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Preparation and characterization of starch-based pH indicator films for anthocyanin release

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Abstract

Perishable foods such as fish, chicken, and milk products lose their characteristic freshness over time, signalling the onset of deterioration. pH-sensitive smart indicator films are practical tools for quality control and delivery of visual information about the freshness of food through colour changes. In this study, *Brassica oleracea* (commonly known as red cabbage) and sago starch (SS) were used to synthesise pH-sensitive films plasticized with glycerol. Sago starch is cheap, abundant, and easily available whereas red cabbage is an excellent source of food colourants. Different concentrations of anthocyanin extracted from red cabbage (RCA) at 8%, 12%, and 16% (w/v) were incorporated into the starch films during fabrication using the solvent casting method. Colour analysis of the films towards food simulants (e.g., 3% acetic acid and 10% ethanol) was conducted. Furthermore, the structural and barrier properties of the films were investigated. Results showed that the addition of RCA increased the water vapour permeability (WVP) by 24% (8% RCA film), whereas the crystallinity index was reduced by 74% (16% RCA film) when compared to the control starch film. Likewise, the concentration of anthocyanins released in 3% acetic acid at room temperature was comparatively lower than the fridge temperature by 0.6% (16% RCA film). Hence, temperature plays an important role in controlling anthocyanin release from the starch matrix, although this depends on the type of food simulant. Hence, the natural pH indicator with visible colour changes is a simple, safe, and cost-effective tool for monitoring perishable foods.

1. Introduction

Food waste can be prevented using intelligent packaging that provides information about the quality of packaged food. Among the abundant natural plant-based pH colourimetric indicators, red cabbage (*Brassica oleracea*) is the most suitable for assessing food freshness and spoilage (Janjarasskul and Suppakul, 2018) due to its high sensitivity towards pH changes. Since food spoilage causes changes in pH, a colourimetric indicator can be used to monitor the condition of packaged food. The utilization of polysaccharides especially starch manufacturing intelligent packaging is due to their abundance sources, low cost, high stability, long-term storage, and adaptability to different storage settings. Starch-based films have similar physical characteristics to synthetic polymers such as translucence, blandness, odourlessness, semi-permeability to carbon dioxide, and lack of permeability to oxygen.

Sago palm (*Metroxylon sagu*) is one of many potentially unexploited crops that thrive in Sarawak and Johor (Azmi *et al*., 2017). Much research has been conducted on sago starch due to its sustainability and high survival rate. The tree can survive where other crops typically fail, including in swampy, acidic, peat, saline, and waterlogged soils (Zhu, 2019). It is interesting to note that the sago palm tree is highly tolerant to adverse conditions including fires, floods, strong winds and drought (Ehara *et al*., 2018). Sago palm is cultivated abundantly in Sarawak, the eastern state of Malaysia. Even though sago starch is produced at a low cost, the yield is higher than other starchy crops like maize, rice and cassava (Rasyid *et al*., 2020). The potential supply of sago starch is substantial given that each sago palm tree could potentially produce over 400 kg of dried starch at maturity which is comparable to

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potatoes (Zhu, 2019).

Red cabbage (*Brassica oleracea*) is a familiar crop widely cultivated around the world, although it is largely found in China, India and Europe (Sarkar and Rakshit, 2021). In the year 2021, China produced 34 million tons of cabbage accounting for 61% of the world's total production, which makes the country the largest producer worldwide. India, the second-largest producer, produced 9.5 million tons followed by Russia (2.3 million tons) in third place, accounting for 4% of the total. Indonesia and Vietnam are the seventh and tenth-largest cabbage producers in the world (adapted from FAOSTAT). Red cabbage is underutilized in Malaysia, where it is commonly used in coleslaw and salad (Muhamad *et al*., 2023). However, the waste can be used for non-food applications such as for the food packaging industry.

