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Biopolymer-based intelligent packaging integrated with natural colourimetric sensors for food safety and sustainability

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Abstract

Increasing concerns about global food safety and security demands innovative solutions, particularly in food packaging technologies. This review paper investigates the advanced integration of natural colourimetric sensors with biopolymer-based packaging materials, with a focus on developments over the past 5 years. These sensors change colour in response to environmental stimuli such as oxygen, temperature, pH and relative humidity, intuitively indicating food freshness and safety. The paper emphasizes the recent advancements in using natural colourants, such as alizarin, anthocyanins, betacyanins, chlorophyll, curcumin and shikonin. When combined with either natural or synthetic biopolymers, these colourants contribute to a sustainable and eco-friendly approach to food packaging. Such technological advances could notably decrease the incidence of foodborne illnesses by signaling potential spoilage or contamination, while also addressing food wastage by providing clear indications of edibility. Although challenges remain in sensor longevity and widespread adoption, the prospects for biopolymer-based food packaging with embedded natural colourimetric sensors are promising.

1 | INTRODUCTION

In today's rapidly expanding global community, the critical convergence of food safety and food security commands urgent attention. Food, as a fundamental human need, is pivotal in sustaining public health and promoting sustainable development. Yet, the significance of food extends beyond mere availability; its safety is paramount for consumption, as food security is incomplete without the assurance of food safety.[1] It is widely acknowledged that for food to fulfil its nutritional promises and for adults to lead an active and healthy life, the consumed food must be safe. Similarly, for children to grow and develop optimally, the nutrition they receive must be uncontaminated and wholesome.[1] Nevertheless, the pressing challenge of ensuring this safety is intensified by escalating global population pressures, raising alarms over potential food shortages and the sustainability of food systems.[2] Compounding this issue, unexpected events such as the economic impacts of the coronavirus disease 2019 pandemic have further strained global efforts to address hunger, food insecurity and poverty.[3]

Food contamination further underscores the links between food security and safety. Fresh produce, due to its nature, is highly susceptible to contamination at multiple points in the supply chain.[4] Such contamination not only diminishes the nutritional value of the food but can also lead to the production of harmful by-products, affecting taste, texture and quality. More concerning is the potential for contaminated food to cause over 200 diseases, stemming from pathogens like bacteria, viruses and parasites, as well as chemical toxins.[1] In light of these interconnected challenges, ranging from the basic need for adequate food to ensuring its safety and combating contamination, it is clear that bridging the gap between food security and food safety is not just necessary, but critical. Therefore, exploring innovative solutions for

Abbreviations: FTIR, Fourier-transform infrared spectroscopy; PLA, Polylactic acid; PVA, Polyvinyl alcohol; SEM, Scanning electron microscope; Ag, Silver; TiO_2, Titanium dioxide; TVC, Total viable count; TVB-N, Total volatile basic nitrogen; ZnO, Zinc oxide.

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food preservation and packaging and understanding the mechanisms that lead to spoilage becomes imperative. By adopting this approach, the interconnected challenges of food safety, contamination and security can be addressed more effectively, ensuring that food not only reaches those in need but also retains its safety and nutritional value for consumption.

While traditional packaging methods, primarily sourced from petroleum, act as barriers to contamination, they fall short in terms of environmental sustainability and fail to provide dynamic information about the food's quality.[5] Consequently, innovative intelligent packaging has been developed, surpassing the conventional role of traditional packaging. This modern packaging, embedded with sensors, not only interacts with the food product but also assesses its surrounding conditions. It provides stakeholders with real-time information and insights, detecting freshness, pathogens, pH levels and other environmental changes, offering more insight than traditional measures such as weight or appearance.[6–10] Such advancements in packaging, especially when combined with responsive colourants, promise a holistic approach to reducing food waste, preventing diseases and ensuring food quality.[5,11]

One emerging innovation that has gained prominence is the use of colour indicators derived from natural sources for real-time monitoring of food quality within intelligent packaging.[12] As the global community becomes more environmentally and health conscious, there is a noticeable shift towards biopolymer-based intelligent packaging, enhanced with natural colourimetric sensors. These not only extend the shelf life of food products but also enable real-time quality checks through visual cues, such as changes in colour due to pH fluctuations or microbial growth.[5] Moreover, they make changes in food quality easily observable and understandable, while also providing protection against environmental hazards, potentially benefiting individuals' health and well-being. Given the critical role of such packaging for perishable foods, including fruits, vegetables, fish, meat and dairy products, delving deeper into this subject is of paramount importance.

This review aims to provide a comprehensive overview of the factors leading to food spoilage the responses of colourimetric sensors and the latest applications of biopolymers in conjunction with colourimetric sensors for intelligent packaging. Additionally, it covers trending research on diverse natural colourimetric sensors over the past 5 years, helping readers gain a nuanced understanding of the current innovations in intelligent packaging and their significance within the broader context of food safety. Notably, biopolymer-based intelligent packaging systems with natural colourimetric sensors have been developed primarily for meat, seafood and dairy products rather than being applied prevalently to a broader spectrum of perishable foods, including vegetables and fruits. Therefore, the discussed application will mainly focus on the utilization of intelligent packaging for these groups.

2 | FACTORS ATTRIBUTING TO FOOD SPOILAGE AND COLOURIMETRIC SENSOR RESPONSES

Food spoilage, referring to the deterioration of food's quality and safety, can pose health risks to consumers. It results from microbial growth, changes in chemical and enzymatic reactions, or physical damage. The main factors contributing to food spoilage, which are crucial in the design and functionality of intelligent packaging systems, predominantly include oxygen, temperature, pH and relative humidity. While colourimetric sensors in the intelligent packaging system are expected to monitor the quality of the food itself, recent studies have focused more on environmental changes caused by food deterioration or alterations in storage conditions rather than directly measuring the food. These intelligent packaging systems utilize indicators for oxygen exposure, temperature fluctuations, pH change and moisture levels to actively monitor and report the state of the packaged food, thereby contributing to enhanced management and prolongation of its shelf life.

2.1 | Oxygen

Oxygen, crucial in oxidative reactions, has a significant impact on food quality. Not only is it essential for the survival and proliferation of aerobic microorganisms, but it also plays a crucial role in various biological processes such as respiration and energy production.[13] The residual oxygen that often remains in food packaging after sealing can provide enough support for aerobic metabolism, fostering an ideal environment for bacterial growth.[14] In the case of packaged meats, oxidation can consequently lead to colour changes and the development of off-flavours. Additionally, in fruits like apples and bananas, the presence of oxygen can activate enzymatic reactions that lead to browning. These reactions can accelerate food deterioration, resulting in spoilage and a decrease in nutritional value. To effectively detect exposure to oxygen in food, which can induce oxidative stress and pose a potential risk of spoilage, intelligent packaging systems employ specific natural colourimetric sensors chosen for their sensitivity to oxidation, thus aiding in the preservation of food quality by indicating the need for corrective actions.

