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Research article

Eco-friendly intelligent packaging film from dragon fruit peel pigment extract for real-time seafood freshness monitoring

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Abstract

Environmental concerns over persistent plastic waste have driven the development of biodegradable alternatives for food packaging. In this study, a pH-responsive biodegradable indicator film was fabricated from sodium alginate and starch, incorporated with a pigment-rich extract (betalain/anthocyanin mixture) obtained from dragon fruit (*Hylocereus spp.*) peel. Films were prepared at various sodium-alginate-to-starch ratios and pigment loadings, and their physicochemical, mechanical, thermal, and biodegradation properties were evaluated. The optimized formulation (SA:S = 3:7) exhibited the highest tensile strength (~21 MPa), balanced water resistance, and favorable swelling behavior. Thermal analysis revealed that the composite films exhibited enhanced stability compared to pure sodium alginate films, while biodegradation assays demonstrated a mass loss of up to 59% within 30 days under natural conditions. The pigment-containing films displayed distinct pH-dependent color changes, enabling real-time visual monitoring of shrimp freshness. Under room-temperature storage, the film changed from red to bluish-yellow after four days, corresponding to a TVB-N value of 30.1 mg/100 g, indicating complete spoilage. These findings suggest that sodium alginate–starch–pigment films are promising, eco-friendly intelligent packaging materials for freshness indication.

Keywords: *Biodegradable film, sodium alginate–starch, dragon fruit peel pigment extract, pH-responsive indicator, Seafood freshness monitoring.*

1. INTRODUCTION

Environmental pollution caused by non-biodegradable plastic waste has become a global concern, particularly in the food packaging industry [1]. Conventional food wraps are commonly made from plastics such as polyethylene, polypropylene, polyvinyl chloride, and polyethylene terephthalate, which take hundreds to thousands of years to decompose [2]. This persistent accumulation of plastic waste not only pollutes ecosystems but also contributes significantly to greenhouse gas emissions during production and disposal [3]. Hence, the development of biodegradable films derived from natural resources is both environmentally urgent and economically promising [4].

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Simultaneously, the issue of seafood freshness poses serious health risks. Spoiled seafood is often associated with pathogenic bacteria such as *Salmonella* and *E. coli*, or the formation of biogenic amines like histamine during decomposition [5]. Consumption of such seafood may lead to food poisoning, inflammatory responses, and even histamine toxicity. Therefore, an intelligent packaging solution that can signal the freshness status of seafood in real-time would greatly benefit both consumers and suppliers [6]. Such smart films could reduce foodborne illness, minimize food waste, and enhance consumer confidence in product quality.

Dragon fruit peel contains a mixture of natural pigments, primarily betalains (betacyanins and betaxanthins), together with minor anthocyanin-like phenolics [7]. These pigments exhibit strong pH-dependent color variation and have therefore attracted interest for use in intelligent packaging systems [8]. Both betalain and anthocyanin families undergo reversible structural transitions under varying pH, producing color changes ranging from red to purple, blue, or yellow [9]. These transitions are influenced by protonation–deprotonation of chromophores, formation of quinonoid or betalamic derivatives, and partial degradation under alkaline conditions. This mixed-pigment behavior allows the extract to function as a natural pH-responsive indicator for monitoring food spoilage, including seafood, where volatile basic nitrogen compounds cause the surrounding microenvironment to become more alkaline.

Several studies have explored anthocyanin-based films and biosensors for food monitoring applications. For instance, anthocyanins extracted from red cabbage [10], hibiscus [11], and purple cabbage [12] have been incorporated into biopolymer matrices such as gelatin, chitosan, or starch to produce pH-responsive indicator films. These studies have demonstrated the potential of anthocyanins as natural colorimetric sensors for detecting food spoilage. However, most reported systems are limited to short-term laboratory tests under ambient conditions and often lack detailed quantitative correlations between film color change and biochemical freshness indicators such as total volatile basic nitrogen (TVB-N) or volatile amines. Moreover, issues of poor color stability, anthocyanin leaching, and inconsistent response at low temperatures (0–4°C), typical of seafood storage, remain unresolved [13, 14].

