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Original article

Active Chitosan Intelligent pH Indicative Films with Anthocyanin from Red Cabbage Extract Against Food Spoilage

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Abstract:

Biomaterial based active membrane development and indicators are recently attracting the researchers and industries to make more ecofriendly approach. The most intriguing area of present study was smart packaging made from biomaterials using red cabbage extract formulated with anthocyanin and chitosan and PVA. The anthocyanin molecules from red cabbage used as functional additives in the current study were used to create pH-sensitive films. Chitosan/Poly (Vinyl Alcohol) blend polymer casting was used to create the films, with two filler loadings (5wt% and 10wt%). Total anthocyanin concentration of 298 ± 1.8 mg/L. was recorded from the extract. According to a pH sensitivity analysis, the smart films' ability to change color by adjusting pH levels from pink at pH levels below 6 to yellow at pH levels above 6 was discovered. FTIR spectral results revealed the presence of anthocyanin, by the indication of oscillations in the C-O-C group which are responsible for the bands from 1030.6 cm^{-1} to 1056.7 cm^{-1}. SEM image of the biofilm indicates the surface morphology before and after anthocyanin absorption by the film. To monitor food spoilage and biomedical applications these pH sensitive biofilms can be used in future.

Keywords: *Poly(vinyl alcohol); chitosan; anthocyanin; pH indicative film; food wrap;*
Food Spoilage

1. Introduction

The food industry is a significant industry that generates billions of dollars annually in the global market. The aim of producing biocompatible, biodegradable, and affordable film-forming materials for biomedical applications has recently increased due to rising environmental awareness and a focus on eco-friendly products. To prevent food contamination and perishing from package to market delivery, the food processing and packaging sectors suffer a significant setback [1]. Food quality and safety must be continuously assessed to sustain the public health issues. Antimicrobial sensitive food packaging is very essential due to increased food spoilage and contamination, microbial risk hazards and adulterations. Antimicrobial based biofilm coatings are much welcomed from the consumers in recent days to ensure that they are preservative free products and freshly packed [2,3,4]. Modern bio-based packaging is now being widely developed around the globe to improve package efficiency and reduce environmental and health hazards [5,6]. To lessen the health concerns posed by petroleum-based polymer residues that migrate to packaged foods, numerous researchers have attempted to develop novel edible food packaging based on different types of natural biopolymers [7-9]. The development of smart packaging techniques, which may be divided into two main categories: active and intelligent packaging is primarily responsible for the innovation in food packaging over the past 20 years. To extend the shelf life of food goods, active packaging systems are created by adding functional bioactive ingredients such as natural plant extracts, essential oils, antimicrobials, etc. [10–12].

Red cabbage (*Brassica oleraceae*) is one of the sources of anthocyanins for coloration of food since its anthocyanins are unique, exhibiting color over a very broad pH range. Red cabbage is naturally abundant and the anthocyanin preparation cost from red cabbage is low in comparison to other materials. The colors of anthocyanins extracted from red cabbage vary from red at low pH to blue and green at high pH [13]. Thus, this broad color change makes it attractive for application as natural pH indicators. The solid support is the second part of an intelligent packaging that has an important role in sensitivity (responsibility to lower amounts of pH change inducing compound), response time (time gap between the pH change and appearance of distinctive color change), reproducibility and reversibility (retrieval of color by removing of pH change inducing compound) of fabricated pH indicator.

Chitosan is a naturally occurring polysaccharide that is created when chitin undergoes an alkaline deacylation process, which results in the partial removal of acetyl groups from acetylamino groups and their replacement by amino groups. In addition to having antibacterial capabilities, chitosan also contains antifungal, mucoadhesive, analgesic, and hemostatic ones [14]. Biopolymer called chitosan (CS) is non-toxic and possesses antibacterial qualities, which are useful for food packaging. Due to their biodegradability, low cost, and wide availability, biodegradable materials made from natural sources are currently thought of as viable replacements for traditional plastic materials. Therefore, to create intelligent films, biodegradable polymers are typically used, together with a pH indicator [14–16]. A synthetic semi-crystalline polymer known as polyvinyl alcohol (PVA) has been employed in biotechnology for tissue regeneration, bandages for wounds, and medication delivery systems. Excellent qualities of PVA-based dressings include their biodegradability, biocompatibility, lack of toxicity, and low cost [7].

