



Recent advances in chitosan-based active and intelligent packaging films incorporated with flavonoids

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ABSTRACT

Biopolymer-based films are promising packaging materials to maintain food quality, reduce food waste and ensure food safety. Chitosan, a biopolymer with excellent film-forming ability, can act as the carrier for plant-derived bioactive compounds. In the past decade, several studies have reported chitosan-based films incorporated with different classes of flavonoids, including flavones, flavanones, isoflavones, flavonols, flavanols and anthocyanidins. These films, based on their functionality, can be divided into chitosan/flavonoid active packaging films and chitosan/anthocyanin (the glycosylated anthocyanidin) intelligent packaging films. This paper presents a comprehensive review on active and intelligent packaging films prepared from chitosan and different classes of flavonoids, with special attention being paid to the preparation, physical and functional properties, stabilization, and application of the films. Factors affecting the physical and functional properties of the films are summarized. In addition, the challenges for the commercial production and application of the films in active and intelligent packaging fields are discussed.

1. Introduction

With the continuous improvement of people's lifestyle, their demands for fresh, natural, healthy and nutritious foods are increasing. In the food supply chain, packaging is an essential process to maintain food quality, reduce food waste and ensure food safety (Verghese, Lewis, Lockrey, & Williams, 2015). The traditional petroleum-based plastic packaging films have been used for several decades, due to their advanced manufacturing technology, low production cost, and ideal barrier and mechanical properties. Despite this, traditional packaging films are hardly degradable in natural environment and have potential food pollution risk. In this context, the development of bioplastics with good biodegradability and compostability has been considered as a promising solution (Asgher, Qamar, Bilal, & Iqbal, 2020).

Till now, numerous emerging biomass-derived packaging films have been developed, especially those with active and intelligent packaging functions (Amin et al., 2022). Active packaging and intelligent packaging are two food packaging techniques with distinct purposes. Similar with traditional packaging films, active packaging can supply barrier and mechanical protections for foods. Moreover, active packaging is

developed to prolong the storage period and improve the quality of foods by releasing antioxidants and antimicrobials or absorbing O₂, CO₂, ethylene and odors (Amin et al., 2022). Intelligent packaging is developed to record or share the information about packaging environment and food quality in different forms, such as data carriers, freshness indicators and biosensors (Firouz, Mohi-Alden, & Omid, 2021). Different from active packaging, intelligent packaging only needs a small piece of intelligent film to be adhered to an intact packaging system.

In recent years, the fabrication of chitosan-based active and intelligent packaging films has received increasing attention (Flórez, Guerra-Rodríguez, Cazón, & Vázquez, 2022). Chitosan is biodegradable and compostable polycationic polysaccharide with no adverse environmental impacts. Due to the presence of amino and hydroxyl groups in its structure, chitosan exhibits many unique physicochemical properties, such as pH-dependent solubility, metal ion-chelating ability, and antioxidant and antimicrobial activities (Wang, Xue, & Mao, 2020). Meanwhile, chitosan has excellent film-forming ability and can act as the carrier of numerous functional ingredients, such as essential oils, pure phenolic compounds, polyphenol-rich plant extracts, nano-sized particles and pH-sensitive colorants (Sarraz et al., 2024). Chitosan films

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reinforced with functional ingredients show good application potentials in active and intelligent packaging fields.

Flavonoids are a group of plant-derived secondary metabolites with numerous health-promoting effects (Ullah et al., 2020). As displayed in Fig. 1A, all flavonoids share the basic C6–C3–C6 skeleton (A–C–B rings) and are generally divided into six classes (i.e. flavones, flavanones, isoflavones, flavonols, flavanols and anthocyanidins), depending on the unsaturation and oxidation degrees of C ring and the linkage pattern between B and C rings (Chen, Wang, Cheng, Gao, & Chen, 2023). Meanwhile, flavonoids are also diverse in the patterns of hydroxylation,

methoxylation, glycosylation and esterification. Considering flavonoids have potent antioxidant and antimicrobial activities, different classes of flavonoids have been added into chitosan matrix to prepare active packaging films (Chen et al., 2023). Anthocyanins, the glycosylated products of anthocyanidins, are pH-sensitive colorants that can be integrated with chitosan matrix to yield intelligent packaging films (Roy & Rhim, 2021a). Chitosan-based active and intelligent packaging films can prolong the storage period and indicate the freshness of different foods. However, there is no relevant review to summarize the recent advances in chitosan-based active and intelligent packaging films incorporated

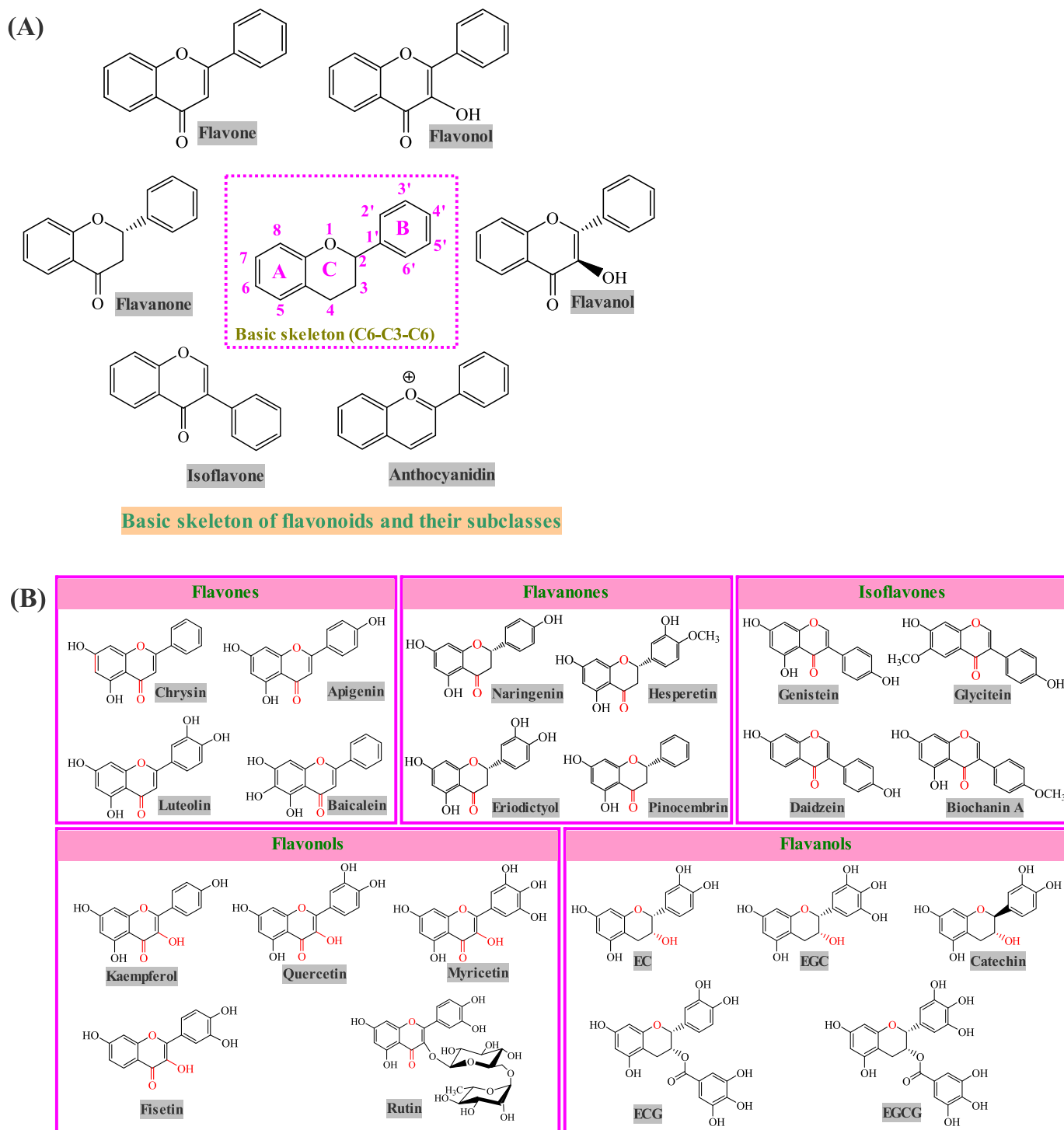


Fig. 1. The molecular basis of different classes of flavonoids (A) and the chemical structures of typical flavonoids used for the production of chitosan/flavonoid active packaging films (B).

with different classes of flavonoids.

In this context, the purpose of this review is to summarize the recent advances in chitosan-based films containing different classes of flavonoids with active and intelligent packaging potentials. The preparation methods, physical and functional properties, stabilization, and application of chitosan/flavonoid active packaging films and chitosan/anthocyanin intelligent packaging films are separately illustrated. At the same time, factors affecting the physical and functional properties of the films, especially the release behavior of chitosan/flavonoid active packaging films and the color changeability of chitosan/anthocyanin intelligent packaging films, are summarized. The challenges for the commercial production and application of the films in active and intelligent packaging fields are also discussed.

2. Chitosan-based active packaging films with flavonoids

2.1. Preparation of the films

Although chitosan has good film-forming ability, its intrinsic antioxidant and antimicrobial activities cannot satisfy the requirement of active packaging (Wang et al., 2020). Considering flavonoids are natural and potent antioxidant and antimicrobial agents, the combined use of chitosan and flavonoids can produce idea active packaging films. As summarized in Table 1, different classes of flavonoids including flavones (apigenin, baicalein, chrysin and luteolin), flavanones (naringenin and naringin), flavanols (fisetin, kaempferol, myricetin, quercetin and rutin) and flavanols (catechin, epicatechin (EC), epicatechin gallate (ECG), epigallocatechin (EGC) and epigallocatechin gallate (EGCG)) have been added into chitosan-based matrix to develop active packaging films. It is worth noting anthocyanidins, a unique class of flavonoids, can be used to develop not only active packaging films but also intelligent packaging films, which will be thoroughly introduced in the next section. The chemical structures of typical flavonoids used for the production of active packaging films are presented in Fig. 1B. Among different flavonoids, quercetin, rutin and catechins are most widely selected by researchers.

In general, flavonoids can be added into chitosan-based films in three different ways (Fig. 2): (1) In most cases, flavonoids are directly added into chitosan-based film-forming solutions by physically mixing to yield packaging films. Notably, most flavonoids, especially flavones, flavanones and flavanols are hydrophobic substances and they have low solubility and compatibility with hydrophilic chitosan-based film-forming solutions. Thus, the addition amount of hydrophobic flavonoids in the films is frequently very low. (2) Flavonoids are first encapsulated in nano-sized carriers, such as nanoparticles (Roy & Rhim, 2021d), nanoemulsions (Bi, Qin, Chen, Kan, & Liu, 2021), nanoliposomes (Ali et al., 2023), nanotubes (Roy & Rhim, 2021a) and nanocapsules (Liang et al., 2017), and the encapsulated flavonoids are then incorporated into chitosan-based film-forming solutions to produce packaging films. Encapsulation can greatly increase the solubility and stability of flavonoids and create films with sustained release of antioxidant and antimicrobial activities. (3) Flavonoids are grafted onto polysaccharides, such as starch aldehyde (Sun et al., 2024; Yong et al., 2020) and chitosan (Mittal et al., 2021; Torres et al., 2012; Wang, Huang, Yun, Yong, & Liu, 2022; Yong, Wang, Huang, & Liu, 2024), through chemical and enzymatic methods to produce polysaccharide-flavonoid conjugates. And then polysaccharide-flavonoid conjugates are added into chitosan-based film-forming solutions to yield packaging films. The graft of flavonoids onto polysaccharides greatly elevates the solubility and stability of flavonoids. The produced polysaccharide-flavonoid conjugates are hydrophilic and have good compatibility with chitosan-based film-forming solutions.

As listed in Table 1, other polymers including pullulan (Qi et al., 2023; Roy & Rhim, 2021b), gelatin (Benbettaieb, Chambin, Karbowskiak, & Debeaufort, 2016; Benbettaieb, Karbowskiak, Brachais, & Debeaufort, 2015; Ponnusamy et al., 2024; Ponnusamy et al., 2024; Roy & Rhim,

2021c, 2021d; Yadav, Mehrotra, Bhartiya, Singh, & Dutta, 2020), hordein (Li, Yan, Guan, & Huang, 2020), pectin (Ali et al., 2023; Fu et al., 2024), polyvinyl alcohol (Narasagoudr et al., 2020; Sabaghi, Maghsoudlou, Kashiri, & Shakeri, 2020; Wang et al., 2024) and zein (Liang et al., 2017) can be blended with chitosan to produce complex film matrix based on their intermolecular hydrogen bond and/or electrostatic interactions. Glycerol, as a plasticizer, is normally added into the film-forming solutions. Besides, different kinds of reinforcing agents, such as D- α -tocopheryl polyethylene glycol 1000 succinate (TPGS) (Bi et al., 2020; Yong et al., 2020; Zhang et al., 2020), nano materials (Qi et al., 2023; Roy & Rhim, 2021b; Wang et al., 2018; Wang et al., 2024), deep eutectic solvent (Jakubowska, Gierszewska, Szydłowska-Czerniak, Nowaczyk, & Olewnik-Kruszkowska, 2023) and essential oils (Roy & Rhim, 2021c, 2021d) can be added into film-forming solutions to produce films with improved physical and functional properties. Genipin, a chemical cross-linking agent, is sometimes added into film-forming solutions in order to enhance the mechanical properties of the films (Roy & Rhim, 2021d).

In most cases, the above mentioned film-forming solutions are poured into moulds and then dried to produce monolayer or bilayer packaging films. This method is well known as solvent casting, which has been widely used to prepare chitosan/flavonoid films (Table 1). This is because solvent casting is easy to handle and does not require specific equipment. In some circumstances, electrospinning has been used to develop chitosan/flavonoid nanofiber films (Li et al., 2020; Ponnusamy et al., 2024; Tian et al., 2016). The nanofibers created by electrospinning have several advantages, such as high specific surface and three-dimensional (3D) fine structure with good flavonoid-loading ability. Notably, the prepared chitosan/flavonoid films can be further exposed to UV (Iturriaga, Olabarrieta, Castellan, Gardrat, & Coma, 2014), electron beam (Benbettaieb et al., 2015; Benbettaieb et al., 2016) and gamma irradiation (Sabaghi et al., 2020) to strengthen the mechanical properties of the films, which is based on the cross-linking between chitosan and other polymers. The procedure for the production of active packaging films based on chitosan and flavonoids is presented in Fig. 2.

2.2. Physical and functional properties of the films

The prepared chitosan/flavonoid films are normally characterized for their physical and functional properties. The physical properties of the films include water resistance (e.g., moisture content (MC), swelling ratio (SR), water solubility (WS), water contact angle (WCA)), barrier properties (e.g., light transmittance (LT), water vapor permeability (WVP) and oxygen permeability (OP)), mechanical properties (e.g., tensile strength (TS), elongation at break (EAB) and elastic modulus (EM)) and thermal properties (e.g., differential scanning calorimetry (DSC) and thermogravimetric analysis (TGA)). The functional properties of the films include antioxidant and antimicrobial activities, which are primarily determined by free radical scavenging and bacteriostatic tests, respectively. Table 1 summarizes the impact of different flavonoids on the physical and functional properties of chitosan/flavonoid films. Notably, the impact of different flavonoids on the films is inconsistent, which is because the physical and functional properties of the films are influenced by different factors.

