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The Development of Intelligent Film From Crosslinked–Acylation Cassava Starch and Purple Sweet Potato Anthocyanin for Monitoring Indian Mackerel Fish Freshness

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ABSTRACT

Edible coatings and films have gained significant interest due to their ability to extend the shelf life of fresh products. This research aimed to develop a novel indicator film using crosslinked–stearic acid acylation-modified cassava starch and anthocyanin extracts, both of which can be utilized to create solid matrices that are safe for consumption. The physical properties of the films improved with the use of modified starch, exhibiting a stronger tensile strength (from 0.96 to 1.02 MPa). Additionally, flexibility increased from 121% to 129%, and the water vapor transmission rate was significantly reduced from 5.86 to 1.79 g/m^2/h. Anthocyanin extracts also contributed to improvements in flexibility and reductions in water vapor transmission rate. Differential scanning calorimetry analysis indicated an increase in the film's thermal stability, with a transition temperature shifting from 276°C to 299°C, attributing to changes in the starch characteristics following modification. Fourier transform infrared spectroscopy revealed alterations in the molecular interactions between the C—H and C═C groups. The application of these edible films for storing and monitoring Indian mackerel fish meat demonstrated consistent results, particularly in terms of color change due to an increase in pH levels (from 7 to 8.5 over 2 days). Furthermore, TVB-N levels in fish meat covered with the edible film were successfully reduced from 71 to 46 mg/100 g over the same period. These findings indicate that the films can function as intelligent food packaging, possessing antioxidant properties beneficial for human health while being biodegradable, thus reducing their environmental impact.

1 | Introduction

Starch is a common agricultural material used in food packaging, sourced from some commodities like corn, rice, potatoes, wheat, and cassava (Pelissari et al. 2009). While starch is primarily extracted from plant sources, it can also be obtained from roots, grains, tubers, and legumes. It serves as a promising biopolymer, exhibiting two microstructural forms—linear and branched—due to its biodegradability, renewability, and abundance. These structures form crystallizable chains that enhance the strength of the polymer (Onyeaka et al. 2022). Among the numerous types of starch, cassava is one of the most widely produced in Indonesia, with 17.7 million tons produced in 2021. Indonesia is also one of the top five cassava producers in the world, making cassava a more captivating crop as a source of starch (Food and Agricultural Organization 2021). Its low cost, renewability, and rapid biodegradability make cassava starch a viable option for developing plastic alternatives (Colivet and Carvalho 2016).

Starch films are odorless, nontoxic, biocompatible, colorless, tasteless, and can withstand permeation by carbon dioxide and oxygen (Shah et al. 2016). Despite these advantages, their mechanical strength and moisture resistance remain limited. The high availability of active hydroxyl groups on the starch surface makes it amenable to chemical modification. Modification of starch has been integrated with novel functional groups on starch molecules or to change the starch molecular size and particle properties, thereby altering the natural properties of starch (Dai, Zhang, and Cheng 2019).

Plasticizers are essential for improving the properties of edible films, particularly those based on polysaccharides and proteins. Glycerol is a common plasticizer due to its low-molecular-weight material that is introduced into polymeric film-forming materials to increase their thermoplasticity, thereby enhancing flexibility and processability. The combination of alginate with glycerol through thermomixing is frequently applied to offset the mechanical weaknesses of glycerol (Gao, Pollet, and Avérous 2017). Moreover, cross-linking and stearic acid acylation can be used to modify the starch's hydrophilic and hydrophobic sides, which attract each other (Hustiany 2017). Amylopectin interactions indicate the properties of the crosslinked–acylation starch. However, providing concrete evidence of where the alteration occurs is quite challenging. This cross-linking acylation enhances shear stability, which relates to the tensile strength (TS) of the film, while film swelling is determined by water penetration into its surface (Liu, Ramsden, and Corke 1999).

The demand for better packaging has resulted in the development of multifunctional smart packaging, which is gaining significant interest for its ability to oversee food safety and aid in the storage and delivery of food products (Jiang et al. 2020; Naghdi, Rezaei, and Abdollahi 2021). Intelligent packaging may provide consumers with reliable data on the quality of food in a simple and rapid manner. Using pH-sensitive packaging is one of the clever methods used to determine the freshness of food goods. The chemical components released because of contamination from degradation by microbial species, such as dimethylamine, trimethylamine, and ammonia, commonly known as total basic nitrogen (TVB-N), can also be used to identify food freshness (Esfahani et al. 2022). Therefore, TVB-N levels and pH variations can be an efficient indicator of food freshness, particularly in protein-containing foods such as nuts, dairy products, meat, or fish products.

Intelligent packaging must be nontoxic, biocompatible, and resistant to degradation. If a color shift is a component that allows the customer to evaluate quality, synthetic pigments such as bromocresol, xylenol blue, and other hazardous materials are usually used in the manufacture of these packaging (González et al. 2022). Therefore, natural pigments must be employed to replace synthetic pigments in intelligent films to indicate food freshness. Natural anthocyanin is responsible for the pink, red, violet, and blue hues of vegetables and fruits, which are nontoxic, biocompatible, and harmless because they are sourced from plants. Usually, food degradation is accompanied by pH changes, and anthocyanin takes on diverse chemical forms and colors as a function of pH; hence, anthocyanin-rich films are seen as promising intelligent indicators for monitoring the freshness of food (Yun et al. 2019). Anthocyanins, on the other hand, are phenolic compounds with antioxidant and antibacterial capabilities that may be used to preserve nutritional qualities, extend shelf life, and maintain the quality of a variety of food items (Oladzadabbasabadi et al. 2022). With these characteristics, anthocyanin could be fortified in food packaging to develop intelligent films.

Purple sweet potato (PSP) (*Ipomoea batatas* sp.) is a nutritious food with purple flesh that is high in anthocyanins. The majority of the anthocyanins in PSP are acylated, making them suitable for developing color-changing food packaging. Chemical compounds containing acylated anthocyanins exhibit intramolecular copigmentation and are more persistent than nonacylated structures (Giusti and Wrolstad 2003). Different authors have reported that intelligent films containing natural pigments from PSP have been investigated with different hydrocolloid matrices (Jiang et al. 2020; Li et al. 2023; Rahmadhia, Sidqi, and Saputra 2023; Wei et al. 2017; Yong et al. 2022; Li et al. 2019b; Zong et al. 2023). Our previous research demonstrated that modified starch-based edible films incorporating beetroot (*Beta vulgaris* L.) extract exhibit excellent mechanical and physical properties and high antioxidant activity. However, its ability to detect pH changes was less sensitive to pH and temperature, especially during storage and processing. This evidence was also supported by another study by Kayın et al. (2019). In this study, anthocyanin was incorporated with modified cassava starch (cross-linking and acylation) to develop a biodegradable intelligent edible film. The structure of the composite films, physical properties, and functional properties were measured and compared with those of unmodified cassava starch and unfortified edible film. Furthermore, the film's potential application in detecting food deterioration in Indian mackerel fish (*Rastrelliger* sp.) was studied. The developed indicator film has a high potential for use in the food industry and consequently has a high practical value.

