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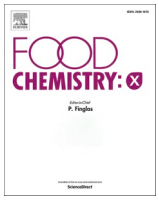

Food packaging films from natural polysaccharides and protein hydrogels: A comprehensive review

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ABSTRACT

The development of innovative, biodegradable food packaging materials to combat plastic pollution has garnered significant attention from scholars and government agencies worldwide. Natural polysaccharides and proteins exhibit excellent modifiability, biodegradability, high ductility, and compatibility with food products, making them ideal candidates for constructing hydrogels. Hydrogel films based on these biopolymers have opened new research horizons in food packaging applications. This review examines natural polysaccharides and proteins commonly used in hydrogel film preparation and explores strategies to improve their packaging performance, including the use of binary mixtures and exogenous additives. To optimize functionality, the cross-linking mechanisms between materials and film-forming methods are summarized. Additionally, recent applications of hydrogel films in food packaging in are discussed, showcasing their ability to extend or monitor food freshness. Despite existing challenges, the current advancements present a promising and sustainable alternative to conventional plastic materials paving the way for innovative packaging solutions.

1. Methodology and visualization

For the present study, relevant research and review articles were retrieved from the Web of Science Core Collection database and analyzed using VOSviewer for visualization (Fig. 1). During the 5-year period, 1699 publications have been reported on hydrogel films, with 257 of these (including 22 reviews) focusing on packaging applications. Visualization results highlighted that key research areas revolve around terms such as “hydrogel film,” “food packaging,” “proteins,” and “polysaccharides,” underscoring their significance in current investigations. Furthermore, terms related to the tensile strength and stability of films appeared with notable frequency, suggesting that these attributes are critical areas of interest. The findings reflect a growing scholarly focus on enhancing these materials’ functional properties. However, a thorough review of existing literature revealed a distinct gap: a detailed and comprehensive analysis addressing the methods to improve the performance of natural polysaccharide and protein hydrogel films, specifically for food packaging applications, is lacking.

2. Introduction

Food packaging serves a vital role in protecting food products from environmental factors such as insects, odors, bacteria, light, air, and moisture, all of which can compromise sensory qualities or nutritional value. Currently, plastic materials dominate the packaging industry due to their affordability, flexibility, and ability to support diverse packaging designs. However, owing to their nonbiodegradable properties, they exert an increasing environmental impact, further contributing to pollution, environmental harm, and global warming. On the 52nd World Environment Day (5 June 2023), the theme is “Beat Plastic Pollution”. On the same day, the United Nations Convention to Combat Desertification (UNCCD) published a report stating that only 9 % of the world’s

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plastics are recycled, and most of these are either discarded or cannot be recycled. The problem of "white pollution" (plastic pollution), caused by a large amount of nondegradable food plastic packaging, has presented a serious risk to the living environment of human beings. Therefore, the development of new eco-friendly packaging materials that can maintain the nutritional value of food, extend the freshness of food, and undergo biodegradation has become an inevitable trend in the development of food packaging.

Edible polymers, which are generally recognized as safe (GRAS) by the Food and Drug Administration (FDA), are gaining prominence as high-quality, biodegradable, and biocompatible alternatives to non-biodegradable, low-quality materials across various industries. Their applications span diverse fields, including flexible wearable sensors, drug delivery systems, medical imaging, and tissue engineering (Manzoor et al., 2022; Rakshit et al., 2024; Yang, Zhang, et al., 2024). In this regard, natural polysaccharide and protein hydrogels have been suggested as potential replacements for currently used packaging due to their wide availability and biodegradability. Hydrogels have three-dimensional cross-linked network structures. These networks, formed by hydrophilic polymers through covalent cross-linking or non-covalent interactions (such as physical entanglement, hydrogen bonding, and ionic interactions), can retain substantial amounts of water(D. Yang, 2022). With the continuous in-depth research on hydrogels in the past decade, hydrogel films, which exhibit swelling and antimechanical properties, can serve as an effective barrier to protect food, while effectively controlling food moisture. As such, they can maintain the nutritional value of food and even show antibacterial properties. Natural polysaccharide and protein hydrogels, while promising have certain limitations. A major disadvantage is their lack of stability in the swollen state under specific environmental conditions(Ma et al., 2020). For example, in perishable fresh foods such as mushrooms, fruits, and vegetables, high transpiration and respiration rates generate excess water vapor, which condenses into droplets on the inner surface of the packaging(Shan et al., 2024). In such cases, the hydrogel film absorbs the excess moisture and swells, compromising its mechanical strength (Y. Cao et al., 2024). This issue can potentially be addressed by incorporating certain substances (e.g., phenols, aldehydes, or nanoparticles) that enhance the mechanical and functional properties of the hydrogel. These substances can cross-link with the hydrogel components or fill structural gaps in the case of nanoparticles, creating a more compact network structure(Batista et al., 2019; Maroufi et al., 2021; Wang, Chen, et al., 2024). Therefore, these enhancements can extend the shelf life of fresh foods, providing a new avenue for developing degradable food packaging solutions.

Despite these advancements, comprehensive and detailed reviews on improving the properties of natural polysaccharide and protein hydrogel films for food packaging are lacking. This review aims to fill that gap by presenting a complete and detailed description of the various aspects of these hydrogels in food packaging and proposing optimization strategies to overcomes their shortcomings and enhance their performance, ultimately creating superior hydrogel packaging films. Among them, binary mixtures and the addition of exogenous additives are proposed as strategies for optimization with respect to the disadvantages of natural polysaccharide and protein materials. In addition, the possible cross-linking mechanisms and film-forming methods used in the formation of hydrogel films are introduced, with emphasis on their application and development in the field of food preservation films, edible films, and intelligent freshness indicator films. At last, some perspectives on the challenges and potential associated with the future development of hydrogel in the field of food packaging are presented, with a view of encouraging additional researchers to do further in-depth research on the subject.

3. Sources for hydrogel films

Natural hydrogels are derived from widely available natural

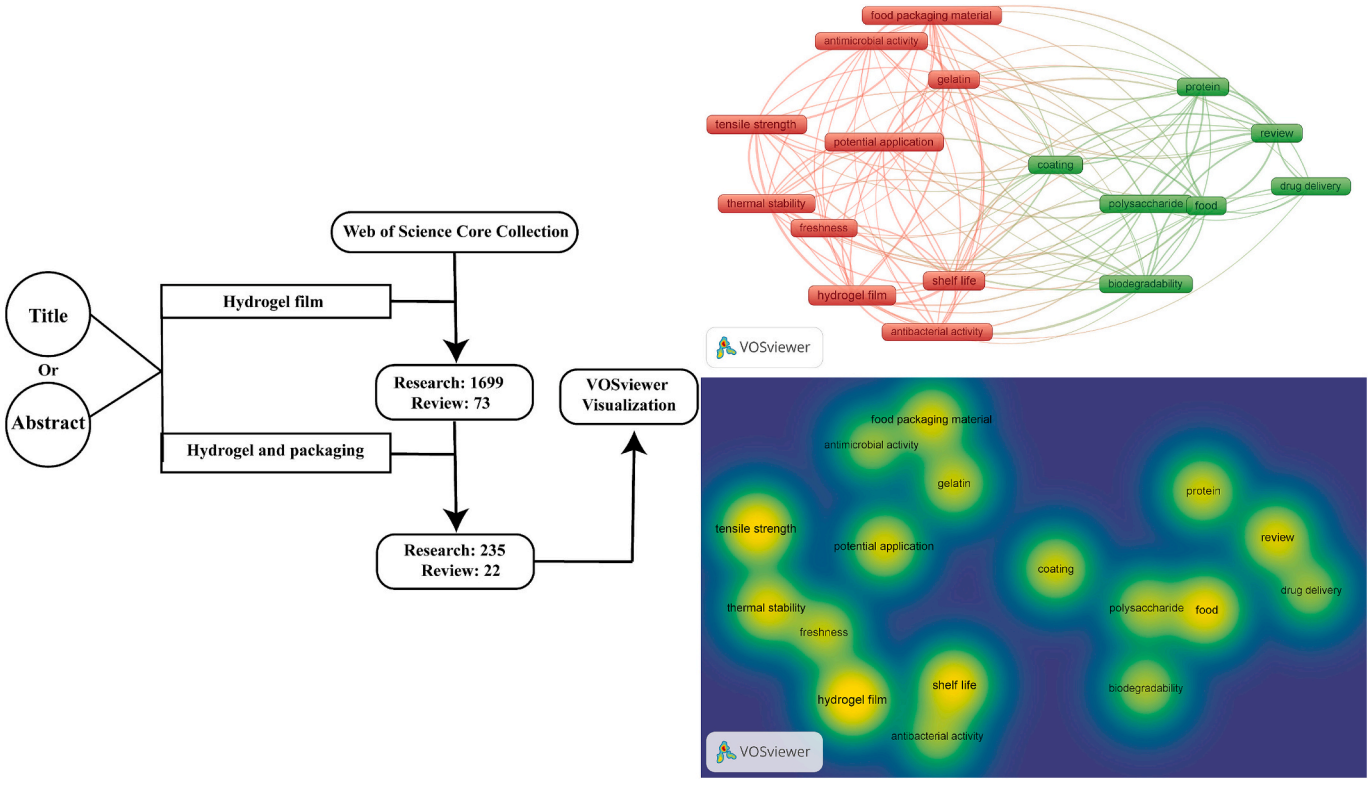


Fig. 1. Search and visualization of relevant literature in the last five years(May 2019–May 2024).
Note: Only articles and review articles are ticked when searching, except for the options of proceeding parper, early access, conference abstracts, letters, meetings, etc.

polymers such as polysaccharides, proteins, lipids, and DNA, offering expanded applications and enhanced biocompatibility. However, while lipids are often incorporated into hydrogel formulations for packaging purposes (e.g., liposomes or wax microspheres), their susceptibility to oxidative deterioration and poor mechanical strength after film formation limit their broader use(Lisuzzo et al., 2021; Shu et al., 2023). Moreover, the high cost of DNA-based hydrogels makes them unsuitable for food packaging applications (Liu, Gou, et al., 2022). By contrast, natural polysaccharides (such as chitosan, starch, and pectin) and proteins (like gelatin, whey proteins, and zein) are more viable candidates for hydrogel films in food packaging. These materials are favored due to their superior properties, including availability, sustainability, structural diversity, biocompatibility, and consumer acceptability.