Recent works have investigated anthocyanin-rich extracts from dragon fruit peel as natural colorants for intelligent films. For example, Saroat Rawdkuen *et al.* developed gelatin–anthocyanin films with promising color responsiveness but without stability assessment during cold storage [15]; Rianita Pramitasari *et al.* used cassava starch–chitosan films incorporating dragon fruit peel anthocyanins for fruit ripeness monitoring rather than seafood freshness [16]; Zhang *et al*. fabricated pectin/cassava starch films based on dragon fruit peel for meat packaging, but did not provide a quantitative correlation with spoilage indicators [17]. Therefore, despite existing efforts, there is still a lack of comprehensive studies combining film performance, colorimetric calibration, and cold-storage validation for seafood freshness detection.

To address these gaps, the objective of this study is to fabricate a biodegradable indicator film based on starch and sodium alginate, incorporated with natural pigments extracted from dragon fruit peel. The film is designed to visually indicate seafood freshness through pH-dependent color changes associated with volatile amine formation during spoilage. The physicochemical properties of the film are evaluated in terms of biodegradability, water vapor permeability, swelling ratio, and tensile strength. Furthermore, the color response of the film is quantitatively correlated with TVB-N values of stored seafood samples, and its indicator functionality is verified under real refrigerated storage conditions.

2. MATERIALS AND METHODS

2.1. Chemicals and materials

In this experiment, the following chemicals and materials were utilized: Sodium hydroxide (NaOH ≥ 99.9%), calcium chloride ($CaCl_2.5H_2O$ ≥ 99.7%), ethanol (C_2H_5OH ≥ 99.7%), glycerol ($C_3H_5(OH)_3$ ≥ 99.7%), sodium alginate ($(C_6H_7NaO_6)_n$ ≥ 99.5%), and starch (, all procured from Xylong Company, China. These substances were of high analytical grade, all with a purity of 99.9%, except for ethanol, which had a purity of 99.7%. The high purity levels ensured minimal interference from impurities during the experimental procedures. Distilled water was used throughout the process to maintain consistency and prevent contamination. Additionally, fresh dragon fruit peels were collected from local markets as a natural source material for bioactive compounds. All materials were handled and stored following standard laboratory protocols to preserve their integrity for the duration of the study.

2.2. The process of extracting pigment-rich from the dragon fruit peels

After collection, the dragon fruit peels were manually separated to remove any remaining pulp, thoroughly washed with distilled water, and subsequently chopped into small pieces. The prepared peels were then homogenized using a blender. For extraction, 100 g of the ground peel was immersed in 500 mL of 70% ethanol and kept at 5°C for 24 h. Following the extraction period, the mixture was filtered to separate the liquid phase. The resulting filtrate was concentrated using a rotary evaporator at 55°C, 60 rpm, and a pressure of 130 mbar to obtain the pigment-rich extract. The obtained pigment-rich extract was stored in amber glass bottles at 4°C until further use. The mild extraction and concentration conditions minimized the co-extraction of impurities such as pectin, sugars, and other phenolics.

2.3. The process of synthesis of the Sodium alginate-Stack film contains pigment-rich extract

A 2% (w/v) starch solution was prepared in distilled water and heated to 80°C under continuous stirring until complete gelatinization. Glycerol, equivalent to 30% of the total polymer dry weight, was added as a plasticizer. Separately, a 2% (w/v) sodium alginate (SA) solution was prepared in distilled water at 50°C until fully dissolved. Both polymer solutions were then mixed at various weight ratios of SA:S (3:7, 5:5, and 7:3, based on dry weight) and stirred for 15 min to ensure homogeneity.

Different volumes of pigment extract (corresponding to 10-50% v/v of the film-forming solution) were incorporated into the polymer mixture under gentle stirring. The resulting homogeneous solutions were cast into flat Petri dishes and left to form films. The wet layer thickness was approximately 3–4 mm, which resulted in final dry films with a measured thickness of 132 ± 8 µm (mean ± SD, n = 5), determined using a digital micrometer (Mitutoyo 293-831-30).

The cast films were immersed in a cross-linking bath containing 1.25% (w/v) $CaCl_2$ and 3% (v/v) glycerol for 10 min, followed by air drying at room temperature for 5 h. Finally, the dried films were conditioned in a desiccator at 52% relative humidity for 3 days before further characterization.

2.4. Characterization of the Films

The films were characterized for their physicochemical, mechanical, thermal, biodegradation, and functional properties.

