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Chitosan for food packaging: Recent advances in active and intelligent films

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ABSTRACT

The massive and uncontrolled use of food packaging derived from petroleum-based plastics has created a serious environmental problem. Hence, the food packaging industry needs to develop packaging from biodegradable polymers. Among the many raw materials studied in the literature, chitosan is one of the most abundant polysaccharides in the nature. Chitosan has attracted attention due to its non-toxicity, antimicrobial, and antifungal properties. Because of this, chitosan is considered a perfect material for the development of films for food use. In this review, recent studies on active and/or intelligent chitosan-based films has been evaluated. Active packaging maintains or improves the condition of packaged food or extends its shelf-life meanwhile intelligent packaging monitors the condition of packaged food or the environment surrounding the food. The effect of the addition of active compounds on the mechanical, barrier and functional properties of chitosan-based films has been assessed. The antimicrobial and antioxidant activity, as well as the potential application of these active and intelligent composite films have also been revised. Literature shows that the presence of phenolic compounds improves both mechanical and barrier properties of chitosan films. The antimicrobial and antioxidant capacity of the films improved significantly by the addition of essential oils, phenolic compounds, and other fruit extracts. Intelligent pH-indicator chitosan-based films have been extensively studied. Further research on chitosan and its combinations with other materials is needed to study which type of foodstuffs could be in contact with chitosan packaging.

1. Introduction

Food packaging plays an important role protecting from chemical, physical, and biological hazards along the food chain. Packaging is essential to deal with the influences (odors, shocks, dust, temperature, light, humidity) that facing food (Kalpana, Priyadarshini, Maria Leena, Moses, & Anandharamakrishnan, 2019). The development of petroleum-based polymers such as polypropylene, polyester and ethylene vinyl alcohol and others allowed the changing of metal, glass or cardboard packaging into plastic packaging (Brody, Bugusu, Han, Sand, & McHugh, 2008). Nowadays, petroleum-based products are the most widely used materials in the food packaging industry because of its good properties at relative low price (Ludwicka, Kaczmarek, & Białkowska, 2020).

Nevertheless, massive use of petroleum-based materials leads to a negative impact on the environment since they are not derived from sustainable sources, not recyclable, compostable or biodegradable (Ludwicka et al., 2020; Motelica et al., 2020). Food packaging research must address the environmental problems derived from the uncontrolled consumption and management of non-biodegradable materials, developing new alternative materials with lower environmental impact. This way, biodegradable natural polymers are the focus of a wide range of research to apply them as alternatives to petroleum-based synthetics. Biodegradable natural polymers widely studied for prospective applications in food packaging industry are chitosan (Priyadarshi & Rhim, 2020), cellulose (Cazón & Vázquez, 2021; da Silva et al., 2021; de Medeiros et al., 2021; Wang, Lu, & Zhang, 2016), starch (Fang, Fu, Tao, Liu, & Cui, 2020), whey protein (Schmid & Müller, 2018) or gelatin (Ramos, Valdés, Beltrán, & Garrigós, 2016) among others. Among the available natural biodegradable materials, chitosan has aroused great interest in recent decades. As shown Fig. 1, a strong increase in the number of publications related to chitosan and packaging industry in recent years can be observed.

Chitosan and its derivatives are biodegradable polysaccharides, nontoxic and biocompatible, as well as with antimicrobial, antifungal and chelating metals properties (Aider, 2010; Bakshi, Selvakumar, Kadirvelu, & Kumar, 2020). Besides, thanks to its extraordinary film-forming properties, chitosan has been investigated for use mainly in the food,

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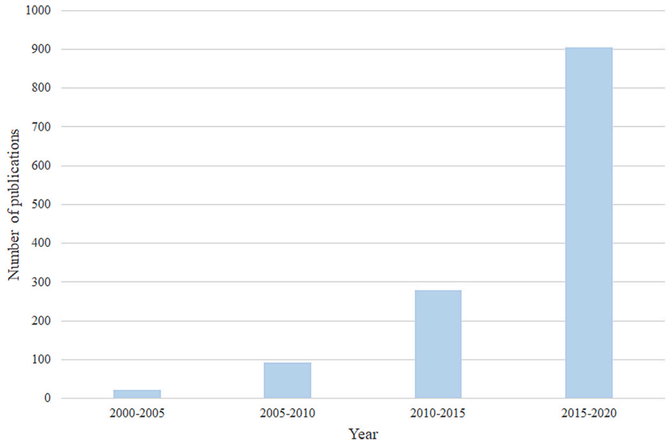


Fig. 1. Scopus indexed publications related to chitosan and the food packaging.

biomedical and chemical industries (Siripatrawan & Harte, 2010).

Chitosan is synthesized by deacetylation of chitin (poly (β-(1 → 4)-N-acetyl-D-glucosamine)), an important natural polysaccharide known since 1884. Chitin is synthesized by many living organisms. It can be obtained from multiple renewable sources, mainly waste from the seafood industry (Cazón & Vázquez, 2019; Younes & Rinaudo, 2015). Hence, chitosan is a cheap and commercially available polysaccharide. In solid phase, chitosan is semi-crystalline, and generally soluble in dilute organic acid such as acetic, citric, formic, lactic, malic or tartaric acid among others (Cazón & Vázquez, 2019; Rhim, Weller, & Ham, 1998). Due to the mentioned chitosan properties, the interest in using it as a material for food packaging films or coatings is growing.

Recent changes in food demand, industrial production trends (fresh, tasty, longer shelf-life and quality controlled products), retail sales and consumer lifestyles (limited time for shopping and cooking) are leading to the evolution of new packaging techniques beyond a passive container and physical barrier tool (Dainelli, Gontard, Spyropoulos, Zondervan-van den Beuken, & Tobback, 2008).

Novel food packaging techniques should target the main causes of food product deteriorations and ensure greater food safety. Thus, the packaging industry is looking for solutions that allow to have functional properties such as specific gas barrier, gas or moisture absorbents, UV protection, antioxidant activity, antimicrobial properties, or monitoring capacity to report product quality. These new packages will extend the food shelf-life, and will allow products to be stored for longer periods (Wyrwa & Barska, 2017). The two mains innovative packaging technologies that have emerged to face this challenge are active and intelligent packaging.

Active packaging is the materials and articles that are intended to maintain or improve the conditions of the packaged food or to extend its shelf-life (European Food Safety Authority (EFSA) (2009). This type of packaging interacts with food, maintaining nutritional quality, inhibiting the growth of pathogenic and non-pathogenic microorganisms or preventing the migration of contaminants. Typical examples of active packaging are oxygen scavengers, carbon dioxide emitters/absorbers, moisture absorbers, ethylene absorbers, ethanol emitters, antioxidants, flavor releasing/absorbing systems, and antimicrobial films (Ozdemir & Floros, 2004).

On the other hand, intelligent packaging is materials and articles that monitor the condition of packaged food or the environment surrounding the food (European Food Safety Authority (EFSA) (2009); Restuccia et al., 2010). These packages can communicate the conditions of the package food. However, these packages do not interact with the food (Müller & Schmid, 2019). This packaging performs intelligent functions such as: detecting, register, tracking and communicating information to extend shelf-life, improve quality, and warn of potential problems (Yam, Takhistov, & M, 2006).

For instance, time-temperature indicators, gas leakage indicators, relative humidity or freshness sensors are different types of intelligent packaging (Müller & Schmid, 2019). The main properties of active and intelligent packaging are summarized in Fig. 2.

Regarding the legislative framework, specific legislation has been developed in the European Union (EC 450/2009) on active and intelligent materials and articles intended to come into contact with food (European Union, 2009). In addition, substances responsible for performing the active or intelligent function of the material must be evaluated to ensure that they are safe and comply with the requirements of Regulation (EC 1935/2004) on materials and articles intended to come into contact with food (European Commission, 2004, pp. 4–17).

Considering the above properties, chitosan can be considered as a biopolymer of great interest and suitable for developing active or intelligent biodegradable food packaging.

The purpose of this review is to present a concise assessment of the latest developments in active and intelligent packaging based on chitosan. Emphasis will be put on the effect of the active additives on the mechanical, barrier and functional properties of the developed active and/or intelligent chitosan-based films. Finally, their application and future perspectives will be assessed.

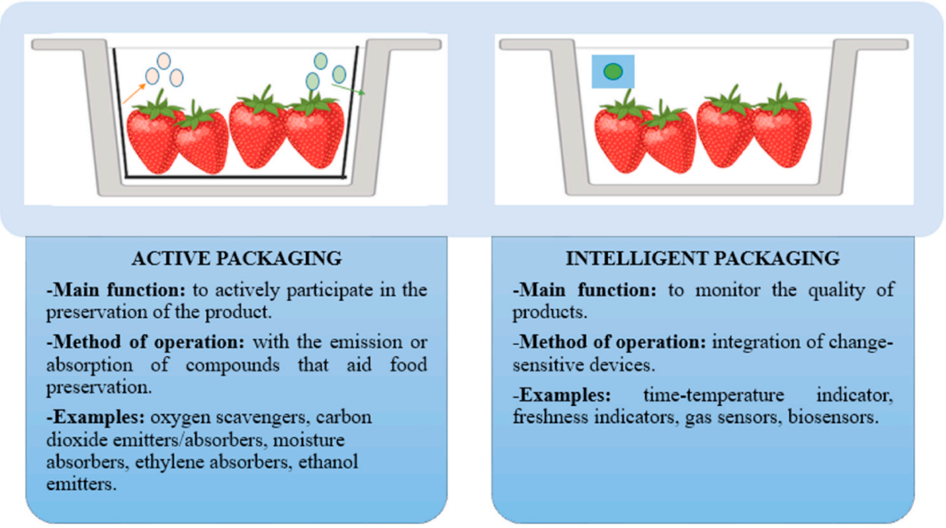
2. Active and intelligent films based on chitosan

Active film is one to which active components are deliberately added, which release or absorb substances with the aim of increasing the shelf-life, maintaining the quality and sensory characteristics of the food (Yildirim et al., 2018). Active packaging can be divided into two sections: active scavenging systems (absorbers) and active release systems (emitters).