3.1. Polysaccharide-based hydrogels

3.1.1. Chitosan

Chitosan (CS) results from partial acetylation of the natural polysaccharide chitin, indicating that both have similar chemical structures. Notably, the amino group in the CS molecule is more reactive than the acetylamino group, endowing CS with the ability to perform key biological functions and chemical modification reactions(Al-Rooqi et al., 2022). CS presents several advantages, including a wide availability and low production cost. Containing free amino groups, CS is the sole alkaline polysaccharide among natural polysaccharides. This property distinctly positions CS for special applications in hydrogel development. Moreover, the exceptional biocompatibility, biodegradability, antimicrobial properties, and precisely controlled degree of deacetylation exhibited by CS present significant advantages for advancing hydrogel technology(Catoira et al., 2019). Therefore, CS has been recognized as a functional biomaterial with higher application potential compared with cellulose. In addition, studies have revealed the significance of CS hydrogels in protecting fruits, vegetables, and meat from microbial decay. This approach has been approved for human consumption globally, ensuring safety(Kumar et al., 2020). Furthermore, films formed from CS hydrogels show better water and moisture resistance than those formed from pectin(Yi et al., 2024), starch(Long et al., 2023), and gelatin(H. Yu et al., 2023), making CS an ideal material for food packaging (Razavi et al., 2020). Nonetheless, CS hydrogel films have certain disadvantages, including low mechanical properties, inadequate water retention, and high sensitivity to humidity. These deficiencies limit the industrial applications of the material.

3.1.2. Starch

The extensive study of starch is a consequence the diversity of its sources, abundance, low cost, non-toxicity, renewability, and film-forming ability. Nevertheless, the utilisation of starch in hydrogel, particularly in hydrogel films, still needs to address concerns, including the limited water solubility at room temperature, high-temperature pasting, and high viscosity, among others(Dutta & Sit, 2024). The majority of current techniques have been employed to enhance the properties of starch hydrogels by incorporating other substances, such as gelatin, CS, glycerol, cellulose. The conversion of the hydroxyl structure of starch into aldehyde groups by oxidizing it to form a double aldehyde starch may be beneficial for the utilisation of starch in the direction of hydrogels. Cui et al. (2022a) prepared a curcumin hydrogel carrier with dialdehyde starch (DS) and gelatin. These hydrogels display self-recovery and fatigue resistance, in addition to exhibiting enhanced their hardness, elasticity, deformation capacity, compressive strength, and modulus increase with an increase in DS content. They also exhibit effective pH-dependent sustained release of curcumin. In addition, the structure and function of starch with polyhydroxy structure can be easily regulated by chemical or enzymatic methods, thus expanding its application(Rosseto et al., 2020; D. Zhao et al., 2024).

3.1.3. Pectin

Pectin is a heteropolysaccharide derived from the cell walls of higher plants. Consequently, pectin has been classified under GRAS by the FDA. The favourable biocompatibility, edibility, and physical characteristics (e.g., gelling and selective air permeability) of pectin render it an appropriate material for the fabrication of edible food packaging films (Espitia et al., 2014). Previous studies have verified the utility of pectin-based films in food packaging. These films have been shown to effectively reduce the respiratory rate and oxidation of packaged foods(W. Ren et al., 2022). However, pure pectin hydrogels films with reduced tensile strength (TS), increased brittleness, and inadequate water repellency are highly susceptible to tearing during preparation and storage(Lorevice et al., 2016).

3.1.4. Xanthan gum

Xanthan gum (XG), a natural heteropolysaccharide, is a high-molecular-weight exopolysaccharide produced by the Gram-negative bacteria *Xanthomonas campestris*. The primary chain of XG engages in hydrogen bonding with the side chains, resulting in a wrapped configuration. This distinct structure endows it with high stability over wide ranges of temperature and pH(Byram et al., 2020). Furthermore, it exhibits various properties such as biocompatibility, pseudoplasticity, and high viscosity, which are conducive to the formation of hydrogels and films. The rheological properties and viscosity of XG are analogous to hyaluronic acid, yet more stable (Han et al., 2012). Confirmed safe by FDA, XG has been used as an emulsifier and stabilizer in food(Kamer et al., 2024). On the other hand, XG, as a hydrophilic colloid, exhibits significant water absorption and swelling in high-humidity environments. This leads to structural instability and a marked reduction in mechanical strength(Aghajanzadeh et al., 2024).

3.1.5. Sodium alginate

Sodium alginate (SA) shows favourable biocompatibility, biodegradability, and stabilization at room temperature. It can also enhance antioxidant and immune activities. Notably, SA is safe and nontoxic and has been recommended for use in food for infants and toddlers by the European Food Safety Authority. As an anionic polymer, SA can rapidly form particles under the condition of polyvalent cations, thereby exhibiting favourable hydrogel(Nezamdoost-Sani et al., 2023). However, SA hydrogel films, when crosslinked ionically, suffer from poor mechanical properties and weak thermal stability(Hadi et al., 2023; Tan et al., 2023). To address these challenges, researchers have explored chemically crosslinking SA by oxidizing its hydroxyl groups to aldehyde groups using sodium periodate, thereby improving the hydrogel's structural integrity. Ge et al. (2022) crosslinked oxidized SA with other substances for fruit preservation and significantly extended the shelf life of strawberries and apples.

3.1.6. Konjac glucomannan

Konjac glucomannan (KGM), a heteropolysaccharide composed of D-glucose and D-mannose linked by 1,4 bonds in a 1:1.6 ratio, is primarily derived from konjac tubers(Manzoor et al., 2022). Characterized by biodegradability, high viscosity, water solubility, nontoxicity, good film-forming properties, and abundant resources, KGM has been widely used in food packaging films, biomedicine, and fat substitutes(W. Lin et al., 2019). However, its application in food packaging is limited because of poor mechanical properties and the absence of antibacterial properties(J. Wang et al., 2019). To address these deficiencies, Kim et al. (2023) enhanced the mechanical properties of hydrogel films by combining κ-carrageenan with KGM and loading them with *Salmonella* phage PBSE191 to improve their ability to inhibit *Salmonella* in chicken meat.

3.1.7. Hyaluronic acid

The hyaluronic acid(HA)-based hydrogel, obtained using HA as the starting material, exhibits good biocompatibility, biodegradability,

mechanical viscoelasticity, and other properties while maintaining the distinctive physiological function of HA(Y. Luo et al., 2023). These beneficial physiological and physical characteristics render HA prominent in biomedicine, particularly in ophthalmic treatment and joint repair(Galarraga et al., 2022). For example, Shang Wang, Lei, et al. (2024) developed an injectable dual network hydrogel composed of sulphated CS and oxidized HA for healing bone defects. S. Zhang et al. (2021) constructed a hydrogel form gelatin and HA deliver curcumin-loaded carboxymethyl CS microspheres for drug delivery. However, relatively few studies have been conducted on the use of HA in food packaging, which may be attributed to the poor stability and high water absorption of HA films.

3.1.8. Potential polysaccharides

In addition, certain naturally occurring polysaccharides with good gelation properties, such as cellulose, salecan, and κ-carrageenan, which possess favourable gelation properties, also exhibit potential for use in food packaging. Yang, Li, et al. (2024) prepared a carboxymethylcellulose-based active hydrogel films for packaging and preservation of fruits. Shan et al. (2024) fabricated a humidity-adjustable active packaging film based on gelatin hydrogel and ethylcellulose. Deng et al. (2024) created a salecan-based hydrogel dressing with self-healing, injectable, and antimicrobial properties. Forghani and Almasi (2024) designed a hydrogel film based on carrageenan/capucel mucilage and combined it with the red cabbage extract for fish freshness indication. However, of note, while these polysaccharide-based hydrogel films show great potential, they have been relatively underexplored in the food packaging field. In particular, salecan-based hydrogel films warrant further investigation and development.

3.2. Protein-based hydrogels

3.2.1. Gelatin

Gelatin is a water-soluble high-molecular-weight peptide derived from collagen hydrolysis. Gelatin has a cost-effective and acceptable safety because of its substantial collagen content in waste products (Toniciolli Rigueto et al., 2022). Recognized for its biodegradability, renewability, and biocompatibility, gelatin also shows potential for creating eco-friendly biocomposite materials. Furthermore, gelatin has a diverse range of functional groups, including -COOH group of aspartic acid terminal -NH_2 and -COOH groups, the -NH_2 group of lysine, the imidazolium group of histidine, and the guanidinium group of arginine. It also features carboxyl and phenolic groups, which serve as potential sites for conjugation and chemical modifications(Mushtaq et al., 2022). These crosslinkable and graftable groups allow gelatin to exhibit excellent responsiveness to structural and functional adjustments. Due to its remarkable crosslinking ability, gelatin is among the most widely used natural materials for developing composite hydrogels or films.

However, despite its versatility, gelatin's hydrophilic nature poses significant challenges. Under high-humidity conditions, it absorbs water, swells and may decompose(Q. Luo et al., 2022). Furthermore, pure gelatin hydrogel films produced under standard drying conditions tend to have low mechanical strength and are brittle, limiting their suitability for various food packaging applications(W. Zhang, Azizi-Lalabadi, et al., 2023).

3.2.2. Whey

Whey is a liquid by-product of the dairy industry, formed during cheesemaking or the production of other coagulated dairy products. Whey proteins possess favourable gelling and emulsifying properties. At lower relative humidity levels, they can form hydrogels with enhanced mechanical and gas barrier attributes(Liu, Zhang, et al., 2020). However, condensed whey protein hydrogels are susceptible to weakening in mechanical strength and rapid degradation by enzymes, hence its limited application in food packaging. This limitation can be effectively overcome by producing hybrid hydrogels. For example, whey proteins can form complexes with one or both of the following substances: CS, gelatin, phenolics, and other substances that exhibit favourable biodegradability, biocompatibility, and mechanical properties(Yan et al., 2023). The aforementioned characteristics render whey viable as natural materials for food packaging.

3.2.3. Zein

Zein is a natural, macromolecular, and biodegradable material derived from renewable resources. It is nonallergenic and classified as GRAS amphiphilic prolamin. In addition, zein is an outstanding material for producing food films because of its low water vapor permeability, oil resistance, biodegradability, and biocompatibility(Ibrahim et al., 2019). Meanwhile, zein molecules generate strong disulfide and hydrophobic interactions that lead to the formation of films, which may serve as synthetic plastic films(Yuan et al., 2022). Zein hydrogel films have been successfully used to encapsulate, transfer, and release active substances. Maize gliadin lacks charged acidic, alkaline, and polar amino acids, resulting in the brittleness of a single membrane, low TS, and weak ductility, thus restricting its functionality(Yuan et al., 2022). Therefore, the aforementioned concerns need to be addressed to obtain superior zein hydrogel films.

3.2.4. Ovalbumin

Ovalbumin (OVA) exhibits exceptional gelling, foaming, and emulsifying capabilities that render it suitable for applications as a delivery method. Using thermal induction and ultrasonic processing, Rao et al. (2020) created an OVA nanoparticle hydrogels and incorporated carvacrol for antibacterial purposes in food applications. This will provide a reference for the development of OVA antibacterial films. However, individual egg proteins are costly to extract and purify, particularly those with low concentrations. Consequently, these individual proteins have not been directly used as encapsulant materials for commercial food applications(Abeyrathne et al., 2013).

