing interest towards chitosan for packaging applications has resulted in many published studies focusing on the production and investigation of properties of films obtained from chitosan blended with other natural and synthetic polymers (Kumar, Mukherjee, & Dutta, 2020). In this study, chitosan blends have been classified into two main groups, respectively *chitosan-natural biopolymers blends* and *chitosan-synthetic polymers blends*. A synopsis of the literature published in the last five years is presented in each subsection considering: type of polymer and its concentration, active compounds, and other additives incorporated in the blend and the main properties of the blend films addressed for food packaging applications.

4.1. Chitosan-natural biopolymer blends

The functional properties of chitosan-based films can be improved by blending with other natural biopolymers such as polysaccharides, proteins, and their derivatives (Aider, 2010; Cazón & Vázquez, 2020; Elsabee & Abdou, 2013). Compatibility between chitosan and these polymers depends on the ability to associate through electrostatic interaction due to chitosan cationic character at appropriate pH conditions and the availability of high-polarity groups, such as NH/NH₂, OH, C=O, C—O—C—, in its backbone to form intermolecular hydrogen bonds or dipole association with the corresponding functional groups of other biopolymers (Bonilla et al., 2018). It has been reported that polysaccharides such as pectin, starch (from rice, corn, potato, cassava, etc.), alginate, carrageenan, xanthan gum, xylan, glucose, kefiran, cellulose, and its derivatives can be blended with chitosan (Wang et al., 2018). A synopsis of recent advances in chitosan-polysaccharide blend films for packaging applications is presented in Table 2.

Protein-based films from animal sources (gelatin, collagen, casein, whey protein, etc.) and plant source (soy protein isolate, corn zein, kidney bean protein isolate, quinoa protein, wheat gluten, etc.) have been studied for the development of biodegradable films due to their

Table 3
Synopsis of research published between 2015–2020 addressing chitosan-protein blend films for food packaging applications. Improvement of the parameters investigated in each contribution is reported as follows: M: mechanical properties, WB: water barrier property, GB: gas barrier permeability, AO: antioxidant activity, AM: antimicrobial activity.