2 | Materials and Methods

2.1 | Materials

PSP samples were purchased from Lion Super Indo Ltd. (Semarang, Indonesia) and stored in a dry place with silica gel. Cassava starch was supplied by Agung Jaya Inc. (Yogyakarta, Indonesia). High-purity glycerol (> 85%) was supplied by Indrasari Lp. (Semarang, Indonesia). Sodium alginate, sodium tripolyphosphate (food grade), stearic acid, hydrochloric acid (HCl) solutions (> 32%), low-concentration ethanol (70%), sodium carbonate powders (Na_2CO_3), and solid caustic soda (NaOH) were purchased from Merck Chemicals and Life Sciences Ltd. (Jakarta, Indonesia).

2.2 | Extraction of Anthocyanin From PSP

Anthocyanin was extracted from PSP according to Jiang et al. (2020), with some modifications. After PSP was peeled and washed, it was cut into small dice. Then, PSPs were

TABLE 1 | Edible film formulation.

Variable	Type of cassava starch	Anthocyanin extract (mL)
Control	Native starch	0
MC 0	Modified starch	0
MC 2	Modified starch	2
MC 4	Modified starch	4
MC 6	Modified starch	6
MC 8	Modified starch	8
MC 10	Modified starch	10

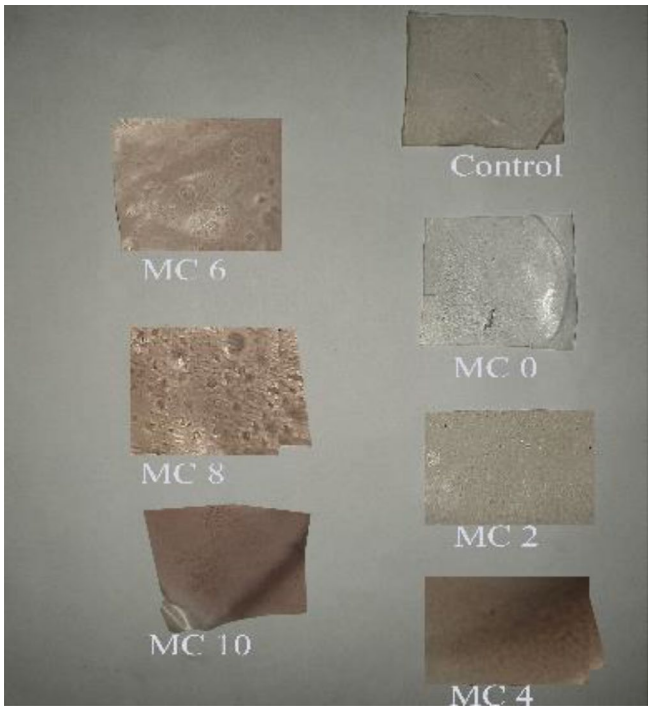

FIGURE 1 | The appearance of the produced film.

steamed in a wok with a steaming rack at 100°C for 5 min. Steamed PSPs were sliced into thin slices, and then were dried at 40°C using Food Dehydrator (RL 202, Runling Inc., China) for 24 h. Dried PSPs were ground into powder and passed through a 40-mesh sieve. Approximately 50 g of PSP powder was mixed with 500 mL of 50% ethanol–water solution (1:10 *v/v*). The solution was heated at 50°C for 5 h and then macerated at room temperature for approximately 12 h. Anthocyanin solution was filtered through filter paper using a vacuum pump, and the extract (anthocyanin) was obtained. The extract was concentrated using a rotary evaporator (RV 10, IKA Scientific, China) at 50°C (based on boiling point of ethanol at 250 mmHg) and the flask was protected using aluminum foil. Concentrated solution was stored and protected from light. Obtained anthocyanin in the PSP was measured to be 68.5 mg/100 g with pH differential method using spectrophotometer UV–vis.

2.3 | Film Preparation

Modified cassava starch was prepared using cross-linking and stearic acid acylation method. This method was referred to Hustiany (2017). Film production was begun by mixing 3 g of cassava starch and 0.5 g of alginate, which was then dissolved in 100 mL of distilled water. Solution was adjusted tp pH 7 by adding hydrochloric acid (1 mol/L) and caustic soda (1 mol/L). Glycerol (7.2 ml) was added and then stirred at 70°C for 90 min. The solution was allowed to cool down to room temperature. Table 1 shows the film's formulation that used in this study. The solution was then put into a glass mold and dried in a 45°C oven for 120 min before being left at desiccant (room temperature) for 2 days (Bergo, Sobral, and Prison 2010). The final appearance of films are shown in Figure 1.

2.4 | Film Characterization

2.4.1 | SEM

The cross-sections of the indicator films were observed using a scanning electron microscope (Philips XL series 30, Netherlands) operating at an acceleration voltage of 20 kV and magnifications ranging from 500 to 1200. The surface and cross-section structures of the composite films were photographed and observed (Wang et al. 2022).

2.4.2 | Fourier transform infrared (FTIR)

The FTIR spectra of the control film and the anthocyanin-modified film were acquired using a PerkinElmer Spectrum IR 10.6.1 (FTIR; Thermo Scientific Diamond Nicolet IS 5, US) spectrometer within the wavelength range of 400–4000 cm^{-1} (Wang et al. 2022).

2.4.3 | DSC

The thermal transitions of the components were assessed through differential scanning calorimetry (Hitachi STA200RV). Approximately 4–5 mg of the dehydrated film samples were enclosed in an aluminum crucible, while an empty aluminum crucible served as the control for background measurements. Nitrogen was employed as the protective gas with a flow rate of 20 mL/min. The heating rate was set at 10°C/min. The DSC analysis covered a temperature range spanning from 30°C to 550°C (Wang et al. 2022).

2.4.4 | Physical Property of Films

The mechanical properties of the film (TS, elongation at break (EAB), and film thickness) were measured using a standard approach. The ASTM-D D412-98 technique was used for TS and elongation analysis (ASTM 2002). Using mechanical universal testing equipment (Lamy Rheology TX-700 Texture Analyzer, France) with a power of 50 N and a speed of 1.0 mm/s was used on a film area size of 5 × 2 cm. A screw micrometer (LISM micrometer,

France) with an accuracy of 0.01 mm was used to measure film thickness. The measurements were taken at five different spots on the edible film. Water vapor transmission rate (WVTr) was determined by the method referred to as ASTM E96/E96M-16 procedure (ASTM 2016). Films with area size of 5 × 5 cm were placed inside desiccator containing silica gel at room temperature. Distilled water, separated with small glass, was placed inside desiccator to maintain humidity. Edible film was weighed every 24 h for 5 days. The slope (the change in mass at each time interval) was computed using Equation (1) to calculate the WVTr value.

$$\mathrm{WVTr} = \frac{\text{slope}}{\text{area}} \cdot \frac{1\ \text{day}}{24\ \text{h}} \left(\frac{\text{g}}{\text{m}^2 . \text{h}} \right) \quad (1)$$

2.4.5 | The Color of Films

Film color was assessed using a portable colorimeter (PCE-CSM 5, USA) with illuminant C (representing average daylight with color temperature near about 6800 K) and a standard 10° observers as reference. The color attributes of control and the anthocyanin-modified films were measured based on parameters such as lightness (L), redness–greenness (a), and yellowness–blueness (b). The overall color difference (ΔE) was computed following the methodology outlined by Choi et al. (2017), employing Equation (2) below.

