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*Review Article*

# Intelligent pH Indicative Film from Plant-Based Extract for Active Biodegradable Smart Food Packing

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*Background*. Biodegradable biopolymers have been developed in response to the growing environmental concern over plastic packaging disposal. The emergence of active and intelligent packaging systems to monitor the quality of packed food is further necessitated by consumer demand and health concerns. Chemical dyes, on the other hand, are not ideal for use as an indicator in smart packaging for food due to their high toxicity and negative impacts on human health and the environment. As a result, the researchers are concentrating on natural pigments produced from plants and food waste as a signaling component in biodegradable packaging as well as the valorization of food waste. This review is focused on the advancement of active packaging from plant pigments.

## 1. Hydrogel

Hydrogels are defined as hydrophilic, three-dimensional polymers crossed along a correlated absorption interval of 10 g/g [1]. The hydrogel has a wide demand in various sectors such as agriculture [2], drug discovery, and water purification [3, 4]. Bashir et al. were the first to write on the use of hydrogels in 1960 [5]. Hydrogel could be a three-dimensional chemical compound structure that swells once exposed to water and includes valency bonds fashioned by the reaction of 1 or additional monomers, attachment bonds like van der Waals interactions, and chemical element bonds between chains. Hydrogels could take the shape of rigid formed forms (soft contact lenses), iron-based powder (pills or tablets for oral ingestion), small particles (as bioadhesive carriers or wound treatment), coatings (on implants or catheters), membranes or sheets, and encapsulated solids and liquids [6].

The analysis is based on making a food packaging film developed with property and perishable materials. As a result, poly(vinyl alcohol) (PVA) is replaced by chitosan (CS) in the packaging film. What is more, anthocyanin

(ATH), a food colorant widely used and extracted from red cabbage during this analysis, is more to the forged film for acclimatization [7].

Chitosan (CS) is a natural polymer derived from chitin and is abundant in crab and shrimp shells [8]. Due to the significant amino and hydroxyl functional groups in chitosan, it has unique traits such as polycationic nature, chelating properties, and film-forming potential. It shows different biological activities as antimicrobial activity, biodegradability, etc. [8]. For further processing of polymer films, chitosan is often blended with other polymers with more versatile chains, such as poly(vinyl alcohol) (PVA) [9].

PVA is an emulsifiable nontoxic synthetic polymer of elasticity and high tensile strength and resilience, as well as low gas permeability, such as $O_2$ and $CO_2$. At extreme temperatures, PVA can have mild water solubility [10]. Due to its excellent film-forming properties, PVA has been widely used in application as thin films, such as food packaging and medicinal application. Anthocyanins are water-soluble pigments found in a variety of plants, including red cabbage, blueberries, eggplants, and flowers. Depending on the pH values, they may use it as a color indicator. It obtained from red cabbage in particular and can range from red to purple and to blue at various pH values [11, 12]. It makes it easier to identify the food quality in terms of pH. To increase the mechanical strength of cast film cross-linking substances such as sodium tripolyphosphate (STPP), glyoxal and glutaraldehyde must be added during the processing of thin polymer films.

pH indicative films were successfully synthesized from hydrogels made by combining 1% poly(vinyl alcohol) (PVA) and 1% chitosan (CS) with anthocyanin (ATH) and sodium tripolyphosphate in this sample (STTP). To improve the mechanical properties of the cast films, ATH extracted from red cabbage was used as the pH indicator, and STTP was used as the cross-linking agent. The presence of the ATH in the cast films was verified by FTIR spectra. The cast film tensile strength, elongation-at-break, and swelling indices were also determined. The compositions of PVA/CS and the STPP dosage applied in the hydrogels had a significant impact on the properties of pH indicative films. If 35 percent of the PVA hydrogel was replaced with CS, the tensile strength of a film cast from pure PVA hydrogel could drop from 43.27 MPa to 29.89 MPa. The cast films were used as a food wrap that could be used to visually track the consistency of the enwrapped food by changing color as the pH values of the enwrapped food change. In practice, a sequential shift in color on the pH suggestive films partially enwrapping the pork belly was successfully observed, signaling the meat's spoilage [7].

## 2. Polymer-Based pH Sensitivity

pH-sensitive polymers are polyelectrolytes with weak acidic or basic groups in their structure that accept or release protons in response to changes in pH. These acidic or basic polyelectrolyte groups can be ionized in the same way as the acidic or basic groups of monoacids or monobasic; however, complete ionization of these structures is more difficult due to their larger size to the electrostatic effects of other nearby ionized groups. It does not contain acid or low acid, i.e., carboxylic, or basic, i.e., ammonia. In response to the pH of the environment, these release protons or accept free protons. Under these pH conditions, the functional groups presented along the backbone and side chains ionized, causing the polymer to swell or dissolve [13]. pH-sensitive anionic polymers are formed on PAA or their derivatives. These systems typically produce anionically charged fractions at pH levels above their pKa, which may attract positively charged therapeutic agents [14].