Chitosan films has low mechanical resistance despite its benefits, hence it is frequently mixed with other polymers like Poly vinyl alcohol (PVA), a non-toxic and biodegradable synthetic polymer, to enhance its mechanical properties [7,8]. However, the low mechanical strength of chitosan necessitates the need for water-soluble, non-toxic polymers such as cellulose derivatives poly ethylene oxide (PEO) and poly vinyl alcohol (PVA) to be blended with. PVA, which is harmless and biocompatible, is referred to as a green material. PVA is water soluble, biocompatible, chemically stable, and environmentally friendly. The polymeric chain of this substance has multiple accessible hydroxyl groups (OH), and it has been the subject of in-depth research in a number of fields. PVA-based hydrogels are thus employed in the pharmaceutical sector for controlled drug delivery [5,6,8,10,14,15,17-21]. Because of this, the suggestive film made from PVA and CS and using ATH as a color indicator could exhibit positive spectroscopic and physicochemical properties and be used in intelligent food packaging [22].

As noted earlier, PVA/CS composites were selected as the model polymers for this investigation of pH Indicative thin-film wraps. In recent years there is a lot of research into the possible uses of PVA/CS hydrogels, including in biomedical applications and biomaterials [23,24]. To ascertain the chemical interactions between the anthocyanin and the polymer components as well, the pH-indicative films were studied by FT-IR and scanning electron microscopy (SEM). Electronic and synthetic based bioindicators can't be accessed easily in terms of its high cost and affordable to all. Natural Biological agent based bioindicators like biofilms and pH indicators are ecofriendly rather than the synthetic modules. Moreover the natural bioindicator agents are more feasible than the artificial which makes the bioindicator based discovery more intriguing in recent years. As a part of the bioindicator development the present study is aimed to evaluate the effects of anthocyanins on the physicochemical properties and biological activities of chitosan/polyvinyl alcohol films, as well as the practical indication effect as food preservative.

2. Materials and Methods

2.1. Material preparation

The following ingredients were bought from Sigma-Aldrich: Chitosan (CS, 75%-85% deacetylated), Poly (vinyl alcohol) (PVA, Mw= 89,000-98,000, Hydrolysis degree: 86.5 to 89%). Hydrochloric acid (37%), Sodium hydroxide (NaOH), Sulfuric acid (H_2SO_4) and glacial acetic acid were supplied by Merck. Red cabbages were purchased fresh from local market. Chemicals were used exactly as received and were all of the analytical quality. In this investigation, deionized water with a resistivity greater than 18.2 MW was used to prepare the samples [40]. All the chemicals used in the present were of analytical grade.

2.2. Preparation of Red Cabbage Extract

Red cabbage (*Brassica oleracea*), obtained from the local market was used to extract the anthocyanin (ATH) for this study, primarily using the solvent extraction technique described by Fuleki and Francis [25]. The extraction method for ATH from red cabbage was slightly modified as follows. Typically, 300 mL of solvents consisting of concentrated HCL (3 mL) in 150 mL of deionized water are added to 150 g of chopped red cabbage and allowed to soak for 60 minutes in an ultrasonic bath [10,18,26]. After extraction, the anthocyanin-rich solution was automatically collected from the solid–liquid mixture through filtration of the nylon bag and collected into a sample bottle for further analysis.