Several researchers have revealed the impact of flavonoid content on the physical and properties of chitosan/flavonoid films, where a low content of flavonoids is favored to produce films with good mechanical and barrier properties but a high content of flavonoids is favored to produce films with high antioxidant and antimicrobial activities (Lipatova, Makarova, & Yusova, 2021; Narasagoudr et al., 2020; Sutharsan, Boyer, & Zhao, 2022; Yong et al., 2020). The interactions between chitosan and flavonoids are mainly based on hydrogen bonds (Fig. 3A). As for the films containing polysaccharide-flavonoid conjugates, their physical and functional properties are normally influenced by the content of polysaccharide-flavonoid conjugates (Mittal et al., 2021) and the grafting ratio of the conjugates (Yong et al., 2024). Mittal

Table 1

Formulations, physical and functional properties, and applications of active packaging films based on chitosan and different classes of flavonoids.

Flavonoids	Film matrix	Reinforcing agents	Film preparation method	Impact of flavonoids on the physical properties of the films	Functional properties of the films	Factors affecting the properties of the films	Application of the films	References
Flavones								
Apigenin	Chitosan	TPGS	Solvent casting	WVP↓, OP↓, LT↓, TS↓, EAB↑	DPPH radical scavenging activity Antimicrobial activity against <i>E. coli</i> , <i>S. aureus</i> , <i>L. monocytogenes</i> and <i>S. typhimurium</i>	The type of flavonoid		Bi et al., 2020
Baicalein	Chitosan	TPGS	Solvent casting	WS↓, WVP↓, OP↓, LT↓, TS↓, EAB↑	DPPH radical scavenging activity	The content of baicalein	Preserving soybean oil	Yong et al., 2020
Chrysin	Chitosan	TPGS	Solvent casting	WVP↔, OP↑, LT↓, TS↓, EAB↑	DPPH radical scavenging activity Antimicrobial activity against <i>E. coli</i> , <i>S. aureus</i> , <i>L. monocytogenes</i> and <i>S. typhimurium</i>	The type of flavonoid		Bi et al., 2020
Luteolin	Chitosan	TPGS	Solvent casting	WVP↓, OP↓, LT↓, TS↓, EAB↑	DPPH radical scavenging activity, Antimicrobial activity against <i>E. coli</i> , <i>S. aureus</i> , <i>L. monocytogenes</i> and <i>S. typhimurium</i>	The type of flavonoid		Bi et al., 2020
	Chitosan	Nanoemulsions	Solvent casting	WVP↓, OP↓, LT↓, TS↑, EAB↑	DPPH radical scavenging activity Antimicrobial activity against <i>E. coli</i> , <i>S. aureus</i> , <i>L. monocytogenes</i> and <i>S. typhimurium</i>	The encapsulation of luteolin		Bi et al., 2021
	Chitosan		Solvent casting	MC↓, WS↑, SR↑, WVP↓, TS↑, EAB↑	DPPH radical scavenging activity Antimicrobial activity against <i>E. coli</i> , <i>S. aureus</i> , <i>L. monocytogenes</i> and <i>S. typhimurium</i>	The type and content of flavonoid		Sutharsan et al., 2022
Flavanones								
Naringenin	Chitosan		Solvent casting		Anti-inflammatory			Quintão et al., 2022
Naringin	Chitosan		Solvent casting		DPPH radical scavenging activity Antimicrobial activity against <i>L. innocua</i>	The treatment of the films with UV irradiation		Iturriaga et al., 2014
Flavanols								
Fisetin	Chitosan/pullulan blend	Nano Ag	Solvent casting	WVP↓, LT↓, TS↔, EAB↔	DPPH radical scavenging activity Antimicrobial activity against <i>E. coli</i> , <i>S. aureus</i> and <i>A. niger</i>	The presence of nano Ag	Preserving litchi	Qi et al., 2023
Kaempferol	Chitosan	TPGS	Solvent casting	WVP↓, OP↓, LT↓, TS↑, EAB↑	DPPH radical scavenging activity Antimicrobial activity against <i>E. coli</i> , <i>S. aureus</i> , <i>L. monocytogenes</i> and <i>S. typhimurium</i>	The type of flavonoid		Zhang et al., 2020
Myricetin	Chitosan	TPGS	Solvent casting	WVP↓, OP↓, LT↓, TS↑, EAB↑	DPPH radical scavenging activity Antimicrobial activity against <i>E. coli</i> , <i>S. aureus</i> , <i>L. monocytogenes</i> and <i>S. typhimurium</i>	The type of flavonoid		Zhang et al., 2020
Quercetin	Chitosan		Solvent casting	MC↓, WS↑, SR↑, WVP↓, TS↑, EAB↑	DPPH radical scavenging activity Antimicrobial activity against <i>E. coli</i> , <i>S. aureus</i> , <i>L. monocytogenes</i> and <i>S. typhimurium</i>	The type and content of flavonoid	Preserving beef	Sutharsan et al., 2022
	Chitosan		Solvent casting	SR↓, WVP↓, WCA↑, LT↓, TS↑, EAB↓	DPPH and ABTS radical scavenging activity Antimicrobial activity against <i>S. aureus</i>	The graft of quercetin with starch aldehyde	Preserving citrus	Sun et al., 2024

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Table 1 (continued)

Flavonoids	Film matrix	Reinforcing agents	Film preparation method	Impact of flavonoids on the physical properties of the films	Functional properties of the films	Factors affecting the properties of the films	Application of the films	References
	Chitosan/polyvinyl alcohol blend	Cu ²⁺ and layered clay nanosheets	Solvent casting	LT↓, TS↑, EAB↓	DPPH and ABTS radical scavenging activity Antimicrobial activity against <i>E. coli</i>	The presence of Cu ²⁺ and layered clay nanosheets		Wang et al., 2024
	Chitosan/gelatin blend		Solvent casting	WVP↓, WCA↓, OP↓, TS↑, EAB↑		The irradiation of the films with electron beam		Benbettaieb et al., 2015, 2016
	Chitosan	Deep eutectic solvent	Solvent casting	WS↓, WVP↓, OP↓, LT↓, TS↑, EAB↓	DPPH radical and H ₂ O ₂ scavenging activity	The content of quercetin and the presence of deep eutectic solvent	Preserving rapeseed oil	Jakubowska et al., 2023
	Chitosan		Solvent casting	WVP↔, OP↓, TS↓, EAB↑	DPPH radical scavenging activity Antimicrobial activity against <i>E. coli</i> , <i>S. sonnei</i> , <i>P. mirabilis</i> , <i>M. organii</i> , <i>S. epidermidis</i> , <i>M. luteus</i> and <i>B. subtilis</i>			Souza et al., 2015
	Chitosan	Chitosan nanoparticles	Solvent casting	WVP↔, WCA↔, LT↓, TS↑, EAB↑	DPPH and ABTS radical scavenging activity Antimicrobial activity against <i>E. coli</i> and <i>L. monocytogenes</i>	The encapsulation of quercetin		Roy & Rhim, 2021c
	Chitosan/gelatin blend	Rosemary essential oil and genipin	Solvent casting	WVP↔, WCA↔, LT↓, TS↑, EAB↓	DPPH and ABTS radical scavenging activity Antimicrobial activity against <i>E. coli</i> and <i>L. monocytogenes</i>	The presence of rosemary essential oil and genipin		Roy & Rhim, 2022
	Chitosan/pectin blend	Soy lecithin nanoliposomes	Solvent casting	MC↓, WVP↓, WCA↑, LT↓, TS↓, EAB↑	Antimicrobial activity against <i>E. coli</i> , <i>L. monocytogenes</i> and <i>S. enterica</i>	The encapsulation of quercetin	Preserving chicken	Ali et al., 2023
	Chitosan/hordein blend		Electrospinning		DPPH and ABTS radical scavenging activity	The encapsulation of quercetin	Preserving apple and potato	Li et al., 2020
	Chitosan/gelatin blend		Solvent casting	WS↓, SR↓, WVP↔, OP↓, TS↑, EAB↑	DPPH and ABTS radical scavenging activity Antimicrobial activity against <i>E. coli</i> and <i>B. subtilis</i>			Yadav et al., 2020
	Chitosan	TPGS	Solvent casting	WVP↓, OP↓, LT↓, TS↑, EAB↑	DPPH radical scavenging activity Antimicrobial activity against <i>E. coli</i> , <i>S. aureus</i> , <i>L. monocytogenes</i> and <i>S. typhimurium</i>	The type of flavonoid		Zhang et al., 2020
Rutin	Chitosan	Gallic acid and coumarin	Solvent casting	LT↓	DPPH radical scavenging activity	The presence of gallic acid and coumarin		Jacobs et al., 2023
	Chitosan		Solvent casting	WS↓, WVP↑, TS↑, EAB↔		The pre-treatment of film-forming solution		Lipatova et al., 2021
	Chitosan/gelatin blend	Cinnamon essential oil	Solvent casting	WVP↔, WCA↑, LT↓, TS↔, EAB↓	DPPH and ABTS radical scavenging activity Antimicrobial activity against <i>E. coli</i> and <i>L. monocytogenes</i>	The presence of cinnamon essential oil		Roy & Rhim, 2021b
	Chitosan/pullulan blend	Chitosan-functionalized halloysite nanotube	Solvent casting	WVP↓, LT↓, TS↑, EAB↑	DPPH and ABTS radical scavenging activity Antimicrobial activity against <i>E. coli</i> and <i>L. monocytogenes</i>	The presence of chitosan-functionalized halloysite nanotube		Roy & Rhim, 2021a
	Chitosan/polyvinyl alcohol blend		Solvent casting	WS↑, WCA↓, WVP↓, OP↓, LT↓, TS↑, EAB↑	Antimicrobial activity against <i>E. coli</i> and <i>S. aureus</i>	The content of rutin		Narasagoudr et al., 2020
Flavanols Catechin	Chitosan		Solvent casting	MC↓, WS↑, SR↑, WVP↓, TS↑, EAB↑	DPPH radical scavenging activity, Antimicrobial activity against <i>E. coli</i> , <i>S. aureus</i> ,	The type and content of flavonoid	Preserving beef	Sutharsan et al., 2022, 2023

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Table 1 (continued)

Flavonoids	Film matrix	Reinforcing agents	Film preparation method	Impact of flavonoids on the physical properties of the films	Functional properties of the films	Factors affecting the properties of the films	Application of the films	References
	Chitosan		Solvent casting	WS↑, WCA↓, WVP↓, OP↓, LT↓, TS↑, EAB↑	<i>L. monocytogenes</i> and <i>S. typhimurium</i> DPPH and ABTS radical scavenging activity	The type of flavonoid grafted onto chitosan	Preserving corn oil	Wang et al., 2022
	Chitosan		Solvent casting	SR↓, WCA↓, WVP↓, OP↓, LT↓, TS↑, EAB↓	DPPH radical scavenging activity	The type of flavonoid grafted onto starch aldehyde	Preserving sunflower seed oil	Yong et al., 2022
	Chitosan/polyvinyl alcohol blend	Chitosan nanoparticles	Solvent casting			The treatment of the films with gamma irradiation		Sabaghi et al., 2020
ECG	Chitosan		Solvent casting	WS↑, WCA↓, WVP↓, OP↓, LT↓, TS↑, EAB↑	DPPH and ABTS radical scavenging activity	The type of flavonoid grafted onto chitosan	Preserving corn oil	Wang et al., 2022
	Chitosan		Solvent casting	WS↑, WCA↓, WVP↓, OP↓, LT↓, TS↑, EAB↑	DPPH and ABTS radical scavenging activity	The grafting ratio of chitosan-ECG conjugate	Preserving corn oil	Yong et al., 2024
	Chitosan		Solvent casting	SR↓, WCA↓, WVP↓, OP↓, LT↓, TS↑, EAB↓	DPPH radical scavenging activity	The type of flavonoid grafted onto starch aldehyde	Preserving sunflower seed oil	Yong et al., 2022
EGC	Chitosan		Solvent casting	WS↑, WCA↓, WVP↓, OP↓, LT↓, TS↑, EAB↑	DPPH and ABTS radical scavenging activity	The type of flavonoid grafted onto chitosan	Preserving corn oil	Wang et al., 2022
	Chitosan		Solvent casting	SR↓, WCA↓, WVP↓, OP↓, LT↓, TS↑, EAB↓	DPPH radical scavenging activity	The type of flavonoid grafted onto starch aldehyde	Preserving sunflower seed oil	Yong et al., 2022
EGCG	Chitosan		Solvent casting	WS↑, WCA↓, WVP↓, OP↓, LT↓, TS↑, EAB↑	DPPH and ABTS radical scavenging activity	The type of flavonoid grafted onto chitosan	Preserving corn oil	Wang et al., 2022
	Chitosan/pectin bilayer matrix		Solvent casting	WCA↑, WVP↓, OP↓, LT↓, TS↑, EAB↔	DPPH and ABTS radical scavenging activity Antimicrobial activity against <i>E. coli</i> , <i>S. aureus</i> , <i>P. extensa</i> and <i>B. cinerea</i>	The protective effect of bilayer film	Preserving strawberry	Fu et al., 2024
	Chitosan/gelatin blend		Solvent casting/ electrospinning	SR↓, WVP↑, TS↑, EAB↓	DPPH and ABTS radical scavenging activity	The film preparation method	Preserving asian seabass depot fat oil	Ponnusamy et al., 2024
	Chitosan	Rectorite	Electrospinning	TS↓, EAB↓	Antimicrobial activity against <i>S. aureus</i>	The presence of rectorite		Tian et al., 2016
	Chitosan	Melanin-like nanoparticles	Solvent casting	WS↔, MC↓, SR↑, WVP↓, WCA↑, LT↓, TS↔, EAB↑	DPPH and ABTS radical scavenging activity Antimicrobial activity against <i>E. coli</i> and <i>S. aureus</i>	The encapsulation of EGCG		Zhao et al., 2022
	Chitosan		Solvent casting	WVP↑, LT↓, TS↓, EAB↓	DPPH and ABTS radical scavenging activity Antimicrobial activity against <i>P. aeruginosa</i> and <i>L. monocytogenes</i>	The content of chitosan-EGCG conjugate	Preserving Asian sea bass slices	Mittal et al., 2021
	Chitosan	Nano-bacterial cellulose	Solvent casting	WS↑, MC↓, SR↓, OP↔, LT↓, TS↓, EAB↓	ABTS radical scavenging activity	The presence of nano-bacterial cellulose		Wang et al., 2018
	Chitosan/zein blend	Chitosan hydrochloride nanocapsules	Solvent casting		DPPH radical scavenging activity	The nature of film matrix		Liang et al., 2017
	Chitosan		Solvent casting	WS↔, MC↓, SR↓, WCA↓, LT↓, TS↑, EAB↓	DPPH radical scavenging activity Antimicrobial activity against <i>E. coli</i> and <i>S. aureus</i>			Dai et al., 2022
	Chitosan		Solvent casting	SR↓, WCA↓, WVP↓, OP↓, LT↓, TS↑, EAB↔	DPPH radical scavenging activity	The type of flavonoid grafted onto starch aldehyde	Preserving sunflower seed oil	Yong et al., 2022

ABTS, 2,2'-azino-bis(3-ethylbenzothiazoline-6-sulfonate); DPPH, 2,2-diphenyl-1-picrylhydrazyl; EAB, elongation at break; LT, light transmittance; MC, moisture content; OP, oxygen permeability; SR, swelling ratio; TS, tensile strength; WCA, water contact angle; WS, water solubility; WVP, water vapor permeability; ↑, increased after flavonoid addition; ↓, decreased after flavonoid addition; ↔, unchanged after flavonoid addition.

