1	Senggani fruit (Melastoma malabathricum Linn.) extract as a natural indicator in
2	pH - responsive PVA - Taro starch plastic packaging
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12	Abstract: Polyvinyl Alcohol (PVA)-Starch-based bioplastics are widely used in many
13	applications. pH-responsive plastic packaging was produced by the addition of senggani
14	(Melastoma malabathricum Linn.) fruit extract into PVA-taro starch-based plastic
15	packaging. This research aimed to study the characteristics of senggani fruit extract at
16	various pH variations and its application as a pH indicator in intelligent packaging.
17	Senggani fruit was extracted by the maceration method using a solvent of 96% ethanol
18	and 3% citric acid with a ratio of $85:15$ (v/v). Senggani fruit extract solution changes
19	color, becoming pink at pH below 6, pale purple at pH 7 - 11, and brownish yellow at pH
20	12 - 14. The color of the senggani fruit extract solution was stable at $pH < 5$. Before the
21	addition of senggani fruit extract, the PVA-taro starch solution produced a brownish-
22	yellow plastic packaging. After the addition of senggani fruit extract, pink plastic was
23	obtained. The addition of senggani fruit extract affected the mechanical properties of
24	plastic packaging, which reduced the swelling from 103.679 \pm 2.456% to 57.827 \pm

3.563%, the tensile strength value from 3.827 ± 0.603 Mpa to 1.991 ± 0.460 Mpa, and
the percent elongation value from 156.250 ± 12.392% to 116 ± 6.722%. Plastic packaging
with the addition of senggani fruit extract changes color at pH 1 - 14 and has different
color parameter values (L, a, b, E, and WI), therefore, it has the potential to be used as
intelligent packaging to monitor food freshness and quality.

6 Keywords: Intelligent packaging, senggani fruit, *Melastoma malabathricum* Linn.

7

8 1. Introduction

9 Packaging for food products is essential, and the guarantee of food safety and quality is 10 maintained by packaging. Plastic packaging from synthetic polymers is widely used 11 however it cannot be decomposed in the short term. The utilization of plastic is relatively 12 high every year, causing plastic waste to accumulate and environmental problems. To 13 overcome these problems, many plastic packaging has been developed with 14 biodegradable materials such as starch, cellulose, wheat gluten, polylactide (PLA), and 15 polyhydroxyalkanoate (PHA) [1].

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Indonesia has abundant biological and natural resources, especially agricultural products.
There are potential resources to be developed into biodegradable plastic such as corn,
cassava [1] taro, wheat, potato, and tapioca, [2]. Taro has the potential to be used as raw
material for making plastic because taro contains starch which consists of amylose and
amylopectin homopolymers. The amylopectin in Taro has a short and branched chain. It
will undergo a long average chain length due to applied heat [3].

Along with consumer needs for the quality and safety of food, packaging innovation has 1 2 developed. One of the innovations is intelligent packaging, which not only provides information about the expiration date of food but also provides information about food 3 quality directly through indicators [4]. There are two types of indicators, namely 4 5 biological indicators and synthetic indicators. Synthetic indicators sold commercially are 6 considered more expensive. Besides, their use can cause environmental pollution and, if 7 applied to food, can damage the human body [5]. Natural indicators are cheaper and environmentally friendly, so they have the potential to be developed further. Indicators 8 usually used on product packaging consist of indicators of changes in pH [6], O₂, CO₂, 9 10 humidity, and temperature [5].

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Anthocyanins are phenolic compounds that give red, purple, and blue colors, generally derived from fruits and vegetables. Anthocyanins can be used as pH indicators because they undergo structural changes and show color variations at different pH conditions [7]. Senggani (*Melastoma malabathricum* Linn.) is a plant that contains anthocyanins. Based on the previous research, the senggani fruit extract gave a sharp color change due to variations in pH [8]. Senggani is easily found in West Kalimantan because it grows wild in bushes and forests.