2.3. Characterization of Red Cabbage Extract

2.3.1. Anthocyanin Concentration

A 200µl aliquot of anthocyanin extract was mixed with 7 ml of a pH 1.0 buffer (0.025 M potassium chloride adjusted with hydrochloric acid) and 7 ml of a pH 4.5 buffer (sodium acetate of 0.4, adjusted with w/w acetic acid).) The anthocyanin material is proportional to the difference in absorbance between the two buffers at 530 nm (the highest absorption wavelength) [34]. On a UV-Vis plate reader, measurements were taken using distilled water as a blank. The absorbance at

700 nm was used to make adjustments to account for the presence of broken compounds or interference-causing chemicals. Since cyanidin glucoside is the source of red cabbage anthocyanins, concentrations were determined using the following equation and expressed as mg cyanidin-3-glucoside per 100 ml extract:

$$\text{Anthocyanin concentration} = \frac{\Delta A.\ \text{MW average}\ .\text{FD}.1000}{\varepsilon.\ \ L}$$

112ΔA = (A 530-A700) pH:1,0-(A530-A700) pH:4.5

where MW normal is the atomic mass of cyanidin-3-glucoside (449.2 g/Mol); FD is the cyanidin-3-glucoside molar absorbance; and A is the difference between the absorbance change at 530 and 700 nm in buffers with pH 1 and 4.5. (26,900) L is the optical path (in cm), and 1000 is the conversion of grams to milliequivalent, multiplied by three.

2.3.2. Confirmatory Anthocyanin Test.

The presence of anthocyanin in red cabbage extract was confirmed by adding 1 mL of sulfuric acid (H_2SO_4) to a test tube containing 2 mL of red cabbage extract and retaining it for a few minutes. Appearance of orange color at the interface indicates the presence of anthocyanin in the sample. 2 drops of 1 N NaOH were added to 2 mL of extract. The presence of anthocyanin would be indicated by blue to bluish-green coloration.

2.4. Preparation of PVA and chitosan film-forming solutions

For the preparation of the PVA film-forming solution, a 50g/L PVA (poly Vinyl alcohol) aqueous solution (100 mL) was stirred at 70-80°C until the solution became transparent. Chitosan film-formation solution was prepared to dissolve by dissolving 1 gram of chitosan (Sigma Aldrich Art No.448877, 80% deacetylation degree) in 100 mL of aqueous acetic acid solution (1 mL/100 mL), under stirring during 24 h at room temperature (25°C).

2.5. Preparation of pH indicator film.

The final Chitosan/PVA/Extract film (TPP hydrogel) was created using the casting procedure with a 3:2 (mL: mL) Chitosan/PVA film-forming solution ratio. 5 mL/100 mL of the hydrogel mixture served as the final extract concentration in the film. To encourage cross-linking between Chitosan and PVA, 1 mL of a Thymine pyrophosphate (TPP) solution containing 10 g/L was added to the finished hydrogel. The hydrogel was cast (50 ml) in Petri plates (60 mm in diameter), and the resulting pH indicator films were dried by the solvent in an oven for 96 hours at 35°C.

2.6. FT-IR spectroscopy analysis

FT-IR spectra were recorded using the attenuated reflectance (ATR) Fourier transform infrared spectroscopy on a PerkinElmer Spectrum 100 spectrophotometer with a resolution of 4 cm^{-1} and operating in the 400-4000 cm^{-1} range.

2.7. Scanning electron microscopy (SEM)

The morphologies and pore structures of the hydrogel were observed by Hitachi S-4700 SEM at an accelerating voltage of 20 kV at the magnification of 50X. Before the observation, the specimen was immersed in liquid nitrogen, fractured, and then sputter coated with a thin layer of gold.

2.8. The coloration of pH Indicative Films

In buffer solutions with pH values ranging from 1 to 12, the color of the pH indicator films was displayed. The films were divided into 2 x 2 cm^2 square coupons and dipped into buffer solutions. Typically, the color shift might be seen and documented within 30 seconds.