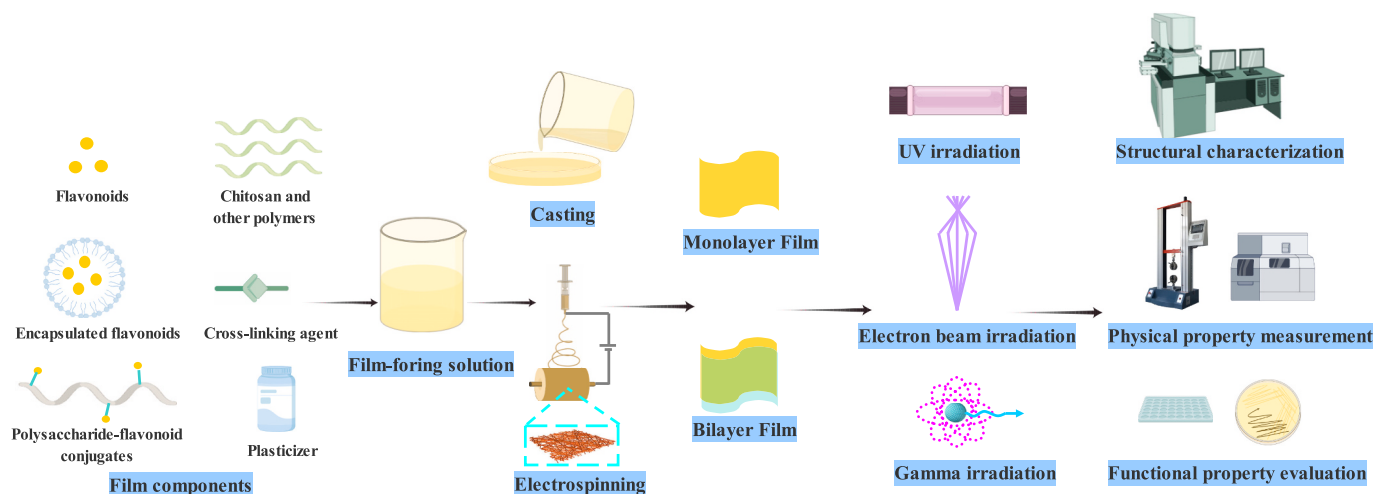


Fig. 2. The scheme illustrating the procedure for the preparation and characterization of chitosan/flavonoid active packaging films.

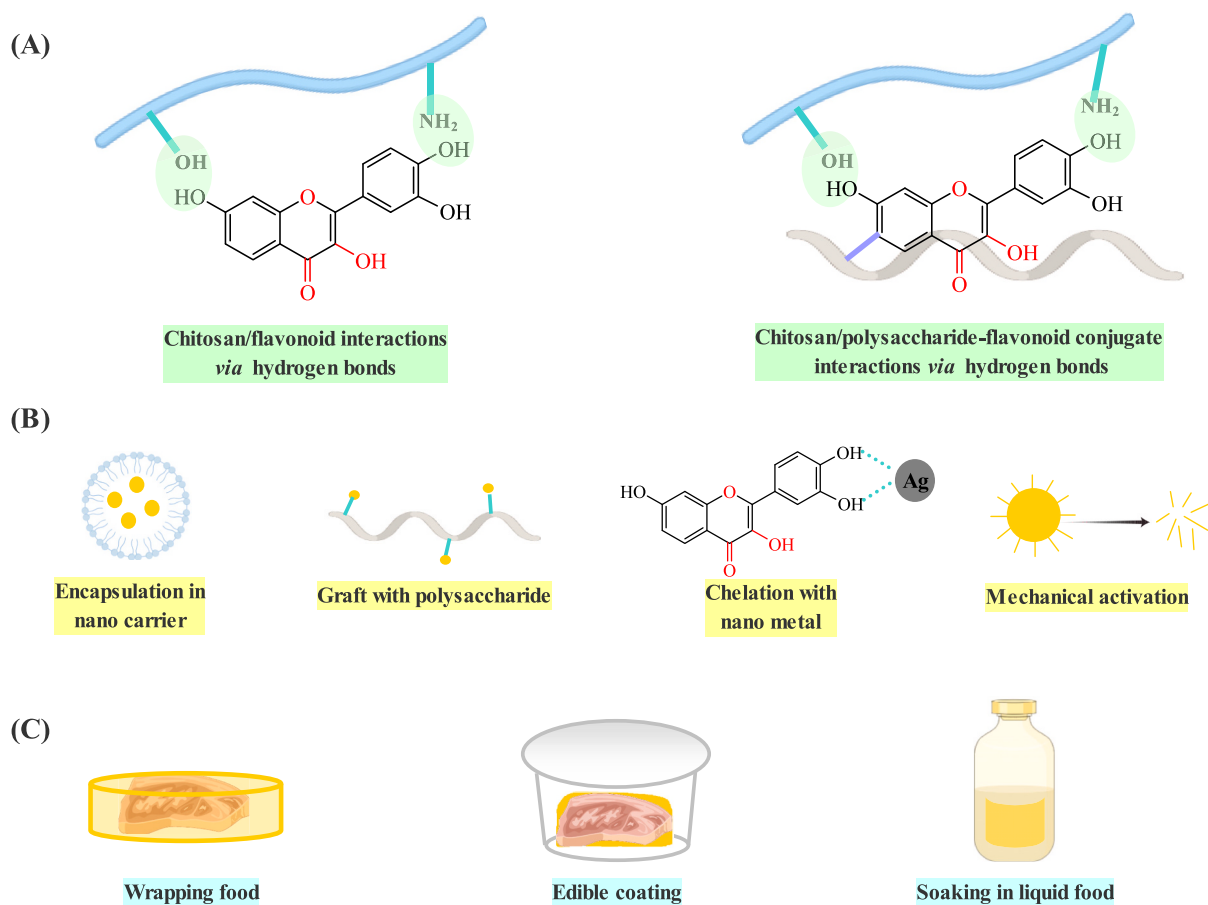


Fig. 3. The diagrams showing the intermolecular chitosan/flavonoid interactions and chitosan/polysaccharide-flavonoid conjugate interactions (A), the strategies to enhance the stability and solubility of flavonoids in chitosan/flavonoid active packaging films (B), and the application forms of chitosan/flavonoid active packaging films (C).

et al. (2021) found the mechanical properties of chitosan/chitosan-EGCG conjugate composite films decreased with the content of chitosan-EGCG conjugate. Yong et al. (2024) revealed the grafting ratio of chitosan-EGCG conjugate was positively correlated with the barrier, mechanical and antioxidant properties of chitosan/chitosan-EGCG conjugate composite films. The interactions between chitosan and

polysaccharide-flavonoid conjugates are primarily based on hydrogen bonds (Fig. 3A).

The physical and functional properties of chitosan/flavonoid films are greatly affected by the type of flavonoids. Sutharsan et al. (2022) compared the physical properties of chitosan-based films containing luteolin (flavone), quercetin (flavonol) and catechin (flavanol). They

found the film with quercetin had the highest mechanical properties but the film with luteolin had the lowest WVP. The same research group further found the film with quercetin had the highest antioxidant activity (Sutharsan, Boyer, & Zhao, 2023). These two studies show different classes of flavonoids significantly influence the physical and functional properties of the films. Zhang et al. (2020) investigated the impact of three flavonols (myricetin, quercetin and kaempferol) on chitosan-based films, and found the film with myricetin had the highest mechanical and barrier properties but the film with kaempferol had the highest DPPH scavenging activity. This study demonstrates different flavonoids within the same class significantly influence the physical and functional properties of chitosan/flavonol films. In a similar study, Bi et al. (2020) revealed that luteolin, apigenin and chrysin, belonging to flavones, affected chitosan films in different degrees. The film containing luteolin had the highest mechanical, barrier and antioxidant properties while the film containing chrysin had the highest antimicrobial activity. Recently, Wang et al. (2022) prepared chitosan-based films containing four kinds of chitosan-catechin conjugates (i.e. chitosan-EC, chitosan-ECG, chitosan-EGC and chitosan-EGCG conjugates), and found the film containing chitosan-ECG conjugate had the highest barrier and antioxidant properties while the film containing chitosan-EGCG conjugate had the highest mechanical properties. Likewise, Yong, Xu, Yun, Hu, and Liu (2022) fabricated chitosan-based films added with four kinds of starch aldehyde-catechin conjugates, and demonstrated the film added with starch aldehyde-ECG conjugate presented the highest mechanical, barrier and antioxidant properties.

Some recent studies have demonstrated the encapsulation of flavonoids can greatly influence the physical and functional properties of chitosan/flavonoid films (Sun et al., 2024). Roy and Rhim (2021d) revealed that chitosan nanoparticles containing quercetin increased the mechanical properties of chitosan-based films. Similarly, Bi et al. (2021) observed the incorporation of luteolin-loaded nanoemulsions improved the mechanical and barrier properties of chitosan films. However, quercetin-loaded nanoliposomes were found to reduce the mechanical properties but increased the hydrophobicity of chitosan/pectin films (Ali et al., 2023). It is worth noting that encapsulation is effective to slow down the release of flavonols from chitosan/flavonoid films, which can make the films exert long-term antioxidant and antimicrobial performances (Bi et al., 2021; Roy & Rhim, 2021d). The physical and functional properties of chitosan/flavonoid films can be elevated by polysaccharide-flavonoid conjugates. Sun et al. (2024) documented starch aldehyde-quercetin conjugates increased the mechanical, barrier, antioxidant and antimicrobial properties of chitosan films, because the bioactive conjugates formed hydrogen bonds and Schiff-base linkages with chitosan. A similar result was documented by Yong, Liu, Kan, and Liu (2022) in chitosan films added with four kinds of starch aldehyde-catechin conjugates.

The physical and functional properties of chitosan/flavonoid films are also affected by the presence of reinforcing agents. For instance, Wang et al. (2018) used nano-bacterial cellulose as a reinforcing agent to increase the water resistance, mechanical and antioxidant properties of chitosan/EGCG films. Qi et al. (2023) prepared fisetin-nano Ag complex based on chelation and found the formed complex could significantly elevate the water vapor barrier and antimicrobial properties of chitosan/pullulan-based films. Jakubowska et al. (2023) applied deep eutectic solvent (choline chloride/citric acid) in chitosan/quercetin films and found the deep eutectic solvent increased the oxygen barrier and antioxidant properties of the films. TPGS, an amphiphilic substance, was added into the chitosan/flavonoid films to improve the miscibility between hydrophobic flavonoid and hydrophilic chitosan (Bi et al., 2020; Yong et al., 2020; Zhang et al., 2020). As reported, chitosan/flavonoid films added with TPGS showed good homogeneity and mechanical properties (Bi et al., 2020; Yong et al., 2020; Zhang et al., 2020).

Interestingly, the physical properties of chitosan/flavonoid films can be improved by the pre-treatment of film-forming solutions. Lipatova

et al. (2021) treated the chitosan/rutin film-forming solutions using a rotor-stator device, which greatly reduced the size of rutin and improved the solubility of rutin in the solutions. As a result, the produced chitosan/rutin films showed uniform appearances. The preparation method of chitosan/flavonoid films is an important factor affecting the physical and functional properties of the films, which is often ignored by researchers. Recently, Ponnusamy, Rajasekaran, et al. (2024) prepared bilayer films by covering chitosan/gelatin/EGCG on polylactic acid via solvent casting and electrospinning techniques. They found the bilayer film prepared by solvent casting had higher TS, WVP and antioxidant activity but higher EAB than the film prepared by electrospinning. As shown in Fig. 2, chitosan/flavonoid films can be subjected to irradiation treatments to further enhance their physical properties. Benbettaieb et al. (2015) documented electron beam irradiation increased the TS and oxygen barrier ability of chitosan/gelatin films containing quercetin. Likewise, Sabaghi et al. (2020) found the TS and water vapor barrier ability of chitosan/polyvinyl alcohol films containing catechin-loaded chitosan nanoparticles were elevated by gamma irradiation. Heat treatment was adopted by Li et al. (2020) to elevate the hydrophobicity of chitosan/hordein/quercetin nanofiber films. The WCA angle of the nanofiber films was increased from 60° to 122° after heat treatment at 150 °C for 6 h. These three studies indicate the post-treatment on chitosan/flavonoid films can improve their physical properties. In the studies of Sutharsan et al. (2022) and Sutharsan et al. (2023), the researchers found the storage time (3 and 6 weeks) and storage temperature (4 and 21 °C) remarkably affected the mechanical and functional properties of chitosan/flavonol films, where the TS of the films increased but the EAB and antioxidant activity of the films decreased during storage. Meanwhile, the changes in the mechanical and antioxidant properties of the films were slowed down by low temperature (4 °C) storage. To sum up, the content and type of flavonoids, the encapsulation and modification of flavonoids, the presence of reinforcing agents, the pre-treatment of film-forming solutions, and the preparation method, post-treatment and storage conditions of the films are important factors affecting the physical and functional properties of chitosan/flavonoid films.

2.3. Release behavior and stability of the films

In practical application, chitosan/flavonoid films normally contact with foods, and flavonoids should be first released from the films and then exert antioxidant and antimicrobial activities. In general, a rapid release of flavonoids is often required for the short-term food storage while a slow release of flavonoids is necessary for the long-term food storage. Therefore, in order to evaluate the active packaging potential of chitosan/flavonoid films, it is very essential to investigate the release behavior of the films. So far, the release behavior of chitosan/flavonoid films are normally tested in different food simulants, such as distilled water (aqueous food simulant), 3 % acetic acid (acidic food simulant), 50 % ethanol (alcohol beverage simulant) and 95 % ethanol (fatty food simulant).

Several studies have revealed the release rate of flavonoids from chitosan/flavonoid films is influenced by the type of flavonoids. For example, Jacobs, Chambin, Debeaufort, and Benbettaieb (2023) compared the release behavior of three antioxidants (rutin, gallic acid and coumarin) from chitosan-based films into distilled water (aqueous food simulant). They found rutin had the lowest release rate, which was because rutin had the largest molecular size and the strongest interactions with chitosan. Meanwhile, the low solubility of rutin also restricted its diffusion into aqueous food simulant (Jacobs et al., 2023). In another study, Zhang et al. (2020) compared the release behavior of three flavonols (myricetin, quercetin and kaempferol) from chitosan-based films into 95 % ethanol (fatty food simulant) and distilled water (aqueous food simulant). They found the release rate of flavonols decreased in the order of kaempferol > quercetin > myricetin in fatty food simulant. However, an opposite trend was observed in aqueous

food simulant (Zhang et al., 2020). This study reveals the release of flavonols was not only influenced by the type of flavonols but also affected by the type of food simulant.