Active release systems include antioxidant releasers, CO_2 emitters and antimicrobial packaging systems (Yildirim et al., 2018). In the literature, most of the active films developed using chitosan as the main polymeric matrix are antioxidant and/or antimicrobial films. It is mainly due to the great interest that have aroused in recent years on the use of active agents from natural sources, such as essential oils and phenolic compounds widely distributed in fruits, vegetables, legumes, seeds, among others. These active agents with powerful antimicrobial and/or antioxidant properties combined with biodegradable polymeric matrices are presented as a promising tool to develop food coatings or films. These active materials directly applied in food allow extending its shelf-life, guaranteeing its quality and food safety. For instances, chitosan films combined with tree tea, bergamot, clove bud, cinnamon or *Eucalyptus globulus* among other essential oils have shown both antimicrobial and antioxidant properties (Hafsa et al., 2016; Perdones, Vargas, Atarés, & Chiralt, 2014; Sánchez-González, Cháfer, Chiralt, & González-Martínez, 2010; Sánchez-González, González-Martínez, Chiralt, & Cháfer, 2010; Xin Zhang, Zou, et al., 2019).

Active scavenging systems include oxygen scavengers, moisture scavenger and ethylene absorbers (Yildirim et al., 2018). Oxygen scavengers in form of film or pad absorb the oxygen present in the headspace of the package, leading to extend the shelf-life and retainment of the food quality (Dey & Neogi, 2019). Singh, Nwabor, Sukri, and Voravuthikunchai (2021) have developed Chitosan-based films with oxygen scavenger properties by adding gallic acid and sodium carbonate to the polymer matrix. On the other hand, ethylene is a growth-stimulating hormone that accelerates maturation and senescence through increasing the rate of food respiration (Ozdemir & Floros, 2004). The incorporation of TiO_2 nanoparticles in chitosan films provides ethylene scavengers properties to the biopolymer (Siripatrawan & Kaewklin, 2018; Xin Zhang, Zou, et al., 2019).

Regarding intelligent films based on chitosan, two main groups of intelligent packaging from chitosan were developed: (1) films with a visual color alteration due to colorimetric reactions and (2) sophisticated biosensors. In the first group time-temperature indicators, freshness indicators or pH indicators are included (Kalpana et al., 2019). This



ACTIVE PACKAGING

-Main function: to actively participate in the preservation of the product.

-Method of operation: with the emission or absorption of compounds that aid food preservation.

-Examples: oxygen scavengers, carbon dioxide emitters/absorbers, moisture absorbers, ethylene absorbers, ethanol emitters.

INTELLIGENT PACKAGING

-Main function: to monitor the quality of products.

-Method of operation: integration of change-sensitive devices.

-Examples: time-temperature indicator, freshness indicators, gas sensors, biosensors.

Fig. 2. Comparison between active and intelligent packaging.

group is the most extensive in the literature of food packaging materials based on chitosan.

Time-temperature indicators are used to monitor a food's condition in real-time, indicating the overall influence of temperature on food product quality. Chitosan-gold nanoparticles composite can indicate the frozen state and thermal history of food based on the visual color change that accompanies the agglomeration of gold nanoparticles due to their localized surface plasmon resonance (Wang, Mohan, Guan, Ravishankar, & Gunasekaran, 2018). The color of the Chitosan-gold nanoparticles composite irreversibly becomes more intense from pink to dark grey as function of the temperature (Wang et al., 2018). Maciel, Yoshida, and Franco (2012) also has developed chitosan colorimetric temperature indicator for temperatures between 40 and 70 °C, using heat-sensitive anthocyanin with irreversible visual color changes. Laccase was immobilized on electrospun chitosan fiber to prepare time-temperature indicator for food quality monitoring. Laccases, mainly catalyzing the oxidation of phenolic compounds such as aminophenols, polyphenols, and phenols, can oxidize guaiacol with visual and color changes from transparent to deep brown or deep purple-brown (Jhuang et al., 2020).

Materials that monitor changes in the pH of food through color change are capable to detect the microbial growth and oxidative degradation of food due to the physical-chemical alterations of the food (Singh, Nwabor, et al., 2021). These materials are used mainly as indicators of pH and freshness of food, although there are also examples of monitoring CO_2 production by establishing a relationship between the pH-CO_2 produced.

Anthocyanins are a major part of phenolic compounds and the most important subclass of flavonoids. They are pigments that show color changes with pH changes (Li et al., 2017; Zhang et al., 2019). Anthocyanins also have powerful antioxidant properties, and it is common to find studies of active and intelligent materials evaluation with both characteristics of anthocyanins. For instance, curcumin loaded chitosan and polyethylene oxide nanofiber film has been developed as freshness indicator of chicken breast package at 4 °C. The color of nanofiber film changed from bright yellow to reddish color which provided an opportunity to detect color changes by even the naked eyes of the untrained consumer (Yildiz, Sumnu, & Kahyaoglu, 2021).

The CO_2 was monitored during the respiration of fruits and vegetables, as well as microorganisms on their surface by adding indicator of bromocresol blue and methyl red to chitosan intelligent label. The color change of the films at different pH values showed a good linear relationship with CO_2 concentration (Wan et al., 2021).

Also based on colorimetric reactions, the addition of quercetin to chitosan films makes it possible to intelligently detect aluminum (Al^{3+}) in food (Bai et al., 2019), since quercetin can form bonds with Al^{3+} producing a colored complex.

On the other hand, chitosan presents potential application for the development of biosensors thanks to its specific sorption properties. The applications of Chitosan-based biosensors in intelligent food packaging are being investigated, although it is still necessary to delve into this topic. Sophisticated humidity and temperature sensors have been developed from chitosan and $CuMn_2O_4$-spinel nanopowder based on impedance change. With rising temperature, the impedance of the sensor decreases and can be attributed to charge carriers generation under the effect of temperature (Chani, Karimov, Khan, Fatima, & Asiri, 2019). Other humidity sensors are based on chitosan-zinc oxide and single-walled carbon nanotube. In this case, the sensing mechanism is attributed to the swelling effect of chitosan surrounding the nanotubes that changes the hopping conduction path between nanotubes (Dai, Feng, Li, Zhang, & Li, 2019).

A quartz crystal microbalance electrode coated with chitosan film has evaluated as a sensor for volatile organic compounds detection (Ayad, Salahuddin, & Minisy, 2014). The coating of the quartz crystal microbalance with appropriate sensing material as chitosan-based allowed to observe the responds to specific analytes in the form of frequency shift, which results from the mass increase caused by sorption of analytes onto the sensing material (Zhang, Hu, Fan, & Li, 2017). Qi, Xu, Zhang, Fei, and Wang (2020) have developed humidity sensors based on quartz crystal microbalance coated with chitosan-multiwalled carbon nanotubes. The optimized sensor possesses high response sensitivity, negligible humidity hysteresis, quick response and recovery time, and remarkable reversibility, repeatability, long-term stability and selectivity. Graphene oxide/chitosan nanocomposite coated quartz crystal microbalance sensor for detection of amine vapors was also studied. At room temperature, the sensor exhibited high sensitivity to aliphatic amines including methylamine, dimethylamine and trimethylamine with detection limits below 3 mg/L (Zhang, Hu, et al., 2017).

Enzymatic and electrochemical biosensors have been developed to detect specific molecules. A cholesterol biosensor was developed by the immobilization of cholesterol oxidase onto the chitosan nanofibers and gold nanoparticles composite network (Gomathi et al., 2011). Lactose biosensor has been obtained from chitosan composites containing Co-hemin metal organic frameworks and cellobiose dehydrogenase (Choi et al., 2020).

The development of active and intelligent chitosan-based materials requires the addition of specific agents that aims to improve certain functional properties. However, the incorporation of new components in the chitosan structure may affect other physicochemical properties such as mechanical and water vapor permeability (WVP) properties, giving modified polymer matrix with respect to pure chitosan films. The new physicochemical properties of the resulting matrix will determine the suitability of each film for each application or the need for new reformulations to modify or reinforce the polymer structure.

In the following sections, it will be discussed how additives added to the chitosan matrix to develop active or intelligent packaging affect the physicochemical (mechanical and water permeability) and functional properties of the polymer. The following sections will focus exclusively on the materials studied in film form.

3. Mechanical properties of active and intelligent chitosan-based films

Measuring mechanical properties allows predicting the behavior of the material under different food processing and handling conditions (Cazón, Velazquez, Ramírez, & Vázquez, 2017). The most widely mechanical properties analyzed and reported in the literature are tensile strength (TS) and percentage of elongation at break (%E). The maximum tension that the film can withstand before it breaks is called tensile strength. On the other hand, the greatest elongation of the film before rupture is expressed as a percentage of elongation at break (Cazón et al., 2017).

According to studies carried out by several authors, the mechanical properties of chitosan films can be modified by multiple factors. The characteristics of the chitosan (degree of deacetylation, molecular weight), the film production method, the chitosan concentration, the storage time, the conditions at the time of the measurements and throughout the test, and the chitosan solvent employed are factors that affect the mechanical properties (Cazón & Vázquez, 2019). Besides, it must be taken into account that the mechanical strength of a film depends on the composition of the composite polymer, their intermolecular forces, the crystallinity degree and the microstructure of the film network (Pastor, Sánchez-González, Chiralt, Cháfer, & González-Martínez, 2013). Hence, the active agents used to enrich the chitosan-based matrix may affect the mechanical properties of the resulted composite. For comparative purposes, Table 1 shows the tensile strength and elongation at break of different chitosan-based films combined with antioxidant agents.

Research reflects that the essential oils used have showed highly variable effect on the mechanical properties of chitosan-based films. The essential oil effect depends on their own composition, the final concentration reached on the chitosan solution and the specific interactions that occur within the polymer matrix. Generally, studies have showed that the incompatibility between the hydrophobic essential oils and chitosan gives a disrupted microstructure. The structural arrangements of the lipid phase in the chitosan network that results in a discontinuous and porous matrix promotes uneven physical interactions. Consequently, the enriched chitosan-films possess lower mechanical resistance properties, such lower TS and %E values than pure chitosan samples (Moalla et al., 2021; Wang et al., 2011). This effect has been observed by the addition of essential oils such as *Perilla frustescens* (Zhang et al., 2018), *Artemisia campestris* (Moalla et al., 2021), *Thymus vulgaris* (Altiok, Altiok, & Tihminlioglu, 2010), *Zataria multifora* Boiss (Moradi et al., 2012), tea tree (Sánchez-González, González-Martínez, et al., 2010), bergamot (Sánchez-González, González-Martínez, et al., 2010), citronella and cedarwood (Shen & Kamdem, 2015), basil (Amor et al., 2021), caraway (Hromiš et al., 2015), cinnamon (Perdones et al., 2014; Wang et al., 2011; Zhang et al., 2019), clove (Wang et al., 2011), ginger (*Pimpinella anisum* L.) (Mahdavi, Hosseini, & Sharifan, 2018), *Artemisia campestris* (Moalla et al., 2021) and other lipophilic compound as α-tocopherol (Martins, Cerqueira, & Vicente, 2012).

