Int. j. adv. multidisc. res. stud. 2024; 4(3):1267-1271

Received: 24-04-2024
Accepted: 04-06-2024

International Journal of Advanced Multidisciplinary Research and Studies

ISSN: 2583-049X

Preparation of Smart Packaging Film based on Cassava Starch and Natural Dye from *Uncaria Gambir* Hunter Leaves

[1] Mutia Devi Hidayati, [2] Arief Rahmatulloh, [3] Sandra Santosa
[1, 2, 3] Department of Chemical Engineering, Politeknik Negeri Malang, Indonesia

Corresponding Author: **Mutia Devi Hidayati**

Abstract
In this research, we reported the fabrication of packaging film based on cassava starch as a matrix crosslink with polyvinyl alcohol as filler we blended with natural dye from *Uncaria Gambir* extract. *U. Gambir* in small amounts can reduce the thickness of the film and be spread evenly distributed in the film structure. The study of moisture content in the Starch/PVA-*U.Gambir* film, is inversely proportional with the amount of *U.Gambir*. On the other hand, the solubility in water is directly proportional with the amount of *U. Gambir*. Based on the result of this research. Starch/PVA-*U. Gambir* film has the potential to be utilized as smart packaging film since it gives different responses toward acid-base condition.

Keywords: *Smart Packaging Film*, Natural Dye, *Uncaria Gambir*, Cassava Starch, Polyvinyl Alcohol

Introduction

Uncaria gambir is one of Indonesia's export commodities in the plantation sector. Based on its phytochemical content, *U. gambir* contains flavonoids, catechins, tannins, and alkaloids (Amir *et al.*, 2012) [1]. This compound has high economic value because it is used as a raw material for the pharmaceutical industry (mouthwash and wound healing), cosmetics, leather tanning, dyes, and food industry ingredients (Lemmens, Wulijarni and Wageningen, 1991; Dhalimi, 2006) [8, 4]. In its development, gambier export commodities are still dominated by raw materials such as leaves and twigs. Diversification and processing of gambier plants towards downstream products that can increase added value has not been implemented optimally (Dhalimi, 2006) [4].

One diversification of gambir processing is making smart packaging is making smart packaging film using natural dyes. Natural dyes have various advantages compared to synthetic dyes, namely they are abundant, cheap, non-toxic, and safe to use. Various natural dye-based compounds that can be used are chlorophyll, anthraquinones, flavonoids, carotenoids, indigoids, and so on (Mansour, 2018) [13]. These dye-based compounds are interactive indicators which could react with food products that contain metabolite compounds. This reaction provides a color change and can be used to indicate food quality and freshness (Arvanitoyannis and Stratakos, 2012; Jiang *et al.*, 2020) [2, 6].

Generally, natural polymers are used as the matrix of the film. Some natural polymers used in making packaging films include chitosan (Yong *et al.*, 2019) [21], starch (Stoll *et al.*, 2016; Lucchese *et al.*, 2018; Qin *et al.*, 2019) [16, 11, 14], and cellulose (Ma and Wang, 2016) [12]. Among these three materials, starch is the material that has the best characteristics to be used as a film matrix. This is because starch has high stability to heat, as well as acidic and alkaline conditions. However, starch couldn't stand alone if being used as a material for making packaging film, it will have disadvantages, namely low mechanical strength (Toro-Márquez, Merino and Gutiérrez, 2018) [17]. Hence, the addition of other biodegradable materials is needed to overcome these physical and functional drawbacks. One alternative material that can be used is polyvinyl alcohol (PVA), a hydrophilic, water-soluble, non-toxic, and biocompatible biodegradable polymer. The combination of starch/polyvinyl alcohol in packaging film can produce films with good transparency (Cano *et al.*, 2016) [3], non-toxic, renewable, and biodegradable (Lu, Xiao and Xu, 2009; Rezaei, Nasirpour and Fathi, 2015) [10, 15].

This research developed natural dye from *U. gambir* as an interactive indicator in smart packaging films. The mass variations of anthocyanin extract that were carried out are 0, 1, 1.5, and 2 (g). Those mass variations were conducted to determine the effect of mass added of *U. gambir* extract on the characteristics of smart packaging film such as thickness, tensile strength, elasticity, and water content. The utilization of a matrix in the form of cassava starch and PVA in this research is expected to

improve the mechanical properties so that it has good performance when applied to smart packaging film.