According to Crevel (2016) report, food packaging solutions are essential to meeting the Sustainable Development Goals (SDGs). Hence, the bioplastics market is getting higher demand with the increasing demand to sustain food supply and food security. Figure 1 shows the global production capacities of bioplastics in 2022. Biodegradable films derived from cellulose and starches account for 3.6% and 17.9%, respectively, of the total production of 51.5%. The data also shows that biodegradable plastics (bioplastics) are becoming more dominant in the market. Sago starch has been studied extensively in Malaysia for various purposes including smart packaging, prebiotics production, and composite film formulation. Other notable applications include nanoparticle formation, lactic acid production and pharmaceutical/medical applications. Lastly, applications of sago starch have also been found in electrolytes, food thickening, aquafeed ingredients, and ceramic foam fillers (Ahmad *et al*., 2020).

In this work, the structural and barrier properties of pH indicator sago starch films incorporated with red cabbage anthocyanin (RCA) were studied. It also examined the impact of various anthocyanin concentrations released into food simulants from the starch matrix.

2. Materials and methods

2.1 Materials

In this study, the food-grade sago starch was purchased from a local supplier in Bintulu (Sarawak, Malaysia). The red cabbage powder was provided by Hana Natural Company (Penang, Malaysia). The ethanol (absolute) and glycerine were supplied by HmbG Chemicals (Hamburg, Germany), whereas the acetic acid was provided by EMSURE.

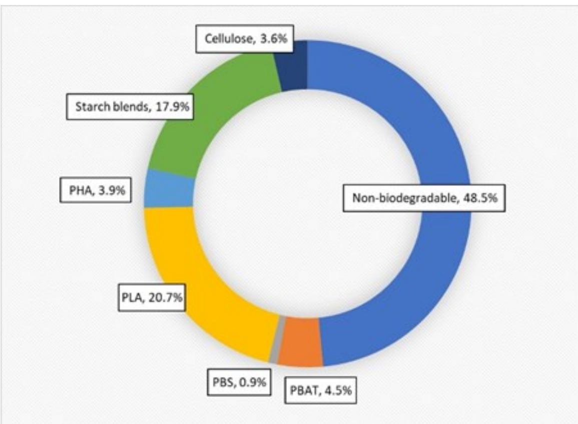
2.2 Film fabrication

The solvent-casting method was used for film fabrication in this study. Firstly, the red cabbage extract (RCE) was prepared at three different weights (8, 12 and 16 g). The RCE was stirred and dissolved in 100 mL of distilled water to obtain a homogeneous solution. The solution was then heated under mild boiling before adding 5 g of sago starch and 3 mL of glycerine into the solution. The solution was stirred and heated continuously to 75°C to obtain a gelatinized and viscous solution. Lastly, the films were cast onto square plates (20 × 20 cm). The casting plate was placed for 3 hrs in an oven (Memmert, Germany) set at 60°C. The control film was prepared without adding RCE to the film solution. The synthesized films were peeled off and stored in a dry desiccator before further analysis.

2.3 Characterization

2.3.1 Physical properties and water vapour permeability

The film thickness was measured using a digital micrometre with 0.001 mm resolution (Insize, model IP54, Vietnam). The density (expressed in mass per unit volume) analysis was evaluated to determine the compactness of the RCA extract in the starch matrix. It was assessed by splitting the samples into 20 × 20 mm squares and measuring the thickness of the films (3



Source	Name	Abbreviation
Biodegradable	PBS	Polybutylene succinate
	PBAT	Polybutyrate adipate terephtalate
	PLA	Poly(lactic acid)
	PHA	Polyhydroxyalkanoate
Non-biodegradable	PE	Polyethylene
	PET	Poly(ethylene terephthalate)
	PA	Polyamide
	PP	Polypropylene

Figure 1. Global production capacities of bioplastics 2022 (adapted from European Bioplastics).

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random measurements). The density of each film was measured as the proportion between the weight and volume (thickness × area) of the film.