However, the new formed interactions depend on the composition of each oil and the presence of other compounds as emulsifying agents. For example, subsequent studies of chitosan-tree tea essential oils (Cazón, Antoniewska, Rutkowska, & Vázquez, 2021) with soy lecithin as emulsifying agents have showed a plasticizing effect of the tea tree due to the homogeneous dispersion of the oil within the chitosan matrix. The resulted active samples have showed higher %E values and lower TS than neat chitosan films. Wang, Zhang, Yang, and He (2021) has observed that the addition of small amounts of clove essential oil (*Eugenia caryophyllata*) (less than 1% w/w) significantly increases the %E (56.7–78.6%) values with slightly variation on the TS (6.7–6.3 N/mm^2) values by increasing the essential oil concentration. In this case, the composition of the essential oil, mainly eugenol and acetyl eugenol which are considered weak acids, easily promote the formation of new stronger interactions. The new polymer-essential oil interactions allow greater deformation of the structure without weakening it (Wang et al., 2021). Similar results were obtained by adding *Eucalyptus globulus* essential oil (Azadbakht, Maghsoudlou, Khomiri, & Kashiri, 2018).

However, when the interaction polymer-essential oil are stronger, manifesting a cross-linking effect, the resulted composite shows higher TS values and lower %E values by increasing the essential oil, as it was observed by the addition of *Thymus capitatus* essential oil (Grande-Tovar et al., 2018), apricot (*Prunus armeniaca*), kernel essential oil (Priyadarshi, Sauraj, Kumar, & Negi, 2018), cinnamon oil (Ojagh, Rezaei, Razavi, & Hosseini, 2010) or the combination among lemon, thyme or cinnamon essential oils (Peng & Li, 2014).

A less common result was obtained by Mahdavi et al. (2018), where the addition of anise (*Pimpinella anisum* L.) essential oil produced an increase in both TS and %E. The authors justify this mechanical behavior due to the hydrophilicity and high molecular weight of the phenolic compounds of anise essential oil. These properties reduce the softening in the film and increase the TS values (Mahdavi et al., 2018). Similar effect has been observed in chitosan films enriched with lavender essential oil (Zhang, Qin, Fan, Zhao, & Cheng, 2013) and rosemary essential oil (Abdollahi, Rezaei, & Farzi, 2012).

The addition of polyphenol-rich extracts to chitosan-based films, such as green tea extracts, black tea extracts (Peng, Wu, & Li, 2013), apple extracts (Riaz et al., 2018; Sun et al., 2017), banana peel extract (Zhang, Liu, Sun, Wang, & Li, 2020), Chinese chive (*Allium tuberosum*) root extract (Riaz et al., 2020), *Lycium barbarum* fruit extract (Wang et al., 2015), honeysuckle flower extract (Wang, Wang, Tong, & Zhou, 2017), leaf *Pistacia terebinthus* extract (Kaya et al., 2018), syringic acid (Yang et al., 2019), pomegranate peel extract, carvacrol (Yuan, Lv, Yang, Chen, & Sun, 2015) protocatechuic acid (J. Liu et al., 2017), purple fleshed sweet potato extract (Yang et al., 2019) resulted in a general trend of decreasing TS and %E values. The presence of polyphenols into chitosan film may interrupt the ordered crystalline structure formation, hindering the polymer–polymer chain interactions and giving weak intermolecular hydrogen bonds. The new interactions chitosan-polyphenol result in the decreased mechanical properties of the polymer structure (Sun et al., 2017).

Other components as purple corn extracts (Qin, Liu, Yuan, Yong, & Liu, 2019), mango leaf extract (Rambabu, Bharath, Banat, Show, & Cocoletzi, 2019), blackberry and blueberry pomace extract (Kurek et al., 2018), pomegranate peel extract (Pirsa, Karimi Sani, Pirouzifard, & Erfani, 2020) or black chokeberry (*Aronia melanocarpa*) pomace extract (Halász & Csóka, 2018) produced the decrease of %E and the increase of TS. These results can be supported by the idea that several compounds with high molecular weight (such as polyphenols, monoterpenes, etc.) present in these extracts can generate bonds in the film structure, which produce a softening of the film but increase its TS.

Nevertheless, the reinforcement of the mechanical properties (higher TS and %E values) has been observed by the addition of polyphenols extract with higher content in flavonoids, such as purple and black eggplant extracts (Yang et al., 2019), black plum peel extract (Zhang , et al., 2019), black soybean seed coat extract (Wang et al., 2019), butterfly pudding extract (Yan, Cui, Qin, Li, & Yuan, 2021), green tea extract (Siripatrawan & Harte, 2010), spirulina alga extract (Balti et al., 2017) and propolis extract (Siripatrawan & Vitchayakitti, 2016). The

Table 1
Mechanical properties (tensile strength, TS and percentage of elongation at break, %E) of active and intelligent chitosan (CH) based films combined with active compounds as essentials oils (EO) and rich polyphenols extracts. RH is relative humidity and T temperature of the previous test storage conditions.

CH-based films	CH (%)	TS (N/mm^2)	%E (%)	T (°C)	RH (%)	References
A) Active						
***Artemisia campestris* EO**	2	2.43–2.19	89.71–65.20	NR	NR	Moalla et al. (2021)
***Thymus vulgaris* EO**	0.5	38.22–32.94	3.6–1.8	NR	NR	Altiok et al. (2010)
Clove (*Eugenia caryophyllata*) EO	1	6.7–6.3	56.7–78.6	NR	NR	Wang et al. (2021)
Lavander EO	1.5	17.54–31.12	17.18–17.83	NR	NR	Zhang et al. (2013)
Cinnamon EO	2	13.35–29.3	16.57–3.58	25	51	Ojagh et al. (2010)
Cinnamon EO	2	56.36–43.11	45.05–28.05	NR	NR	Zhang, Zou, et al. (2019)
***Zataria multiflora* Boiss EO**	2	24–3	29–10	NR	NR	(Moradi et al., 2012)
Tea tree EO	1	75–54	20–8	20	54.4	Sánchez-González, González-Martínez, et al. (2010)
Bergamot EO	1	65–22	7–1.7	20	54.4	Sánchez-González, González-Martínez, et al. (2010)
***Thymus capitatus* EO**	2	2.25–19.48	20.46–14.69	NR	NR	Grande-Tovar, Chaves-Lopez, Serio, Rossi, & Paparella (2018)
Apricot (*Prunus armeniaca*) kernel EO	2	13.92–19.36	11.03–3.76	NR	NR	Priyadarshi, Sauraj, Kumar, Deeba, et al. (2018)
Citronella EO	1	36.54–22.29	25.8–5.07	22	30	Shen and Kamdem (2015)
Cedarwood EO	1	33–17.12	14.5–8.25	22	30	Shen and Kamdem (2015)
***Eucalyptus globulus* EO**	1.5	34.5–26.6	25.24–35.74	NR	NR	Azadbakht et al. (2018)
Basil EO[b]	3	13–10.5	23–22	NR	NR	Amor et al. (2021)
Turmeric EO	2	40.18–32.92	14.20–9.64	NR	NR	Li et al. (2019)
Caraway EO	1	44.47–2.04	31.53–5.55	NR	NR	Hromiš et al. (2015)
***Perilla frutescens* (L.) Britt. EO**	2	11.76–12.47	13.26–9.36	23	53	Zhang et al. (2018)
Lemongrass EO	1.5	14.61–7.93	37.47–65.34	NR	NR	Han Lyn & Nur Hanani (2020)
Ginger (*Zingiber officinale*) EO	1	31.9–30.9	18.18–18.20	NR	NR	Remya et al. (2016)
Anise (*Pimpinella anisum* L.) EO	2	15.85–21.38	7.81–12.32	NR	NR	Mahdavi et al. (2018)
Green tea extracts	2	27–29	5–1	25	53	Peng et al. (2013)
Black tea extracts	2	25–27	6–2	25	53	Peng et al. (2013)
Apple peel polyphenols extracts	2	24–16	25–14	25	NR	Riaz et al. (2018)
Banana peels extract	2	35.05–21.56	33.05–10.85	NR	NR	Zhang et al. (2020)
Anthocyanin black plum peel extract	2	14.93–19.72	46.87–55.07	NR	NR	Zhang, Zou, et al. (2019)
Chinese chive (*Allium tuberosum*) root extract	2	25–15	51–35	NR	NR	Riaz et al. (2020)
Purple eggplant extracts	2	29.42–39.78	30.97–57.98	NR	NR	Yong et al. (2019)
Black eggplant extracts	2	33.91–24.74	60.26–48.96	NR	NR	Yong et al. (2019)
Purple rice extracts	2	26.48–21.46	42.26–57.06	NR	NR	Yong et al. (2019)
Black rice extracts	2	24.39–18.76	51.13–61.16	NR	NR	Yong et al. (2019)
Propolis extract	2	16–22	17–13	NR	NR	Siripatrawan and Vitchayakitti (2016)
Purple corn extract	2	31.68–34.69	49.55–31.69	NR	NR	Qin et al. (2019)
Mango leaf extract	3	19–23	25–15	NR	NR	Rambabu et al. (2019)
Oak (Quercus robur) extract[a]	1.5	14.2–12.5	10.5–14	NR	NR	Bajić et al. (2019)
Hop (Humulus lupulus) extract[a]	1.5	14.2–10	10.5–23	NR	NR	Bajić et al. (2019)
Brown algae (Laminaria hyperborean) extract[a]	1.5	14.2–5.5	10.5–30	NR	NR	Bajić et al. (2019)
***Lycium barbarum* fruit extract**	1	19.5–10	13–7	NR	NR	Wang et al. (2015)
Honeysuckle flower extract	4	8.42–4.42	111.06–53.29	NR	NR	Wang, Wang, Tong, and Zhou (2017)
Spirulina alga extract	2	22.45–29.65	27.45–34.29	NR	NR	Balti et al. (2017)
Stem *Pistacia terebinthus* extract	1	26.5–4	4–72	25	NR	Kaya et al. (2018)
Leaf *Pistacia terebinthus* extract	1	26.5–15	4–2	25	NR	Kaya et al. (2018)
Seed *Pistacia terebinthus* extract	1	26.5–3	4–80	25	NR	Kaya et al. (2018)
Protocatechuic acid	2	49.59–26.71	35.10–7.86	NR	NR	Liu et al. (2017)
Syringic acid	2	22–12	15–8	NR	NR	Yang et al. (2019)
Phenolic acid	2	31.53–10.75	27.42–14.55	NR	NR	Liu et al. (2021)
Ferulic acid	2	31.53–25.93	27.42–23.31	NR	NR	Liu et al. (2021)
Gallic acid	2	31.53–28.57	27.42–24.82	NR	NR	Liu et al. (2021)
Vanillic acid	2	31.53–15.94	27.42–16.82	NR	NR	Liu et al. (2021)
Salicylic acid	2	31.53–19.32	27.42–18.92	NR	NR	Liu et al. (2021)
Gallic acid and sodium carbonate (O_2)	1	19.50–8.45	29.26–14.98	NR	NR	Singh, Nwabor, et al. (2021)
Nanosized TiO_2	2	12.3–13.8	51–49	25	50	(Siripatrawan & Kaewklin, 2018)
TiO_2	2	14.93–23.98	46.87–49.55	NR	NR	Zhang, Zou, et al. (2019)
B) Intelligent						
Black soybean seed coat extract	2	20.64–23.24	67.71–73.88	NR	NR	Wang et al. (2019)
Purple corn extract	2	31.68–34.69	49.55–31.69	NR	NR	Qin et al. (2019)
Purple rice extracts	2	26.48–21.46	42.26–57.06	NR	NR	Yong et al. (2019)
Black rice extracts	2	24.39–18.76	51.13–61.16	NR	NR	Yong et al. (2019)
Blueberry pomace extract	2	21.97–19.35	50.16–37.24	25	50	Kurek et al. (2018)
Blackberry pomace extract	2	26.22–32.35	49.20–53.30	25	50	Kurek et al. (2018)
Purple-fleshed sweet potato extract	2	17.73–13.67	26.22–9.47	NR	NR	Yong et al. (2019)
Pomegranate peel extract	1.5	30–190	3.3–2.5	25	55	Pirsa et al. (2020)
Black chokeberry (*Aronia melanocarpa*) pomace extract	1	60–82	5–3	23	50	Halász and Csóka (2018)
Purple eggplant extracts	2	29.42–39.78	30.97–57.98	NR	NR	Yong et al. (2019)
Black eggplant extracts	2	33.91–24.74	60.26–48.96	NR	NR	Yong et al. (2019)
Butterfly pudding extract	2	17.7–31.9	24.5–40.5	NR	NR	Yan et al. (2021)
Banana peels extract	2	35.05–21.56	33.05–10.85	NR	NR	Zhang et al. (2020)
Purple corn extract	2	31.68–34.69	49.55–31.69	NR	NR	Qin et al. (2019)