Polymers that respond to pH can be linear, branched, or networked. Depending on their systems, they can have different responses to solution conditions and different self-assembly behaviors. pH change induces deswelling in hydrogel and dendrimer-like structures. Surface modified with polymers allows for the creation of ionic surface and thin/thick layers as a result of pH changes. pH modifications cause changes in polymers of various architectures [15]. These pH-sensitive polymeric systems' intelligent properties are appealing for application in life sciences and the chemical industry, with possible applications in managed drug delivery, personal care, industrial coatings, oil exploration, and water remediation, among others [16].

Several popular polymerization methods can be used to make pH-sensitive polymers. Depending on the form of polymerization, functional groups need to be prevented from reacting. The masking is often removed to revive polymerization to restore pH-sensitive functionality. Because of the molecular weight distribution, living polymerization is commonly used to create pH-sensitive polymers. Some examples include group transfer polymerization (GTP), atom transfer radical polymerization (ATRP), and reversible addition-fragmentation chain transfer (RAFT). Graft polymers, which have a backbone with branches, are a common form of synthesis. The branch structure can be modified to achieve various properties [15].

## 3. Food Packing

Food packaging is one of the most important aspects of a commodity from the consumer's viewpoint as well as one of the most important in modern commercial trade [17, 18] since it ensures food quality and protection, aids in transportation, allows safe storage, avoids product harm and loss, reduces economic losses, aids in product marketing, and indirectly preserves consumer's health [19]. The typical and traditional food packaging, which is comprised of petroleum compounds, is a safe device. It protects the food product from microbial/physicochemical deterioration, as well as ambient conditions and external stimuli, extending its shelf life [20].

Biopolymers such as proteins, polysaccharides, and their derivatives are natural polymers that degrade in the environment as a result of natural physical, chemical, and biological processes especially microorganism metabolism. It could be separated by a variety of natural resources. Plant-based polysaccharides such as chitosan, starch, cellulose, alginate, agar, carrageenan, pectin, and various gums are commonly used

[21, 22]. While one of the functions of food packaging is to ensure food quality and safety, modern packaging must also notify the customer about food quality and sustainability for consumption. Several smart packaging systems based on colorimetric indicators will provide consumers with real-time quality monitoring for food items through quality sensors/indicators for this reason of nowadays vast development [23].

Due to their low nontoxicity, eco-friendliness, easy preparation, biodegradability, low cost, availability, sustainability, and pollution-free properties, smart packaging based on natural colors and biodegradable films has recently emerged as an attractive alternative for use in food packaging among various indicators' and sensors' freshness applied in food systems. These natural colorants embedded in the biopolymeric film matrix appear to change color as the physiological condition of the food change during spoilage, thereby informing the customer of the packaged food's consistency and suitability for consumption. [24]

## 4. Plant Pigment

Pigments are found in every organism in the world, and plants are the major producers; therefore, pigments are responsible for the colors we see every day. Leaves, fruits, vegetables, and flowers, as well as the skin, eyes, and other animal tissues, as well as germs and fungus, contain them (Figure 1). Medicines, foods, clothing, furniture, cosmetics, and other products employ natural and synthetic pigments [25]. Pigments are chemical substances that absorb light in the visible wavelength range. The color is produced by a molecule-specific structure called a chromophore. This structure absorbs the energy, causing an electron to jump from an exterior orbital to a higher orbital. The nonabsorbed energy is reflected and/or refracted to be captured by the eye, and the resulting neural impulses are transferred to the brain, where they can be interpreted as a color [26].

Natural, synthetic, and inorganic pigments are classed according to their origin. Living species such as plants, animals, fungi, and bacteria produce natural pigments. Laboratory-made synthetic pigments are available. Organic pigments, both natural and manmade, are organic substances. Inorganic pigments are present in nature or can be made synthetically [27]. To give the desired functional qualities of a pH indicator, natural colors such as anthocyanins can be incorporated to biodegradable starch films. Anthocyanins are secondary metabolites found in a wide range of fruits and vegetables (e.g., red cabbage, sweet potato, bean husk, and grapes), making them a viable source of natural pH indicators [28].

## 5. Red Cabbage Pigment

The red cabbage (*Brassica oleracea L.* var. capitata F. rubra) is such a cabbage, which is recognized by its shading. The leaf of the red cabbage is more somewhat blue-purple versus red tone and is orbicular and firmly wrapped with waxy leaves. Red cabbage is currently developed and exchanged universally as a colorant in the food business; notwithstanding, it has been utilized for restorative purposes [29]. The red cabbage was mixed with phinolic content with antocyanins are the significant properties [30]. Anthocyanins can be delivered via the phenylpropanoid pathway and are classified as a flavonoid of class [31]. Red cabbage is a rich birthplace of anthocyanins, which can use for common tinge. Its tone is pH-subordinate which is red to blur tone over an expansive pH range [32]. There are 24 different types of anthocyanins in red cabbage, as well as sweet-smelling and aliphatic acids [33].