The water vapour permeability test was used to evaluate the water vapour transfer through the samples (semi-permeable/permeable) based on ASTM method E96-00 (ASTM, 2002) with some modifications. A container was used to determine the water vapour permeability. The container surface area was determined before placing distilled water inside with a small gap (3 to 4 cm) of air space between the specimen and the water. Next, each film sample was tightly sealed over a circular opening of the container using parafilm strips and cellophane tape. The weight of the sealed container was measured using a measuring balance before heating it at 38°C in the incubator. The water vapour transport was determined by taking the initial weight of the apparatus and then periodically weighed over time until the results become linear. The water vapour transmission rate (WVTR) and water vapour permeability (WVP) were calculated as shown in the equation:

$$WVTR = \mathrm{Q/At} \tag{1}$$

$$WVTR = (WVTR \times l)/\Delta \mathrm{P} \tag{2}$$

Where Q = net weight (g), A = sample permeation area (m^2), T = time (day), L = mean sample thickness (m) and ΔP = difference in water vapour pressure through the sample for pure water

2.3.2 Swelling index

The gravimetric approach proposed by Vo *et al.* (2019) was employed to determine the swelling index (SI) based on Equation 3.

$$\mathrm{SI} = \frac{\mathrm{m_1 - m_0}}{\mathrm{m_0}} \times 100 \tag{3}$$

Where SI stands for the swelling index, which represents the amount of water that the film has absorbed, whereas m_0 and m_1 represent the weights (g) of the film at both the start and the time t in question, respectively.

2.3.3 X-ray diffraction

X-ray diffractometer D8 Advance (Bruker AXS GmbH, Germany) was used to measure the film's X-ray diffraction (XRD) pattern. The XRD technique was applied to determine the crystalline character of the starch film and starch incorporated with RCE films to the reference. The target radiation was CuKα operating at a 30 kV voltage, and a current of 15 mA. The XRD spectra were recorded at 2θ from 5 to 70° at a scanning speed of 1°/min and a step size of 0.02°. The crystallinity index (CI) is the area of all the crystalline peaks divided by the area of all the crystalline and amorphous peaks. The (CI) of the biofilms were calculated according to Equation 4 (Moradi *et al.*, 2019):

$$\text{Crystallinity Index (CI)} = (\mathrm{I_{002}} - \mathrm{I}_{am})/\mathrm{I_{002}} \times 100 \tag{4}$$

Where I_{max} is the maximum intensity of the largest crystal plane reflection of the films and I_{am} is the intensity of X- ray scattering broad band due to the amorphous part of the sample (diffraction intensity at 2θ = 18°).

2.4 Release study

The release study was conducted to determine the amount of anthocyanin released from the starch matrix into the food simulants. Due to the complexity of food matrices, food simulants that mimic the physicochemical characteristics of foods are used as an alternative in migration assays. In this study, 3% (v/v) acetic acid was utilized to simulate acidic food, while 10% (v/v) ethanol represented aqueous/acidic food. The prepared films were cut into 10 × 2 cm and immersed in 30 mL food simulants for one hour at 4°C and at 30°C before UV-vis analysis.

3. Results and discussion

3.1 Characterization

3.1.1 Physical properties and water vapor permeability

Film thickness is a crucial factor that directly influences mechanical strength, water vapour permeability, light transmittance, and opacity (Wang *et al.*, 2019). The addition of RCA has a significant effect on the films' thickness as it is directly proportional to the amount of anthocyanin, as shown in Table 1. A significant enhancement in film thickness was observed because of the high concentration of the solid loadings in the RCA, which created a heterogeneous film structure. It was reported that the thickness of a film manufactured from butterfly peas was unaffected by modifying the amount of extract (Rawdkuen *et al.*, 2020). However, the anthocyanin extract of torch ginger (TGE) was significantly influenced by the higher concentrations of TGE incorporated into the film resulting in highly thick films. The density of the films also increased albeit slightly with an increase in anthocyanin content, which agrees with the results obtained by Xue Mei *et al.* (2020).

Table 1. Composition of RCE anthocyanin in each film.

Films	Ratio starch to RCE	Starch (g)	Glycerine (g)	RCE (g)
Control	-	5	3	0
8% RCA	1: 1.6	5	3	8
12% RCA	1: 2.4	5	3	12
16% RCA	1: 3.2	5	3	16

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WVP is a significant and crucial indicator of the applicability of food packaging. One of the main purposes of food packaging is to prevent or reduce moisture transfer between the food and the environment. The addition of RCA extracts significantly affected the WVP values of the starch films (Table 2). WVP values of the starch film (control) were from 6.934×10^{-5} g.mm/mm^2.day.kPa, while the WVP of the 8% RCA film was 9.158×10^{-5} g.mm/mm^2.day.kPa.