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The natural indicators incorporated with bioplastic for pH-responsive materials have attracted recently. The PVA-Starch-based materials are interesting due to their low cost and biodegradability. Starch-PVA composites with ZnO and Jamun extract as the additives gave a good response to the pH change [9]. Another anthocyanin source, red cabbage, successfully acted as an active and intelligent packaging on starch-PVA matrix

with propolis as the additives [10]. The mechanical properties of the matrix are one of
the important factors for packaging materials. According to [11], the mechanical
properties of PVA were influenced by the presence of starch due to its brittle natural
properties. To improve the mechanical properties, the presence of a crosslinking agent is
important. Glutaraldehyde is one of the common crosslinking agents for PVA-Starch
[12]. Another low-cost crosslinking agent is citric acid. Citric acid could bridge the
backbone of starch and PVA [13] [14].

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9 Many factors affect the environmental pH of food. Monitoring changes in pH can be used 10 as an effective method to identify spoilage in food [15]. Anthocyanins can be used to 11 identify food spoilage based on changes in pH. Sitanggang et al. (2020) have used 12 anthocyanin extract in gelatin as a pH indicator for smart packaging. Therefore, this 13 research was conducted by the addition of anthocyanin extract to the plastic packaging of 14 PVA-taro starch as an indicator that responds to pH changes and thus can be developed 15 as smart packaging.

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Starch as the matrix component has great attention to its biodegradability, food safety, 17 18 low cost, and ease of preparation. Taro tuber is one of the natural resources that produce starch. There were various applications of taro tuber as biodegradable plastic. Shanmathy 19 20 et al. (2021) introduced the bentonite as the reinforced taro starch biodegradable matrix. 21 The taro tuber starch also gave good performance in incorporated with chitosan [16]. The 22 PVA-Taro starch bioplastics had biodegradability up to 65% for 8 weeks in aquatic conditions [17] [18]. On the other hand, the extract of Senggani has various applications, 23 24 there are in food additives and colorants, pharmaceutical industries, and coating [19]. Due

to the anthocyanin content of Senggani fruit extract, it turns out to have potency as a pH
indicator [20] and detection for shrimp freshness [21]. For the application pH-responsive
material has not developed properly. The novelty in this research, is the Senggani fruit
extract incorporated with PVA-taro starch as a pH-responsive material to monitor the
freshness of fish.

6

7 2. Materials and methods

8 2.1. Materials

The materials used in this study were hydrochloric acid (HCl) p.a, citric acid 9 10 $(C_6H_8O_7.H_2O)$ p.a, disodium phosphate heptahydrate (Na_2HPO_4) p.a, glycerol $(C_3H_8O_3)$ p.a, potassium chloride (KCl) p.a, sodium hydroxide (NaOH) p.a, sodium bicarbonate 11 (NaHCO₃) p.a purchased from Merck. Polyvinyl Alcohol (PVA) was purchased from 12 13 Sigma Aldrich. All of the chemicals were analytical grade. For pigment extraction, ethanol (C₂H₆O) 96% was in technical grade. Senggani fruit (Melastoma malabathricum 14 Linn.) as the source of the pigment was collected at Ayani Street, Pontianak City, and 15 taro as the source of starch was purchased at Flamboyan Market Pontianak, Indonesia. 16

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2.2. Extraction of Senggani Fruit

The Senggani fruit was collected at Ayani Street, Pontianak City. It was separated from the dirt and rinds, weighed at 600 grams, and mashed with a blender. It was extracted by maceration for 3 x 24 hours. The solvent was changed every 24 hours using ethanol solvent until all samples were submerged with a ratio of 96% ethanol solvent : 3% citric acid 85 : 15 (v/v). The filtrate was collected and evaporated using a rotary evaporator. The extract was obtained after a vacuum drying treatment.

2.3. pH Sensitivity Test on Senggani Fruit Extract

Senggani fruit extract was weighed at 125 mg and dissolved in 25 mL of 96% ethanol and 3% citric acid with a ratio of 85 : 15 (v/v). An amount of 1 mL extract solution was put into 14 test tubes, then 5 mL of buffer solution pH 1 - 14 was added [22]. The preparation of buffer solution with a pH range of 1 - 14 would be described below. The color changes were observed, and the maximum wavelength spectrum was measured using a UV-Vis spectrophotometer (Shimadzu UV 2600) in the wavelength range of 400 - 800 nm [23].