2.2 | Temperature

Temperature stands as a crucial factor in the context of food spoilage. It directly influences the rate of enzymatic reactions within the food and affects the viability of microorganisms present. Sudden shifts in temperature can set off irreversible reactions that might alter or harm the physical characteristics of food and influence its ripening process. Notably, microbial growth is minimal at temperatures below freezing, but it rises exponentially in the range of 0–40°C (32–104°F).[15] As an illustration, dairy products like milk and cheese are particularly susceptible to spoilage at elevated temperatures due to increased bacterial activity. The frequent temperature variations that occur during food transit, storage and preparation can hasten spoilage.[13] Consequently, vigilant monitoring of temperature fluctuations is critical to prevent deterioration and ensure the safety of food, especially for perishable items that cannot be stored at room temperature. Intelligent packaging incorporates temperature-sensitive indicators that are designed to change colour based on the thermal history of the product, enabling

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more informed decisions regarding its safety and usability based on past storage information.

2.3 | pH

The alteration in pH during food deterioration is a result of microbial metabolism and protein degradation. Microorganisms that ferment glucose produce organic acids, such as lactic and acetic acid, contributing to a decrease in pH. Their activity also leads to the formation of carbon dioxide, which, in the presence of water, creates carbonic acid, further lowering the pH by increasing hydronium ions.[13] Conversely, the degradation of proteins by certain types of microorganisms that produce proteases results in the accumulation of total volatile basic nitrogen (TVB-N). The breakdown of proteins into ammonia and volatile nitrogen compounds, triggered by oxidation and bacterial activities, increases TVB-N, potentially raising the food's pH.[16] Therefore, the pH shift resulting from microbial activities and degradation processes plays a pivotal role in determining the freshness and eventual spoilage of food products. To track these pH changes, intelligent packaging systems integrate pH-sensitive colourimetric sensors that visually indicate the freshness and potential spoilage of food products, thus enhancing consumer caution on microorganism proliferation and reducing food waste.

2.4 | Relative humidity

High humidity increases water activity, creating a favourable environment for microbial growth, which in turn accelerates food spoilage and poses safety risks. It also causes food products to absorb moisture, leading to softening and a consequent reduction in shelf life.[13] For instance, bread tends to mould faster in humid conditions due to increased moisture absorption. Additionally, humid conditions facilitate certain chemical reactions. Ammonia in the atmosphere can react with water to form ammonium and hydroxide ions, creating an alkaline environment. This reaction can lead to pH changes in foods, triggering further responses from compounds that chemically interact with hydroxide ions.[17,18] Consequently, elevated humidity promotes physical, chemical and biological changes that deteriorate both the quality and safety of food. By incorporating humidity indicators within intelligent packaging, the system can alert to adverse storage conditions, thereby empowering proactive measures to maintain food quality and extend its shelf life.

3 | BIOPOLYMERS IN RECENT DEVELOPMENTS OF INTELLIGENT PACKAGING

Biopolymers serve as versatile carriers for colourimetric sensors, taking diverse forms such as gels, solutions, films, coatings and solids.[19] Their adaptability enhances their use in intelligent packaging, demonstrating innovative ways to monitor and display changes in packaged products. Derived from living organisms, biodegradable polymers such as proteins, lipids and polysaccharides offer eco-friendly solutions due to their compostability.[19–21] Additionally, biodegradable polymers can be chemically synthesized from either biomass or petrochemicals.[22] All these polymers naturally degrade under conditions like moisture and temperature, leaving no harmful residues behind.[22] The shift toward biopolymer-based food packaging is driven by environmental concerns over non-biodegradable plastics and a growing consumer demand for sustainable, high-quality products.[20,21,23] Biopolymer-based films possess mechanical and barrier properties stemming from intermolecular interactions, each exhibiting unique characteristics. This diversity allows for the development of tailored packaging solutions, optimizing the preservation conditions for specific food types and addressing their unique shelf-life challenges. However, no single biopolymer encompasses all desired attributes, such as barrier effects, solubility, mechanical strength and antimicrobial activity.[19–20] The biopolymer-based packaging often lacks strength, water or thermal resistance.[23,24] Being nearly colourless, biopolymer-based films are ideal for incorporating colourimetric sensors without obscuring sensor responses to environmental changes. Their integration can also improve the films' solubility, strength and other properties.[21] Depending on the desired attributes of the food packaging, various materials will be applied, designed for specific food types and may utilize nanomaterials to optimize preservation and maintain freshness. While these biopolymers have the potential to extend food shelf life, it should be noted that the change is slight rather than dramatic. The nuanced approach to intelligent packaging design, emphasizing the selective use of materials and sensors, caters to the specific needs of different food categories, thereby optimizing shelf-life extension in a tailored manner. This customization underscores the shift towards more sophisticated packaging strategies that not only consider the environmental impact but also the specific preservation needs of various food products. In this review, only certain natural and synthetic biopolymers are discussed, in alignment with the focus of recent studies. Figure 1 provides a summary of the classification of commonly used biopolymers in intelligent packaging over the past 5 years, outlining their types and sources.

3.1 | Natural biopolymers

3.1.1 | Polysaccharides

Polysaccharides, as carbohydrate polymers, exhibit excellent gas barrier properties, effectively protecting against the permeation of gases such as carbon dioxide and oxygen.[19,25] This is largely due to their tightly packed network structure, which contributes to their gas impermeability. The gas barrier property is a critical mechanism by which polysaccharides extend food shelf-life, as they minimize the exchange of gases that accelerate spoilage and degradation. However, their inherent hydrophilic nature leads to the ready absorption of moisture, rendering them less effective as barriers against water vapour.[20,23] Polysaccharide biopolymers are increasingly popular in food packaging because of their ability to minimize and block oxygen penetration. This