4. Strategies for enhancing the quality of hydrogels

An ideal material for food packaging must have robust mechanical properties, serve as effective barriers to oxygen and humidity, and prevent microbial contamination during transportation and storage (Manzoor et al., 2022). However, single component hydrogel films composed of polysaccharides or proteins often fail to meet these requirements. Their inherent weaknesses, such as poor mechanical strength, limited stability, high water absorption, and insufficient antimicrobial properties restrict their application in food packaging. Specific strategies need to be undertaken to enhance the quality of the hydrogel and thus achieve the desired goal. This approach may entail combining multiple natural polymers, adding active ingredients or nanoparticles, among other measures (Table.1).

4.1. Binary biopolymer mixtures

Binary biopolymer mixtures comprising two natural polymers can be broadly categorised into three groups: polysaccharide–polysaccharide, protein–protein, and polysaccharide–protein mixtures. The process of gelation can be enhanced by combining two natural polymers. Both protein–protein and polysaccharide–polysaccharide combinations have been used successfully in research to build and optimize food hydrogels with altered structural properties(X. Yang et al., 2021). The study of protein–polysaccharide hydrogels has been a prominent area of research in recent years. Compared with a single type of hydrogel, protein–polysaccharide hydrogels offers various benefits, including superior water-holding capacity(Le et al., 2017). These mixed hydrogels also distinctly offer various microstructures. This advantage may be attributed to proteins and polysaccharides being separate types of biopolymers with extremely different chemical structures, physical characteristics, and activities(Hou et al., 2015). The combination of

Table 1
Strategies to enhance the properties of natural protein and polysaccharide hydrogel films.

Enhancement Strategies		Materials	Additives	Properties	Applications	References
Binary biopolymer mixtures		XG/hydroxypropyl methylcellulose	-	Visible light barrier performance was enhanced; 0.2 g of XG addition increased the TS and EAB of the film by about 20 MPa and 7 %; decreased the weight loss of banana after 18 days of storage.	Fruit preservation	(M. Zheng et al., 2022)
		High amylose corn starch/KGM	-	TS (7.58 MPa → ~9.5 MPa) and EAB (35.17 % → 54.11 %) increased significantly; WVP decreased	Food packaging film	(Zou et al., 2021)
		Fish gelatin/Casein phosphopeptides	-	0.1 g/100 mL: TS: 9.6 MPa → 18.1 Mpa; EAB: 23.4 % → 84.2 %; WVP decreased; UV-blocking performance, transparency and thermal performance enhancement	Edible bioactive film	(Khedri et al., 2021)
Additives	Bioactive materials	CS, gelatin	Curcumin	0.2 wt%: breaking strength: 2.38 MPa → 2.79 Mpa; DPPH radical scavenging activity: 8.95 % → 41.32 %; enhanced antibacterial activity; provides color responsiveness to ammonia	Freshness indication for protein-rich foods	(M. Duan et al., 2023)
		Gelatin	Eugenol	Reduced light transmission; enhanced film hydrophobicity and antioxidant activity; significant inhibitory effect on *S. aureus*; available for preservation of chicken breast meat	Edible film; food preservation	(L. Lin et al., 2023)
		Amphiphilic CS, carboxymethyl GG	Mustard essential oil	Amphiphilic CS: carboxymethyl GG = 5: 2, the composite membrane had the best thermal stability (249.58 °C) and TS, EAB (1.34 MPa and 86.67 % → 3.35 MPa and 111.0 %); reduced WVP and oxygen permeability; 2.0 μL/mL of mustard essential oil: TS: 3.35 MPa → 3.72 MPa; better inhibition of Gram-positive bacteria; 10 days longer shelf life of mango	Fruit preservation	(Z. Yang et al., 2023)
Additives	Nanoparticles	CS	Titanium dioxide (TiO_2)	TS: 24.43 MPa → 46.33 MPa; EAB: 15.23 % → 25.77 %; WCA: 98.0° → 44.4°; decrease in light transmission in the visible area; extended shelf life of red grapes (CS film: 15 days, CS-TiO_2 film: 22 days)	Food preservation	(Zhang et al., 2017)
		κ-carrageenan, 2-hydroxyethyl cellulose	SiO_2, Ag	10 wt%: TS: 23.8 MPa → 37.1(+Ag: 41.5) Mpa; EAB: 22.3 % → 27.5 %(+Ag: 28.9); WCA: 60.1° → 73.6°(+Ag: 76.4°); the transmission rate of visible light and the transmission rate of water vapor decrease; thermal stability enhancement	Antibacterial film	(Zhang et al., 2017)
		Carrageenan	zinc oxide (ZnO), copper oxide (CuO)	TS: 55.2 Mpa → 48.2(1 % ZnO)/32.9(0.5 % ZnO, 0.5 % CuO)/ 30.4(1 % CuO) Mpa; EAB: 6.6 % → 10.3 %(1 % ZnO)/18.2 %(0.5 % ZnO, 0.5 % CuO)/ 15.9 %(1 % CuO); stronger antibacterial activity against *E. coli* than L. *monocytogenes*	Active food packaging	(Oun & Rhim, 2017)

Note: "/": film materials/reinforcing substances; TS: tensile strength; EAB: elongation at break; WVP: water vapor permeability; WCA: water contact angle.

polysaccharides and proteins can lead to a wider range of phase behaviors. These variable phase behaviors can arise from varying affinities between polymers and solvents (water), in addition to long- or short-term interactions between polysaccharides and proteins(X. Yang et al., 2021). Consequently, multiple alternatives for designing distinctive hydrogel structures exist when combinations of polysaccharides and proteins are used, facilitating the preparation and application of hydrogel films.

4.2. Additives for hydrogels

Additives are chemical compounds added to polymers to reinforce their inherent properties. Moreover, additives derived from natural substances are highly attractive material that is safe and renewable. The performance and functional aspects of hydrogel films are currently improved using additives (Fig. 2), such as phenols, aldehydes, nanoparticles, and essential oils. The incorporation of these substances into hydrogel films can enhance their antioxidant or antibacterial properties, thus promoting the preservation of food quality during the food packaging.

4.2.1. Bioactive materials

4.2.1.1. Phenolic compounds. Phenolic compounds with at least one aromatic group attached to numerous hydroxyl groups can promote hydrogen bonding across amino, amide, and hydroxyl groups along the polymerization chain(Leyva-Jiménez et al., 2023). These electrostatic interactions may stabilize the hydrogel structure, whilst simultaneously increasing the toughness, stiffness, and expansion capacity of the material.

Phenols have recently gained prominence in the production of hydrogel films. Biao et al. (2019) developed tea polyphenol-reinforced calcium alginate hydrogel edible films developed with enhanced mechanical characteristics. When added within the range of 1 %–5 %, tea polyphenols may improve the TS and EAB of films. The mechanical characteristics of the films were also significantly affected to varying degrees, depending on the quantity, by the addition of polyphenols. Moreover, rutin is used to enhance the mechanical qualities of natural polymer-based hydrogel films. Wang et al. (2020b) used glucomannan and gelatin as the gelling base to prepare composite hydrogel films with rutin as an additive. In the study, TS increased significantly in the experimental group relative to the control group($P < 0.05$); in addition, it substantially decreased the water vapor transmission rate of the film. The reason is that rutin promoted the formation of hydrogen bond-based electrostatic interaction and Schiff bond-based intermolecular interaction in the hydrogel films. The addition of plant extracts rich in phenolic compounds yielded similar effects, except when phenolics were directly introduced into the hydrogel films. For instance, incorporating 5 % (*w*/w, hydrogel) grape seed extract increased the EAB of carboxymethylcellulose-based films by 20 %(Oun & Rhim, 2020); similarly, the addition of 10 % (*v*/v) lychee shell extract raised the EAB of guar gum/carboxymethylcellulose films from 29.93 % to 62.12 % (Deshmukh et al., 2022); however, high concentrations of extracts(> 10 %) can aggregate within hydrogel films, disrupting the polymer chains. This aggregation compromises the film's structure and negatively affects its mechanical properties (Wang, Yin, et al., 2024).

Moreover, these polyphenols can inhibit the growth and

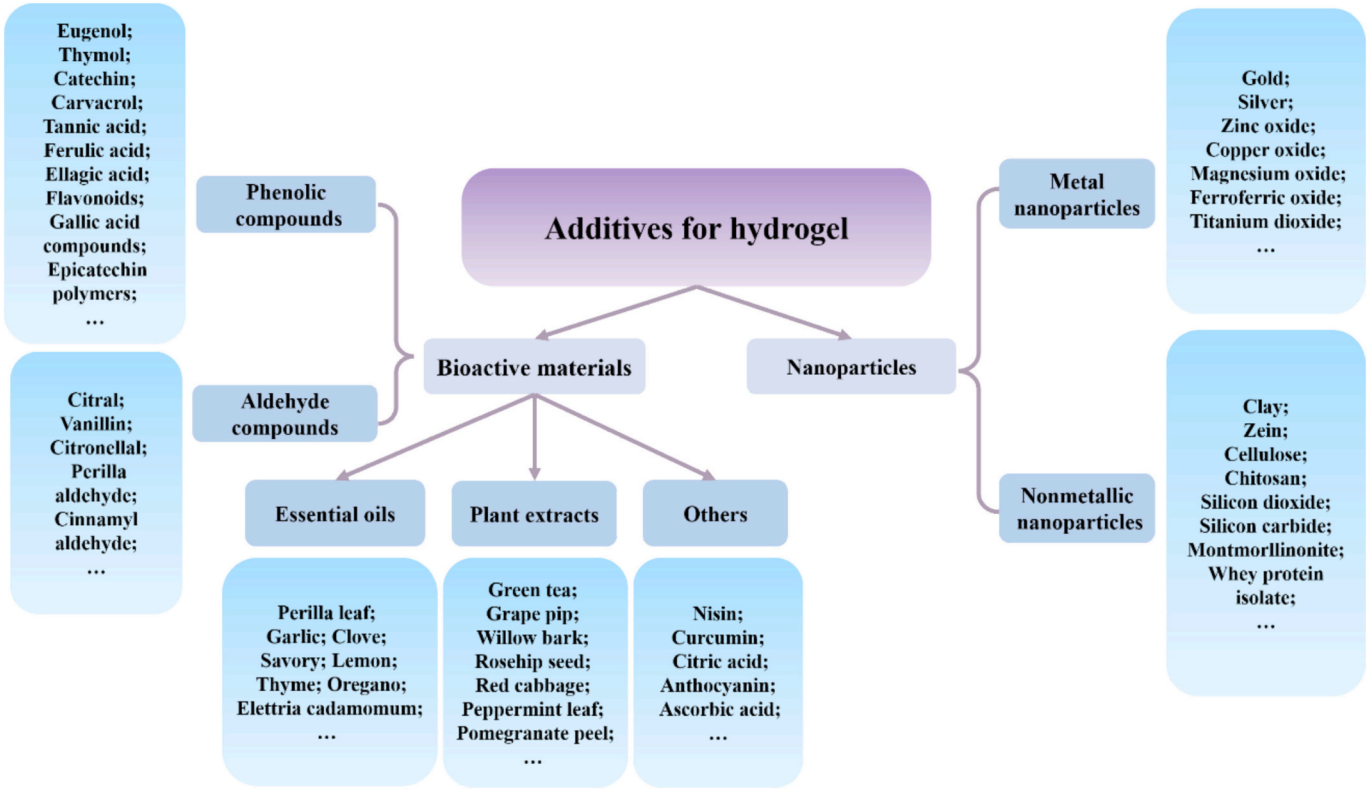


Fig. 2. Common additive ingredients for hydrogel performance enhancement.

reproduction of microorganisms through various mechanisms. These mechanisms include inactivating enzymes; inhibiting the synthesis of DNA, RNA, and proteins; and disrupting bacterial membrane proteins. The addition of tea polyphenols in alginate films by Biao et al. (2019) significantly improved the antioxidant capacity of the films. In addition, the hydrogel containing pomegranate extract more strongly inhibited the growth of *Escherichia coli*(*E. coli*) and *Staphylococcus aureus* (*S. aureus*), which was mainly attributed to the phenolic compounds such as gallic acid, ellagic acid, and pomegranate glycosides in pomegranate extracts (Maroufi et al., 2021).

