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Polysaccharide-based pH-responsive intelligent halochromic food packaging materials: a review

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The pH-responsive intelligent food packaging materials change color in response to pH variations induced by food spoilage and the release of microbial metabolites. Recently, polysaccharides have gained significant attention as potential matrices for such systems due to their numerous benefits compared to conventional fossil fuel-based packaging materials, including renewability, biodegradability, biocompatibility, relative abundance, film-forming ability, nontoxicity, and tailorable surface properties. Natural halochromic dyes have emerged as food freshness indicators (FFIs) due to their low cost, nontoxicity, eco-friendliness, rapid responsiveness, and ease of use. Numerous studies have reported the use of polysaccharides and natural halochromic dyes for fabricating pH-responsive polysaccharide packaging (PRPP) materials; however, the roles and mechanisms of action of these systems in monitoring food freshness have not been systematically analyzed. Therefore, the current study aims to fill a gap in an underexplored area of the literature by consolidating recent findings on PRPP materials and natural dyes for real-time detection of food freshness. This review comprehensively covers the structure and properties of polysaccharides and natural halochromic dyes, film fabrication techniques, mechanisms of food spoilage detection, and state-of-the-art intelligent food packaging applications of PRPP materials. Furthermore, this review discusses current regulations, safety considerations, challenges, and future perspectives of PRPP materials.

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Sustainability spotlight

Polysaccharide-based food packaging has recently emerged as an alternative to petroleum-based synthetic plastics owing to their renewability, nontoxicity, eco-friendliness, relative abundance, and sustainability. Polysaccharide-based intelligent food packaging incorporating natural halochromic dyes will enable real-time monitoring of food freshness. These packaging materials can act as an early indicator of food spoilage, helping consumers use food, especially perishables, without discarding them after long periods in refrigerators. These packing materials will contribute to sustainability by supporting the achievement of numerous United Nations (UN) sustainability development goals (SDGs), including zero hunger (2), good health and well-being (3), sustainable cities and communities (11), responsible consumption and production (12), and climate action (13).

1 Introduction

The primary functions of food packaging include containment, communication, convenience, and protection, which are important for slowing down food deterioration.[1] Currently, non-renewable fossil-based packaging materials (FPMs) are widely used in the food packaging industry due to their versatility, durability, low cost, tailorable design, excellent food preservation capabilities, and ease of manufacturing and handling.[2] Recently, the demand for FPMs has skyrocketed, with 3.2×10^6 tons being manufactured yearly.[3] However, FPMs can cause severe "white pollution" and high carbon dioxide (CO_2)

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emissions, contributing to the greenhouse gas (GHG) effect and consequently affecting the global climate.[2] Moreover, the excessive use of FPMs in food packaging has led to detrimental environmental and health implications over the past few years. As a result, there has been considerable interest in alternative food packaging materials to address the overreliance on FPMs.[4] Biodegradable natural polymers (BNPs), including polysaccharides, lipids, and proteins, have recently emerged as substitutes for FPMs due to their intriguing characteristics, such as renewability, relative abundance, nontoxicity, high mechanical and chemical properties, and switchable structures.[5]

Among many BNPs, polysaccharides are the most prevalent macromolecules in the biosphere, and their source can be of animal, plant, algae, or microbial origin.[6] Fig. 1 illustrates the major sources of polysaccharides and their examples used in the manufacturing of food packaging materials. Polysaccharides are generally composed of similar or different kinds of monomers linked together to form linear, branched, or crosslinked chains.[6] The presence of reactive functional groups, including hydroxyl (–OH), amine (–NH_2), amide (–$CONH_2$), carboxylic (–COOH), and carbonyl (–C=O) in their structure qualifies them for derivatizing into various products for packaging applications.[7,8] Polysaccharides exhibit improved mechanical and chemical properties, aesthetics, and ability to act as semi-permeable barriers to water and O_2, thereby extending the shelf life and quality of foods.[9] The application of polysaccharide-based packaging materials (PPMs) in the food industry also offers several advantages, as they help reduce the carbon footprint of the food industry. Some PPMs are incorporated with phytochemicals, prolonging the freshness of food.[2] Polysaccharide-based food packaging is approved by the United States Food and Drug Administration (US FDA), permitting modifications, including edible films, coatings, aerogels, and active and intelligent packaging, to preserve the quality and safety of food products during storage and transportation.[10] The sources, structures, properties, and food packaging applications of PPMs are summarized in Table S1 (see the SI).

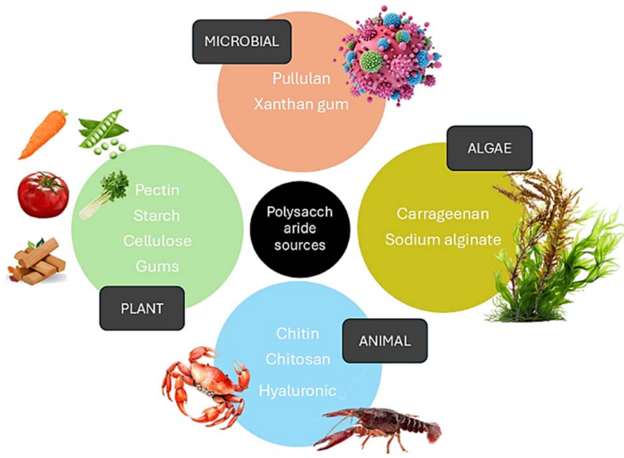
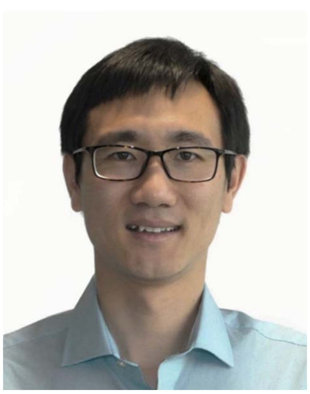


Fig. 1 Different categories of polysaccharides and their sources.

Over the past few years, polysaccharide-based intelligent packaging materials have garnered immense interest, as they not only preserve food but also monitor its physicochemical properties, including moisture content, pH, temperature, and food quality.[11,12] The properties of PPMs can be further enhanced by incorporating antimicrobial agents to protect against microbial contamination.[11,13] According to the European Union (EU), intelligent packaging is defined as packaging that tracks the quality of food being packed or surrounded.[7] Intelligent packaging enables communication with producers, supply chains, and stakeholders regarding the quality of the food inside.[14] Polysaccharide-based intelligent packaging materials generally increase shelf life, enhance food quality, and reduce the weight of the packaging.[7] Intelligent packaging also monitors the conditions of food without any interaction.[15] Several sensors have been embedded in intelligent packaging materials, operating on a mechanical, chemical, enzymatic, or immunochemical basis.[16]

Among many intelligent packaging materials, PRPP has recently attracted attention. Interestingly, PRPP can be



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manufactured by amalgamating one or more biopolymers and is highly popular among retailers, the food industry, and consumers because it is cost-effective, non-invasive, and highly efficient, and many of its products are food-grade.[17] The PRPP provides real-time information about food quality through pH-sensitive indicators.[18] There are two main types of pH sensors being employed: electrochemical and colorimetric systems. Colorimetric systems utilize natural colorants or synthetic dyes to detect pH changes within the packaging, whereas electrochemical systems employ ion-selective electrodes (ISEs) prepared from carbon, polymeric materials, and metals and metal oxides to detect pH-active chemicals, thereby generating an oxidation or reduction current.[19] Electrochemical systems offer numerous advantages, including high sensitivity and selectivity, rapid response times, and simple detection.[20] However, *in situ* pH measurement is not feasible with electrochemical methods due to their indirect detection. On the other hand, colorimetric systems with FFIs have been considered for direct pH detection in food.[21] FFIs are halochromic dyes that change color with pH; hence, they are widely employed in PRPP.[22] The integration of halochromic properties and reversible color changes in response to environmental stimuli that alter the pH introduces a dynamic dimension to the perishable food packaging industry, addressing the issue of food wastage and promoting food security.

Halochromic dyes are sensitive, affordable, and more favourable for use, enabling easy freshness monitoring.[23] Both synthetic and natural halochromic substances have been employed in intelligent food packaging to monitor the freshness of foods inside by observing the color change.[24] Synthetic halochromic dyes used in food packaging include polyaniline,[25] bromocresol green, 2,4,6-trinitrophenyl,[26] bromophenol blue, bromocresol purple,[27] chlorophenol red, and cresol red.[28] However, these synthetic colorants exhibit several issues, such as the potential to cause health issues in consumers, the ability to alter the sensory properties of the food,[27] and risks to the environment upon disposal.[29] In contrast, natural halochromic dyes are gaining more attention among consumers due to their aforementioned features.[24,30,31] Food packaging systems utilize the halochromic effect of natural pigments to detect the real-time freshness of food.[32] Examples of natural halochromic dyes used in PRPP are curcumin, anthocyanin, phycocyanin, betalain, quercetin, shikonin, and alizarin.[24]

This review aims to provide a comprehensive overview of recent advancements in PPRPs, with a preliminary focus on intelligent packaging systems, covering the period from 2020 to 2025. The major polysaccharide sources, the mechanism related to the pH sensitivity of PRPPs, the natural halochromic dyes used as FFIs, and their properties have also been discussed. Most importantly, this article highlights the current fabrication techniques, applications related to the PRPPs, regulations, safety concerns, and challenges and prospects in this emerging field. Therefore, to the best of our knowledge, this is the first attempt to discuss the unexplored area of using PRPP in intelligent natural halochromic food packaging applications. Moreover, the findings from the recent literature will guide process engineers, material scientists, researchers, and food technologists in the packaging industry in designing sustainable packaging solutions to enhance real-time monitoring of food safety and quality.