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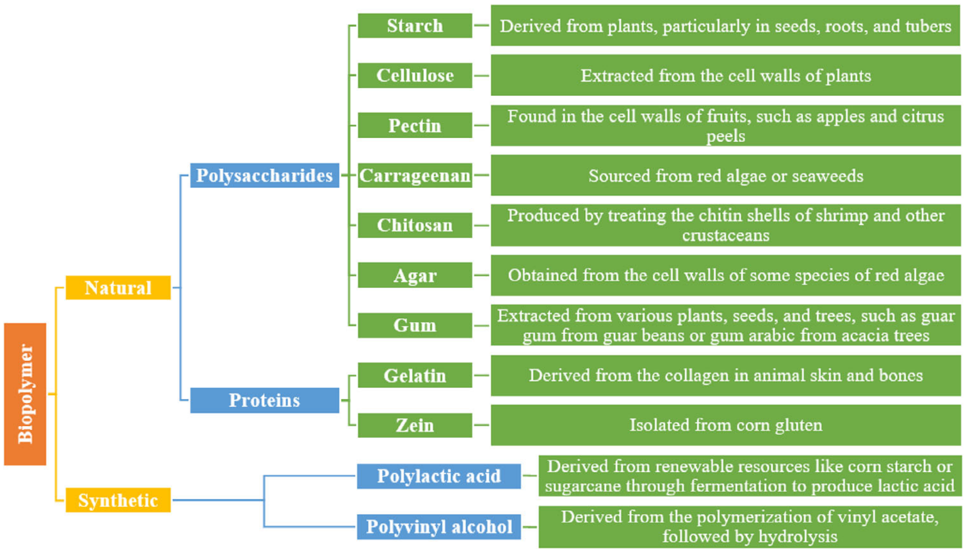

FIGURE 1 Classification of biopolymers with corresponding sources.

characteristic makes them ideal for use in food packaging materials, where preventing oxidation is critical. Their mechanical strength also makes them a reliable choice for packaging, as they effectively create a barrier that minimizes the risk of breakage and exposure to spoilage factors, crucial for protecting food inside the packaging.[25] Moreover, the growing interest in polysaccharides is driven by their abundant availability and cost-effectiveness.[23] Notably, agricultural wastes provide a sustainable source of cellulose and other polysaccharides, further promoting their use in eco-friendly packaging solutions.[21] The formation mechanism of polysaccharide biopolymers involves disintegrating polymer segments and reconstituting them into a film matrix or gel, primarily through solvent evaporation. This process enhances hydrophilic properties and facilitates hydrogen bonding.[23] There is a wide variety of polysaccharide biopolymers used, including starch, cellulose, pectin, carrageenan, chitosan, agar and various gums.[20,23] Among these, chitosan is the most extensively studied and utilized in creating coatings and films with colourimetric sensors for intelligent packaging.

Chitosan, derived from chitin, is a cationic polysaccharide known for its abundance, biocompatibility and biodegradability. It stands out for its antimicrobial and antioxidant properties, making it an exceptional choice for food packaging. These properties of chitosan contribute to shelf-life extension by inhibiting microbial growth and preventing oxidation, which are major factors in food spoilage. Chitosan excels in film production, showcasing impressive mechanical strength and barrier properties.[20,23,25] When used as food packaging, it effectively lowers oxygen levels in packaging, regulates moisture transfer, delays enzymatic browning in fruits, controls respiration and reduces dehydration.[20] Consequently, chitosan coatings can significantly extend the shelf-life of perishable fruits like apples or pears by reducing oxidation, and moisture loss and suppressing mould growth. A study by Mannozzi et al.[26] assessed the impact of a chitosan coating derived from mushrooms on the quality and microbial growth of blueberries over 14 days at 4°C. The chitosan-based coatings significantly inhibited yeast and mould growth compared to uncoated samples, underscoring its potential as an excellent food packaging material. Further, the mechanical strength and water-resistant properties of chitosan can be enhanced by incorporating colourimetric sensors for intelligent packaging. The study by Li et al.[27] demonstrated that integrating purple potato extracts, rich in anthocyanin, into the chitosan-based film enhanced its tensile strength, water resistance and roughness, while also displaying significant antioxidant activities, making it suitable for packaging oxidizable substances.

3.1.2 | Proteins

Protein-based films are emerging as a sustainable option in food packaging, offering superior mechanical properties compared to polysaccharide films. However, they possess lower strength and higher water vapour permeability compared to other synthetic polymers.[20] Their capacity for network formation, coupled with inherent plasticity and elasticity, provides significant packaging benefits. This physical resilience not only enhances food preservation but also serves as a protective barrier, mitigating damage during handling and transportation, thereby further extending food shelf life. Additionally, these biopolymers are favoured for creating biodegradable intelligent films due to their excellent film-forming abilities and effective gas barrier characteristics, which are similar to polysaccharides. The film formation process involves partial denaturation of polypeptide chains through solvent addition, pH alteration or heat application, resulting in a robust protein matrix. Moreover, these proteins blend efficiently with various natural additives, enhancing the functionality of biodegradable films. This makes them a promising eco-friendly alternative for food-intelligent packaging.[23,25] However, it is noteworthy that protein biopolymers are currently less common than polysaccharide polymers in the latest research on intelligent packaging featuring colourimetric

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sensors. Among the protein biopolymers, gelatin and zein are the most frequently used.

Gelatin, a popular protein biopolymer, is a choice in intelligent packaging films because of its abundance, biodegradability, renewability and biocompatibility. It is also prized in food packaging for its excellent film-forming capability, transparency and UV-barrier properties.[25] Gelatin-based films can be specifically tailored for UV-sensitive products, providing a protective shield that extends shelf-life by minimizing photodegradation. Combining gelatin-based films with colourimetric sensors can sense environmental changes and enhance protection for food products. In an experiment by Musso et al.,[28] they found that gelatin films with added curcumin, particularly those prepared from hydroalcoholic dispersions, exhibited high antioxidant properties. Additionally, the study by Zhao et al.[29] reported that curcumin-infused chitosan/gelatin-based films changed colours at different pH levels, responded to alkaline gases and extended the shelf life of fresh meat by 3 days at 4°C by inhibiting bacterial growth and reducing volatile basic nitrogen production.

Zein, a polyamine extracted from corn, is distinguished by its hydrophobic nature and thermoplastic properties, manifesting as a water-insoluble, bacterially resistant, antioxidative material capable of forming adhesive films.[20] This makes zein particularly effective in packaging applications requiring moisture and oxygen barriers, such as for nuts and snacks, where its properties help maintain crispness and prevent rancidity. In a notable development, Aghaei et al.[30] engineered a zein-based halochromic sensor by innovatively embedding alizarin into zein electrospun nanofibers. This process effectively trapped the alizarin within the zein matrix through hydrogen bonding. The resulting packaging system demonstrated rapid and noticeable colour changes in response to alterations in pH and TVB-N. These dynamic characteristics underscore Zein's potential for real-time monitoring applications with colourimetric sensors in intelligent food packaging.