4.2.1.2. Aldehydes. Some aldehydes are also used to enhance the quality of hydrogel films. For instance, cinnamaldehyde(CIN) is employed to improve the practical utility of high-amylose cornstarch films(Wan et al., 2023). The addition of 5 % (*w*/w) of CIN resulted in the films exhibiting favourable mechanical properties. Moreover, the diameter of the inhibition zone of high-amylose corn starch films containing 20 %–60 % CIN against *E. coli* and *S. aureus* exceeded 16 mm, and increased with an increase in the mass ratio of CIN(Wan et al., 2023). The antibacterial effectiveness is categorised into three levels based on the diameter of the antibacterial zone: weak (diameter $<$ 10 mm), moderate (10–16 mm), and strong ($\geq$ 16 mm)(Gul et al., 2022). This classify cation indicates that 20 %–60 % of CIN exhibits high antibacterial activity. When the mass fraction of CIN is $\geq$40 %, the preservation period of strawberries can be extended to 15 d. Moreover, the aldehyde functional group based on the substance can react with the amino group to form a Schiff base, rendering the hydrogel structure more compact.

4.2.1.3. Others. In addition to this phenolic material, essential oil is also widely used in hydrogel films. With strong antibacterial and antioxidant qualities, essential oils derived from plants and spices are ideal for various applications. They may enhance the physicochemical characteristics and functional performance of edible films(W. Chen et al., 2022). Nevertheless, the strong aroma and volatility of essential oils are present a challenge for the packaging because of their adverse effects on the sensory perception of food.

Other kinds of active ingredients can also be incorporated into hydrogel films to amplify their functional attributes. Examples include curcumin, anthocyanins, and antimicrobial peptides(Liu, Liu, et al., 2020; Wu et al., 2021). This improvement opens up more application prospects for hydrogel films, including intelligent freshness indicators, edible films, and more.

4.2.2. Nanoparticles

Nanoparticles are also called ultrafine particles, which generally refers to particles with sizes in the 1–100 nm range. Moreover, nanoparticles improve the material properties of biopolymers. Nanocellulose, silicon dioxide, titanium dioxide, and some other organic or inorganic substances can enhance the mechanical properties (TS, toughness, shear strength, etc.) and barrier properties of biopolymers(Mihindukulasuriya & Lim, 2014). In addition, precious metal nanoparticles (such as gold and silver) can provide good antibacterial properties for composite materials(Atta et al., 2022).

4.2.2.1. Gold/silver nanoparticles. Gold nanoparticles (AuNPs) can increase cellular oxidative stress by generating ROS and releasing intracellular lactate dehydrogenase into extracellular media, thereby exerting antibacterial effects(Q. Yu et al., 2016). Despite the low toxicity of AuNPs to *Pseudomonas aeruginosa* and *Candida albicans*, nanoparticles have been demonstrated to prevent the formation of biofilms and the invasion of root canal stem cells(Atta et al., 2022). By contrast, silver nanoparticles (AgNPs) damage the cell membrane, leading to the leakage of cell contents and ultimately bacterial death(Marambio-Jones & Hoek, 2010). Olivier salad packaged with different concentrations of AgNPs can exhibit better sensory characteristics and less microbial growth. Moreover, when the nanosilver concentration is 404.93 ppm, good antibacterial effects can be achieved, and salad freshness can be

maintained for about 14 d(Valipour Motlagh et al., 2021). Notably, the antibacterial performance of AgNPs is superior to that of AuNPs; with regard to safety, AgNPs (low concentration safety) are weaker than AuNPs (nontoxic).

In addition to their antibacterial properties, hydrogels combined with gold nanoparticles can serve as indicators. Goal-response hydrogels containing AuNPs have been synthesized in some studies(Z. Zhu et al., 2014). Upon contact with the target, the hydrogel quickly dissolves, releasing an Au core/Pt shell nanoparticle (Au@PtNP). It can efficiently catalyze the decomposition of H_2O_2, generating a large amount of O_2 that facilitates the generation of quantitative bar graphs to quantify the target content. Moreover, AgNPs exhibit electrochemical and bioluminescent properties, rendering them suitable for use in nanosensors and biomarkers (Bhardwaj et al., 2013). With the aforementioned information considered, AuNPs and AgNPs can not only confer antibacterial properties on composites but also exert distinctive effects in monitoring and detecting food contamination. This process can potentially lead to new directions in food packaging applications.

4.2.2.2. Oxidized nanoparticles. Some oxidized nanoparticles, such as CuO, ZnO, TiO_2, and SiO_2, have drawn attention for their bacteriostatic characteristics and enhanced material properties. Moreover, metal oxide nanoparticles demonstrate enhanced stability relative to conventional antimicrobial drugs under extreme conditions and are considered nontoxic to humans(Krol et al., 2017).

4.2.2.2.1. Titanium dioxide nanoparticles. Titanium dioxide nanoparticles (TiO_2NPs), characterized by high thermal stability and inertness, represent good photocatalyst materials in the energy and environment sector. Their uses encompass air and water purification, antibacterial treatment, self-cleaning surfaces, and water separation. These functionalities are closely related to their cost-effectiveness, nontoxicity, and other characteristics. Similar to studies on zinc oxide nanoparticles, research on titanium dioxide nanoparticles focuses on antibacterial properties. Without light, TiO_2NPs can directly adsorb with cells, leading to a loss of membrane integrity; when exposed to UV light, TiO_2NPs exhibit direct activation of their antibacterial activity(Khan et al., 2015). It effectively targets common foodborne pathogens such as L. *monocytogenes*, *V. parahaemolyticus*, and *S. choleraesuis*(Atta et al., 2022). In other aspects, Oleyaei et al. (2016) prepared starch (potato)-TiO_2NP composite films and investigated their physical properties. TiO_2NPs improved the optical transparency of the films and slightly increased TS and the contact angle; however, they significantly decreased water–vapor permeability (WVP) and EAB. Goudarzi et al. (2017) also prepared composite films by using starch (wheat) and TiO_2NPs. In their study, the TiO_2NP content, hydrophobicity, EAB, and fracture tensile energy increased, while TS, WVP, and Young's modulus decreased. This result slightly varied from the TS and EAB findings by Oleyaei et al. (2016).

4.2.2.2.2. Silicon dioxide nanoparticles. Silicon dioxide (SiO_2) is a common element extensively found in the surface soil of the Earth and is the predominant form occurring naturally. Recent studies have shown that SiO_2 nanoparticles (SiO_2NPs) can be used as a good exogenous additive to improve the physical and mechanical properties of various natural polymer hydrogel films, such as CS, gelatin, starch, and so on because of their high specific surface area, low toxicity, biocompatibility, optical transparency, and high stability(W. Zhang, Ahari, et al., 2023). Moreover, it is beneficial for improving the sustained release properties of other active substances in the hydrogel films, extending the duration of their action. Manli Yang et al. (2016) added SiO_2NPs to the SA film and found that adding 6 % (*w*/w) of SiO_2NPs increased TS by 30 MPa, whereas adding 9 % (w/w) of SiO_2NPs increased EAB by about 5 % only. With further addition, the TS and EAB of the SA membrane were negatively affected. M. Yang et al. (2018) found that the polyvinyl alcohol/SA–SiO_2NPs (0–9 %, w/w) composite film exhibited maximum values of TS and EAB at a SiO_2NP content of 6 % (w/w). This result suggests that elevated concentrations of SiO_2NPs may result in the collapse of the mechanical properties of the film, which may be caused by the aggregation of SiO_2NPs in the film matrix. The structural integrity of the membrane matrix network is then disrupted, reducing the cross-linking between the matrix components(W. Zhang, Ahari, et al., 2023). By contrast, Surendhiran et al. (2022) used SiO_2NPs to load turmeric essential oil (TEO) and combine CS to prepare composite films. Research indicates that the existence of SiO_2NPs influences the extension of active substance effectiveness over time. This distinctive characteristic feature can facilitate its development and utilisation in antibacterial or antioxidant food packaging.

4.3. Limitations of strategies

While the strategies discussed in the aforementioned studies can enhance the physical and functional properties of hydrogel films, recognizing their limitations is crucial. Not all enhancement methods are universally applicable, and inappropriate approaches can inadvertently weaken the mechanical strength of the films. For instance, the TS and EAB values of the composite films decreased to varying degrees when CS was mixed with Sardinella protein isolate, ZnONPs, bamboo leaf antioxidants, prululan polysaccharides, or carvacrol(Azaza et al., 2022; J. Liu et al., 2021; Xiao et al., 2022). A possible reason is that the unsuitable materials hindered the cross-linking of CS polymerization chains, reducing the interchain interactions. This process led to the deterioration of the mechanical properties of the films(Xiao et al., 2022). Therefore, in selecting an enhancement method to attain enhanced properties, the characteristics of the materials and the cross-linking mechanism should be fully considered.

