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Original research

Developing a smart colorimetric indicator film by incorporating pomegranate peel powder into cassava starch film

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ABSTRACT

The purpose of this study was to investigate the effect of pomegranate peel powder on the physicochemical, barrier, and pH sensitivity of the films based on cassava starch. The pomegranate peel powder (PPP) was used at levels of 2, 4, 6, and 8% w/v in starch film formulation, and the thickness, water solubility, water vapor (WVP) and oxygen permeability, and opacity were determined, and the color variations of the films were also measured after immersion in different pH buffers (pH 4.0-9.0). The results demonstrated the addition of PPP to film formulation and increasing its levels, the water solubility, opacity, WVP, and oxygen permeability were increased ($p<0.05$), so that the values of these parameters in the control sample were 33.78%, 0.65 mm^{-1}, 6.92×10^{-10} g/s.m.Pa, and 4.06 cc-mil/m^2.Day, respectively, and in the film containing 8% of PPP reached 41.32%, 1.25 mm^{-1}, 7.72×10^{-10} g/s.m.Pa, and 5.63 cc-mil/m^2.Day, respectively. The results of examining the sensitivity of films to pH showed that the color of the produced films changed against changes in solution pH so that with increasing the pH of the solution to the neutral and alkaline range, the color of the films changed from reddish pink to green. According to obtained results, it can be concluded that the intelligent films based on cassava starch containing PPP can be used as pH-sensitive indicators to display the freshness of food products.

Keywords: Intelligent film; pH sensitive; Starch films; Anthocyanin; Barrier properties

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1. Introduction

The application of biopolymer materials obtained from natural sources instead of petroleum materials to produce packaging films is a very suitable and effective solution to reduce the risks of synthetic and plastic packaging and improve the safety of packaged food (Javidi et al., 2022). The application of these natural packaging due to having an environmentally friendly relationship and being recyclable also reduces environmental pollution (Salajegheh et a., 2020). Polysaccharides, lipids, proteins, or combinations of these biopolymers can be used to manufacture biodegradable food packaging (Tongdeesoontorn et al., 2020).

Starch is a major stored polysaccharide, which is naturally found in abundant in different plant sources. Starch can film form, and the films produced from it have a low price and are recyclable, tasteless, odorless, biodegradable, non-toxic, and biocompatible (Cui et al., 2021). Cassava (tapioca) starch is one of the important sources of starch for packaging film preparation because it has a low gelatinization temperature and the gel formed by this starch indicates good stability (Tamimi et al., 2021). However, packaging based on polysaccharides has some disadvantages, such as their high solubility in water and poor barrier properties against water vapor and oxygen. Therefore, to develop their application in food packaging, an attempt is made to overcome these limitations by using appreciative additives and agents (Moosavian et al., 2017).

Spoilage of food products not only causes waste of foods but also raises issues related to consumer safety, so determining the quality of food products during transportation, handling and storage is very important (Huang et al., 2021). Since pH is one of the major chemical factors associated with food spoilage (Singh et al., 2018), pH-sensitive colorimetric packing films based on natural materials display the freshness or spoilage of food products and increase the quality and safety of foods have attracted the attention of various

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researchers (Gasti et al., 2021; Hematian et al., 2022; Oladzadabbasabadi et al., 2022). Colorimetric intelligent packaging consists of two main parts including a solid film matrix and a pH-sensitive pigment (Yang et al., 2021). Since chemical and synthetic dyes are often carcinogenic and their use is not recommended, pigments obtained from natural sources are considered an alternative to synthetic types (Oladzadabbasabadi et al., 2022).

Anthocyanins belong to the flavonoids group and are one of the most important and abundant pigments found in nature that are soluble in water and cause red, purple, or blue colors in various flowers, fruits, and other parts of plants (Azlim et al., 2022). These natural pigments also often indicate remarkable antioxidant and antimicrobial activity, and their color changes against pH changes have been confirmed by researchers (Fernández-Marín et al., 2022; Oliveira Filho et al., 2021; Xue Mei et al., 2020). The color of anthocyanins is greatly influenced by their chemical structure, temperature, ultraviolet irradiation, pH, presence of oxygen, and co-pigmentation (Oladzadabbasabadi et al., 2022).