Biopolymer	Additives	Key findings	M	WB	GB	AO	AM	Reference
Sheep bone collagen (1.5 % w/v) Skin gelatin (1.5 % w/v) Chitosan (1.5 % w/v)		<ul style="list-style-type: none"> • Addition of chitosan into bone collagen improved transparency and tensile strength of bone collagen film. • Increasing proportion of chitosan over 50 % in the biopolymers blend improved the elasticity value. • Blending chitosan and bone collagen led to an improvement in UV barrier property, water solubility, and thermal stability while water vapor permeability increased. 	✓	✓				Hou et al. (2020)
Hordein nanofiber (11 % w/v) Chitosan (0.4 % w/v)	• Quercetin (5 % w/w based on hordein)	<ul style="list-style-type: none"> • Addition of quercetin into the nano biopolymers blend showed antioxidant activity and treating films with different temperatures (90, 120, 150, and 180 °C) did not influence the antioxidant activity. • Heat treatment enhanced the water resistance of nano biopolymers blend. • Covering apple and potato samples with heat treated nano fiber films delayed the rate of enzymatic browning and preserved their fresh color after 6 and 12 h, respectively. 	✓	✓				Li, Yan, Guan, and Huang (2020)
Zein (2 % w/v) Chitosan (2 % w/v)	• α -tocopherol (50 % w/w based on the content of dry materials)	<ul style="list-style-type: none"> • Active zein-chitosan films containing α-tocopherol reduced the postharvest deterioration of mushroom (<i>Agaricus bisporus</i>) at 4 °C for 12 days. This was mainly due to the excellent gas barrier property and antioxidant activity of films. 	✓	✓				Zhang, Liu, Sun, Wang, and Li (2020)
Gelatin (2 % w/v) Chitosan (2 % w/v)	• Polyphenols from the fruits of Chinese hawthorn (2, 4, and 6 % w/w on the total biopolymer weight)	<ul style="list-style-type: none"> • Addition of the polyphenol extract into the biopolymers blend increased thickness, tensile strength, elasticity, opacity, and total color difference, while water content and water vapor permeability reduced. • Antioxidant activity of films significantly improved by increasing the concentration of polyphenol extract. 	✓	✓				Kan et al. (2019)
Gelatin (5 % w/v) Chitosan (3, 6 and 9 % w/w based on gelatin)	• Citric acid (10 and 20 % w/w based on gelatin weight)	<ul style="list-style-type: none"> • Incorporation of citric acid into the biopolymer blend improved elasticity values. • Higher concentration of chitosan and citric acid in biopolymers blend led to better antibacterial activity against <i>E. coli</i>. 	✓	✓				Uranga et al. (2019)
Fish myofibrillar protein (0.45, 0.8, 1.3, 1.8, and 2.14 % w/v) Chitosan (13.18, 20, 30, 40, and 46.81 % w/w)		<ul style="list-style-type: none"> • The optimum formulation to produce biodegradable film contained 1.3 % (w/v) fish myofibrillar proteins, 30 % (w/w) chitosan, and 40 % (w/w) glycerol. • Adding chitosan in the biopolymers blend increased elasticity, thermal stability, UV barrier properties while solubility, swelling degree, and water vapor permeability decreased. • The porcine plasma protein/chitosan blend film had lower transparency than neat porcine plasma protein and chitosan film. • Blending porcine plasma protein and chitosan increased thermal stability. • Blend films (ratio 1:1) showed improvement in water resistance and water vapor permeability, solubility, and mechanical properties compared to the neat porcine plasma protein film. 	✓	✓				Samsalee and Sothornvit (2019)
Gelatin (2 % w/v) Chitosan (1 % w/v)	• Porcine plasma protein (3 % w/w)	<ul style="list-style-type: none"> • Cinnamom, citronella, pink clove, nutmeg and thyme essential oils (1 % w/w based on weight) 	✓	✓				Haghghi, Biard et al. (2019)
Gelatin (1 % w/v) Chitosan (1 % w/v)	• Cinnamon essential oil (0.4 % w/w based on a dry biopolymer weight)	<ul style="list-style-type: none"> • Cinnamom essential oil into the biopolymer blend improved elasticity, thermal stability, water vapor permeability, UV barrier, and contact angle values. • Antibacterial activity against <i>C. jejuni</i>, <i>E. coli</i>, <i>L. monocytogenes</i>, and <i>S. Typhimurium</i>. • Addition of cinnamon essential oil into the biopolymer blend improved elasticity, thermal stability, water vapor permeability, UV barrier, and contact angle values. 	✓	✓				Guo et al. (2019)
Collagen (3.5 % w/v) Chitosan (1 % w/v)	• Pomegranate peel extract (1, 3, and 5 % v/v)	<ul style="list-style-type: none"> • Active films containing cinnamon essential oils showed antibacterial activity against <i>E. coli</i> and <i>S. aureus</i>. • Addition of 5 % pomegranate peel extract into the biopolymers blend caused a reduction in water solubility and enhanced antibacterial activity against <i>B. sphaerophylax</i>, <i>B. subtilis</i>, <i>S. typhi</i>, and <i>E. coli</i>. 	✓	✓				Bhuimbar, Bhagwat, and Dandge (2019)

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Table 3 (continued)