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

where: $\Delta L^* = L^* — L_0$; $\Delta a^* = a^* — a_0$; $\Delta b^* = b^* — b_0$; L_0, a_0, and b_0 are the initial values of edible film.

2.4.6 | UV–vis Spectra

UV–vis spectra of edible film were measured at pH values ranging from 1 to 14 using a SHIMADZU UV-1900i spectrophotometer (Kyoto, Japan). This measurement aimed to assess the impact of pH changes on anthocyanin structure. The absorbance of edible film solutions with 10-mL anthocyanin extract as a reference (before the film dried) at pH values 1–14 was recorded within the wavelength range of 400–600 nm (Choi et al. 2017).

2.4.7 | Antioxidant Activity

Antioxidant activity contained in edible films was estimated by in vitro analysis using 1,1-diphenyl-2-picrylhydrazyl (DPPH) assay according to Esfahani et al. (2022). A set of solution of DPPH was mixed with edible film solution (before the film dried) and the absorbance was recorded using a spectrophotometer (GBC Cintra 101, Australia) at a wave of 517 nm. The DPPH free radical-scavenging activity was calculated using Equation (3):

$$\%\,\mathrm{AA} = \frac{\left(\mathrm{A_0} - \mathrm{A_t}\right) \mathrm{x}\ 100}{\mathrm{A_0}} \quad (3)$$

where A_0 is the initial absorbance of the DPPH solution and A_t is the absorbance of the leftover DPPH after the steady-state reaction with the film sample. Each analysis was performed three times ($n = 3$).

2.5 | Film Practical Application

2.5.1 | Fish Spoilage Trial

Indian mackerel fish were prepared by peeling the skin and cutting the fish meat into strips of 10 g. Every piece of fish meat was placed and wrapped inside eight different variable edible films measuring 10 × 5 cm. Each sample was stored in an incubator at room temperature. The color of the film specimen and the TVB-N content of the fish meat were recorded every 24 h for 2 consecutive days.

2.5.2 | pH Measurement

Ten grams of fresh long-jawed mackerel fish were prepared and placed on sterilized petri dishes. These fish samples were stored at room temperature and subjected to analysis every 24 h, for a total duration of 48 h to detect pH changes. Each fish sample was thoroughly blended with 90 mL of distilled water and pH value was tested by a digital pH meter.

2.5.3 | Biodegradability of Film

Biodegradability tests were conducted by referring to a method from Jaramillo et al. (2016). Natural soil with humus characteristic (obtained from Semarang, Indonesia, which is composed of dark-brown Mediterranean soil and gray alluvial association soil) was poured into a tray measuring 10 × 20 × 5 cm^3. Samples of each system measuring 2 × 2 cm were initially weighed and then buried in the soil to a depth of approximately 1 cm. The tray was placed in open storage at room temperature. Each sample was weighed every 24 h for 15 days (after the dirt on the film was cleaned). The mass data obtained from the test were plotted graphically to show mass changes over the course of 15 days.

2.6 | Statistical Analysis

All measurements were taken in triplicate, and the data were analyzed using IBM SPSS 25.0 and Origin 8.5.0 software. The findings were reported as average value ± standard deviation. Three distinct components of the analysis—control film, modified cassava starch, and 10 mL of anthocyanin inserted into the modified cassava starch—were compared using SEM, FTIR, and DSC. To observe the distinct impact of the greatest anthocyanin content, 10 mL was selected.

3 | Results and Discussions

3.1 | SEM Analysis

The morphology of the cross-sections of the starch films is shown in Figure 2. The matrix in all films is consistent and smooth. The film created from unmodified cassava starch (Figure 2a), on the other hand, has a fractured/cracked matrix (shown in red circle) which suggests weaker structural integrity likely due to insufficient bonding between starch molecules. This is not observed in

films created from modified cassava starch (Figure 2b); it is hypothesized that this phenomenon happens as a consequence of changes/alteration of the starch structure in cassava starch as a result of cross-linking from sodium tripolyphosphate which binds additional starch molecules, resulting in a rigid film structure with modified starch that is stronger and denser (Bajner 2005; M. Li et al. 2022). The film with modified starch (Figure 2c) likewise had a tight structure when anthocyanin extract was applied, demonstrating that anthocyanin from PSP was properly incorporated into the film with modified starch. This integration is facilitated by the inclusion of alginate which is mixed homogeneously in the film and serves to encapsulate the anthocyanin (Romruen et al. 2022; Schlindweinn et al. 2022). These structural changes contribute to the enhanced mechanical strength and potential functional performance of the modified films.

3.2 | FTIR Analysis

The intermolecular interactions that occur in edible films with unmodified and modified cassava starch are illustrated in Figure 3a. Films with control variables exhibit an absorbance peak at 3289 cm^{-1}, corresponding to the presence of the normal "polymeric" OH stretch from polysaccharides. Strong absorption band at 2886 cm^{-1} is attributed to the stretching vibration of C—H (Nandiyanto, Oktiani, and Ragadhita 2019). Then, the bands at 1647, 1410, and 1020 cm^{-1} were ascribed to the stretching of C═C, bending of C—O—H, and C—O stretching. The FTIR spectrum of MC 0 did not change significantly after the starch component was replaced by cross-linking–acylated-modified starch. However, in the modified starch edible film, there was a peak shift at around 3295 and 2891 cm^{-1}, suggesting changes in the interaction between OH^- and C—H groups. These interactions between polysaccharide molecules that are connected to one other as a result of crosslink modification, which binds additional starch molecules by sodium tripolyphosphate, produce this alteration (Bajner 2005).