Encapsulation is an effective way to slow down the release of flavonols from chitosan/flavonoid films. Roy and Rhim (2021d) documented the release of quercetin from chitosan-based films was delayed after quercetin was encapsulated in sodium tripolyphosphate cross-linked chitosan nanoparticles. Similarly, Bi et al. (2021) also observed the migration of luteolin from chitosan films was inhibited by nano-emulsion encapsulation. Except for flavonols themselves, film matrix is an important factor affecting the release rate of flavonols. Liang et al. (2017) demonstrated the release rate of EGCG from chitosan/zein films was influenced by the weight ratio of film matrix, with the chitosan/zein (3/1, w/w) film having the highest release rate. The release behavior of flavonols from chitosan/flavonoid films is also affected by the presence of reinforcing agents. Wang et al. (2018) found the release of EGCG from chitosan-based films was retarded by adding nano-bacterial cellulose, which was ascribed to hydrogen bond interactions between EGCG and nano-bacterial cellulose.

The preparation method of chitosan/flavonoid films is a non-negligible factor affecting the release of flavonols. Ponnusamy et al. (2024) prepared bilayer films by covering chitosan/gelatin/EGCG on polylactic acid via solvent casting and electrospinning techniques. They found the bilayer film fabricated via electrospinning technique had relatively slower release rate of EGCG, which was because EGCG was encapsulated in nanofibers during electrospinning. The post-treatment of chitosan/flavonoid films has a big impact on the release behavior of flavonols. Benbettaieb et al. (2016) investigated the impact of electron beam irradiation on the release of quercetin from chitosan/gelatin films. They found electron beam irradiation treatment slowed down the release of quercetin, which was because irradiation treatment created cross-linking between film matrix and encapsulated quercetin in the matrix. Likewise, Sabaghi et al. (2020) demonstrated gamma irradiation decreased the release rate of catechin from chitosan/polyvinyl alcohol films. Based on above findings, it is concluded that the type and form of flavonols, the nature of film matrix, the presence of reinforcing agents, the preparation method and post-treatment of the films, and the type of food simulant can affect the release behavior of chitosan/flavonoid films.

Notably, although chitosan/flavonoid films have outstanding antioxidant and antimicrobial activities, their active packaging efficiency is seriously restricted by the poor solubility and stability of flavonoids. On one hand, most flavonoids are hydrophobic and have low compatibility with hydrophilic chitosan-based matrix. On the other hand, flavonoids are easily oxidized under natural conditions, resulting in functionality loss. So far, different means have been utilized to improve the solubility and stability of flavonoids in chitosan/flavonoid films (Fig. 3B).

Most existing studies have successfully improved the solubility and stability of flavonoids by encapsulating flavonoids in nano-sized carriers, such as nanoparticles (Roy & Rhim, 2021d), nanoemulsions (Bi et al., 2021), nanoliposomes (Ali et al., 2023), nanotubes (Roy & Rhim, 2021b), nanocapsules (Liang et al., 2017) and nanofibres (Li et al., 2020). The flavonoids encapsulated in nano-sized carriers often show antioxidant and antimicrobial activities in a sustained manner. In recent years, some researchers have managed to increase the solubility and stability of flavonoids by grafting them with polysaccharides, such as starch aldehyde (Sun et al., 2024; Yong, Xu, et al., 2022) and chitosan (Mittal et al., 2021; Torres et al., 2012; Wang et al., 2022; Yong et al., 2024). As compared with the films containing unmodified flavonoids, the films containing polysaccharide-flavonoid conjugates show better swelling ability, stability, and antioxidant and antibacterial activities (Sun et al., 2024). Meanwhile, the films containing polysaccharide-flavonoid conjugates normally have good homogeneity and storage stability. Recently, the formation of self-assemble flavonoid-nano metal chelate complex has been considered as a feasible way to increase the stability of flavonoids (Qi et al., 2023; Wang et al., 2024). The formed

flavonoid-nano metal chelate complex can be further adsorbed onto the layered clay nanosheets to achieve better stability (Wang et al., 2024). Nonetheless, the impact of flavonoid-nano metal chelate complex on the stability of the films has not been evaluated in existing studies (Qi et al., 2023; Wang et al., 2024), which should be deeply investigated in future. To increase the solubility of rutin, chitosan/rutin film-forming solutions were treated with a rotor-stator device (Lipatova et al., 2021). As a result, rutin particles were reduced to sub-micron size and well dispersed in chitosan solutions. This study reveals that mechanical activation can increase the homogeneity of chitosan/flavonoid film-forming solutions. In summary, the solubility and stability of flavonoids in chitosan/flavonoid films can be improved by the encapsulation, structural modification, nano metal chelation and mechanical activation of flavonoids (Fig. 3B).

2.4. Applications of the films

Nowadays, with increasing consumer demands for high-quality and safe foods, the researchers have managed to utilize chitosan/flavonoid films for active packaging. As summarized in Table 1, chitosan/flavonoid films have been widely used in the preservation of different food products, such as fruits (e.g., apple, citrus and litchi), vegetables (e.g., potato), aquatic products (e.g., sea bass), meat products (e.g., beef, chicken and pork), and edible oils (e.g., corn oil, rapeseed oil, soybean oil and sunflower seed oil).

To exert antioxidant and antimicrobial activities, chitosan/flavonoid films should directly contact with foods and sustainably release flavonoids from the films. In general, chitosan/flavonoid films have been used for active packaging in different manners (Fig. 3C). The first approach is to wrap or cover foods with chitosan/flavonoid films, where flavonoids can gradually migrate from the films into the foods. This method has been widely adopted by researchers (Ali et al., 2023; Mittal et al., 2021; Ponnusamy et al., 2024). The second approach is based on the edible coating of foods with chitosan/flavonoid film-forming solutions, forming a protective semi-permeable membrane around foods. The coated foods are often sealed in commercial plastic packaging boxes or bags (Fu et al., 2024; Guo et al., 2022; Qi et al., 2023; Sun et al., 2024). The third approach is to immerse chitosan/flavonoid film in liquid foods all the time and the flavonoids are gradually released from the films into the foods during storage (Jakubowska et al., 2023; Torres et al., 2012).

To evaluate the active packaging performance of chitosan/flavonoid films on different foods, the quality change of foods during storage should be thoroughly measured. For fruits and vegetables, it is essential to measure their physiological and biochemical indexes (e.g., weight loss, hardness, decay degree, pH, total soluble solid, titratable acid, conductivity, respiration rate and enzyme activity) and nutritional indexes (e.g., total polyphenol and ascorbic acid contents) (Fu et al., 2024; Qi et al., 2023; Sun et al., 2024). Chitosan-flavonoid films can extend the shelf life of fruits by retaining moisture, maintaining cell membrane integrity, delaying senescence and browning, reducing nutritional loss, and inhibiting microbial induced decay (Fu et al., 2024; Qi et al., 2023; Sun et al., 2024). For aquatic and meat products, their physiological and biochemical indexes (e.g., total viable count (TVC), texture, thiobarbituric acid reactive substance (TBARS), total volatile basic nitrogen (TVB-N) and free fatty acid) and sensory attributes (e.g., color, smell and consumer acceptance) are often determined (Guo et al., 2022). The protective effects of chitosan/flavonoid films on aquatic and meat products are mainly based on the inhibition of lipid/protein oxidation and microbial proliferation (Guo et al., 2022). As for edible oils, their oxidative degrees are often evaluated by measuring the formation of primary and secondary oxidation products, such as peroxide value, anisidine value, TBARS and fatty acid profile (Jakubowska et al., 2023; Ponnusamy et al., 2024; Wang et al., 2022; Yong et al., 2022; Yong et al., 2024). It is found that chitosan/flavonoid films can effectively inhibit the lipid oxidation of edible oils by retarding the progress of primary and

secondary oxidation (Jakubowska et al., 2023; Ponnusamy et al., 2024; Wang et al., 2022; Yong et al., 2022; Yong et al., 2024).

Notably, several problems should be solved before the commercial production and application of chitosan/flavonoid films. (1) There are several classes of flavonoids that can be used to produce active packaging films. However, existing studies have mainly focused on the films with limited flavonoids, especially quercetin, rutin and catechins. More experiments are needed to compare the physical and functional properties of the films containing different classes of flavonoids. Meanwhile, the relationship between the structure of flavonoids and the properties of chitosan/flavonoid films should be established. Notably, when selecting a certain flavonoid, its cost should be carefully considered. (2) The formula of chitosan/flavonoid films should be optimized. Except for chitosan and flavonoids, different kinds of polymers, cross-linking agents, plasticizers and reinforcing agents can be also added into the films to achieve better performances. These additives certainly have different impacts on the films, which should be deeply investigated. (3) Existing chitosan/flavonoid films are mainly prepared by solvent casting, which is not suitable for industrial production. More attention can be paid on more efficient film preparation methods, such as electrospinning and extrusion. Besides, the properties of the films prepared by different methods should be compared. (4) Especial attention should be paid to elevate the stability of flavonoids in the films. Among different stabilization techniques, encapsulation and structural modification are two effective methods that should be deeply investigated. For instance, the wall materials used to encapsulate flavonoids and the polysaccharides used to conjugate flavonoids need to be screened. Meanwhile, the stability of chitosan/flavonoid films during storage and application needs to be evaluated. (5) The application of chitosan/flavonoid films in active packaging has not received enough attention from the researchers. Since chitosan/flavonoid films directly contact with foods, the safety of the films and the residue of film components in foods should be carefully evaluated. The release behavior of chitosan/flavonoid films needs to be determined. In addition, the relationship between the properties and the preservation efficiency of chitosan/flavonoid films on different foods should be established. (6) Some new functional properties of chitosan/flavonoid films can be explored. For instance, viruses are responsible for a wide range of diseases (e.g., hepatitis and severe acute respiratory syndrome coronavirus 2) and have caused global food safety crises. Despite ongoing research, effective treatments for many foodborne viral infections remain elusive. As reported, flavonoids are phytochemicals with antiviral activity. These compounds act at multiple stages of viral infection, including viral entry, replication, and protein translation (Badshah et al., 2021). However, the

antiviral activity of chitosan/flavonoid films has not been evaluated yet.

3. Chitosan-based intelligent packaging films with anthocyanins

3.1. The structure and pH-responsive property of anthocyanins

Anthocyanins are the glycosylated products of anthocyanidins, a unique class of flavonoids (Fig. 1A). Like other classes of flavonoids, anthocyanins from different sources own certain antioxidant and antimicrobial activities that are useful for active packaging (Roy & Rhim, 2021a). Moreover, anthocyanins show a wide array of natural colors (e. g., red, purple and blue), which makes anthocyanins be suitable to fabricate intelligent packaging films (Roy & Rhim, 2021a). Up to date, more than 600 structurally different anthocyanins have been found in nature. Basically, anthocyanins are the glycosylated anthocyanidins. The most common anthocyanidins are cyanidin, delphinidin, malvidin, pelargonidin, peonidin and petunidin (Fig. 4A), whose structures are different in the pattern of methoxylation and hydroxylation (Roy & Rhim, 2021a). The color differences of natural anthocyanins are mainly attributed to different structured anthocyanidins, with pelargonidin presenting brick-red color, cyanidin and peonidin presenting purplish red color, and delphinidin, petunidin and malvidin presenting violet color (Roy & Rhim, 2021a). In some plants, anthocyanins are acylated by aliphatic or aromatic acids, which further contributes to the structural diversity of anthocyanins (Jokioja, Yang, & Linderborg, 2021). Acylation effectively reduces the polarity of anthocyanins and creates steric hindrance towards anthocyanins, thereby elevating the structural stability of anthocyanins (Zhao et al., 2017). Meanwhile, anthocyanidins and acyl groups within single acylated anthocyanin molecule can form intramolecular copigmentation, resulting in the hyperchromic effect on anthocyanins (Zhao et al., 2017).

Notably, anthocyanins are not only nature colorants but also pH-responsive color changeable substances. Anthocyanins can change their intrinsic colors when external pH status changes (Zhao et al., 2022). The pH-responsive color changeability makes anthocyanins be distinctive raw materials to construct intelligent packaging films (Roy & Rhim, 2021a). As displayed in Fig. 4B, the pH-responsive color changeable mechanisms of anthocyanins are associated with the structural transition of anthocyanins at different pHs. Different molecular forms of anthocyanins including flavylium cation, neutral quinone base, anionic quinoidal base, carbinol pseudobase and cis-/trans-chalcone are transformed with the variation of pH status, where hydration, proton transfer and isomerization reactions are involved in the transition processes (Xu et al., 2023). Since anthocyanins from different plants consist

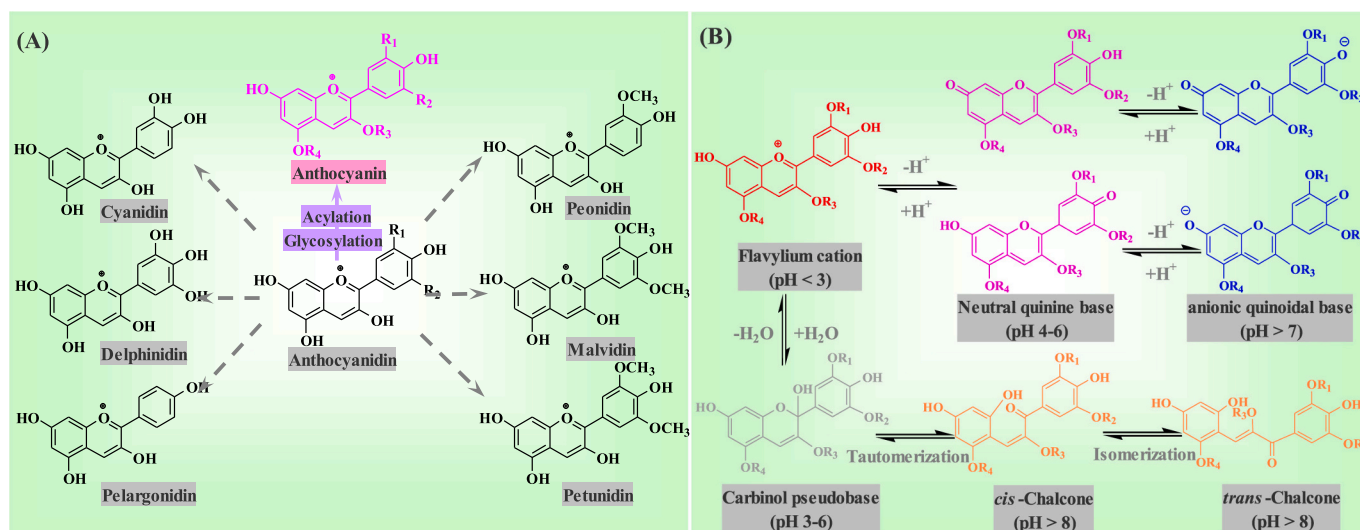


Fig. 4. The chemical structures of six common anthocyanidins (A) and the molecular basis for the pH-sensitive color changeability of anthocyanins (B).

of different structured anthocyanin monomers, the pH-responsive color changeability of anthocyanins greatly varies with their sources. Despite this, comparative studies on the pH-responsive color changeability of anthocyanins from different plants are very scarce (Cheng et al., 2022; Kan et al., 2022). It has been demonstrated that anthocyanins extracted from black wolfberry, red cabbage and purple sweet potato have better pH-responsive color changeability, because anthocyanins in these plants are highly acylated (Kan et al., 2022). In this regard, it is very necessary to screen the highly acylated anthocyanins from plant kingdom for preparing intelligent packaging films.