Biopolymer	Additives	Key findings	M	WB	GB	AO	AM	Reference
• Zein (10 % w/v) • Chitosan (6 wt%)	• TiO ₂ nanoparticles (0.05, 0.1, 0.15, 0.2, and 0.25 % w/w)	<ul style="list-style-type: none"> • Addition of TiO₂ nanoparticles up to 0.2 % into the biopolymers blend enhanced water absorption and water vapor permeability. • Biopolymers blend containing TiO₂ nanoparticles showed antibacterial activity against <i>S. aureus</i>, <i>E. coli</i>, and <i>S. enteritidis</i>. • Blending chitosan and gelatin caused an improvement in mechanical and water barrier properties. • Incorporation of ethyl lauroyl arginate into the biopolymer blend conferred antibacterial activity against <i>C. jejuni</i>, <i>E. coli</i>, <i>L. monocytogenes</i>, and <i>S. typhimurium</i>. 	✓	✓	✓	✓	✓	Qu et al. (2019)
• Gelatin (2 % w/v) • Chitosan (2 % w/v)	• Ethyl lauroyl arginate (0.1 % v/v)	<ul style="list-style-type: none"> • Addition of silver nanoparticles into the biopolymers blend enhanced elasticity values while tensile strength and light transmittance in the visible light region were reduced. • Incorporation of silver nanoparticles into the biopolymers blend showed antimicrobial activity. • Shelf life of red grapes fruits wrapped with gelatin-chitosan blend enriched with silver nanoparticles was extended for additional two weeks. • Addition of β-carotene loaded starch nanocrystals into the biopolymers blend caused a significant reduction in water solubility while antioxidant activity increased. • Addition of gallic acid into the biopolymers blend increased opacity and elasticity. • Incorporation of β-cyclodextrin and gallic acid into the biopolymers blend reduced water barrier properties. 	✓	✓	✓	✓	✓	Haghghi, De Leo et al. (2019)
• Gelatin (1 % w/v) • Chitosan (2 % w/v)	• Silver nanoparticle (0.05 and 0.1 % w/w)	<ul style="list-style-type: none"> • Shelf life of red grapes fruits wrapped with gelatin-chitosan blend enriched with silver nanoparticles was extended for additional two weeks. • Addition of β-carotene loaded starch nanocrystals into the biopolymers blend caused a significant reduction in water solubility while antioxidant activity increased. • Addition of gallic acid into the biopolymers blend increased opacity and elasticity. • Incorporation of β-cyclodextrin and gallic acid into the biopolymers blend reduced water barrier properties. 	✓	✓	✓	✓	✓	Kumar, Shukla, Baul, Mitra, and Halder (2018)
• Gelatin (3 % w/v) • Chitosan (1 % w/v)	• β-Carotene loaded starch nanocrystals (1mg/1 mL)	<ul style="list-style-type: none"> • Gallic acid (1 % w/w total dry weight of film) • Tween 80 (50 and 100 % w/w based on the weight of the gallic acid) • β-cyclodextrin • Ethanol • Eugenol and ginger essential oils (0.5 % w/w based on dry biopolymers weight) 	✓	✓	✓	✓	✓	Hari, Francis, Rajendran Nair, and Nair (2018)
• Gelatin (4 % w/v) • Chitosan (1 % w/v)	• Procyandin (0.25, 0.5, 0.75, and 1 mg/mL)	<ul style="list-style-type: none"> • Addition of essential oils into the biopolymers blend improved elasticity and UV barrier properties. • Incorporation of essential oils into biopolymers blend showed significant antioxidant activity. • Addition of procyandin into the biopolymers blend improved elasticity, water vapor permeability, water solubility, swelling index, and UV barrier properties while tensile strength was reduced. • Incorporation of procyanidin into the biopolymers blend showed antioxidant activity and antibacterial activity against <i>S. aureus</i> and <i>E. coli</i> strains. • Elasticity, thermal stability, and homogeneity of chitosan film increased by blending with soy protein isolate. 	✓	✓	✓	✓	✓	Ramzia, Ma, Yao, Wei, and Huang (2018)
• Gelatin (3 % w/v) • Chitosan (1 % w/v)	• Cu nanoclusters (20 mmol/L)	<ul style="list-style-type: none"> • Increasing chitosan proportion in biopolymers blend improved the mechanical properties and water vapor permeability, contact angle, and thermal stability. • Blending chitosan and gelatin showed an improvement in elasticity, water solubility, and water barrier property. 	✓	✓	✓	✓	✓	Xing, Zhang, Li, Li, and Shi (2018)
• Eggshell membrane gelatin (3 % w/v) • Chitosan (1.5 % w/v) • Gelatin (4 % w/v) • Chitosan (1 % w/w)	• Cinnamon, guarana, rosemary and boldo-dochile ethanolic extract (1 % v/v)	<ul style="list-style-type: none"> • Increasing chitosan proportion in biopolymers blend improved the mechanical properties and water vapor permeability. • Addition of ethanolic extracts into the biopolymers blend enhanced antioxidant and antibacterial activity against <i>S. aureus</i> and <i>E. coli</i>. • Blending chitosan and gelatin showed UV barrier property. • Addition of boric acid into the biopolymer blend improved tensile strength and water solubility, moisture content, and water vapor permeability. • Addition of polyethylene glycol into the biopolymers blend caused an increase in elasticity, water content, and water solubility. 	✓	✓	✓	✓	✓	Ahmed and Ikram (2016)
• Gelatin (10 % w/v) • Chitosan (2 % w/v)	• Boric acid (2, 3, 4 and 5 % w/w) • Polyethylene glycol (5, 10 and 20 % v/v)	<ul style="list-style-type: none"> • Addition of sorbitol into the chitosan-starch or chitosan-gelatin blend films enhanced water vapor permeability compared with films containing glycerol. 	✓	✓	✓	✓	✓	Badawy, Rabea, and El-Noutby (2016)

