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Development of Environmentally Friendly and Intelligent Food Packaging Bio-Nanocomposite Films

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ABSTRACT

Researchers are actively exploring biodegradable biocomposite films as environmentally friendly packaging solutions. Increasing consumer demand for a healthy and secure lifestyle led to a serious recent study into the development of intelligent food packaging bio-nanocomposite films aiming not only contribute to sustainability but also possess advanced functionalities through the integration of nanotechnology and intelligent features. This research focuses on the development of active and pH-responsive bio-nanocomposite films by incorporating various concentrations of SPE anthocyanins into the nanoparticle of the CH-PSPS matrix using the solvent-casting method. Thorough examination and characterization of the films revealed a smooth and compact surface, indicative of a uniform distribution of SPE anthocyanins within the matrix as observed through AFM analysis. The inclusion of SPE anthocyanins resulted in a significant increase in antioxidant activity, ranging from 16.37% to 26.44%. Additionally, all films containing SPE anthocyanins exhibited excellent UV barrier properties and demonstrated sensitivity to pH levels within the range of 1 to 10 in buffer solutions. Moreover, the films effectively preserved the freshness of the shrimp during storage. Consequently, these developed films showcase promising potential as active and intelligent packaging materials for food products.

Keywords: bionanocomposite film, chitosan nanoparticle, purple sweet potato starch; anthocyanin.

INTRODUCTION

Food packaging plays an important role in protecting food during the supply chain by shielding it from numerous risks and external causes (Perveen et al., 2023). The use of petroleum-based polymers such as polypropylene, polyester, and ethylene vinyl alcohol has replaced traditional materials with plastic packaging, which has dominated the industry due to its low cost (Pang et al., 2023). Nevertheless, since these materials are not easily recyclable, compostable, or biodegradable, their extensive use promotes ecological problems (Lee & Liew, 2020). Researchers are actively exploring biodegradable biocomposite films as environmentally friendly packaging solutions (Ratna et al., 2023; Lou & Chen, 2023). Biodegradable films are frequently developed by combining hydrocolloids derived from proteins and polysaccharides such as starch, chitin, chitosan (CH), gelatin, cellulose, alginate, pectin, and gum (Fitriani et al., 2023). These films, incorporating natural components, offer eco-friendly alternatives with enhanced functionalities (Baghi et al., 2022).

Starch and chitosan, two widely accessible biopolymers, have recently acquired importance in the fabrication of bio-nanocomposite packaging films. Starch provides intriguing film-forming abilities in addition to its low cost and availability in nature (Matheus et al., 2023), while chitosan offers barrier properties, non-toxicity, and biocompatibility (Al-Hilifi et al., 2023). Several investigations have been conducted on biocomposite films made from chitosan and starch. (Aguirre-Loredo et al., 2023) synthesized thermoplastic biofilms using achira starch, chitosan, and nanoclays, and discovered that sonication time affected the chemical and physicomechanical properties of the films. (Hao et al., 2023) produced biodegradable composite films by combining chitosan and maize starch, noticing better tensile characteristics and water vapor barrier performance with the addition of chitosan. (Yu et al., 2023) fabricated biodegradable films by incorporating chitosan nanoparticles into a mixture of hydroxypropyl methylcellulose and hydroxypropyl starch, noting that the nanoparticle size influenced the films' barrier and mechanical qualities. (Suhartini et al., 2023) developed a biodegradable film out of a plasticizer, nano-chitosan, and avocado seed starch, and the final film displayed remarkable barrier qualities against water vapor.

Increasing consumer demand for a healthy and secure lifestyle led to a serious recent study into the development of smart food packaging aiming at real-time food quality monitoring. This intelligent packaging has the potential to track the inside package conditions and provide information to consumers concerning both food and the environment (Diksha et al., 2023; Kalpana et al., 2019). Active intelligent films can be developed by adding pH-responsive active compounds derived from flowers, fruits, vegetables, and tubers, such as curcumin, chlorophyll, betalains, and anthocyanins (Bhargava et al., 2020). Anthocyanins are active compounds that are often used in the production of active intelligent films due to their strong antibacterial and antioxidant properties as well as their ability to change color with pH (Qin et al., 2019). Anthocyanins are bioactive compounds, water-soluble pigments, and phenolic metabolites that can change the color of materials (red, orange, blue, and purple) by chemical structural changes. Incorporating anthocyanin-rich mayberry extract into a cassava starch matrix raises the TS, functions as a UV-vis light barrier, gives antioxidant characteristics, and can be utilized to evaluate pork freshness (Yun et al., 2019). According to (Wang et al., 2023), CH/chitin nanofiber bio-nanocomposites containing eggplant anthocyanins possess relatively strong physical, antibacterial, and antioxidant capabilities; the color sensitivity of these composites can be used to properly assess pork freshness. Anthocyanins from Jacaranda cuspidifolia were discovered to have strong antibacterial and antioxidant action in CH/polyvinyl alcohol composite films and can be utilized to reliably assess fish freshness (Amaregouda et al., 2022). To improve antibacterial and antioxidant activity, anthocyanin generated from sweet potato was mixed with quercetin-loaded CH nanoparticles; the pH-sensitive features of the resulting material can be used to check shrimp freshness (Dong et al., 2023).