Experimental

Materials and chemical reagents

Dried leaves of *Uncaria gambir* were obtained from South Sumatra, Indonesia. Cassava starch was obtained from the oro-oro dowo market in Malang, Indonesia. Ethanol (99.6%) was purchased from Smart-Lab Company, in Indonesia. Glycerol (99%) hydrochloric acid (HCl, 36-38%), and Polyvinyl alcohol were purchased from Merck. Germany. The other reagents were of analytical grade.

Instrumentals

This study used several tools to do the experiments for instance scale, thermometer, electric heater, erlenmeyer, pH indicators, rotary evaporator, volume pipette, magnetic stirrer, beaker glass, FTIR, Tensile strength, and elongation test.

Extraction of *U. Gambir*

Dry leaves of *U. gambir* (100 g) were extracted twice with 250 mL of 80% ethanol solution containing 0.5 % HCl (v/v) at 4 °C for 24 h. The filtrate was centrifuged at 4000 g for 15 min and the supernatant was combined, and concentrated with a rotary evaporator by IKA-RV-8-Basic under pressure to obtain *U.gambir* extract.

The Preparation of pure starch/PVA film

Starch (2 g) and PVA (1 g) were incorporated into distilled water (100 mL). The suspension was heated on a hot plate with a magnetic stirrer at 80 °C and 500 rpm for 1 h until gelatinization. Then, 1 (w/w) % of glycerol was mixed with starch/PVA solution for 30 min. Finally, the solutions were degassed and cast into a Plexiglas plate (20 cm x 20 cm) and dried for 48 h at 40 °C.

The Preparation *Starch/PVA-UG film*

The dried UG extract was mixed with 100 mL of distilled water and stirred at 50 °C for 30 min on a magnetic stirrer at 500 rpm. The suspension was centrifuged at 4000 g for 15 min to obtain a pure UG suspension. The starch (2 g) and PVA (1 g) were added into the pure UG suspension and then dissolved at 80 °C using a magnetic stirrer at 500 rpm for 1 h. Furthermore, 1(w/w) % of glycerol was mixed with starch/PVA solution for 30 min. The film-forming solutions were similarly dried in a drying oven for 48 h at 40 °C. Before characterization, all dried films were stored in a desicator with a relative humidity of 50%. The samples were prepared with UG 1, 1.5, and 2 g, respectively.

Film thickness and moisture content

Film thickness was measured by digital micrometer at randomly selected 10 points on each film. The moisture content of the film (approximately 50 mg) was determined by measuring the weight loss of the film upon drying at 105 °C until the weight is constan. Three times replicates were performed for each sample. Moisture content (%) was calculated by the following equation:

$$\text{Moisture content (\%)} = \frac{(M_i - M_t)}{M_i} \text{ x } 100$$

Where Mi and Mt presented the weights of the film sample before and after drying, respectively.

Mechanical properties

The film sample was cut into a rectangular piece (6 cm x 1 cm) and measured on a Stograph VG10-E with an initial distance of 4 cm and a testing speed of 2 mm/s. Each test was repeated at least six times. The tensile strength was calculated by the following equation:

$$\text{Tensile strength} = \frac{F}{X \; x \; W}$$

Where F was the maximum stretching strength (N), x was the film thickness (mm), and W was the width of film piece (mm).

Furthermore, elongation at break was calculated by the following equation:

$$\text{Elongation at break (\%)} = \frac{L}{L_o} \text{ x } 100$$

Where L was the elongation at the moment of rupture (mm), and L_o is the initial length of the film piece (mm).