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Table 3 (continued)

Biopolymer	Additives	Key findings	M	WB	GB	AO	AM	Reference
● Quinoa protein (6.7 % w/v) ● Chitosan (1.5 and 2 % w/v) ● Chitosan-tri polyphosphate nano particles (0.3 w/v)	● Thymol nanoparticles (0.1 % w/v)	<ul style="list-style-type: none"> • Addition of glycerol and sorbitol into the biopolymers blend showed antioxidant activity. • Addition of chitosan-tri polyphosphate nanoparticles into the biopolymers blend improved water vapor permeability. • Incorporation of chitosan-tri polyphosphate-thymol nanoparticles into the biopolymers blend enhanced antibacterial activity against <i>L. innocua</i>, <i>S. aureus</i>, <i>S. typhimurium</i>, <i>E. aerogenes</i>, <i>P. aeruginosa</i>, and <i>E. coli</i>. 	✓	✓	✓	✓	✓	Caro et al. (2016)
● Gelatin (3 % w/v) ● Chitosan (2 % w/v)	● Red grape seed extract (1 and 2 % w/w) ● <i>Ziziphora clinopodioides</i> essential oil (1 and 2 % w/w)	<ul style="list-style-type: none"> • Addition of red grape seed extract and <i>Ziziphora clinopodioides</i> into the biopolymers blend showed antibacterial activity against <i>L. monocytogenes</i>, total mesophilic and psychrotrophic bacteria, <i>Pseudomonas</i> spp., <i>P. fluorescens</i>, <i>S. puurificans</i>, lactic acid bacteria and <i>Enterobacteriaceae</i> family. • Packing minced rainbow trout fillets with chitosan-gelatin blend enriched with red grape seed extract and <i>Ziziphora clinopodioides</i> essential oil extended the shelf life at refrigerated condition due to the delay of lipid oxidation and inhibition of bacterial growth. • Blending brewer's spent grain protein with chitosan caused an improvement in water vapor permeability and mechanical properties. • Blend films showed antioxidant and antibacterial activity against <i>S. aureus</i>, <i>E. coli</i>, <i>L. monocytogenes</i>, and <i>S. typhimurium</i>. 	✓	✓	✓	✓	✓	Lee, Lee, Yang, and Song (2015)
	● Brewer's spent grain protein (3 % w/v) ● Chitosan (2 % w/v)							

high abundance, acceptable mechanical properties, excellent gas barrier properties to non-condensable gases (oxygen, carbon dioxide, and nitrogen) and aromas (Arfat, Ahmed, Hiremath, Auras, & Joseph, 2017). The chitosan-protein blend film could render better functional properties than single proteins and chitosan film, thus promoting their application in food packaging (Basta, Khwaldia, Aloui, & El-Saied, 2015; Ma et al., 2012; Wang et al., 2018). A synopsis of recent advances in chitosan-protein blend films for packaging applications is presented in Table 3.

4.2. Chitosan-synthetic polymers blends

Blending chitosan with synthetic polymers (polyvinyl alcohol - PVA, polyvinyl pyrrolidone - PVP, polylactic acid - PLA, etc.) has been extensively studied for the positive effects on the physical, mechanical, and biological features of composite films. The success of synthetic polymers as biodegradable materials depends on their diverse range of mechanical properties, chemical resistance, and low production costs compared to natural polymers (Bourakadi et al., 2019). Chitosan is potentially miscible with some synthetic polymers mainly due to the formation of intermolecular hydrogen bonds between hydroxyl groups of synthetic polymer and hydroxyl and amine groups of chitosan (Bonilla, Fortunati, Atarés, Chiralt, & Kenny, 2014). Depending on the interactions between polymer components, blending chitosan with synthetic biopolymers can enhance the mechanical and water barrier properties of films in some cases. A synopsis of recent advances in chitosan-synthetic polymer blend films for packaging applications is presented in Table 4.