3.2 | Synthetic biopolymers

Alongside natural biopolymers, synthetic biopolymers have garnered significant attention in the development of intelligent packaging solutions with diverse smart sensors. Derived from either biomass or petrochemicals, these polymers undergo a polymerization process that transforms monomer units into long chains, a crucial step in their fabrication. As eco-friendly alternatives to traditional petroleum-based plastics, synthetic biopolymers stand out for their biodegradability. Their chemical structures are susceptible to breakdown through hydrolysis or microbial action into smaller entities, such as oligomers, dimers and monomers, making them appealing as sustainable packaging options.[31,32]

Commonly used synthetic biopolymers, including polycaprolactone, polylactic acid (PLA), polyvinyl alcohol (PVA), polyglycolic acid and polybutylene succinate, are highly valued in food packaging for their elasticity, transparency and processability. Among these, PLA and PVA have emerged as leading materials in the field of intelligent packaging. PLA, derived from the fermentation of lactic acid from sources such as corn, sugarcane, or sugar beet, is particularly appreciated for its clarity and rigidity. This makes it especially suitable for packaging fresh fruits and vegetables, where visual appeal is crucial. While PLA is biodegradable and adaptable to a variety of packaging needs, it has limitations in terms of sensitivity to heat, water vapour permeability, tensile strength and potentially high costs.[31,32] PVA, despite facing challenges similar to those of PLA, is distinguished by its ability to form films that are clear, flexible and smooth.[31,33–34]

To enhance the functional properties of PVA-based packaging, nanotechnology has been employed to incorporate nanoparticles, such as zinc oxide (ZnO), silver (Ag) and titanium dioxide (TiO_2), thereby improving mechanical and barrier properties to meet specific packaging demands.[24,31,33,34] These modifications have been shown to enhance water vapour and oxygen permeability, thermal stability and mechanical strength, creating a more robust network structure.[24,31,34] Such advancements make synthetic biopolymer-based films highly adaptable to a variety of food products, from meat and poultry requiring high barrier properties against oxygen to bakery items needing moisture control. Furthermore, various studies have demonstrated that ZnO and Ag nanoparticles endow PVA-based films with antimicrobial properties, particularly effective against pathogens like *Salmonella enterica* and *Staphylococcus aureus*.[24,31,33] This antimicrobial property is crucial for packaging applications where the inhibition of microbial growth can significantly extend the shelf-life and safety of food products, such as in animal-based products or for ready-to-eat meals. As Jayakumar et al.[24] discussed, lipid peroxidation and microbial spoilage are the primary causes of meat spoilage. The pH value of chicken stored in pure PVA film rose from 5.8 to 7.6 after 12 days, compared to the 5–7 days under normal storage conditions, demonstrating PVA's limited capability to postpone meat spoilage. In contrast, chicken wrapped in a nanocomposite film exhibited a smaller increase in pH to 6.3 over the same period. This finding highlights the nanocomposite film's effectiveness in preserving meat quality by acting as a microbial barrier. Essentially, the inclusion of nanoparticles significantly enhances PVA's role in extending the shelf life of perishable goods and improving food safety. The integration of nanoparticles into PVA-based packaging marks an advancement, augmenting its mechanical, barrier and antimicrobial properties. These enhancements not only extend the shelf life of packaged foods but also contribute to the sustainability and safety of the packaging industry, leading the change toward more eco-friendly and effective solutions in packaging technology.

4 | NATURAL COLOURIMETRIC SENSORS AND THEIR APPLICATIONS

Colourimetric sensors have become essential for the advancement of intelligent packaging, significantly improving food safety. Their effectiveness lies in their ability to undergo chemical structural changes in response to environmental variations. Broadly, these sensors are categorized into two groups: natural and synthetic colourants. Synthetic colourants, manufactured artificially, have raised safety concerns due to their potential migration into food.[12,35] Research indicates that

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such artificial colourants are associated with health hazards, including allergic reactions and instances of intoxication.[5,35] On the other hand, natural colourants, derived from sources such as plants, animals, fungi and minerals, are increasingly preferred by consumers.[5,35,36] These natural options have demonstrated their efficiency in monitoring the freshness of meats, seafood and dairy products. Nevertheless, their utility is limited due to their low sensitivity to minor environmental changes and inherent instability.[12] As the trend shifts towards solutions that are cost-effective, easy to use, accurate, reliable, eco-friendly and safe for detecting food spoilage, it is important to further investigate the capabilities and applications of natural colourimetric sensors.[10,36] While the primary objective of natural colourimetric sensors is to detect food spoilage indicators present in the packaging environment or food itself, their incorporation with biopolymers can also help mitigate deficiencies in the packaging's fundamental properties, thereby maintaining food quality. The practical uses of natural colourimetric sensors in conjunction with biopolymers in intelligent packaging are depicted in Table 1. Additionally, Figure 2 provides a summary of various natural colourants used as colourimetric sensors in intelligent packaging over the past 5 years, detailing the changes in their chemical structures and the corresponding colour responses.

4.1 | Alizarin

Alizarin, also known as 1,2-dihydroxyanthraquinone, is increasingly recognized for its application in food safety as a freshness indicator in packaged foods. It can be naturally derived from the roots of the madder plant. Alizarin exhibits pH-responsive discolouration, turning yellow under highly acidic conditions when the hydroxyl groups are protonated and shifting to a red hue under alkaline conditions as these groups become deprotonated. The colour change mechanism, involving proton transfer and the potential for hydrogen bond rearrangements between hydroxyl and carbonyl groups, can be influenced by increases in TVB-N, which is commonly associated with microbial growth.[12,21] In the food industry, alizarin is used as an indicator of meat and fish freshness, responding to changes in pH and TVB-N levels. The research conducted by Aghaei et al.[30,37] demonstrated its effectiveness in indicating spoilage in rainbow trout fillets, with colour transitions from yellow to magenta or violet correlating to rises in pH, TVB-N and total viable count (TVC).