For the FTIR spectrum with variations in anthocyanin levels shown in Figure 3b, it can be observed that there is a shift in the absorption peak in the spectrum range of 2900–2880 cm^{-1}, which indicates C—H stretching when anthocyanin is added. Additionally, the spectrum range of 1500–1650 cm^{-1} suggests there is a broad absorption when anthocyanin from PSP is added, suggesting interaction of C═C group stretching and is also related to the interaction of the aromatic group, likely resulting from π-π interactions between anthocyanin's aromatic

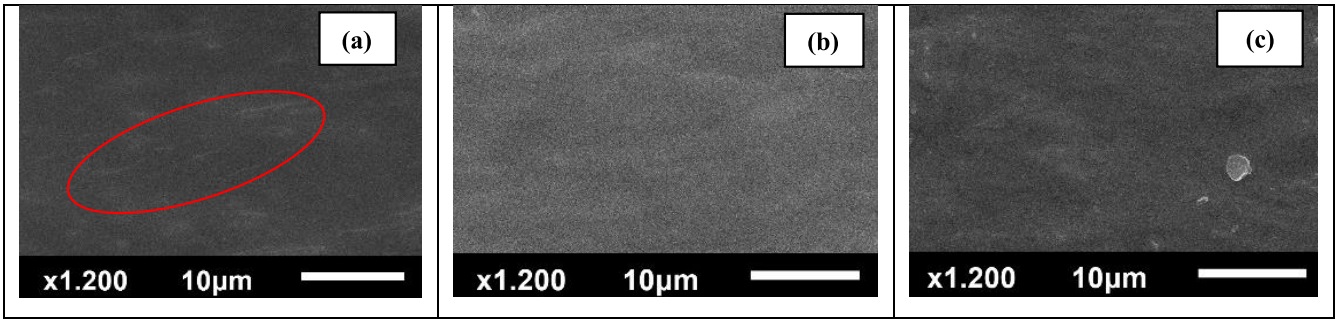

FIGURE 2 | Cross section images of the (a) control variable, (b) MC 0, and (c) MC 10 at 1200x magnification.

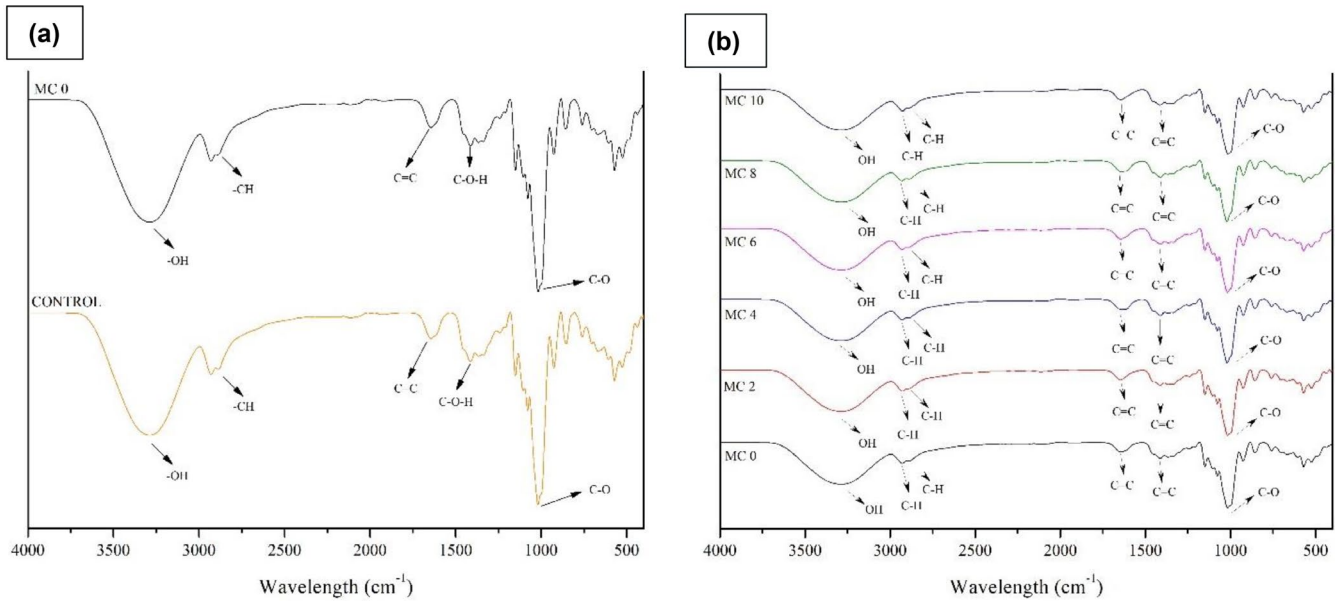

FIGURE 3 | FTIR spectra comparison of (a) unmodified starch edible film and modified starch edible film; and (b) modified starch edible film with different PSP anthocyanin volume.

rings and starch components (Jiang et al. 2020). The addition of PSP anthocyanin to the films led to an alteration in physical and chemical interaction. This interaction could be attributed to the cross-linking of anthocyanin by hydrogen bonds with the support of alginate and starch, making it a promising candidate for applications in food packaging (Choi et al. 2017).

3.3 | DSC Analysis

DSC analysis is used to evaluate the thermal resistance of edible films and to offer insight into changes in the intermolecular structure of the film brought on by temperature variations (Nisar et al. 2018). Figure 4a presents the DSC results of native starch and modified cassava starch edible films with two peaks: the endothermic peak of native starch and modified cassava starch edible films at 82.98°C and 76.09°C, respectively, as the melting temperature (Tm) values, and the exothermic peak of native starch and modified cassava starch edible films at 278.16°C and 299.54°C, respectively, as signs of sample decomposition. This phenomenon is similar to the research of Kadzińska et al. (2020), where exothermic peaks and endothermic peaks are also found in edible films made from sodium alginate. The edible film made from modified cassava starch has a lower Tm value than native starch. This is consistent with studies by Choi et al. (2022) and Ali and Dash (2023), which found that the modified edible film's Tm was lower than the native starch edible films. Furthermore, the exothermic peaks of the native starch and modified cassava starch films exhibit noticeable differences, indicating that the film made with modified starch is more thermally stable. It might result from the modification caused by STPP cross-linking, which can remove inter- and intramolecular interactions between starch chains and attract hydrophilic functional groups to starch molecules, reducing the crystallinity of the modified film (Ali and Dash 2023; Choi et al. 2022).

TGA analysis was performed to determine the effect of thermal stability on the interaction of ingredients in the film (Thakur et al. 2017). Figure 4b indicates the thermal change and weight loss events of the native starch and modified cassava starch edible films. Based on Figure 4b, both edible films showed weight loss in three stages with a temperature range of 31°C–550°C. The first weight loss occurs between 31°C and 200°C due to the loss of moisture content of the edible film. The modified cassava starch edible film has a greater weight loss than the native starch edible film. The second weight loss occurs between 200°C and 300°C, which is caused by the evaporation of the glycerol plasticizer, and other covalent bonds formed during cross-linking will also be broken at this temperature (Sharma, Sharma, and Saini 2018). The final weight loss occurs after 300°C–550°C caused by the decomposition of the starch crystallinity (Thakur et al. 2017). The TGA results show that the modified cassava starch edible film has fairly low thermal stability up to 300°C, but the thermal stability increases at temperatures above 300°C. Edible films modified by cross-linking have better thermal stability because they have a more complex structure and delay the decomposition of polysaccharides at high temperatures (Guevara et al. 2022). The DSC and TGA results show that cross-linking cassava starch reduces the Tm and improves thermal stability at high temperatures, making the modified film better suited for applications that require stability under heat.