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10 A buffer solution of pH 1 was prepared by mixing 100 mL KCl 0.02 M and HCl 2 N until pH 1 \pm 0.1 was obtained (Purwaniati et al., 2020). Buffer solution of pH 2 was prepared 11 12 by 25 mL KCl 0.2 M and HCl 0.2 M and mixed until pH 2 \pm 0.1 was obtained. Buffer 13 solutions of pH 3 - 8 were prepared by citric acid solution 0.1 M and Na₂HPO₄ solution 0.2 M with various volumes until the desired pH was obtained. Buffer solutions of pH 9 14 - 10 were prepared by mixing NaHCO₃ 0.05 M and Na₂CO₃ solution. Buffer solutions of 15 pH 11 and 12 were obtained by mixing 50 mL Na₂HPO₄ 0.05 M with NaOH 1 M. Buffer 16 solutions of pH 13 were prepared by mixing 25 mL KCl 0.2 M with NaOH 0.2 M until 17 pH 13 reached (Mulyono, 2006). Last, buffer solutions of pH 14 were obtained by NaOH 18 1 M solution (Indira, 2015). All processes were monitored by a pH meter (Mettler Toledo 19 20 S220 basic).

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22 2.4. Taro Starch extraction

The taro was peeled, washed thoroughly, and weighed 825 grams. Taro was blended with water in a ratio of 1: 2 (w/v) and then filtered using a cloth. After that, the pulp was added to 1 L of water, blended, and filtered again. The two liquids are combined and deposited
for 24 hours so that the liquid and starch residue are separated and decanted. Furthermore,
mashed with a blender and sieved through a 100 - mesh sieve. The starch precipitate was
dried under the sunlight.

5 2.5. Preparation of Plastic Packaging Made of PVA-Taro Starch with Senggani 6 Fruit Extract

7 Preparation of bioplastics was carried out by *slip casting* method. An amount of 2.5 grams 8 of PVA was weighed and dissolved in 25 mL of distilled water in a beaker glass and heated at 90 °C while stirring with a magnetic stirrer until dissolved. An amount of 2.5 9 grams of taro starch, 1.25 grams of citric acid, and 1 mL of glycerol were dissolved in 25 10 mL of distilled water using another beaker glass. After the PVA was dissolved, a solution 11 12 of starch, citric acid, and glycerol dissolved in distilled water was added and heated at 90 13 °C for 10 minutes and then stirred with a magnetic stirrer for 50 - 60 minutes until viscous solution was obtained. The same procedure was also carried out with the addition of 14 Senggani fruit extract, referring to [24], 1 gram of Senggani fruit extract was weighed 15 and dissolved in 10 mL of 96% ethanol and 3% citric acid in a ratio of 85 : 15 (v/v). The 16 diluted extract was taken with a pipette of up to 2.5 mL into a cold bioplastic solution and 17 18 stirred using a magnetic stirrer. Furthermore, then printed on a 15 cm x 21 cm acrylic plate and allowed to stand for 2 x 24 hours at room temperature (28 °C). After drying, the 19 bioplastic was removed from the acrylic plate and tested. 20

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1 2.6. Characterization of Plastic Packaging Made of PVA-Taro Starch with

2 Senggani Fruit Extract

Characterization of the PVA-Taro Starch was carried out by pH sensitivity test, water
resistance test, mechanical properties, and application of the bioplastic for food
packaging. All of the characterizations are described below.

6 2.6.1 pH Sensitivity Test of Plastic Packaging Made of PVA-Taro Starch

Plastic packaging made of PVA-taro starch without Senggani fruit extract and with the addition of Senggani extract was cut to a size of 2 cm x 1.5 cm. Furthermore, the plastic packaging was placed into a petri dish containing a buffer solution of pH 1-14, and color changes were observed. Then the sample is placed on white paper as a background [25]. The value of L* = 93.698, a* = 1.123 and b* = - 5.310. The values of L measure the color changes that occur; a, b using a colorimeter (Linshang LS170), and the color difference (ΔE) and whiteness index (WI) values are calculated using the following formula [26]:

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WI = $100 - \sqrt{(100 - L)^2 + a^2 + b^2}$(2)

16 2.6.2 Water Resistance Test

Test swelling by weighing the bioplastic cut to a size of 2.5 cm x 2.5 cm, then placed into
a container containing 20 mL of distilled water for 1 hour. Furthermore, the bioplastic
samples were removed from the water and weighed, and water resistance was calculated
[25]. The water resistance test was repeated three times.