Pomegranate (*Punica granatum* L.) is one of the oldest known fruits that contain high amounts of functional compounds (Kaderides et al., 2015). This fruit is mainly used in the food industry to prepare pomegranate drinks or fruit juices. Since the yield of pomegranate juice is less than half the weight of the fruit, a large number of by-products such as peel are formed every year, which is often consumed in animal feed (Zahed & Farahmandfar, 2021). Anthocyanins are the most abundant natural pigments in pomegranate peel so these pigments contained 98% of the polyphenols content in pomegranate peel (Azarpazhooh et al., 2019). In previous studies, pomegranate peel extract has been used to develop active films based on different biopolymers, and the functional activity of these films has been reported (Cui et al., 2020; Esfahani et al., 2022; Moghadam et al., 2020; Zeng et al., 2021). Due to the presence of high levels of anthocyanins in pomegranate peel, this study aimed to develop intelligent colorimetric films based on cassava starch by using pomegranate peel powder.

2. Material and Methods

2.1. Materials

Cassava starch was obtained from SIM Supply Company (Penang, Malaysia). Malas variety of pomegranate was purchased from the local market (Saveh, Iran) and its peel was washed and then dried in the shade. Glycerol plasticizer and other chemicals were prepared by Merck Company (Darmstadt, Germany).

2.2. Preparation of intelligent films based on cassava starch and pomegranate peel powder

At first, cassava starch (3% w/v) was added to cold distilled water and dispersed. The mixture was stirred for one min at room temperature (23 ± 2°C) and then the glycerol (at 30% w/v of starch) and different levels of pomegranate peel powder (PPP) (2%, 4%, 6%, and 8% w/w) was incorporated into the starch suspensions. The mixtures were then heated for 25 min on a magnetic stirrer to form the gel. 100 mL of each gel sample (100 mL) was uniformly spread on a Teflon (300µ) and dried for 48 h at room temperature (Chi et al., 2020).

2.3. The thickness measurement

A digital micrometer (QLR IP54, America) was used to determine the thickness of the film samples at five random parts of each film.

2.4. The water solubility measurement

The pieces of the samples were cut and placed in a desiccator containing calcium chloride (0% RH), and in an oven (Memmert, Germany) for 24 h. After that, the films were weighted and mixed with 100 ml of deionized water. The container was covered with aluminum foil and was stirred for 24 h at room temperature (23 ± 2°C). The samples were then filtered using filter paper and dried in the oven (at 40°C) to stabilize the weight. The water solubility of the film samples was obtained through the following equation, where: W_1 was the weight of dry film (g), and W_2 was the weight of swollen film (g) (Maizura et al., 2008).

$$Water\ solubility\ (\%) = \frac{W_2 - W_1}{W_2} \times 100 \qquad (1)$$

2.5. The water vapor permeability (WVP) measurement

The WVP of the film samples was measured using ASTM E96/E96M-16 Standard with some modifications (ASTM, 2016). Initially, the thickness of film samples was measured and fitted to the standard glass cups containing activated silica gel. The cups were covered with Parafilm and placed in a desiccator containing saturated magnesium nitrate (55% RH and 25°C). During one week, the weight of the cups was recorded at certain times. After that, the curve of changes in moisture content was drawn and the line's slope was determined to calculate the rate of water vapor transmission (WVTR) (g/day). The WVP of the films was determined by multiplying the steady state WVTR by the thickness of the film and dividing that by the area of film and the difference in water vapor pressure between the inside and outside of the cups.