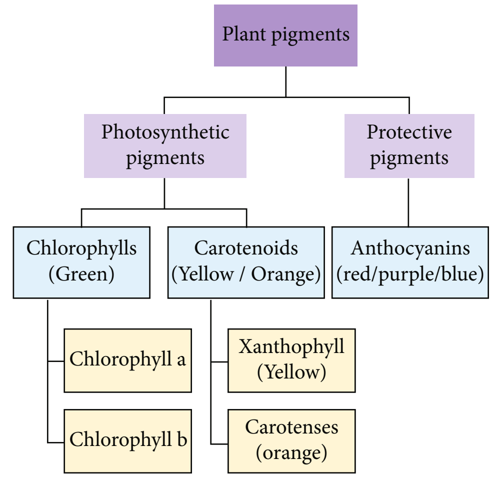


Figure 1: Types of plant pigments.

Antihypertensive, cancer prevention agent limit, hepatoprotective, and antihyperglycemic effects are some of the physiological properties of anthocyanins [34]. Red cabbage's most common anthocyanin is cyanidin-3-diglucoside-5-glucoside, which can be nonacylated, monoacylated, or diacylated with caffeic, p-coumaric, sialic, and ferulic acids [35]. Anthocyanins are the most abundant water-soluble colorants, responsible for the blue, purple, and red hues found in many blooms and plant leafy foods. In this approach, red cabbage could be used as a distinctive colorant in the food industry [36].

Red cabbage is abundant in minerals, nutrients, oligosaccharides, and bioactive chemicals such anthocyanins, flavonols, and glucosinolates, all of which are beneficial [37]. Crimson cabbage is also valued by buyers for its flavour and as a source of a deep red color that enhances the food's taste. As a result, red cabbage is a popular and frequently used vegetable as a fresh cut portion of mixed greens. Similarly, red cabbage is characterized by a lengthy period of usage, implying that it might be easily stored and available in a new structure for the duration of the year [38].

## 6. Pigment, Gel, and Film

Red cabbage (*Brassica oleracea L.*) is a palatable source of anthocyanins with a high content and prospective production per unit area [39]. Anthocyanin removal from red cabbage is known to result in high levels of mono- or diacylated cyanidin anthocyanins [40]. Anthocyanin type and acylation are two important factors that determine their shading properties at different pH levels [41]. Because of the pH of the environment, red cabbage anthocyanin concentrates can display a wide spectrum of shading from orange to red to purple and blue due to its anthocyanin structures. Anthocyanin acylation affects their cancer-preventive qualities as well as their food strength [42]. Anthocyanins make up the largest group of water-soluble colorants and are responsible for the blue, purple, and red hues found in many soil-borne flowering plant products. Red cabbage could be used as a flavouring agent in food production and as a pH indicator [43].

Because of the arrangement of covalent bonds, ionic communication, hydrogel holding, and hydrophobic connections, three-dimensional organizations of hydrophilic polymers in hydrogels stood out towards the fuse of a greater number of medications for their application in lethargic maintained and controlled medication conveyance [44]. Chitosan (CS) is a characteristic polymer got from chitin and discovered richly in the shells of crabs and shrimps [8]. Chitosan is not generally best in polymer handling for film creation attributable to its mediocre mechanical properties like lower elasticity, less lengthening-at-break, and Young's modulus. Poly(vinyl alcohol) (PVA) is a water-dissolvable biodegradable engineered polymer with huge pliable strength, and the composites mixed with PVA and chitosan are known to have improved steadiness, biocompatibility, and mechanical strength, comparative with those of unadulterated PVA and unadulterated CS polymers [45].

Dissolvable projecting and hot softened expulsion are presently two of the most prominently applied preparing strategies for the creation of polymer flimsy movies. The hot liquefy expulsion measure regularly requires the contribution of high energy, for example, mechanical energy as extremely high shear pressure as well as nuclear power, to guarantee the prepared polymer in the softened state. In that capacity, this technique is not appropriate for preparing a polymer with heat and additionally shear delicate particles. Conversely, dissolvable projecting is the prevalent technique for assembling films containing temperature-delicate fixings, for example, anthocyanin, because the temperature needed to vanish the dissolvable is regularly lower than the cycle temperature of the hot liquefy expulsion [46].

## 7. Food Packing-pH Film

In recent years, public concern over the disposal of traditional synthetic plastics has grown, particularly when the entire time of deterioration is long. As a result, biodegradable film research has seen a boom in interest. Many studies investigated biodegradable films, such as edible films and coatings manufactured from edible ingredients, in the hopes of increasing food quality and extending shelf life [47]. Chitosan is a natural cationic polysaccharide and a diacylated chitin derivative derived primarily from shellfish processing waste. Chitosan has antimicrobial properties, which means it can stop fungi, yeast, and bacteria from growing. Chitosan film has been discovered to have high mechanical strength, flexibility, biodegradability, and antibacterial properties [48].

The food industry has invested in smart packaging in shelf in response to customer demand for fresher items with a longer shelf life (Figure 2). The packaging smart with pH indicators was created with the aim of encouraging a deliberate relationship between food and packaging in order to improve quality characteristics [49]. As it is understood that food deterioration is linked to pH changes in the product, consumers can detect these changes in the food by simply adjusting the color of the packaging [50].