Purple sweet potatoes, which are common in Aceh, Indonesia, contain anthocyanins with high antioxidant activity 59.25% (Husna et al., 2013). Despite the antioxidant benefits and pH-responsiveness of anthocyanin extract, there is a limitation of data on the development of active intelligent films applying CH nanoparticles and anthocyanin extract from purple sweet potatoes. The main objective of this study is to investigate how incorporating PSPE anthocyanin into the chitosan/purple sweet potato starch bio-nanocomposite matrix affects various aspects involving film surface topography, thermal properties, antioxidant activity, color, UV-barrier properties, and shrimp freshness monitoring. This is the first report of active intelligent films made using anthocyanins CH, PSPS, and PSPE anthocyanin. The topographical surface and thermal properties of these films were characterized using atomic force microscopy (AFM) and differential scanning calorimetry (DSC). The antioxidant activity and pH sensitivity of these films were studied. To establish the optical properties of the film, color analysis, and UV-barrier tests were carried out. Monitoring shrimp freshness was used to evaluate the functionality of the composite films.

MATERIAL AND METHODS

Materials

Purple sweet potato starch (PSPS) was purchased from a local market in Banda Aceh, and chitosan powder was purchased from Chimultiguna Co., Ltd. (Indramayu, Indonesia) with a particle size of 200–300 mesh and a molecular mass of 102 kDa. Sigma Aldrich (Darmstadt, Germany) provided the 2,2-diphenyl-1-picrylhydrazyl (DPPH), glycerol, and glacial acetic acid

Film preparation

The film was created using a solvent-casting technique described in a prior publication (Hasan et al., 2022). In the study, four types of films were generated by adding various concentrations of PSPE anthocyanin (0–15 wt%) into the bio-nanocomposite matrix CH/PSPS (60%:40% w/w). The resulting films were designated as CH/PSPS, CH/PSPS-PSPE5%, CH/PSPS-PSPE10%, and CH/PSPS-PSPE15%.

Atomic force microscopy

AFM (Veeco Instruments, Santa Barbara, CA, USA) was used to measure the surface roughness of the film samples. The test was performed on a 1.5 × 1.5 cm section of the sample film, and the film roughness was calculated in terms of the root mean square roughness (Rq) and average roughness (Ra).

Antioxidant activity test

The antioxidant activity of the film was measured in terms of the capacity to capture DPPH radicals. A 0.1-mM DPPH solution was prepared by diluting 1.97-mg DPPH with 50 mL of ethanol and placed in a dark bottle. A mass of 50 mg of each film was incorporated into 10 ml of 96% ethanol and agitated for 1 min. A volume of 4.0 mL of 0.1-mM DPPH was mixed with 1.0 mL of each film solution and incubated in the dark bottle for 30 minutes (Zheng et al., 2023). The absorbance of the resulting solution was measured using a UV–Vis spectrophotometer at 517 nm, the wavelength of maximum absorbance for DPPH. The DPPH radical scavenging activity was calculated as a percentage using Equation 1:

$$Antioxidant\ activity\ (\%) = \frac{A_0 - A_1}{A_0} \times 100 \quad (1)$$

where: A_0 and A_1 denote the absorbances of the control and test films, respectively.

UV-barrier capacity and opacity of films

The UV-barrier capacity of the films was determined according to the method of (Cazon et al., 2019). Films with dimensions of 0.8×4 cm were scanned over a wavelength range of 200 to 800 nm using a UV-vis spectrophotometer (Perkin Elmer Inc., MA, USA). The film opacity was determined from the absorbance measured at a wavelength of 600 nm using Equation 2:

$$Opacity = \frac{A}{x} \quad (2)$$

where: A and x – the absorbance at 600 nm and the thickness of the film, respectively.

Differential scanning calorimetry

Thermal properties of the films were investigated through Differential Scanning Calorimetry (DSC) analysis using a PT 1600 simultaneous thermogravimetric analyzer (Linseis Inc., USA). The samples were loaded into an aluminum pan and subjected to a heating rate of 10 °C/min under a nitrogen atmosphere (20 mL/min), ranging from 30°C to 600°C

Color analysis

The color of the films was analyzed in terms of L, a, and b values using a CIE colorimeter (Hunter Associates Laboratory, Inc., USA). The difference in the full color (ΔE) of each film was calculated using the formula given below (Equation 3):

$$\Delta E = \sqrt{(L^* - L)^2 + (a^* - a)^2 + (b^* - b)^2} \quad (3)$$

where: L^*, a^*, and b^* are the color parameters of a background white plate and have values of 93.49, −0.25, and −0.09, respectively (Agunos et al., 2020).

Characterization of the pH-sensitivity of the CH/PSPS-PSPE films

The pH sensitivity of the films was determined by immersion in buffer solutions with pHs of 1–10. A CH/PSPS film with dimensions of 20×20 mm was prepared and immersed for 5 min in buffer solutions with pHs of 1–10. The film sample was removed from the solution, and the color parameters (L, a, b, and ΔE) were determined using Equation 3.

Application of CH/PSPS-PSPE films for monitoring shrimp freshness

A package containing fresh shrimp was covered with a CH/PSPS-PSPEs film (7×7 cm) to monitor the shrimp's freshness. The package was stored at room temperature and analyzed every two days for six days. The analysis consisted of measuring the color parameters (L, a, b, and ΔE) and pH.

RESULTS AND DISCUSSION

Atomic force microscopy

AFM is a standard method used to investigate the surface characteristics of different materials and was used to analyze the surface roughness of the films fabricated in this study. Figure 1. shows three-dimensional AFM images of the investigated films, and the corresponding surface roughness is shown in Table 1. The surface roughness of the films changed with the quantity of added PSPE anthocyanin. Specifically, adding 5% PSPE anthocyanin increased the surface roughness because the interaction between the anthocyanin and matrix disrupted the bonds in the polymer (Qin et al., 2019). However, the roughness gradually decreased upon the addition of 10% to 15% PSPE anthocyanin because of the formation of hydrogen bonds and van der Waals forces between the anthocyanins and matrix (Kannatt, 2020). The results of this study align with previously reported results (Xu et al., 2023).