(*continued on next page*)

Table 1 (*continued*)

CH-based films	CH (%)	TS (N/mm^2)	%E (%)	T (°C)	RH (%)	References
Grapes anthocyanins	2	49.98–47.07	3.89–2.05	25	75	Yoshida, Maciel, Mendonça, & Franco (2014)
Purple tomato anthocyanins	1	12–10	45–55	NR	NR	Li et al. (2021)
Black rice bran anthocyanins	1	33.60–20.32	18.82–27.38	25	50	(Wu et al., 2019)
TiO_2 nanoparticles	2	14.93–23.98	46.87–49.55	NR	NR	Zhang, Zou, et al. (2019)

NR = Data not reported.

[a] Lactic acid used as solvent.

[b] Hydrochloric acid used as solvent.

superior mechanical properties observed are attributed to the hydrogen bonds formed between the hydroxyl/amino groups of the chitosan and the polyphenols contained in the extract.

On the other hand, other phenolic compounds as steam and seed *Pistacia terebinthus* extracts, ellagic acid (Vilela et al., 2017) and the pH-sensitives purple and black rice extracts (Yong et al., 2019) and purple tomato anthocyanins (Li, Wu, Wang, & Li, 2021) act as plasticizers, promoting the increase of the %E and the decrease of TS (Kaya et al., 2018).

Singh, Nwabor, et al. (2021) added gallic acid and sodium carbonate to the chitosan film to develop oxygen scavenger material. Gallic acid is antioxidant but also absorbs a high amount of oxygen when exposing to the alkaline environment. The result is a decrease in both mechanical parameters as the concentration of the compounds added increase. This may be due to the large amount of sodium carbonate cracking the internal matrix of the chitosan film (Singh, Nwabor, et al., 2021) since chitosan/gallic acid/glycerol films showed a mechanical reinforcement with higher TS values (Zarandona, Puertas, Dueñas, Guerrero, & de la Caba, 2020).

The incorporation of TiO_2 nanoparticles in chitosan films to obtain ethylene absorber matrix result in TS reinforcement and %E reduction (Siripatrawan & Kaewklin, 2018; Xin Zhang, Zou, et al., 2019). The increase in TS was related to the fact that TiO_2 probably strengthens the network structure of the film. However, the decrease in %E values could have been caused by the hydrogen bonding of Ti with the amino and hydroxyl functional groups of chitosan, resulting in a less flexible film (Siripatrawan & Kaewklin, 2018; Xin Zhang, Zou, et al., 2019). Similar trend has been observed by the addition of other nanoparticles, such silver nanoparticles as antimicrobial agent (Qin et al., 2019).

In the literature, there is a significant lack of studies evaluating the effect of ethanol or CO_2 emitting/absorbing, moisture absorbing and flavor releasing/absorbing agents on the mechanical properties of chitosan-based films, being necessary more in-depth research in this area.

4. Barrier properties of active and intelligent chitosan-based films

Barrier properties play an important role in the preservation of food products, since one of the main functions of the packaging films is separating foods from surrounding atmosphere to retard food deterioration. Among these properties, the most studied is water vapor permeability (WVP). The WVP indicates the amount of water that permeates per unit of area and time (g/m·s·Pa). Generally, the main goal is to develop films that keep the WVP values as low as possible to avoid moisture transfer between food and the surrounding atmosphere (Riaz et al., 2018). Most of the WVP results obtained on biodegradable films available in the literature have been calculated gravimetrically following the American Society for Testing and Materials Standard Test Method E96/E96M (ASTM), known as the “cup method” (Cazón et al., 2017).

Considering the hydrophilic nature of chitosan films, they consequently have a low water barrier property (Aider, 2010; Park, Daeschel, & Zhao, 2004). The WVP varies based on a variety of factors such as molecular weight, degree of deacetylation and chitosan concentration. In addition, external factor such as the measurement method used, the measurement conditions, storage time and conditions affect the results as well (Cazón & Vázquez, 2020).

For comparative purposes, Table 2 shows WVP values of chitosan-based films enriched with several active compounds.

Lipidic compounds as the essential oils have been added to chitosan matrix with the aim of improving their barrier properties. Nevertheless, the addition of essential oil does not produce a general downward trend. The effect on the barrier properties is strongly affected by the composition of the lipid component used as addictive and the interactions that take place into the structure. For instance, the WVP values of chitosan/essential oils showed a decrease by the addition of *Perilla frustescens* (L.) Britt. essential oil (Zhang et al., 2018), *Thymus capitatus* essential oil (David Grande-Tovar et al., 2018), *Zataria multifora* Boiss essential oil (Moradi et al., 2012), bergamot essential oil (Sánchez-González, Cháfer, et al., 2010), apricot (*Prunus armeniaca*) kernel (Priyadarshi, Sauraj, Kumar, Deeba, et al., 2018), anise (*Pimpinella anisum* L.) essential oil (Mahdavi et al., 2018), tea tree essential oil (Sánchez-González, González-Martínez et al., 2010), clove (*Eugenia caryophyllata*) (Wang et al., 2021), citronella and cedarwood oil (Shen & Kamdem, 2015), or lemon and thyme (Peng & Li, 2014). It is believed that this effect may be due to covalent interactions between the matrix and the constituent components of the essential oil (mainly oxygenated terpenes) which causes a cross-linking reaction (Shen & Kamdem, 2015).

However, the WVP values increase by the addition of *Eucalyptus globulus* essential oil (Azadbakht et al., 2018), rosemary essential oil (Abdollahi et al., 2012), thyme oil (Altiok et al., 2010), cinnamon essential oil (Ojagh et al., 2010.; Perdones et al., 2014; Wang et al., 2011), turmeric essential oil (Li et al., 2019), or α-tocopherol (Martins et al., 2012). Despite the hydrophobicity nature of these oils, the presence of essential oil on the chitosan network create a series of micropores on the hydrophilic surface of the film, through which water passed (Altiok et al., 2010; Li et al., 2019; Wang et al., 2011).

In the case of the addition of phenolic compounds, a predominant trend can be observed in the effect of these compounds on the permeability properties of chitosan. Generally, the presence of phenolic compounds leads to a decrease in WVP values and less permeable film is achieved. This occurs by the addition of propolis extract (Siripatrawan & Vitchayakitti, 2016), apple extracts (Riaz et al., 2018; Sun et al., 2017), banana peel extract (Zhang, et al., 2020), pine needles (*Cedrus deodara*) extract (Kadam, Singh, & Gaikwad, 2021), Chinese chive root extract (Riaz et al., 2020), curcumin extract (Rachtanapun et al., 2021), green and black tea extract (Peng et al., 2013; Siripatrawan & Harte, 2010), spirulina extract (Balti et al., 2017), *Lycium barbarum* fruit extract (Wang et al., 2015), black plum peel extract(Xin Zhang, Zou, et al., 2019), purple corn extract (Qin et al., 2019), blueberry and blackberry pomace extract (Kurek et al., 2018), propolis extract (Siripatrawan & Vitchayakitti, 2016) black soybean seed coat extract (Wang et al., 2019), mango leaf extract (Rambabu et al., 2019), syringic acid (Yang et al., 2019), protocatechuic acid (Liu et al., 2017), purple and black rice extracts (Wu et al., 2019; Yong et al., 2019), or pomegranate peel and carvacol extract (Pirsa et al., 2020; Yuan et al., 2015). The hydrogen and covalent interactions between the chitosan network and the extract’s

Table 2
Water barrier properties (WVP) of active and intelligent chitosan (CH) based films with active compounds as essentials oils (EO) and rich polyphenols extracts. RH is relative humidity and T temperature of the previous test storage conditions.