Since plastic is commonly used in many applications, including food packaging, the development of pH indicator packaging can come from either fossil or renewable sources. Plastic, on the other hand, has a detrimental effect on the climate because it pollutes the environment and is not biodegradable plastics using starch, alginate, and natural fibers, for example, which can be used as food coatings due to their degradable properties and conservation ability and is a revolutionary solution to this problem aimed at environmental preservation and partial replacement of traditional polymers derived from crude oil [51].

Haghighi et al. [51] explained that the recent sharp rise in exposure to environmental concern arising from plastic packaging has sparked interest in more environmentally friendly packaging materials. This latest trend encourages the commercialization of information through the use of chitosan-based films. Because of its unusual biological and functional properties, chitosan has been extensively researched and used. However, inherent flaws such as a low mechanical properties and high susceptibility to humidity restrict its industrial applications, which include food packaging. The scientific literature addressing chitosan-based films for their potential application in the food packaging industry has been extensively reviewed in the current research. The paper summarizes the various techniques used to resolve inherent flaws in chitosan-based films and enhance their properties, with a focus on blending with natural and synthetic biopolymers [52].

Wang et al. explained that chitosan and poly(vinyl alcohol) (PVA) were used to create a semi-interpenetrating polymeric network that was cross-linked with glutaraldehyde. The chitosan had a molecular weight of 612 kDa and a degree of deacetylation of 72 percent, respectively. The chemical bonds formed by cross-linking reaction were investigated, as well as their transformation in different pH media. The mechanical properties of the hydrogel and the gelatin property of the chitosan-PVA gel solution were investigated. The formation of Schiff's base (C=N) and $-NH3+$ was suggested by the FTIR spectra of the hydrogel before and after swelling at pH 3 and pH 7. They also demonstrated the pH-induced transformation of C=N to C-N and –NH3+ to –NH2, as well as the Schiff's base instability. The chitosan is required from hydrogel formation due to the Schiff's base reaction between the chitosan amino groups and the glutaraldehyde aldehyde groups. The addition of PVA enhanced the hydrogel's mechanical properties. PVA, on the other hand, appears to leach out in the acidic medium during longer swelling periods due to hydrolysis of the gel networks, Schiff's foundation [53].

Alizadeh-Sani et al. clarified that the new ascent in familiarity with safe food and changing buyer perspectives has

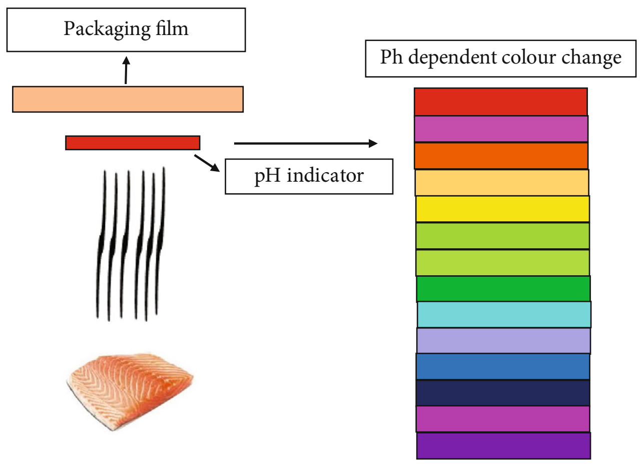


FIGURE 2: pH-dependent color change of food material.

acquired development bundling innovation. Customers are progressively requesting common food colorants like carotenoids, betadine, anthocyanins, and chlorophylls instead of engineered colors for food applications. Accordingly, shrewd bundling dependent on characteristic colorants and biopolymers has been presented as the most recent innovation in the food bundling field. Keen items shield food from natural risks, yet additionally convey ongoing messages (colorimetric, synthetic, or electrical) to shoppers for changes in the bundling climate and food quality [21].

Carvalho et al. explained that anthocyanin shades are appropriate as regular colors for food, makeup, and dietary enhancements, because of the interest for better items and their cancer prevention agent properties. This work meant to extricate the anthocyanin colors from red cabbage and its partition of the arrangement by adsorption activity onto chitosan films. The anthocyanins were extricated from red cabbage in hot water at 90°C by 15 min. Chitosan was got from shrimp waste, and its films were created by projecting procedure (rigidity of $25.1 \pm 1.9$ MPa, stretching of $10 \pm 3.5$ %, and thickness of $103.1 \pm 1.3\,\mu$m). The anthocyanin adsorption tests were acted in cluster, and the most noteworthy adsorption limit was around 140 mg $g^{-1}$ [30].

Pereira and colleagues elaborate on this food packaging with time-temperature indicators as one of the so-called intelligent packages. They use a gadget that monitors the state of food in real time, demonstrating the overall impact of temperature on food quality. The goal of this study was to develop and characterize a temporal temperature indicator (TTI) based on a PVA/chitosan polymeric doped with anthocyanins that may be utilized to detect changes in the pH of packaged foods exposed to incorrect storage temperatures. To manufacture the TTI, chitosan, PVA, and anthocyanins were taken from Brassica oleracea var. capitata (red cabbage).TG-DSC, FTIR, UV-Vis, and swelling index (Si) methods were used to describe the TTI. The color variance following activation by various pH values was calculated using the CIELAB scale. The mechanical parameters of the TTI were determined using stress/strain tests. Despite having a lower modulus of elasticity than commercial polymers used in food packaging, the produced TTI has physicochemical qualities that make it desirable for use in intelligent food packaging. An activation test on pasteurised milk with obvious changes in the coloring of the film, which is crucial for signaling to consumers that the food has been subjected to changes in its chemical composition, is supported by the TTI shown here [54].