2 Structures, properties, sources, and food freshness indicator (FFI) mechanisms of natural halochromic dyes

Natural halochromic dyes are highly conjugated aromatic systems that contain various functional groups, including –OH, –COOH, and –C=O, which absorb light, causing a pH-dependent shift in their electronic structure and altering their color. Shikonin, curcumin, phycocyanin, quercetin, betalain, and alizarin are the most common natural halochromic dyes used as FFIs. Table S2 summarizes the major source(s), structural features, and color variations against pH of the most common natural halochromic dyes used in pH detection (see the SI).

Halochromic pH-responsive dyes generally serve as smart detectors, indicating real-time food freshness and ensuring food safety and quality, similar to biosensors.[22] In biosensor technology, there are two major components: receptors and transducers. Receptors recognize target analytes, allowing transducers to measure biochemical signals through their interactions. The smart detectors are categorized into two types: direct detectors and indirect detectors. Among them, halochromic dyes are classified as direct detectors because they switch color in response to pH variations resulting from changes in the chemical environment.[33]

When food spoils, microorganisms grow inside, and their metabolic activities alter pH through protein decomposition, enzymatic reactions, or chemical processes associated with spoilage. Furthermore, the biochemical reactions produce volatile –NH_2 compounds, organic acids, and carbon dioxide (CO_2), which also contribute to shifts in pH.[34] For instance, fresh poultry generally has a pH range of 5.30–6.50 after being slaughtered. When poultry is stored at room temperature for 24 hours post-slaughter, the pH can reach the spoilage threshold of 8.45, primarily due to microbial breakdown of meat proteins, which releases volatile ammonia (NH_3) and alters the pH of both the external and internal environments of the food.[35] These pH changes can be detected by the halochromic dyes.[29] Fig. 2 depicts the general principle of how a halochromic dye incorporated into a PPM changes color in response to the release of volatile chemicals during food spoilage.

The pH-responsive halochromic PPMs comprise two major components: a supporting matrix and a filler or pH-responsive halochromic dye. Generally, the solid support, or the base material on which the dye is immobilized, is represented by polysaccharide matrices due to their high surface area, uniform dye diffusion, hydrophobicity, and accessible microstructure.[36] The structural integrity of the polysaccharide support directly affects the color stability of the halochromic dye.[37] The pH-sensitive halochromic dye anchored onto the supported matrix should be pure, nontoxic, and stable for long-term storage purposes.[35,37] Moreover, the halochromic dye should strongly interact with food spoilage indicators, such as acids or bases, to produce a clear, distinct color change quickly and easily. The ability of the halochromic dye to respond in the presence of other halochromic dyes under spoilage conditions, with enhanced sensitivity and repeatability across a broad pH range, is also crucial for its real-time response in indicating food freshness.[38]

3 Preparation of PRPP materials

Several techniques are currently employed to prepare PRPP materials. The three most commonly used techniques include casting, tape casting, and thermocompression. In addition, extrusion blow moulding, layer-by-layer assembly, and electrospinning techniques are also used to fabricate PRPPs.[29,40] The following section discusses the aforementioned major PRPP preparation techniques.

3.1 Casting

In the casting method, polysaccharides are dissolved in a suitable solvent, and a pH-responsive dye is added to form a homogeneous film-forming solution. The solution is poured onto a Petri or glass plate, where the solvent is allowed to gradually evaporate at a low temperature (~25–90 °C) in an oven or at room temperature in air, subsequently forming a uniform film.[1,41,42] The casting method is most commonly used in laboratory-scale applications due to its simplicity, limited space requirements, and low cost.[1,43] However, scaling up is one of the major concerns of this technique.[43]

Numerous studies have reported the development of PRPPs using the casting method for food packaging applications. For instance, Sani and coworkers fabricated an edible coating consisting of a chitin/methyl cellulose nanofiber matrix and red barberry anthocyanin as a pH indicator. This edible film was manufactured using casting and developed to monitor the freshness of fish and meat products.[44] The authors also designed a green halochromic active and smart packaging material using the casting method by incorporating saffron and barberry anthocyanin, as well as TiO_2 nanoparticles (NPs) into a gelatin and κ-carrageenan matrix.[45] Thuy-Vi Vo *et al.* fabricated an intelligent film by blending chitosan, polyvinyl alcohol (PVA), and red cabbage anthocyanin through the casting method. They incorporated sodium tripolyphosphate as a crosslinking agent to improve the mechanical strength of the film. The chitosan and PVA hydrogel solutions were mixed in a 3 : 7 (v/v) ratio to fabricate the hydrogel matrix. Anthocyanin extract, accounting for 25% of the total volume, was added to the solution, and the final pH was adjusted to 6.1. The solvent mixture was then cast into the mold and placed in an oven at 35 °C for 48 hours to evaporate the solvent.[46] In another study, a packaging film for salmon was developed using the casting method with a mixture of cowpea starch, sorbitol, and maqui berry extract (MBE). Cowpea starch and sorbitol were mixed in distilled water, stirred for 10 minutes, and then sonicated for 5 minutes. Then MBE was added to the mixture and stirred for an additional 20 minutes at 85 °C. The mixture was then filtered and cast into a mold. The resulting packaging material exhibited the best antioxidant properties for salmon.[47]

3.2 Tape casting

Tape casting is primarily used on an industrial scale, where a solution of a polysaccharide and a natural colorant is passed through a casting machine to form films with uniform thickness.[1] In a general setup, a refrigerated film-forming solution is poured onto acrylic plates using the application tool.[48] Then the tool is activated to move at a set speed, and the solution is poured as the application tool moves, covering the entire film area.[48] The dimensions of the film can be predetermined and set on the application tool.[48] Fig. 3 shows the fabrication of a biodegradable film using Shenyang Kejing tape casting equipment. This tape casting technique can be used to manufacture biodegradable films with dimensions considerably larger than those obtained by conventional casting.[49] The tape casting technique holds several advantages over conventional methods in film-forming because it allows extending the polysaccharide mixture over a large area, making it easy to

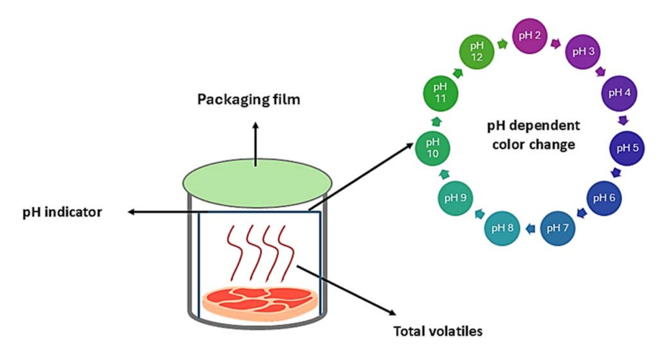

Fig. 2 Schematic representation of how halochromic dye incorporated PRPP changes color upon the release of volatile compounds during food spoilage.[39] Reproduced (adopted) with permission from ref. 39.