CH-based films	CH (%)	WVP ($\cdot 10^{-10}$ g/m·s·Pa)	T (°C)	RH (%)	References
A) Active					
Thymus capitatus EO	2	1.11–0.88	NR	42	(Grande-Tovar et al., 2018)
Lemon EO	2	93.6–88.6	25	0–75	Peng and Li (2014)
Thyme EO	2	93.6–91.4	25	0–75	Peng and Li (2014)
Cinnamon EO	2	93.6–88.9	25	0–75	Peng and Li (2014)
Turmeric EO	2	4.76–5.07	25	0–100	Li et al. (2019)
Clove bud EO	2	0.15–0.25	37.8	10–100	Wang et al. (2011)
Cinnamon EO	2	0.15–0.32	37.8	10–100	Wang et al. (2011)
Rosemary EO	2	0.80–0.68	20	0	Abdollahi et al. (2012)
Perilla frustescens (L.) Britt. EO	2	61.9–63.8	38	0	Zhang et al. (2018)
Citric acid	2	327–232	25	75	Priyadarshi, Sauraj, Kumar, Deeba, et al. (2018)
Ellagic acid	1	33.56–43.98	25	75	Vilela et al. (2017)
Phenolic acid	2	0.17–0.168	27	NR	Liu et al. (2021)
Ferulic acid	2	0.17–0.158	27	NR	Liu et al. (2021)
Gallic acid	2	0.17–0.158	27	NR	Liu et al. (2021)
Vanillic acid	2	0.17–0.161	27	NR	Liu et al. (2021)
Salicylic acid	2	0.17–0.165	27	NR	Liu et al. (2021)
Olive oil emulsion	2	1.17–1.03	25	67	Pereda, Amica, & Marcovich (2012)
Propolis extract	2	0.57–0.55	25	75	Siripatrawan and Vitchayakitti (2016)
Chinese chive root extract	2	1.32–0.78	NR	75	Riaz et al. (2020)
Blueberry pomace extract	2	1.99–1.95	25	70	Kurek et al. (2018)
Blackberry pomace extract	2	2.24–1.97	25	70	Kurek et al. (2018)
Pomegranate peel extract	2	2.77–2.5	25	50	Yuan et al. (2015)
Carvarol extract	2	2.77–1.11	25	50	Yuan et al. (2015)
Curcumin extract	1.5	22.39–21.18	25	52	Rachtanapun et al. (2021)
Green tea extract	2	260–100.8	25	75	Siripatrawan and Harte (2010)
Spirulina extract	2	0.54–0.43	25	75	Balti et al. (2017)
Lycium barbarum fruit extract	1	50.9–40.5	25	75	Wang et al. (2015)
Gallic acid	1	8.63–6.41	23	55	Singh, Nwabor, et al. (2021)
B) Intelligent					
Black rice bran anthocyanins	1	1.67–2.05	25	75	(Wu et al., 2019)
Purple eggplant anthocyanins	2	0.96–1.24	20	100	Yong et al. (2019)
Black eggplant anthocyanins	2	1.07–1.22	20	100	Yong et al. (2019)
Purple rice extracts	2	49.8–61.1	NR	100	Yong et al. (2019b)
Black rice extracts	2	50.4–61.7	NR	100	Yong et al. (2019b)
Purple-fleshed sweet potato extract	2	151.17–170.1	20	100	Yong et al. (2019)
	2	144.6–130	25	NR	Qin et al. (2019)
Purple corn extract					
TiO_2 nanoparticles	2	6.65–5.82	20	NR	Zhang, Zou, et al. (2019)

NR = Data not reported.

own phenolic compounds reduce the availability of hydrophilic groups, decreasing the affinity of the chitosan matrix to water molecules (Balti et al., 2017; Kurek et al., 2018; Peng et al., 2013).

Slightly WVP values variations have been observed by the addition of anthocyanins from purple and black eggplant extracts (Yong et al., 2019) or a WVP increase by the addition of ellagic acid (Vilela et al., 2017).

Regarding ethylene scavenger, Siripatrawan and Kaewklin (2018) and Xin Zhang, Zou, et al. (2019) have developed chitosan film to which TiO_2 nanocomposites were added. The authors have observed that the water insoluble TiO_2 can block water vapor micro-paths in the microstructure of the film, promoting the decrease in WVP values (Xin Zhang, Zou, et al., 2019). This behavior has also been repeated by the addition of another particles such as zeolite as ethylene adsorption (do Nascimento Sousa et al., 2020) and silver nanoparticles as antimicrobial agents (Qin et al., 2019).

The same trend was found for the O_2 scavenger. Gallic acid/sodium carbonate reduce the WVP values of the chitosan samples from $8.63 \cdot 10^{-10}$ to $6.41 \cdot 10^{-10}$ g/m·s·Pa (Singh, Nwabor, et al., 2021). This can be explained by the fact as the concentration of gallic acid in the chitosan amine increases, the benzene ring group of the acid become more present. This hinders the intermolecular and intramolecular hydrogen bonding of chitosan (Singh, Nwabor, et al., 2021), making the membrane less permeable.

Regarding the other type of active chitosan films with properties like ethanol emitting, CO_2 emitting/absorbing, moisture absorbing or flavor releasing/absorbing capabilities, further studies are necessary to delve into the effect of these actives' agents on chitosan permeability properties.

5. Functional properties of active and intelligent chitosan-based films

5.1. Antioxidant activity

The ability of an antioxidant to block or delay the oxidation of other molecules is called antioxidant activity (Zhang, Zhang, Chen, Ma, & Xia, 2021). The main values to be analyzed for the antioxidant capacity of the film are DPPH• radical scavenging, ABTS•+ radical scavenging and total phenolic content (TPC).

Pure chitosan-based film has a very low radical scavenging activity, which can be explained by the fact that chitosan does not have any hydrogen atoms that can be easily supplied to serve as a good antioxidant (Schreiber, Bozell, Hayes, & Zivanovic, 2013). Another fact that is related to the poor antioxidant activity of chitosan films is the weak free radical scavenging ability of free amino groups at the C-2 position of chitosan chains (Genskowsky et al., 2015). To improve the antioxidant properties of chitosan, some natural active compounds such as essential oils and polyphenols have been added by physical mixing into the chitosan matrix to produce composite films. For comparative purposes, Table 3 shows the antioxidant property values of various enriched chitosan -based films.

It is worth highlighting the components that have shown the greatest antioxidant capacity with promising applications in the food industry. The addition of 0.5–2% (w/v) of green or black tea extracts have been

Table 3
Functional properties of antioxidant active and intelligent chitosan-based films. TPC is total phenolic content.

Antioxidant agent added	TPC	DPPH	ABTS	References
	mg GA/g	(%)	(%)	
A) Active				
Carvacrol EO	4–70	NR	NR	Yuan et al. (2015)
α-tocopherol	NR	97.42–97.71	NR	Martins et al. (2012)
Gallic acid	NR	51.4–96.4	NR	Zarandona et al. (2020)
***Zataria multifora* Boiss EO**	<8	20–35	NR	(Moradi et al., 2012)
Apple polyphenols	NR	70–90	70–90	(Riaz et al., 2018; Sun et al., 2017)
***Zataria multiflora* Boiss EO**	4–6	12–35	NR	Moradi et al. (2011)
Honeysuckle flower extract	NR	5–9	NR	(Wang, Wang, Tong, & Zhou, 2017b)
***Eucalyptus globulus* EO**	10–40	25–45	NR	Hafsa et al. (2016)
Apricot (*Prunus armeniaca*) kernel EO	NR	25–35	NR	Priyadarshi, Sauraj, Kumar, Deeba, et al. (2018)
Caraway EO	NR	80–85	NR	Hromiš et al. (2015)
***Artemisia campestris* hydroalcoholic extract**	269.98	96.79	NR	Moalla et al. (2021)
***Artemisia campestris* aqueous extract**	101.19	70	NR	Moalla et al. (2021)
Grape seed extract	4–76	12–38	NR	Moradi et al. (2011)
Banana peel extract	2.8–4.8	52–89	42–96	Zhang et al. (2020)
Pine needles (*Cedrus deodara*) extract	0.9–2.9	10–63	17–48	Kadam et al. (2021)
Green tea extract	6–33	29–51	NR	Siripatrawan and Harte (2010)
Pomegranate peel extract	4–15	NR	NR	Yuan et al. (2015)
***Lycium barbarum* fruit extract**	NR	4–13	NR	Wang et al. (2015)
Chinese chive (*Allium tuberosum*) root extract	NR	20–46	28–55	Riaz et al. (2020)
Thyme spice extract	NR	19–55	NR	Liu et al. (2021)
Clove spice extract	NR	19–57	NR	Liu et al. (2021)
Prickly ash spice extract	NR	19–70	NR	Liu et al. (2021)
Fennel spice extract	NR	19–48	NR	Liu et al. (2021)
Geranium spice extract	NR	19–86	NR	Liu et al. (2021)
Cinnamon spice extract	NR	19–80	NR	Liu et al. (2021)
Pine nut shell extract	NR	11–26	NR	Zhang et al. (2020)
Peanut shell extract	NR	11–33	NR	Zhang et al. (2020)
Jujube leaf extract	NR	11–45	NR	Zhang et al. (2020)
Procyanidin	NR	53–88	55–90	Zhang et al. (2021)
Protocatechuic acid	35–65	70–95	NR	(Liu et al., 2017)
Gallic acid	NR	51.9–96.4	NR	Zarandona et al. (2020)
Mango leaf extract	0.005–0.012	20–23	4–8	Rambabu et al. (2019)
Propolis extract	4–6	30–55	NR	Siripatrawan and Vitchayakitti (2016)
Ascorbate	NR	80–95	NR	Tan et al. (2019)
B) Intelligent				
Purple eggplant extracts	NR	12–44	NR	Yong et al. (2019)
Black eggplant extracts	NR	21–46	NR	Yong et al. (2019)
Purple rice extracts	NR	15–29	NR	(Yong et al., 2019a)
Black rice extracts	NR	24–59	NR	(Yong et al., 2019a)
Blueberry pomace extract	4.17–5.63	NR	NR	Kurek et al. (2018)
Blackberry pomace extract	5.02–6.21	NR	NR	Kurek et al. (2018)
Black soybean seed coat extract	NR	35–52	NR	Wang et al. (2019)
Purple-fleshed sweet potato extract	NR	51–79	NR	Yong et al. (2019)
Black plum peel extract	NR	18–70	NR	Zhang, Zou, et al. (2019)
Purple corn extract	NR	16–43	NR	Qin et al. (2019)
Purple potato extract	NR	21.21–45.32	NR	Li et al. (2019)
Procyanidin	NR	54–89	55–90	Zhang et al. (2021)
TiO_2 nanoparticles	NR	18–19	NR	Zhang, Zou, et al. (2019)
Silver nanoparticles	NR	16–18	NR	Qin et al. (2019)

NR = Data not reported.

enough to obtain chitosan films with DPPH$^{\bullet}$ radical scavenging activity higher than 90% (Peng et al., 2013). Other plant extracts as caraway oil, at low concentrations 1% (v/v) can reach around 80% DPPH$^{\bullet}$ value (Hromiš et al., 2015). The antioxidant capacity of *Artemisia campestris* hydroalcoholic extract and aqueous extract has been evaluated. Results demonstrated that the hydroalcoholic fraction has higher antioxidant capacity (96.79% DPPH$^{\bullet}$) due to the higher total phenolic and flavonoid contents than the aqueous fraction (70% DPPH$^{\bullet}$) (Moalla et al., 2021).