The pH-responsiveness of pigment extracted from dragon fruit peel was examined by recording UV–Vis spectra (350–800 nm, StellarNet Inc., USA). The maximum absorption wavelength (λ_max) was identified for each pH condition to determine color variation. Buffer solutions with pH 1–13 were prepared using 0.1 M HCl–KCl (pH 1–2), citric acid-Na_2HPO_4 (pH 3–6), Tris-HCl (pH 7–9), and Na_2CO_3-NaOH (pH 10–13). Visible color changes were also photographed for each pH condition.

Water solubility (WS) and swelling degree were determined following the method of Gontard *et al*. [18]. Square specimens (2 × 2 cm) were dried to constant weight (W_0) and then immersed in 50 mL of distilled water at 25 ± 1°C for 24 h. For WS, the remaining undissolved film was dried and weighed (W_1), while for SD, the swollen film was gently wiped and weighed (W_2). WS and SD were calculated using the following equations:

$$WS(\%) = \frac{W_o - W_1}{W_o} x100 \ (1)$$

$$SD(\%) = \frac{W_2 - W_o}{W_o} x100 \ (2)$$

All experiments were performed in triplicate.

Mechanical properties, including tensile strength (TS) and elongation at break (EAB), were measured using a universal testing machine (Instron, USA) in accordance with ASTM D882-10. Film samples (10 × 100 mm) with an average thickness of 0.08 ± 0.01 mm were conditioned at 52% relative humidity and 25°C for 48 h before testing. Measurements were conducted at a crosshead speed of 50 mm/min, and each test was repeated three times.

The thermal stability of the films was evaluated by thermogravimetric analysis and differential scanning calorimetry (TGA–DSC, NETZSCH STA 449 F3 Jupiter, Germany). Samples (approximately 10 ± 0.5 mg) were heated from 30 to 600°C at a rate of 10°C/min under nitrogen flow (50 mL/min).

The biodegradability of the films was assessed by soil burial at a depth of 5 cm in loamy soil with a pH of 6.8 and 20% moisture content, maintained at room temperature for 30 days. The percentage of weight loss was calculated based on the initial (W_0) and final (W_t) dry weights using the following equation:

$$Biodegradation\ (\%) = \frac{W_o - W_t}{W_o} x100 \quad (3)$$

2.5. Evaluate the ability to indicate fresh seafood

Fresh shrimp with an average length of 10 ± 1 cm and a weight of 12 ± 2 g per individual were purchased from a local supermarket and immediately transported to the laboratory in an ice box maintained at 4 ± 1 °C. Approximately 150 g of shrimp (12–13 individuals) were placed in polystyrene trays (12 × 10 cm). The trays were covered with pigment-extract indicator films of identical area (12 × 10 cm), ensuring that the films did not directly contact the shrimp surface. Plastic food wrap was used as a control. All samples were stored in a household refrigerator maintained at 4 ± 1°C, equipped with an air-circulating fan and no internal light source, to minimize external effects on the color stability of anthocyanins.

The color change of the films was monitored over a 6-day period. UV–Vis–NIR spectra were recorded using a StellarNet spectrophotometer (USA) in the wavelength range of 350–800 nm to observe visible color variations of the anthocyanin-based films. The maximum absorption peaks (λ_max) corresponding to the anthocyanin chromophores were determined, and color parameters (L*, a*, b*) were measured using the SpectraWiz software. The total color difference (ΔE) was calculated using Eq. (4). Each measurement was performed in triplicate, and results were expressed as mean ± standard deviation [19].

$$\Delta E = \sqrt{(\Delta a)^2 + (\Delta b)^2 + (\Delta L)^2} \quad (4)$$

The freshness of the shrimp samples was evaluated in parallel through pH and total volatile basic nitrogen (TVB-N) analyses according to the method of Goulas and Kontominas [20]. For TVB-N determination, 10 g of shrimp flesh were homogenized with 50 mL of distilled water, followed by distillation with 2 g MgO and one drop of silicone to prevent foaming. The distillate was collected in a 3% boric acid solution containing mixed indicators (methyl red and methylene blue) and titrated with 0.1 N HCl. TVB-N content (mg/100 g shrimp flesh) was calculated according to Eq. (5). All measurements were performed in triplicate.

$$TVB - N\ \left(\frac{mg}{100g}\right) = \frac{VxCx14x100}{10} \quad (5)$$

3. RESULTS AND DISCUSSION

3.1. Physicochemical properties

Figure 1 presented the UV–Vis spectra of the dragon-fruit-peel pigment extract at different pH values.