TABLE 2 | Physical properties of edible films.

Variable	Tensile strength (MPa)	Elongation at break (%)	WVTr $\left(\frac{g}{m^2.h}\right)$
Control	0.96 ± 0.04^{bc}	121.87 ± 0.1^{a}	5.86 ± 0.21^{d}
MC 0	1.02 ± 0.23^{c}	129.87 ± 0.24^{a}	1.79 ± 0.33^{c}
MC 2	0.83 ± 0.12^{b}	134.13 ± 0.2^{a}	0.12 ± 0.11^{a}
MC 4	0.61 ± 0.32^{a}	164.80 ± 0.24^{bc}	0.32 ± 0.14^{bc}
MC 6	0.67 ± 0.19^{a}	173.73 ± 0.1^{c}	0.19 ± 0.19^{a}
MC 8	0.72 ± 0.31^{b}	178.80 ± 0.63^{c}	0.31 ± 0.41^{b}
MC 10	1.20 ± 0.21^{c}	159.60 ± 0.33^{b}	0.21 ± 0.37^{ab}

Note: Number of trials = 3. Values quoted are mean values ± standard deviations.
[a–d]Within a column, significant differences ($p < 0.05$) for edible films.

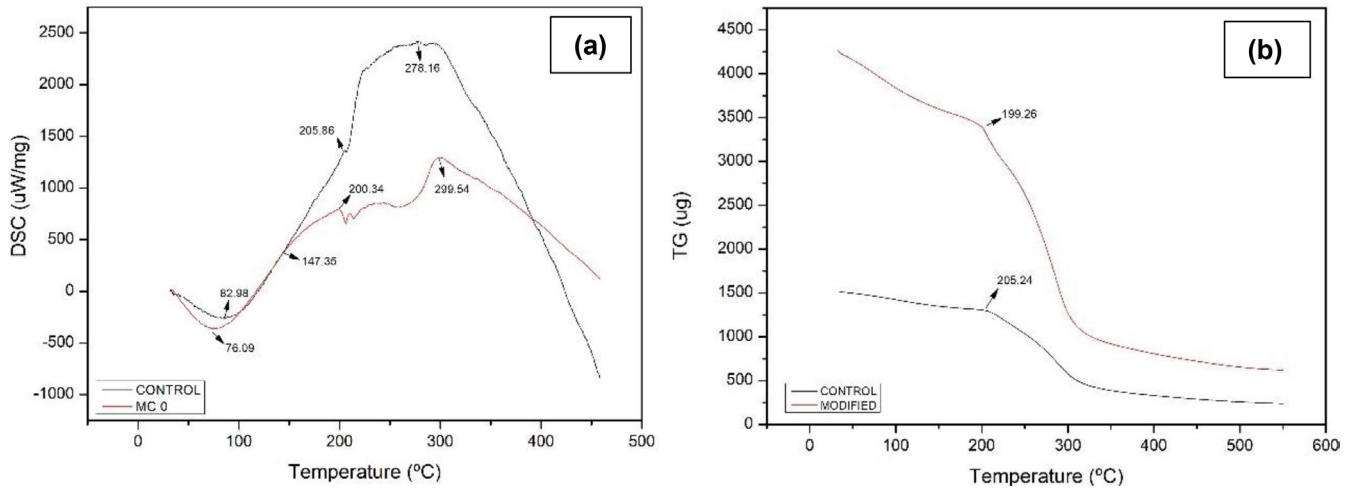

FIGURE 4 | (a) DSC thermograms of edible films from native starch and modified cassava starch, and (b) TGA curves of native starch edible film and modified starch edible film.

3.4 | The Effect of Modified Cassava Starch in Edible Film Properties

The mechanical properties of the edible films were measured in terms of TS and EAB as shown in Table 2. The TS of the edible films from native starch and modified starch increased from 0.96 to 1.02, and the elongation of the edible film increased from 121.87 to 129.87. The TS value of modified cassava starch-based edible film was higher than that of unmodified edible film. Based on research by Yıldırım-Yalçın, Şeker, and Sadıkoğlu (2019), the TS value of modified starch films was also higher than that of unmodified starch films. The increase in TS value was caused by an increase in the crosslink density in the starch film, which resulted from the crosslink reaction between the hydroxyl groups and the cross-linking agent (Detduangchan, Sridach, and Wittaya 2014). Modification of starch through cross-linking strengthened the hydrogen bonds between starch phosphate and plasticizer, consequently increasing the elongation value (Gutiérrez et al. 2015). These crosslinks result in a denser network structure, enhancing the film's mechanical integrity. Additionally, increasing EAB value may also result from intermolecular interactions resulting from STPP cross-linking modifications (Dai, Zhang, and Cheng 2019).

The WVTr value was an important parameter because it can be used to determine the shelf life of packaged products. Lower WVTr can extend the shelf life of perishable food products by providing a better barrier against moisture (Dai, Zhang, and Cheng 2019). Table 2 indicates the WVTr value decreased from 5.86 to 1.79 for edible film with native starch and modified starch. The phosphate group from sodium tripolyphosphate will interact with the hydroxyl group in starch, leading to cross-linking that hinders the infiltration of water vapor into the material. Modifying the starch also causes the amylose chains to become linear, resulting in the formation of links between starch molecules, reducing the film's hydrophilic characteristics (Detduangchan, Sridach, and Wittaya 2014). As a result, the modified edible film exhibits a lower WVTr value, indicating its strong moisture-protective properties. The cross-linking process leads to a more compact molecular structure, thereby reducing the options for water vapor to pass through. The tighter the structure, the lower the films' ability to be penetrated. (Ebrahimi et al. 2016).