3. Results

3.1. Characterization of Anthocyanin

Here, a system for evaluating food freshness is created by combining back propagation (BP) neural network technology with red cabbage anthocyanin markers. Red cabbage anthocyanins were used as confirmatory test for the presence of color response pigments, and chitosan/PVA was used as the solid matrix. Silva and his team (2015) reported about the use of anthocyanins from red cabbage for pH monitoring [23]. A pH indicator film made of chitosan/corn starch and red cabbage extract was created as a visual indicator of fish deterioration. A PVA/Chitosan polymeric doped with anthocyanins derived from red cabbage was developed and studied. The Total anthocyanin concentration of 298 ± 1.8mg/L was recorded. The creation of the reddish-orange color of the solution upon the addition of H_2SO_4 (Figure 1), along with the dark-bluish solution noticed upon the addition of 1 N NaOH, which swiftly changed to a reddish-orange color, provided chemical confirmation of the presence of anthocyanin (Figure 2). Confirmatory assays for anthocyanins employ acidic and basic solutions since anthocyanin types depend on pH. Anthocyanins go through molecular changes when they interact with acids and bases. The most prevalent sort of ions in an acidic medium is flavylium ions, which give off a red tint.

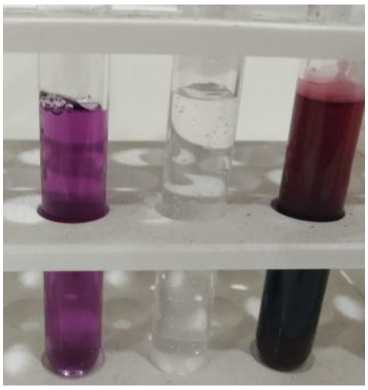

Figure 1: Sulfuric acid induced colour change

Figure 2: NaOH Test induced colour change

3.2. Synthesis and chemical characteristics of pH indicative films:

Chitosan has been extensively studied and used because of its extraordinary natural and advantageous qualities. To create a semi-interpenetrating polymeric structure that was cross-linked with glutaraldehyde, chitosan and poly vinyl alcohol (PVA) were used [40]. The Chitosan deacetylation level was 72%, and its atomic weight was 612 kDa [28]. The red cabbage was successfully used to extract the anthocyanin (ATH). Figure.3 demonstrated the color shift in buffer solutions with an ATH content of 86.67 mg/L. The fact that anthocyanins change color visibly when exposed to different pH levels is one benefit linked with their usage as pH indicators. For instance, they appear red or pink in acidic solutions and blue or green in alkaline ones. As a result, the ATH was developed to cast pH-indicative films made of PVA and CS.

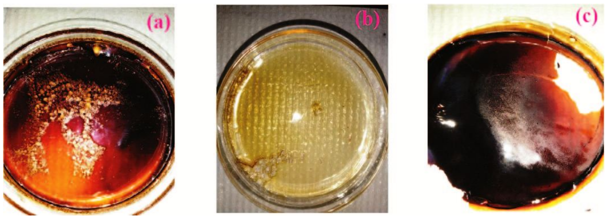

Figure 3. Color change of anthocyanin extract from left to right.

The nature and color change of the chitosan/PVA/TPP/Red cabbage extract gradually changed after each chemical entity addition. The intensity and type of the color of anthocyanins are dependent on the presence of hydroxyl and methoxyl groups. The presence of many hydroxyl groups changes the color to a bluish tint, while the presence of more methoxyl groups indicates an increase in redness [29]. Figure. 4 (a-c) shows the perspective color change after the addition of each constituent from chitosan to the red cabbage extract. Interestingly, the color of the resultant film changed from brown to pale dark red finally.

Figure.4: (a) Red Cabbage extract film layer, (b) Formation of PVA/TPP/Red cabbage extract. (c) Biofilm of composite chitosan/PVA/TPP/Red cabbage extract.