Castillo et al. explained that chitosan and starch are biodegradable polymers with excellent film-forming capabilities and a wide range of food-related applications, including active and smart packaging that can track and inform customers about food conditions in real time. As a result, we provide a pH monitoring system based on chitosan, corn, starch, and red cabbage extract, all of which are inexpensive and renewable. Cornstarch, a medium-molecular-weight chitosan, and Brassica oleracea var. capitata phytochemical extract (red cabbage), TG-DSC, FTIR, water vapour transmission rate, and light microscopy were used to characterize the device. The color variance following activation in various pH ranges was calculated using the CIELAB approach. To confirm the device's utility as a fish spoilage detection sensor, application tests using fish fillets were conducted. The gadget exhibits strong optical and morphological features, as well as being particularly sensitive to pH changes, according to these findings. During the application test, the equipment visually displayed pH changes. As a result, the system responds quickly to changes in sample pH. As a result, it might be utilized as a visual indicator of how food is stored and consumed [55].

Balbinot-Alfaro et al. said intelligent packaging can emit a signal (electric, colorimetric, etc.) in real time in response to any improvement in the initial packaging conditions and food quality, in addition to acting as a food safety barrier. The colorimetric sensor in pH indicators or pH sensors is normally made up of two parts: a solid base and a dye that are sensitive to pH change. The dyes are derived from a variety of fruits and vegetables, as well as synthetics. The pH of food changes at the start of the degradation process; this transition is one of the measures of product quality. Packaging with a pH indicator is a safety measure that can signify the consistency of the food at the time of purchase prior to consumption. The aim of this research is to improve the characteristics and applicability of this indicator. This review paper includes the studies on pigments, polymers, food, and packaging solution, as well as an overview of the materials/technologies used in the production and the perspectives/challenges that this new technology brings [24].

Hydrogels consisting of cellulose are hydrophilic materials that can absorb and hold a considerable quantity of water in their interstitial locations. They contain a variety of organic biopolymers such as cellulose, chitin, and chitosan. These polymers exhibit a wide range of outstanding features, including reactivity to pH, time, temperature, chemical species, and biological circumstances, as well as a high propensity to absorb water. Biopolymer hydrogels can be modified and created for a wide range of uses, prompting a recent increase in scientific research. Researchers all over the world are focused on naturally generated hydrogels in response to increasing environmental challenges and demand because of their biocompatibility, biodegradability, and availability. Biocompatible materials, such as cellulose hydrogels, can be utilized in medical devices to treat, complement, or replace any tissue, organ, or biological function. These hydrogels could be employed in agriculture, as smart materials, and in a variety of other applications. This review summarizes recent and ongoing research on the physiochemical properties of cellulose-based hydrogels, as well as their uses in biomedical fields like drug delivery, tissue engineering, and wound healing, healthcare and hygienic products, agriculture, textiles, and industrial applications as smart materials [56].

Fernandez et al. explain that red cabbage is a vegetable known for its enriched bioactive constituents. Generally, among the population, it is used as an ingredient in raw salads or coleslaws, pickle, and boiled and steamed dishes for its impact on human health and low calories and high fiber composition. It is widely used in food production to improve the aesthetic value of food and to provide health benefits as a natural colorant in drinks, candies, and gums. It has many health benefits, including protection against cancer and diabetes, as well as strengthening the immune system, aiding in body detoxification, promoting weight loss, improving skin, reducing inflammation, and relieving constipation. Red cabbage's antioxidant content aids in the prevention of chronic illness and the treatment of conditions such as Alzheimer's and depression. This paper examines the scientific approach to red cabbage as well as its pharmacological function [57].

## 8. Nanomaterial Food Packaging

From better packaging material with improved mechanical strength, barrier properties, and antimicrobial films to nanosensing for pathogen detection and alerting consumers to the safety status of food, nano-based "smart" and "active" food packaging offer several advantages over traditional packaging methods.

Nanoparticles are not just employed in antimicrobial food packaging; nanocomposite and nanolaminates have also been used in food packaging to create a barrier against high temperature and mechanical shock, hence prolonging food shelf life. Incorporating nanoparticles into packaging materials provides high-quality food with a longer shelf life. Polymer composites were developed to provide more mechanical and thermostable packing materials. In order to improve polymer composites, a variety of inorganic and organic fillers are used.

## 9. Conclusion

The active and intelligent packaging materials develop in the search of environment-friendly packaging solutions. In recent years, extensive research into the creation of novel active packaging technologies with natural pigments has resulted in a wide range of active packaging systems that can be used to increase the shelf life of food products.

## Data Availability

The data used to support the finding of this study are included within the article.

## Conflicts of Interest

The authors have no conflicts of interest to declare.