Antioxidant activity

The antioxidant activity of a substance is its ability to inhibit or delay oxidation and is an essential factor in determining the effectiveness of packaging films in preserving the quality and shelf life of food products. Table 1 shows that lower antioxidant activity was measured in this study compared to previous measurements of 72% for CH/cornstarch films (Hasan et al., 2022). This difference is attributed to the enhanced interaction between nanoparticles that alters the CH/PSPS film structure. However, the addition of 5%, 10%, and 15% PSPE anthocyanin significantly increases the antioxidant activity of the CH/PSPS film. The presence of phenolic hydroxyl groups is crucial for an anthocyanin-rich film to exhibit antioxidant activity. These hydroxyl groups can capture free radicals through a donation of hydrogen atoms and electron transfer [Amaregouda et al., 2022). Therefore, the more abundant phenolic hydroxyl groups are in a film, the higher the antioxidant activity of the film is expected to be (Ahidar et al., 2023). Thus, the film fabricated in this study is active and has excellent potential as a food packaging film.

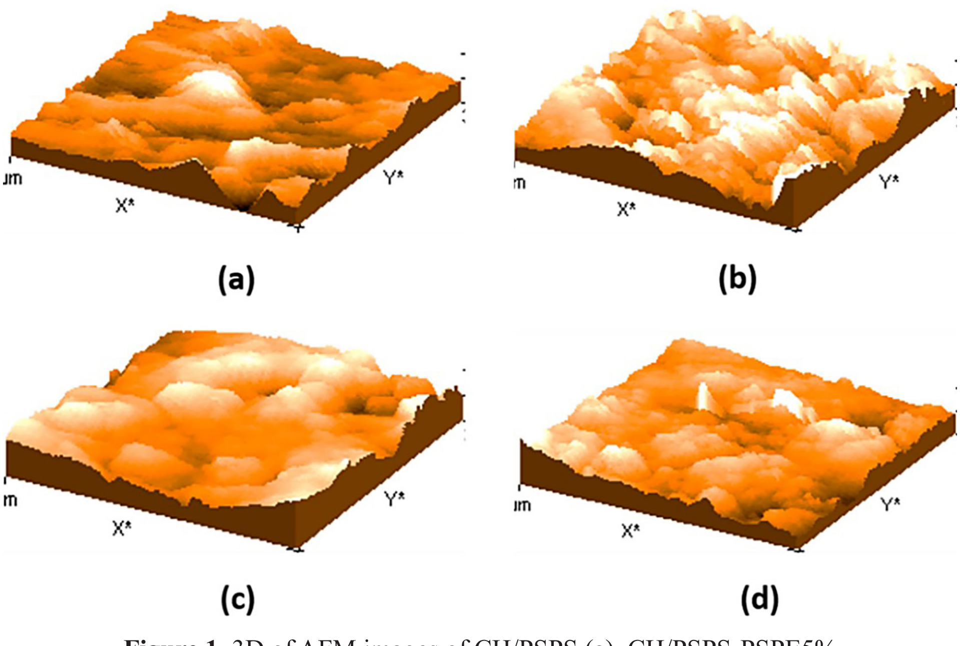


Figure 1. 3D of AFM images of CH/PSPS (a), CH/PSPS-PSPE5% (b), CH/PSPS-PSPE10% (c), and CH/PSPS-PSPE15%(d) films

Table 1. Antioxidant activity, surface roughness, and opacity of all CH/PSPS films

Film samples	Antioxidant activity (%)	Opacity (mm^{-1})	Surface Roughness (nm)	
			Ra	*Rq*
CH/SPS	16.37 ± 0.08^{a}	0.90 ± 0.03^{a}	300	390
CH/PSPS-PSPE5%	23.67 ± 0.07^{b}	2.11 ± 0.12^{b}	370	420
CH/PSPS-PSPE10%	25.77 ± 0.17^{c}	2.62 ± 0.77^{bc}	290	400
CH/PSPS-PSPE15%	26.44 ± 0.17^{d}	2.71 ± 0.30^{c}	160	200

Note: different letters within the same column indicate significantly different at a significance level ($p < 0.05$).

UV-barrier capacity and opacity

The opacity and UV-barrier capacity are critical factors for active intelligent films because UV radiation can cause food degradation, leading to nutrient loss, color fading, and even the formation of toxic substances. A high-quality film should have a robust light-barrier capacity to protect from UV radiation, maintain safety quality, and extend the shelf life of food products. Figure 2 shows the transmittance of the investigated films within the 200–800 nm range. The CH/PSPS film and all the CH/PSPS-based PSPE anthocyanin films exhibit a low transmittance in the UV range, indicating that the CH/PSPS-PSPE films possess a high UV-barrier capacity. This phenomenon occurs due to the strong UV radiation absorption capabilities of anthocyanins' phenolic compounds. The aromatic ring of anthocyanin contains chromophoric groups like C=C and C=O, which facilitate n-π* interactions, leading to effective UV absorption (Zheng et al., 2023). The opacity values of the investigated films are presented in Table 1. These results indicate that the addition of PSPE anthocyanin to the CH/PSPS matrix has a significant effect on the opacity of the resulting films, that is, the overall film opacity increases with the quantity of added PSPE anthocyanin because PSPE anthocyanin is darker than the CH/PSPS matrix. This behavior may result from the formation of a dense polymeric complex structure between CH/PSPS and PSPE anthocyanin (Capello et al., 2021).