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1 2.6.3 Mechanical Properties

Bioplastic mechanical properties were tested by preparing a sample measuring 5 cm x 1
cm for tensile strength and elongation using the Universal Testing Machine at a speed of
1 mm/s. The data obtained then calculated the value of tensile strength and elongation.

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6 2.6.4 Application of Plastic Packaging on Freshness of Fish

Tuna is cleaned and weighed 25 grams, then put into a plastic container. Then put it into
the refrigerator with variations of storage time of 24 hours, 48 hours, 72 hours, 96 hours,
and 120 hours, and observe color changes in the plastic packaging. Then the plastic
packaging of PVA-taro starch without addition and with the addition of Senggani fruit
extract was cut 3 cm x 2 cm; then it was placed on top of the fish, and the container was
closed.

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14 **3** Results and Discussion

To the application as a pH - sensitive material, here we evaluate the characteristics of Senggani fruit extract including the properties at different pH conditions, followed by physical and mechanical properties study when the pigment is incorporated into PVA taro starch matrix. PVA - taro starch without Senggani fruit extract addition was used as a reference. The performance of pH - responsive materials was observed as the fish packaging.

21 3.1. Characteristics of Senggani Fruit Extract

Senggani fruit extract was successfully extracted with ethanol : citric acid 3% (85 : 15)
as the solvent. Citric acid addition to the solvent was to maintain the condition to prevent
the degradation of anthocyanin in the extract. Anthocyanin extract from Vietnamese

Carissa carandas L. fruit showed good color stability and less anthocyanin degradation 1 2 with citric acid added [27]. Melastoma malabathricum fruit extraction with acidified ethanolic solvent gave high mean anthocyanin yields of 880.923 mg/100 g [19]. 3 Extraction of Hibiscus subdariffa with acidified ethanol (85:15) showed that the acidic 4 5 condition of the solvent increased the efficiency of anthocyanin extract. It can be 6 concluded that acid addition to the extraction medium had a great effect in stabilizing 7 anthocyanins, thus the extraction efficiency increased. Acid conditions provide a favorable medium for the formation of flavylium chloride salts from simple anthocyanins 8 and improve the efficiency of anthocyanins extractions [28]. Senggani fruit extract at a 9 10 pH range of 1 - 14 resulted in a color change, as shown in Figure 1. Senggani fruit extract is pink at pH 1 - 3 and fades at pH 4 - 6. The senggani fruit extract has a pale purple color 11 at pH 7 - 11, slowly turning brownish-yellow from pH 12 to pH 14 and forming a 12 13 precipitate. Senggani fruit extract solution reacted with a high pH (alkaline) solution, causing the solution to be unstable, therefore precipitation occurred at pH 14, indicating 14 the solution's instability. The precipitate formed can occur as a result of aggregation [29]. 15

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Figure 1. Changes in the color of the senggani fruit extract solution at (a) pH 1 - 7 and (b) pH 8 - 14

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The color change of the Senggani fruit extract solution at various pH shows the absorption peaks of different wavelengths in **Figure 2**. At pH 1 - 8, the peak of the maximum absorption wavelength increased (bathochromic shift) and decreased the absorbance value to pH 6. At pH 7 and 8, the absorbance value increased. At pH 9, the maximum wavelength decreased to pH 11 along with a decrease in the absorbance value; at pH 12, the maximum wavelength increased with an increase in the absorbance value. At pH 13 and pH 14, there is no maximum wavelength spectrum.



