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Eco-friendly Polymeric Films for Smart Food Packaging: Preparation and Applications

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Abstract

With the increasing demand for environmentally friendly, safe, preservative and intelligent food packaging, there is a growing trend towards using plant-derived natural colorants that possess green, non-toxic, antioxidant, antibacterial, and pH-sensitive properties. As a result, the development of active intelligent packaging films containing plant-derived natural colorants has become a research priority in the realm of food packaging. The utilization of natural pigment-based films in packaging provides valuable insights for future development.

1. Introduction

Increasing attention is now focused on developing active and intelligent packaging films by incorporating pH-sensitive anthocyanins from various plant sources into biopolymer matri- ces (such as polysaccharides and proteins), incorporating plasticizers, nanofillers, and an anthocyanin extract for functional smart packaging applications. The core materials in- vestigated include Starch, Lipids, Agar-Agar (as biopolymers), and Cellulose Acetate (as a semi-synthetic polymer). These materials are selected due to their natural origin abun- dance for forming thin films. The films are designed to integrate an anthocyanin extract (red cabbage extract) as a pH-sensitive component, which is critical for their intelligent functionality. The final film properties are modulated by the inclusion of plasticizers (glycerol sorbitol) to enhance flexibility and reduce brittleness, and nanofillers (cellulose nanofibrils, ZnO, TiO_2 to improve mechanical strength, thermal stability, and barrier properties.

2. Description of Color Pigments

Anthocyanins are a major class of water-soluble pigments belonging to the parent class of molecules called flavonoids, which are synthesized via the phenylpropanoid pathway. They are considered polyphenols. The name derives from the Greek words ´anthos (flower) and kua´neos (dark blue). These pigments are found in the vacuoles of plant cells, particularly in the leaves, stems, roots, flowers, and fruits of higher plants. Anthocyanins are responsible for the vibrant red, pink, purple, blue, or black colors observed in many fruits and vegetables, such as berries, grapes, and red cabbage. Chemically, anthocyanins are the glycosides of their aglycone forms, which are known as anthocyanidins. The basic structural core of an antho- cyanin is the $C_6 - C_3 - C_6$ skeleton, specifically the flavylium ion (2-phenylbenzopyrylium cation), which has a positive charge on the oxygen atom of the C-ring. This highly conjugated structure is key to their coloration.

Anthocyanins are crucial in plants for attracting pollinators and seed dispersers, and they also protect the plant from UV damage and cold stress. Their color and stability are highly dependent on the surrounding pH, making them excellent natural pH indicators (a characteristic utilized in the current film development). They typically appear red in acidic conditions ($pH < 7$) and shift toward purple and blue as the pH increases.

Classification of anthocyanins is primarily based on their structure. The simplest clas- sification divides them into the sugar-free anthocyanidin aglycones and the anthocyanin glycosides (which have one or more sugar moieties attached). The most common natural an- thocyanidins (aglycones) are the C_{15} compounds that differ by the number and arrangement of hydroxyl (-OH) and methoxyl (-OCH_3) groups on the B-ring. The six most common and widely distributed anthocyanidins found in food sources are : Cyanidin (Cy) Delphinidin (Dp) Pelargonidin (Pg) Peonidin (Pn) Petunidin (Pt) Malvidin (Mv) . Anthocyanins are further classified by the number and nature of the sugar groups (e.g., glucose, rhamnose) attached to the aglycone (often at the C-3 position) and whether these sugars are further modified by aliphatic or aromatic carboxylic acids (acylated anthocyanins). As of 2006, over 550 different anthocyanins have been reported.

2.1.Bacteriostatic Efficacy

Anthocyanins exhibit significant bacteriostatic activity, meaning they inhibit the growth and reproduction of various bacteria, including common foodborne pathogens such as Staphy- lococcus aureus and Escherichia coli. This antimicrobial effect stems from the compounds belonging to the flavonoid group of plant polyphenols, which interfere with microbial func- tions. The primary mechanism of action involves disrupting the integrity and function of the bacterial cell membrane. Anthocyanins have been shown to increase the surface hy- drophobicity of bacterial cells and subsequently increase the cell membrane permeability. This action leads to the leakage of essential intracellular contents, such as ions and cyto- plasmic material, thereby inhibiting the bacteria's ability to generate energy and synthesize proteins. Furthermore, these compounds can reduce the formation of biofilms, a critical factor in microbial contamination and resistance.