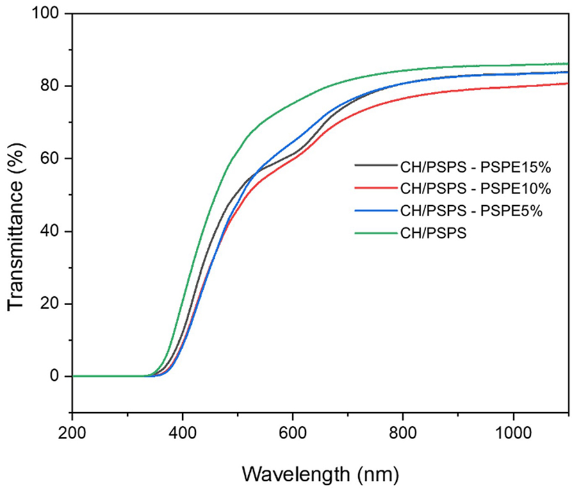


Figure 2. UV- barrier capacity of CH/PSPS and CH/PSPS-PSPEs films at different amounts of PSPE anthocyanin

Differential scanning calorimetry

Thermal properties characterization is essential for understanding the correlation between structure and properties in individual polymers and polymer composites (Rachmina et al., 2024; Lei et al., 2024). The DSC curves (Figure 3) of the CH/PSPS films showed two corresponding endothermic peaks and one exothermic peak. The first endothermic peak, which ranged from 58.71 to 88.38 °C, was associated with the loss of water in the chitosan nanocomposite films (Syahbazi et al., 2017). The addition of anthocyanin to the chitosan/starch matrix decreases the water vapor evaporation temperature of the film. This effect is caused by the interaction of

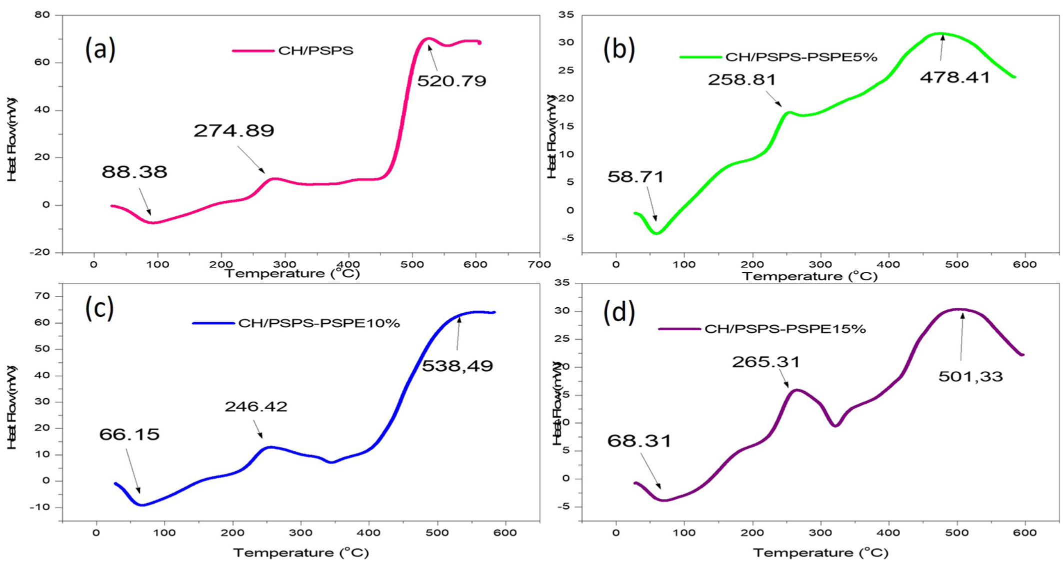


Figure 3. DSC thermogram of CH/PSPS film samples

anthocyanin molecules with matrix components, causing changes in the molecular arrangement and structure of the chitosan/starch matrix, resulting in a reduction in the temperature required for water molecules to evaporate from the film (Lozano-Navarro et al., 2018). The second endothermic peak, which correlates to the melting point (Ginting et al., 2023), occurs at temperatures ranging from 246.2 to 274.9 °C. The higher melting point observed in CH/PSPS compared to other films is attributed to the use of nano-sized chitosan raw materials (Shapi'i et al., 2022). This enables for a more efficient interaction between chitosan molecules and starch by forming hydrogen bonds (Khan et al., 2023).

Color analysis

Table 2 displays the CH/PSPS film color with and without added anthocyanin. The addition of PSPE anthocyanin affects the film color. The dark purple color of the extract intensifies the color of the film, which has a low brightness or appears dark. For the CH/PSPS film, anthocyanin addition significantly affects the L value but not the ΔE value. The brightness of the film, as indicated by the L value, decreases as the quantity of added PSPE anthocyanin increases. By contrast, the a, b, and ΔE values tend to increase with the quantity of added PSPE anthocyanin, such that the film (appears reddish and yellowish in the presence of anthocyanin.