3.5 | The Effect of Anthocyanin on Modified Edible Film Properties

Table 2 indicates the mechanical parameters of the edible films in terms of TS and EAB. The TS of the edible film initially decreased from 1.02 to 0.61 when anthocyanin ranged from 0 to 4 mL. Anthocyanins reduce intramolecular forces between biopolymer molecules and increase the irregularity of the biopolymer structure, thereby reducing the TS value (San Lee, Soloi, and How 2021). However, when the anthocyanin was added in amounts from 6 to 10 mL, the TS value increased from 0.67 to 1.20. Conversely, It was caused by the abundance of hydroxyl groups in anthocyanin which could form hydrogen bonds with the hydroxyl groups in starch, resulting in stronger interfacial adhesion between starch and anthocyanin (Yun et al. 2019). The elongation of the edible film increased from 129.87 to 178.80 with the addition of 2–8 mL of anthocyanin. Anthocyanins increase film compatibility, which could increase extensibility (Ge et al. 2020). Similar changes in TS and E values were found when anthocyanins from *Lycium ruthenicum* Murr were added to packaging films made from cassava starch (Qin et al. 2019). According to research by Ge et al. (2020), the TS value of oxidized chitin nanocrystalline nanocomposite films with the addition of anthocyanin from black rice bran decreased and the ΔE value increased with the addition of anthocyanin. Therefore, the mechanical properties of starch films with the addition of anthocyanins could be related to their properties. However, there was a subsequent decrease in EAB from 178,80 (MC 8) to 159.60 (MC 10). Plummeted in elongation value was probably caused by the formation of strong hydrogen bonds between starch and anthocyanin when 10 mL of anthocyanin was added to the film (Qin et al. 2019). The table showed that the WVTr value experienced fluctuations with varying anthocyanin concentrations. Specifically, there was an increase of 0.32 ± 0.14bc and 0.31 ± 0.41b when adding 4 and 8 mL of anthocyanin, respectively. However, there was a decrease of 0.12 ± 0.11a, 0.19 ± 0.19a, and 0.21 ± 0.37ab when adding 2, 6, and 10 mL of anthocyanin, respectively. The increase in the WVTr value was attributed to the formation of anthocyanin aggregates, which produce more free volume in the film network, thereby facilitating moisture transfer (Yun et al. 2019). The decrease in the WVTr value is caused by intermolecular interactions between the anthocyanin and the film, which reduce the film's affinity for water vapor (Wang et al. 2022). As a result, the modified edible film with the addition of 10 mL of anthocyanin exhibited a lower WVTr value, indicating its strong moisture-protective properties, making it suitable for applications requiring extended shelf life (Ebrahimi et al. 2016).

3.6 | Application of Intelligent Films

In order to analyze the differences in physical properties, antioxidant activity, and biodegradation rates among the various formulations of edible films, a one-way ANOVA test was employed. This statistical method was chosen to determine whether there were significant differences between multiple groups, such as different modified starch formulations and varying concentrations of anthocyanin, at different time intervals. The ANOVA analysis provided insight into the influence of these factors on the performance and functionality of the developed edible films. As seen in Figure 5b, the ΔE values progressively increase with the addition of anthocyanin, highlighting the film's responsiveness to pH changes due to fish spoilage. Table 3 and Figure 1 indicate the initial color of the edible film for all samples, with a color difference between unmodified and modified cassava starch edible film, as well as a change in film color when anthocyanin extract is added, which turns the film purple. Color changes in native starch edible film emerged in the first 24 h with a value of 45.65 and then plummeted after 24 h to 43.87, which was caused by the color change of the native starch film from white into blackish white. This is because films containing native starch have poor performance at keeping moisture from outside the film, as seen in the physical parameters of the film in Table 2. The poor moisture barrier properties between the fish meat and the surrounding environment will cause fish meat to start to spoil rapidly (Niranjana Prabhu and Prashantha 2018). This is also corroborated by concrete evidence that the film was overrun with white mold in the first 24 h and darkened

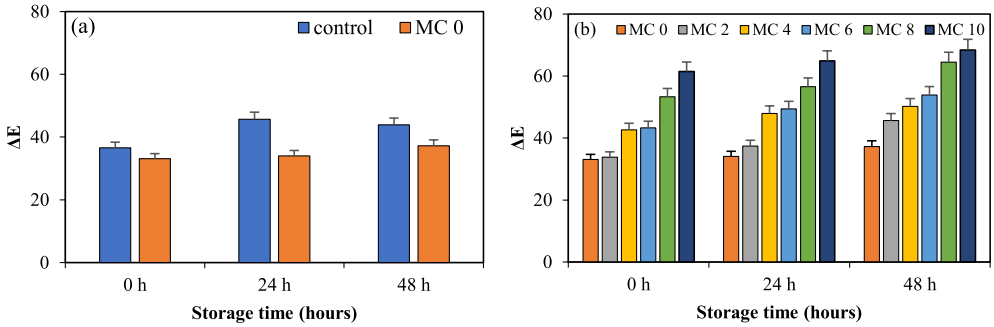

FIGURE 5 | Comparison of ΔE index mean values of fish meat covered with (a) unmodified and modified cassava starch edible films, and (b) variety of anthocyanin levels in modified cassava starch.

TABLE 3 | Initial color and appearance of all edible films.

Sample	Color parameter			Film appearance
	L	A	b	
Control	70.5 ± 0.51^{c}	1.6 ± 0.32^{a}	5.8 ± 0.44^{a}	
MC 0	74.5 ± 0.54^{c}	1.5 ± 0.41^{a}	7.4 ± 0.46^{b}	
MC 2	69.7 ± 0.75^{bc}	1.6 ± 0.34^{a}	5.8 ± 0.41^{a}	
MC 4	70.5 ± 0.43^{c}	4.8 ± 0.42^{ab}	7.5 ± 0.53^{b}	
MC 6	45.5 ± 0.54^{a}	15.7 ± 0.37^{c}	11 ± 0.32^{c}	
MC 8	52.5 ± 0.61^{b}	14.3 ± 0.33^{c}	9.8 ± 0.37^{c}	
MC 10	69.7 ± 0.66^{bc}	8.4 ± 0.46^{b}	4.2 ± 0.41^{a}	

Note: Number of trials = 3. Values quoted are mean values ± standard deviations. $^{a-c}$Within a column, significant differences ($p < 0.05$) for edible films.

after that time. Films made from modified cassava starch, on the other hand, may survive moisture better than native starch edible films. Films with modified cassava starch also saw a steady color shift (in the ΔE range of 33.07–37.24), indicating that the film was more resistant to film storage, even though the film with modified starch began to grow mold after 48 h. Films containing anthocyanin extract from PSP exhibit noticeable color changes, as seen by variations in ΔE values that tend to ascend, as shown in Figure 5b. The film color shifts from purple to greenish purple, followed by E shifts to 45.61, 50.22, 53.91, 64.45, and 68.43 for MC 2, 4, 6, 8, and 10, respectively. The alteration in acidity exhibited in Figure 6 suggests that fish spoilage releases alkaline compounds, which cause the pH to increase from 7 to 8.5. The pH changes from 7.0 to 8.5 are consistent with the color variations in fish meat covered with anthocyanin edible film, which changes from purple to blackish purple.

This finding is confirmed by the color variation of anthocyanin extract in Figure 8a, where the hue of the anthocyanin extract turns blackish purple between pH 8 and 9.