5. Conclusions and future perspectives

Packaging is an essential item responsible for the protection of the product and provides food safety assurance during marketing. Conventional plastic packaging material due to non-biodegradability and insufficient waste management system has labeled the food industry as a source of pollution and social concerns. Therefore, bio-based, and biodegradable materials have received considerable attention to address these issues in recent years. The adoption of chitosan as packaging material could contribute to mitigating the environmental concern, despite some drawbacks in terms of thermal stability, barrier and mechanical properties, and production costs. Blending chitosan with other natural and synthetic polymers is an effective way to overcome these limitations, making the films suitable for specific uses. This approach appears to have a bright future for innovative food packaging design since it will allow the partial replacement of the existing synthetic plastic packaging materials presently available in the market. Potential applications of chitosan blend-based films are for fresh products (vegetable, meat and fish) and foods with short to medium shelf life. Such films may represent an interesting alternative to conventional plastic films especially when recycling is not possible or is compromised due to presence of food residues: in these cases, the feature of biodegradability/compostability offers a valid end-of-life alternative. Nanotechnologies certainly represent a promising complementary tool for further improvement of mechanical and barrier properties of chitosan-based films and for the addition of other functionalities (antimicrobial and antioxidant capacity), but their development and application faces resistance due to toxicological issues. In particular, nanocomposite materials based on the incorporation of biobased nanomaterials such as nano-cellulose, emerge for their potential to provide a barrier and mechanical strength maintaining the full biodegradability of the material. Finally, despite contributions retrieved in literature in the considered timeframe, the aspect of biodegradability remains mostly disregarded. This omission has two reasons: the first is linked to the difficulty of applying a real holistic and multidisciplinary approach, necessary for the development of food packaging; the second arises from the consideration that making a new material from

Table 4
Synopsis of research published between 2015 – 2020 addressing chitosan-synthetic biopolymer blend films for food packaging applications. Improvement of the parameters investigated in each contribution is reported as follows: M: mechanical properties, WB: water barrier properties, GB: gas barrier permeability, AO: antioxidant activity, AM: antimicrobial activity.

Biopolymer	Additives	Key findings	M	WB	GB	AO	AM	Reference
● PVA (0.5, 1, 1.5, and 2 % w/v) ● Chitosan (0.5, 1, 1.5, and 2 % w/v)	● Ethyl vanillin (2 % w/v)	<ul style="list-style-type: none"> ● Addition of ethyl vanillin into the biopolymers blend caused an improvement in the tensile strength, surface hydrophobicity, and UV barrier properties while water vapor transmission rate and oxygen transmission rate were reduced. ● Blend films containing ethyl vanillin showed antimicrobial activity against <i>S. aureus</i> and <i>E. coli</i>. ● Addition of glycerol reduced the extent of chitosan blends while polyethylene glycol. 	✓	✓	✓	✓	✓	Narasgoudar, Hegde, Vanjeri, Chougale, and Masti (2020)