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## References

[1] E. M. Ahmed, "Hydrogel: preparation, characterization, and applications: a review," *Journal of Advanced Research*, vol. 6, no. 2, pp. 105–121, 2015.

[2] W. Abobatta, "Impact of hydrogel polymer in agricultural sector," *Adv. Agric. Environ. Sci. Open Access*, vol. 1, no. 2, pp. 59–64, 2018.

[3] S. Mantha, S. Pillai, P. Khayambashi, A. Upadhyay, and Y. Zhang, "Smart hydrogels in tissue engineering and regenerative medicine," *Materials*, vol. 12, no. 3323, p. 33, 2019.

[4] S. Thakur, P. P. Govender, M. A. Mamo, S. Tamulevicius, Y. K. Mishra, and V. K. Thakur, "Progress in lignin hydrogels and nanocomposites for water purification: future perspectives," *Vacuum*, vol. 146, pp. 342–355, 2017.

[5] S. Bashir, M. Hina, J. Iqbal et al., "Fundamental concepts of hydrogels: synthesis, properties, and their applications," *Polymers*, vol. 12, no. 11, p. 2702, 2020.

[6] D. Rico-García, L. Ruiz-Rubio, L. Pérez-Alvarez, S. L. Hernández-Olmos, G. L. Guerrero-Ramírez, and J. L. Vilas-Vilela, "Lignin-based hydrogels: synthesis and applications," *Polymers (Basel).*, vol. 12, no. 81, pp. 1–23, 2020.

[7] T. Vo, T. Dang, and B. Chen, "Synthesis of Intelligent pH Indicative Films from Chitosan/Poly(vinyl alcohol)/anthocyanin extracted from red cabbage," *Polymers*, vol. 11, no. 7, p. 1088, 2019.

[8] K. Halász and L. Csóka, "Black chokeberry (Aronia melanocarpa) pomace extract immobilized in chitosan for colorimetric pH indicator film application," *Food Packaging and Shelf Life*, vol. 16, pp. 185–193, 2018.

[9] P. Oikonomou, M. Sanopoulou, and K. G. Papadokostaki, "Blends of poly(vinyl alcohol) and poly(vinyl pyrrolidone): interrelation between the degree of hydration and thermal and mechanical properties," *Industrial & Engineering Chemistry Research*, vol. 60, no. 39, pp. 14203–14212, 2021.

[10] M. Teodorescu, M. Bercea, and S. Morariu, "Biomaterials of poly(vinyl alcohol) and natural polymers," *Polymer Reviews*, vol. 58, no. 2, pp. 247–287, 2018.

[11] S. Pourjavaher, H. Almasi, S. Meshkini, S. Pirsa, and E. Parandi, "Development of a colorimetric pH indicator based on bacterial cellulose nanofibers and red cabbage (Brassica oleraceae) extract," *Carbohydras*, vol. 156, pp. 193–201, 2017.

[12] C. M. Yoshida, V. B. V. Maciel, M. E. D. Mendonça, and T. T. Franco, "Chitosan biobased and intelligent films: Monitoring

pH variations," *LWT - Food Science and Technology*, vol. 55, no. 1, pp. 83–89, 2014.

[13] J. Schoeller, F. Itel, K. Wuertz-Kozak, G. Fortunato, and R. M. Rossi, "pH-responsive electrospun nanofibers and their applications," *Polymer Reviews*, pp. 1–49, 2021.

[14] F. Reyes-Ortega, "pH-responsive polymers: properties, synthesis and applications," in *Smart Polymers and Their Applications*, pp. 45–92, Woodhead Publishing, 2014.

[15] G. Kocak, "pH-responsive polymers," *Polymer Chemistry*, vol. 8, no. 1, pp. 144–176, 2017.

[16] K. A. Brun-Graeppi, A. Silva, C. Richard, M. Bessodes, D. Scherman, and O.-W. Merten, "Cell microcarriers and microcapsules of stimuli-responsive polymers," *Journal of Controlled Release*, vol. 149, no. 3, pp. 209–224, 2011.

[17] A. Bahrami, R. Delshadi, E. Assadpour, S. M. Jafari, and L. Williams, "Antimicrobial-loaded nanocarriers for food packaging applications," *Advances in Colloid and Interface Science*, vol. 278, p. 102140, 2020.

[18] Z. Pilevar, A. Bahrami, S. Beikzadeh, H. Hosseini, and S. M. Jafari, "Migration of styrene monomer from polystyrene packaging materials into foods: characterization and safety evaluation," *Trends in Food Science & Technology*, vol. 91, pp. 248–261, 2019.

[19] N. D. Steenis, E. van Herpen, I. A. van der Lans, T. N. Ligthart, and H. C. van Trijp, "Consumer response to packaging design: the role of packaging materials and graphics in sustainability perceptions and product evaluations," *Journal of Cleaner Production*, vol. 162, pp. 286–298, 2017.

[20] E. Mohammadian, M. Alizadeh-Sani, and S. M. Jafari, "Smart monitoring of gas/temperature changes within food packaging based on natural colorants," *Comprehensive Reviews in Food Science and Food Safety*, vol. 19, no. 6, pp. 2885–2931, 2020.