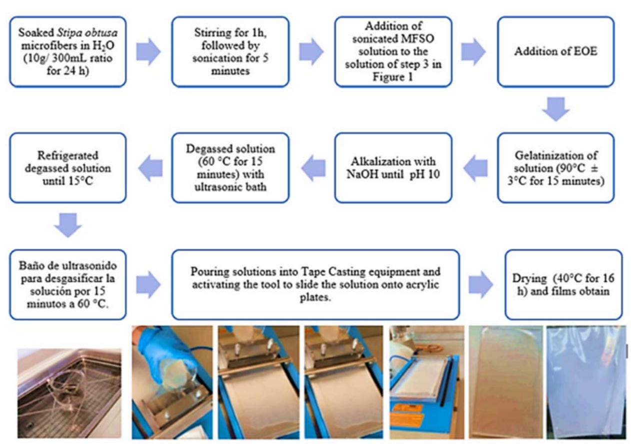

Fig. 3 Large-scale fabrication of starch-based biodegradable films using the tape casting method.[48] Reproduced (adopted) with permission from ref. 48.

manufacture films with controlled thickness, being homogeneous, and also facilitating scaling up the production.[48]

Several studies have reported the development of PRPPs using the tape casting technique. For instance, a starch-based packaging material was fabricated by tape casting to store products with dry surfaces.[49] Paola and coworkers used the tape casting technique to manufacture an edible film incorporating a glycerol/starch suspension.[50] A large-scale biodegradable film derived from potato starch (chuño) and nanocellulose derived from *Stipa obtusa* microfibers (MFSO), incorporated with eucalyptus essential oil (EEO) as the antimicrobial agent, was manufactured using Shenyang Kejing tape casting equipment (Model MSK-AFA-I) by Parada-Quinayá and coworkers.[48] The speed of the application tool was 10 mm s^{-1}. For each batch, 250 mL of gelatinized film-forming solution was prepared and continuously poured as the application tool moved. The film thickness was predetermined and set on the equipment, and the film's total area was 280 mm × 160 mm. The final dimensions of the scaled-up film were 250 mm × 150 mm with a film thickness of around 0.11 mm.[48] The flow chart of the film fabrication methodology is shown in Fig. 3. Moreover, a nanocomposite film was fabricated by corn starch and cassava starch reinforced by cellulose nanofibers (CNFs).[36] Films were prepared by the tape casting technique using a 2.5 mm blade opening. This technique enabled the fabrication of smooth, homogeneous, and uniform starch-CNF films with enhanced tensile strength and surface properties. However, the corn starch nanocomposite films exhibited the best mechanical properties, characterized by low surface roughness and increased tensile strength.[36] In another study, the tape casting technique was utilized to formulate a κ-carrageenan-based, biodegradable, antibacterial film incorporating Wells–Dawson polyoxometalate as an antibacterial agent against food-borne bacterial species.[51]

3.3 Thermocompression

The thermocompression method involves heating the biopolymer to its melting point and then exerting pressure to form a thin solid film. Generally, a polymer or polymer mixture is placed between two Teflon or metal plates and heated to its melting temperature. It is then pressed under a hydraulic press at a relatively high pressure for a few minutes to mould it into a thin film. Then the film is removed and conditioned at room temperature and pressure. This method is fast processing and most suitable for large-scale production.[1] The thermocompression method is a useful technique due to its simplicity, the absence of solvent requirements, and enhanced hydrophobicity and water resistance.[52]

Few studies have reported the production of PRPP using the thermocompression method. For instance, Miaoqi Dai and others utilized thermocompression molding to fabricate a composite film from spine grape pomace and methyl cellulose, owing to its high efficiency, simplicity, eco-friendly nature, and high productivity.[53] The agar-xanthan gum-carboxymethyl cellulose blend thermoplastic was also fabricated using the thermocompression technique.[54] In this study, thermocompression was performed between polytetrafluoroethylene-coated fiberglass with 0.15 mm thickness and a steel mold with dimensions of 130 mm × 130 mm × 4.1266 mm. The thermoplastic films were prepared by pressing 3.5 g of the blend at 140 °C for 3 minutes without pressure, followed by 6 minutes at 140 °C with a constant pressure of 300 kN. Finally, the cooling hydraulic press was operated to cool the material and peel off the polytetrafluoroethylene-coated foil.[54] In another study, the thermocompression method was employed to develop biodegradable corn starch thermoplastic films. Those films were fabricated by incorporating chitin and chitosan. Here, the thermostatized hydraulic press was used to fabricate the films, with processing conditions of 140 °C for 6 minutes, during which the pressure was increased every 2 minutes. A 1 mm-thick aluminum frame was used as the mold with a relation of 1.9 g sample per cm^3. After the thermocompression process, the material was cooled up to 50 °C, the pressure was released, and the films were removed from the frame. The fabricated films exhibited a uniform thickness and good appearance. The absence of unmalting starch granules and visible agglomerates of chitosan and chitin confirmed the effectiveness of the thermocompression technique.[55] Andrea C. Galvis-Sánchez and colleagues prepared a chitosan biofilm using the thermocompression molding technique. The authors utilized a circular mold with a 5 cm diameter and a 2 mm thickness. The film-forming solution containing mold was placed between two stainless steel plates covered with aluminum foil. Before compression, the mold was removed, and a hydraulic press was used for thermocompression at 120 °C.[56]

3.4 Extrusion blow molding

Extrusion is mainly used to produce packaging materials from petroleum-based polymers because high working temperatures, like 180–290 °C, can cause degradation of biopolymers.[29] The extrusion process can also be used to process biopolymers with good thermal stability, high melt strength, and limited swelling, as the parison must support its own weight before the mold can hold it.[57] Usually, there are three main steps in the extrusion process. First, the screw advances the polymer in the feeding zone, where it is combined and uniformly compressed under controlled pressure. Next, the material moves into the kneading zone, where it is thoroughly mixed and homogenized. High temperatures and screw action help release trapped air and ensure uniform melting during this stage. The now fully molten and highly viscous polymer is finally stabilized and metered in the equalization zone, allowing it to be extruded through the machine die in a controlled and consistent manner.[29] Rodríguez-Castellanos and coworkers used the extrusion technique to process the starch–gelatin polymer matrix reinforced with cellulose. Here, the hydrolyzed starch–gelatin combination (with or without 5% cellulose) was fed into an extruder with a 20 : 1 L/D ratio to manufacture the biopolymer blends *via* single-screw extrusion. To create a homogeneous melt, the material was pushed forward at 30 rpm while being heated through a regulated temperature profile of 35–50 °C. After being formed using a 3 mm die, the extrudate was pelletized for further usage. According to this study, adding gelatin to starch during extrusion improves the properties of starch under shear and temperature conditions, including film formation, barrier properties, and tensile and elongational strengths. As a reinforcing additive, cellulose improves mechanical performance and dimensional stability. Cellulose and gelatin work together to fortify starch-based products and improve their suitability for extrusion processing.[57] Jie Zhu and colleagues used extrusion blow molding to fabricate the starch-based films incorporating nano-ZnO and nano-SiO_2. The incorporation of NPs improves the tensile strength, thermal stability, surface hydrophobicity, and barrier properties of the film. The compounding was performed using a twin-screw extruder with a 2.17 mm screw diameter and a 40D length, and the extrusion temperatures from the barrel to the die were 60 °C, 90 °C, 105 °C, 110 °C, 115 °C, and 115 °C, respectively, at a screw speed of 150 rpm. A single-screw extruder (25 mm screw diameter, 30D length; Lianjiang Machinery Co., Ltd, Zhangjiagang, China) has been used for film blowing, with a film-blowing die and six temperature-controlled zones. While the screw ran at a steady speed of 25 rpm, the temperature profile from the feeding zone to the die was progressively increased from 70 °C to 130 °C (70 °C, 100 °C, 115 °C, 120 °C, 125 °C, and 130 °C).[58] Fig. 4 illustrates the extrusion-blow molding technique for preparing PRPPs.

3.5 Electrospinning

Electrospinning is a useful and adaptable technique for producing continuous, nonwoven polymer nanofibers, with additional benefits in terms of orientation, superior porosity, and fiber homogeneity.[29] This technique is also capable of improving the sensitivity and stability of halochromic films.[59] An electrospinning apparatus consists of a capillary tube or syringe, serving as a reservoir for the polymer solution, a high-voltage source, a metallic needle for dispensing the solution, and a collector for collecting the nanofibers (Fig. 5). The term "needle assembly" refers to the combination of the needle and syringe. The process consists of three stages: jet initiation, elongation, and solidification. During the electrospinning process, a high electric field causes the polymer solution or melt at the needle tip to transform from a sphere into a cone (a Taylor cone) and to extend from the tip, creating fiber filaments. The essential elements of the electrostatic spinning process are typically a high-voltage electric field, a nozzle, and a metal collection plate.[29] Duan and coworkers fabricated pullulan/chitin nanofibers incorporating anthocyanin and curcumin *via* electrospinning. Those nanofibers exhibited the best pH responsiveness for the spoilage of *Plectorhynchus cinctus* at room temperature, and antioxidant, and antibacterial properties.[60] Qi *et al.* developed an alginate, polyvinyl alcohol, and Blackwolf berry anthocyanin-incorporating nanofiber film by the electrospinning technique, and the film exhibited high pH responsiveness and rapid color change, even at 1s.[61] Fig. 5 illustrates the schematic representation of the electrospinning technique.

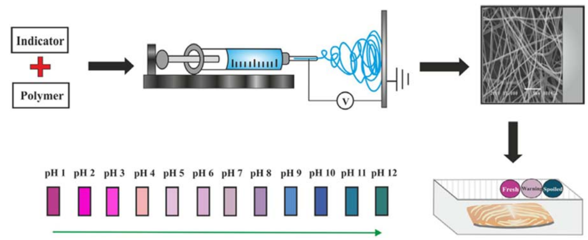

Fig. 5 A schematic representation of the electrospinning technique.[61] Reproduced (adopted) with permission from ref. 61.