2.6. The oxygen permeability measurement

The oxygen permeability of the films was measured by using a Mocon Oxtran 2/21 system (Minneapolis, USA) and the ASTM D3985-17 Standard Method. After conditioning the films at room temperature (23 ± 2°C), 55% RH, and atmospheric pressure for 2 days, their thickness was determined. After that, the films were placed into the equipment diffusion cell, and the oxygen permeability of samples was estimated by using the convergent method and WinPermTM permeability software (ASTM, 2017).

2.7. The opacity determination

Initially, the thickness of the film pieces (dimensions of 4 × 1 cm) was recorded, and the samples were then placed inside the quartz cell of the UV-Vis spectrophotometer (Shimadzu, Japan) and their absorbance was recorded at 600 nm. The opacity of the samples was calculated through the following equation, where: A600 and X are the absorbances at 600 nm, and the thickness of the sample (mm), respectively (Yan et al., 2012).

$$Opacity\ (mm^{-1}) = \frac{A600}{X} \tag{2}$$

2.8. *Assessing the sensitivity of films to pH changes*

The color response of the films was investigated using the method expressed by Yong et al. (2019) with some modifications. Initially, the color indexes of film samples including L* (lightness), a* (red-green), b* (yellow-blue), and ΔE (total color changes) was determined by a colorimeter (ColorFlex, America) and then the films (dimensions of 30 mm × 15 mm) were immersed in buffer solutions with different pHs (pH= 4-9). After removing the buffer solutions, the color indexes of the films were determined at three points randomly. The ΔE was calculated using the following equation, if the ΔE is more than 3.5, a clear difference in the color will be observed (Halász & Csóka, 2018).

2.9. *Statistical analysis*

Statistical analysis of data obtained from the tests was done using IBM SPSS Statistics 22.0 (Chicago, IL, USA). One-way analysis of variance (One-way ANOVA) followed by Duncan multi-range post hoc test was used to compare means at $p < 0.05$ significance level among different samples.

3. Results and Discussion

3.1. *The thickness of the films*

Fig. 1 compares the average thickness values of cassava starch films containing different levels of PPP and shows that despite a slight increase in the thickness of the films due to the increase in the level of PPP, this change was not significant. The average thickness of the film samples was in the range of 0.115 to 0.122 mm.

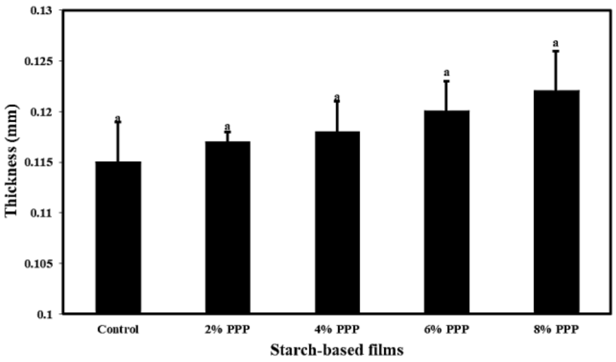

Fig. 1. Comparison the thickness (mm) mean values of cassava starch-based films containing different levels of PPP. Bars represent mean (n = 3) ± *SD*. Different letters on the bars indicate a significant difference at 5% level of probability among film samples. PPP: pomegranate peel powder.

The results of examining the thickness of the cassava starch-based films containing PPP demonstrated that due to the use of a similar production method (solvent casting method) and similar base materials for preparing film samples (starch and glycerol), no significant differences were observed between the thickness of the produced films and the slight increase due to the addition of PPP is probably due to the presence of fibers in this additive and increase in the water absorption capacity. Similarly, Bilgiç et al. (2019) and Andretta et al. (2019) found that by adding eggplant anthocyanin extract and blueberry residue to the starch-based films, no significant change in the film thickness was observed. Emam-Djomeh et al. (2015) and Veiga-Santos et al. (2011) also achieved similar results.

3.2. *The water solubility of the films*

The water solubility values of the cassava starch-based films containing different levels of PPP are compared in Fig. 2. The lowest solubility was for the control sample (33.78%) and by adding PPP at a level of 2%, an increase in water solubility was observed but was not statistically significant. Increasing the level of PPP in the film formulation from 2% to 8% led to a significant increase in the water solubility of the films ($p < 0.05$) so that the highest water solubility value was observed in the film containing the highest level of PPP (41.32%).