3.3. FT-IR spectroscopy analysis:

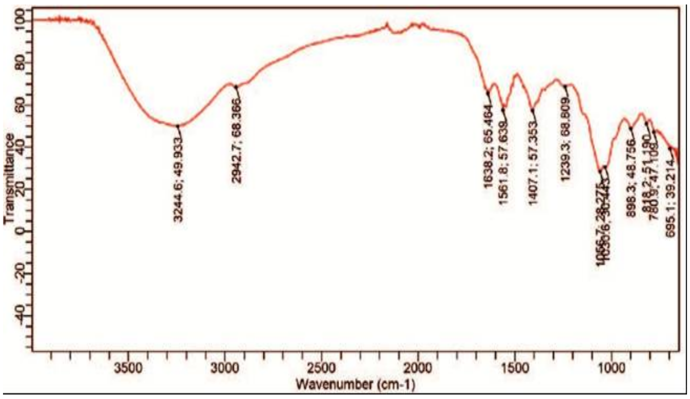

Figure.5: Red Cabbage extract film layer, chitosan/TPP film formation.

Figure.5 displays the FT-IR spectra of the chitosan/PVA/TPP/film. The hydrogel containing ATH was used to create the test film, with a PVA/CS volume ratio of 35/65. Peaks at 3244.6 cm1 and 1407.1 cm^{-1} are attributed to PVA's FT-IR spectra. The bands at 1407.1 cm^{-1} and 2942.7 cm^{-1} are the result of -CH2 stretching vibrations, whereas the -OH group's stretching and bending vibrations are responsible for the bands at 1407.1 cm^{-1} and 2942.7 cm^{-1}. Anthocyanin that have crystallized in the polymer are what cause the peak to appear at 1638.2 cm^{-1}. The FT-IR spectrum for the anthocyanin extract shows a strong absorption band with a maximum at 1015 cm^{-1} assigned to aromatic ring C-H deformation, as well as bands at 1650 and 1455 cm^{-1} corresponding to the stretching vibration of the C=C aromatic ring. The absorption band with a maximum at 1233 cm^{-1} is assigned to stretching of pyran rings, typical of flavonoid compounds. Bands appearing between 1300 cm^{-1} and 1380 cm^{-1} are assigned to C=O angular deformations of phenols.

In bands ranging from 898.3 to 1056.7 cm-1, the oscillations of the C-O-H group may be seen [12,27]. For pure chitosan (CS), the absorption at 3244.6 cm-1 relates to the stretched -OH group, while the band at 2942.7 cm-1 is a hallmark of the -CH_2 group bound to the -OH group. The peak at 1638.2 cm-1 is due to the C-O stretching of the acetyl group (amide I). Additionally, the peak at 1407.1 cm-1 is caused by the bending vibration of the -C-OH group, whereas the peak at 1239.3 cm-1 is caused by the knife in the plane of the -OH group [18,27]. The C=C aromatic ring is represented by the peak at 2942.7 cm-1 in the FT-IR spectra of anthocyanin, while oscillations in the C-O-C group are responsible for the bands from 1030.6 to 1056.7 cm-1 [18]. The peak for the -OH group is 3244.6 cm-1. As shown in Figure.3, the FT-IR spectra of the PVA/CS/ATH pH-indicating film contain the PVA, chitosan, and anthocyanin characteristics.

3.4. Scanning electron microscopy (SEM)

SEM was used to examine the polymer film's surface morphology. The SEM images of chitosan film before and after anthocyanin pigment adsorption was shown in (Figure.6). The PVA/CS film's surface was remarkably fibrous (Figure.6). At 60,000 times magnification, no crystal grains could be seen, indicating that the membrane's constituent parts were thoroughly mixed and formed as gel matrix which is a prominent feature for the biofilm membrane development. In Figure.6 PVA/CS/ATH demonstrate that the film surface was coated and the film roughness was decreased. This is because the anthocyanin molecules that have accumulated on the material's surface are responsible for this.