[21] M. Alizadeh-Sani, E. Mohammadian, J. W. Rhim, and S. M. Jafari, "pH-sensitive (halochromic) smart packaging films based on natural food colorants for the monitoring of food quality and safety," *Trends in Food Science & Technology*, vol. 105, pp. 93–144, 2020.

[22] J. W. Rhim, H. M. Park, and C. S. Ha, "Bio-nanocomposites for food packaging applications," *Progress in Polymer Science*, vol. 38, no. 10-11, pp. 1629–1652, 2013.

[23] F. E. Tirtashi, M. Moradi, H. Tajik, M. Forough, P. Ezati, and B. Kuswandi, "Cellulose/chitosan pH-responsive indicator incorporated with carrot anthocyanins for intelligent food packaging," *International Journal of Biological Macromolecules*, vol. 136, pp. 920–926, 2019.

[24] E. Balbinot-Alfaro, D. V. Craveiro, K. O. Lima, H. L. G. Costa, D. R. Lopes, and C. Prentice, "Intelligent packaging with pH indicator potential," *Food Engineering Reviews*, vol. 11, no. 4, pp. 235–244, 2019.

[25] F. Delgado-Vargas, A. R. Jimenez, and O. Paredes-Lopez, "Natural pigments: carotenoids, anthocyanins, and betalains—characteristics, biosynthesis, processing, and stability," *Critical Reviews in Food Science and Nutrition*, vol. 40, no. 3, pp. 173–289, 2000.

[26] H. Paliwal, S. Goyal, S. Singla, and S. Daksh, "Pigments from natural sources: an overview," *International Journal of Research in Pharmacy and Pharmaceutical Sciences (IJRPPS)*, vol. 1, no. 3, pp. 1–12, 2016.

[27] M. Ahmad and B. P. Panda, "Optimization of red pigment production by Monascus purpureus MTCC 369 under solid-state fermentation using response surface methodology," *Songklanakarin Journal of Science and Technology*, vol. 36, no. 4, pp. 439–444, 2014.

[28] L. Prietto, T. C. Mirapalhete, V. Z. Pinto et al., "pH-sensitive films containing anthocyanins extracted from black bean seed coat and red cabbage," *LWT- Food Science and Technology*, vol. 80, pp. 492–500, 2017.

[29] B. Shankaranarayanan and E. Nakkeeran, "Purification of anthocyanins from red cabbage using semi interpenetrating network hydrogel beads in a packed bed column," *Separation Science and Technology*, vol. 54, no. 5, pp. 675–682, 2019.

[30] V. V. Carvalho, J. O. Gonçalves, A. Silva, T. R. Cadaval Jr., L. A. Pinto, and T. J. Lopes, "Separation of anthocyanins extracted from red cabbage by adsorption onto chitosan films," *International Journal of Biological Macromolecules*, vol. 131, pp. 905–911, 2019.

[31] H. Berland, N. W. Albert, A. Stavland et al., "Auronidins are a previously unreported class of flavonoid pigments that challenges when anthocyanin biosynthesis evolved in plants," *Proceedings of the National Academy of Sciences*, vol. 116, no. 40, pp. 20232–20239, 2019.

[32] N. Mahmad, R. M. Taha, R. Othman, S. Abdullah, N. Anuar, and H. Elias, "Anthocyanin as potential source for antimicrobial activity inClitoria ternateaL. andDioscorea alataL," *Pigment & Resin Technology*, vol. 47, no. 6, pp. 490–495, 2018.

[33] M. I. Aksu, E. Turan, I. G. Sat, E. Erdemir, F. Oz, and M. Gürses, "Improvement of quality properties of cemen paste of pastirma by lyophilized red cabbage water extract," *Journal of Food Processing and Preservation*, vol. 44, no. 9, pp. 1–10, 2020.

[34] H. Yong and J. Liu, "Active packaging films and edible coatings based on polyphenol-rich propolis extract: a review," *Comprehensive Reviews in Food Science and Food Safety*, vol. 20, no. 2, pp. 2106–2145, 2021.

[35] S. Fang, F. Lin, D. Qu, X. Liang, and L. Wang, "Characterization of purified red cabbage anthocyanins: improvement in HPLC separation and protective effect against $H_2O_2$-induced oxidative stress in HepG2 cells," *Molecules*, vol. 24, no. 1, p. 124, 2019.

[36] M. Nowacka, M. Dadan, M. Janowicz et al., "Effect of non-thermal treatments on selected natural food pigments and color changes in plant material," *Comprehensive Reviews in Food Science and Food Safety*, vol. 20, no. 5, pp. 5097–5144, 2021.

[37] A. Demirbas, *"Red Cabbage Extract as a Natural Antioxidant with Application to Packaged Fresh Tilapia and Enhancement of Silver and Iron Nanoparticles," Doctoral dissertation*, University of Florida, 2016.

[38] J. Kapusta-Duch and B. Kusznierewicz, "Young shoots of white and red headed cabbages like novel sources of glucosinolates as well as antioxidative substances," *Antioxidants*, vol. 10, no. 8, p. 1277, 2021.