5. Formation mechanisms of hydrogel films

5.1. Physical crosslinking

Hydrogels that form polymerization networks by physical forces are called physical hydrogels. These forces encompass hydrogen bonding, ion interaction, and van der Waals forces(Manzoor et al., 2022). Hydrogen bonds frequently play a part in hydrogel systems because polysaccharides and proteins generally include -COOH, -OH, or $-NH_2$ functional groups. Ion interaction refers to the mutual attraction between polymers or small molecules carrying opposite charges to form hydrogel systems, such as CS and SA hydrogels(Aksu Demirezen et al., 2023). In addition, proteins or polysaccharides may generate van der Waals forces due to their different functional groups, promoting the stability of polymer networks. Notably, physical hydrogels can be dissolved at a certain pH value, temperature, or ionic strength, directly influencing their mechanical properties and film development(Leyva-Jiménez et al., 2023). From another perspective, physical hydrogels can potentially stimulate responsiveness—for instance, cationic starch-carrageenan-sodium alginate hydrogels exhibit varying swelling properties depending on pH and temperature conditions(Cai et al., 2024). Similarly, carboxymethyl CS-based hydrogels that encapsulate tannins showed significantly higher release rates when the pH shifted from 5.5 to 7.4(Xiong et al., 2024). This pH-sensitive behavior suggests the potential for using such hydrogels as indicators of meat spoilage, where spoilage is often accompanied by a transition from acid to alkaline conditions. However, further investigation is required to refine these applications and validate their effectiveness.

5.2. Chemical crosslinking

The distinction between chemically and physically cross-linked hydrogels is determined by the presence or lack of covalent bonds in the three-dimensional network and the production of new polymers. Owing to differences in bond energies, chemically cross-linked hydrogels show greater strength. During chemical cross-linking, crosslinking

agent (e.g., glutaraldehyde, glutamine aminotransferase, formaldehyde, and dialdehyde) are commonly used to form covalent bonds resulting in hydrogels with strong physical strength and chemical stability(Saqib et al., 2022). However, there will also be potential safety issues for food packaging. Therefore, selecting the appropriate chemical crosslinking agent, which is influenced by the cross-linking mechanism, is particularly significant. Currently, hydrogels prepared by chemical cross-linking such as free radical polymerization (Ji et al., 2020a), Schiff base reaction(Ngwabebhoh et al., 2021a) and disulfide bonding(F. Cao et al., 2021) are promising candidates for packaging films (Fig. 3).

5.2.1. *Free radical polymerization*

Free radical polymerization is a well-established approach for hydrogel synthesis. This technique only requires changing the target monomer to prepare multifunctional hydrogels. The technique begins with the introduction of free radicals. These free radicals generate active sites to which monomer units bind to form chains. Subsequent steps involve continuous chain growth and polymerization to form macromolecules(Manzoor et al., 2022). Ji et al. (2020b) found that silver ions in the presence of ammonium persulfate can convert hydrogen into hydroxyl radicals. These silver ions catalyze radical polymerization to

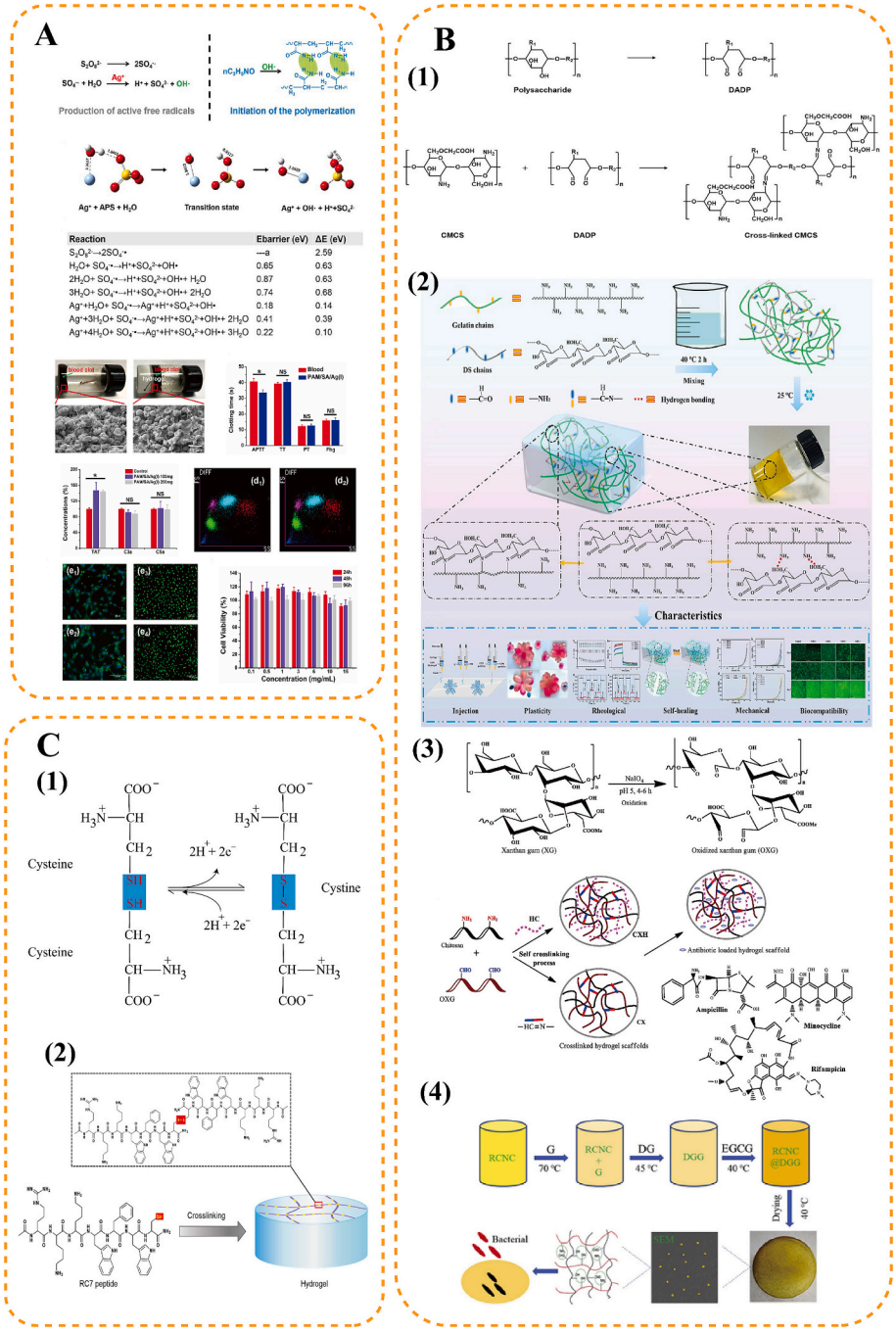


Fig. 3. (A) Hydrogel formed based on free radical polymerization: silver ion induced free radical(Haifeng Ji et al., 2020) Copyright 2020 ACS. (B) Hydrogels (films) formed based on Schiff base reaction:(1) DADP – CMCS (Wang, Huang, Zheng, et al., 2023b) Copyright 2023 Elsevier. (2) DS - Gelatin(Tianqi Cui, Yue Wu, Chunlei Ni, Yuxue Sun, & Jianjun Cheng, 2022) Copyright 2022 Elsevier. (3) Dialdehyde XG - CS(Fahanwi Asabuwa Ngwabebhoh et al., 2021) Copyright 2021 Elsevier. (4) Dialdehyde KGM – Gelatin(Wang et al., 2020a) Copyright 2022 Elsevier. (C) Hydrogel formed based on disulfide bonding reaction: cysteine as an example(F. Cao et al., 2021) Copyright 2021 Elsevier.

form hydrogels at room temperature. Hydrogels prepared using this method exhibit good histocompatibility, hemocompatibility, cytocompatibility, and immunocompatibility (Fig. 3A).

5.2.2. Schiff base reaction

Schiff base reaction, a nucleophilic addition reaction of carbonyl-containing aldehydes and ketones with primary amines, can produce a substance with dynamic covalent bonds. Studies have demonstrated that the aldehyde groups obtained after oxidation of some natural substances enable the preparation of hydrogels with satisfactory biocompatibility via Schiff base reaction. Wang, Huang, Zheng, et al. (2023a) prepared dialdehyde cross-linkers from corn starch (A-type starch), potato starch (B-type starch), alginate, pectin, and dextran and then crosslinked them with carboxymethyl CS via Schiff base reaction to form hydrogels (Fig. 3B). The experimental results showed that the cross-linking effect of these dialdehyde polysaccharides increased with their oxidation, which could be used to cross-link biomolecules containing amino groups and with sufficient biocompatibility and hemocompatibility.

Moreover, hydrogels prepared through these reactions can also be referred to as dynamic covalent cross-linked hydrogels with reversible covalent bonds. Their nature endows hydrogels with robust self-healing capabilities. Cui et al. (2022b) prepared a hydrogel carrier by oxidizing starch with sodium periodate to DS and reacting it with the Schiff base of $-NH_2$ in gelatin to generate dynamic imine bonds while cross-linking it synergistically with hydrogen bonds (Fig. 3B). In their study, dynamic rheology and structural analysis corroborated that this hydrogel has a stable and strong reticular structure. This structure exhibited the ability to regenerate rapidly after decomposition and collapse, showing self-healing ability. The hydrogel showed good pH-dependent controlled release of the active ingredient curcumin, which has a greater potential for drug or nutrient delivery. In addition to starch, dialdehyde XG was also synthesized by sodium periodate oxidation, and interpenetrating network hydrogels were prepared via Schiff base reaction with CS, combined with hydroxypropyl methylcellulose, in the study by Ngwabebhoh et al. (2021b) (Fig. 3B). Notably, Schiff base reactions have been studied for use in food packaging. L. Wang et al. (2020a) prepared films via Schiff base reaction of dialdehyde KGM with gelatin. The mechanical and functional properties of the material were enhanced through the use of rutin-functionalised cellulose nanocrystals and epigallocatechin gallate, respectively(Fig. 3B). The experimental results demonstrated that the introduction of rutin-functionalised cellulose nanocrystals improved the mechanical and barrier properties of the films. The films exhibited notable antibacterial activity against foodborne pathogens because of the presence of epigallocatechin gallate. These studies fully demonstrate that hydrogel systems constructed via Schiff base reaction can be potentially used in food packaging and thus deserve further investigation.

5.2.3. Disulfide bonding reaction

Disulfide bonding reaction is also a common method for constructing dynamic covalent cross-linked hydrogels. Disulfide bonds usually form via the coupling of two thiol groups under oxidizing conditions. Their formation can be reversed or prevented in the presence of a high concentration of reducing agents(Ye et al., 2020). Fig. 3C illustrates this mechanism using Cys as an example. Based on this, a PAF26 peptide derivative with Cys amino acids at the c-terminus was designed by F. Cao et al. (2021). An antibacterial hydrogel was synthesized by cross-linking the thiols in the Cys residues to each other to form disulfide bonds (neutral solution, oxidation). The experimental results demonstrated that the hydrogel had a strong cross-linked structure and good antibacterial ability against *Candida albicans* and inhibited the growth of *E. coli* and *S. aureus* (Fig. 3C). This finding provides a possibility for the application of disulfide bonding reaction in antimicrobial food packaging.