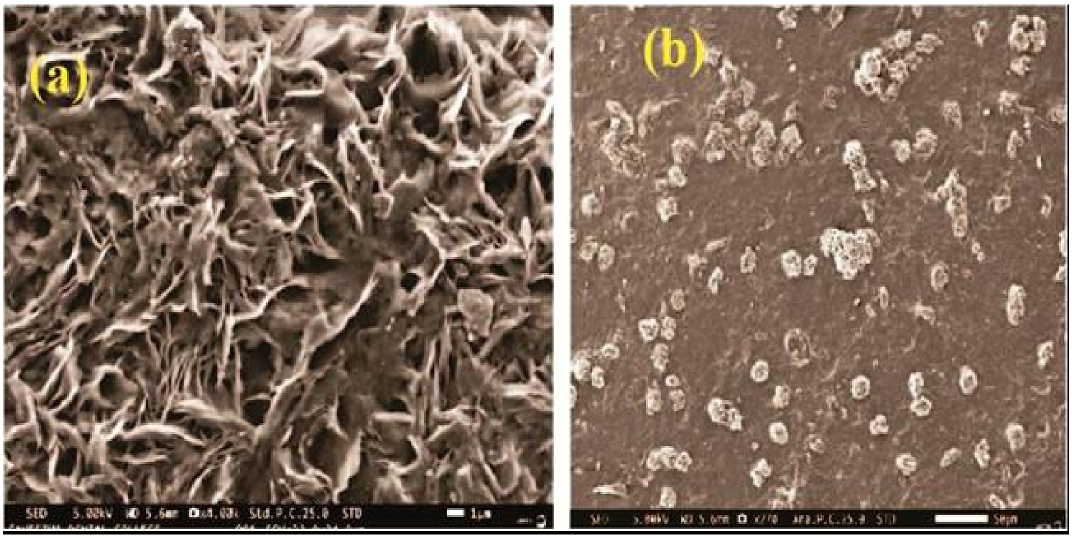

Figure 6. SEM micrograph of PVA/CS/TPP film synthesized from the ATH-containing hydrogel. a) PVA/CS without TPP. b) PVA/CS with TPP.

3.5. Plausible Application of PVA/CS/ATH pH Indicative Film for Food Packaging

The PVA/CS/ATH prepared biofilm may be applied as bioindicator after rigorous testing and analysis in future perspect since the amount of *PVA/CS/ATH* obtained in the present study is meagre which cannot be subjected for qualitative and quantitative assays. Intelligent packaging films are usually composed of matrix material and a pH indicator. Due to their biodegradability, non-toxicity and biological properties, many natural polymers, such as chitosan, starch, clay, pectin and other biopolymers have been used in food packaging [30-32]. Despite its ability to indicate food decomposition, the chitosan-PVA film has limitations in its application for dry food types of products. The film must directly contact the food surface and have enough water in it to penetrate the polymer film. Furthermore, to protect the anthocyanin extract from being harmed by high temperatures, this film should be put on a product that is kept in a chiller or freezer. In contrast, PVA appears to filter out in the acidic medium over prolonged swelling times as a result of the hydrolysis of the gel structures, according to Schiff's theory. According to Alizadeh-Sani et al., [33], advances in packaging creation were brought forth by the underutilized growth in nature with secure nutrition and shifting consumer attitudes. Because of the fascination with better things and their cancer prevention specialist capabilities, anthocyanin shades are appropriate as standard colors for food, cosmetics, and dietary changes [31,32]. Using an adsorption process on chitosan films, this work involved extracting the anthocyanin hues from red cabbage and its portion of the process. Reasonable information regarding the anthocyanin molecule's immobilization onto chitosan films was presented in this study, and the results may have a significant impact on how these colors are used in combination with chitosan in many areas. Therefore, as compared to films made solely from chitosan or PVA, the films produced by combining the two materials have different physical properties. Additionally, in order to enhance properties like antibacterial activity, chitosan blend films can be combined with additional materials like natural extracts or inorganic metal particles in future applications.