Significant differences ($p < 0.05$) were observed between the treatments at 24 and 48 h, as shown in Figure 6. For TVB-N value from this experiment, two different variables had TVB-N values of 7.07–8.05 mg/100 g at beginning, but there was a change in TVB-N after 24–48 h. Fish flesh coated with edible film resists spoiling, resulting in low nitrogen levels released of up to 46.39 mg/100 g compared to fish meat not coated with edible film, which reaches 71.75 mg/100 g after 48 h. The chemical compound nitrogen is assumed to be present in alkaline chemicals generated by meat during deterioration, and its value is determined using TVB-N parameters (Esfahani et al. 2022). The chemical compounds released were created by decomposition of protein into high-level TVB-N components such as ammonia, dimethylammonium, and trimethylamine (Liu et al. 2018). The TVB-N value was computed and is shown in Figure 6, which demonstrates a rise in TVB-N values due to fish decay, although there is a substantial difference in TVB-N values between fish maintained openly and fish coated with edible film. These results are consistent with color changes that occur in edible films with modified starch or with the addition of anthocyanin extracts. Anthocyanins were added to indicate discoloration caused by increases in pH level caused by food decomposition, in this case, was the spoiling of Indian mackerel fish. These color shifting of edible film were related to the properties of anthocyanin molecules in different pH of the solution (Choi et al. 2017). According to the research's results, smart edible films consisting of modified cassava starch and anthocyanin extract from PSP are excellent for real-time monitoring of fish freshness via color change. Color shifts from film, on the other hand, are relatively difficult to detect, especially for persons with minor color retention abnormalities such as deuteranomaly or tritanomaly, as well as those with other visual impairments. This is assumed to be related to an excess amount of anthocyanins, which cause the film to slow down color changes during degradation (Jiang et al. 2020). To address this challenge, future studies could explore optimizing anthocyanin concentrations or integrating digital sensors for more precise monitoring of color changes, ensuring accessibility for all users.

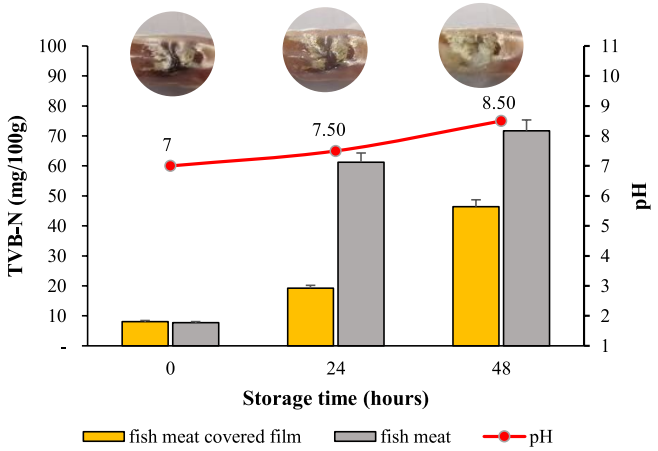

FIGURE 6 | Acidity and TVB-N values of stored Indian mackerel fish with/without edible film (MC 10) cover within 48 h at room temperature (25°C).

3.7 | Antioxidant Level of Intelligent Films

The DPPH test was performed to assess the antioxidant capacity of edible film containing anthocyanin from PSP. As demonstrated in Table 4, antioxidant capacity of modified cassava starch based edible film fortified with anthocyanin increased significantly. The enhanced antioxidant capacity of the films is mostly due to the inclusion of anthocyanin, which has potential antioxidant properties (Yun et al. 2019). Anthocyanins offer a variety of functions, including antioxidant, antitumor, antimutagenic, and cardiovascular disease prevention (Li et al. 2019a). These characteristics were supported by the chemical component anthocyanin, which is a group of naturally occurring phenolic compounds known as phenolic flavonoids (with chemical structure C_6-C_3-C_6). Anthocyanin's structure contains phenolic hydroxyl groups, which may donate free electrons or hydrogen atoms to reactive free radicals, thereby quenching free radicals (Contreras-Lopez et al. 2014). Adding anthocyanins to the films not only increases their antioxidant capability but also improves their ability to be used in food preservation by reducing oxidative damage in packaged foods. This characteristic is especially beneficial for prolonging the freshness of perishable items, providing a natural substitute for artificial antioxidants.

3.8 | Color Response in Anthocyanin With pH Changes

Color changes in PSP anthocyanin extracts need to be investigated in order to validate the usage of anthocyanins as pH change monitors. Sensitivity of anthocyanin extract to pH changes is crucial during its role as pH indicator in edible film, providing a visual signal of freshness or spoilage in food products. Figure 7a depicts the apparent color shift of anthocyanins as pH varies from 2 to 12. Variation results show a shift in anthocyanin color from strong purple to green at pH 2 to 5, then purple discoloration at pH above 5, and afterward green at pH 11 and 12. The UV–vis spectral analysis shown in Figure 7b demonstrates that anthocyanin spectra at pH 2 and 3 have an absorption peak at 525 nm. The anthocyanin spectra then showed a shift in absorption peaks at pH 5 and 6–545 nm. After the pH rises above 7, the absorbance peak shifts back to between 575 and 600 nm (Choi et al. 2017). Shifting that happens in anthocyanins as acidity/pH conditions change causes the greatest peak of absorption

TABLE 4 | Antioxidant level of edible film with the addition of anthocyanin extract.

Sample	Inhibition (%)
MC 0	9.45 ± 0.45^{a}
MC 2	30.88 ± 0.21^{b}
MC 4	55.95 ± 0.12^{b}
MC 6	59.52 ± 0.41^{b}
MC 8	70.76 ± 0.92^{c}
MC 10	79.54 ± 0.64^{c}

Note: Number of trials = 3. Values quoted are mean values ± standard deviations.
[a–c]Within a column, significant differences ($p < 0.05$) for edible films.