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Figure 2. UV-Vis spectrum of senggani fruit extract pH 1-14

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11 Changes in pH can cause structural changes in the anthocyanin compound molecules that 12 cause color changes. In an acidic solution, the anthocyanin structure changes to flavylium 13 cation (red) and quinoidal anhydrase (purple); in an alkaline solution, it changes to 14 chalcone (yellow) [26]. The anthocyanin structures at pH 1 - 3 presented as flavylium 15 cations, and pH 4 - 5 as carbinol pseudo-base. At pH 6 - 7, anthocyanin had a quinoidal 16 base structure, however at pH 7 - 8 it presented as an anionic quinoidal base. At pH 9, 17 anthocyanin had a chalcone structure [30]. Phytochemical screening showed that *Melastoma malabathricum* gave a strong response
to presented flavonoids and phenolic [31]. Anthocyanin remains the major component in *Melastoma malabathricum* fruit extract. Anthocyanin identification using UPLC-ESIMS/MS showed that on *Melastoma malabathricum* fruit extract contains cyanidine and
delphinidine compound [19]. Figure 3 presents the cyanidine and delphinidine structure
[32].



7

8 **Figure 3** Anthocyanidine structure (a) Cyanidine (R1 = -OH; R2 = -H);

9 (b) Delphinidine (R1 = -OH; R2 = -OH) [32]

10 Plastic packaging made from PVA-taro starch produced without and with senggani fruit

11 extract added produced brownish-yellow and pink plastic, respectively. It is shown in

12 **Figure 4**.



- 15 Figure 4. PVA plastic packaging-taro starch (a) without Senggani fruit extract (b) with
- 16 the addition of Senggani fruit extract

1 The IR spectrum was produced from plastic packaging made from PVA-taro starch 2 without the addition of senggani fruit extract and with the addition of senggani fruit 3 extract in **Figure 5**.



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Figure 5. The spectrum of plastic packaging made of PVA-starch (a) without the addition
of senggani fruit extract and (b) with the addition of Senggani fruit extract

Figure 5 shows a shift in the absorption band between the functional groups without the 7 addition of Senggani fruit extract and the addition of Senggani fruit extract that occurs 8 9 due to the interaction between Senggani fruit extract and PVA-starch. Absorption peaks at wave numbers 3555 cm⁻¹ and 3539 cm⁻¹ represent the OH stretching vibration. The 10 wavenumbers at 2978 cm⁻¹ and 2928 cm⁻¹ indicate the absorption peak of the CH 11 asymmetric stretching vibration Sitanggang et al., 2020). 12 The presence of C=O stretching was interpreted by the peaks at 1730 cm⁻¹ and 1755 cm⁻¹ [13]. The absorptions 13 at 1404 cm⁻¹ and 1408 cm⁻¹ indicate the CH-CH₂ vibration [33]. Furthermore, it was 14

observed at wave numbers 1026 cm⁻¹ and 1032 cm⁻¹, which showed the absorption peak
of the C-O-C stretching vibration [13].

There were no significant shifts in absorption between with or without senggani fruit 3 extract. However, there were changes in intensity of peak absorption at 3539 cm⁻¹ and 4 1730 cm⁻¹. The peak intensity of hydroxyl groups and C=O groups of PVA-Taro Starch 5 with Senggani fruit extract addition remains lower than the other one. The decreased 6 7 absorption intensity might be caused by the functional groups of organic compounds in 8 Senggani fruit extract that had reacted with hydroxyl groups of PVA and starch and were involved in crosslinking reactions in the presence of citric acid. The formation of 9 hydrogen bonding involves the OH groups of the PVA, starch, and Senggani fruit extract 10 with the COOH groups of citric acid [13]. 11

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13 **3.2. Physical Characteristics of Plastic Packaging**

The mechanical properties of plastic packaging made from PVA-taro starch are shown in **Table 1**. The addition of Senggani fruit extract to plastic packaging lowered both tensile strength and elongation values. The decrease in tensile strength caused by fruit extract addition into the film thus changes its mechanical properties [34]. Senggani fruit extract contains many organic compounds with major components from anthocyanin groups.