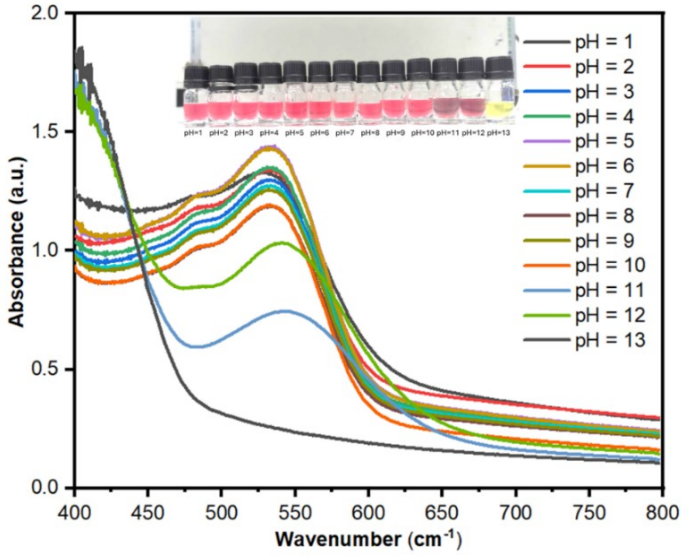

Figure 1. *Color change of pigment extract according to pH and its UV-Vis spectrum*

The extract contains both betalain and minor anthocyanin-type pigments; therefore, the spectral response represents the combined behavior of these chromophores. In acidic media (pH 1–3), the extract appears red with a dominant absorption band around 525–540 nm, attributed mainly to betacyanins. As pH increases to 4–6, the color shifts to pink or purple due to partial conversion of betacyanins and hydrated/neutral forms of other phenolics [21]. In moderately alkaline conditions, a bluish or yellowish tone emerges, consistent with the formation of betalamic derivatives and degradation pathways typical for betalains at high pH. At strongly alkaline pH (≥12), the absorbance decreases sharply, reflecting pigment instability [21]. These results confirm that the mixed pigment extract shows clear pH sensitivity suitable for indicator applications [22].

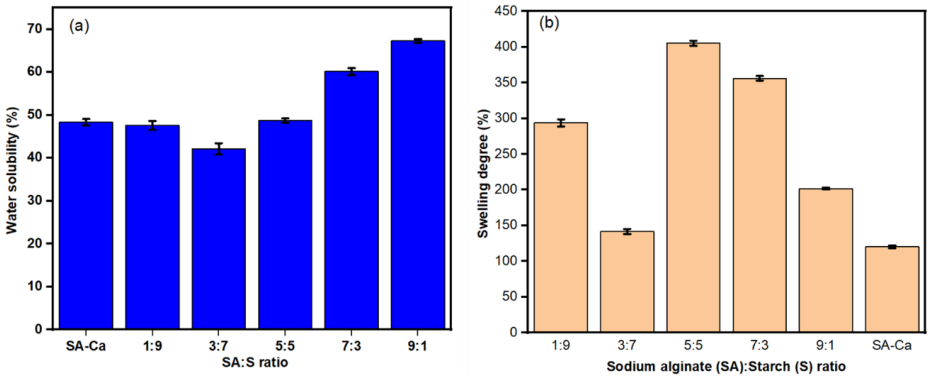

Figure 2. *Effect of sodium alginate and starch ratio on the (A) water permeability and (B) swelling degree of film*

Evaluating the water solubility and swelling degree of films is crucial for predicting their stability in humid environments, protective performance for the packaged product, and potential applicability in food packaging and biodegradable material systems [23]. The water solubility (**Figure 2A**) of the SA–Ca film was approximately 48%, which was lower than that of most SA:S films due to the strong ionic crosslinking between Ca^{2+} and sodium alginate chains, leading to a more compact network that resists water penetration. Among the SA:S films, the 3:7 ratio exhibited a relatively low water solubility (~43%), suggesting that the starch-rich composition reduced the available hydrophilic sites of SA and enhanced structural integrity in aqueous environments. By contrast, increasing the SA content from 5:5 to 9:1 markedly increased water solubility, reaching about 67% at 9:1, likely due to the reduced density of intermolecular interactions and the predominance of hydrophilic SA chains that readily dissolve in water.