3.2. Preparation of the films

As summarized in Table 2 and Fig. S1, anthocyanins have been isolated from different fruits (blackberry, black chokeberry, black plum, black wolfberry, blood orange, blueberry, blue honeysuckle, elderberry, grape, hawthorn, jaboticaba, jambolan, karonda, mulberry, pancoli and raspberry), vegetables (black carrot, eggplant, onion, red cabbage, red radish and Yang-He), grains (black peanut, black rice, black soybean, purple corn, purple potato, purple rice, purple sweet potato and purple tomato) and flowers (blue portulaca, butterfly pudding, jacaranda, mallow, red poppy, rose and roselle) before they are used to fabricate chitosan-based intelligent packaging films.

Anthocyanins from various plants have been added into chitosan-based matrix to develop intelligent packaging films (Table 2). In several early studies, chitosan was often used alone as the film matrix. Nowadays, chitosan is frequently blended with other biodegradable polymers (e.g., alginate, Arabic gum, basil seed gum, carrageenan, cellulose, collagen, fucoidan, gelatin, glucomannan, hydroxypropyl methylcellulose, konjac methylcellulose, pectin, polyvinyl alcohol, starch, whey protein and zein) to yield more stable film matrix, based on the hydrogen bonds and/or electrostatic interactions between chitosan and other polymers (Roy & Rhim, 2021a). It is worth noting the proportion of chitosan and other polymers is a key factor affecting the properties of the films. However, this factor is often ignored by researchers. In most existing studies, a fixed chitosan/polymer proportion is selected casually. To find the optimal chitosan/polymer proportion, nano-laser particle size analyzer has been recently used to determine the particle size, polydispersity index and ζ -potential of film-forming solutions (Li et al., 2024). This study demonstrates the formation of small-sized polyelectrolyte complexes is beneficial to maintain the stability of hydrocolloid system, supplying a route to optimize the formula of multiple polymer matrix (Li et al., 2024). In some circumstances, cross-linking agents (e.g., sodium tripolyphosphate and citric acid) are added into film matrix to strengthen the interactions of chitosan and other polymers (Chen et al., 2024; Duan et al., 2022; Zhang et al., 2023). Except for anthocyanins and chitosan-based matrix, plasticizer (e.g., glycerol) is an essential element to produce intelligent packaging films with good elasticity. Interestingly, anthocyanins themselves are good plasticizers (Lu, Zhou, Yu, Chen, & Yuan, 2022).

In recent studies, different types of functional ingredients including metal nanoparticles (Cao et al., 2023; Li, Li, Wang, Sun, & Pei, 2023; Lu et al., 2022; Liu, Huang, Ying, Hu, & Hu, 2021; Luo, Xia, Liu, Ji, & Luo, 2022; Qin, Liu, Yuan, Yong, & Liu, 2019; Sun et al., 2020; Yi et al., 2024; Zhang et al., 2019; Zheng et al., 2023), nanoclay (Capello, Leandro, Gagliardi, & Valencia, 2021; Koosha & Hamed, 2019), nanocrystal (Chen, Yan, Huang, Zhou, & Hu, 2021; Fernández-Marín et al., 2022; Oun et al., 2022; Wu et al., 2019; Zheng et al., 2024), nanofiber (Wang et al., 2023), essential oils (Hao et al., 2023; Li et al., 2022; Rajendran et al., 2024; Yan et al., 2021; Zhang et al., 2023; Zhao et al., 2023), bacteriocin (Zhang et al., 2023) and protein hydrolysate (Ren et al., 2023; Zhang et al., 2023) have been added into chitosan/anthocyanin film-forming solutions to produce intelligent packaging films with elevated functionality (Table 2). The pH value of final film-forming solutions is often uncontrolled, which is because chitosan is often dissolved in acetic acid solution that benefits the stabilization of

anthocyanins.

As displayed in Fig. 5, several methods including casting, dip-coating, electrospinning and 3D-printing have been used to fabricate chitosan/anthocyanin films. Casting, the most widely used method, is based on the solvent evaporation of film-forming solution on a leveled plate under drying. Considering anthocyanins are heat-sensitive substances, the temperature should be seriously controlled during the drying process of the films. Although casting is simple and cost-saving, this method is hardly scaled-up and is much suitable for laboratory scale (Roy & Rhim, 2021a). Extrusion, the industrial scale-up film preparation method, has not been used to fabricate chitosan-based intelligent packaging films till now. This is because the high temperature (nearly 200 °C) applied in extrusion has a decomposition effect on anthocyanins (Almasi, Forghani, & Moradi, 2022). Similar with casting, dip-coating is a convenient approach to prepare intelligent packaging films. This method is based on the penetration and adsorption of anthocyanins onto cellulose-based filter paper, where filter paper serves as the solid support for anthocyanins (Tirtashi et al., 2019). Since anthocyanins in the solution are not totally absorbed onto filter paper, the loading amount of anthocyanins in the films cannot be accurately controlled. Meanwhile, the films developed by dip-coating method is not transparent. Electrospinning is a novel technique to prepare anthocyanin-entrapped nanofibers for intelligent packaging use (Zhang et al., 2020). During electrospinning process, chitosan-based polymeric solutions containing anthocyanins are stretched under high voltage and the solvent is evaporated simultaneously to form ultra-fine nanofibers on a metal collector. The prepared nanofiber films are well known for their large specific surface areas and high porosity. Notably, anthocyanins can be effectively entrapped in nanofibers to avoid the decomposition of anthocyanins (Huang et al., 2023; Shavisi, 2024; Shavisi & Shahbazi, 2022). Meanwhile, due to their high porosity, electrospun nanofiber films with anthocyanins have good pH-responsive color changeability. Despite this, as compare with the films prepared by casting, the electrospun fiber films have low barrier and mechanical properties (Forghani et al., 2022). 3D-printing is an emerging platform to fabricate intelligent packaging films via continuous physical stacking. During 3D-printing, the film-forming solution is loaded into a cylinder, extruded from the nozzle under the pressure of air flow, and then deposited onto the plat-form layer by layer (Li et al., 2022). The biggest merit of 3D-printing is the personalized design for sample shapes and patterns, where the loading amount of anthocyanins can be accurately controlled. However, 3D-printing is not suitable to produce intelligent packaging films in an industrial scale.

The above mentioned methods are normally used alone to prepare monolayer films. Moreover, these methods can be used alone or combined used to produce multiple layer films. The main purpose to develop multiple layer films is to supply one or more protective layers for anthocyanins. In most cases, solvent casting method has been used to prepare bilayer films. For example, Zhang et al. (2023) developed a bilayer film with konjac glucomannan/elderberry anthocyanin as the indicator layer and chitosan/oregano essential oil as the hydrophobic and antibacterial layer. Likewise, bilayer films were prepared through solvent casting by adding carbon dots (Liu et al., 2024) and nano TiO₂ (Cao et al., 2023) in the protective layers. The protective agents can not only protect anthocyanins from light-induced degradation but also greatly elevate the antioxidant and/or antimicrobial activity of the films. Except for solvent casting method, other methods have been also used to prepare bilayer films. For example, Huang et al. (2023) prepared a bilayer film consisted of zein/gelatin/carvacrol antibacterial layer and chitosan/polyvinyl alcohol/blueberry anthocyanin indicator layer through electrospinning technique. The antibacterial layer provided a hydrophobic barrier for anthocyanin in the indicator layer. Another bilayer film was prepared by Luo et al. (2022) using two different film preparation methods. First, an antibacterial layer was prepared by casting chitosan/hydroxyethyl cellulose/nano TiO₂ solution on glass plate. Then, an indicator layer was prepared by stacking

Table 2.

Formulations, physical and functional properties, and applications of packaging films based on chitosan and anthocyanins from different plants.

Source of anthocyanins	Film matrix	Reinforcing agents	Film preparation method	Impact of anthocyanins on the physical properties of the films	Functional properties of the films	Factors affecting the properties of films	Application of the films	References
Arabica coffee (<i>Coffea arabica</i> L.)	Chitosan/sodium alginate blend		Solvent casting	WS↓, WVP↓, LT↓, TS↑, EAB↓	pH-sensitivity	The content of anthocyanins	Monitoring beef freshness	Hu et al., 2022
Blackberry (<i>Vaccinium corymbosum</i> L.)	Chitosan		Solvent casting	MS↑, WVP↓, OP↑, TS↑, EAB↓	Ferric reducing antioxidant power pH-sensitivity	The source of anthocyanins and the content of anthocyanins		Kurek et al., 2018
Black carrot (<i>Daucus carota</i> L.)	Chitosan/polyvinyl alcohol blend	Bentonite nanoclays	Solvent casting	WVP↔, LT↓, TS↑	Antioxidant activity (DPPH radical scavenging activity) Antimicrobial activity against <i>S. aureus</i> , <i>E. coli</i> and <i>P. aeruginosa</i> pH-sensitivity	The presence of bentonite nanoclays		Koosha & Hamed, 2019
	Chitosan/cellulose blend		Dip-coating	SR↑, WS↑	pH-sensitivity		Monitoring milk freshness	Tirtashi et al., 2019
Black chokeberry (<i>Aronia melanocarpa</i>)	Chitosan		Solvent casting	WS↓, SR↓, TS↑, EAB↓	pH-sensitivity	The content of anthocyanins		Halász & Csóka, 2018
	Chitosan/polyvinyl alcohol blend		Solvent casting	MC↓, SR↓, WVP↓, LT↓, TS↑, EAB↑	DPPH scavenging activity Antimicrobial activity against <i>E. coli</i> , <i>P. aeruginosa</i> and <i>S. aureus</i>	The content of anthocyanins	Preserving and monitoring shrimp freshness	Long et al., 2024
	Chitosan/polyvinyl alcohol blend	Cellulose nanocrystals and grapefruit seed extract	Solvent casting	WVP↑, TS↑, EAB↑	DPPH and ABTS radical scavenging activity Antimicrobial activity against <i>E. coli</i> and <i>L. monocytogenes</i> pH-sensitivity	The presence of cellulose nanocrystals and grapefruit seed extract		Oun et al., 2022
Black peanut (<i>Arachis hypogaea</i> L.)	Chitosan/gelatin blend	Nano ZnO	Solvent casting	MC↓, WS↓, WVP↓, LT↓, TS↑, EAB↑	DPPH radical scavenging activity Antimicrobial activity against <i>S. aureus</i> and <i>E. coli</i> pH-sensitivity	The content of anthocyanins and the presence of nano ZnO	Monitoring shrimp freshness	Lu et al., 2022
Black plum (<i>Prunus salicina</i> Lindl.)	Chitosan	Nano TiO ₂	Solvent casting	MC↓, WS↑, WVP↓, LT↓, TS↑, EAB↑	DPPH radical scavenging activity Antimicrobial activity against <i>S. aureus</i> , <i>E. coli</i> , <i>Salmonella</i> , <i>L. monocytogenes</i> pH-sensitivity Ethylene scavenging ability	The presence of nano TiO ₂		Zhang et al., 2019
Black rice (<i>Oryza sativa</i> L.)	Chitosan	Oxidized chitin nanocrystals	Solvent casting	WVP↑, OP↓, LT↓, TS↓, EAB↑	DPPH radical scavenging activity pH-sensitivity	The content of anthocyanins and the presence of oxidized chitin nanocrystals	Monitoring shrimp and fish freshness	Wu et al., 2019
	Chitosan		Solvent casting	MC↓, WVP↓, TS↑, EAB↑	DPPH radical scavenging activity pH-sensitivity	The variety of plant and the content of anthocyanins	Monitoring pork freshness	Yong et al., 2019
	Chitosan	Oregano essential oil	Solvent casting	MC↓, WS↑, WVP↔, WCA↓, TS↓, EAB↑	DPPH radical scavenging activity Antimicrobial	The content of anthocyanins and the presence of	Monitoring pork freshness	Hao et al., 2023

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Table 2. (continued)