Table 1. Mechanical properties of plastic packaging

Sample	Tensile Strength (Mpa)	Elongation (%)	Swelling (%)
PVA-Taro Starch	3.827 ± 0.603	156.250 ± 12.392	103.679 ± 2.456
PVA-Taro Starch	1.001 ± 0.460	116 ± 6.722	57.827 ± 3.563
Senggani Fruit Extract	1.991 ± 0.400	110 ± 0.722	

20 Information: data presented in the form of mean \pm standard deviation

Table 1 shows the swelling percentage of the sample after Senggani fruit extract addition.

When the organic compound is incorporated, there are two possibilities. First, at lower concentrations of anthocyanin, the mechanical properties of the bioplastic could be improved. On the contrary, another research showed that at higher concentrations, the mechanical properties had decreased due to the unstabilized polymeric network by excessive anthocyanin [33]. This phenomenon decreased both mechanical properties, the tensile strength, and the percentage of elongation in this research.

The swelling value for PVA-starch with Senggani fruit extract had lowered due to the 7 presence of additives thus restricting the water penetration into the bioplastic [35]. This 8 restriction was confirmed by FTIR data (Figure 5) which presented the decreased 9 intensity of the hydroxyl group after the Senggani fruit extract incorporated had lowered 10 swelling capacity. The decreased intensity of hydroxyl groups will reduce the interaction 11 of bioplastic with water [23]. Moreover, the Senggani fruit extract in this research was a 12 13 crude extract, which contains waxy and resinous components, contributed to hydrophobic surfaces, and contributed to lower swelling capacity [33]. Swelling capacity influenced 14 the biodegradability of bioplastic. Plastics with a lower percentage of swelling capacity 15 are better due to easily damaged or decomposed [36]. 16

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18 **3.3. pH-responsive Plastic Packaging**

The color response in the pH range of 1 - 14 of plastic packaging made from PVA - taro starch is shown in **Figure 6**. At pH 1, the plastic packaging is pink and slowly fades to pH 5; at pH 6, the plastic packaging shows an orange color change. Furthermore, at pH 7, it is brownish yellow, and until pH 14, there is a change in the color of the plastic packaging, which is getting brown.

Color changes in plastic packaging made from PVA - taro starch with the addition of
 Senggani fruit extract result from changes in the anthocyanin structure at different pHs.
 The color change comes from flavylium cations under acidic conditions turning into
 quinoidal anions under alkaline conditions [26]. Meanwhile, in plastic packaging without
 the addition of Senggani fruit extract, there was no color change.

pН	PVA - Taro Starch Senggani Fruit Extract	PVA - Taro Starch	рН	PVA - Taro Starch Senggani Fruit Extract	PVA - Taro Starch
1		4	8	5	
2			9		
3	and the second sec	-	10		
4			11	100	11
5			12		
6			13		
7	Et.		14		



Figure 6. Changes in the color of plastic packaging at pH 1-14

1	The color parameters L and WI values at 0 - 50 indicate a dark-colored film and 51 - 100
2	a light color. a positive value (a +) indicates red a negative (a -) is green, b positive (b +)
3	means yellow and b negative (b -) is blue [26]. Table 2 shows the values of L and WI
4	with brightness levels that tend to decrease with increasing pH. A positive value indicates
5	a reddish color which tends to decrease to pH 7, increase from pH 8 to 12, then decrease
6	to pH 14. At the same time, a positive b value indicates a yellow color at $pH > 8$ with a
7	value of b greater than $pH < 8$. The WI value, which tends to be lower with increasing
8	pH, indicates that the plastic packaging is getting darker. The total color difference value
9	(Δ E) shows the difference from plastic packaging, which tends to decrease to pH 7 and
10	increase at pH 8 to 14. These color parameters show that plastic packaging with the
11	addition of Senggani fruit extract has a color change response to changes in pH.