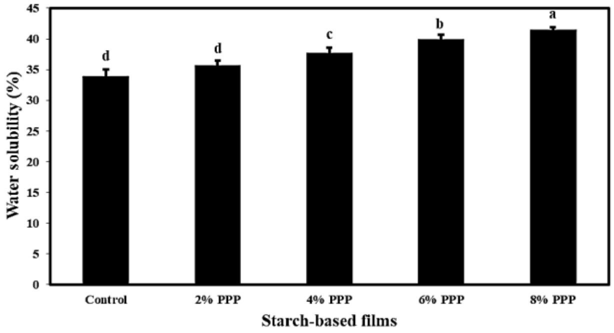

Fig. 2. Comparison the water solubility (%) mean values of cassava starch-based films containing different levels of PPP. Bars represent mean (n = 3) ± *SD*. Different letters on the bars indicate a significant difference at 5% level of probability among film samples. PPP: pomegranate peel powder.

Increasing the level of PPP in the starch film formulation led to an increase in the water solubility of the produced films compared to the control film, which can be due to less reaction between phenolic compounds in pomegranate peel and hydroxyl groups of starch, which leads to the formation of a structure with less compaction and weaker bonds (Choi et al., 2017; de Moraes Crizel et al., 2018). Pomegranate peel also contains fibers that can increase the solubility of the films. Despite the increase in solubility of the films due to the addition of PPP, all the films produced in this study had a relatively low water solubility in general due to strong intermolecular bonds between the starch chains, which prevents the disintegration of the film structure (Kim et al., 2015). Andretta et al. (2019) also showed that the incorporation of the blueberry residue into the cassava starch films caused a significant increase in the solubility of the samples, and these researchers attributed the observed solubility increase to the presence of hydrophilic fibers in this additive. Veiga-Santos et al. (2011) achieved similar results in examining the effect of the grapes and spinach extracts on the water solubility of cassava starch-based films. Emam-Djomeh et al. (2015) and Mehdizadeh et al. (2012) also reported that the solubility of sodium caseinate and chitosan-starch films increased due to the increase in pomegranate peel extract, respectively.

3.3. *The water vapor permeability (WVP) of the films*

Fig. 3 compares the average WVP values of cassava starch-based film containing different levels of PPP and indicates that by adding PPP and increasing its level in the film samples from 2% to 8%, a significant increase in WVP of the samples was observed ($p < 0.05$). So, the WVP of the control sample was 6.92×10^{-10} g/s.m.Pa, and in the film sample containing 8% PPP, the WVP reached 7.72×10^{-10} g/s.m.Pa.

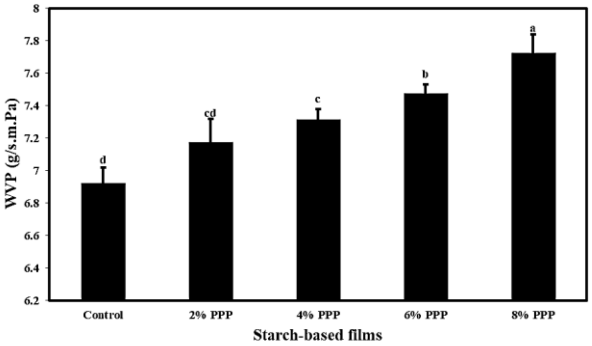

Fig. 3. Comparison the WVP (g/s.m.Pa) mean values of cassava starch-based films containing different levels of PPP. Bars represent mean ($n = 3$) ± *SD*. Different letters on the bars indicate a significant difference at 5% level of probability among film samples. PPP: pomegranate peel powder, WVP: water vapor permeability.