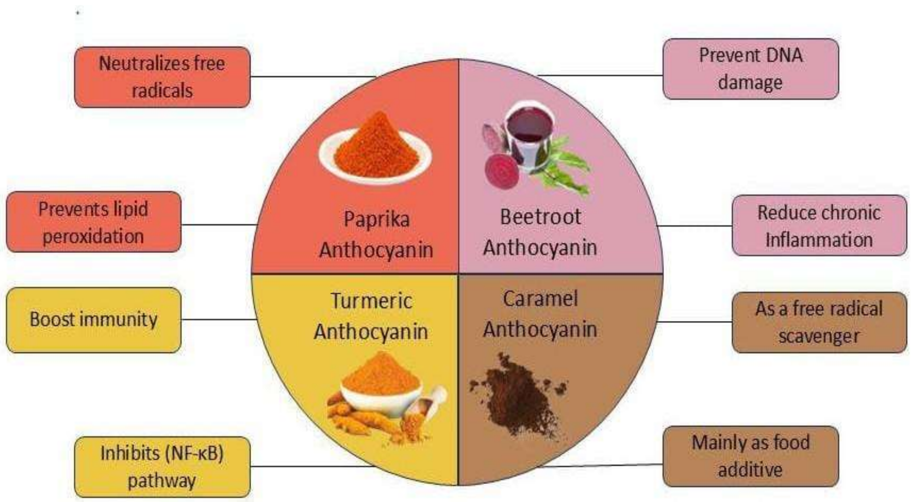

Figure 1: Anti-inflammatory and Antioxidant properties of different anthocyanins

Generally, Gram-positive bacteria (e.g., S. aureus) tend to be more susceptible to an- thocyanin action than Gram-negative bacteria (e.g., E. coli), likely due to differences in the complexity and structure of their respective cell walls. This powerful, natural inhibitory capacity makes anthocyanins, such as those derived from red cabbage, promising ingredients for developing active and intelligent food and packaging films.

2.2.Anti-inflammatory Activity

Anthocyanins are potent antioxidants that directly scavenge harmful free radicals, reducing oxidative stress in the body. These combined effects help prevent chronic diseases linked to inflammation and oxidative damage. Their anti-inflammatory action involves inhibiting pro-inflammatory pathways, such as the NF-κB pathway, and modulating enzymes like cyclooxygenase (COX).

2.3.Antioxidant Activity

Anthocyanins are potent antioxidants that combat oxidative stress through two main mechanisms. First, they act as direct free radical scavengers, readily donating a hydrogen atom or an electron to neutralize reactive oxygen species (ROS) and prevent cellular damage. Second, they exert indirect effects by modulating gene expression, such as activating the Nrf2 pathway. This pathway enhances the synthesis and activity of the body's own antioxidant enzymes, like catalase and superoxide dismutase (SOD), boosting the overall defense system.

3.Fabrication Techniques

The fabrication of anthocyanin-based intelligent active packaging primarily involves incorporating the pH-sensitive anthocyanin extract into a suitable biopolymer matrix. The most common and effective techniques fall into the following categories:

3.1.Solvent Casting Method (Solution Casting)

This is the most widely reported and simplest method for preparing thin, flexible films, especially on a lab scale. The anthocyanin extract is mixed thoroughly with a solution of the chosen polymer (e.g., chitosan, starch, pectin, gelatin, or PVA) and a plasticizer (like glycerol). The homogeneous solution is then poured into a flat mold and allowed to slowly evaporate at room or controlled temperature, leaving a solid film. Excellent for creating indicator films that are highly responsive to pH changes (volatile compounds like ammonia released during food spoilage).

3.2.Extrusion

Extrusion is a continuous, large-scale industrial process that offers better control over film thickness and mechanical properties, but requires specialized equipment. The polymer, plasticizer, and anthocyanin are mixed and fed into an extruder. This process involves heating and shearing the mixture while forcing it through a die to form a continuous film or sheet. Used for commercial-scale production of films. The challenge is that the high heat used in extrusion can sometimes degrade the heat-sensitive anthocyanin, requiring careful process optimization.

3.3.Electrospinning

This technique is used to create nanofibers (ultrafine fibers) which result in high-surface-area films, potentially improving sensitivity and gas permeability. The polymer-anthocyanin solution is fed through a syringe and subjected to a high voltage electrostatic field. This draws the solution out, causing the solvent to evaporate and leaving a non-woven mat of solid nanofibers on a collector plate. Films with enhanced mechanical strength and extremely fast color response due to the high surface area of the nanofibers.

4.Applications of Anthocyanin-based Smart Packaging

Anthocyanin-based intelligent packaging plays a dual role in food storage, acting as both an active preservative and a smart freshness indicator. particularly for protein-rich products like meat and seafood, In its indicator function, the pH-sensitive anthocyanin pigment, often incorporated into biodegradable polymer films (like chitosan or starch), provides a real-time visual assessment of spoilage. As poultry products and livestock meat products (such as beef or pork) deteriorate, spoilage microorganisms break down proteins into volatile compounds like ammonia and total volatile basic nitrogen (TVB-N), which increases the local pH within the package headspace. The anthocyanin pigment responds to this alkaline shift by undergoing a visible color change, typically transitioning from an initial pink or red (acidic pH) to purple, blue, or green (neutral/alkaline pH), allowing consumers or retailers to rapidly and non-destructively determine the product's safety status.