Source of anthocyanins	Film matrix	Reinforcing agents	Film preparation method	Impact of anthocyanins on the physical properties of the films	Functional properties of the films	Factors affecting the properties of films	Application of the films	References
	Chitosan/pectin blend		Solvent casting	TS↑	activity against <i>S. aureus</i> and <i>E. coli</i> pH-sensitivity and ammonia-sensitivity	oregano essential oil The content of anthocyanins	Monitoring pork and beef freshness	Zeng, Ye, Liu, & Fei, 2023
Black soybean (<i>Glycine max</i> (L.) Merr.)	Chitosan		Solvent casting	MC↓, WS↑, WVP↓, LT↓, TS↑, EAB↑	DPPH radical scavenging activity pH-sensitivity	The content of anthocyanins		Wang et al., 2019
Black wolfberry (<i>Lycium ruthenicum</i> Murr.)	Chitosan/whey protein blend sodium tripolyphosphate Chitosan		Solvent casting	MC↔, WCA↑, LT↓, TS↑, EAB↑	Ammonia-sensitivity		Monitoring shrimp and fish freshness	Chen et al., 2024
		Nano ZnO	Solvent casting	MC↑, TS↑, EAB↓, WCA↔	DPPH radical scavenging activity Antibacterial activity against <i>E. coli</i>	The source of anthocyanins and the presence of nano ZnO	Monitoring pork freshness	Li et al., 2023
Blood orange (<i>Citrus sinensis</i> (L.) Osbeck)	Chitosan/Arabic gum blend	Thyme oil	Solvent casting	MC↔, WS↔, WCA↓, WVP↔, LT↓, TS↑, EAB↑	pH-sensitivity ABTS and DPPH radical scavenging activity Antimicrobial activity against <i>S. aureus</i> and <i>E. coli</i> , pH-sensitivity	The content of anthocyanins and the presence of thyme oil	Preserving and monitoring milk freshness	Zhao et al., 2023
Blueberry (<i>Rubus fruticosus</i>)	Chitosan		Solvent casting	MS↑, WVP↓, OP↑, TS↑, EAB↓	Ferric reducing antioxidant power pH-sensitivity	The source of anthocyanins and the content of anthocyanins		Kurek et al., 2018
	Chitosan	Cellulose nanocrystal	Solvent casting	MC↑, WS↑, SR↑, WVP↓, OP↓, LT↓, TS↓, EAB↑	DPPH scavenging activity, Antimicrobial activity against <i>E. coli</i> and <i>S. aureus</i>	The presence of cellulose nanocrystal	Preserving and monitoring shrimp freshness	Zheng et al., 2024
Blueberry (<i>Vaccinium</i> spp.)	Chitosan/polyvinyl alcohol and zein/gelatin bilayer matrix	Carvacrol	Electrospinning		pH-sensitivity Antimicrobial activity against <i>S. aureus</i> , <i>E. coli</i> , <i>L. monocytogenes</i> and <i>S. enterica</i> , Ammonia-sensitivity	The other layer	Preserving and monitoring fish freshness	Huang et al., 2023
	Chitosan/sodium alginate bilayer matrix	Nano TiO ₂	Solvent casting (bilayer film)	WVP↓, LT↓, TS↑, EAB↑	DPPH scavenging activity, Antimicrobial activity against <i>E. coli</i> and <i>S. aureus</i>	The other layer	Preserving and monitoring pork freshness	Cao et al., 2023
Blue honeysuckle (<i>Lonicera caerulea</i>)	Chitosan/potato starch blend		Solvent casting		Ammonia-sensitivity	The pH of film-forming solution	Monitoring shrimp freshness	Li et al., 2022
	Chitosan/polyvinyl alcohol blend		Solvent casting	MC↓, WVP↓, LT↓, TS↑, EAB↑	ABTS and DPPH scavenging activity pH-sensitivity	The encapsulation of anthocyanins	Preserving pork	Li et al., 2024
Blue porterweed (<i>Stachytarpheta jamaicensis</i>)	Chitosan/polyvinyl alcohol blend		Solvent casting	MC↓, WS↑, WVP↓, OP↓, LT↓, TS↑, EAB↓	DPPH radical scavenging activity pH-sensitivity	The content of anthocyanins	Monitoring chicken freshness	Amaregouda & Kamanna, 2023
Butterfly pudding (<i>Clitoria ternatea</i> L.)	Chitosan	Cinnamon essential oil	Solvent casting	MC↓, SR↓, WCA↓, TS↑, EAB↑, WVP↑	pH-sensitivity	The content of anthocyanins	Monitoring fish freshness	Yan et al., 2021
	Chitosan/gelatin blend		Solvent casting	MC↓, WS↓, WVP↔, WCA↓, LT↓, TS↓, EAB↑	pH-sensitivity and ammonia-sensitivity	The pH of film-forming solution	Monitoring beef freshness	Li et al., 2024
	Chitosan/polyvinyl alcohol blend		Solvent casting	MC↔, WS↓, SR↓, WVP↑	DPPH and ABTS radical scavenging	The source of anthocyanins	Monitoring beverage (fruit juice)	Singh et al., 2021

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Table 2. (continued)

Source of anthocyanins	Film matrix	Reinforcing agents	Film preparation method	Impact of anthocyanins on the physical properties of the films	Functional properties of the films	Factors affecting the properties of films	Application of the films	References
Coleus (<i>Plectranthus scutellarioides</i>)	Chitosan/fucoidan blend		Solvent casting	WCA↑, LT↓, TS↓, EAB↑	activity Ammonia-sensitivity	The content of anthocyanins	and milk) freshness	Wang et al., 2023
				MC↓, WS↓, WVP↓, WCA↑, OP↓, LT↓, TS↑, EAB↑	DPPH and ABTS radical scavenging activity Antimicrobial activity against <i>E. coli</i> and <i>S. aureus</i> pH-sensitivity		Monitoring fish freshness	
Eggplant (<i>Solanum melongena</i> L.)	Chitosan	Laponite® nanoclay	Solvent casting	MC↔, WS↓, WVP↔, WCA↔	pH-sensitivity	The presence of Laponite® nanoclay	Monitoring beef freshness	Capello et al., 2021
	Chitosan	Esterified chitin nanofibers	Solvent casting	MC↓, WS↓, WVP↓, WCA↑, OP↓, LT↓, TS↑, EAB↑	DPPH and ABTS radical scavenging activity Antimicrobial activity against <i>E. coli</i> and <i>S. aureus</i> pH-sensitivity and ammonia-sensitivity	The content of anthocyanins and the presence of esterified chitin nanofibers	Monitoring pork freshness	Wang et al., 2023
	Chitosan		Solvent casting	MC↓, SR↓, WVP↓, LT↓, TS↑, EAB↑	DPPH radical scavenging activity pH-sensitivity	The variety of plant and the content of anthocyanins	Monitoring milk freshness	Yong et al., 2019
Elderberry (<i>Sambucus nigra</i> L.)	Chitosan/konjac glucomannan bilayer matrix	Oregano essential oil	Solvent casting	MC↔, WS↔, WVP↔, WCA↑, LT↓, TS↑, EAB↑	DPPH radical scavenging activity Antimicrobial activity against <i>E. coli</i> and <i>S. aureus</i> pH-sensitivity		Preserving cheese	Zhang et al., 2023
Grape (<i>Vitis</i> spp.)	Chitosan/polyvinyl alcohol blend		Solvent casting		pH-sensitivity and ammonia-sensitivity	The pH of film-forming solution		Xiang et al., 2024
	Chitosan/sodium alginate blend, Gelatin/sodium alginate blend		Solvent casting	WCA↓	pH-sensitivity	The nature of matrix	Monitoring fish freshness	de Azevedo & Noreña, 2024
Hawthorn (<i>Crataegus scabrifolia</i>)	Chitosan/gelatin blend	Nanocellulose	Solvent casting	MC↓, SR↓, WVP↑, LT↓, TS↑, EAB↑	pH-sensitivity	The content of anthocyanins	Monitoring shrimp freshness	Yan et al., 2021
Jacaranda (<i>Jacaranda cuspidifolia</i>)	Chitosan/polyvinyl alcohol blend		Solvent casting	MC↓, WS↑, WCA↑, WVP↓, OP↓, LT↓, TS↑, EAB↑	DPPH radical scavenging activity Antimicrobial activity against <i>S. aureus</i> and <i>E. coli</i> pH-sensitivity	The content of anthocyanins	Monitoring fish freshness	Amaregouda, Kamanna, & Gasti, 2022
Jambolan (<i>Syzygium cumini</i>)	Chitosan/polyvinyl alcohol blend		Solvent casting	MC↔, WS↑, WAC↑, Opacity↓	pH-sensitivity	The content of anthocyanins	Monitoring shrimp freshness	Merz et al., 2020
Karonda (<i>Carissa carandas</i> L.)	Chitosan/polyvinyl alcohol blend		Solvent casting	MC↔, WS↓, SR↓, WVP↑, WCA↑, LT↓, TS↓, EAB↑	DPPH and ABTS radical scavenging activity Ammonia-sensitivity pH-sensitivity	The source of anthocyanins	Monitoring beverage (fruit juice and milk) freshness	Singh et al., 2021
Mallow (<i>Malva sylvestris</i>)	Chitosan/carrageenan blend		Electrospinning	MC↓, WS↓, WVP↓, OP↓, TS↓, EAB↑	pH-sensitivity		Monitoring fish freshness	Shavisi, 2024
Mulberry (<i>Morus</i> sp.)	Chitosan/hydroxyethyl cellulose blend	Nano TiO ₂	3D-printing (bilayer film)	WVP↓, TS↑, EAB↑	Antimicrobial activity against <i>S. aureus</i> and <i>E. coli</i>	The filling rate of 3D-printing	Monitoring litchi freshness	Luo et al., 2022
	Chitosan	Lemongrass essential oil	3D printing	WVP↓, OP↔, TS↔, EAB↓	DPPH radical scavenging activity Antimicrobial activity against <i>S. aureus</i> and <i>E. coli</i>	The presence of lemongrass essential oil	Monitoring pork freshness	Li et al., 2022

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Table 2. (continued)

Source of anthocyanins	Film matrix	Reinforcing agents	Film preparation method	Impact of anthocyanins on the physical properties of the films	Functional properties of the films	Factors affecting the properties of films	Application of the films	References
	Chitosan/konjac glucomannan blend	Nano ZnO	Solvent casting	WVP↓, LT↓, TS↔, EAB↓	DPPH radical scavenging activity Antimicrobial activity against <i>S. aureus</i> and <i>E. coli</i>	The content of anthocyanins and the presence of nano ZnO		Sun et al., 2020
	Chitosan/collagen blend	Nano ZnO	Solvent casting	WVP↔, LT↓, TS↑, EAB↔	pH-sensitivity DPPH radical scavenging activity pH-sensitivity	The content of anthocyanins and the presence of nano ZnO	Monitoring pork freshness	Zheng et al., 2023
Onion (<i>Allium cepa</i> L.)	Chitosan	Cinnamon essential oil	Solvent casting	MC↑, WS↑, SR↓, LT↓, TS↑, EAB↓	ABTS and DPPH scavenging activity Antimicrobial activity against <i>E. coli</i>			Rajendran et al., 2024
Pancoli (<i>Phyllanthus reticulatus</i>)	Chitosan/methylcellulose blend		Solvent casting	MC↓, WS↓, WVP↓, WCA↑, LT↓, TS↑, EAB↓	DPPH scavenging activity Antimicrobial activity against <i>E. coli</i> and <i>S. aureus</i>	The content of anthocyanins	Monitoring pork freshness	Gasti et al., 2021
Purple corn (<i>Zea mays</i> L.)	Chitosan	Nano Ag	Solvent casting	MC↓, WS↑, WVP↓, LT↓, TS↑, EAB↓	pH-sensitivity DPPH radical scavenging activity Antimicrobial activity against <i>S. aureus</i> , <i>E. coli</i> , <i>L. monocytogenes</i> and <i>S. typhimurium</i>	The content of anthocyanins and the presence of nano Ag		Qin et al., 2019
Purple potato (<i>Solanum tuberosum</i> L.)	Chitosan/polyvinyl alcohol blend	Nano ZnO	Solvent casting	MC↓, WVP↔, LT↓, TS↑, EAB↓	Antimicrobial activity against <i>S. aureus</i> and <i>E. coli</i>	The source and content of anthocyanins in the films, and the presence of nano ZnO	Monitoring shrimp freshness	Liu et al., 2021
Purple rice (<i>Oryza sativa</i> L.)	Chitosan		Solvent casting	MC↓, WVP↓, TS↑, EAB↑	DPPH radical scavenging activity pH-sensitivity	The variety of plant and the content of anthocyanins	Monitoring pork freshness	Yong et al., 2019
Purple sweet potato (<i>Ipomoea batatas</i> L.)	Chitosan		Solvent casting	MC↓, WS↑, WVP↓, LT↓, TS↓, EAB↓	DPPH radical scavenging activity pH-sensitivity	The content of anthocyanins		Yong et al., 2019
	Chitosan/polyvinyl alcohol, locust bean gum/polyvinyl alcohol, κ-carrageenan/polyvinyl alcohol		Solvent casting	WVP↓, LT↓, TS↓, EAB↓	DPPH radical scavenging activity pH-sensitivity and ammonia-sensitivity	The source of anthocyanins and the nature of matrix	Monitoring shrimp freshness	Yong et al., 2022
	Chitosan/hydroxypropyl methylcellulose blend	ε-Polylysine blend	Solvent casting	MC↓, WVP↑, LT↓, TS↑, EAB↑	Antimicrobial activity against <i>S. aureus</i> and <i>E. coli</i>	The presence of ε-polylysine	Monitoring fish freshness	Pang et al., 2023
	Chitosan/polyvinyl alcohol blend		Solvent casting	MC↔, WS↔, WCA↓, LT↓	pH-sensitivity	The source of anthocyanins	Monitoring beef freshness	Capello et al., 2021
Purple tomato (<i>Solanum lycopersicum</i> L.)	Chitosan	Nano ZnO	Solvent casting	MC↑, TS↑, EAB↓, WCA↔	DPPH radical scavenging activity Antibacterial activity against <i>E. coli</i>	The source of anthocyanins and the presence of nano ZnO	Monitoring pork freshness	Li et al., 2023
	Chitosan		Solvent casting	SR↑, TS↓, EAB↑	pH-sensitivity	The content of anthocyanins	Monitoring milk and fish freshness	Li et al., 2021

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Table 2. (continued)

Source of anthocyanins	Film matrix	Reinforcing agents	Film preparation method	Impact of anthocyanins on the physical properties of the films	Functional properties of the films	Factors affecting the properties of films	Application of the films	References
Raspberry (<i>Rubus idaeus</i> L.)	Chitosan/starch/gelatin blend	Curcumin	Solvent casting	MC↑, WS↑, SR↓, WVP↑, LT↓, TS↑, EAB↑	ABTS radical scavenging activity	The presence of other colorants (curcumin)	Monitoring chicken freshness	Duan et al., 2022
Red cabbage (<i>Brassica oleracea</i> L.)	Chitosan/gelatin blend	Bacteriocin CHQS	Solvent casting	WS↓, SR↓, WVP↓, LT↓, TS↔, EAB↔	ABTS and DPPH scavenging activity Antimicrobial activity against <i>B. subtilis</i> , <i>S. enterica</i> and <i>E. coli</i>	The content of citric acid and the presence of bacteriocin		Zhang et al., 2023
	Chitosan	Oxidized-chitin nanocrystals	Solvent casting	WVP↓, OP↓, LT↓, TS↑, EAB↑	DPPH radical scavenging activity pH-sensitivity and ammonia-sensitivity	The content of anthocyanins	Monitoring fish and shrimp freshness	Chen et al., 2021
	Chitosan/basil seed gum blend		Solvent casting	MC↓, WS↑, WVP↑, TS↓, EAB↑	DPPH radical scavenging activity Antimicrobial activity against <i>S. aureus</i> and <i>E. coli</i>	The content of anthocyanins	Monitoring fish freshness	Nadi, Razavi, & Shahrapour, 2023
	Chitosan	Chitin nanocrystals and curcuma oil	Solvent casting	MC↓, WS↓, WCA↑, LT↓, TS↑, EAB↓	DPPH radical scavenging activity pH-sensitivity and ammonia-sensitivity	The content of anthocyanins and the presence of curcuma oil		Fernández-Marín et al., 2022
	Chitosan	Collagen hydrolysate	Solvent casting	WS↑, WVP↑, LT↓, TS↔, EAB↑	DPPH radical scavenging activity Antimicrobial activity against <i>S. aureus</i> and <i>E. coli</i>	The content of anthocyanins	Monitoring shrimp freshness	Ren et al., 2023
Red poppy (<i>Papaver rhoeas</i> L.)	Chitosan/polyvinyl alcohol, locust bean gum/polyvinyl alcohol, κ-carrageenan/polyvinyl alcohol		Solvent casting	WVP↓, LT↓, TS↓, EAB↓	DPPH radical scavenging activity pH-sensitivity and ammonia-sensitivity	The source of anthocyanins and the nature of matrix	Monitoring shrimp freshness	Yong et al., 2022
	Chitosan		Solvent casting	WVP↔, TS↔, EAB↓	DPPH radical scavenging activity Ammonia-sensitivity		Monitoring shrimp freshness	Tavassoli, Khezerlou, Firoozy, Ehsani, & Punia Bangar, 2023
Red radish (<i>Raphanus sativus</i> L.)	Chitosan/zein blend		Solvent casting	SR↓, WVP↓, WCA↑, OP↓, TS↑, EAB↓	pH-sensitivity and ammonia-sensitivity	The content of anthocyanins	Monitoring mushroom freshness	Yi et al., 2023
	Chitosan/zein blend	Nano TiO ₂	Solvent casting	SR↓, WCA↑, LT↓, TS↑, EAB↓	pH-sensitivity and ammonia-sensitivity	The presence of nano TiO ₂	Monitoring mushroom freshness	Yi et al., 2024
Riceberry (<i>Oryza sativa</i> L.)	Chitosan		Solvent casting	WS↓, SR↓, WCA↑, WVP↓, LT↓, TS↑, EAB↑	DPPH and ABTS radical scavenging activity pH-sensitivity and ammonia-sensitivity	The content of anthocyanins	Monitoring shrimp freshness	Eze, Jayeoye, & Singh, 2022
Rose (<i>Rosa damascena</i> Mill.)	Chitosan/Arabic gum blend		Electrospinning	MC↓, WS↓, TS↓, EAB↑, WVP↓	DPPH radical scavenging activity pH-sensitivity and ammonia-sensitivity		Monitoring chicken breast freshness	Shavisi & Shahbazi, 2022