3.6 Layer-by-layer assembly

Layer-by-layer (LBL) technology is a flexible method for creating multilayer films by sequentially depositing multivalent molecules and macromolecules *via* electrostatic, hydrogen-bonding, and hydrophobic interactions. Because of their improved functional qualities and adjustable structure, polyelectrolyte multilayer films made using this technique have demonstrated broad utility. Crosslinking further increases mechanical strength and thermal stability and decreases hydrophilicity, thereby improving the performance of LBL films. Even though typical chemical crosslinkers such as formaldehyde, boric acid, and glutaraldehyde work well, their high cost and toxicity prevent their use in food and medicinal applications.[62] Chen *et al.* (2025) used LBL assembly to improve the properties of metal NP-incorporating polysaccharide-based films, which exhibited enhanced surface hydrophobicity with a 98.5° contact angle.[63] He and coworkers fabricated a chitosan/sodium alginate/anthocyanin halochromic film using LBL assembly technology. The film was composed of components with the best mechanical properties and the highest barrier properties against moisture and ultraviolet light.[64] Fig. 6 shows a schematic representation of the major fabrication methods for PRPPs.

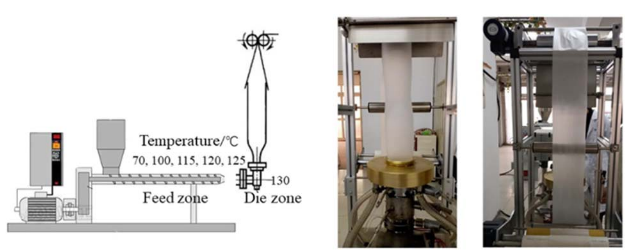

Fig. 4 Extrusion blow molding technique for preparing PRPPs.[58] Reproduced (adopted) with permission from ref. 58.

4 Applications of PRPPs

Polysaccharides have been investigated for food applications, including the fabrication of pH-responsive packaging materials. Natural colorants are widely tested as halochromic agents in PRPPs.[65] The following section discusses the most recent applications of polysaccharides and natural colorants in food packaging.

In a study by C. Wang and coworkers, a PRPP material was fabricated *via* solution casting. The process involved blending carrageenan (CA) and carboxymethyl cellulose (CMC) to prepare

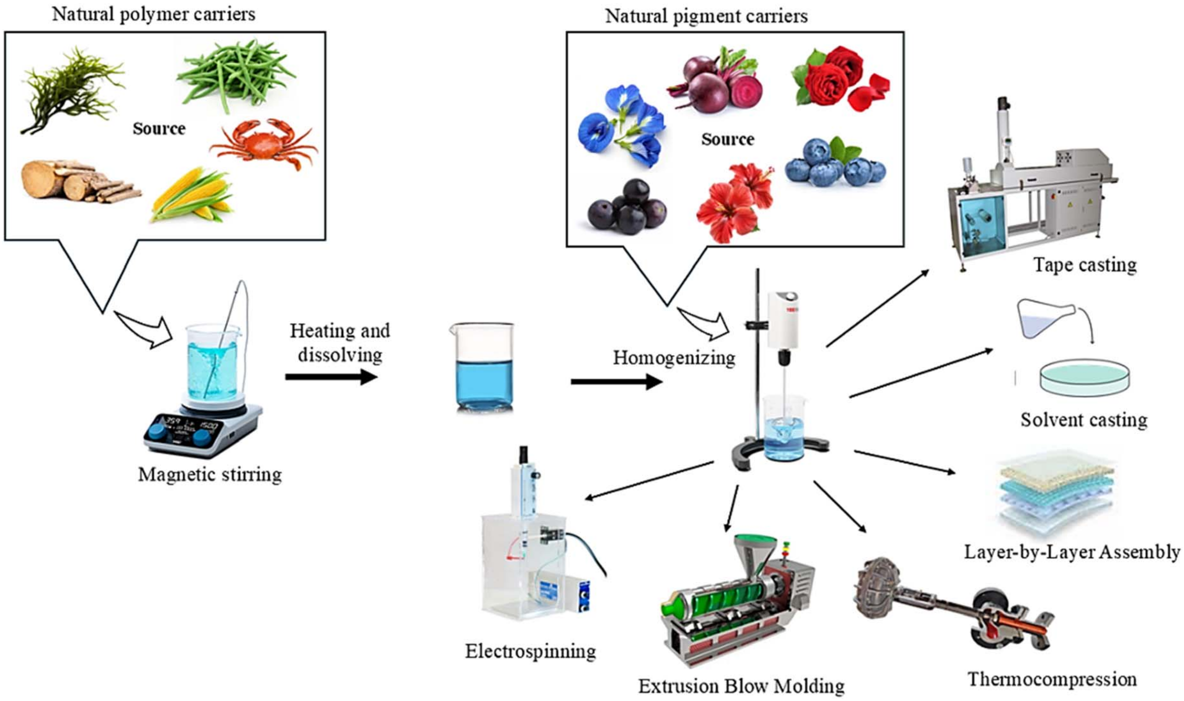

Fig. 6 Schematic representation of the major preparation methods of PRPP materials.

the polysaccharide-based film, followed by the incorporation of rose anthocyanin (RA) as a halochromic dye. Fig. 7 (A) shows the halochromic properties of RA, along with its UV-vis spectra at pH levels ranging from 2 to 11. RA displayed increased antioxidant properties, enhanced crystallinity, and higher surface roughness of the film, in addition to its pH responsiveness. The film was evaluated for its ability to detect freshness in hairtail fish under refrigerated conditions. Fish spoilage released R–NH_2 and NH_3, causing a color change in the dye inside the package. In this study, the authors analyzed the relationship between pH and total volatile basic nitrogen (TVB-N).[65] Fig. 7 (B) exhibits the relationship between pH and TVB-N released during the spoilage of hairtail fish. Initially, the fresh hairtail fish exhibited a TVB-N value of 21.98 mg/100 g with a pH of 6.38. Under storage conditions, the spoilage indicators increased. By the fifth day, the TVB-N content had risen to 27.58 mg/100 g, while the pH also increased to 6.46. On the seventh day, the fish reached spoilage levels, with TVB-N content increasing to 30 mg/100 g and the pH reaching 7. Fig. 7(C) illustrates the color variation of the packaging film in response to pH changes. The film gradually shifted from light pink, representing freshness, to yellow at the sub-fresh stage, and finally to light yellow, indicating fish spoilage.[65]

Adımcılar *et al.* fabricated an intelligent film by incorporating anthocyanin extracted from purple basil to monitor the freshness of chicken breast under refrigerated conditions.[66] The film matrix was developed using pectin and alginate.[66] Fig. 8(A) exhibits the halochromic nature of the purple basil extract across a pH range of 2 to 12, as well as the reaction of the film under different pH conditions. The film was red at pH levels below 6, and at pH 6, the color changed to vibrant violet. Under basic conditions (pH 8), the film turned blue, and it turned green above pH 8. The freshness study conducted on chicken breast showed that after 15 days of refrigerated storage conditions, the color of the chicken breast packed film changed from bluish-violet to green due to the release of volatile alkaline compounds (see Fig. 8(B)).[66]

In another study, Lu Mu and colleagues developed an intelligent (H–K–G–B) film by blending κ-carrageenan, hydroxypropyl methylcellulose, and gelatin with the blueberry anthocyanin FFI (see Fig. 9(A)) using a casting technique. The authors used a smartphone-based visual detection platform for real-time monitoring of food freshness. Fig. 9(B) and (C) demonstrate the halochromic nature of the H–K–G–B intelligent film from pH 2 to pH 12, and a schematic representation of the spoilage detection of salmon meat with increasing pH, TVB-N, and thiobarbituric acid reactive substances (TBARS). Fig. 9(D) shows the system for visual monitoring of salmon meat freshness, integrating the H–K–G–B film with the precise color recognition function of the smartphone.[67]

Si Tan and coworkers prepared an anthocyanin-incorporating intelligent film to detect the freshness of shrimp by blending a polymer matrix comprising gelatin, sodium alginate (SA), and nanoclay, and curcumin as the pH-responsive colorant.[68] The incorporation of nanoclay into the film enhanced its hydrophobicity and barrier properties, controlled the release of curcumin, and confirmed the biodegradability of the film within 30 days. Curcumin acted as both a halochromic dye and an antioxidant, prolonging the oxidative spoilage of shrimps. The film changed its color from yellow to orange to red under acidic, neutral, and alkaline conditions, respectively, during the spoilage process.[68] Ezati and coworkers