We describe here the development and characterization of a biofilm sensor based on a PVA and chitosan polymer blend that contains anthocyanins, taking into account that chitosan and PVA can be used safely in food packaging and that natural extracts can be integrated into chitosan/PVA blends. The goal of this biofilm is to determine pH variations indirectly from changes in the pH of food products over time when the food is stored at temperatures other than those advised for storage. The present study provides a summary of earlier and ongoing research on the physiochemical properties of cellulose-based hydrogels, as well as information on how they are used in biomedical fields like drug delivery, tissue engineering and wound healing, healthcare, and clean products, horticulture materials, and mechanical applications as clever materials [34]. A vegetable with enhanced bioactive components is red cabbage. Basic servings of mixed greens serve as a fixer [35]. It is widely used in food production to advance the fashionable value of food and to provide health benefits as a common colorant in beverages, candies, and gums. It has several health advantages, such as protection against cancer and diabetes, as well as strengthening the immune system, aiding in bodily purification, promoting weight loss, improving skin, reducing irritation, and relieving clogging [36] Numerous investigations have shown that PVA and CS have great pH sensitivity. Other common uses for these CS/PVA nanocomposite films include water treatment, medication delivery, and bone tissue engineering [37]. With good thermal and mechanical qualities, CS/PVA transforms into a stable and biocompatible composite. Due to the high electrostatic attraction and hydrogen bonds that exist between the positively charged polysaccharides groups in CS and the negatively charged PVA sheets films made of uniformly dispersed CS/PVA might be produced under the right pH conditions. It is well known that the amino group of CS interacts with the PVA's oxygen functions through hydrogen bonding, improving the material's mechanical characteristics [36-38].

4. Discussion

The results confirm that red cabbage anthocyanins were successfully extracted and incorporated into a PVA/Chitosan biofilm matrix. The chemical confirmation assays validated anthocyanins' pH-sensitive stimuli, while FT-IR analysis showed that intermolecular hydrogen bonding facilitated strong interactions among CS, PVA, and anthocyanins. SEM micrographs confirmed structural homogeneity of the films, indicating successful formation of a stable biopolymeric matrix.

The distinct color changes observed across pH values are consistent with the structural transitions of anthocyanins: Flavylium cations (red–pink) at acidic pH, Quinonoidal base forms (purple–blue) at neutral pH, Chalcone/pseudobase forms (green–yellow) at alkaline pH.

This reversible color change makes the films suitable for intelligent packaging. Previous studies (Silva et al., 2015; Alizadeh-Sani et al., 2018) reported similar systems with starch or other biopolymers; however, blending PVA and CS enhanced both mechanical properties and pH responsiveness.

Nevertheless, the study demonstrates that natural anthocyanins, immobilized in a CS/PVA matrix, can serve as eco-friendly, non-toxic, and biodegradable pH indicators for food packaging and freshness monitoring.

5. Conclusion

In the food supply chain, packaging plays a crucial role in ensuring that food quality is maintained during transportation, storage, and final usage. It shields the product from external factors such oxygen, water, light, pests, and chemicals, and it delays quality deterioration, improving the effectiveness of distribution and marketing. Intelligent packaging is currently the subject of deeper research. However, there are still many obstacles to overcome in the implementation of anthocyanin intelligent film. Future studies on the construction of a precise correlation between indicators and food freshness as well as the improvement of composite membranes' hydrophobicity may broaden the potential applications for anthocyanin intelligent composite films. The present study evaluated the hydrogel film formation of a composite PVA/Chitosan/ATH and its perspective pH changes. By combining 1% PVA solution and 1% CS solution in different volume ratios, doping the mixture with ATH as an indicator, and mixing in STPP as a cross-linker, pH-indicative films of PVA/CS/ATH have been effectively cast from hydrogels. The PVA/CS/ATH pH indicative films can be used for food indicators in packaging and food testing's with further analysis in future works. However, further studies are needed to determine its performance in real foodstuffs. Also, future developments include the study of the label response in various temperatures and the study of extract stability in the label is needed.

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