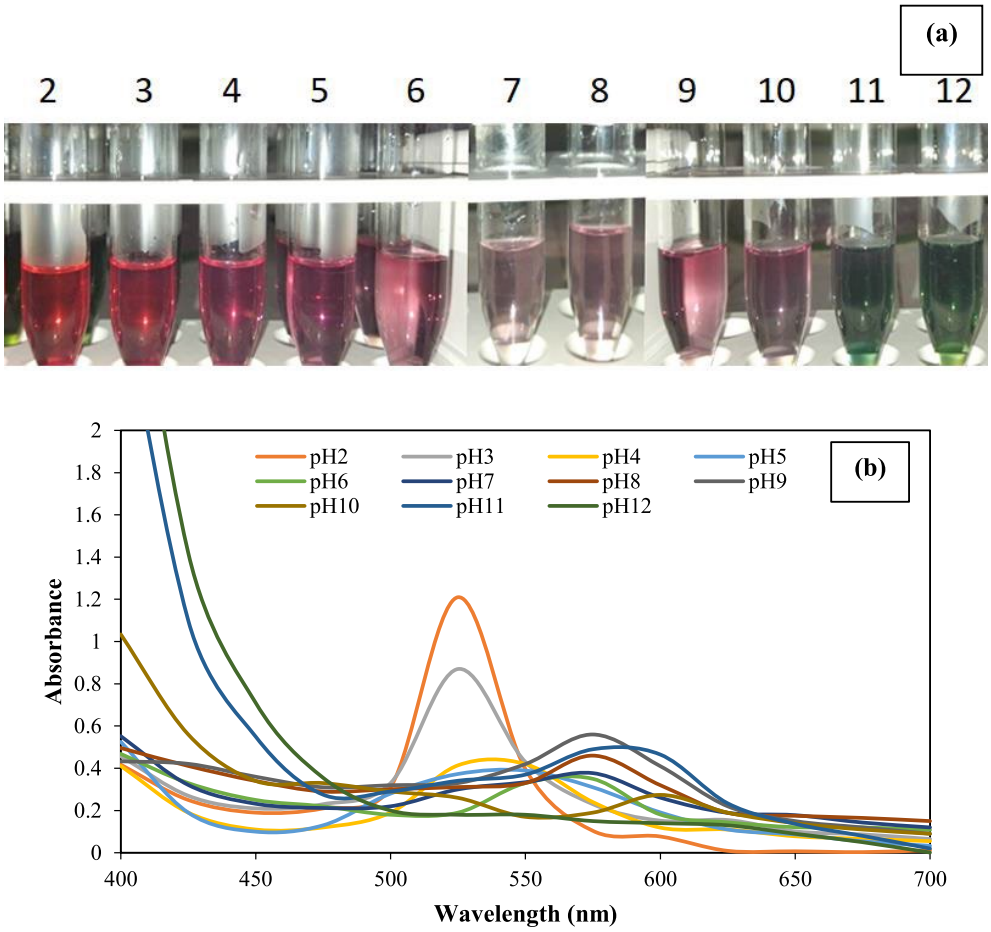

FIGURE 7 | (a) Color variations of anthocyanin extract from PSP in range of pH 2 to 12, and (b) UV–vis spectra of anthocyanin extract from PSP at different pH.

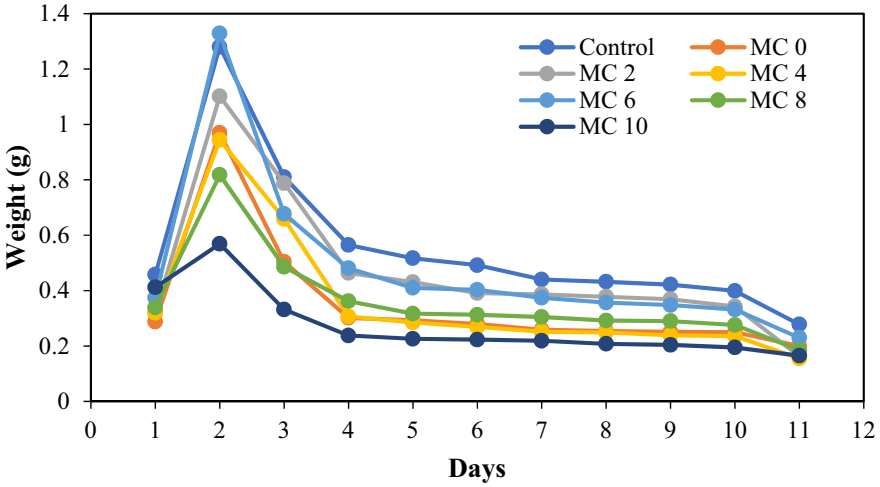

FIGURE 8 | Mass degradation of edible films during storage time.

to alter, indicating a phenomena known as bathochromic shift (Bąkowska, Kucharska, and Oszmiański 2003). Such changes in absorption peaks at different pH levels can be particularly useful in detecting changes in pH that occur during the decomposition of perishable foods (He et al. 2015). Flavylium cation, quinoidal base, carbinol base, and chalcone will take place at lower pH levels as changes in anthocyanin structure in acidic conditions. On the other hand, at alkaline pH solutions, anthocyanin will shift toward anhydro bases. Further alkalinity conditions will degrade anthocyanin (Grajeda-Iglesias et al. 2017).

3.9 | Biodegradable Properties of Film

The biodegradation analysis of edible film made from cassava starch, whether modified or not with cross-linking and with

anthocyanin added, aims to determine the decomposition time of the edible film when it is disposed of into the environment. The biodegradation analysis process was carried out by burying it in natural soil with humus characteristics and then observing mass changes in the edible film for about 11 days (Rahmatullah, Putri, et al. 2022). Figure 8 indicates mass degradation from day 1 to day 11, with rate of decomposition of 0.11 g/day for the control edible film, and 0.085 g/day for the modified edible film. It has been shown that film with modified starch has slow degradation percentage as compared to the control film, which can be attributed to the crosslinked structure formed. This indicates that the modification of starch with STPP has an effect on biodegradation because phosphate groups will interact with hydroxyl groups in starch, resulting in enhanced durability of the film that will prevent water from entering into tissues and inhibiting biodegradation (Detduangchan, Sridach, and Wittaya 2014). However, the mass degradation of the edible film on the second day increased due to the dampness of the soil, which caused the soil to stick to the edible film. Then, from the 3rd day until the 11th day, there was a decrease in mass for each edible film variable.

On the other hand, it can be seen that the mass degradation value of the edible film that was added by anthocyanin has a lower rate of decomposition. For MC 2, 4, 6, 8, and 10 consecutively, it decomposed with rate of decomposition of 0.094, 0.087, 0.087, 0.07, and 0.045 g/day. Films with added anthocyanin showed a further decrease in mass loss, suggesting that anthocyanin forms hydrogen bonds with the starch structure, further reducing the film's affinity for water vapor and limiting its biodegradation (Wang et al. 2022). These findings demonstrate the use of modified cassava starch films containing anthocyanin in applications requiring controlled breakdown. The greater resistance to moisture and biodegradation may extend the shelf life of the packaging material.

4 | Conclusion

Edible films have gained interest due to their ability to extend food shelf life and detect freshness. This study developed a smart edible film using modified starch through cross-linking and stearic acid acylation, aimed at enhancing molecular structure and physical properties. The film includes natural anthocyanin for freshness detection through color changes and added antioxidant properties, while glycerol and alginate act as plasticizers. Modified starch films displayed improved TS, increasing from 0.96 to 1.02 MPa, and with 2 mL anthocyanin addition, TS rose to 1.20 MPa. While anthocyanin slightly weakened some physical properties, EAB and WVTr improved. SEM, FTIR, and DSC analyses confirmed better molecular structure and thermal stability due to tight binding from the cross-linking and acylation processes. The films effectively indicated spoilage in fish through pH-responsive color changes due to TVB-N release. Antioxidant testing (DPPH) showed improved inhibition rates (30.88% to 55.95%) with added anthocyanin. Despite the enhancements, the film remains biodegradable, decomposing in local Semarang soil, offering environmental benefits for commercialization. This smart film holds potential in the food industry for reducing plastic waste while providing freshness detection for consumers.

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Conflicts of Interest

There are no conflicts of interest to declare.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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