Table 2. Color parameters (L, a, b, and ΔE) of all CH/PSPS films

Film samples	Color parameters			
	L	*a*	*b*	*ΔE*
CH/PSPS	79.90 ± 1.12[c]	-5.07 ± 1.24[a]	33.80 ± 2.21[a]	78.14 ± 28.36[a]
CH/PSPS-PSPE5%	71.63 ± 1.79[b]	-3.93 ± 2.00[a]	46.57 ± 5.57[b]	84.79 ± 26.08[a]
CH/PSPS-PSPE10%	59.00 ± 0.77[a]	4.67 ± 1.51[c]	61.23 ± 0.77[c]	93.45 ± 18.13[a]
CH/PSPS-PSPE15%	58.20 ± 1.41[a]	1.23 ± 0.37[b]	58.33 ± 1.31[c]	93.41 ± 17.83[a]

Note: different letters within the same column indicate significantly different at a significance level ($p < 0.05$)

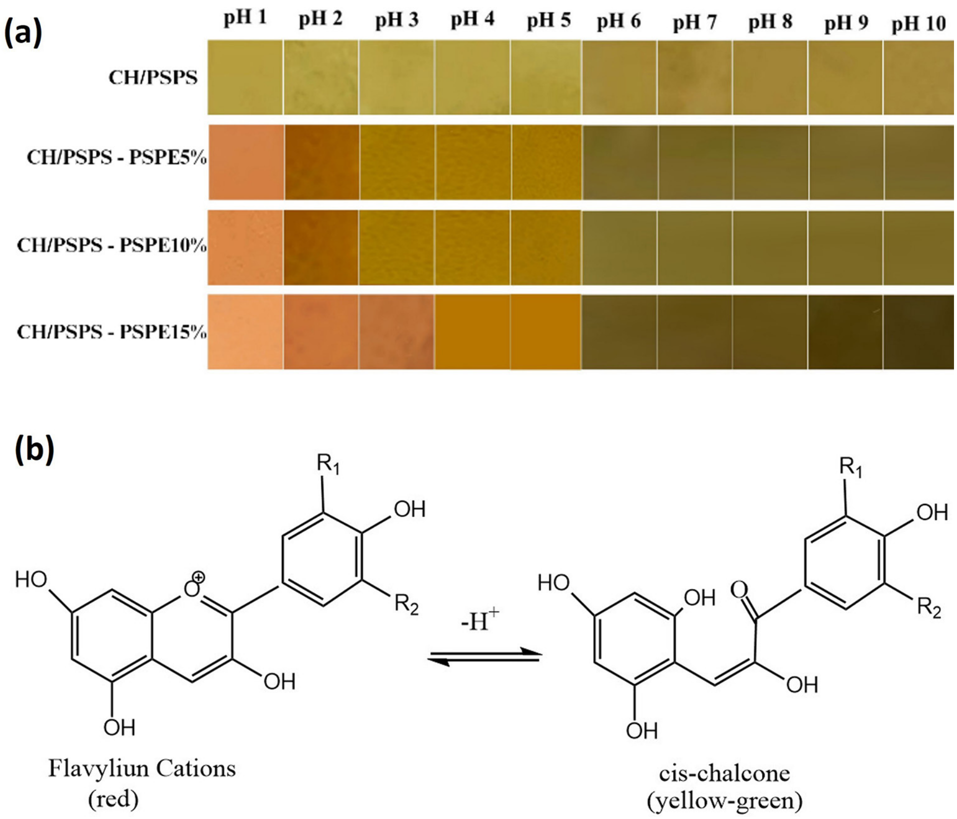
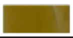
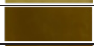
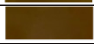
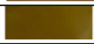


Figure 4. (a) Color changes of CH/PSPS films with and without PSPE anthocyanin after being immersed in buffer solution (pH 1–10) for 1 min; (b) structural change of anthocyanin

Table 3. Apparent color change and color parameters (L, a, b, and ΔE) of films as freshness indication for shrimp preservation at room temperature from 0, 2, 4, and 6 days

Sample	Time (day)	Color response	pH	Color parameter			
				L	a	b	ΔE
CH/PSPS	0		6.6	79.90 ± 1.12^{d}	-5.07 ± 1.24^{a}	33.80 ± 2.22^{a}	35.53 ± 2.10^{a}
	2		7.0	71.60 ± 3.18^{c}	-7.33 ± 0.98^{a}	64.87 ± 2.60^{b}	67.77 ± 2.82^{b}
	4		7.5	64.90 ± 1.57^{b}	-8.27 ± 1.47^{a}	60.83 ± 1.67^{b}	66.27 ± 1.07^{b}
	6		7.9	60.13 ± 0.85^{a}	0.57 ± 1.71^{b}	61.43 ± 0.40^{b}	68.40 ± 0.57^{b}
CH/PSPS-PSPE5%	0		6.6	71.63 ± 1.79^{b}	-3.93 ± 2.00^{a}	46.57 ± 5.57^{a}	50.21 ± 5.73^{a}
	2		6.8	62.07 ± 1.27^{a}	-4.57 ± 0.82^{a}	59.10 ± 0.96^{b}	65.51 ± 1.39^{b}
	4		7.5	59.93 ± 1.26^{a}	-2.63 ± 1.89^{a}	61.53 ± 0.33^{bc}	68.58 ± 0.74^{b}
	6		7.8	69.13 ± 2.20^{b}	-4.90 ± 1.73^{a}	66.90 ± 2.33^{c}	70.23 ± 2.24^{b}
CH/PSPS-PSPE10%	0		6.6	59.00 ± 0.77^{c}	4.67 ± 1.52^{c}	61.23 ± 0.77^{c}	68.95 ± 0.42^{b}
	2		6.7	51.33 ± 0.37^{b}	5.43 ± 1.79^{b}	54.233 ± 0.51^{a}	67.06 ± 0.35^{a}
	4		7.4	46.90 ± 1.28^{a}	7.233 ± 0.91^{a}	50.733 ± 0.81^{b}	67.24 ± 0.77^{a}
	6		7.8	48.13 ± 1.29^{a}	12.03 ± 0.87^{b}	54.733 ± 0.77^{b}	70.28 ± 0.75^{b}
CH/PSPS-PSPE15%	0		6.6	58.20± 1.41^{c}	1.23 ± 0.37^{c}	58.333 ± 1.32^{c}	66.57 ± 0.47^{a}
	2		6.7	42.333 ± 1.71^{b}	11.033 ± 1.91^{b}	48.933 ± 0.79^{b}	69.575 ± 1.25^{b}
	4		7.4	36.767 ± 1.83^{a}	17.367 ± 1.17^{a}	41.733 ± 1.35^{a}	70.341 ± 1.33^{b}
	6		7.7	64.267 ± 2.92^{d}	-4.133 ± 1.70^{d}	64.000 ± 0.94^{d}	69.152 ± 0.83^{b}

Note: at a significance level of $p< 0.05$, different letters within a common column suggest statistical significance.