[39] N. Ahmadiani, R. J. Robbins, T. M. Collins, and M. M. Giusti, "Anthocyanins contents, profiles, and color characteristics of red cabbage extracts from different cultivars and maturity stages," *Journal of Agricultural and Food Chemistry*, vol. 62, no. 30, pp. 7524–7531, 2014.

[40] W. Wiczkowski, D. Szawara-Nowak, and J. Topolska, "Red cabbage anthocyanins: profile, isolation, identification, and

antioxidant activity," *Food Research International*, vol. 51, no. 1, pp. 303–309, 2013.

[41] H. E. Khoo, A. Azlan, S. T. Tang, and S. M. Lim, "Anthocyanidins and anthocyanins: colored pigments as food, pharmaceutical ingredients, and the potential health benefits," *Food & Nutrition Research*, vol. 61, no. 1, p. 1361779, 2017.

[42] N. B. Stebbins, *"Characterization and Mechanisms of Anthocyanin Degradation and Stabilization," ProQuest Diss. Theses*, University of Arkansas, 2017.

[43] P. Brudzyńska, A. Sionkowska, and M. Grisel, "Plant-derived colorants for food, cosmetic and textile industries: a review," *Materials*, vol. 14, no. 13, p. 3484, 2021.

[44] X. Qi, W. Wei, J. Shen, and W. Dong, "Salecan polysaccharide-based hydrogels and their applications: a review," *Journal of Materials Chemistry B*, vol. 7, no. 16, pp. 2577–2587, 2019.

[45] L. S. Casey, "Investigation of chitosan-PVA composite films and their adsorption properties," *Journal of Geoscience and Environment Protection*, vol. 3, no. 2, pp. 78–84, 2015.

[46] S. Karki, H. Kim, S. J. Na, D. Shin, K. Jo, and J. Lee, "Thin films as an emerging platform for drug delivery," *Asian Journal of Pharmaceutical Sciences*, vol. 11, no. 5, pp. 559–574, 2016.

[47] J. H. Song, R. J. Murphy, R. Narayan, and G. B. H. Davies, "Biodegradable and compostable alternatives to conventional plastics," *Philosophical Transactions of the Royal Society, B: Biological Sciences*, vol. 364, no. 1526, pp. 2127–2139, 2009.

[48] J. Wu, S. Ge, H. Liu et al., "Properties and antimicrobial activity of silver carp ( _Hypophthalmichthys molitrix_ ) skin gelatin-chitosan films incorporated with oregano essential oil for fish preservation," *Food Packaging and Shelf Life*, vol. 2, no. 1, pp. 7–16, 2014.

[49] T. M. Vedove, B. C. Maniglia, and C. C. Tadini, "Production of sustainable smart packaging based on cassava starch and anthocyanin by an extrusion process," *Journal of Food Engineering*, vol. 289, p. 110274, 2021.

[50] P. Zeng, X. Chen, Y. R. Qin et al., "Preparation and characterization of a novel colorimetric indicator film based on gelatin/polyvinyl alcohol incorporating mulberry anthocyanin extracts for monitoring fish freshness," *Food Research International*, vol. 126, p. 108604, 2019.

[51] E. Syafri, E. Yulianti, M. Asrofi et al., "Effect of sonication time on the thermal stability, moisture absorption, and biodegradation of water hyacinth (Eichhornia crassipes) nanocellulose-filled bengkuang (Pachyrhizus erosus) starch biocomposites," *Journal of Materials Research and Technology*, vol. 8, no. 6, pp. 6223–6231, 2019.

[52] H. Haghighi, F. Licciardello, P. Fava, H. W. Siesler, and A. Pulvirenti, "Recent advances on chitosan-based films for sustainable food packaging applications," *Food Packaging and Shelf Life*, vol. 26, p. 100551, 2020.

[53] T. Wang, M. Turhan, and S. Gunasekaran, "Selected properties of pH-sensitive, biodegradable chitosan–poly(vinyl alcohol) hydrogel," *Polymer International*, vol. 53, no. 7, pp. 911–918, 2004.

[54] V. A. Pereira, I. N. Q. de Arruda, and R. Stefani, "Active chitosan/PVA films with anthocyanins from Brassica oleraceae (red cabbage) as time-temperature indicators for application in intelligent food packaging," *Food Hydrocolloids*, vol. 43, pp. 180–188, 2015.

[55] L. A. Castillo, S. Farenzena, E. Pintos et al., "Active films based on thermoplastic corn starch and chitosan oligomer for food packaging applications," *Food Packaging and Shelf Life*, vol. 14, pp. 128–136, 2017.

[56] S. M. F. Kabir, P. P. Sikdar, B. Haque, M. A. R. Bhuiyan, A. Ali, and M. N. Islam, "Cellulose-based hydrogel materials: chemistry, properties and their prospective applications," *Progress in Biomaterials*, vol. 7, no. 3, pp. 153–174, 2018.

[57] D. L. Fernandez, K. Hedge, and A. R. Shabaraya, "Scientific approaches on red cabbage: a review," *International Journal of Pharma And Chemical Research*, vol. 5, no. 3, pp. 123–130m, 2019.