The acid selected for the preparation of the chitosan film-forming solution could affect the antioxidant properties of the added essential oil. Analysis of chitosan - tree tea oil comparing lactic or malic acid as solvent have demonstrated that chitosan films contained malic acid possess 6.5–7.5 higher DPPH$^{\bullet}$ scavenging capacity (61.24% DPPH$^{\bullet}$) than that contained lactic acid (8.31% DPPH$^{\bullet}$) (Cazón et al., 2021). Malic acid has showed a synergic effect as promoter agents for antioxidant activity of the tree tea oil terpens. The lipophilic compound α-tocopherol from edible oils, such as palm oil, sunflower oil, has been one of the components that has shown the highest scavenging activity with DPPH$^{\bullet}$ values of 97% for very small concentrations of compound in the final film (0.1–0.2% w/v) (Martins et al., 2012).

The effect of active chitosan films added with some spices extracts as thyme, clove, prickly ash, fennel, geranium and cinnamon on antioxidant capacity has been studied (Liu, Liu, Gong, Chi, & Ma, 2021). Among them, geranium spice extract showed the highest antioxidant capacity (80% DPPH value), quadrupling the antioxidant capacity of a pure chitosan film.

The high capacity to increase total phenolic content and radical scavenging activity is related to the high presence in the spices of flavonoids and phenols with excellent antioxidant properties (Liu et al., 2021). Other examples of this effect was observed by Zhang, et al.,

(2020) with the addition of plant extracts such as pine nut shell extract, peanut shell extract and jujube leaf extract. The film with the best result was the one with the addition of jujube leaf extract, which was related to the high amount of phenolic compounds and flavonoids in its composition (Zhang et al., 2020).

Fruit extracts have shown to have a high content of polyphenolic agents with powerful antioxidant capacity. For example, ABTS$^{\bullet+}$ and DPPH$^{\bullet}$ values ranged from 70 to 90% for chitosan samples enriched with apple extract at a concentration between 0.25 and 1% have been reported (Riaz et al., 2018; L.; Sun et al., 2017). The chitosan-banana peel extract films have demonstrate a DPPH$^{\bullet}$ values ranged 52–89%, ABTS values ranged 42–96%, TPC values ranged 2.8–4.8 mg GAE/g (Zhang et al., 2020). Protocatechuic acid, which is commonly found in fruits, edible plants and vegetables, allow developing CH films with DPPH$^{\bullet}$ values range between 40 and 100% (Liu et al., 2016).

Phenolic compounds used to obtain pH-sensitive material also possess antioxidant capacity that allow developing antioxidant films as well. Some examples are the active and intelligent chitosan-based material developed by Yong et al. (2019) who has observed that purple and black eggplant extracts in chitosan films possess a DPPH$^{\bullet}$ scavenging activity up to 44–46%. Black and purple rice extracts on chitosan samples have shown similar values (Yong et al., 2019a). In this case, the black rice extracts show a better antioxidant effect than the purple one. This was attributed to the fact that the polyphenols present in the extract are able to scavenge free radicals through the donation of phenolic hydrogen atoms (Yang et al., 2019).

In films with O_2 scavenger capacity, the addition of gallic acid has been studied by Zarandona et al. (2020). Results show an increase in DPPH$^{\bullet}$ radical scavenging capacity (up to 96.4%), concluding that gallic acid can also be considered as an active agent to be taken into account in the development of active films (Zarandona et al., 2020). Chitosan/gallic acid/sodium carbonate films have shown an outstanding improvement of the O_2 scavenging capacity from 1.82 to 19.55 mL O_2/g by increasing the sodium carbonate concentration (Singh, Singh, Kumar, Kirtiraj, et al., 2021).

5.2. Antimicrobial activity

On the other hand, antimicrobial activity of the films is part of the functional properties. The degree of deacetylation, molecular weight, film forming conditions, pH, temperature, and other parameters influence chitosan's antibacterial effectiveness. Previous studies have shown that chitosan can prevent the growth of a wide range of fungi and bacteria, being more effective against Gram-positive bacteria than Gram-negative bacteria (Riaz et al., 2018).

For comparative purposes, Table 4 shows the antimicrobial activity of active chitosan films with some agents added. The formulation of chitosan-based films with essential oil enhanced the antimicrobial activity against Gram-negative bacteria such as *Escherichia coli* (Amor et al., 2021; Sani, Pirsa, & Tağı, 2019; Wang et al., 2011), ***Klebsiella pneumoniae*** (Altiok et al., 2010), *Pseudomonas aeruginosa* (Hafsa et al., 2016), *Shewanella baltica, Shewanella putrefaciens, Serratia* spp. and *Pseudomonas fluorescens* (David Grande-Tovar et al., 2018). Gram-positive bacteria are likewise inhibit by the addition of essential oil, for instance, *Staphylococcus aureus* (Hafsa et al., 2016; Wang et al., 2011), and *Staphylococcus saprophyticus* (Amor et al., 2021).

Molds, fungi, and yeast are also microorganisms that should be avoided. For instance, chitosan films combined with tree tea or bergamot essential oil have been tested against *Penicillium italicum* but the power of inhibition of the composite films decreased throughout the

Table 4
Functional properties of antimicrobial active and intelligent chitosan-based films.

Agent added	Gram – bacteria	Gram + bacteria	Fungi	Yeasts	References
A) Active					
***Eucalyptus globulus* EO**	*E. coli, P.aeuruginosa*	*S.aureus, C.albicans*	NR	NR	Hafsa et al. (2016)
Clove bud EO	*E. coli*	*S.aureus*	*A.oryze, P. digitatum*	NR	Wang et al. (2011)
Cinnamon EO	*E. coli*	*S.aureus*	*A.oryze, P. digitatum*	NR	Wang et al. (2011)
***Thymus capitatus* EO**	*S.baltica, S.putrefaciensSerratia spp, P.fluorescens*	NR	NR	NR	(Grande-Tovar et al., 2018)
Tea tree EO	NR	NR	*P.italicum, Penicillium*	NR	(Sánchez-González, González-Martínez, et al., 2010)
Bassil EO	*E. coli*	*S.saprophyticus*	NR	NR	Amor et al. (2021)
Melissa officinialis EO and Zinc oxide	*E. coli*	NR	NR	NR	Sani et al. (2019)
Turmeric extract	*Salmonella*	*S.aureus*	NR	NR	Kalaycıoğlu et al. (2017)
***Pistacia terebinthus* extract**	*P.microbilis, P.vulgaris, P.aeruginosa, E.coli*	*NR*	NR	NR	Kaya et al. (2018)
Apple peel polyphenols extract	*E.coli,*	*S.aureus, B.cereus*	NR	NR	Riaz et al. (2018)
Green tea extract	*E.coli*	*L.innocua*	NR	NR	Amankwaah et al. (2020)
Pomegranate peel extract and carvacrol	*E. coli*	*S.aureus*	NR	NR	Yuan et al. (2015)
Pullulan	NR	*S.aureus*	NR	NR	Li et al. (2020)
Vanillin	*E. coli*	NR	NR	NR	Stroescu et al. (2015)
TiO_2 nanoparticles	*E. coli*	*S.aureus*	*C.albicans* *A.niger*	NR	Zhang, Xiao, et al. (2017)
TiO_2 nanosized	*E. coli, S.typhimurium, P.aeruginosa*	*S.aureus*	*Aspergillus, Penicillium*	NR	Siripatrawan & Kaewklin (2018)
Gallic acid	*E. coli, S.typhimurium*	*B.subtilis, L.innocua Lactobacillus sakei*	NR	NR	Sun et al. (2014); Zarandona et al., 2020)
N-halamine	*E. coli*	*S.aureus*	NR	NR	Li et al. (2013)
Nanocellulose	*E. coli, S.enteriditis*	*S.aureus*	NR	NR	Dehnad et al. (2014)
B) Intelligent					
Purple corn extract	*E. coli, Salmonella*	*S.aureus, L.monocytogenes*	NR	NR	Qin et al. (2019)
Silver nanoparticles	*E. coli, Salmonella*	*S.aureus, L.monocytogenes*	NR	NR	Qin et al. (2019)
TiO_2 nanosized	*E. coli, S.typhimurium, P.aeruginosa*	*S.aureus*	*Aspergiullus, Penicillium*	NR	(Siripatrawan & Kaewklin, 2018)

NR = Data not reported.

storage time (Sánchez-González, Cháfer, et al., 2010; Sánchez-González, González-Martínez et al., 2010). Inhibition of *Aspergillus niger*, *Aspergillus oryzae, Botrytis cinerea*, *Rhizopus stolonifera* and *Penicillium digitatum* fungi has been observed by the addition of clove bud and cinnamon essential oils to chitosan film (Perdones et al., 2014; Wang et al., 2011; Zhang et al., 2019). The incorporation of turmeric essential oil into chitosan allowed developing films with satisfactory and significant anti-aflatoxigenic activity thanks to the observed antifungal properties against *Aspergillus flavus* (Li et al., 2019).

Yeasts such as *Candida albicans* and *Candida parapsilosis* are also successfully inhibited by the application of *Eucalyptus globulus* essential oil on chitosan matrix (Hafsa et al., 2016). The strong antimicrobial effect of essential oils to some extent due to their content of terpenes, which affect the permeability and other functions of the bacterial membranes. Namely, monoterpenes would case cell death by increasing the quantity of lipidic peroxides such as hydroxyl, alkoxyl and alkoperxyl radicals (Amor et al., 2021).