This result is in line with those reported for films made by loading red dragon fruit peel anthocyanin into cassava starch/chitosan films (Pramitasari et al., 2022) (which appear red to yellow).

pH-sensitivity of CH/PSPS films

Films containing anthocyanins are generally pH-sensitive: changes in the anthocyanin structure with pH manifest as a change in the film color (Zeng et al., 2022). Figure 4 shows there were no significant color changes in the CH/PSPS film (control) with pH because no anthocyanin was added to the film; thus, CH was the dominant component of the film, which therefore appeared yellow (Zhao et al., 2023). By contrast, remarkable color variations were observed for the films with added PSPE anthocyanin in various pH buffers. Similar color changes were observed for the CH/PSPS-PSPE5% and CH/PSPS-PSPE10% films: pink (pH = 1), dark red (pH = 2), yellow (pH = 3–5), and green (pH = 6–10). The CH/PSPS-PSPE15% film appeared pink (pH = 1), dark red (pH = 2–3), and brownish-yellow (pH = 4–5). When soaked in a buffer solution with higher pHs, the color of the CH/PSPS-PSPE15% film changed to greenish-brown (pH = 6–8) and eventually became transparent green (pH = 9–10). These color changes were related to the quantity of added anthocyanin (Li et al., 2022). The observed color variation in the PSPE anthocyanin-rich CH/PSPS films was attributed to the change in the anthocyanin structure from the flavylium cation form in acidic solutions (pHs below 7) to the quinoidal anhydrase and anionic quinoidal base forms in neutral and alkaline solutions (Figure 4b) (Tang et al., 2019). Therefore, the film fabricated in this study is a potential intelligent film for food packaging.

Application of CH/PSPS films for shrimp freshness monitoring

Seafood spoilage is characterized by the generation of volatile nitrogen-based compounds, such as ammonia, dimethylamine, and trimethylamine, from protein breakdown by enzymes and microorganisms, leading to pH elevation (Chen et al., 2022). Anthocyanin-rich films are pH-sensitive and exhibit different colors depending on the anthocyanin content. Therefore, these films can be used as active intelligent films to indicate the freshness of protein-rich animal foods, such as shrimp, fish, and milk (Hu et al., 2022). Table 3 shows a gradual increase in the pH of shrimp from 6.63 to 7.91 during six days of storage. This increase was attributed to the generation of volatile nitrogen compounds, including ammonia and amines, by protein degradation (Chen et al., 2022). There were significant color changes in the CH/PSPS films containing PSPE anthocyanins at different concentrations during shrimp storage for up to 6 days. The brightness of the films, as indicated by the L* value, decreased over time. As the storage time increased, there was an increase in the a* value for all the films, except for the control film, whereas the b* value of all the films increased, corresponding to a color change to yellow. The total color difference (ΔE) gradually increased with the storage duration for all the films. On the sixth day, the film pH exceeded a threshold value, and the film color changed to green, indicating severe denaturation of the shrimp. This result demonstrates that CH/PSPS-PSPE films (anthocyanin-rich films) possess intelligent properties and are therefore suitable for monitoring the freshness of shrimp in real-time.

CONCLUSIONS

This research involved the fabrication of active and pH-responsive bio-nanocomposite films through the incorporation of different concentrations of SPE anthocyanins into the nanoparticle of the CH-PSPS matrix using the solvent-casting method. The films were thoroughly examined and characterized. According to the AFM analysis, the resulting films exhibited a smooth and compact surface, suggesting a uniform distribution of SPE anthocyanins within the matrix. The addition of SPE anthocyanins increased the antioxidant activity of the film from 16.37% to 26.44%. All films containing SPE anthocyanins exhibited excellent UV barrier properties were sensitive to pH 1–10 buffer solutions, and effectively demonstrated the freshness of shrimp during storage. Therefore, the resulting films can be used as active and intelligent packaging films for food products.