The swelling degree (**Figure 2B**) showed a distinct trend. The SA:S 3:7 film demonstrated a balanced swelling capacity (~150%), significantly higher than SA-Ca (~100%) but lower than 5:5 (~410%) and 7:3 (~350%). Excessive swelling at 5:5 and 7:3 could compromise mechanical stability and dimensional integrity in wet conditions, whereas the 3:7 composition provided moderate water uptake while maintaining film cohesion. This balance between water solubility and swelling suggests that the 3:7 SA:S ratio offers the most desirable combination of water resistance and dimensional stability for practical applications, particularly in food packaging and biodegradable material systems. These results are consistent with previous studies indicating that starch incorporation into sodium alginate matrices can reduce water solubility and control swelling by modifying polymer–polymer interactions and network porosity [24, 25].

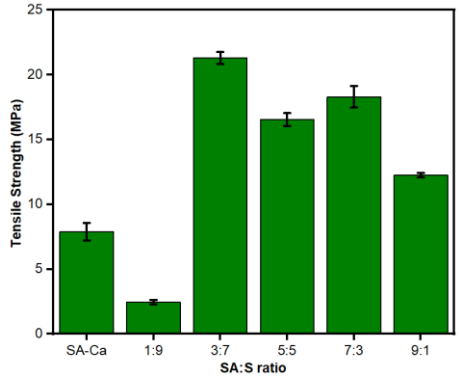

Figure 3. *Effect of sodium alginate and starch ratio on the tensile strength mechanical strength is a critical factor in determining the handling, durability, and potential application of biodegradable films in packaging*

As shown in **Figure 3**, the tensile strength of the SA:S films was significantly influenced by the polymer ratio. The 3:7 SA:S ratio exhibited the highest tensile strength (~21 MPa), indicating an optimal balance between the film-forming ability of sodium alginate and the reinforcing effect of starch granules. This synergistic interaction likely resulted from strong hydrogen bonding and interfacial compatibility between SA and S, which enhanced stress transfer under tensile load. The tensile strength decreased when the starch content was either too high (1:9, ~2.5 MPa) or too low (9:1, ~12 MPa). At high starch content, the discontinuous starch granules disrupt the polymer matrix, leading to weak points that reduce mechanical integrity. Conversely, when SA content dominates, the film becomes more rigid and brittle, limiting its load-bearing capacity. Intermediate ratios, such as 5:5 (~16.5 MPa) and 7:3 (~18 MPa), showed relatively high tensile strength but still lower than the 3:7 composition. The SA–Ca film exhibited moderate tensile strength (~8 MPa), likely due to ionic crosslinking between SA and Ca^{2+}, which increases rigidity but reduces extensibility, thus limiting overall tensile performance compared to the optimal SA:S blend. These results are in agreement with Rhim *et al.,* [24] who reported that incorporating starch into sodium alginate films can significantly improve tensile strength when the starch content is balanced to enhance intermolecular interactions without compromising film uniformity. Similarly, Talja *et al.,* [25] observed that potato starch–alginate films reached maximum tensile strength at intermediate ratios, attributing this to a denser and more homogeneous polymer network. Moreover, the tensile strength value obtained for the 3:7 SA:S film (~21 MPa) in this study is comparable to the range reported by Jiménez *et al.,* [26] for biodegradable starch-based films (18–25 MPa), indicating that the optimized formulation in the present work can match the performance of other bio-based packaging materials.