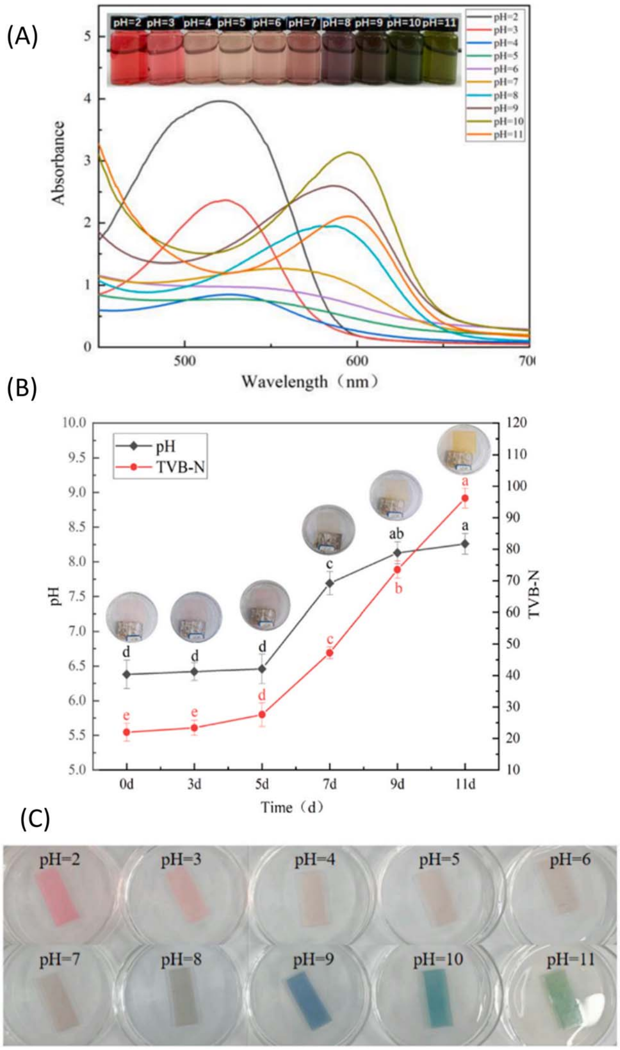

Fig. 7 (A) UV-vis spectra of RA at different pH values (2–11) and the color of the dye at different pH values, (B) relationship between pH and TVB-N released during the fish spoilage, and (C) the pH response of the PRPP film material in the pH range of 2–11.[65] Reproduced (adopted) with permission from ref. 65.

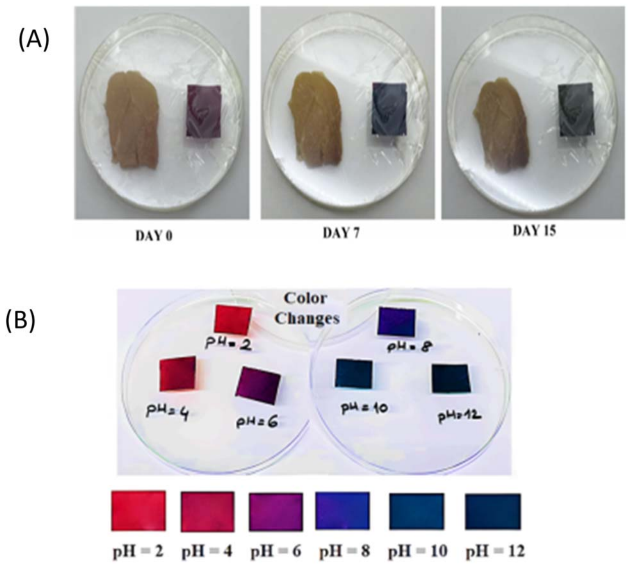

Fig. 8 (A) The halochromic properties of purple basil anthocyanin across the pH range of 2 to 12, and (B) the color variation of the film during the spoilage of chicken breasts under refrigerated conditions.[66] Reproduced (adopted) with permission from ref. 66.

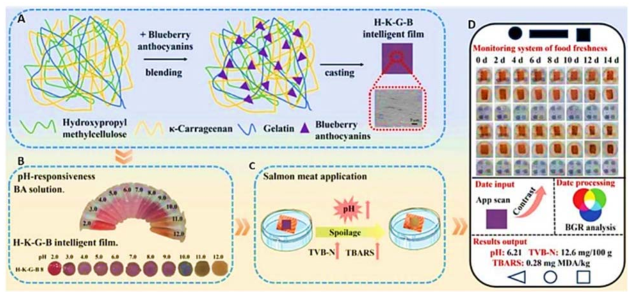

Fig. 9 (A) Schematic diagram of the intelligent film prepared from blueberry anthocyanin (BA) incorporated κ-carrageenan, hydroxypropyl methylcellulose, and gelatin, (B) the color variation of blueberry anthocyanin from pH 2 to pH 12, (C) spoilage detection of salmon meat using intelligent film, and (D) a smartphone-based salmon meat freshness monitoring system.[67] Reproduced (adopted) with permission from ref. 67.

developed a PRPP film by incorporating shikonin as the halochromic dye. The film was formed by incorporating CMC, cellulose nanofibers, and glycerol, which was also added as a plasticizer to detect the real-time freshness of seafood. The film was formed using the casting technique, which involved preparing a film-forming solution through vigorous stirring at 90 °C for 30 minutes. The hydrophobicity of the film improved with the addition of shikonin, without altering the other properties. The initial color of the shikonin-incorporating indicator film was reddish pink, and it turned into bluish purple when exposed to ammonia vapor, and turned into dark reddish pink upon exposure to acetic acid vapor.[69] Recently, Joseph Robert Nastasi and coworkers devised pH-sensitive pectin films by incorporating anthocyanins from two different sources: Mountain Pepper Berry (MPB) and Queen Garnet Plum (QGP). The films were designed to capture the freshness of raw animal products. This study also employed the casting technique for film preparation. Films were fabricated by incorporating anthocyanins from two different sources, and each film matrix consisted of high-methoxy pectin and glycerol. Film-forming solutions were prepared by shaking the solution mixtures at 2000 rpm for 4 hours under dark conditions. This study highlighted that the MPB anthocyanin-incorporating film exhibited the best mechanical properties, while the QGP anthocyanin-incorporating film responded more effectively to the pH changes. The authors reported that the source of anthocyanins could affect the mechanical properties of the film.[70] Fig. 10(A) and (B) show the UV-vis spectra and halochromic properties of MPB and QGP anthocyanin in the pH range of 2 to 11. Fig. 10(C)

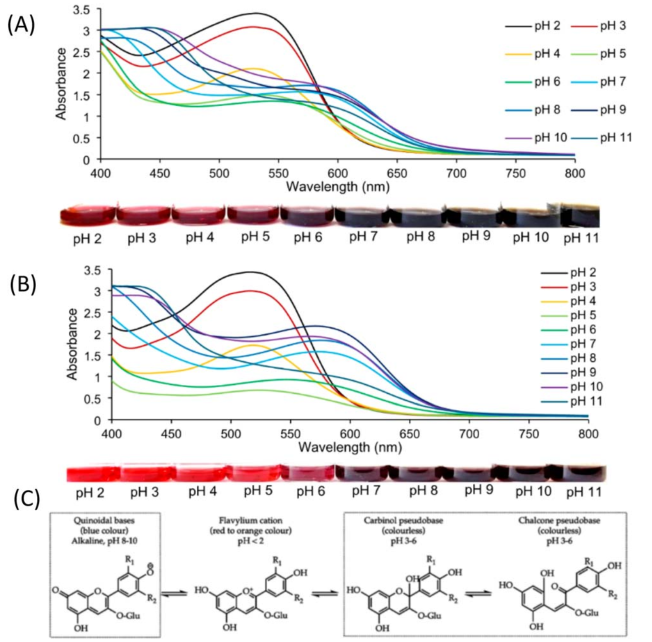

Fig. 10 UV-vis spectra and pH responsiveness of (A) MPB and (B) QGP anthocyanin from pH 2 to pH 11 and (C) the structural variations of anthocyanin at different pH values.[70] Reproduced (adopted) with permission from ref. 70.

shows variations in the molecular structure of anthocyanin at different pH values.

Chen *et al.* fabricated an intelligent film incorporating alizarin as the FFI, using cationic guar gum and κ-carrageenan as a supporting matrix *via* the casting method. The fabricated intelligent film detected the freshness of milk and shrimp. The authors mentioned that the alizarin-incorporating intelligent film showed high sensitivity to volatile NH_3 and trimethylamine ($(CH_3)_3N$). The film monitored freshness at three chromatic stages: fresh, spoiling, and spoiled, with a reusability of up to six months. Fig. 11(A) and (B) illustrate the preparation method of the film-forming solution and the film formulation through hydrogen bonding and electrostatic interactions between the film-formulated materials, including cationic guar gum, κ-carrageenan, and alizarin. Additionally, Fig. 11(C) depicts a schematic representation of how the alizarin-incorporating intelligent film changes its color in response to the spoilage of milk and shrimp.[71]

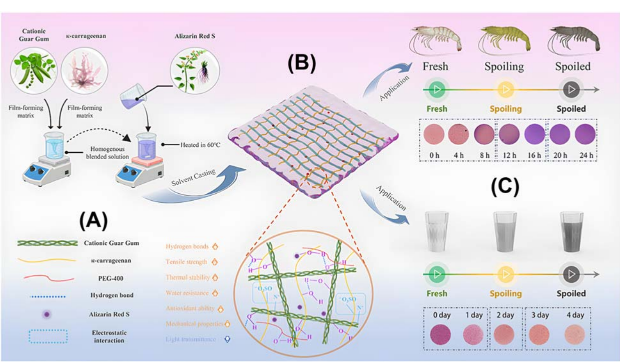

Fig. 11 Schematic representation of an intelligent film incorporating alizarin/cationic guar gum/κ-carrageenan. (A) The preparation method of the film-forming solution, (B) the mechanism of film formulation through hydrogen bonding and electrostatic interactions between film-formulated materials, and (C) the color change of the alizarin-incorporating intelligent film in response to the spoilage of milk and shrimp.[71] Reproduced (adopted) with permission from ref. 71.