6. Preparation of hydrogel films

6.1. Casting forming

Casting forming is a process whereby a film-forming liquid is injected into the mold under atmospheric pressure. The material undergoes curing and molding into a film with the same shape as the inner cavity of the mold by polymerization (Fig. 4A). In a laboratory setting, Petri dishes often serve as moulds for the preparation of films. The procedure is divided into three steps: (i) preparation of the film-forming liquid, which needs to be cross-linked and without insoluble particles and air bubbles; (ii) casting of a fixed amount of the film-forming liquid in a mold (Petri dish, glass, etc.) to control the thickness and size of the film; (iii) drying of the film-forming liquid in natural conditions or in an oven to remove the solvent and form the film. This method is frequently selected for laboratory research due to its simplicity, cost-effectiveness, ease of operation, and capacity to yield uniform thin films(H. Yong, Xu, et al., 2022). However, the extended drying period and limited film formation involved in this method impede the smooth transition from laboratory settings to industrial-scale applications.

6.2. Layer-by-layer self-assembly

Layer-by-layer self-assembly requires the use of the alternating layer-by-layer deposition method, relying on weak interactions between molecules within each layer (electrostatic gravitational force, hydrogen bonding, ligand bonding, etc.). These interactions facilitate the spontaneous combination of layers, leading to the formation of fully structured and stable aggregates with specific properties(Kumar et al., 2020). In the preparation of polymer films, the electrostatic interaction between ions serves as the driving force for film formation. This technique is called electrostatic layer-by-layer self-assembly, also known as the electrostatic alternating deposition technique. This approach involves the following steps: (i) cleaning the substrate (commonly quartz flakes) and surface activation; (ii) submerging the substrate in alternating cationic and anionic charge solutions; and (iii) repeating (ii) as needed (Fig. 4B). Surface activation involves modifying the substrate surface to render it either hydrophilic or hydrophobic and either positively or negatively charged. The treated substrates are stored in ultrapure water until needed. During preparation, if the substrate has a positive (negative) charge, the initial monolayer will be polyanionic (cationic). Following each immersion, the substrate is rinsed with ultrapure water to eliminate any excess material. This method allows the precise control of the structure and thickness of the self-assembled film. Biofunctional macromolecules, conductive polymers, and photoreactive polymers can be easily introduced into the film owing to the nonspecific nature of electrostatic interactions, resulting in a film with biofunctional, conductive, and photoactive properties(Almasi et al., 2021).

Moreover, the films produced via this method offer enhanced control of the release of active compounds from the film into the foodstuff. B. Liu et al. (2023) used maize alcohol-soluble protein nanoparticles and pectin as encapsulation carriers. Layer-by-layer self-assembly was employed to encapsulate probiotics in pectin, investigating the viability of cells under different conditions (freeze-drying, gastrointestinal digestion, heating, and storage). A high survival rate (>95 %) was obtained during preparation; activity could be improved during heating and storage; during gastrointestinal digestion, the dissolution of pectin could expose the cells to harsh environments faster if the coating layer was insufficient; however, this adverse effect could be counteracted if the coating layer was adequate. Thus, layer-by-layer self-assembly plays an important role in maintaining the activity of substances within the film.

6.3. Electrospinning

Electrospinning technology, also known as electrostatic spinning

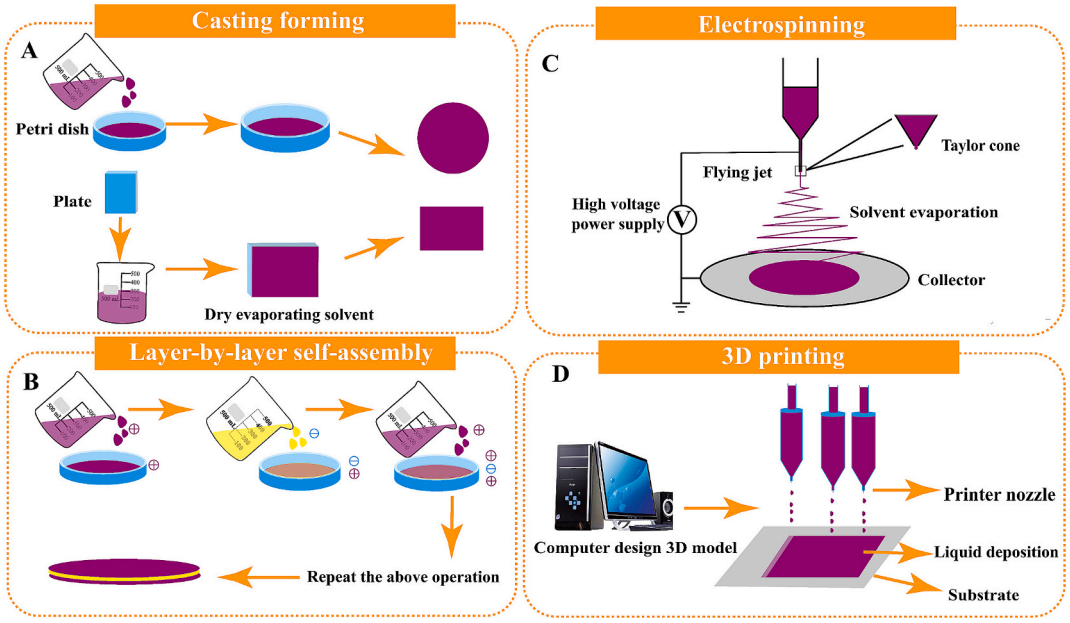


Fig. 4. Illustration of film formation method.

technology, is an innovative technique for the preparation of nanofiber films. This method entails the utilisation of polymer solutions to generate jets under the influence of a strong electric field, resulting in the formation of continuous fibers with diameters in the submicron to the nanometer range(Fig. 4C). Electrospun fibers are characterized by small pores, high specific surface area, and adjustable porosity. These characteristics are conducive to the immobilization and release of active compounds(W. Zhu et al., 2022). Electrostatic spinning substrates usually consist of polymers or biomolecules such as polysaccharides and proteins. Thus, electrostatically spun fibers demonstrate considerable potential for use in reactive food packaging.

In addition, electrostatic spinning is a nonthermal process that facilitates the maintenance of structural stability, particularly for substances with low thermal stability at high temperatures, such as essential oils and anthocyanins. Zhang, Zhang, et al. (2022) prepared polyvinyl alcohol/β-cyclodextrin nanofiber active packaging films wrapped with *Zanthoxylum bungeanum* essential oil by electrostatic spinning for the storage and preservation of strawberries and cherries. This active packaging film extended the freshness of strawberries and cherries to 10 days. Vafania et al. (2019) also prepared CS/gelatin nanofiber films containing thyme essential oil by needle-less electrostatic spinning. In their study, the nanofiber film exhibited up to 92 % encapsulation efficiency of the essential oil, and the loading capacity increased with the concentration of the essential oil; the encapsulated essential oil was bactericidal to *C. perfringens* and exerted no effect on the color and sensory properties of the sausage. The utilisation of electrostatic spinning for the preparation of activated food packaging films has demonstrated immense potential across diverse domains, both (i) fruit and vegetable and (ii) meat products.

6.4. 3D printing

3D printing is a rapid prototyping technology based on 3D mathematical model data(Fig. 4D). In contrast to layer-by-layer self-assembly process, 3D printing constructs objects by physically stacking them layer by layer. The prominent characteristic of this technique is its customization capacity, including shape designs, sizes, structures, and porosity levels. Li, Jiang, Zhou, et al. (2022) prepared CS, mulberry anthocyanin, and lemongrass essential oil interlayers by 3D printing. Tapioca starch was used as a protective layer to form the indicator film. In the study, the indicator film showed significant antioxidant and antimicrobial properties; in addition, the color changed from red to gray-blue when the pork was cooled and spoiled. The RGB(Red, Green, Blue) values of the indicator film were analyzed in conjunction with smartphone software to automatically analyze and determine the freshness of the pork. Li, Jiang, Wang, et al. (2022) devised a type of preservation card by 3D printing. This insurance card was prepared by co-precipitating encapsulating lemongrass essential oil with β-cyclodextrin and then mixing them with SA to produce a biological multifunctional preservation card by 3D printing. This technique can realize air-conditioning packaging for strawberries and extend the shelf life. Built upon the principles of 3D printing, this method can mitigate shape and size errors in each test sample. However, 3D printing has certain material requirements. Natural hydrogels have better biodegradability and biocompatibility, whereas synthetic hydrogels exhibit superior mechanical properties and have less batch-to-batch variability, rendering them suitable for 3D printing films. Therefore, the selection of suitable hydrogel film materials is particularly important for 3D printing.

7. Applications in food packaging

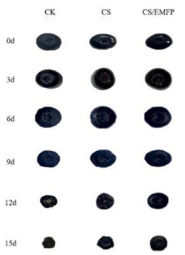
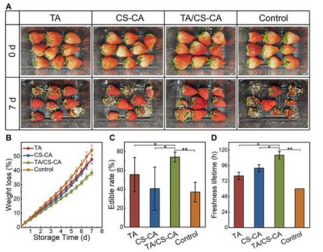
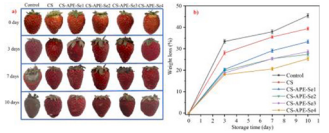
The development of hydrogel films prepared from polysaccharides and proteins has attracted significant attention from researchers in the field of food packaging. Research in this field is divided into enhanced film mechanical properties, antibacterial preservation, edibility, and intelligent indication aspects (Table.2).

7.1. Food preservation films

The freshness of food encompasses a comprehensive set of indicators, including the color, odor, taste, and nutritional value of food. Food degradation occurs during storage, transport, and sales. This process is attributed to various factors, including microorganisms, oxygen exposure, and enzymatic activity, thus reducing its freshness and causing food waste. As previously mentioned, hydrogels have a complex three-dimensional structure and can be used as carriers of freshness-preserving substances (active substances, nanoparticles, etc.). Therefore, hydrogel films have a great potential for both development and improvement within the food supply chain.

Catechins, a class of phenolic active compounds extracted from

Table 2
Research on common applications of natural polysaccharide/protein hydrogel films in the food field.