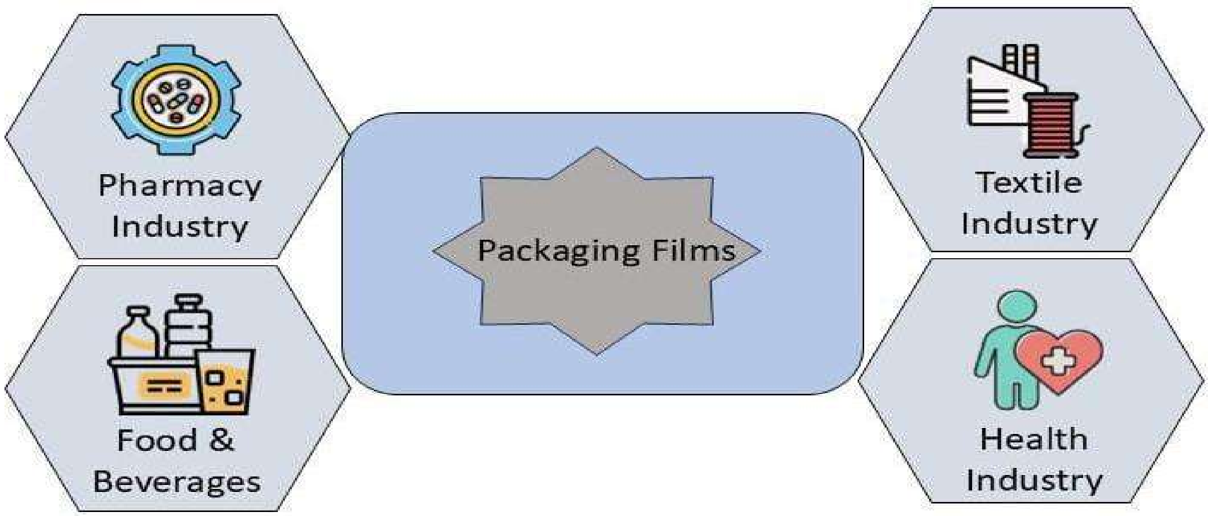

Figure 2: Multidisciplinary Functional Roles of Sustainable Smart Polymeric Films.

Similarly, for fish products and crustacean products like shrimp, which are highly perishable, this intelligent system is crucial as their spoilage also results in the rapid generation of alkaline TVB-N; an anthocyanin-laden film quickly changes color (e.g., from red to green/yellow), directly correlating with the loss of freshness.

5. Constraints of Real Time Monitoring

The primary challenge lies in the low chemical stability of anthocyanin pigments. They are highly susceptible to degradation and color fading when exposed to light, heat (especially during film fabrication processes like extrusion), oxygen, and enzymes. This instability undermines the indicator's reliability over long storage periods. Further challenges include cost and industrial scalability. Extracting and purifying natural anthocyanins consistently is expensive and often yields batch-to-batch variability. Integrating these sensitive extracts uniformly into polymer matrices at a large commercial scale, while maintaining the film's essential mechanical and barrier properties, proves technically difficult. Finally, regulatory approval and ensuring consumer comprehension of the color-change signal across diverse markets are also significant hurdles for commercial adoption.

Beyond technical issues, consumer acceptance and interpretation fidelity pose adoption challenges. The slight color variations due to different food matrices or background colors can lead to misinterpretation of the freshness signal by the end-user. Moreover, the higher production cost compared to conventional packaging hinders its widespread commercial viability, especially for low-margin products, requiring strong evidence of tangible benefits to justify the added expense.

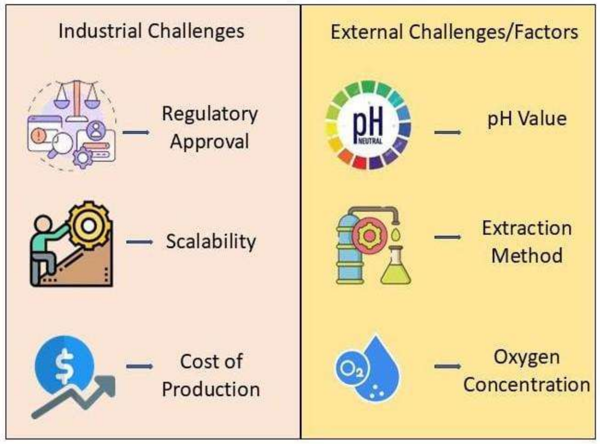

Figure 3: Challenges of application of anthocyanins

6. Conclusion

Smart packaging solutions utilizing anthocyanins are emerging as a dual-action approach for monitoring food quality and extending shelf life, primarily through their natural pH-sensitive properties. This analysis examines current advancements in manufacturing processes like electrospinning and microencapsulation, which are critical for maintaining the bioactive integrity of these pigments. By embedding these natural indicators into sustainable biopolymer frameworks, researchers can achieve a balance between ecological responsibility and functional precision. The study concludes that the effectiveness of these intelligent systems is dictated by the specific interaction between the active agents and the polymer matrix, necessitating more standardized protocols for commercial application.

Advancing eco-friendly smart packaging from a concept to a market-ready product requires a deeper focus on how biodegradable matrices behave under fluctuating environmental stressors, such as varied humidity and mechanical handling, By reflecting critically on both technological progress and existing bottlenecks, this review underscores the need for interdisciplinary innovation to fully realize the potential of anthocyanin-based packaging. With continued development, these systems could serve as a sustainable and multifunctional alternative to conventional indicators, aligning food packaging innovation with safety, functionality, and environmental responsibility.

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