12 Table 2. Color parameter of plastic packaging PVA-taro starch with the addition of13 Senggani fruit extract

pН	L	a	b	ΔΕ	WI
1	72.463 ± 1.255	17.683 ± 0.681	25.209 ± 0.370	40.742 ± 1.227	58.646 ± 1.380
2	71.281 ± 2.690	11.915 ± 1.970	22.701 ± 3.544	37.473 ± 4.803	61.482 ± 4.672
3	76.583 ± 3.532	11.521 ± 1.818	20.459 ± 1.197	32.740 ± 3.213	66.785 ± 3.729
4	77.755 ± 1.921	10.497 ± 1.004	13.927 ± 3.820	26.867 ± 4.181	71.466 ± 3.707
5	74.573 ± 1.276	11.293 ± 0.531	19.704 ± 2.232	33.335 ± 1.030	65.520 ± 0.721
6	75.511 ± 1.575	9.227 ± 1.310	21.924 ± 5.606	33.817 ± 5.636	65.719 ± 4.973
7	78.052 ± 0.788	7.687 ± 0.477	15.482 ± 0.939	26.857 ± 1.200	72.047 ± 1.143
8	76.717 ± 5.868	9.233 ± 3.461	17.879 ± 8.753	29.884 ± 11.057	69.065 ± 10.411
9	69.797 ± 0.561	9.119 ± 0.347	18.615 ± 1.349	34.759 ± 1.359	63.354 ± 1.201
10	65.816 ± 1.737	11.191 ± 1.129	26.163 ± 1.763	43.250 ± 2.569	55.508 ± 2.549
11	69.035 ± 1.159	8.294 ± 1.456	19.525 ± 2.999	35.822 ± 2.433	62.362 ± 2.050
12	51.907 ± 2.440	19.512 ± 1.652	34.662 ± 4.800	60.732 ± 4.982	37.526 ± 4.692
13	62.175 ± 5.124	11.167 ± 2.128	27.533 ± 0.994	46.815 ± 3.364	51.751 ± 4.061
14	56.684 ± 4.385	17.368 ± 2.059	39.776 ± 1.728	60.679 ± 2.167	38.570 ± 2.736

The PVA - Taro Starch with Senggani fruit extract was applied to observe the
performance in evaluating the spoilage of fish at chiller temperature (~ 5°C). Figure 7
showed the applications of the bioplastics. It had shown a pink color at the beginning and 17

turned to pale pink after 24 hours. A clear brownish color of the bioplastic was obtained 1 2 after 48 hours followed by pH testing to the fish reach pH 8. The physical visualization of the fish also showed spoilage had started after 48 hours. On the other hand, the plastic 3 packaging of PVA - taro starch without Senggani fruit extract addition had no color 4 5 change from the beginning up to 120 hours of storage. By this phenomenon, it could be concluded that the addition of Senggani fruit extract into PVA - Taro Starch bioplastic 6 7 gave a response to the change in the condition of the fish, hence it had potency as an 8 intelligent packaging material.



^{9 *}h denoted as hours

Figure 7 Changes in the color of plastic packaging with time variations

11 The color changes that occur indicate that plastic packaging with the addition of Senggani

12 fruit extract is responsive to alkaline compounds that evaporate from spoilage that occurs

13 in fish. The compounds were produced by bacterial activity in protein decomposition.

¹⁰

This activity produces alkaline and volatile nitrogen (Total Volatile Bases), influencing
the increase in pH and foul odor [9][37]. Plastic packaging with the addition of Senggani
fruit extract has a fast response to Total Volatile Bases (TVB) [23], so it can show the
freshness of fish during storage in real-time.

5

6 4. Conclusion

7 The PVA - Taro Starch was prepared with and without the Senggani fruit extract addition. 8 Senggani fruit extract gave a response to different pH conditions (pH 1 - 14) due to the presence of anthocyanin. The FTIR data showed that the addition of senggani fruit extract 9 didn't shift the wavenumber, however, there were reduced peak intensities at 3539 cm⁻¹ 10 and 1730 cm⁻¹. This phenomenon might be caused by hydrogen interaction presented in 11 the bioplastic. Both mechanical properties and swelling degree had lowered for the PVA 12 13 - Taro Starch with Senggani fruit extract addition. A study of the PVA-Taro starch in different pH conditions showed that pigment addition had contributed to the color change 14 of the bioplastic. It also gave a response when tested on fish products. Therefore, it had 15 potency as intelligent packaging to monitor the freshness and quality of food, especially 16 fish products. 17

18

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