Materials	Additives	Film-forming method		Application Research		References
CS, golden mushroom foot polysaccharide	–	Solution casting technique	Blueberries	**15 days**: weight loss rate, total number of colonies, decay rate and respiration intensity were lower than the control group.		(J. Zhao et al., 2023)
CS	Citric acid, tannic acid	Dipping method	Strawberries	**7 days**: minimum rate of weight loss (38.4 %) and maximum rate of edibility (74.1 %).		(Chang et al., 2023)
CS	*Andrographis paniculata* extract, selenium nanoparticles	Solution-casting method	Strawberries	**10 days**: lower spoilage and weight loss of strawberries compared to the control group.		(Thanh Huong et al., 2023)

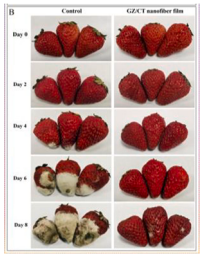
Materials	Additives	Film-forming method		Application Research		References
Gelatin, zein	Cinnamaldehyde, thymol	Electrospinning	Strawberries	**4 days**: Control group started to decay; **6 days**: Decay deepened in the control group, the packed group maintained a good appearance, color and fullness; **8 days**: complete decay in the control group and the beginning of decay in the packaging group; shelf life extended by 4 days.		(X. Wu et al., 2023)

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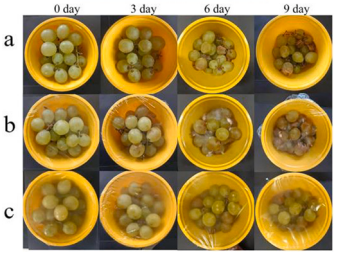
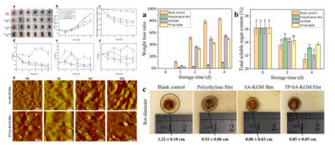
Table 2 (*continued*)

Materials	Additives	Film-forming method		Application Research		References
CS, pullulan	*Artemisia annua* essential oil	Flow casting method	Grape berries	**10 days**: maintain freshness in appearance, less mold spots; lower weight loss rate & nice hardness.		(Wendan Chen et al., 2023)
SA, KGM	Tea polyphenols	Casting forming	Beef, apple	**Beef, 10 days**: total colony count meets spoilage criteria, extending freshness by 2 days compared to control; **Apple, 4 days**: maintenance of high soluble solids content and reduced wound decay diameter.		(Shancan Wang, Li, He, et al., 2023)

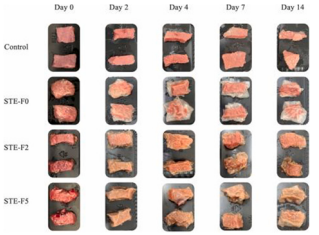
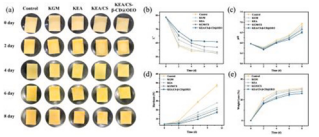
Materials	Additives	Film-forming method		Application Research		References
Gelatin, GG, carboxymethyl cellulose	Shallot tunic extracts	Casting forming	Beef	**14 days**: good inhibition against *E. coli*, *Coliforms*, *S. aureus*; better inhibition of lipid oxidation; flesh stays bright red.		(Nguyen et al., 2023)
KGM, CS	Oregano essential oil, anthocyanins	Solvent casting method	Cheese	**8 days**: relatively minor color change; slow pH rise probably due to inhibition of yeast and mold growth; lower hardness and better texture maintenance; lower weight loss rate.		(Zhang, Cao, et al., 2023)

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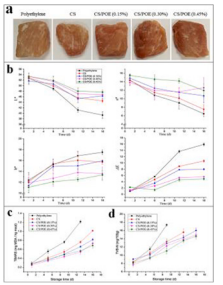
Table 2 (*continued*)

Materials	Additives	Film-forming method		Application Research		References
CS, wheat starch and pea starch	*Portulaca oleracea* extract	Casting forming	Frozen pork	**6 days**: **Polyethylene film group**: meat samples had turned off-white, the red color disappeared and gave off a strong rotten smell; **CS film group**: meat had turned slightly white with a slight rotten smell; **Composite film group**: meat was redder, no rotten smell, TVB-N and thiobarbituric acid lipid were significantly lower than other experimental groups.		(Fan et al., 2023)

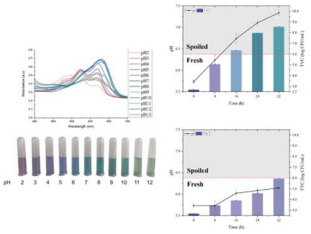
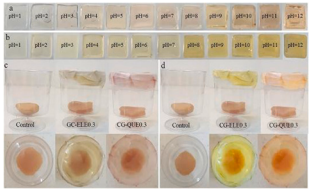
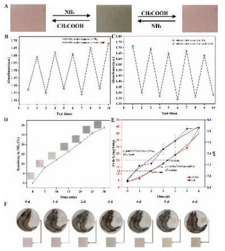
Materials	Additives	Film-forming method		Application Research		References
D. zingiberensis starch	Butterfly bean flower anthocyanin	Solution casting method	Chicken	**Room temperature**, **≥ 16 h**: deterioration, blue-violet → blue → blue-green; **4 °C, 32 h**: no deterioration, maintain violet.		(Wang, Yang, Deng, et al., 2023)
kappa-carrageenan (CG)	Quercetin(QUE) / eucalyptus leaf extract(ELE)	Casting forming	Chicken	**25 °C, 3 days:** **CG-QUE 0.3:** red to orange; **CG-ELE 0.3:** green to yellow.		(Mirzaei et al., 2024)
CS, starch	Rose anthocyanins	Casting forming	Alive shrimp (*Litopenaeus Vannamei*)	**4 °C, ≥2 days**: deterioration, pink → colorless → yellow.		(L. Zheng et al., 2023)

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Table 2 (*continued*)

Materials	Additives	Film-forming method		Application Research	References
Materials	Additives	Film-forming method		Application Research	References
CS, chitin	Eggplant anthocyanins	Casting forming	Pork	**25 °C, ≥24 h:** deterioration, light blue → dark blue →green.	(Wang, Xie, Tang, et al., 2023)
Collagen hydrolysate, CS	Red cabbage extracts	Casting forming	Shrimps (*Penaeus chinensis*)	**Control group, 4 °C, ≥2 days:** red (fresh) → purple (imminent spoilage) → blue (spoilage); **Coated group, 4 °C, ≥5 days:** film turns purple, shelf life extended by 3 days.	(G. Ren et al., 2023)

natural plants such as tea leaves, exhibit high antioxidant capacity. Huimin Yong, Hu, et al. (2022) found that four catechin monomers ((−)-epigallocatechin, (−)-epigallocatechin, (−)-epigallocatechin gallate, and (−)-epigallocatechin gallate) could be bound to dialdehyde starch via an acid-mediated coupling reaction and as such are stable antioxidants. In alignment with this study and the underlying mechanism, H. Yong, Xu, et al. (2022) prepared a novel antioxidant packaging film with catechin-dialdehyde starch coupling and CS as used in the antioxidant preservation of sunflower oil. The films had a higher UV–visible light barrier, water vapor and oxygen barrier, lower swelling, stronger TS, thermal stability, and antioxidant activity. In addition, the conjugated composite membranes effectively suppressed the elevated levels of peroxide value and thiobarbituric acid reactive substance (TBARS) in sunflower oil during storage. This observation suggests that the approach demonstrates good antioxidant capacity and potential application in food preservation.

Meanwhile, antimicrobial films can also effectively be used to maintain food freshness and extend shelf life. Liu, Wang, et al. (2022) fabricated antibacterial hydrogel packaging films by electrostatic spinning. Gelatin and CS were used as film-forming materials, with 3-phenyllactic acid as the additive. The hydrogel film with 1 % 3-phenyllactic acid exerted a significant inactivation effect on foodborne pathogens *S. aureus* and *E. coli*, as well as lengthened the shelf life of cold chicken to 4 days. Zhang, Chen, et al. (2022) devised bilayer membranes with CS and zein via layer-by-layer casting. CS was enveloped with pectin-based nanoparticles loaded with curcumin and Nisin, forming an inner hydration layer to provide broad-spectrum antibacterial properties. Meanwhile, zein was used as a hydrophobic outer layer to provide moisture resistance. The incorporation of nanoparticles was found to enhance antioxidant properties and exhibit efficacious antibacterial activity against *S. aureus* and *E. coli*, thereby contributing to the preservation of food.

Extending the storage life of food by improving the antioxidant and antibacterial capacity of packaging films mainly constitutes is main current research direction. In some studies, additives that can absorb or release gases such as oxygen, carbon dioxide, and ethylene, apart from other gases, into the film have been incorporated to maintain the freshness and extend the shelf life of foods(Mao et al., 2023). The mechanism entails creating a gas environment that hinders enzymatic reactions (lipid hydrolases, lipoxygenases, etc.) within the food or the growth of spoilage bacteria. This process effectively prolongs the freshness duration of the food (Wang, Liu, Zhao, et al., 2023). In essence, this approach is also aimed at antioxidant or bacterial inhibition. The application of hydrogel films for modified atmosphere packaging requires more research to fill this part.

7.2. Edible films

As the name suggests, edible film packaging incorporates edible components into the packaging. Edible packaging can exist as a thin layer formed directly on the surface of the food product (edible coating), or individually formed as a sheet/film (edible film). It is then wrapped around the surface of the food product(Jeya Jeevahan et al., 2020). The coating may also be referred to as a “film” because of its formation in situ, covering the surface of the food product. In summary, edible films represent a functional and edible protective layer wrapped around the surface of a food product. The introduction of the edible film concept represents a transformative improvement in packaging, pushing it toward a new standard that increasingly blurs the boundaries between food, preservation, and packaging.

Edible films are prepared from natural polymers of polysaccharides, proteins (animal or plant), lipids, or a combination of these materials. The natural polysaccharide and protein hydrogel films described exhibit significant potential in edible film field. Wang, Li, Meng, et al. (2023) used KG and gelatin as film-forming base materials. Honeysuckle leaf extract was then added as an active substance to prepare an active edible

film by casting forming. With the addition of honeysuckle leaf extract, the inhibitory rates of *E. coli* and L. *monocytogenes* approached 90 %, and the inhibitory rate of *S. aureus* reached 96 %. These results demonstrate the potential of the active edible film to extend the shelf life of foods by providing desirable antioxidant and antibacterial effects. Dursun Capar (2023) (using SA, grape leaf extract, and quercetin), Molnar et al. (2023) (using CS, gum Arabic, and grape seed extract), and Karkar et al. (2023) (using CS and *Nigella sativa* L. extract) prepared different composite edible films, all of which exhibited positive antimicrobial and antioxidant activities for maintaining the texture and prolonging the freshness of food.