● PVA (4 % w/w) ● Chitosan (2 % w/w)	● Glycerol ● Polyethylene glycol ● Glycerol/ Polyethylene glycol	<ul style="list-style-type: none"> ● Combination of glycerol and polyethylene glycol led to the highest level of compatibility. Flexibility of the blend plasticized with the combination of polyethylene glycol and glycerol was improved five-times of the blend plasticized with glycerol. However, the antibacterial activity of the chitosan/PVA blend plasticized with polyethylene glycol /glycerol was significantly reduced compared to that of glycerol or polyethylene glycol. ● Addition of silica into the biopolymers blend up to 0.2% (wt.) improved the tensile strength and elasticity values. ● Addition of silica into the biopolymers blend up to 0.2% (wt.) improved the browning index of fresh cherries. ● Compared with synthetic food packaging materials, the blend films degraded quickly in the soil while addition of silica slightly reduced the biodegradability. ● Addition of ethyl lauroyl arginate into the biopolymers blend negatively influenced elasticity, tensile strength, and water barrier properties while barrier properties to UV light improved. ● Chitosan-gelatin blend enriched with ethyl lauroyl arginate exhibited antibacterial activity against <i>C. jejuni</i>, <i>E. coli</i>, <i>L. monocytogenes</i> and <i>S. typhimurium</i>. 	✓	✓	✓	✓	✓	Haghghi et al. (2020)
● PVA (4 % w/w) ● Liquidified chitin (1 % w/w)	● Silica (0.1, 0.2, 0.4, 0.8, and 1.6 wt/w)	<ul style="list-style-type: none"> ● Ethyl lauroyl arginate (1, 2.5, 5, and 10 % w/w) 	✓	✓	✓	✓	✓	Vadivel et al. (2019)
● PVA (5 % w/v) ● Chitosan (1 % w/v)		<ul style="list-style-type: none"> ● Curcumin (0.01 w/v) 	✓	✓	✓	✓	✓	Hasanpour Ardakani-Zadeh and Hosseini (2019)
● PVA (0.5 % w/v) ● Xylan (0.5 % w/v) ● Chitosan (1 % w/v) ● Nano hydroxyapatite ● (0.01 w/v)		<ul style="list-style-type: none"> ● Oregano essential oil (1, 3, and 5 % w/w) 	✓	✓	✓	✓	✓	Ghaderi, Hosseini, Keyvani, and Gómez-Guillén (2019)
● Poly(ϵ -capro-lactone) (6 % w/v) ● Chitosan (1 % w/v)		<ul style="list-style-type: none"> ● Thiabendazole-ium-montmorillonite (5 % w/w) 	✓	✓	✓	✓	✓	Bourakadi et al. (2019)
● PVA (2 % w/v) ● Fish gelatin (2 % w/v) ● Chitosan (1.5 % w/v) ● PVA (4 % w/v) ● Chitosan (1.25 % w/v)		<ul style="list-style-type: none"> ● PVA (2 % w/v) ● Increasing chitosan proportion in biopolymers blend caused an increase in water vapor permeability, water absorption, and opacity values while water solubility, tensile strength, and elasticity values reduced. ● Addition of thiabendazole into the biopolymers blend caused an increase in tensile strength and elastic modulus values. Chitosan-PVA blend films containing thiabendazole showed antibacterial activity against <i>P. aeruginosa</i>, <i>S. aureus</i>, and <i>E. coli</i>. ● Increasing chitosan proportion in the biopolymer blend caused a reduction in tensile strength and elasticity values while UV barrier properties improved. 	✓	✓	✓	✓	✓	Wu, Ying, Liu, Zhang, and Huang (2018)