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Table 2. (continued)

Source of anthocyanins	Film matrix	Reinforcing agents	Film preparation method	Impact of anthocyanins on the physical properties of the films	Functional properties of the films	Factors affecting the properties of films	Application of the films	References
	Chitosan/starch blend		Solvent casting	WVP↓, WCA↑, LT↓, TS↑, EAB↓	pH-sensitivity and ammonia-sensitivity	The encapsulation of anthocyanins	Monitoring shrimp freshness	Zheng et al., 2023
Roselle (<i>Hibiscus sabdariffa</i> L.)	Chitosan		Solvent casting	MC↓, SR↓, WVP↓, WCA↓, LT↓, TS↑, EAB↓	pH-sensitivity and ammonia-sensitivity		Monitoring fish freshness	Khezerlou, Tavassoli, Alizadeh Sani, Ehsani, & McClements, 2023
	Chitosan/polyvinyl alcohol blend	Nano ZnO	Solvent casting	MC↓, WVP↔, LT↓, TS↑, EAB↓	Antimicrobial activity against <i>S. aureus</i> and <i>E. coli</i>	The source and content of anthocyanins in the films, and the presence of nano ZnO	Monitoring shrimp freshness	Liu et al., 2021
	Chitosan		Solvent casting	LT↓, MC↑, WS↑	pH-sensitivity	The nature of film matrix		Peralta et al., 2019
	Chitosan/polyvinyl alcohol blend, Chitosan/starch blend		Solvent casting		DPPH radical scavenging activity	The nature of film matrix	Monitoring pork freshness	Zhang et al., 2019
Yang-He (<i>Zingiber striolatum</i> Diels)	Chitosan/gelatin blend		Solvent casting	MC↔, LT↓, TS↓, EAB↑	Ammonia-sensitivity	The content of anthocyanins	Monitoring fish freshness	Qin et al., 2024

ABTS, 2,2'-azino-bis(3-ethylbenzothiazoline-6-sulfonate); DPPH, 2,2-diphenyl-1-picrylhydrazyl; EAB, elongation at break; LT, light transmittance; MC, moisture content; OP, oxygen permeability; SR, swelling ratio; TS, tensile strength; WCA, water contact angle; WS, water solubility; WVP, water vapor permeability; ↑, increased after anthocyanin addition; ↓, decreased after anthocyanin addition; ↔, unchanged after anthocyanin addition.

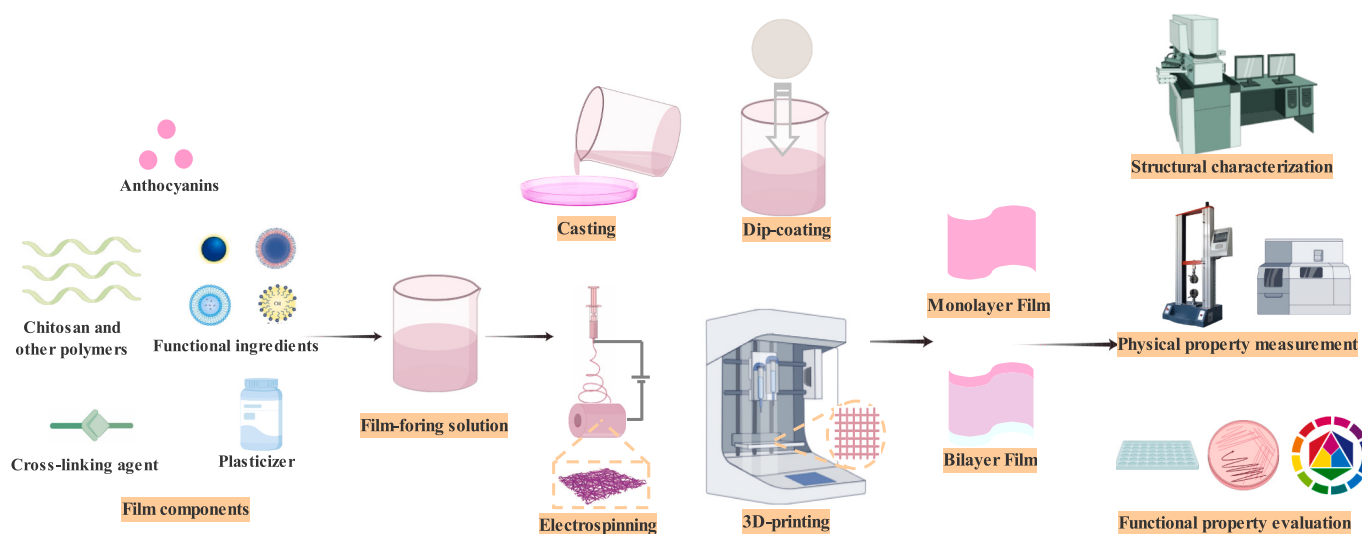


Fig. 5. The scheme illustrating the procedure for the preparation and characterization of chitosan/anthocyanin intelligent packaging films.

chitosan/hydroxyethyl cellulose/mulberry anthocyanin on the anti-bacterial layer through 3D-printing. A recent trend is to prepare triple layer film by embedding the indicator layer in the inner. Li et al. (2022) fabricated a sandwich-like triple layer intelligent packaging film, where the chitosan/lemongrass essential oil/mulberry anthocyanin indicator layer was prepared by 3D-printing and then heat-sealed between two pieces of starch layers. The starch layers were made by solvent casting. Although the indicator layer was embedded in the inner, it still maintained the good color changeability (Li et al., 2022).

3.3. Physical and functional properties of the films

The prepared chitosan/anthocyanin intelligent packaging films are often balanced under an environment with medium relative humidity

and then characterized for their physical and functional properties. The physical properties of chitosan/anthocyanin intelligent packaging films include optical (color), water resistant (e.g., MC, SR, WS and WCA), mechanical (TS, EAB and EM), barrier (LT, WVP and OP) and thermal properties (TGA and DSC), which are the same as the physical properties of chitosan/flavonoid active packaging films. As listed in Table 2, anthocyanins are able to change the physical properties of chitosan-based packaging films, which can be explained by the following reasons. First, anthocyanins are colorants that can make chitosan-based films show vibrant colors and light barrier ability. Secondly, anthocyanins are highly hydrophilic substances that can alter the water resistance of chitosan-based packaging films. Finally, anthocyanins embedded in chitosan-based film matrix can alter the inner compactness of the films. Meanwhile, anthocyanins can interact with chitosan and other polymers

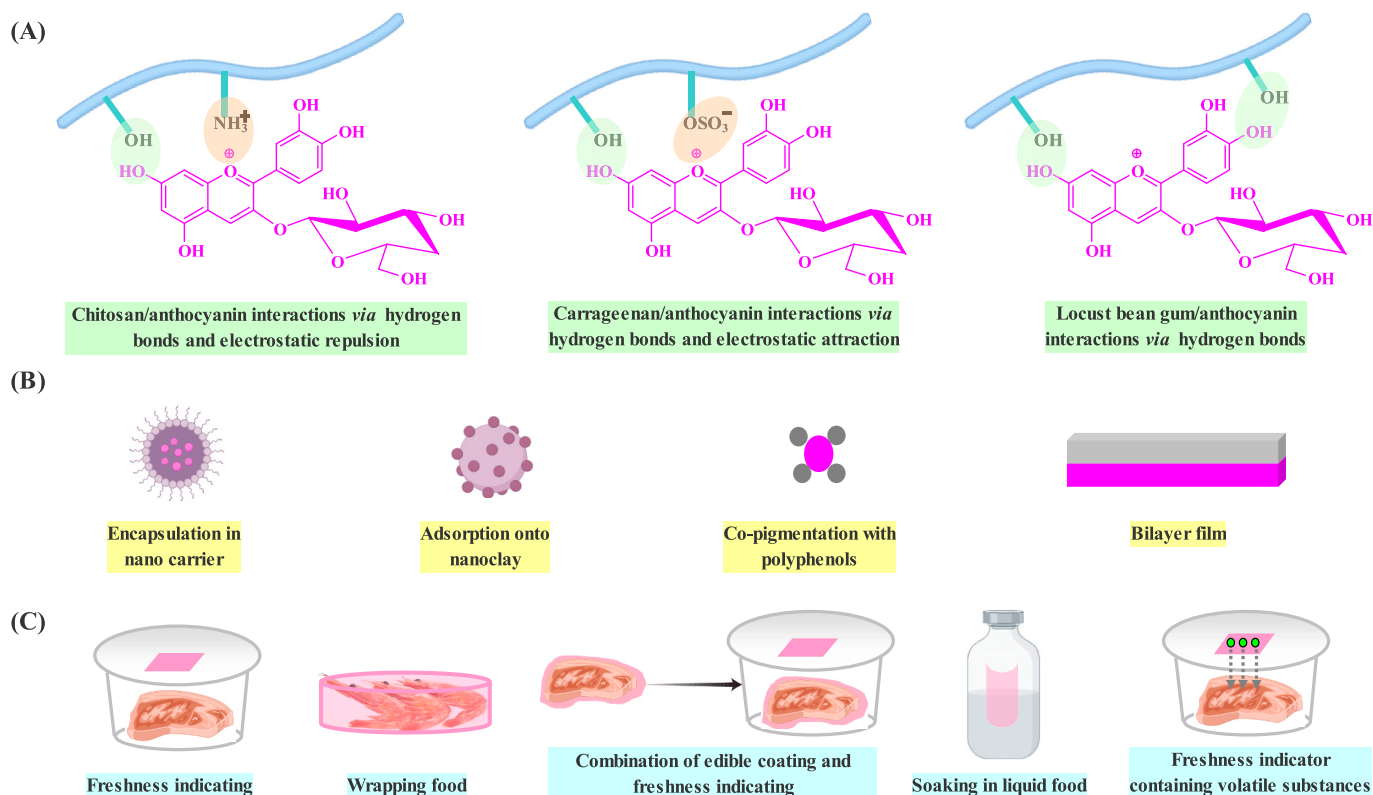


Fig. 6. The diagrams showing the intermolecular chitosan/anthocyanin interactions, carrageenan/anthocyanin interactions and locust bean gum/anthocyanin interactions (A), the strategies to enhance the stability and solubility of anthocyanins in chitosan/anthocyanin intelligent packaging films (B), and the application forms of chitosan/anthocyanin intelligent packaging films (C).

through hydrogen bond and/or electrostatic interactions (Fig. 6A), thereby changing the arrangement of chitosan-based polymers. As a result, the optical, mechanical, barrier and thermal properties of the films are changed by anthocyanins. Except for anthocyanins, the presence of other polymers can also affect the physical properties of chitosan/anthocyanin films, which is attributed to the intermolecular interactions between chitosan and other polymers (Li et al., 2024). Notably, reinforcing agents have big impacts on the physical properties of chitosan/anthocyanin films. For instance, the cross-linking agents and nano fillers can greatly increase the mechanical and barrier properties of chitosan/anthocyanin films (Chen et al., 2024; Duan et al., 2022; Zhang et al., 2023). Essential oils can increase the water resistance of chitosan/anthocyanin films (Hao et al., 2023; Li et al., 2022; Rajendran et al., 2024; Yan et al., 2021). In addition, bilayer films often display elevated mechanical and barrier properties than monolayer films (Cao et al., 2023; Huang et al., 2023; Zhang et al., 2023).

As compared with physical properties, the functional properties of chitosan/anthocyanin films including antioxidant activity, antimicrobial activity and color changeability have attracted more attention. Since anthocyanins are antioxidant and antimicrobial agents, chitosan/anthocyanin films present DPPH and ABTS radical scavenging activity and antimicrobial activity (Table 2). Despite this, the antioxidant and antimicrobial activities of chitosan/anthocyanin films are incomparable to those of other chitosan/flavonoid films. So, many researchers have managed to increase the antioxidant and antimicrobial activities of chitosan/anthocyanin films by adding reinforcing agents, such as metal nanoparticles (Cao et al., 2023; Li et al., 2023; Liu et al., 2021; Lu et al., 2022; Luo et al., 2022; Qin et al., 2019; Sun et al., 2020; Yi et al., 2024; Zhang et al., 2019; Zheng et al., 2023), nanoclay (Capello et al., 2021; Koosha & Hamed, 2019), nanocrystal (Chen et al., 2021; Fernández-Marín et al., 2022; Oun et al., 2022; Wu et al., 2019; Zheng et al., 2024), essential oils (Hao et al., 2023; Huang et al., 2023; Li et al., 2022;

Rajendran et al., 2024; Yan et al., 2021; Zhang et al., 2023; Zhao et al., 2023), curcumin (Duan et al., 2022) and bacteriocin (Zhang et al., 2023). It should be noted that the active packaging potential of chitosan/anthocyanin films is very limited, which is because the hydrophilic and colored anthocyanins can easily leak out from the films into the contacted foods (Zhang et al., 2019). As a result, the appearance of foods is negatively affected. In this respect, the release of anthocyanins is often not required for chitosan/anthocyanin films, which is significantly different from chitosan/flavonoid active packaging films.

Different from antioxidant and antimicrobial activities, color changeability is essential for chitosan/anthocyanin films in intelligent packaging field. The color changeability of chitosan/anthocyanin films is often tested under liquid and gaseous environment, where the films are immersed in solutions with different pHs or exposed to HCl/ammonia vapor with different concentrations. As listed in Table 2, the color changeability of chitosan/anthocyanin films is influenced by many factors, such as the content and source of anthocyanins, the nature of matrix, the presence of reinforcing agents, and the pH of film-forming solution. In most existing studies, different amounts of anthocyanins are added into the films, and the results show the appropriate amount of anthocyanins make the films display an ideal color changing performance. Since anthocyanins from different plants have different chemical compositions, the color changeability of chitosan/anthocyanin films is remarkably impacted by the source of anthocyanins (Kurek, Garofulić, Bakić, Šcetar, & Uzelac, 2018; Li et al., 2023; Liu et al., 2021; Singh, Nwabor, Syukri, & Voravuthikunchai, 2021; Yong et al., 2022). However, existing studies have only focused on the impact of anthocyanins from two different plants, which is far more enough. In future, deeper comparative studies should be carried out to investigate the impact of anthocyanins from more plants. Meanwhile, the influence of plant variety on the color changeability of chitosan/anthocyanin films should be also considered (Yong et al., 2019).