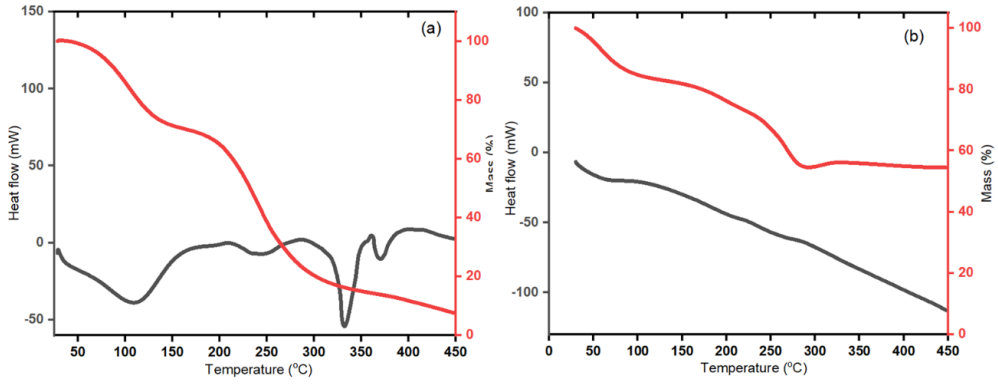

Figure 4. *TGA-DSC pattern of (A) sodium alginate film, (B) sodium alginate: starch: pigment-extract film*

The TGA–DSC results (**Figure 4**) reveal distinct differences in the thermal characteristics of the sodium alginate (SA) film and the SA:Starch: Pigment-extract film. For the pristine SA film (**Figure 4A**), the initial weight loss stage (0-150°C) is primarily attributed to the evaporation of free and weakly bound water, accompanied by a minor endothermic peak in the DSC curve. The main degradation stage occurs between 150 and 300°C, where more than 60% of the mass is lost, coinciding with a pronounced exothermic peak that reflects the scission of polymer backbones and oxidative degradation of organic chains. Above 300°C, the

sample continues to lose mass at a slower rate, corresponding to the carbonization and combustion of the residual char. In contrast, the SA:Starch: Pigment-extract film (**Figure 4B**) exhibits improved thermal stability. The initial mass loss is slower and less pronounced, indicating that the presence of starch and pigment-extract compounds reduces the rate of water evaporation. The main degradation stage (150-300°C) results in a mass loss of only 30-35%, which is considerably lower than that of pristine SA. Additionally, the DSC profile shows smoother transitions with less distinct exothermic peaks, suggesting a more gradual degradation process. Above 300°C, the composite retains a higher residual mass compared to pure SA, indicating enhanced char formation. This improvement can be attributed to the formation of a hydrogen-bonded and possibly π–π stacked network between SA, starch, and pigment-extract compounds, which may restrict polymer chain mobility and hinder thermal decomposition [27]. These results demonstrate that the incorporation of starch and anthocyanin into SA films not only improves their functional properties but also significantly enhances their thermal resistance, which is advantageous for applications requiring higher thermal endurance.

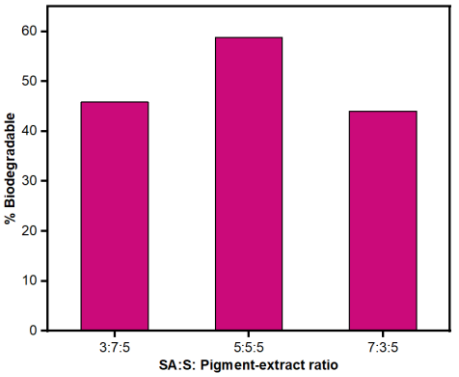

Figure 5. *Biological decomposition of sodium alginate: starch: pigment-extract film*

The biodegradation results confirm that Sodium Alginate: Starch: Pigment-extract (SA:S: pigment-extract) films exhibit high biodegradability under natural conditions due to their entirely bio-based composition. Sodium alginate and starch are polysaccharides that can be efficiently degraded by microbial enzymes such as alginate lyase and amylase, while pigment-extract, a plant-derived polyphenolic compound, does not hinder this process. The degradation mechanism is initiated by the absorption of moisture from soil or water, leading to film swelling, followed by microbial colonization and enzymatic cleavage of the polymer chains into smaller fragments, which are subsequently mineralized into CO_2, water, and microbial biomass [28]. As shown in **Figure 5**, the biodegradation percentages of the films ranged from approximately 44% to 59% after 30 days, with the SA:S: Pigment-extract ratio of 5:5:5 achieving the highest rate (~59%). This enhanced degradation may be attributed to an optimal balance between porosity and polymer network cross-linking, which facilitates water penetration and microbial access.