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REFERENCES

1. Aguerre-Loredo, R.Y., Fonseca-Garcia, A., Calambas, H.L., Caicedo, C.M. 2023. Improvements of thermal and mechanical properties of achira starch/chitosan/clay nanocomposite films. Heliyon, 9(6), e16782.
2. Agunos, R.I.F., Mendoza, D.V.M., Rivera, M.A.S., 2020. Anthocyanin colorimetric strip for volatile amine determination. International Journal of Food Science, 2020, 1672851.
3. Ahidar, N., Labhar, A., Benamari, O., Ahari, M., Salhi, A., Elyoussfi, A., Amhamdi, H. 2024. Phenolic content and antioxidant activity of *Cannabis* sativa L. flower from Ketama Region in Northen Marocco. Ecological Engineering & Environmental Technology, 25(1), 209–215.
4. Al-Hilifi, S.A., Al-Ibresam, O.T., Al-Hatim, R.R., Al-Ali, R.M., Maslekar, N., Yao, Y., Agarwal, V. 2023. Development of chitosan/whey protein hydrolysate composite films for food packaging application. Journal of Composites Science, 7(3), 94.
5. Amaregouda, Y., Kamanna, K., Gasti, T. 2022. Fabrication of intelligent/active films based on chitosan / polyvinyl alcohol matrices containing Jacaranda cuspidifolia anthocyanin for real-time monitoring of fish freshness, International Journal of Biological Macromolecules, 218, 799–815.
6. Baghi, F., Gharsallaoui, A., Dumas, E., Ghnimi, S. 2022. Advancements in biodegradable active films for food packaging: effects of nano/microcapsule incorporation. Foods, 11(5), 760.
7. Bhargava, N., Sharanagat, V.S., Mor, R.S., Kumar, K. 2020. Active and intelligent biodegradable packaging films using food and food waste-derived bioactive compounds: A review. Trends in Food Science and Technology, 105, 385–401.
8. Capello, C., Leandro, G.C., Gagliardi, T.R., Valencia, G.A. 2021. Intelligent films from chitosan and biohybrids based on anthocyanins and laponite: physicochemical properties and food packaging applications. Journal of Polymers and the Environment, 29, 12, 3988–3999.
9. Cazon, P., Vazquez, M., Velazquez, G. 2019. Composite films with UV-barrier properties based on bacterial cellulose combined with chitosan and poly(vinyl alcohol): Study of puncture and water interaction properties. Biomacromolecules, 20(5), 2084–2095.
10. Chen, M., Hu, L., Hu, Z., Li, G., Chin, Y., Hu, Y. 2022. Effect of chitosan coating combined with hypotaurine on the quality of shrimp (Litopenaeus vannamei) during storage, Fisheries and Aquatic Sciences, 25(2), 64–75.
11. Diksha, T., Yogesh, K.. Vijay, S.S., Tanuja, S., Saxena, D. 2023. Development of pH-sensitive films based on buckwheat starch, critic acid and rose

petal extract for active food packaging, Sustainable Chemistry and Pharmacy, 36, 101236.

12. Dong, S., Zhang, Y., Lu, D., Gao, W., Zhao, Q., Shi, X. 2023. Multifunctional intelligent film integrated with purple sweet potato anthocyanin and quercetin-loaded chitosan nanoparticles for monitoring and maintaining freshness of shrimp, Food Packaging and Shelf Life, 35, 101022.
13. Fitriani, F., Bilag, M.R., Aprilia, S., Arahman, N. 2023. Biodegradable hybrid polymer film for packaging: A review. Journal of Natural Fibers, 20(1), 2159606
14. Ginting, E.M., Motlan, Sani, R.A., Bukit, B.F. 2023. Utilization of Eco-Friendly Rice Husk Ash Waste as Reinforcement in LDPE Thermoplastics, Ecological Engineering & Environmental Technology, 24(8), 240–246.
15. Hao, Y., Cheng, L., Song, X., Gao, Q. 2023. Functional properties and characterization of maize starch films blended with chitosan. Journal of Thermoplastic Composite Materials, 36(2), 089270572211422.
16. Hasan, M., Khaldun, I., Zatya, I., Rusman, R., Nasir, M. 2022. Facile fabrication and characterization of an economical active packaging film based on corn starch-chitosan biocomposites incorporated with clove oil. Journal of Food Measurement and Characterization, 17, 306–316.
17. Hu, A., Xia, B.P., Li, T-S., Chen, M., Wang, S., Dong, W. 2022. Polysaccharide biomacromolecule film integrated with lycium ruthenicum anthocyanins for food freshness monitoring, Starch-staerke, 74(11), 2200147.
18. Husna, N.E., Novita, M., Rohaya, S. 2013. Anthocyanins content and antioxidant activity of fresh purple fleshed sweet potato and selected products. Agritech, 33, 296–302.
19. Kalpana, S., Priyadarshini, S.R., Maria Leena, M., Moses, J.A., Anandharamakrishnan, C. 2019. Intelligent packaging: Trends and applications in food systems, Trends in Food Science and Technology, 93, 145–157.
20. Kanatt, S.R. 2020. Development of active/intelligent food packaging film containing Amaranthus leaf extract for shelf life extension of chicken/fish during chilled storage. Food Packaging and Shelf Life, 24, 100506.
21. Khan, A., Ezati, P., Rhim, J-W. 2023. Chitosan/starch-based active packaging film with N, P-doped carbon dots for meat packaging. ACS Applied bio materials, 6(3), 1294–1305.
22. Lee A., and Liew, M.S. 2020. Ecologically derived waste management of conventional plastics. Journal of Material Cycles and Waste Management, 22(1), 1–10.
23. Lei, X., Ou, Y., Wang, Y., Ma, J. 2024. Thermal decomposition behavior of polylactic acid-based polyurethane resin. Polish Journal of Environmental Studies, 33(2), 1–12.
24. Li, B., Bao, Y., Li, J., Chen, Q., Cui, H., Wang, Y., Tian, J., Shu, C., Wang, Y., Lang, Y., Zhang, W., Tan, H., Huang, Q., Si, X. 2022. A sub-freshness monitoring chitosan/starch-based colorimetric film for improving color recognition accuracy via controlling the pH value of the film-forming solution, Food Chemistry, 388, 132975.
25. Li, N., Zhou, Z., Wu, F., Lu, Y., Jiang, D., Zhong, L., Xie, F-J. 2022. Development of pH-indicative and antimicrobial films based on polyvinyl alcohol/starch incorporated with ethyl lauroyl arginate and mulberry anthocyanin for active packaging. Coatings, 12(10), 1392.
26. Lou, L., Chen, H. 2023. Functional modification of gelatin-based biodegradable composite films: a review. Food Additives & Contaminants: Part A, 40, 928–949.
27. Lozano-Navarro, J.I., Diaz-Zavala, N.P., Velasco-Santos, C., Melo-Banda, J.A. 2018. Chitosan-starch films with natural extracts: physical, chemical, morphological and thermal properties. Materials, 11(1), 120.
28. Matheus, J.R.V., Dalsasso, R.R., Rebelatto, E.A., Andrade, K.A., de Andrade, L.M., de Andrade, C.J., Monteiro, A.R., Fai, A.E.C. 2023. Biopolymers as green-based food packaging materials: A focus on modified and unmodified starch-based films, Comprehensive Reviews in Food Science and Food Safety, 22(2), 1148–1183.
29. Pang, G., Ding, M., Jiang, S., Chen, P., Zhao, Z., Gao, R., Song, B., Shen, Q., Cai, F., Druzhinina, I.S. 2023. The distinct plastisphere microbiome in the terrestrial-marine ecotone is a reservoir for putative degraders of petroleum-based polymers, Journal of Hazardous Materials, 453, 131399.
30. Perveen, S., Anwar, M.J., Ismail, T., Hameed, A., Mahomoodally, M.F., Saeed, F., Imran, A., Hussain, M., Rehman, H.U., Khursheed, T., Tufail, T., Ali, S.W., Jbawi, E. 2023. Utilization of biomaterials to develop the biodegradable food packaging, International Journal of Food Properties, 26(1), 1122–1139.
31. Pramitasari, R., Gunawicahya, L.N., Anugrah, D.S.B. 2022. Development of an indicator film based on cassava starch–chitosan incorporated with red dragon fruit peel anthocyanin extract. Polymers, 14(19), 4142.
32. Qin, Y., Liu, Y., Yong, H., Liu, J., Zhang, X., Liu, J. 2019. Preparation and characterization of active and intelligent packaging films based on cassava starch and anthocyanins from Lycium ruthenicum Murr, International Journal of Biological Macromolecules, 134, 80–90.
33. Rachmina, R., Hasan, M., Hasanah, U., Halim A. 2024. Sugar palm starch/chitosan bionanocomposite films incorporated with anthocyanin and curcumin thermal properties and release kinetics. Journal