Water vapor permeability (WVP) is one of the key characteristics of starch-based films. This feature demonstrates the ability of packaging film to control the transfer of water vapor between food and the environment. The starch-based films often show high WVP (Bhat et al., 2013). In general, hydrophobic and hydrophilic compounds affect the WVP of the packaging films. The presence of hydrophilic compounds such as phenols in high amounts in pomegranate peel leads to an increase in the WVP of the film samples. Reducing the strength and cohesion of the film structure by adding PPP also makes water molecules pass through the film more easily and thus increases the WVP of the film samples. Due to the different nature of functional additives and the type and amount of bonds in their structure, adding these compounds to packaging films can have a different effect on the WVP. So, Bilgiç et al. (2019) found that the incorporation of aqueous extract of eggplant anthocyanin to the starch film reduced the WVP, while incorporating the ethanolic extract showed a significant increase in the WVP of the film samples. Yun et al. (2019) stated that the presence of high amounts of anthocyanins in the bayberry extract could lead to a lower density of the starch film structure and thus increase the WVP. Increased WVO of sodium caseinate films was observed after adding different levels of the pomegranate peel extract (Emam-Djomeh et al., 2015).

3.4. *The oxygen permeability of the films*

Fig. 4 compares the oxygen permeability values of the cassava starch-based films containing different levels of PPP. As can be seen in the figure, the control sample had the lowest oxygen permeability value (4.06 cc-mil/m^2.Day) and with increasing the level of PPP in the samples, the oxygen permeability increased ($p<0.05$). So in the sample containing the highest level of PPP (8% level), the highest oxygen permeability was observed (5.63 cc-mil/m^2.Day).

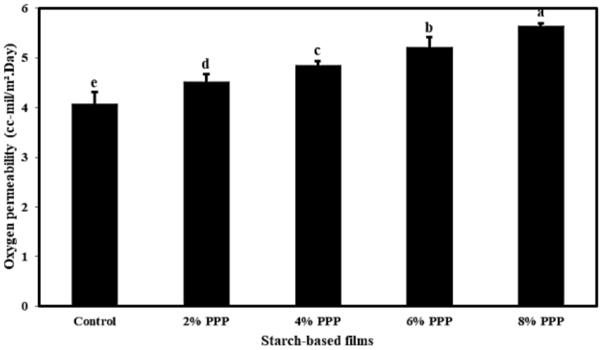

Fig. 4. Comparison the oxygen permeability (cc-mil/m^2.Day) mean values of cassava starch-based films containing different levels of PPP. Bars represent mean ($n = 3$) ± *SD*. Different letters on the bars indicate a significant difference at 5% level of probability among film samples. PPP: pomegranate peel powder.

The oxygen permeability of the biopolymer films is one of the important features for storing packaged food products. Since the presence of high amounts of oxygen leads to increased oxidation of lipids and loss of nutritional quality and organoleptic properties of food products, it is necessary that food packaging has good oxygen barrier characteristics. Increasing the oxygen permeability of starch films due to the addition of PPP in this study is probably related to the lower compaction of the film structure and the creation of more holes between the polymer chains, which facilitates the entry of oxygen into the films. Liu et al. (2018) showed that the addition of low levels of curcumin to the kappa-carrageenan film reduced the oxygen permeability, however, at high levels of it a significant increase in the oxygen permeability was observed. Andretta et al. (2019) also found that the oxygen permeability of cassava starch-based films containing blueberry residues was higher than the control film. Kurek et al. (2014) also reported similar results in investigating the oxygen permeability of the chitosan films containing carvacrol.

3.5. *The opacity of the films*

The effect of adding different levels of PPP on the opacity of cassava starch-based films is shown in Fig. 5. The control sample had the lowest opacity (0.65 mm^{-1}), and the addition of PPP to the starch films caused a significant increase in the opacity of the samples and with increasing the level of this additive, an increase in the opacity of the films was observed ($p < 0.05$). The highest opacity was for the film sample containing 8% PPP (1.25 mm^{-1}).