Similarly, Ezati et al. investigated the use of alizarin as a colourimetric sensor in polysaccharide biopolymer-based films for monitoring the freshness of minced beef and fish. Their study explored alizarin's ability to detect meat spoilage by observing that the TVB-N content in minced beef progressively increased from an initial value of 11.66 mg/100 g to over 17 mg/100 g by day 4 at 4°C. This indicated spoilage, correlating with an increase in TVC from 5.58 $\log_{10}$ CFU/g to over 7 $\log_{10}$ CFU/g by day 4, thus highlighting microbial proliferation as a key factor in meat spoilage. The colour changes in the colourimetric indicator were visibly altered from brown to purple, marking the initiation of spoilage after 4 days of storage. The colour parameter (ΔE) values ranged from 17.16 to 46.32, showing a strong correlation ($R = 0.86$) between TVB-N contents and ΔE values.[38] Additionally, alizarin's application in detecting fish freshness was examined. During storage at 4°C, the pH of rainbow trout samples increased from an initial value of 6.24–6.71 by day 6, with a further rise to approximately 7 ± 0.1 by day 8. This was accompanied by an increase in TVB-N from an initial value of 13.53–19.6 mg/100 g by day 6, surpassing the spoilage threshold of 25 mg/100 g by day 8. Concurrently, the colour of the alizarin indicator changed from orange to reddish-brown, with a significant increase in ΔE values from being low at the fresh stage to 43.38 by day 8, indicating a strong correlation ($R = 0.87$) between the ΔE of the alizarin incorporated packaging and the TVB-N content of the fish.[39] Their studies confirmed the indicator's colour change in response to spoilage.

Beyond its sensory applications, Ezati et al.[40] developed a chitosan-based, pH-responsive functional film incorporating alizarin for active and intelligent food packaging. This alizarin-enriched film displayed enhanced UV-blocking properties, improved elasticity, surface hydrophobicity and thermal stability, as confirmed by scanning electron microscope (SEM) and Fourier-transform infrared spectroscopy (FTIR) analyses. While its antibacterial effect was minimal, the film's antioxidant activity was significantly boosted with the addition of alizarin, making it a promising tool in food safety and preservation.

4.2 | Anthocyanins

Recent scholarly investigations into natural colourimetric sensors have predominantly centred on anthocyanins. Anthocyanins, flavonoid compounds derived from 2-phenylchromen, display various colours depending on the pH. In acidic environments, anthocyanins appear as red flavylium cations. As the pH increases, anthocyanins reduce the red hue. Under neutral or alkaline conditions, they adopt purple or blue quinone base structures and may eventually transition to yellow chalcone structures. The stability and colour of anthocyanins, pivotal for their role as freshness indicators, are significantly influenced by pH.[12,21] This effect is further enhanced through interactions with polysaccharide biopolymers such as chitosan, starch, cellulose and gum. These interactions, often mediated by electrostatic forces, extend their π–π conjugate systems, intensifying the colour change.[21]

Anthocyanins are sensitive to pH fluctuations in their environment and packaged food, often resulting from microbial growth and associated metabolites. They can detect not only increases in pH due to TVB-N but also decreases caused by acetic acids and carbon dioxide.[41,42] Beyond their function as colourimetric sensors, anthocyanins have demonstrated antimicrobial and antioxidant properties in numerous studies. For instance, research by Liu et al.[43] focused on immobilizing anthocyanins within starch/PVA-based biopolymer composites to create cohesive films. These films serve as colourimetric sensors responsive to pH changes and exhibit antimicrobial effects against microorganisms such as *Bacillus subtilis*, *Aspergillus niger* and *Staphylococcus aureus*. Their study further tested the films' intelligent properties on pasteurized milk, monitoring spoilage over a 48-h period at room temperature. Spoilage was indicated by a change in colour to red, corresponding to a decrease in pH from 7.0 to 6.0, suggesting

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TABLE 1 Intelligent packaging practical examples: biopolymer films with natural colourimetric sensors for perishable food products.

Colourimetric sensor	Biopolymer	Environmental change	Response in colour	Food product	Reference
Alizarin	Zein	↑ pH (6.46–6.90)	Dark yellow → magenta	Rainbow trout fillets	[30]
	Cellulose-chitosan	↑ pH (6.25–7.06)	Brown → purple	Minced beef	[38]
	Starch-cellulose	↑ pH (6.24–7.00)	Orange → reddish brown	Rainbow trout fillets	[39]
Anthocyanin	Bacterial cellulose	↑ pH (6.36–7.09) (6.24–7.22)	Deep carmine → jelly bean Blue and khaki	Rainbow trout fillets Common carp fillets	[50]
	Cellulose-chitosan	↓ pH (6.60–5.70)	Blue to lilac → violet rose	Pasteurized milk	[51]
	Starch	↓ pH (6.50–5.70)	Deep blue → purple	Pasteurized milk	[52]
	Chitosan	↑ pH (6.85 – 7.08)	Reddish-purple → yellow	Hairtail	[56]
	Chitosan-cellulose	↑ pH (5.70-7.30)	Violet → violet to green/grey	Lamb	[60]
	Starch	↑ pH (5.96–7.45)	Pink/red/purple →green/yellow	Pork	[61]
Betacyanin	Pectin	↑ pH (6.90–7.84)	Light red → brown-yellow	Shrimp	[63]
	Starch-PVA	↑ TVB-N (7.07–35.18 mg/100 g)	Pink → pale yellow	Shrimp	[64]
	Glucomannan-PVA	↑ TVB-N (2.4–39.74 mg/100 g)	Purple → yellow	*Osphronemus goramy*	[65]
	Locust bean gum-PVA	↑ TVB-N (7.01–50.24 mg/100 g)	Reddish-purple →brown/yellow	Shrimp	[66]
	Chitosan-PVA	↑ TVB-N (4.7–54.3 mg/100 g)	Purple → yellow	Shrimp	[67]
Chlorophyll	Wheat gluten	↑ oxidation	Green → yellow	Sesame oil	[72]
Curcumin	Tara gum-PVA	↑ TVB-N (14.86–60.02 mg/100 g)	Yellow → orange-red	Shrimp	[17]
	Chitosan-gelatin	↑ TVB-N (6.41–16.3 mg/100 g)	Yellow → red	Pork tenderloin	[29]
	Chitosan	↑ TVB-N (6.21–35.15 mg/100 g) (3.48–54.25 mg/100 g)	Yellow → orange-red	Pomfrets meat Shrimp	[73]
	K-carrageenan	↑ TVB-N (4.91–31.11 mg/100 g) (7.15–41.53 mg/100 g)	Yellow → red	Pork Shrimp	[74]
	Lallemantia iberica seed gum	↑ TVB-N (6.66 – 43.89 mg/100 g)	Yellow → red	Shrimp	[75]
Shikonin	Cellulose	↑ pH (5.6–6.9) (5.9–6.5)	Red → dark purple	Rainbow trout fillets Pork	[78]
	Starch and agar	↑ pH (6.6 – 7.2)	Reddish-pink → blue-violet	Shrimp	[80]

anthocyanins' potential applications in detecting spoilage among dairy products.