The control film (without RCA) exhibited the lowest WVP, while the highest was observed in the film with 8% RCA ($p<0.05$). The higher WVP of the RCA-containing films may be attributed to the increase in the free volume space of the starch films due to the addition of anthocyanin or RCA extracts. The films with higher anthocyanin content showed lower WVP values, possibly due to the intramolecular and intermolecular interactions between the RCA extract, starch, and glycerine which reduced the gap in the film. The results suggest that the diverse chemical structures of the RCA extracts may have interacted differently with the starch molecule. Likewise, the bulky aromatic rings of the anthocyanins could obstruct the inner networks of RCA films and reduce the water vapour affinity of films (Ge *et al*., 2020). According to these results, it can be concluded that the incorporation of RCA extracts improved the water barrier properties of the starch films. The lower WVP was achieved with a higher concentration of anthocyanin, which created a denser and more compact network to inhibit moisture transfer.

3.1.2 Swelling index

The SI is a measure or indicator of the effectiveness of stored water absorption. It has a substantial impact on how well smart pH-sensitive indicators respond to colour since a greater swelling index rapidly releases the dyes (Wei *et al*., 2017). Figure 2 (a) and (b) shows the SI of different RCA concentrations incorporated in the starch films and the final SI at the 20th minute of the swelling test respectively. The swelling degree of 8% RCA film is significantly different from other films. Once immersed in the distilled water, the film starts to swell and continues to the maximum level compared to others. Other films initially showed lower SI before increasing to as high as 30% and the equilibrium state. It was reported that a higher weight fraction of solids decreases the SI of films (Muhamad *et al*., 2011). The films with 12% and 16% RCA contain a higher weight fraction of solid than films with 8% RCA, which explains the lower IS values. The control film also shows the swelling process occurred only once after the immersion and the values were maintained for 20 mins.

The swelling properties of starch were different from chitosan as a smart indicator film base medium. When the anthocyanin concentration increased, its polyphenolic compounds weakened the intermolecular connections in chitosan, which resulted in the loss of network cohesiveness and increased swelling characteristics (Tirtashi *et al*., 2019). However, the incorporation of anthocyanin in the starch formulation decreased the swelling degree of the films as the concentration increased. This result can be explained by the different

Table 2. Physical and WVP properties of films.

Films	Thickness (mm)	Density (g/m^3)	WVP ($\times 10^{-5}$ g.mm/mm^2.day.kPa)
Control	0.124±0.039^a	0.010±0.005^a	6.934±0.789^a
8% RCA	0.169±0.013^b	0.012±0.004^b	9.158±0.331^b
12% RCA	0.215±0.022^c	0.013±0.002^{bc}	8.808±1.437^{bc}
16% RCA	0.231±0.008^{cd}	0.013±0.001^{bcd}	8.694±0.193^{bcd}

Values are presented as mean±SD. Values with different superscript within the same column are statistically significantly different between samples ($p<0.05$).