Results and Discussion

The Preparation *Starch/PVA-UG film*

Each powder of *U. gambir* with mass variations of 1, 1.5, and 2 (g) was mixed with 100 mL of distilled water. The solubility of dry *U. gambir* powder in water is very low, so a stirring process is required at a temperature of 50 °C for 30 minutes using a magnetic stirrer at a speed of 500 rpm. Further, the centrifugation process was continued at 2000 rpm for 30 minutes to obtain a pure *U. gambir* suspension. The film fabrication was carried out by mixing the *U. gambir* suspension with varying mass (1, 1.5, and 2 g) starch, and 1 g PVA dissolved at a temperature of 80 °C using a magnetic stir at a speed of 500 rpm for 1 hour until gelatinization occurred. Then, the resulting gel was added with 1% glycerol (v/v) and homogenized for 1 hour without heating. Next, the resulting gel was cast on a glass plate and dried for 48 hours at 40 °C using an oven. The results of starch/PVA-UG films with various masses can be seen in Fig1.

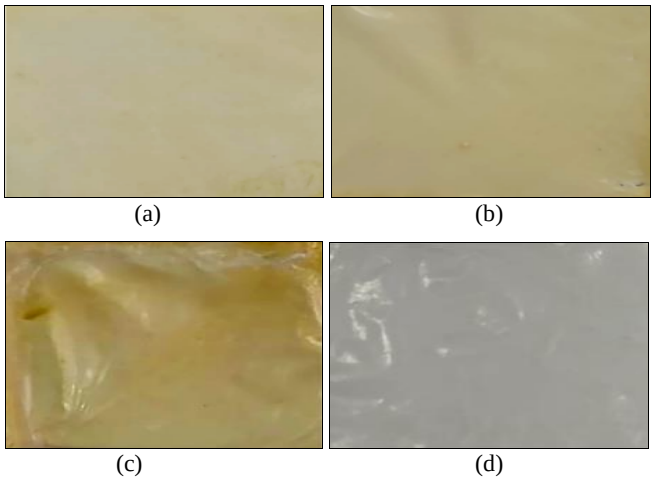

Fig 1: Starch/PVA-UG I (a), Starch/PVA-UG II (b), Starch/PVA-UG III (c), Pati/PVA (d) film

Thickness and moisture contents of starch-UG films

Thickness is an important factor that influences water vapor permeability, transparency, and tensile strength of packaging films. In Fig 2, Starch/PVA-UG II and Strach/PVA-UG III films have relatively higher thickness than Starch/PVA and Starch/PVA-UG I ($p < 0.05$). However, there was no significant difference in film thickness between Starch/PVA, Starch/PVA-UG I, Starch/PVA-UG II, and Starch/PVA-UG III. This shows that small amounts of *U. Gambir* have little impact on film thickness. Similar results were observed in *L. ruthenicum* starch extract and starch/PVA roselle film (Zhai *et al.*, 2017; Qin *et al.*, 2019) [22, 14].

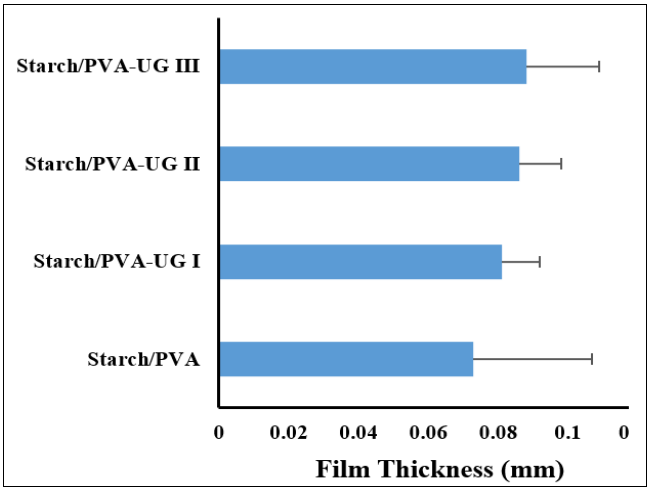

Fig 2: Film thickness of Starch/PVA-UG

U. Gambir in small amounts will be able to distribute well in the space between the starch and PVA molecules so that it can reduce the thickness of the film and more evenly induce the film structure. The moisture content of the Starch/PVA-UG films are shown in Fig 2. The moisture content of Starch/PVA-UG film decreased significantly from 26.68% to 21.65% as the content of *U. Gambir* added to the Starch/PVA mixture increased. Meanwhile, the Starch/PVA film showed a much higher moisture content of 31.47%.