Furthermore, incorporating anthocyanin-rich natural sources like purple potato and various berries (blackberry, blueberry, black chokeberry, jaboticaba) into these films not only enhances their efficacy for intelligent packaging applications but may also amplify their antioxidant activity, potentially contributing to the prolonged freshness of perishable goods.[44–49] Notably, anthocyanins extracted from red cabbage and black carrots are widely used in the creation of intelligent packaging. Numerous studies have demonstrated their efficacy in detecting spoilage in seafood such as fish and shrimp, pasteurized milk and in sensing environmental changes.[50–57] Although anthocyanins can increase the water vapour permeability of films, their impact on tensile strength varies depending on their interaction with biopolymers. For example, a film developed by Liang et al.[54] incorporating *Artemisia sphaerocephala* Krasch. gum and carboxymethyl cellulose sodium embedded with red cabbage anthocyanins showed decreased tensile strength and light transmission but improved elongation at break and transparency. This film underwent colour transitions from pink to green across a pH range of 3.0–10.0 and responded distinctly

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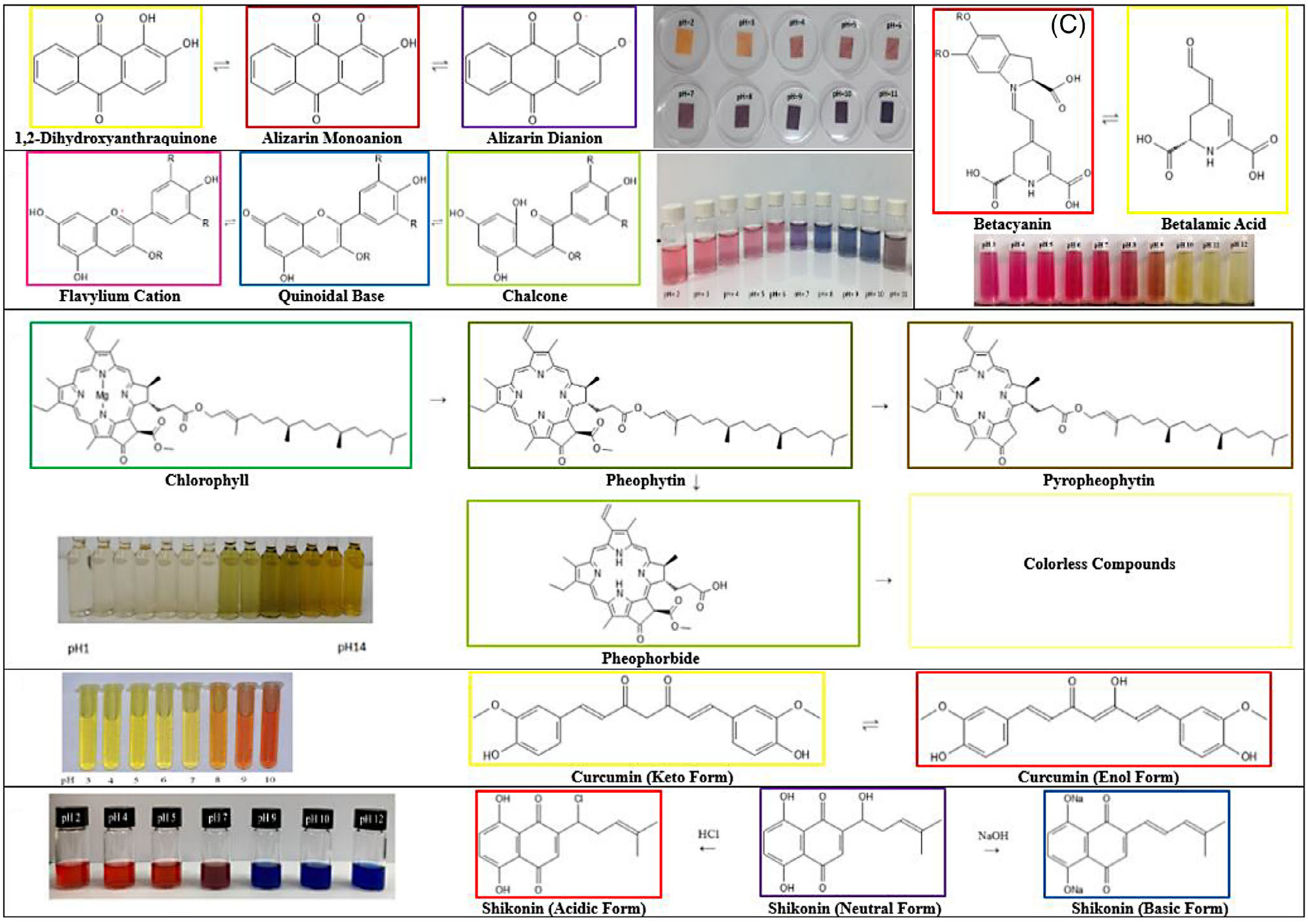

FIGURE 2 Chemical structure and associated colour changes in (A) alizarin,[38] (B) anthocyanins,[51] (C) betacyanins,[64] (D) chlorophyll,[70] (E) curcumin,[74] and (F) shikonin.[78]

to ammonia and changes in relative humidity at 33%, 75% and 90%. Conversely, Koosha et al.[53] focused on chitosan/PVA films infused with black carrot anthocyanins, using them as natural pH indicators and integrating bentonite as a nano-filler for intelligent packaging applications. The incorporation of anthocyanins led to enhanced tensile strength due to hydrogen bonding interactions and increased water vapour permeability. A notable colour change was also observed in the anthocyanin-infused films in response to pH variations. Other recent studies have explored anthocyanins from various natural sources, such as black/purple rice, eggplant, saffron petal, *Lycium ruthenicum* and roselle. Results indicate that anthocyanins combined with biopolymer-based films are an excellent choice for pH-sensitive intelligent packaging. They demonstrate improved gas barrier and water permeability, along with antioxidant and antimicrobial properties, reinforcing the insights gained from existing knowledge.[58–62]

4.3 | Betacyanins

Betacyanins, a group of red and purple pigments within the betalain family, are predominantly found in beets and pitayas, known for their vibrant crimson hues. These compounds are structurally stable in both acidic and neutral environments. However, in alkaline conditions, as well as under varying temperatures and light exposures, their structure and colour undergo significant changes. Specifically, betacyanin molecules transform into betalamic acid due to structural changes. This conversion affects their light absorption properties, which in turn alters their visible colour. Under strong alkaline conditions, betacyanins degrade into colourless and yellow compounds, resulting in a colour transition from red in acidic environments to orange and yellow as the alkalinity increases.[12,21] Beyond their role as natural colourimetric sensors, betacyanins exhibit a range of functional properties, including antibacterial and antioxidant activities, which are beneficial in intelligent packaging to preserve food quality. Since they are generally non-toxic or exhibit low toxicity, they are in line with the increasing preference for natural colourants in the food industry.[12,21] Together, these attributes make betacyanins suitable candidates for colourimetric indicators across various acid-base conditions, particularly for monitoring food freshness.