of Ecological Engineering, 25, 300–308.

34. Ratna, Yusmanizar, Aprilia, S., Munawar, A.A. 2023. Development of biocomposite edible film food packaging based on gelatin from chicken claw waste, Case studies in chemical and environmental engineering, 8, 100371.
35. Shahbazi, M., Rajabzadeh, G., Ahmadi, S.J. 2017. Characterization of nanocomposite film based on chitosan intercalated in clay platelets by electron beam irradiation, Carbohydrate Polymers, 157, 226–235.
36. Shapi'i, R.A., Othman, S.H., Basha, R.K., Naim, M.N. 2022. Mechanical, thermal, and barrier properties of starch films incorporated with chitosan nanoparticles, Nanotechnology Reviews, 11(1), 1464–1477.
37. Suhartini, Solihat, I., Foliatini, Setyawati, S.R., Nurdiani, Sulistiawaty, L.2023. Synthesis and characterization of nano chitosan-avocado seed starch as edible films, Jurnal Kimia Riset, 8(1), 49–58.
38. Tang, B., He, Y., Liu, J., Zhang, J., Li, J., Zhou, J., Ye, Y., Wang, J., Wang, X. 2019. Kinetic investigation into pH-dependent color of anthocyanin and its sensing performance, Dyes and Pigments, 170, 107643.
39. Wang, F., Xie, C., Tang, H., Hao, W., Wu, J., Sun, Y., Sun, J., Liu, Y., Jiang, L. 2023. Development, characterization and application of intelligent/active packaging of chitosan/chitin nanofibers films containing eggplant anthocyanins. Food Hydrocolloids, 139, 108496.
40. Xu, H., Shi, Y., Shi, N., Yang, J., Hao, R. 2023. Preparation and characterization of PH-responsive polyvinyl alcohol/chitosan/anthocyanin films, Food Science and Technology, 43, 98022.
41. Yu, X.M., Li, Q., Jin, Z., Jiao, A. 2023. Preparation and characterization of hydroxypropyl methylcellulose/hydroxypropyl starch composite films reinforced by chitosan nanoparticles of different sizes, Materials Today Communications, 35, 105714.
42. Yun, D., Cai, H., Liu, Y., Xiao, L., Song, J., Liu, J. 2019. Development of active and intelligent films based on cassava starch and Chinese bayberry (Myrica rubra Sieb. et Zucc.) anthocyanins, RSC Advances. 9, 30905–30916.
43. Zeng, F., Ye, Y., Liu, mJ., Fei, P. 2022. Intelligent pH indicator composite film based on pectin/chitosan incorporated with black rice anthocyanins for meat freshness monitoring, Food Chemistry: X, 17, 100531.
44. Zhao, Y., Wang, J., Zhang, M-L., Ma, X., Lv, S. 2023. Preparation and Application of pH-Sensitive Film Containing Anthocyanins Extracted from Lycium ruthenicum Murr, Materials, 16(10), 3828.
45. Zheng, L., Liu, L., Yu, J., Faraq, M.A., Shao, P. 2023. Intelligent starch/chitosan-based film incorporated by anthocyanin-encapsulated amylopectin nanoparticles with high stability for food freshness monitoring, Food Control, 151, 109798.