For comparison, typical polylactic acid (PLA) films show relatively slow degradation under ambient soil conditions, often requiring 6-12 months to achieve 50–60% mass loss due to their high crystallinity and limited water permeability [29]. Polycaprolactone (PCL) films degrade even more slowly, taking more than 12 months under similar conditions [30]. Chitosan-based films, although biodegradable, generally exhibit 25-40% weight loss after 30 days, depending on the deacetylation degree and environmental factors [31]. Therefore, SA:S: Pigment-extract films demonstrate both rapid biodegradation and the potential to serve as environmentally friendly alternatives to conventional plastic films, thereby contributing to the reduction of environmental pollution.

3.2. Evaluate the ability to indicate fresh seafood

To investigate the seafood freshness-indicating ability of the films, three types of films were used: commercial plastic film, sodium alginate: starch film, and a sodium alginate: starch: pigment-extract film. In **Figure 6**, can be seen that the commercial film and the SA:S (sodium alginate: starch) film showed no color change after four days, whereas the SA:S: pigment-extract film exhibited a distinct color change from red to a slightly bluish-yellow colorless after 4 days. This color change is likely due to the accumulation of volatile organic amines in the shrimp, such as ammonia, dimethylamine, etc. The possible mechanism is as follows:

NH_3 diffuses into the film, forming $NH_3 \cdot H_2O$, which is then hydrolyzed into NH_4^+ and OH^-, creating an alkaline environment and thereby increasing the pH value [32].

A key parameter for accurately determining the nitrogen content in shrimp is the total volatile basic nitrogen (TVB-N) measurement - a standard indicator of shrimp freshness. According to DB33451-2003 (China), fresh shrimp have a TVB-N value < 15 mg/100 g, incipient spoilage occurs when TVB-N = 20–30 mg/100 g, and complete spoilage is indicated by TVB-N > 30 mg/100 g. In this experiment, the total nitrogen content on day 4 was measured at 30.1 mg/100 g, indicating that the shrimp were completely spoiled by day 4. From the observed color change of the pigment-containing film in assessing shrimp freshness, it can be concluded that the developed film can be effectively applied as a freshness indicator for seafood.

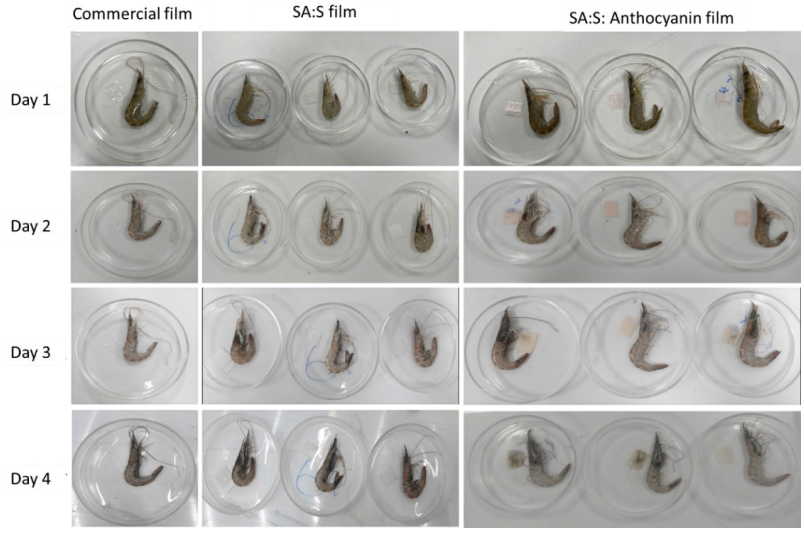

Figure 6. *Image of the ability to indicate the fresh shrimp of commercial wrap, sodium alginate: starch (SA:S), and sodium alginate: starch: pigment-extract (SA:S: pigment-extract)*

4. CONCLUSION

This study successfully developed a biodegradable, pH-sensitive film from sodium alginate and starch, functionalized with pigment extracted from dragon fruit peel. The optimized formulation (SA:S = 3:7) provided superior tensile strength, controlled water permeability, and moderate swelling capacity, alongside improved thermal resistance and rapid biodegradability. Importantly, the film demonstrated effective seafood freshness indication, with a visible color transition aligning with established TVB-N spoilage thresholds in shrimp. By integrating waste-derived natural pigments into biodegradable polymer matrices, this work offers a dual environmental benefit: reducing plastic pollution and valorizing agricultural by-products. The developed film holds strong potential for application in intelligent packaging systems, enhancing food safety, reducing waste, and promoting sustainable packaging solutions.

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