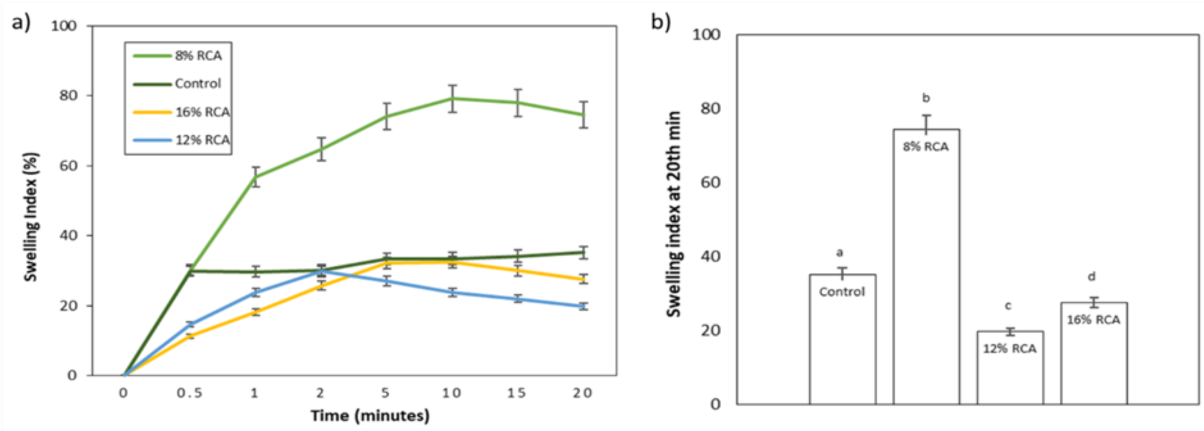

Figure 2. a) Swelling index over time, b) swelling index at 20 mins for control and RCA films. Bars with different notations are statistically significantly different at $p<0.05$ (Tukey's Test).

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hydrophilic properties of starch and chitosan that affect the swelling degree of both biofilms. Likewise, the higher RCA content reduced the swelling degree of the starch films, which also prevented rapid migration of the anthocyanin from the starch matrix (Issa *et al*., 2017).

3.1.3 X-ray diffraction

XRD analyses were performed to assess the crystallinity or amorphous nature of the biofilms. The XRD spectra of starch (control) and RCA blend films (8%, 12%, and 16%) are shown in Figure 3, whereas the crystallinity Index (CI) is presented in Table 3. The CI was used to examine the structural changes and degree of crystallinity in the starch film (control) and RCA-containing films.

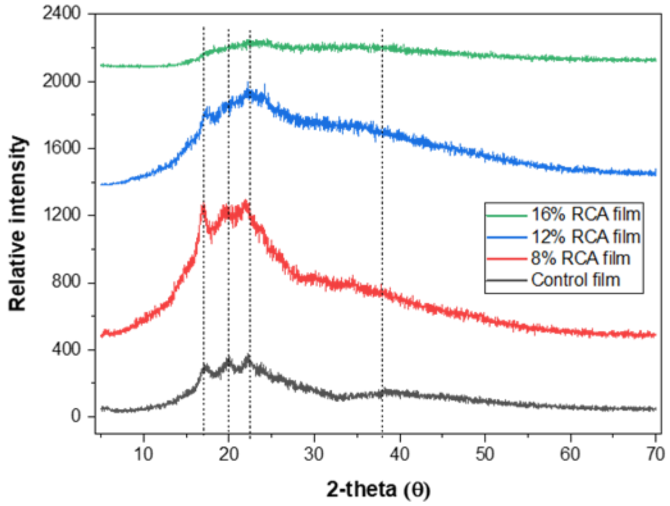

Figure 3. XRD spectra of control and RCA films (8%, 12%, and 16%).

Table 3. Crystallinity index of the starch film (control) and RCA-containing films.

Films	Crystallinity Index (%)
Control	58.01
8% RCA	53.12
12% RCA	36.12
16% RCA	15.08

The starch film (control) showed three strong diffraction peaks at 17.2°, 20.0°, and 22.3° which corresponds to the C-type diffraction spectrum of native sago starch, as similarly in Adawiyah *et al*. (2013). During the gelatinization process, the semi-crystalline structure of the original starch granules was completely dissolved, resulting in the amorphous form of the starch film (Pelissari *et al*., 2018). The 8% RCA film showed similar amorphous patterns to the control film but with stronger peak intensities. The enhancement in the peak intensity of starch-RCA films is probably because the hydrogen bonds formed between starch and RCA resulted in a strong three-dimensional network (Wu *et al*., 2009). However, the higher RCA concentrations reduced the crystallinity of the films as evidenced by the significantly reduced peak intensities. This indicated that there was less rearrangement of anthocyanin molecules to crystallization because the intermolecular hydrogen bond formed between RCA and starch interactions resulted in a semi-crystalline structure (Pourjavaher *et al*., 2017).