Some researchers have compared the color changeability of chitosan/anthocyanin films with that of other polymer/anthocyanin films, revealing the type of matrix is an important factor influencing the color changeability of the films (de Azevedo & Noreña, 2024; Peralta, Bitencourt-Cervi, Maciel, Yoshida, & Carvalho, 2019; Yong et al., 2022). For example, Yong et al. (2022) demonstrated the charged nature of matrix was a key factor affecting polymer/anthocyanin intermolecular interactions and the color of the films. As displayed in Fig. 6A, chitosan with positively charge could interact with anthocyanins through hydrogen bonds and electrostatic repulsion, causing a bathochromic shift in anthocyanins. Similarly, κ -carrageenan with negative charge could interact with anthocyanins through hydrogen bonds and electrostatic attraction. However, neutral locust bean gum could only interact with anthocyanins through hydrogen bonds, which had little impact on the color of anthocyanins (Yong et al., 2022). The color changeability of chitosan/anthocyanin films is impacted by the presence of other colorants, such as curcumin (Duan et al., 2022). Duan et al. (2022) found the color changeability of chitosan/starch/gelatin/raspberry anthocyanin film was improved by adding curcumin. However, the addition of nano fillers (Li et al., 2023; Lu et al., 2022; Qin et al., 2019; Zhang et al., 2019) and essential oils (Hao et al., 2023; Li et al., 2022; Zhang et al., 2023) has little impact on the color changeability of chitosan/anthocyanin films. Recently, a few researchers adjusted the pH of film-forming solutions and compared the color changeability of the formed films (Li et al., 2022; Li et al., 2024; Xiang et al., 2024). They demonstrated the films prepared at lower pH had better color changeability. These studies reveal the pH of film-forming solutions has a big impact on the color changeability of chitosan/anthocyanin films.

3.4. Stability of the films

Since anthocyanins are less stable than other classes of flavonoids, the stability of chitosan/anthocyanin films is a big concern of researchers. The stability of chitosan/anthocyanin films is frequently evaluated under different storage conditions by varying storage temperature, light/dark and storage time (Duan et al., 2022; Li et al., 2024; Yong et al., 2022; Zheng et al., 2023). Existing studies have revealed that low storage temperature and dark environment benefit the long-term storage of the films. Moreover, several researchers have managed to increase the stability of chitosan/anthocyanin films by different means, including encapsulation, adsorption, co-pigmentation and bilayer film (Fig. 6B). For example, Zhao et al. (2023) prepared chitosan/Arabic gum-based intelligent packaging films by adding blood orange anthocyanin-loaded thyme oil emulsions, which was based on the enhanced stability of the encapsulated anthocyanins. In a similar way, rose anthocyanin-loaded amylopectin nanoparticles were added into chitosan/starch blend to yield stable intelligent packaging films (Zheng et al., 2023). A biohybrid was fabricated by Capello et al. (2021), who adsorbed eggplant anthocyanin onto Laponite® nanoclay. Then the biohybrid was added into chitosan matrix to produce a stable intelligent packaging film. This study indicates adsorption is a good choice to enhance the stability of chitosan/anthocyanin films. Recently, Lu et al. (2024) prepared chitosan/xanthan gum-based intelligent packaging films containing red cabbage anthocyanin and rosmarinic acid, and found rosmarinic acid increased the pH sensitivity and stability of the films. This study is mainly based on the co-pigmentation effect of rosmarinic acid on anthocyanins, providing a new route to elevate the stability of chitosan/anthocyanin films. Nowadays, the construction of bilayer films is widely accepted as an effective way to increase the stability of chitosan/anthocyanin films, where the anthocyanins in the indicator layer are protected by the reinforcing agents in the protective layer (Cao et al., 2023; Huang et al., 2023; Liu et al., 2024; Luo et al., 2022; Zhang et al., 2023). Existing studies have demonstrated the reinforcing agents, such as carvacrol (Huang et al., 2023), oregano essential oil (Zhang et al., 2023), carbon dots (Liu et al., 2024) and nano TiO₂ (Cao et al., 2023; Luo et al., 2022), can protect anthocyanins from

oxidation and light irradiation.

3.5. Applications of the films

In practical application, food spoilage-related volatile (e.g., volatile ammonia, biogenic amine, CO₂ and hydrogen sulfide) often causes the acidity and alkalinity variations in the packaging container, which can be sensed by pH-responsive color changeable anthocyanins (Roy & Rhim, 2021a). In this respect, the freshness of foods can be indicated by the color of chitosan/anthocyanin films (Fig. 6C). In most existing studies, only a small piece of chitosan/anthocyanin label is attached to the inner cover of packaging container to fulfill the mission of freshness indicating. In this sense, the application cost of chitosan/anthocyanin intelligent packaging films is much lower than that of chitosan/flavonoid active packaging films. In other studies, chitosan/anthocyanin films have been simultaneously used in preserving and monitoring the freshness of foods in different approaches. The first approach is to wrap food with the film. In this way, the freshness-keeping and freshness-indicating functions are fulfilled by the food-contacted film (Cao et al., 2023; Hao et al., 2023). The second approach is to first coat food with film-forming solution and then place the coated food in a container attached with a chitosan/anthocyanin label. The film-forming solution can form a thin layer around the food and provide antioxidant and antimicrobial protections, and thus showing a good freshness-keeping effect. Meanwhile, the freshness of food can be judged by the chitosan/anthocyanin label attached to the container (Long et al., 2024). The third approach is to paste chitosan/anthocyanin label in the container and soak the label in the liquid food. The label can extend the shelf life of food by releasing antioxidant and antimicrobial functions and simultaneously indicate the freshness of food (Zhao et al., 2023). It should be noted that, no matter which kind of approach is applied, if the film-forming solution or film directly contacts with food, the migration of anthocyanins into the food is inevitable. In this regard, the impact of anthocyanins on the sensory attributes (especially the appearance) of food must be assessed beforehand. Meanwhile, the safety of film-forming solution or film should be evaluated by cytocompatibility assay (Amaregouda & Kamanna, 2023; Singh et al., 2021). The fourth approach is to add volatile antioxidant and antimicrobial agents (e.g., carvacrol) into chitosan/anthocyanin label. The volatile substances can be gradually released from the label, and thus exhibiting antioxidant and antimicrobial effects on the packaged food (Huang et al., 2023). This approach effectively avoids the direct label-food contact and simultaneously maintains the freshness indicating function of the label, which is more recommended.

As summarized in Table 2, chitosan/anthocyanin films have been used to indicate the freshness of different food products, such as meat products (beef, chicken and pork), aquatic products (fish and shrimp), dairy products (cheese and milk), fruits (litchi), fruit juice and mushrooms. Among these food products, meat and aquatic products have received the biggest attention. It should be noted the spoilage mechanisms of different food products are very different. For example, meat and aquatic products produce volatile alkaline substances during storage, which elevate the pH status, TVB-N level and TVC value of these food products. So chitosan/anthocyanin films generally turn green and even brown when they are used to monitor the freshness of meat and aquatic products (Hu, Liu, Qin, Yan, & Yang, 2022; Long et al., 2024). In a recent study, Hao et al. (2023) thoroughly evaluated the profiles of volatile compounds and bacterial community of pork during chilled storage by gas chromatography-ion mobility spectrometer and high-throughput sequencing analysis, respectively. This study provides a new route to deeply understand the spoilage mechanisms of meat products. Oppositely with meat and aquatic products, milk and fruit juice normally present a decreasing pH trend during their rancidity processes (Li, Wu, Wang, & Li, 2021; Singh et al., 2021; Zhao et al., 2023). As a result, chitosan/anthocyanin films usually turn red when they are used to monitor the freshness of milk and fruit juice (Singh

et al., 2021; Zhao et al., 2023). As for fruits, their soluble solid and titratable acid contents are gradually exhausted during storage, resulting in the color change of the films towards red (Luo et al., 2022). Mushrooms, due to their high respiration rates, can produce a large amount of CO₂. The accumulated CO₂ in the packaging container can reduce the pH status and make the films become red (Yi et al., 2023). Therefore, to evaluate the effectiveness of chitosan/anthocyanin films in intelligent packaging, not only the color changes of the films should be recorded but also the physiological and biochemical indexes of food products should be determined.

In some studies, the correlation between the color values of chitosan/anthocyanin films and the physiological/biochemical parameters of foods is analyzed (Chen et al., 2021; Li et al., 2022; Li et al., 2024; Lu et al., 2022; Yan et al., 2021; Zheng et al., 2024). A high correlation is found between the color value of the films and the TVB-N level of meat and aquatic products. The freshness degree of food can be sub-divided into fresh level (edible state), sub-fresh level (stale/start to spoilage) and spoiled level (inedible state) (Li et al., 2022; Luo et al., 2022). Notably, the color changes of chitosan/anthocyanin films can be not only observed by the naked-eye but also accurately evaluated by two color systems. One is Lab color system consisting of *L* (lightness), *a* (redness/greenness), *b* (yellowness/blueness) and ΔE (color difference) values, which can be detected by a colorimeter (Chen et al., 2021; Lu et al., 2022; Yan et al., 2021; Zheng et al., 2024). The other is RGB color system containing *R* (red), *G* (green) and *B* (blue) values, which can be determined by a specified software in the smartphone (Li et al., 2022; Li et al., 2024). However, the detection of Lab color values of the films by colorimeter is unrealistic for the consumers in the market. By contrast, the acquirement of RGB color values is more convenient for the consumers through a smartphone.

Nowadays, the commercial production and application of chitosan/anthocyanin films have several limitations. Firstly, existing studies have mainly used anthocyanin-rich plant extracts for the production of chitosan/anthocyanin films. The anthocyanin composition of the plant extracts are influenced by different factors, such as plant source, plant variety, growth condition, extraction method and extraction solvent, etc. That is why the films containing anthocyanins from the same plants frequently show different physical and functional properties (Table 2). In order to realize the standardized production of chitosan/anthocyanin films, it is necessary to obtain pure anthocyanin monomers beforehand. (2) Most existing studies have prepared chitosan/anthocyanin films by casting, which is unsuitable for large scale production of the films. Since anthocyanins are heat-labile substances, the industrially used film preparation methods, such as extrusion, blown molding, compression molding and injection molding are not suitable to prepare chitosan/anthocyanin films. In future, new and advanced equipment needs to be explored for the large scale production of chitosan/anthocyanin films. (3) To maintain the stability of chitosan/anthocyanin films during storage and application is always a big challenge. Even when the films are stored under dark and low temperature, the decomposition of anthocyanins is inevitable. Meanwhile, chitosan/anthocyanin films are too hydrophilic and might lose their integrity under highly humid packaging environment. Therefore, it is essential to seek more effective stabilization and hydrophobization methods for chitosan/anthocyanin films. (4) The application performance of chitosan/anthocyanin films is not satisfactory. Many films only show minor color changes in indicating food freshness, which cannot be easily judged by the naked eye. Moreover, the accuracy and reproducibility of the films in indicating food freshness need to be strengthened. In this sense, the formulation of the films needs to be optimized.

4. Conclusions and future perspectives

The biodegradable and compostable chitosan can be combined with bioactive and colorful flavonoids to produce chitosan/flavonoid active packaging films and chitosan/anthocyanin intelligent packaging films.

For the production of chitosan/flavonoid active packaging films, different classes of flavonoids with three forms (i.e. unmodified, modified and encapsulated forms) can be added into chitosan-based film-forming solutions. In order to improve the physical and functional properties of the films, other polymers, plasticizers, reinforcing agents and cross-linking agents can be added into chitosan-based film-forming solutions. Active packaging films are normally produced by solvent casting and electrospinning. Flavonoids can elevate the antioxidant and antimicrobial activities of the films. The active packaging potential of chitosan/flavonoid films relies on the release of flavonoids from the films, which is influenced by several factors, such as the type and form of flavonoids, the nature of film matrix, the presence of reinforcing agents, the preparation method and post-treatment of the films, and the type of food simulant. The stability of chitosan/flavonoid films can be improved by the encapsulation, structural modification, nano metal chelation and mechanical activation of flavonoids. Nowadays, chitosan/flavonoid films have been widely used in the preservation of fruits, vegetables, aquatic products, meat products and edible oils. In future, more experiments are needed to deeply compare the physical and functional properties of chitosan-based films containing different classes of flavonoids. The formula of chitosan/flavonoid films should be optimized to achieve better active packaging performances. Especial attention should be paid to elevate the stability of chitosan/flavonoid films by encapsulating flavonoids in nano-sized carriers or by grafting flavonoids onto polysaccharides. In addition, the relationship between the properties and the preservation efficiency of chitosan/flavonoid films on different foods should be established.

For the production of chitosan/anthocyanin intelligent packaging films, anthocyanins isolated from different fruits, vegetables, grains and flowers can be added into chitosan-based film-forming solutions along with other polymers, plasticizers, reinforcing agents and cross-linking agents. Intelligent packaging films can be produced by solvent casting, dip-coating, electrospinning and 3D-printing. Anthocyanins can supply chitosan-based films with pH-sensitive color changeability, which is important for intelligent packaging. The color changeability of chitosan/anthocyanin films is affected by the content and source of anthocyanins, the nature of matrix, the presence of reinforcing agents, and the pH of film-forming solution. The stability of chitosan/anthocyanin films can be elevated by different means, such as encapsulation, adsorption, copigmentation and bilayer film preparation. Chitosan/anthocyanin films are useful to indicate the freshness of meat products, aquatic products, dairy products, fruits, fruit juice and mushrooms. The integration of active and intelligent packaging functions is a new trend for chitosan/anthocyanin films. In future, it is necessary to obtain pure anthocyanin monomers to realize the standardized production of chitosan/anthocyanin films. New and advanced equipment needs to be explored for the large scale production of chitosan/anthocyanin films. It is essential to seek more effective stabilization and hydrophobization methods for chitosan/anthocyanin films. The intelligent packaging performance of chitosan/anthocyanin films needs to be optimized to achieve good accuracy and reproducibility.

CRediT authorship contribution statement

Xuanzhuo Liu: Writing – original draft, Investigation, Formal analysis, Data curation. **Fengfeng Xu:** Investigation, Data curation. **Huimin Yong:** Visualization, Conceptualization. **Dan Chen:** Visualization. **Chao Tang:** Investigation. **Juan Kan:** Writing – review & editing. **Jun Liu:** Writing – review & editing, Supervision, Project administration, Funding acquisition, Conceptualization.

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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Appendix A. Supplementary Data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.fochx.2025.102200>.

Data availability

Data will be made available on request.

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