Since light can stimulate the oxidation of food products, films used for food packaging should be able to limit the entry of light into the package as much as possible (Wang et al., 2019). The increase in the opacity of starch films containing PPP is due to the ability of light absorption by anthocyanins in pomegranate peel (Peralta et al., 2019). The higher the concentration of anthocyanins leads to the higher the opacity of the film samples. Due to the red color of pomegranate peel, the increase in opacity of the films due to the incorporation of it is not unexpected. Sun et al. (2017) also stated that anthocyanins have light-absorbing properties. Bilgiç et al. (2019) also reported that the addition of eggplant anthocyanin extract to starch films increased the opacity of the films. Other researchers have found similar results in studying the effect of anthocyanin extracts on the opacity of the packaging films (Capello et al., 2021; Merz et al., 2020; Qin et al., 2019).

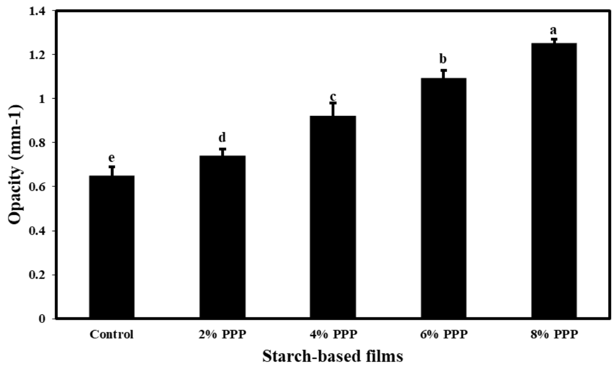

Fig. 5. Comparison the opacity (mm^{-1}) mean values of cassava starch-based films containing different levels of PPP. Bars represent mean (*n* = 3) ± *SD*. Different letters on the bars indicate a significant difference at 5% level of probability among film samples. PPP: pomegranate peel powder.

3.6. *The sensitivity of the films to pH changes*

The color parameters of different cassava starch-based films containing PPP after drying and after placement in buffer solutions with different pHs were investigated and the results are presented in Table 1. With the addition of PPP in starch films and increasing its levels from 2% to 8%, the L* index (lightness) was significantly reduced and the a* and b* indexes, as well as ΔE index (total color change) of the films was increased and the color of produced samples were redder ($p < 0.05$). Since the ΔE index of all film samples was higher than 3.5, their color changes compared to the control film can be seen with the human eye. Exposure of the active films in solutions with different pHs significantly reduced the color lightness ($p < 0.05$). The most changes due to exposure to different buffer solutions were related to the a* color index. The color of starch-based films containing PPP was pink to red, and the films in pH = 4 also showed a light pink color. The color of the film samples at pH = 7 turned green and the color of these films at pH = 9 was completely green.

The active starch films containing PPP demonstrated different colors at different pHs. Since in buffer solutions with different pHs, the ΔE index of films containing 4, 6, and 8% of PPP was more than 3.5, the color changes in these pHs were visible to the human eye. At different pHs, different structures of anthocyanins are observed, including the flavonoid cation structure at acidic pH, the colorless chalcone or *carbinol pseudobase* structure at relatively acidic pH, and the quinoidal base structure at alkaline pH (Meng et al., 2021). In their research, Andretta et al. (2019) developed pH-sensitive films using blueberry residues and observed that at acidic pHs (pH = 2-5), the color of the film samples was reddish orange, and in alkaline pHs (pH = 6-12), the color of these films changed to yellowish green. Similarly, Choi et al. (2017) reported that due to the incorporation of flashed sweet potato anthocyanin extract into biopolymer-based films, the a* index values decreased with changing pH from acidic to alkaline, and the color of the films changed from red to green. In studies conducted by other researchers, discoloration of pH-sensitive film samples containing anthocyanins from various sources such as black beans (Prietto et al., 2017), red cabbage (Pereira et al., 2015), grape peel (Ma & Wang, 2016), eggplant (Bilgiç et al., 2019), etc. was also observed. Yong et al. (2019) also demonstrated that chitosan-based films containing purple-fleshed potato extract were sensitive to pH change and with changing the pH from acidic to alkaline, their red-pink color changed to purple-brown color.