In addition, the diffraction peaks of the films shifted slightly after the incorporation of RCA in the 12% RCA film. The film with the highest RCA concentration (16%) became highly amorphous as typified by the absence of the characteristic starch granule peak in the sample. The finding indicates that starch was well-dispersed in the film without crystalline structure. This phenomenon most likely occurred because heating and mechanical stirring during gelatinization damaged the crystalline regions of the starch granules (Zhai *et al*., 2017). Likewise, the CI was reduced with the incorporation of RCA in the starch films. Generally, the CI of RCA films decreased drastically when the anthocyanin extract was incorporated (Pourjavaher *et al*., 2017; Wang *et al*., 2019).

3.2 Release study

Release studies have been examined, through the migration of antioxidant compounds in food simulants, to verify the effectiveness of active packaging (Da Costa Monção *et al*., 2022). The release kinetics of the active compound into the food throughout the storage time is, therefore, a crucial factor when guaranteeing antimicrobial effectiveness and food safety (Fernández-Pan *et al*., 2015). In this way, the time needed to reach a concentration level of activity in the food greater than the minimum inhibitory concentration (MIC) must be predicted to ensure food safety. This approach provides useful information about the ability of active packaging to exert an antimicrobial function in foodstuffs (Requena et *al*., 2017). The pigment-holding potential of high RCA concentration films was investigated by determining the release of anthocyanin (coloured pigments) after immersion in several food simulants. Figure 4 shows the concentration of anthocyanin released into the acetic acid and ethanol-based food simulants at different temperatures.

As observed, the concentration of anthocyanins released in 3% acetic acid at room temperature was lower in general compared to 10% ethanol at 30°C and 4°C temperatures. These result contradicts with the findings by Zabihzadeh Khajavi *et al*. (2019), who reported that increasing the temperature, enhances the migration rate of the migrants that reside in the space. The findings of the present study indicate that temperature is an important factor in anthocyanin

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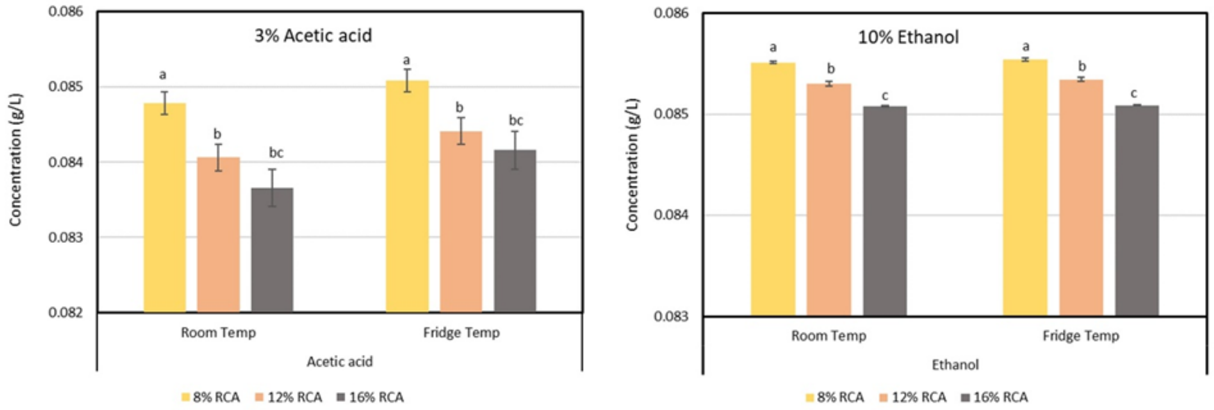

Figure 4. Anthocyanin concentrations released into acetic acid and ethanol food simulants. Bars with different notations are statistically significantly different at $p<0.05$ (Tukey's Test).

immobilization in starch-based films but depends on the type of food simulants. The immobilization of anthocyanin was also longer in 10% ethanol compared to 3% acetic acid as the release concentration was smaller, as shown in Figure 4. Thus, the addition of other nanoparticles such as cellulose nanofiber (CNF) will prolong the immobilization of anthocyanin in the starch films (Mohammadalinejhad *et al*., 2020). Moreover, a higher amount of RCA also contributes to the lower release of anthocyanin (Pourjavaher *et al*., 2017), which is an essential parameter for consumers to receive appropriate visual feedback on smart packaging applications.