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Table 4 (continued)

Biopolymer	Additives	Key findings	M	WB	GB	AO	AM	Reference
● Microcrystalline cellulose (3, 4 and 5 % w/w)		● Addition of chitosan-PVA blend into the cellulose based films caused an improvement in mechanical properties.	✓					Cazón, Vázquez, and Velazquez (2018)
● PVA (2 and 4 % w/w)		● Blending chitosan and cellulose improved barrier properties to the UV light.						
● Chitosan (0.5 and 1 % w/w)		● Addition of PVA into the biopolymers blend caused an improvement in light transmittance values.						
● PVA (3 % w/v)		● Addition of cellulose nanocrystals into PVA-chitosan blend improved tensile strength and thermal properties.	✓					Perumal, Sellamuthu, Nambiar, and Sadiku (2018)
● Chitosan (1.5 % w/v)	% w/w base on polyvinyl alcohol-chitosan blend)	● PVA-chitosan blend films enriched with cellulose nanocrystal from rice straw showed antifungal activity against <i>C. gloeosporioides</i> and antibacterial activity against <i>S. mutans</i> , <i>S. aureus</i> , <i>E. coli</i> , and <i>P. aeruginosa</i> .	✓					Yu, Ji, Chu, and Zhang (2018)
● PVA (1 % w/v)	● SiO ₂ (0.3, 0.6, and 0.9 %w/w)	● Addition of silicon dioxide into the biopolymers blend caused an enhancement in mechanical, water and oxygen barrier properties.	✓					Liu, Wang, and Lan (2018)
● Chitosan (2 % w/v)		● In comparison to neat PVA film, blending PVA and chitosan caused an improvement in elasticity and oxygen barrier properties while water barrier properties decreased. Increasing proportion of chitosan in biopolymers blend showed better antibacterial activity against <i>S. aureus</i> and <i>E. coli</i> .	✓					
● PVA (10 %w/v)		● Addition of sulfosuccinic acid into the biopolymers blend caused an enhancement in tensile strength, elasticity, swelling degree, water solubility, thermal stability, and optical properties.	✓					Yun, Lee, Kim, and Yoon (2017)
● Chitosan (2.5, 3, and 3.5 % w/v)								
● PVA (2 % w/v)	● Sulfosuccinic acid ● Glycerol (0–60 wt%)	● Increasing chitosan-gallic acid concentration into the biopolymers blend and treating with UV caused an increase in tensile strength while elasticity values were reduced.	✓					Do Yoon et al. (2017)
● Chitosan (2 % w/v)	● Xyitol (0–60 wt%)	● Films containing chitosan-gallic acid (1 %w/v) showed antibacterial activity against <i>E. coli</i> , <i>S. typhimurium</i> , <i>S. aureus</i> , and <i>B. cereus</i> .	✓					Zhang et al. (2017)
● PVA (5 % w/v)		● Montmorillonite (0, 5, 10, 15, and 20 % w/w, based on the dry weight of CS/PVA)	✓					
● Sodium lactate loaded chitosan (2 % w/v)		● Montmorillonite concentration up to 15 % in biopolymers blend improved tensile strength and elastic modulus while elasticity values decreased.	✓					
● PVA (5 % w/v)	● Carvacrol (5 % w/v)	● Addition of montmorillonite into the biopolymers blend enhanced barrier properties to water vapor, oxygen, and carbon dioxide. Blend films showed antibacterial activity against <i>E. coli</i> .	✓					
● Chitosan (0.15 % w/w)	● Cellulose nanocrystals (3 % w/w)	● Blend films showed improvement in mechanical properties while color and transparency were not affected.	✓					
● PVA (2 % w/v)		● Addition of carvacrol and cellulose nanocrystal into the biopolymer blend caused an improvement in antioxidant activity and antimicrobial activity against <i>P. carotovorum</i> subsp., <i>Oidiniform</i> , and <i>X. axonopodis</i> .	✓					
● Chitosan (2 % w/v)		● Blend films showed antioxidant activity and antimicrobial activity against <i>E. coli</i> , and <i>S. typhimurium</i> .	✓					
● PVA (10, 20, and 30 % w/w)		● Increasing proportion of PVA in biopolymers blend caused an increase in tensile strength and elasticity.	✓					Hajji et al. (2016)
● Montmorillonite (5 %w/v)		● Increasing proportion of chitosan in the blend improved antioxidant activity and antibacterial activity against <i>S. aureus</i> , <i>B. cereus</i> , <i>M. luteus</i> , <i>S. enterica</i> , <i>E. coli</i> , and <i>S. typhimurium</i> .	✓					
● Chitosan (2 % w/v)		● Incorporation of montmorillonite into the biopolymer blend enhanced the mechanical and antimicrobial activities while barrier properties to water and oxygen improved.	✓					
● EVOH (4 % w/v)	● Nano zinc oxide (1 and 2 %w/w)	● Addition of nano zinc oxide into the biopolymers blend caused an improvement in barrier properties against water vapor and oxygen.	✓					Sadeghi and Shahedi (2016)
● Chitosan		● Presence of chitosan and nano zinc oxide caused excellent antimicrobial activity against <i>A. niger</i> and <i>E. coli</i> . Adding nano zinc oxide improved barrier, mechanical, and antimicrobial properties.	✓					

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Biopolymer	Additives	Key findings	M	WB	GB	AO	AM	Reference
PVA	Potassium nitrate ● (0.1, 0.2, 0.3, 0.4, and 0.5 % w/w)	● Addition of potassium nitrate into the biopolymers blend caused an improvement in tensile strength and elasticity due to its crosslinking effect. ● The degradation behavior can be improved with addition of potassium nitrate.	✓					Jahan et al. (2016)
Chitosan (1 % w/v)	Methylidiphenyl diisocyanate (0.2, 1, 2, and 3 % w/w of the final PLA/chitosan solution)	● Increasing concentration of methylidiphenyl diisocyanate in biopolymers blend improved tensile strength and contact angle values.	✓					Gartner, Li, and Almenar (2015)
PLA	Chitosan (1 % w/v)							

biodegradable components would result in a new biodegradable material. This should not be taken for granted and requires verification especially in the case of film incorporation of antimicrobial compounds. Indeed, compounds which inhibit microbial food spoilage might also cause negative effects in the composting process. Hence, future research on chitosan-based films and sustainable materials, in general, should include biodegradability among the targeted parameters.

Declaration of Competing Interest

The authors report no declarations of interest.

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Table 4 (continued)

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