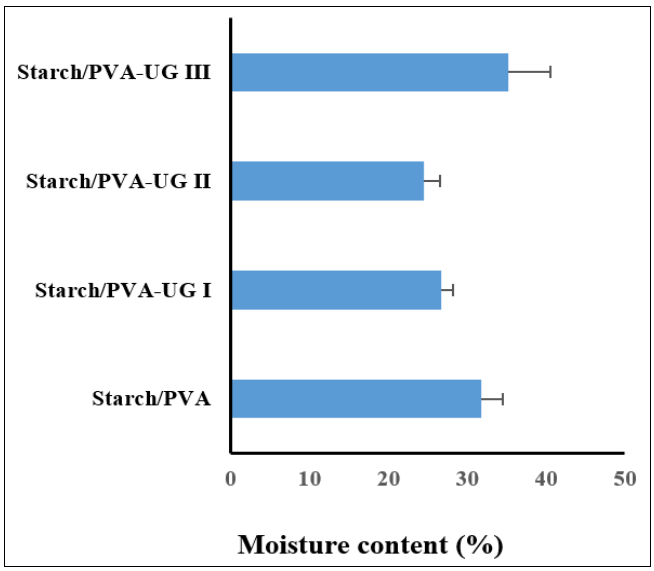

Fig 3: Moisture content of Starch/PVA-UG

This condition is due to the abundance of hydrophilic groups in starch, PVA, and *U. Gambir* molecules, especially OH groups. When *U. Gambir* was added to the Starch/PVA layer, the hydroxyl group in *U. Gambir* formed an intermolecular hydrogen bond with the hydrophilic hydroxyl groups on Starch and PVA. As a result, the interaction occurs between hydrophilic hydroxyl groups in starch/PVA, consequently, the moisture content is reduced. Therefore, the starch/PVA-UG III films have lower water content compared to the starch/PVA films (Wang *et al.*, 2013) [18]. This is confirmed by the study of moisture content in the Starch/PVA-roselle film, which decreased significantly along with the increase in *U. Gambir*. It is also associated with the interaction between starch/PVA and *U.Gambir*, which reduces the availability of hydroxyl groups in Starch/PVA interacting with water vapor (Zhai *et al.*, 2017) [22].

Mechanical properties of starch-UG films

Based on Fig 4 and 5, it can be seen that the presence of *U. Gambir* extract in the film affects the tensile strength and elasticity of the film. The tensile strength of the film is directly proportional with the content of *U. Gambir* in film. The starch/PVA-UG III film has the highest tensile strength and elasticity values among the others. Tensile strength enhancement of starch/PVA-UG film was caused by the abundant hydroxyl groups in the *U.gambir* extract, thereby forming hydrogen bonds with the hydroxyl groups in the starch and producing a stronger interface (adhesion) between starch, PVA, and *U.gambir* (Koosha and Hamedi, 2019; Wang *et al.*, 2019) [7, 19].

The mechanical properties of different Starch/PVA-UG films can also be seen in Fig 5. The elasticity of the film is directly proportional with *U. gambir* levels in the film. This is due to the *U. gambir* extract which contains hydroxyl groups is improve the performance of Starch and PVA. As a result, the film becomes more homogeneous, which will increase the elasticity value.