4. Conclusion

In this work, RCA was successfully incorporated into the sago starch to synthesize highly sensitive films for different food simulants. It was observed that the starch films released anthocyanin at different rates under various temperature and RCA concentration conditions. The highly amorphous films and slow release of anthocyanin from the starch matrix are important criteria for producing sensitive food indicator films. Similarly, the temperature has minimal effect on anthocyanin release, which makes it suitable at various storage temperatures. The fabricated indicator displayed different structural and barrier properties due to the different RCA concentrations incorporated into the films. The findings showed that the addition of RCA improved the physical performance of the starch-based films for potential application as packaging films.

Conflict of interest

The authors declare no conflict of interest.

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References

Adawiyah, D.R., Sasaki, T. and Kohyama, K. (2013). Characterization of arenga starch in comparison with sago starch. *Carbohydrate Polymers*, 92(2), 2306–2313. https://doi.org/10.1016/j.carbpol.2012.12.014

Azmi, A.S., Malek, M.I.A. and Puad, N.I.M. (2017). A review on acid and enzymatic hydrolyses of sago starch. *International Food Research Journal*, 24, 265–273.

Crevel, R.V. (2016). Bio based food packaging in Sustainable Development. Challenges and opportunities to utilize biomass residues from agriculture and forestry as a feedstock for bio-based food packaging. Rome, Italy: Food and Agriculture Organization of the United Nations (FAO).

Da Costa Monção, É., Grisi, C.V.B., De Moura Fernandes, J., Souza, P.S. and Souza, A.L. (2022). Active packaging for lipid foods and development challenges for marketing. *Food Bioscience*, 45, 101370. https://doi.org/10.1016/j.fbio.2021.101370

Ehara, H., Toyoda, Y. and Johnson, D.V. (Eds.) (2018). Sago palm: Multiple contributions to food security and sustainable livelihoods. In Sago Palm: Multiple Contributions to Food Security and Sustainable Livelihoods. Springer E-Book. https://doi.org/10.1007/978-981-10-5269-9

European Bioplastics. Bioplastics market development update 2023 Retrieved in 2022 from European Bioplastics Website: https://www.european-bioplastics.org/market

Fernández-Pan, I., Maté, J.I., Gardrat, C. and Coma, V. (2015). Effect of chitosan molecular weight on the antimicrobial activity and release rate of carvacrol-enriched films. *Food Hydrocolloids*, 51, 60–68. https://doi.org/10.1016/j.foodhyd.2015.04.033

Issa, A., Ibrahim, S.A. and Tahergorabi, R. (2017). Impact of sweet potato starch-based nanocomposite films activated with thyme essential oil on the shelf-life of baby spinach leaves. *Foods*, 6(6), 43. https://doi.org/10.3390/foods6060043

Janjarasskul, T. and Suppakul, P. (2018). Active and intelligent packaging: The indication of quality and safety. *Critical Reviews in Food Science and Nutrition*, 58(5), 808–831. https://doi.org/10.1080/10408398.2016.1225278

Mohammadalinejhad, S., Almasi, H. and Moradi, M. (2020). Immobilization of *Echium amoenum* anthocyanins into bacterial cellulose film: A novel colourimetric pH indicator for freshness/spoilage monitoring of shrimp. *Food Control*, 113, 107169. https://doi.org/10.1016/j.foodcont.2020.107169

Muhamad, I.I., Khairuddin, N. and Ling, L.H. (2011). Effect of lauric acid addition on the microbial efficacy of chitosan-based film. *Jurnal Teknologi*, 54, 101–109. https://doi.org/10.11113/jt.v54.94

Pelissari, F.M., Ferreira, D.C., Louzada, L.B., Dos Santos, F., Corrêa, A.C., Moreira, F.K.V. and Mattoso, L.H. (2018). Starch-based edible films and coatings: An eco-friendly alternative for food packaging. In Clerici, M.T.P.S. and Schmiele, M. (Eds.) Starches for Food Application: Chemical, Technological and Health Properties. USA: Academic Press. https://doi.org/10.1016/B978-0-12-809440-2.00010-1