Table 1. The color parameters of cassava starch-based films containing different levels of PPP immersed in different pH buffers.

pH	Samples	L*	a*	b*	ΔE
-	Control	95.29 ± 0.38 a	1.05 ± 0.04 e	3.39 ± 0.28 e	-
	2% PPP	93.95 ± 0.46 b	6.58 ± 0.08 d	5.76 ± 0.20 d	6.16 ± 1.05 d
	4% PPP	91.92 ± 0.52 c	10.46 ± 0.07 c	8.41 ± 0.24 c	11.18 ± 0.84 c
	6% PPP	88.50 ± 0.24 d	11.99 ± 0.15 b	13.45 ± 0.33 b	16.34 ± 0.93 b
	8% PPP	86.64 ± 0.41 e	13.67 ± 0.10 a	17.15 ± 0.27 a	20.58 ± 1.12 a
pH 4	Control	34.82 ± 0.16 d	1.78 ± 0.06 e	-1.94 ± 0.11 a	-
	2% PPP	35.54 ± 0.39 c	5.23 ± 0.11 d	-2.01 ± 0.14 a	3.52 ± 0.83 c
	4% PPP	36.07 ± 0.61 bc	7.02 ± 0.24 c	-1.98 ± 0.28 a	5.40 ± 0.75 b
	6% PPP	36.79 ± 0.55 ab	9.10 ± 0.22 b	-2.09 ± 0.19 a	7.58 ± 0.69 a
	8% PPP	37.29 ± 0.52 a	10.56 ± 0.17 a	-2.10 ± 0.32 a	9.12 ± 0.88 a
pH 7	Control	40.28 ± 0.49 a	-0.49 ± 0.10 a	-2.32 ± 0.23 a	-
	2% PPP	37.70 ± 0.43 b	-0.57 ± 0.15 a	-0.64 ± 0.33 b	3.08 ± 0.25 b
	4% PPP	35.14 ± 0.55 c	-0.63 ± 0.26 a	0.08 ± 0.14 a	5.61 ± 0.40 a
	6% PPP	35.57 ± 0.36 c	-0.60 ± 0.19 a	-0.11 ± 0.22 ab	5.20 ± 0.44 a
	8% PPP	35.33 ± 0.47 c	-0.64 ± 0.20 a	0.02 ± 0.26 a	5.46 ± 0.37 a
pH 9	Control	36.95 ± 0.40 a	-6.38 ± 0.16 b	-2.64 ± 0.18 b	-
	2% PPP	35.41 ± 0.29 b	-10.46 ± 0.20 a	-0.34 ± 0.22 a	4.93 ± 0.55 a
	4% PPP	34.03 ± 0.37 c	-10.59 ± 0.13 a	-0.56 ± 0.15 a	5.53 ± 0.47 a
	6% PPP	34.39 ± 0.46 c	-10.65 ± 0.16 a	-0.39 ± 0.19 a	5.46 ± 0.52 a
	8% PPP	34.78 ± 0.24 c	-10.55 ± 0.22 a	-0.44 ± 0.14 a	5.19 ± 0.67 a

Values represent mean (n=3) ± SD. Different letters represent significant difference at 5% level of probability among sample the samples in each column and each pH buffers. PPP: pomegranate peel powder.

4. Conclusion

The results of this study generally indicated the color sensitivity of the cassava starch-based films containing PPP to pH changes. If the environment changes from acidic to alkaline, the color of the films was changes from reddish pink to green. Due to the total color change index of these films, their color changes are visible with the naked eye. Other properties like water vapor permeability and oxygen permeability of the films increased by the addition of the PPP. However, it is not important for an intelligent film. Therefore, the produced films in this study can be used as intelligent pH-sensitive packages.

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Conflict of interest

The authors declare that they have no known competing financial interests.

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