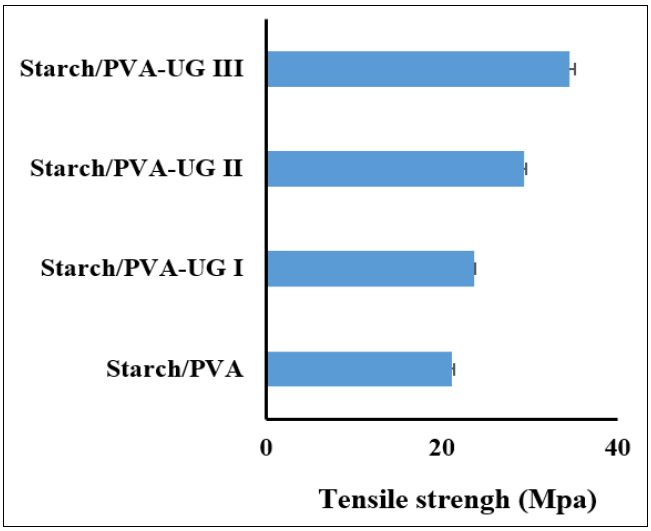

Fig 4: Tensile strength of Starch/PVA-UG film

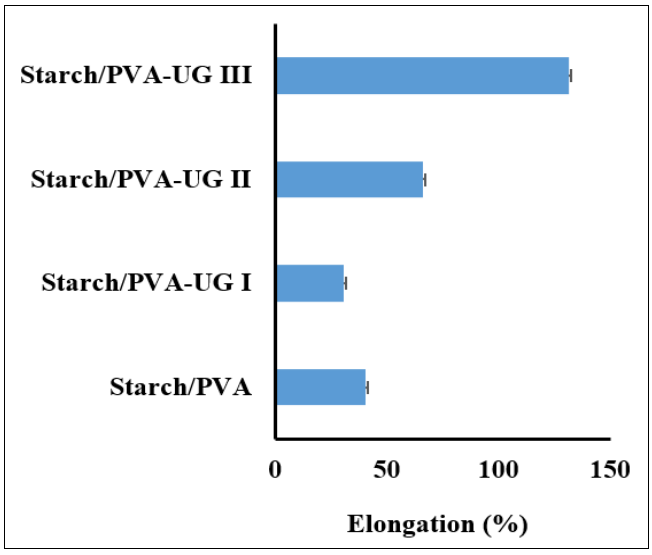

Fig 5: Elongation of Starch/PVA-UG film

Conclusion

In summary, natural dye indicators were extracted from *U. gambir* leaf and used in the fabrication of smart packaging films. The addition of various *U. gambir* extracts have affected the mechanical, thermal, water solubility, moisture content, and thickness. The concentration of extract added is directly proportional to the properties of the film, namely tensile strength, elasticity, water solubility, and thickness. The abundance of hydroxyl groups in *U. gambir* extracts forms hydrogen bonds with the hydroxyl groups in starch, PVA, and UG. In the future, starch/PVA-UG films have potential to be utilized as a freshness monitor of food in real-time.

Acknowledgements

The author would like to express their gratitude to the Chemical Engineering Department of Politeknik Negeri Malang for extending their support and providing the infrastructure to carry out the research work.