Shah *et al.* fabricated an edible film to detect real-time shrimp freshness by incorporating phycocyanin as a colorimetric dye into a polymer matrix prepared using gallic acid, SA, and CMC. The authors observed that the color of the film changed from blue to bluish grey to dark grey, depending on the storage period. Additionally, the incorporation of gallic acid and phycocyanin increased the film thickness and tensile strength while reducing water vapor permeability, oxygen permeability, moisture content, and water solubility. Gallic acid was responsible for the antioxidant and antimicrobial properties of the film, and its incorporation also improved the pH sensitivity of phycocyanin.[72]

Baohua Liu and coworkers developed an intelligent active packaging film by incorporating blueberry anthocyanin into an oxidized sodium alginate–gelatin matrix. The film showed excellent mechanical properties, UV-blocking ability, and antibacterial, and antioxidant activity. The film was used to monitor the freshness of shrimp, and the color of the film changed from orange to green on shrimp spoilage. The shrimp spoilage was also controlled by the packaging films from 2 days to nearly 4 days upon the incorporation of 16% blueberry anthocyanin.[73]

The study conducted by Akbar Mirzaei and coworkers fabricated a pH-responsive film with collagen and CMC to detect the freshness of packaged fish, incorporating quercetin as a halochromic dye. The addition of quercetin enhanced the mechanical, antioxidant, and antimicrobial properties of the PRPP. After 3 days, the film color changed to a bright yellow.[74] Several investigations have reported the use of PRPP materials for food spoilage. Table 1 provides an overview of the most recent applications of PRPP materials in food spoilage monitoring, summarizing the food type, film formulation, pH responsiveness, and film properties.

5 Regulations and safety considerations

Although the PRPPs are considered nontoxic and environmentally friendly, they are still subject to several regulations and safety compliances. As biopolymers and natural colourants are increasingly employed in food packaging, the intersection of food packaging technology, safety regulations, and policy frameworks must be thoroughly addressed to ensure consumer health and address environmental concerns. The following section discusses major regulations and safety considerations for PRPP materials.

5.1 Food contact material (FCM) safety regulations

The Framework Regulation (EC) No 1935/2004 must be fully complied with by all materials and items intended for food contact in the EU.[105] This regulation establishes the overall safety requirement, stating that materials should not transfer components into food in amounts that could harm human health, cause

Table 1 Applications of PRPP intelligent films in food spoilage monitoring

Field of application	PRPP film formulation		Natural dye source(s) and its pH responsiveness		Properties	Application(s)	Ref
	Matrix	Filler(s)	Source	pH responsiveness			
Dairy products	Gelatin/carrageenan polymer mixture	Propolis and shikonin	Shikonin solution from gromwell root extract (*Lithospermum erythrorhizon*)	Color changed across a wide pH range of 2–12, and film color changed from purple, violet to blue color with the pH increase	✓ UV blocking ✓ Mechanical, vapor, and thermal barrier ✓ pH responsive ✓ Good gas sensing ability ✓ Antimicrobial and antioxidant properties	Detection of packed milk freshness	75
	Starch matrix	Black carrot anthocyanin	Black carrot anthocyanin	The film turned pink under acidic pH conditions and then turned into purple and deep blue with the increase in pH	✓ pH responsive	Freshness of milk	76
Meat and fish (seafood)	Konjac glucomannan matrix incorporating gelatin-cellulose nanocrystals as a solid stabilizer	Anthocyanin and curcumin Thyme essential oil as an antibacterial agent	Anthocyanin from purple sweet potato	The film color changed from yellow to yellowish brown to reddish brown with time	✓ pH responsive ✓ Light and water vapor/oxygen barrier properties ✓ Thermal stability ✓ Hydrophobicity ✓ Antibacterial properties	Freshness of shrimp and yogurt	77
	Polyvinyl alcohol, chitosan, and starch blend	Anthocyanin	Ghangaru (*Pyracantha crenulata*)	The film color changed from dark red to brown over 36 hours with meat spoilage	✓ pH responsive ✓ Antioxidant activity ✓ High opacity ✓ High moisture retention	Freshness of meat	78
	Gelatine and quaternary ammonium chitosan	Betalain	Betalain from *Amaranthus tricolor* L.	The film color changed from purple to yellow with the increase in pH	✓ Water vapour barrier ✓ Light barrier ✓ Antioxidant and antimicrobial properties ✓ pH responsive	Monitor the freshness of shrimp	79
	Alginate-cellulose	Betalain	Betalain extracted from red prickly pears	The color of the film appeared purple at pH values below 10 and turned to yellow with pH increase (above 10)	✓ UV stable ✓ pH responsive	Detect salmon spoilage	80
	Cinnamon essential oil incorporating chitosan matrix	Anthocyanin	Anthocyanin (butterfly pudding extract)	The film showed a visible color change from purple, blue, to dark green in the 1–14 pH range	✓ Improved mechanical properties ✓ High pH-sensitive film	Monitor the freshness of fish (tilapia)	81
	Cellulose paper	Shikonin	Shikonin	The film color shifted from red to dark blue in the 2–12 pH range	✓ Water resistant ✓ Thermal stability ✓ Antioxidant activity ✓ pH responsive	Fish and pork freshness	82
	Oxidize chitin nanocrystals and a gelatin nanocomposite film	Anthocyanin	Anthocyanin (black rice bran)	The film shifted from rose–carmine, pink, purple, gray–blue, and yellow green within the pH range from 2–12	✓ Antioxidant activity ✓ pH responsive	Detect the spoilage of shrimp and hairtail	83
	Starch/gelatine film	Anthocyanin	Anthocyanin (red radish extract)		✓ Good mechanical properties ✓ pH responsive	Real-time meat quality	30

Table 1 (*Contd.*)

Field of application	PRPP film formulation		Natural dye source(s) and its pH responsiveness				
	Matrix	Filler(s)	Source	pH responsiveness	Properties	Application(s)	Ref
				The color of the film changes from orange, grey to purple with the pH shift from 2–12			
	Film consists of konjac glucomannan and bacterial cellulose nanofibers	Curcumin	Curcumin	The packaging film gradually turned red with the deterioration of pork	✓ Thermal stability ✓ Antioxidant activity ✓ Anti-bacterial activity ✓ pH responsive	Monitor the freshness of meat and pork	84
	Pullulan/gellan gum film	Anthocyanin	Anthocyanin (leaves and fruit extract of *Broussonetia papyrifera*)	The color of the film gradually turned into red, violet, vat het blue, and finally yellow with the pH change from 2–12	✓ Thermal stability ✓ UV blocking properties ✓ Antioxidant activity ✓ pH, and ammonia-responsive	Monitor fish freshness (*Pelteobagrus fulvidraco*)	85
	Chitosan film	Alizarin	Alizarin	The color of the composite film gradually changed from yellow to purple in response to the pH change from 4–10, and with the fish spoilage, its color turned from khaki to light brown	✓ Thermal stability ✓ Hydrophobicity ✓ UV blocking properties ✓ Antioxidant activity ✓ pH, and ammonia responsiveness	Monitor fish freshness	86
	Konjac glucomannan and carrageenan matrix with diatomite	Anthocyanin	Anthocyanin (extracted from blueberry)	The composite film changed its color from bright pink to bluish violet with the spoilage of shrimp	✓ Detect volatile amines ✓ High tensile strength ✓ Good barrier properties ✓ Hydrophobicity	Monitor the freshness of shrimp	87
	Konjac glucomannan and hydroxypropyl methyl cellulose film	Mulberry extract	Mulberry extract	The film color was changed from purple to grey to yellow due to the spoilage of fish	✓ UV blocking ✓ pH responsive ✓ Antimicrobial and antioxidant properties ✓ Increased mechanical properties	Monitor real-time fish freshness	88
	Konjac glucomannan/pullulan-based film	Anthocyanin	Anthocyanin (Acai berry extract)	The film color was changed from purple to blue to yellow due to the spoilage of fish	✓ UV blocking ✓ Mechanical ✓ Vapor, and thermal barrier ✓ pH responsive ✓ Antimicrobial and antioxidant properties	Real-time monitoring of fish freshness	89
	κ-carrageenan film with rice straw lignin	Anthocyanin	Anthocyanin (*Padus virginiana* peel extract)	The film color was changed from pink to greyish yellow	✓ High sensitivity for ammonia and pH ✓ Thermal stability ✓ Antioxidant properties	Real-time monitoring of the freshness of chicken breast meat	90
	1-Butyl-3-methylimidazolium chloride (BmimCl), cellulose nanocrystal (CNC), and hydroxypropyl	Anthocyanin	Anthocyanin	The film color was changed from light purple to green with the deterioration of seafood	✓ High stability ✓ Excellent sensitivity ✓ Low detection limit ✓ High sensitivity to pH and ammonia	Detect real-time freshness of seafood in cold chains and other fields	91