Regarding polyphenolic extracts, the antimicrobial properties of chitosan films are improved by the addition of green tea extracts (Amankwaah, Li, Lee, & Pascall, 2020), pomegranate peel extract (Yuan et al., 2015), *Spirulina* extract (Balti et al., 2017), apple peel extract (Riaz et al., 2018), black plum peel extract (Xin Zhang, Zou, et al., 2019), propolis extract (Siripatrawan & Vitchayakitti, 2016), purple corn extract (Qin et al., 2019). The chitosan - polyphenolic extract samples have been successfully tested against a wide group of Gram-negative bacteria, such as *E. coli, Salmonella typhimurium*, *Salmonella enterica*, *Proteus mirabilis, Proteus vulgaris,* and *P. aeruginosa* (Balti et al., 2017; Kalaycıoğlu, Torlak, Akın-Evingür, Özen, & Erim, 2017; Kaya et al., 2018; Riaz et al., 2018). Regarding Gram-positive bacteria, the antimicrobial activity is mainly exerted against *S. aureus, Streptococcus mutans, Bacillus subtilis, Bacillus cereus, Bacillus thuringiensis, Listeria monocytogenes, Listeria innocua*, *Lactobacillus sakei* and *Lactobacillus plantarum* (Amankwaah et al., 2020; Balti et al., 2017; Kalaycıoğlu et al., 2017; Kaya et al., 2018; Ojagh et al., 2010; Riaz et al., 2018, 2020; Yuan et al., 2015; Zarandona et al., 2020; Xin Zhang, Zou, et al., 2019).

Polyphenols from apple extract enhance the antimicrobial properties of the chitosan samples against molds (*Colletotrichum fructicola*, *Botryosphaerial dothidea* and *Alternaria tenuissima*) but hardly show inhibitory activity against yeast strains (*Saccharomyces cerevisiae*, baker's yeast and tropical candida) (Sun et al., 2017).

Other agents were added to exert an antimicrobial effect on Gram-negative bacteria as *E. coli, S. typhimurium, P. aeruginosa* and *Salmonella enteriditis*. For instance, the addition of vanillin (Stroescu et al., 2015), TiO_2 nanoparticles (Zhang, Xiao, et al., 2017), silver nanoparticles (Qin et al., 2019) and N-halamine (Li, Hu, Ren, Worley, & Huang, 2013) caused an inhibition of these type of bacteria. For Gram-positive bacteria, the inhibition of *S. aureus* was achieved with the addition of pullulan (Li, Yi, Yu, Wang, & Wang, 2020), TiO_2 nanoparticles (Xiaodong Zhang, Xiao, et al., 2017), N-halamine (R. Li et al., 2013) and chitosan-nanocellulose biocomposites (Dehnad, Mirzaei, Emam-Djomeh, Jafari, & Dadashi, 2014).

6. Applications of films based on chitosan

6.1. Applications of active films

The application of active CH-based films has been studied in multiple foodstuffs. For comparative purposes, Table 5 shows different applications of chitosan active films with the addition of different active components.

In the literature, the most studied chitosan-based active films are those with antioxidant capacity. The antioxidant chitosan-based films enriched with olive pomace (de Moraes Crizel et al., 2018) and mango leaf extract (Rambabu et al., 2019) have successfully tested as a high protective film against the oxidation of nuts for 31 and 28 days, respectively. The authors verified the powerful antioxidant effect of the

Table 5
Applications of active and intelligent chitosan-based films.

Active components	Function	Foodstuff	Reference
A) Active			
Banana peels extract	Antioxidant	Apple	Zhang et al. (2020)
Mango leaf extract	Antioxidant	Cashew nuts	Rambabu et al. (2019)
Olive pomace	Antioxidant	Nuts	(de Moraes Crizel et al., 2018)
Citric acid	Antioxidant	Green chilli	Priyadarshi, Sauraj, Kumar, Deeba, et al. (2018)
α-tocopherol	Antioxidant	Mushroom	Zhang et al. (2020)
Spice extracts	Antioxidant	Pork	Liu et al. (2021)
***Zataria multiflora* Boiss EO and grape seed extract**	Antioxidant/ Antimicrobial	Mortadella sausage	Moradi et al. (2011)
Cinnamon and ginger EO	Antioxidant/ Antimicrobial	Pork	Wang, Wang, et al. (2017)
ϵ-polylysine	Antioxidant/ Antimicrobial	Beef fillet	Alirezalu et al. (2021)
Thinned young apple polyphenols	Antioxidant/ Antimicrobial	Grass carp	Sun et al. (2018)
Apricot kernel EO	Antioxidant/ Antimicrobial	Bread	Priyadarshi, Sauraj, Kumar, Deeba, et al. (2018)
***Thymus moroderi* and *Thymus piperella* EO**	Antioxidant/ Antimicrobial	Cooked cured ham	Ruiz-Navajas et al. (2015)
Red grape seed extract and *Ziziphora clinopodioides* EO	Antioxidant/ Antimicrobial	Minced rainbow troutfillet	Kakaei and Shahbazi (2016)
Potassium sorbate/ Vainillin	Antimicrobial	Butter cake	Sangsuwan et al. (2015)
Peanut skin and pink pepper residue	Antimicrobial	Chicken restructured	Serrano-León et al. (2018)
Gelatin	Antimicrobial	Peeled shrimp	Mohebi and Shahbazi (2017)
Chestnut extract	Antimicrobial	Fresh pasta	Kõrge et al. (2020)
***Eucalyptus globulus* EO**	Moisture absorbers/ Antimicrobial	Sliced sausage	Azadbakht et al. (2018)
Ginger EO	Moisture absorbers/ Antimicrobial	Barracuda fish	Remya et al. (2016)
Neat chitosan	Oxygen scavenger	Ground meat	Chounou et al. (2013)
Neat chitosan	Oxygen scavenger	Sliced sucuk	Şahin et al. (2017)
TiO2	Ethylene absorber	Tomato	Kaewklin et al. (2018)
Aloe vera gel	Ethylene absorber/ Antimicrobial	Mango	Shah and Hashmi (2020)
***Thymus vulgaris* EO**	Flavor absorber/ Antimicrobial	Cooked pork	Quesada et al. (2016)
B) Intelligent			
Alizarin	pH	Fish	Ezati & Rhim (2020)
Butterfly pudding extract	pH indicator	Fish	Yan et al. (2021)
Red cabbage extracts	pH indicator	Fish	Silva-Pereira et al. (2015)
***Bauhinia blakeana* Dunn dyes**	pH indicator	Pork and fish	Zhang et al. (2014)
Purple and black rice extracts	pH indicator	Pork	Yong et al. (2019)
Carrot anthocyanins	pH indicator	Pasteurized milk	(Ebrahimi Tirtashi et al., 2019)
Melissa officinalis essences and pomegranate peel extract	pH indicator	Cream cheese	Pirsa et al. (2020)
		Pork belly	

(*continued on next page*)

Table 5 (*continued*)

Active components	Function	Foodstuff	Reference
Red cabbage anthocyanins	pH-freshness indicator		Vo and Dang (2019)
Purple and black eggplant anthocyanins	pH-freshness indicator	Milk	Yong et al. (2019)
Jambolan fruit anthocyanins	Time-temperature indicator	Shrimp	Merz et al. (2020)
Jabuticaba fruit and purple sweet potato anthocyanins	Time-temperature indicator	Meat	Capello et al. (2020)
Red cabbage anthocyanins	Time-temperature indicator	Pasteurized milk	Pereira et al. (2015)
Roselle anthocyanins	Freshness indicator	Pork	Zhang, Zou, et al. (2019)
Purple tomato anthocyanins	Freshness indicator	Milk and fish	Li et al. (2021)
Black rice bran anthocyanins	Freshness indicator	Pomfret and shrimp	Wu et al. (2019)

developed chitosan active films by analyzing the peroxide value of the oil of the nuts (de Moraes Crizel et al., 2018; Rambabu et al., 2019).

Assays on physical parameters such as color, firmness, weight loss or respiration rate during storage time of fresh fruit or vegetables also indicate the positive effect of antioxidant chitosan films. For instance, films enriched with clove (*Eugenia caryophyllata*) essential oil (Wang et al., 2021), banana peels extract (Zhang et al., 2020), ascorbic acid (Sun, Liang, Xie, Lei, & Mo, 2010; Özdemir & Gökmen, 2017; Özdemir, Sultan, & Gökmen, 2019), aloe vera (Shah & Hashmi, 2020), procyanidins (Mannozzi et al., 2018) improve certain parameters for the storage time of apples, pomegranate arils, blueberry, mango, litchi fruit. Similar results have showed films with citric acid to preserve the color and texture of green chilli (Priyadarshi, Sauraj, Kumar, & Negi, 2018), or with the addition of α-tocopherol on mushroom to allow a maintained of firmness and alleviation of browning of these products (Zhang et al., 2020).

The effectiveness of chitosan active antioxidant films has been evaluated in meat products as well. Moradi, Tajik, Razavi Rohani, and Oromiehie (2011) has observed that in mortadella sausages packaged by neat chitosan films during refrigerated storage exhibited a rapid increase in thiobarbituric acid (used to measure the lipid oxidation) like control samples without film. However, oxidation in the samples wrapped with chitosan films formulated with grape seed extract and *Zataria multiflora Boiss* essential oil rapidly decreased during first 6 days (Moradi et al., 2011). Thyme, cloves, prickly ash, fennel, geranium, and cinnamon extracts on chitosan-based films have been evaluated to prolong the shelf-life of refrigerated pork. The results reflect that after 7 days the active film allows decreasing the total viable count of bacteria and effectively preventing the meat discoloration (Liu et al., 2021). Besides, active chitosan films enriched with syringic acid exert a preservative effect on quail eggs, observing a slower decrease of the weight loss rate, yolk index and Haugh unit values than those of the uncoated eggs (Yang et al., 2019).

The additional antimicrobial properties of many of these tested oils have shown a growth inhibitory effect of certain pathogenic microorganisms such as *L. monocytogenes* and delays the oxidation of lipids present in these foodstuffs. This desired effect has been observed in cooked cured ham (*Thymus moroderi* and *Thymus piperella* essential oil) (Ruiz-Navajas et al., 2015), pork (cinnamon and ginger essential oils) (Wang, Wang, et al., 2017), sliced sausage (*Eucalyptus globulus* essential oil) (Azadbakht et al., 2018) and mortadella sausages (*Zataria multiflora* and grape seed extract) (Moradi et al., 2011). The improvement of the chemical properties of chicken burger and delayed lipid oxidation have been reached by the application of chitosan/anise (*Pimpinella anisum* L.) essential oil films (Mahdavi et al., 2018).