Betacyanins, extracted from pitaya peel waste, have been innovatively utilized in the development of intelligent packaging materials. These films change colour, shifting from red to brown-yellow, in

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response to pH and ammonia level changes, indicating spoilage by correlating with increases in TVB-N and pH values. Recent research has applied these films primarily to seafood, such as fish and shrimp, highlighting their potential for freshness monitoring. While betacyanins enhance the films' UV barrier properties, antioxidant and antimicrobial functions, they can reduce thermal stability, water resistance and mechanical strength. However, hydrogen bonding between polysaccharide biopolymers and betacyanins can improve water vapour barriers and mechanical properties.[63–65] Other studies using different betacyanin-rich sources, like cockscomb and cactus pears, report similar improvements and demonstrate the feasibility of utilizing betacyanin as an effective colourimetric sensor in intelligent packaging. The study by Wu et al.[66] illustrates that when shrimp spoilage, characterized by TVB-N levels, exceeds the permissible limit of 20 mg/100 g within 24 h, visible colour changes in the attached betacyanin-containing films, derived from cockscomb and transitioning from reddish-purple to brown/yellow, correlate with this spoilage. Similarly, films containing betacyanin, derived from cactus pears, are suggested as effective tools for monitoring the freshness of perishable seafood products. In the research conducted by Yao et al.,[67] these films are attached to the headspace of containers holding fresh shrimp to evaluate their sensitivity to TVB-N. The results reveal a gradual increase in TVB-N content, reaching 20.1 mg/100 g at 24 h, which surpasses the safety threshold for freshness, indicating spoilage. Significant colour changes in the films, from purple to orange and yellow, were observed, demonstrating their ability to monitor shrimp freshness effectively. Previous research also supports the use of films containing betacyanin-rich extracts from various sources on foods other than shrimp, such as milk, chicken and beef, highlighting their potential for broad application in food safety and quality control.[66] Overall, betacyanins provide enhanced pH and ammonia sensitivity in intelligent packaging and improve film properties when combined with polysaccharide biopolymers.

4.4 | Chlorophyll

Chlorophyll, the green pigment in plants, algae and cyanobacteria, shows promise as a colourimetric sensor due to its response to various stimuli. It reacts to heat exposure, oxidation, alkaline environments and metabolites produced by microorganisms, such as carbon dioxide, lactic acid and acetic acid. Upon exposure to heat and alkaline conditions, chlorophyll transforms from green to brown through a multi-step process. Initially, chlorophyll is altered into olive-green pheophytin by replacing its central magnesium ion with hydrogen. Subsequently, pheophytin further degrades into pyropheophytin through decarboxylation. In contrast, when stored with abundant oxygen, especially under acidic conditions, chlorophyll gradually fades, losing its bright green colour and resulting in a pale-yellow product. This loss of colour intensifies as pheophytin breaks down into pheophorbides and eventually into colourless compounds.[68,69] Notably, in alkaline environments, chlorophyll turns brown irreversibly. Immersing the films in an acidic solution leads to a complete colour washout, rendering the films transparent and colourless. This dramatic change is attributed to the total destruction of the colour pigments.[70]

Compared to other natural food colourants, the colour of chlorophyll is relatively stable, making it a less sensitive and observable colourimetric indicator in intelligent biodegradable packaging. This suggests that it may be less applicable for providing real-time information. In the study by Latos-Brozio et al.,[71] natural colourimetric indicators such as lutein, curcumin, beta-carotene and chlorophyll were utilized in combination with biodegradable substances like polylactide and polyhydroxybutyrate to develop intelligent packaging materials. The researchers compared the tolerance and colourimetric responses of each colourant under conditions of weathering, UV and thermo-oxidative ageing. They observed that polyesters containing chlorophyll exhibited less pronounced colour changes. This could explain why chlorophyll has seen limited study, with only one recent application on food products found. The study by Chavoshizadeh et al.[72] introduces a wheat gluten-based biodegradable film incorporating chlorophyll, highlighting its role in enhancing the shelf life of sesame oil and indicating expiration dates. The film reduces oil oxidation, evident from the halved peroxide value and changes colour from green to yellow in response to oil quality after a prolonged storage period. The underlying mechanism is that chlorophyll interacts with oxidants, causing the pigment to undergo a colour change from green to yellow due to oxidation reactions over time. While chlorophyll's transformative responses to various stimuli demonstrate its potential as a colourimetric sensor, its relatively stable colouration and irreversible chemical reactions under certain conditions limit its practical application in real-time intelligent packaging, necessitating further investigation in this field.

4.5 | Curcumin

Curcumin, extracted from the rhizomes of turmeric (*Curcuma longa*), plays a crucial role in intelligent packaging due to its unique properties. It exists in two tautomeric forms: enol and keto. In acidic and neutral environments, the keto form is predominant, while the enol form prevails in alkaline conditions. This molecular versatility leads to a significant pH-responsive colour change, transitioning from yellow in acidic or neutral environments to red in alkaline ones.[12,21] This colour shift is highly valuable in intelligent packaging, particularly within the food industry.

Recent studies have consistently shown curcumin's rapid and noticeable colour reaction to pH changes and rising TVB-N levels, which are readily observable to the naked eye. Both experimental results and practical applications in meat and seafood packaging clearly illustrate this phenomenon.[17,28,29,73–75] A study by Wu et al.[73] investigated the effectiveness of biopolymer-based films, incorporated with curcumin, in monitoring the freshness of pomfret meat and shrimp. During storage, the TVB-N levels in pomfret meat and shrimp significantly increased from 6.21 to 35.15 mg/100 g and from 3.48 to 54.25 mg/100 g, respectively, indicating a decline in seafood freshness due to microbial degradation. The films used to monitor this freshness changed colour from yellow to orange-red over five days, with a

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strong correlation (Pearson's correlation coefficient > 0.95) between TVB-N levels and the films' colour change, highlighting their practical application in the food industry for visual freshness assessment.