Table 1 (*Contd.*)

Field of application	PRPP film formulation		Natural dye source(s) and its pH responsiveness				
	Matrix	Filler(s)	Source	pH responsiveness	Properties	Application(s)	Ref
	guar (HPG) incorporating composite film						
	Polyvinyl alcohol/okra mucilage polysaccharide (PVA/OMP) composite film	Anthocyanin	Rose anthocyanin	The color of the film gradually turned into red, pink, blue, green, and finally yellow–green with the pH change from 2–12	✓ Mechanical and barrier properties ✓ High sensitivity for ammonia and pH	Monitor real-time freshness of shrimp, seafood, and meat products	92
	Composite film incorporating glycerol, starch, and polyvinyl alcohol	Anthocyanin or curcumin	Anthocyanin or curcumin		✓ The curcumin incorporating composite films were more stable than the anthocyanin incorporating composite films at room temperature ✓ The 2 : 8(v/v) ratio of anthocyanin: curcumin suited best for monitoring fish freshness	Real-time freshness of fish products	93
	Gellan gum with heat-treated soy protein isolate composite film	Anthocyanin	Anthocyanin (extract of *Clitoria ternatea*)	The film color was changed from red, violet, blue, blue–green, green, and finally brownish yellow in the 1–14 pH range	✓ Modified tensile strength ✓ Swelling capacity ✓ Water vapour permeability	Freshness of shrimp	94
	Pectin composite film with zinc alginate	Anthocyanin	Anthocyanin (peel extract of dragon fruit)	The film's color was changed from pink, blue, and finally yellow in the 2–12 pH range	✓ UV blocking ✓ Antimicrobial activity ✓ Water vapor barrier ✓ Flame retardancy	Freshness of shrimp (room and cold storage conditions)	95
	Methylcellulose and chitin nanofiber	Anthocyanin	Anthocyanin (red barberry)	The film's color was changed from pink to yellow	✓ UV-visible screening ✓ Moisture barrier ✓ pH and ammonia responsive ✓ Improved mechanical properties ✓ Antimicrobial activity ✓ Antioxidant activity	Freshness of fish fillet	44
	Nano-fibrillated cellulose/cellulose nanocrystals	Anthocyanin	Anthocyanin (red cabbage extract)	The film's color was changed from purple to green	✓ Enhanced tensile strength ✓ Flexibility	Seafood packaging	96
	Soluble soybean polysaccharide/SiO_2 nanocomposite film	Curcumin	Curcumin	The film's color was changed from yellow, orange to orange–red	✓ Sense the spoilage ✓ pH changes ✓ Antibacterial activity	Monitor the freshness of shrimp	97
	Methylcellulose film	Anthocyanin	Anthocyanin (extract of Jambolao (*Syzygium cumini*))	The film's color was changed from red, pink, violet to dark purple with pH change from 1–10	✓ Barrier and mechanical properties ✓ Resistant to oxidation and biodegradable	Aquatic products and meat	98
	Film formed by crosslinking κ-carrageenan and gelatin together	TiO_2 NPs and anthocyanin	Anthocyanin (red barberry and saffron)	Red barberry anthocyanin incorporating film- peach pale pink to yellow color, on fish spoilage	✓ Inhibit bacterial growth and oxidation ✓ Resistance to moisture ✓ Biodegradable ✓ High mechanical strength	Track the freshness of fish	45

Table 1 (*Contd.*)

Field of application	PRPP film formulation		Natural dye source(s) and its pH responsiveness		Properties	Application(s)	Ref
	Matrix	Filler(s)	Source	pH responsiveness			
				Saffron anthocyanin incorporating film – violet to green color, on fish spoilage	✓ Barrier properties against UV and visible light		
	Sugarcane wax fixes on agar	Anthocyanin	Anthocyanin (butterfly pea flower extract – pH and ammonia vapor sensitivity)	The film's color was changed from red, blue, and green with pH change from 2–12	✓ UV blocking protects the value of the product from visible light	Track the freshness of shrimp	99
	Dual-modified cassava starch film (oxidized hydroxypropyl starch, acetylated di-starch phosphate (ADSP), and oxidized-acetylated starch)	Anthocyanin	Anthocyanin (red cabbage extract	Changes color from pink, violet, purple, blue, green, and yellow across a wide pH range, 2–12, and responds to volatile ammonia	✓ Barrier for water vapor ✓ Water resistance ✓ Transparency ✓ pH responsive	Intelligent packaging for perishables	100
Fruits and vegetables	Salinized hemicellulose, polyvinyl alcohol, and oxalic acid film	Blueberry anthocyanin	Blueberry anthocyanin	The film's color was changed from pink to blue–green with the pH increase	✓ Water vapor barrier ✓ Antibacterial properties ✓ Antioxidant properties ✓ Enhanced mechanical properties	Monitor the freshness of the strawberry	33
	CMC/chitosan film	Blueberry anthocyanin	Blueberry anthocyanin	The film's color was changed from pink to blue–green with the pH increase	✓ Hydrophobicity ✓ Mechanical strength ✓ pH responsiveness ✓ Barrier properties	Monitor the real-time freshness of fruits and pork also	101
Other	Konjac glucomannan and oxide chitin nanocrystal film	Red cabbage anthocyanin	Red cabbage anthocyanin		✓ Antibacterial properties ✓ Antioxidant properties ✓ Barrier properties against UV light	Active and intelligent packaging for foods	37
	SA/nano ZnO/polyvinyl-alcohol chitosan film	Blueberry anthocyanin	Blueberry anthocyanin	The film's color was changed from pink to blue–green with the pH increase	✓ Antibacterial properties ✓ Moisture barrier ✓ pH sensitivity ✓ Better transmission of light ✓ UV blocking properties	For foods with low moisture content	102
	Chitin nanofibres/bacterial cellulose nanofibre nanocomposite film	Curcumin nano/micro particles	Curcumin nano/micro particles	The film's color was changed from yellow, orange to red color with the pH change from 1–13	✓ Antibacterial activity ✓ Antioxidant activity ✓ pH responsive	Smart and active food packaging	38
	Polyvinyl alcohol/corn starch matrix incorporated with ovalbumin-CMC nanocomplexes	Anthocyanin	Anthocyanin	The color of the film changes from purple to pink under the storage conditions of mushrooms	✓ High thermal stability ✓ Barrier for water ✓ High mechanical strength	Monitor the freshness of mushrooms	103
	Gellan gum film	Anthocyanin (red cabbage extract)	Anthocyanin (red cabbage extract)	The color was changed from red, pink, purple, blue, and finally to green in the 2–12 pH range	✓ Thermal stability ✓ Hydrophobicity ✓ Detects ammonia, and pH changes	Monitor the real-time freshness of mushrooms (also able to detect the freshness of fruits and vegetables)	104

an unacceptable change in the food's composition, or negatively affect its sensory qualities under normal or foreseeable conditions of use. Additionally, as specified in Regulation (EC) No 2023/2006, the production of these materials must adhere to the guidelines of good manufacturing practice (GMP). Polysaccharide-based materials fall within this general framework because they are typically classified as bio-based food contact materials (BBFCMs) made from natural polymers.[105]

Based on their intended function, pH-responsive materials are specifically categorized as active and intelligent materials under Article 3 of Regulation (EC) No 1935/2004.[105] These are subject to their own specific measure, Regulation (EC) No 450/2009, which sets out rules for the safety assessment and authorization of their components. If the polysaccharide material functions as a main structural component, it may also be required to comply with the specific rules for plastic materials set out in Regulation (EU) No 10/2011 (as amended, including Regulation (EC) No 2019/1338). This plastic regulation establishes an authorized list (positive list) of starting substances in Annex I, accompanied by defined Specific Migration Limits (SMLs) that must be met.[105] Current analytical methods and risk assessment processes for traditional plastics are generally considered appropriate or adaptable for these bio-based alternatives.

5.2 Biodegradability regulations

The EU waste laws, such as the Packaging and Packaging Waste Directive 94/62/EC and the Waste Framework Directive, are particularly important for the downstream treatment of packaging materials. The main objective of these laws is to prevent the creation of packaging waste, including guidelines for recycling, reusing, and recovering packaging waste to reduce ultimate disposal. The explicit integration of biodegradation into the circular economy strategy is one of the ambitious new goals proposed by the recent modification of the EU waste regulation.

Packaging must be able to break down chemically, physically, thermally, or biologically into CO_2, biomass, and water to be considered biodegradable. Materials must fulfil the requirements specified in international standards such as EN 13432 (European standard titled: "Packaging – Requirements for packaging recoverable through composting and biodegradation") and/or EN 14995 (European standard titled: "Plastics – Evaluation of compostability. Test scheme and specifications") in order to be certified as compostable. For instance, no more than 10% of the material pieces may be smaller than 2 mm after 12 weeks to comply with the EN 13432 standard for the degree of disintegration.[105] This offers a precise, quantitative way to assess the actual environmental benefit of packaging made from biodegradable polysaccharides.