Pourjavaher, S., Almasi, H., Meshkini, S., Pirsa, S. and Parandi, E. (2017). Development of a colourimetric pH indicator based on bacterial cellulose nanofibers and red cabbage (*Brassica oleraceae*) extract. *Carbohydrate Polymers*, 156, 193–201. https://doi.org/10.1016/j.carbpol.2016.09.027

Rawdkuen, S., Faseha, A., Benjakul, S. and Kaewprachu, P. (2020). Application of anthocyanin as a colour indicator in gelatin films. *Food Bioscience*, 36, 100603. https://doi.org/10.1016/j.fbio.2020.100603

Requena, R., Vargas, M. and Chiralt, A. (2017). Release kinetics of carvacrol and eugenol from poly (hydroxybutyrate-co-hydroxyvalerate) (PHBV) films for food packaging applications. *European Polymer Journal*, 92, 185–193. https://doi.org/10.1016/j.eurpolymj.2017.05.008

Sarkar, D. and Rakshit, A. (2021). Bio-priming in combination with mineral fertilizer improves the nutritional quality and yield of red cabbage under Middle Gangetic Plains, India. *Scientia Horticulturae*, 283, 110075. https://doi.org/10.1016/j.scienta.2021.110075

Tirtashi, F.E., Moradi, M., Tajik, H., Forough, M., Ezati, P. and Kuswandi, B. (2019). Cellulose/chitosan pH-responsive indicator incorporated with carrot anthocyanins for intelligent food packaging. International Journal of Biological Macromolecules, 136, 920–926. https://doi.org/10.1016/j.ijbiomac.2019.06.148

Vo, T., Dang, T. and Chen, B.-H. (2019). Synthesis of Intelligent pH Indicative Fims from Chitosan / Poly (vinyl alcohol) / Anthocyanin Extracted from Red Cabbage. *Polymers*, 11(7), 1088. https://doi.org/10.3390/polym11071088

Wang, X., Yong, H., Gao, L., Li, L., Jin, M. and Liu, J. (2019). Preparation and characterization of antioxidant and pH-sensitive films based on chitosan and black soybean seed coat extract. *Food Hydrocolloids*, 89, 56–66. https://doi.org/10.1016/j.foodhyd.2018.10.019

Wei, Y.C., Cheng, C.H., Ho, Y.C., Tsai, M.L. and Mi, F.L. (2017). Active gellan gum/purple sweet potato composite films capable of monitoring pH variations. *Food Hydrocolloids*, 69, 491–502. https://doi.org/10.1016/j.foodhyd.2017.03.010

Wu, Y., Geng, F., Chang, P.R., Yu, J. and Ma, X. (2009). Effect of agar on the microstructure and performance of potato starch film. *Carbohydrate Polymers*, 76(2), 299–304. https://doi.org/10.1016/j.carbpol.2008.10.031

Xue Mei, L., Mohammadi Nafchi, A., Ghasemipour, F., Mat Easa, A., Jafarzadeh, S. and Al-Hassan, A.A. (2020). Characterization of pH-sensitive sago starch films enriched with anthocyanin-rich torch ginger extract. *International Journal of Biological Macromolecules*, 164, 4603–4612. https://doi.org/10.1016/j.ijbiomac.2020.09.082

Zabihzadeh Khajavi, M., Mohammadi, R., Ahmadi, S., Farhoodi, M. and Yousefi, M. (2019). Strategies for controlling release of plastic compounds into foodstuffs based on application of nanoparticles and its potential health issues. *Trends in Food Science and Technology*, 90, 1–12. https://doi.org/10.1016/j.tifs.2019.05.009

Zhai, X., Shi, J., Zou, X., Wang, S., Jiang, C., Zhang, J., Huang, X., Zhang, W. and Holmes, M. (2017). Novel colourimetric films based on starch/polyvinyl alcohol incorporated with roselle anthocyanins for fish freshness monitoring. *Food Hydrocolloids*, 69, 308–317. https://doi.org/10.1016/j.foodhyd.2017.02.014

Zhu, F. (2019). Recent advances in modifications and applications of sago starch. *Food Hydrocolloids*, 96, 412–423. https://doi.org/10.1016/j.foodhyd.2019.05.035