Furthermore, curcumin's responsiveness to increasing relative humidity has been established, along with changes in colour parameters in response to levels of ammonium and hydroxide ions, as evidenced by research conducted by Ma et al.[17] and Cvek et al.[18] When integrated into biopolymer composite films, curcumin serves not only as a colourimetric sensor but also enhances mechanical strength, UV protection and antibacterial qualities. It boosts the films' tensile strength, thermal stability and barrier capabilities due to robust interfacial interactions like electrostatic forces and hydrogen bonds.[73,74] The inclusion of curcumin prompts visible colour changes in response to different pH levels, serving as indicators of freshness, while simultaneously improving film properties to enhance food preservation and extend shelf life. Moreover, curcumin's notable features, including its non-toxicity, heightened antibacterial and antioxidant properties and health benefits such as anti-cancer and anti-inflammatory effects, make it an outstanding additive in food packaging.

4.6 | Shikonin

Shikonin, a naphthoquinone compound, possesses a highly reactive naphthazarin core. This reactivity makes it prone to polymerization when exposed to environmental factors such as oxygen, light, heat, acids, or bases. Shikonin can be derived from natural sources, most notably the roots of *Lithospermum erythrorhizon* and *Arnebia euchroma*.[76,77] In intelligent packaging, shikonin acts effectively as a colourimetric sensor due to its pH-sensitive nature. It is notably unstable and undergoes a significant colour shift from red-violet to dark blue in alkaline conditions, while it remains more stable in acidic environments. These pH-dependent colour transitions allow shikonin to serve as a reliable indicator of product freshness or spoilage in intelligent packaging applications.[12,21]

In a study conducted by Ezati et al.,[78] shikonin was adsorbed onto cellulose paper to create a pH-responsive colour indicator. This indicator changed colour in response to pH variations and maintained its effectiveness for 4 months. It also reacted sensitively to ammonia gas. The study confirmed its successful application in monitoring the freshness of fish and pork, with colour changes corresponding to pH fluctuations in the food. The pH of fish and pork samples stored at 25°C increased from 5.6 to 6.9 and from 5.9 to 6.5 respectively, over 36–48 h, correlating with significant changes in the colour of a shikonin indicator from red to dark purple, indicating spoilage. The indicator's colour change demonstrated a high correlation with pH levels, showing correlation coefficients of 0.98 and 0.99 for fish and pork respectively, effectively indicating the quality change and spoilage of the meat and seafood products. Moreover, incorporating non-toxic shikonin can enhance the UV-light barrier properties, mechanical strength and water vapour permeability of films.[76,79,80] The biopolymer composite film additionally exhibited antioxidant and antibacterial properties, especially against *Listeria monocytogenes*, as demonstrated by the experimental results of Roy et al.[77] and Ezati et al.[80] These characteristics make shikonin a compelling candidate for use in the development of intelligent packaging to preserve food safety.

5 | POTENTIAL BENEFITS AND CHALLENGES

There are several benefits associated with the integration of biopolymers and natural colourimetric sensors in packaging systems. Biopolymer-based food packaging, especially those made from polysaccharides and proteins, offers significant economic advantages, such as cost savings due to the use of renewable resources and the potential for recycling. Additionally, these packaging materials provide ecological benefits by reducing the carbon footprint, promoting biodegradability and utilizing renewable resources, thereby contributing to environmental sustainability.[22] However, it must be acknowledged that their properties, including mechanical strength, water vapour permeability, gas barrier properties and thermal resistance, are not as competitive as those of non-biodegradable packaging materials. While the use of nanoparticles or other synthetic materials can mitigate these drawbacks, concerns about the potential migration of metal nanoparticles and synthetic substances into food products and their unclear health impacts, such as toxicity, genotoxicity and carcinogenicity, remain.[31] On the other hand, natural colourimetric sensors offer a more eco-friendly and low-toxicity solution for smart sensing in intelligent packaging systems. Their inherent antioxidant and antimicrobial properties can further aid in effectively preventing food spoilage. However, responsive indicators based on natural sources suffer from lower sensitivity compared to synthetic indicators, exhibit poor colour stability under unfavourable conditions such as fluctuating temperatures or high humidity that can induce chemical reactions, primarily reflect the deterioration-inducing environmental conditions rather than accurately monitoring the actual food quality and face challenges in large-scale processing.[21,36] These issues highlight the limitations and gaps in the current use of colourimetric sensors in intelligent packaging solutions. Further research is needed to address these shortcomings and enhance the application of these components in intelligent packaging systems.

6 | CONCLUSION AND FUTURE PROSPECTS

The concept of embedding colourimetric sensors into biopolymer-based packaging represents an innovative breakthrough. It addresses critical challenges like global food shortages, contamination and food-borne diseases. These sensors undergo colour changes when exposed to various environmental conditions such as oxygen, temperature, pH and relative humidity, signalling the freshness and safety of the packaged food. Notably, recent developments have exploited the potential of natural colourants such as alizarin, anthocyanin, betacyanins, chlorophyll, curcumin and shikonin. These colourants are particularly suited for this purpose due to their natural origin, sensitivity to changes and compatibility with biodegradable polymers. The colour shift provided

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by these sensors offers consumers an intuitive and immediate indication of the food's condition, especially for perishable items. This innovation promises to significantly reduce foodborne illnesses by alerting consumers to spoilage or contamination before consumption. Beyond health benefits, this technology is crucial in combating food waste, thus contributing to the fight against food insecurity. It allows for more accurate judgment of food edibility, ensuring that consumable items are not discarded unnecessarily. Despite challenges in ensuring the longevity of these sensors and achieving widespread acceptance, this approach holds considerable potential for transforming the packaging sector into a key ally for food safety and environmental sustainability. Looking ahead, the future prospects in this domain are vast. Advancements in materials science and sensor technology may enable the development of even more sensitive and durable sensors. Additionally, as consumers become increasingly conscious of food safety and sustainability, the demand for such intelligent packaging solutions is expected to increase exponentially. Collaborative efforts among industry stakeholders, researchers and policymakers can accelerate the refinement and adoption of these innovations, solidifying their role in the next generation of food packaging solutions.

AUTHOR CONTRIBUTIONS

The manuscript was written through the contributions of all authors. All authors have given approval to the final version of the manuscript.

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CONFLICT OF INTEREST STATEMENT

The authors declare no conflicts of interest.

DATA AVAILABILITY STATEMENT

Data are available on request from the authors.

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