From these studies, the edible films may be similar to the aforementioned preservation films but are distinct entities. Edible films tend to be more of a subset of preservation films. Because of variations in material, functionalities, and the inherent nature of their roles. Compared with preservation films, edible films require more comprehensive consideration and rigorous processing with regard to the material selection, preparation, and performance optimization of the film. The reason is that the safety of edible films must be ensured as they are intended for direct human consumption. These films also alleviate the waste disposal problems. Further, even if not ingested, the film exhibits faster degradation, compared with synthetic and biodegradable packaging materials, significantly mitigating environmental hazards(Jeya Jeevahan et al., 2020). However, edible films also share challenges associated with their weak mechanical properties and barrier properties. Notably, the dosage of natural additives and the use of nanoparticles in edible films have to be carefully considered. Although natural additives are safer than synthetic additives, they may be toxic if reactions occur or if their concentrations are excessively high during preparation(Gaspar & Braga, 2023). Moreover, the utilisation of nanoparticles in edible film research has thus far been relatively limited. A study indicates that the ability of nanoparticles to penetrate human cells increases with decreasing diameter. For example, particles measuring 100 nm can easily penetrate cells, those measuring 40 nm can enter the cell nucleus, and those smaller than 35 nm can cross the blood-brain barrier (Bumbudsanpharoke et al., 2015). Compared with larger-sized particles, smaller-sized particles can exhibit higher catalytic activity and may have greater enhanced adsorption rates, binding capacities, and propensity to produce reactive oxygen species(Vlachogianni & Valavanidis, 2014).

Indeed, nanotechnology provides significant potential for enhancing the mechanical properties and preservation capacity of edible films. However, publicly available data remain scarce, particularly studies on the safety and toxicological effects of nanoparticles. Therefore, the development of a safe and high-performing edible film would represent a substantial contribution to the food packaging field.

7.3. Intelligent freshness indicator films

By definition, intelligent food packaging refers to a packaging system designed to exhibit observable changes, such as swelling, degradation, or color change in the packaging component when the food is subjected to external stimuli. These stimuli include temperature, pH, and light. This real-time response is indicative of the quality or freshness of the food within the package. As the food produces interacts with endogenous enzymes and exogenous microorganisms, it produces carbon dioxide, volatile alkaline nitrogen, and hydrogen sulfide. The pH of the surrounding environment directly changes(P. Shao et al., 2021). Thus, the use of pH-responsive indicators to signify has drawn research attention. This method facilitates a straightforward change in package color, leading to a more intuitive and visibly discernible indication of freshness (Fig. 5).

In smart indication packaging, indicators are categorised into chemically synthesized and natural substances. Among them, bromophenol red, bromophenol blue, bromocresol violet, and bromocresol green–methyl red are commonly used chemical class colorants. They

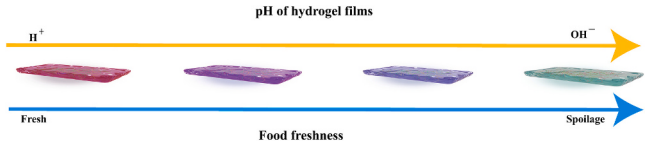

Fig. 5. Schematic diagram of pH-responsive hydrogel film color indication of freshness.

offer several advantages, such as sensitivity of reaction, apparent change in color, and high stability, among others(P. Shao et al., 2021). However, chemical reagents are accompanied by potential safety because of the possibility of leaching, migration, and subsequent contamination of food. With this problem considered, noncontact packaging seems to be a preferable method to avoid direct contact between chemical reagents and food. Nonetheless, this review advocates the preference for safe and biodegradable natural indicators. The reason is that consumers favor natural substances, facilitating the commercialization of intelligent packaging. With their wide availability and high level of safety, natural pigments (anthocyanins, curcumin, betaine, etc.) are a well-chosen option for chromogenic agents. However, their stability and indication sensitivity should be improved compared to synthetic pigments. For instance, anthocyanins are easily degraded by light, heat, and oxygen, thus shortening the service life of their indication function; some natural pigments exhibit reversible color changes leading to inaccurate results, which limits the further development of natural pigments in intelligent packaging(Z. Shao et al., 2024). Consequently, improving the stability and sensitivity of natural pigments is a key research issue.

Currently, some methods can improve the stability of natural pigments, such as liposomes, microcapsules, Pickering emulsions, and nanoparticle encapsulation systems. In addition, the natural polysaccharide or protein hydrogels proposed in this review are often used in research. Xie et al. (2020) embedded blueberry anthocyanin–chondroitin sulfate nanocomplexes in a hydrogel system prepared using κ-carrageenan. The stability test results revealed that nanocomplexes effectively delayed the degradation of anthocyanins at lower pH levels, and the presence of κ-carrageenan hydrogels further enhanced their retention in various adverse environments (temperature, pH, and metal ions).

The modification of natural pigments can also lead to a more stable indicator. Y. Wu and Li (2023) modified roselle anthocyanins with acetic acid, which significantly enhanced their thermal stability, antioxidant capacity, photostability, and pH stability. The retention of the modified anthocyanins increased from 30 %–50 % to 50 %–90 % under alkaline conditions. A bilayer film with both indication and antimicrobial properties was prepared by combining modified roselle anthocyanin. GG served as the internal indicating layer, and sodium carboxymethyl cellulose-starch-Nisin acted as the external antimicrobial layer. The experimental results proved that the bilayer film exhibited enhanced mechanical properties, barrier properties, and hydrophobicity. In addition, the film detected the freshness of chicken breast in real time and extended its shelf life by 2 days. Thus, the bilayer film showed potential as an antimicrobial indicator packaging film. Notably, the internal indicator layer was not obscured by the external antimicrobial layer, demonstrating enhanced sensitivity(Y. Wu & Li, 2023). This improvement could be attributed to the Nisin nanoparticles with enhanced color rendering. Liu, Zhang, et al. (2020) similarly concluded that bilayers containing *Clitoria ternatea Linn* anthocyanin with TiO_2 nanoparticles showed a pronounced color change. Therefore, the presence of nanoparticles can potentially heighten the color change sensitivity of anthocyanins.

In addition, binary complex pigments have superior color change indication effects compared with single pigments. In the study by A. Duan et al. (2022), the CS–starch–gelatin composite film acquired superior EAB, TS, WVP, antioxidant properties, freshness retention, and color change aspects when raspberry anthocyanin (*Rubus idaeus L.*) and

curcumin (*Curcuma longa L.*) were combined, exhibiting potential as a smart packaging and indicator. The stability of the coupling compound (bovine serum albumin/casein) has also been improved by combining ultrasound and glycosylation. A complex has also been formed using carboxymethyl cellulose to encapsulate anthocyanins, indirectly and effectively enhancing the thermal stability of anthocyanins(Cui et al., 2023). This technique provides insights into improving the stability of natural pigments.

In conclusion, pH-indicating intelligent packaging can advance toward commercialization when methods to preserve natural colors for long periods are developed. The utilisation of film color with mobile phone software has also been integrated to obtain more detailed information about food quality within the packaging. These data include food freshness ratings(Lee et al., 2019), which offer a more intuitive shopping guide than traditional food shelf life, reflecting the convenience and novelty of intelligent indicator packaging.

8. Advantages and challenge

In light of current environmental challenges, traditional plastic packaging is increasingly inadequate to meet the needs of sustainability. The primary advantage of using natural polysaccharides and proteins to form hydrogel films for food packaging is their biodegradability, aligning with the growing emphasis on eco-friendly and sustainable practices. Furthermore, the unique three-dimensional mesh structure of hydrogels provides an excellent matrix for loading and controlled release of active substances, making them ideal for antimicrobial and intelligent packaging applications. Further research and innovation in natural hydrogel films hold the potential to revolutionize food packaging.

However, acknowledging the current state of research on natural hydrogel films remains largely confined to the laboratory is crucial. Significant challenges must be addressed before these materials can be successfully deployed in production, transportation, and distribution systems. These challenges primarily concern the film-forming materials, methods of production, and mechanical strength of hydrogel films, all of which must meet the rigorous requirements at each stage of the supply chain. According to study results presented in Table 1, the mechanical and functional properties of hydrogel films often fail to meet the minimum criteria established by Chinese standards for food packaging or lidding films, such as GB/T 28118–2011, GB/T 30768–2014, GB/T 41220–2021, and DB43/T 1168.4–2019. For instance, these standards demand a TS of $\geq$30 MPa, EAB of $\geq$30 %, WVP of $\leq$25 $g{\cdot}m^{-2}{\cdot}h^{-1}$, OP of $\leq$12 $cm^3{\cdot}m^{-2}{\cdot}h^{-1}{\cdot}0.1\ MPa^{-1}$, etc. Achieving these benchmarks necessitates improvements in the inherent properties of natural polysaccharides and proteins and the optimization of film-forming techniques. Developing hydrogel films with enhanced mechanical strength, flexibility, and barrier properties is a fundamental priority for researchers in this field. Another major obstacle is scaling up the industrial production methods for hydrogel films. Most film-forming techniques explored thus far are suited for small-scale or laboratory-scale applications. Industrial production, however, requires processes capable of high throughput and consistent quality. Current plastic packaging is primarily manufactured using techniques such as blister, injection, blow and extrusion molding. These processes impose stringent demands on the thermal stability and oxidative resistance of raw materials, properties that natural polysaccharides and proteins often lack in their unmodified state. The rich functional groups inherent in natural polysaccharides and proteins provide opportunities for chemical and physical modifications. Tailored modifications can enhance thermal and oxidative stability, enabling these materials to withstand the demands of industrial-scale processing, while expanding the range of available packaging forms. The issue of water activity of hydrogel films encountered by their industrialization also needs to be addressed. Hydrogel films inherently possess certain water activity, which facilitates the migration of substances from packaging materials to packaged foodstuffs, leading to undesired interactions and limiting their use for moist products. One proposed solution involves the development of contactless packaging of speciality food products. However, this approach significantly narrows the range of applications.

On the other hand, although the demand for greener packaging alternatives is growing among consumers, convincing the average consumer of the safety and efficacy of hydrogel food packaging films is a critical challenge. More literature needs to be published on the safety, sustainability, and non-contamination of hydrogel food packaging films. They should answer questions such as how long can the films keep food fresh, are edible films actually edible, what are the detection limits of smart indicator films, and does a “dose-effect relationship” exists between the indicator and food.

9. Conclusions

Natural polysaccharide and protein hydrogel films offer significant potential as innovative food packaging solutions, addressing the environmental pollution caused by petroleum-based plastic packaging. These materials are notable for their availability, biodegradability and compatibility with food products. In recent years, advances in research on natural polysaccharides, proteins, active substances, and nanoparticles have enabled the development of high-performance hydrogel films for applications such as antimicrobial preservation, edibility and intelligent packaging. Additionally, diverse cross-linking mechanisms and film-forming techniques present exciting opportunities for exploration in this field. Notably, while the concept of hydrogel food packaging films is groundbreaking, their practical implementation faces challenges particularly regarding durability and industrial scalability. The current limitations of small-scale production methods must be addressed, emphasizing the development of thermoplastic production techniques to enable large-scale industrial production. In conclusion, although natural polysaccharide and protein hydrogel films are not yet widely commercialized as food packaging solutions, existing research demonstrates their promise and effectiveness. Continued efforts to refine their design strategies and improve film-forming methods are essential for achieving sustainable, non-polluting food packaging solutions in the future.

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CRediT authorship contribution statement

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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.2025.102174.

Data availability

No data was used for the research described in the article.

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