The growth inhibition of *L. monocytogenes* and the antioxidant effect of active chitosan films have also been studied in fishery products such as minced trout fillet (red grape seed extract and *Ziziphora clinopodioides* essential oil) (Kakaei & Shahbazi, 2016) and grass carp fillets (apple polyphenols) (Sun et al., 2018). In these cases, the application of the films led to a delay in the oxidation of lipids and proteins present in the food. In peeled shrimp, there was a decrease in the total bacterial count of bacteria, such as *L. monocytogenes* when applying the active chitosan/gelatin film (Mohebi & Shahbazi, 2017). The application of chitosan/ginger essential oil films on Barracuda fish improve the moisture barrier properties and the antimicrobial effect against psychotropic and mesophilic bacteria (Remya et al., 2016). Chitosan films with microcapsules of grape seed extract and carvacrol have showed beneficial effects on fresh salmon fillets, maintaining the luminosity values closer to those of fresh salmon and lower total volatile basic nitrogen, pH and bacterial counts for a longer period of time (Alves et al., 2018).

In active chitosan films where antimicrobial effect is exerted, studies on foodstuffs are very varied. For instance, butter cake (Sangsuwan, Rattanapanone, & Pongsirikul, 2015) was studied, resulting in a tripling of the shelf-life of the product when chitosan film was applied with vanilla and potassium sorbate (Sangsuwan et al., 2015). Chitosan/apricot (*Prunus armeniaca*) kernel essential oil films have shown no fungal growth on bread slides after 10 days, being this type of food highly vulnerable to *Rhizopus stolonifera* (Priyadarshi, Sauraj, Kumar, & Negi, 2018).

Restructured chicken has been studied, and the use of chitosan films resulted in a significantly lower counts of psychotropic microorganisms than in those treated without active film (Serrano-León et al., 2018). Also, microbial spoilage in these samples decreased significantly compared to the control sample (Mahdavi et al., 2018). Chitosan-*Thymus vulgaris* essential oil films has been designed for the shelf-life extension of ready-to-eat meat products. The presence of *Thymus vulgaris* essential oil reduced yeast populations, whereas aerobic mesophilic bacteria, lactic acid bacteria, and enterobacteria have not been affected. Meat color preservation and the moisture condensation are enhanced in the presence of *Thymus vulgaris* essential oil, giving a better appearance to the packaged meat (Quesada, Sendra, Navarro, & Sayas-Barberá, 2016). Wrapping of cooked ham samples with chitosan/basil encapsulated essential oil films have been find to decrease mainly the growth of aerobic mesophilic bacteria and the enhancement of food pH for 10 days of the storage time analysis (Amor et al., 2021). The same antimicrobial effect has been produced in fresh pasta, where the application of chestnut extract to the chitosan films generated a process of retrogradation of the pasta, allowing it to maintain a hard texture after 9 days of conservation in the film (Kõrge, Bajić, Likozar, & Novak, 2020).

On the other hand, it was studied the O_2 scavenging capacity of active chitosan films applied on ground meat, where an increase in the shelf life of 5–6 days was obtained (Chounou et al., 2013). Other authors studied the same effect on active chitosan films applied on sliced sukuk, where a delay in the deterioration of the color of the meat product was also observed due to the O_2 scavenger (Şahin, Çarkcıoğlu, Demirhan, & Candoğan, 2017).

Ethylene absorption by active chitosan films leads to an increase of the post-harvest storage life and a delay of the ripening process mainly in fruits and vegetables, such as tomato fruit (Kaewklin, Siripatrawan, Suwanagul, & Lee, 2018) and mango (Shah & Hashmi, 2020).

Flavor absorption is also an effect that is being studied. It was found that the application of thyme essential oil to the active chitosan film generated a masking process of undesirable odors in cooked pork (Quesada et al., 2016).

Studies of chitosan active films are abundant, however most of them do not study the application of these films in food. This is important to target their use as food packaging.

6.2. Applications of intelligent films

Chitosan has been evaluated as raw material for intelligent films as Table 5 summarizes. Intelligent pH indicator films are used to monitor the freshness of foodstuffs. The indicator changes the color when the pH of the product changes. In this way, consumers can immediately see whether the food they are about to eat is in good condition. Intelligent pH-indicator chitosan-based films have been extensively studied over the last few years. Numerous studies have been carried out on fishery products. For instance, the effect of the addition of alizarin to chitosan film was studied in fish. It was observed that the color of the film changed vividly from slightly yellow to purple upon a pH change in the range 4–10 (Ezati & Rhim, 2020). The application of butterfly pudding extract caused a color change in chitosan films from purple-blue to dark green during fish preservation (Yan et al., 2021). Other added extracts, but in this case from red cabbage, resulted in a change of the film color from light blue to yellow after 7 days of storage. This indicates that pH changes have been detected and thus contamination of the fish fillets occurred (Silva-Pereira, Teixeira, Pereira-Júnior, & Stefani, 2015). A pH-sensitive chitosan-based film with dyes extract from the flower of *Bauhinia blakeana Dunn* has been also applied as a sticker sensor to test the freshness and pH changes of fish (Zhang, Lu, & Chen, 2014). This resulted in a change from purple to green as volatile amines are released from the feed (Zhang et al., 2014).

As for the meat products, a chitosan/black and purple rice extract samples have been developed, from which it is possible to determine the pH change in the pork samples studied (Yang et al., 2019). Results indicated that chitosan-purple rice extracts is more suitable for controlling the deterioration of the pork than chitosan-black rice extract, due to the moderate anthocyanin content (Yang et al., 2019). The chitosan, poly(vinyl alcohol), and anthocyanin extracted from red cabbage has been cast to serve as a wrapper for pork belly slices. The film turns yellowish with a pale green color when in a slightly alkaline range (Vo & Dang, 2019). The sensitive chitosan-*Bauhinia blakeana Dunn* film also successfully works to detect contamination of pork meat (Zhang et al., 2014). It was observed that after 48 h, the color of the detection film changes from brown to green, indicating an obvious pH change in the sample (Zhang et al., 2014).

Studies were also conducted on dairy products. Cellulose-chitosan films with the addition of carrot anthocyanins demonstrated that they could be used as a food grade biomaterial to control the freshness and spoilage of milk (Ebrahimi Tirtashi et al., 2019). After 48 h of storage at 20 °C, a color change from blue to purplish-pink can be observed. Similar results were obtained by chitosan-purple and black eggplant extracts films, where after 10 h of storage at 40 °C, the milk acidified promote the color change from light blue to dark blue. Specifically, black eggplant extract has showed a more significant color alteration (Yang et al., 2019). The cream cheese was also evaluated, where the chitosan/*Melissa officinalis* essences/Pomegranate peel extract film, as the time and temperature of the cheese increased, the samples became acidic and this pH change resulted in a color change from blue to red (Pirsa et al., 2020).

In the literature, the main studies about intelligent packaging based on chitosan, usually use anthocyanins to develop indifferently temperature and freshness indicators. A chitosan-based film with anthocyanins from Jambolana fruit that shows changes from red to blue when used to monitor the freshness of shrimp at temperatures between −20 °C and 20 °C has been developed (Merz et al., 2020). In meat products, a similar material with anthocyanins from fruit and sweet potato have been tested on meat samples under three different temperatures (−20, 4 and 20 °C) for 72 h (Capello et al., 2020). From the color change of the film from red to blue, it can be concluded that meats stored at 4 °C and 20 °C are considered as contaminated after 72 h and 24 h, respectively. Anthocyanins from roselle have been evaluated to control pork meat. A yellowish pigmentation of the film is generated as contamination of the meat begin to occur (Zhang, Zou, et al., 2019).

Studies have been carried out on pasteurized milk as well. The red cabbage anthocyanins in chitosan-samples turn from grey to dark pink when the milk is contaminated. This color change occurs when the milk reaches a pH value of 4.6, after being subjected to temperatures above refrigeration for time intervals ranging from 0 to 4 days (Pereira, de Arruda, & Stefani, 2015). Purple tomato anthocyanins have been studied on milk. In this case, the film color changes from green to pink, after a decrease in pH from 6.8 to 5.0 is generated after 48 h of storage (Li et al., 2021). The same authors have studied what effect these same films would have on fish samples. The film turns from dark green to light green. Fish samples stored after 48 h generate biogenic amines, which increase the pH of the product (Li et al., 2021). Seafood (pomfret fish and shrimp) spoilage has been monitored with chitosan-films enriched with oxidized chitin nanocrystals and black rice bran anthocyanins. The films show a color change from purple to blue-grey after 24 h of storage due to the increase of total volatile basic nitrogen in the packaging film (Wu et al., 2019).

Further studies of chitosan-based intelligent films in foodstuffs are necessary to delve into applications in other food groups such as fruits and vegetables, as they are highly perishable foods and can be easily contaminated.

7. Conclusions and future perspectives

The application of biodegradable natural polymers for the manufacture of food packaging is a technique that is becoming increasingly important. Due to the massive use of synthetic polymers, changes in food demand, industrial production trends, retail sales and consumer lifestyles, the packaging industry needs to evolve its packaging techniques.

Multiple studies have shown that the addition of other active compounds to chitosan films have interesting effects on their mechanical, barrier and functional properties. Revealing results have shown that the presence of phenolic compounds improves both mechanical and barrier properties. As for the functional properties of the films, through the addition of essential oils, phenolic compounds and fruit extracts, the antimicrobial and antioxidant capacity of the films improved significantly.

Studies are needed to determine the mechanical, barrier and functional properties of other types of chitosan-based films, such as ethylene scavenging, oxygen and CO_2 absorption/release, and flavor and odor absorption/release.

However, much more research is needed to be able to create chitosan-based packaging that can meet practical criteria and thus compete with petroleum-based packaging. The aspects to be worked on are: (1) Further research on chitosan and its combination with other materials to meet practical requirements. (2) Conduct studies on chitosan films and know what kinds of hazards we can expose ourselves to with its use. (3) To study which types of foodstuffs could be in contact with chitosan packaging.

Author contributions

Patricia Cazón: Visualisation, Methodology, Writing – review and editing. **Maria Florez**: Methodology, Investigation, Formal analysis, Writing – original draft. **Esther Guerra-Rodriguez:** Methodology, Writing – review and editing. **Manuel Vázquez**: Formal analysis, Writing – review and editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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