6 Challenges and future directions

The PRPP materials enhance sustainability, reduce carbon footprints, and improve food security and safety, while also indicating food freshness promptly. In addition, the multifunctional nature of natural colorants also offers improved mechanical properties and antibacterial, and antioxidant benefits to the intelligent films. Amid those advantages, they also present several challenges. When considering intelligent films fabricated using polysaccharides, they can degrade over time upon exposure to illumination and heat. Due to their compact internal structure, most films exhibit high water vapor and oxygen permeability, which can lead to deterioration of the product inside.[106,107] Despite the relative abundance and other features of many biopolymers, there are still constraints on cost-effectiveness and scalability for industry-scale production. In the case of pigments/natural colorants used as FFIs, they become inactive over time due to their instability compared to synthetic dyes, leading to changes in their original characteristics in response to changes in both external and internal environments.[1,106] When the FFI films come into contact with food, they can interact with the food surface, leading to loss the sensory properties of the food.[1] The PRPPs are more hygroscopic, so they cannot be used to pack meat products. Lipids, such as essential oils, waxes, and other lipids, can be used to increase the hydrophobicity of the films.[105] Additionally, the nanocomposites and NPs are incorporated with the film-forming solutions to improve the water vapor barrier properties. Saurabh Bhatia and coworkers fabricated an alginate/acacia gum hydrogel-based film loaded with cinnamon essential oil and found that its incorporation increased thickness, elongation at break, and antioxidant properties while decreasing water vapor permeability, tensile strength, and moisture content.[108]

Several solutions have been proposed to overcome the current issues associated with PRPPs. For instance, nanotechnology-based approaches have been employed to mitigate many drawbacks and enhance the performance of PRPPs.[107,109] The incorporation of nanomaterials into PRPPs has been widely investigated as they can improve the mechanical and physicochemical properties, as well as their ability to function as oxygen scavengers, antimicrobial agents, and gas barriers.[110] The PRPPs have poor water vapor barrier properties and limited heat stability, and are more brittle than fossil-based plastic materials. These limitations can be addressed by incorporating nanocomposites.[105] Cellulose nanofibers, nanoclays, or metal-based NPs are used to improve water vapor barrier properties.[3] Additionally, nano-SiO_2 and nano-ZnO are stable, nontoxic, and durable inorganic compounds mainly used in the packaging industry to improve the barrier and mechanical properties of PRPPs.[58] Salarbashi and colleagues fabricated a PRPP material incorporating SiO_2 NPs. They reported that increasing the NP concentration decreased film thickness, water vapor permeability, and water solubility, while improving mechanical performance and antibacterial properties.[97] Fan Wang *et al.* (2021) developed a superhydrophobic pH-responsive coating by combining starch NPs with poly(dimethylsiloxane). The micro- and nanostructures formed between starch NPs and poly(dimethylsiloxane) confer superhydrophobic properties to the coating (water contact angle > 152.0°).[111] Nanosensors have recently been incorporated into polysaccharide-based intelligent food packaging and tested as pH change indicators for freshness monitoring due to their high sensitivity, high surface area-to-volume ratio, and optical properties.[112] Zhai and others developed a nanocomposite film by incorporating silver NPs (AgNPs) as a pH sensor, capped and synthesized using guar gum (GG). GG-stabilized AgNPs changed their color from yellow to colorless, responding to the volatile H_2S with a limit of detection of 0.81 μM under optimum conditions at pH 7. This nanocomposite film was also tested over chicken breast and silver carp during spoilage.[113]

Crosslinking is another way to improve the mechanical and barrier properties of PRPPs. Creating stronger, more closely connected three-dimensional networks entails the formation of chemical linkages between multiple polymer chains, either intra- or intermolecularly. Crosslinking is frequently categorized by the type of bond or interaction, including covalent, ionic, van der Waals, or H-bonds or by the mechanism of action, such as chemical, physical, or enzymatic.[105]

Multilayer packaging with two or more distinct layers has also been introduced to improve the performance of films, including their protective, mechanical, and barrier features.[103] Specifically, active ingredients with antimicrobial and antioxidant properties can be added to one or more layers of packaging materials to improve their ability to protect food. For instance, Z. Yang *et al.* developed a bilayer film by adding gelatin and zinc oxide NPs (ZnONPs) to the upper layer and mulberry anthocyanin-incorporating gellan gum to the lower layer; the film exhibited high NH_3 sensitivity and good mechanical properties.[103] Recently, Li and coworkers developed a multifunctional double-layer intelligent packaging film by a layer-by-layer casting strategy to monitor food freshness and preservation. The outer layer of the film is made from konjac glucomannan with a Pickering emulsion containing cinnamon essential oil, which provides strong antioxidant and antibacterial properties. The inner alginate–alizarin layer functioned as a pH/NH_3-responsive indicator. The overall double-layer film exhibits high mechanical strength, hydrophobicity, and light-blocking ability. The film has been tested on shrimp packaging and showed small TVB-V after 72 hours and a rapid response to the shrimp's freshness.[114]

Plasticizers can improve the elasticity, flexibility, mechanical properties, and water vapor permeability of the packaging films. There are two types of plasticizers: water-soluble and water-insoluble plasticizers. Esters are the main water-insoluble plasticizer, and fatty acids and polyhydric alcohols are mainly included in the group of water-soluble plasticizers.[115] Water-soluble plasticizers retain moisture in the film and increase its water vapor permeability. However, water-insoluble plasticizers can seal the micropores in the films, thereby improving their barrier properties.[115] However, it is important to consider the quantity and type of additives when adding them to packaging materials to ensure safety, as these additives can migrate from the PRPPs and pose a risk of food contamination.[59]

Composite films prepared from two or more polymer carriers can enhance mechanical properties and balance the performance of films. For example, Wu and others manufactured a film incorporating gellan gum with heat-treated soy protein isolates to reduce the swelling capacity. The addition of soy protein isolates improved the physical and mechanical properties of the film.[94] Yun Wang and colleagues also investigated the effect of crosslinking on the hydrophobicity and mechanical strength of the starch-based polysaccharide labels. The labels incorporating soybean proteins exhibited high pH sensitivity, structural stability, and low pigment leakage.[116]

Artificial intelligence (AI) can also be integrated with the PRPP industry to optimize the process conditions for effective, efficient, and high-quality production. For example, Gao and colleagues recently designed an AI-driven framework to optimize the performance of polysaccharide-based packaging film manufacturing processes. Active learning technologies have been utilized to reduce design time, material waste, and costs, while enabling the customization of film properties.[117] In another study, a machine learning (ML) model has been developed to predict the properties of raw materials, optimize formulations, and reduce experimental effort in the manufacturing of xanthan gum-based foam materials.[118]

7 Conclusion

The pH-responsive polysaccharide-based intelligent packaging films have recently emerged as eco-friendly smart alternatives to petroleum-based plastic packaging materials. The current review discussed the use of polysaccharide-based films and natural halochromic dyes in food packaging. The film fabrication techniques, mechanisms of food spoilage detection, and recent intelligent food packaging applications of PRPP materials have also been discussed. By incorporating natural halochromic dyes into renewable polysaccharide matrices, they represent a step toward a sustainable, circular bioeconomy in the food industry. Although current applications of PRPP materials are limited to laboratory-scale research, further efforts can be made to scale up production to an industrial level by improving stability, enhancing material performance, and standardizing methods. Future research must focus on cost-effective, multifunctional, and environmentally adaptable PRPPs to facilitate their broader utilization, for instance, developing integrated intelligent food packaging systems incorporating nanocomposites, time–temperature indicators, and gas sensors to improve their multifunctionality, applying direct polysaccharide modifications, such as carboxymethylation and graft copolymerization, to enhance antimicrobial, antioxidant, and pH-responsive properties of packaging materials without adding additives, and furthermore, the use of agricultural wastes to extract polysaccharides for the manufacture of low-cost packaging materials and to promote the circular bioeconomy.

Author contributions

R. S. Dassanayake: conceptualization, supervision, reviewing, writing, editing and proofreading. G. H. P. Ganegoda: conceptualization, writing the manuscript, collecting and analyzing the data. Danushika C. Manatunga: supervision. K. K. A. Sanjeewa: supervision. G. L. R. Jayathunge: supervision. Renuka N. Liyanage: supervision. Yang Zhou: reviewing and editing. Yuanyuan Liu: reviewing and editing.

Conflicts of interest

There are no conflicts to declare.

Data availability

This manuscript is a systematic review of previously published literature. No new primary data were generated in the course of this research. All data supporting the conclusions and analyses presented herein are available within the cited articles and their associated supplementary information (SI) files. Supplementary information: Tables S1 and S2, structures, sources, properties, and food applications of major polysaccharides and the structure, pH responsiveness and properties of major natural halochromic dyes. See DOI: https://doi.org/10.1039/d5fb00953g.

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