References

1. Amir M, *et al*. Phytochemical analysis and *in vitro* antioxidant activity of Uncaria gambir. International Journal of Green Pharmacy. 2012; 6(1):67-72. Doi: 10.4103/0973-8258.97136.
2. Arvanitoyannis IS, Stratakos AC. Application of Modified Atmosphere Packaging and Active/Smart Technologies to Red Meat and Poultry: A Review. Food and Bioprocess Technology. 2012; 5(5):1423-1446. Doi: 10.1007/s11947-012-0803-z.
3. Cano A, *et al*. Physical and antimicrobial properties of starch-pva blend films as affected by the incorporation of natural antimicrobial agents. Foods. 2016; 5(1):1-17. Doi: 10.3390/foods5010003
4. Dhalimi A. Permasalahan Gambir (Uncaria gambir L.) di Sumatera Barat dan Alternatif Pemecahannya. Perspektif. 2006; 5(1):46-59.
5. Gutiérrez TJ, *et al*. Hydrogen-bonding interactions and compostability of bionanocomposite films prepared from corn starch and nano-fillers with and without added Jamaica flower extract. Food Hydrocolloids. Elsevier Ltd. 2019; 89(October 2018):283-293. Doi: 10.1016/j.foodhyd.2018.10.058.
6. Jiang G, *et al*. Preparation and characterization of indicator films from carboxymethyl-cellulose/starch and purple sweet potato (Ipomoea batatas (L.) lam) anthocyanins for monitoring fish freshness. International Journal of Biological Macromolecules. Elsevier B.V. 2020; 143:359-372. Doi: 10.1016/j.ijbiomac.2019.12.024.
7. Koosha M, Hamedi S. Intelligent Chitosan/PVA nanocomposite films containing black carrot anthocyanin and bentonite nanoclays with improved mechanical, thermal and antibacterial properties. Progress in Organic Coatings. Elsevier. 2019; 127(November 2018):338-347. Doi: 10.1016/j.porgcoat.2018.11.028.
8. Lemmens J, Wulijarni N, Wageningen P. Plant Resources of South-East Asia, 1991.
9. Liu J, *et al*. Morphology, structural and physicochemical properties of starch from the root of Cynanchum auriculatum Royle ex Wight. International Journal of Biological Macromolecules. Elsevier B.V. 2016; 93:107-116. Doi: 10.1016/j.ijbiomac.2016.08.063.
10. Lu DR, Xiao CM, Xu SJ. Starch-based completely biodegradable polymer materials. Express Polymer Letters. 2009; 3(6):366-375. Doi: 10.3144/expresspolymlett.2009.46.
11. Luchese CL, *et al*. Evaluation of blueberry residue incorporated cassava starch film as pH indicator in different simulants and foodstuffs. Food Hydrocolloids. Elsevier Ltd. 2018; 82:209-218. Doi: 10.1016/j.foodhyd.2018.04.010.
12. Ma Q, Wang L. Preparation of a visual pH-sensing film based on tara gum incorporating cellulose and extracts from grape skins. Sensors and Actuators, B: Chemical. Elsevier B.V. 2016; 235:401-407. Doi: 10.1016/j.snb.2016.05.107.
13. Mansour R. Natural Dyes and Pigments : Extraction and Applications, 2018. Doi: 10.1002/9781119407850.ch5.
14. Qin Y, *et al*. International Journal of Biological Macromolecules Preparation and characterization of active and intelligent packaging fi lms based on cassava starch and anthocyanins from Lycium ruthenicum Murr. International Journal of Biological Macromolecules. Elsevier B.V. 2019; 134:80-90. Doi: 10.1016/j.ijbiomac.2019.05.029.
15. Rezaei A, Nasirpour A, Fathi M. Application of cellulosic nanofibers in food science using electrospinning and its potential risk. Comprehensive Reviews in Food Science and Food Safety. 2015; 14(3):269-284. Doi: 10.1111/1541-4337.12128.
16. Stoll L, *et al*. Microencapsulation of Anthocyanins with Different Wall Materials and Its Application in Active Biodegradable Films. Food and Bioprocess Technology. 2016; 9(1):172-181. Doi: 10.1007/s11947-015-1610-0.
17. Toro-Márquez LA, Merino D, Gutiérrez TJ. Bionanocomposite Films Prepared from Corn Starch With and Without Nanopackaged Jamaica (Hibiscus sabdariffa) Flower Extract. Food and Bioprocess Technology. Food and Bioprocess Technology. 2018; 11(11):1955-1973. Doi: 10.1007/s11947-018-2160-z.
18. Wang L, *et al*. Preparation and characterization of active films based on chitosan incorporated tea

polyphenols. Food Hydrocolloids. Elsevier Ltd. 2013; 32(1):35-41. Doi: 10.1016/j.foodhyd.2012.11.034.
19. Wang X, *et al*. Preparation and characterization of antioxidant and pH-sensitive films based on chitosan and black soybean seed coat extract. Food Hydrocolloids. Elsevier Ltd. 2019; 89(August 2018):56-66. Doi: 10.1016/j.foodhyd.2018.10.019.
20. Wu S, *et al*. Electrochemical writing on edible polysaccharide films for intelligent food packaging. Carbohydrate Polymers. 2018; 186(January):236-242. Doi: 10.1016/j.carbpol.2018.01.058.
21. Yong H, *et al*. Effects of anthocyanin-rich purple and black eggplant extracts on the physical, antioxidant and pH-sensitive properties of chitosan film. Food Hydrocolloids. Elsevier Ltd. 2019; 94(January):93-104. Doi: 10.1016/j.foodhyd.2019.03.012.
22. Zhai X, *et al*. Food Hydrocolloids Novel colorimetric fi lms based on starch / polyvinyl alcohol incorporated with roselle anthocyanins for fi sh freshness monitoring. Food hydrocolloids. Elsevier Ltd. 2017; 69:308-317. Doi: 10.1016/j.foodhyd.2017.02.014.