



Microencapsulation of three natural dyes from butterfly pea, Sappan wood, and turmeric extracts and their mixture base on cyan, magenta, yellow (CMY) color concept

Musdalifa¹, Muthia Chairany¹, Nur Haliza¹, Februadi Bastian^{2*}

¹Undergraduate Student, Study Program of Food Science and Technology, Department of Agriculture Technology, Faculty of Agriculture, Hasanuddin University, Indonesia

²Lecturer, Study Program of Food Science and Technology, Department of Agriculture Technology, Faculty of Agriculture, Hasanuddin University, Indonesia

Abstract

Synthetic dyes in food can cause severe problems for health, so they need to be replaced by natural dyes. However, natural dyes are unstable, and encapsulation is one way to maintain the stability of natural dyes. This study was conducted to determine the best microencapsulation coating, storage stability, and color variations produced by butterfly pea, sappan wood, and turmeric extracts. The coating materials used were maltodextrin, carrageenan, and carboxy methyl cellulose (CMC) using the following formulations: 85% maltodextrin and 15% carrageenan (formula A) and 90% maltodextrin and 10% carrageenan (formula B) for coating butterfly pea and sappan wood extracts. Turmeric extracts were coating using 85% maltodextrin and 15% carrageenan (formula A) and 75% CMC and 25% starch (formula C). The encapsulation with maltodextrin (90%) and carrageenan (10%) was the best of encapsulation formula for butterfly pea and sappan wood extract. However, the encapsulation with maltodextrin (85%) and carrageenan (15%) was the best of encapsulation formula for turmeric extract. The green color was obtained from mixing butterfly pea and turmeric dyes in 1:4 ratio, purple from mixing butterfly pea and sappan dyes in 1:8 ratio, and orange from mixing turmeric and sappan dyes in 1:2 ratio.

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1. Introduction

Color is the first sensory attribute that will be assessed and provide a taste perception of a food product and has a significant role in attracting consumer interest (1). Food producers usually use dyes to improve their visual product quality. However, many food producers in Indonesia, especially small and medium scale food industries with generally produce traditional foods are not concerned with risks associated with the use of unauthorized coloring agents that impact consumers' health. Synthetic dyes are widely used because they are easy to produce, more stable, and provide many color variants (1). Kobylewski and Jacobson's research (2) indicated that some permitted synthetic dyes contain substances that can cause hypersensitivity reactions, carcinogenic, and genotoxicity. In addition, several types of synthetic dyes may cause food poisoning (3) and brain function decline (4). In response, the Minister of Health of the Republic of Indonesia issued Regulation Number 722/Menkes/Per/IX/88, prohibiting hazardous substances in food, including dangerous synthetic dyes.

* Correspondence : **Februadi Bastian**  februadi@unhas.ac.id

Synthetic dyes can be replaced with natural dyes from plant extracts such as butterfly pea (*Clitoria ternatea*), sappan wood (*Caesalpinia sappan* L.), and turmeric (*Curcuma domestica* V.). Butterfly pea flowers contain delphinidin of 2.16 mg/g (5) which can be used for cyan or blue pigment; sappan wood contains 45 mg/g of brazilin pigment that gives red color (6); and Turmeric contains 60-70% curcumin which provides yellow color (7–9). These pigments are basic colors which can be used to produce various colors such as the principle of CMY (cyan, magenta, and Yellow) color model used in printer ink.

Hidalgo et al. (2018) suggested that color-forming bioactive compounds in natural dyes are susceptible to oxidation during storage and are less stable than synthetic dyes (9). In addition, pH change and sunlight exposure affect the delphinidin (10), curcumin in turmeric (9) and brazilin in sappan wood (11). Several studies have been conducted to maintain the color stability of natural materials by applying microencapsulation (12–14). Microencapsulation can be carried out through continuous coating of material particles using polymers. Microencapsulation is considered an effective method in protecting bioactive materials from adverse environmental conditions, and at the same time, maintaining physical stability to facilitate the handling and storage process (15).

The types and compositions of coating materials must be examined when using microencapsulation since it has a significant effect on the result of the encapsulated dye product. Several types of films from a mixture of maltodextrin, carrageenan, carboxymethyl cellulose (CMC), and starch with a specific concentration ratio can be used as coating materials in microencapsulation (16,17). Based on the review provided above, this research aimed to determine the best coating formulation; to evaluate storage stability of microencapsulated butterfly pea, sappan wood, and turmeric extracts; and to determine pigment mixture ratio to produce the desired color variations.

2. Materials and Methods

2.1. Extraction of Butterfly Pea Extract

Two kilogram of dried butterfly peas was mashed and then extracted using maceration method. Dried butterfly peas were extracted using Ethanol 70% (1:4 w/v) at room temperature for 24 hours. The liquid and solid phase was separated by filtration using filter paper. The butterfly pea residues were re-macerated for 24 hours with ethanol 70% (1:3 w/v), then filtered. The extract was then concentrated using a rotary evaporator at 40°C for four hours, then dried using a vacuum oven at 50°C for four hours to obtain powdered extracts (19).

2.2. Extraction of Sappan Wood Extract

Two kilograms of dried powdered sappan woods were macerated in water at a 1:4 w/v ratio for 24 hours at room temperature. The mixture was occasionally stirred during maceration and filtered at the end of the maceration process. The sappan wood residue was re-macerated for 24 hours with water at a 1:3 w/v ratio, then filtered with filter paper. The extracts were then concentrated using a rotary evaporator at 40°C for four hours and dried using a vacuum oven at 50°C for four hours to get the powdered extracts (20).

2.3. Extraction and Solvent Evaporation of Turmeric Extract

Two kilograms of fresh turmeric were peeled and minced then macerated in ethanol 96% at 1:3 w/v ratio for 24 hours at room temperature. The mixture was occasionally while occasionally stirred during maceration and filtered at the end of maceration process. The turmeric residue was re-macerated for 24 hours with water at a 1:2 w/v ratio, then filtered with filter paper. The extracts were then concentrated using a rotary evaporator at 40°C for four hours and dried using a vacuum oven at 50°C for four hours to get the powdered extracts (21).

2.4. Extract Microencapsulation

2.4.1. Butterfly Pea and Sappan Wood extracts microencapsulation

In this study, to formulation of coating materials (formula A: 85% maltodextrin and 15% carrageenan; formula B: 90% maltodextrin and 10% carrageenan) were used. The coating material (10g) was dissolved in 100 ml of distilled water and homogenized at 3000 rpm for 5 minutes. After homogeneous condition was reached, 5g of powdered butterfly pea extracts and sappan wood extracts were added gently and homogenization was continued for 5 minutes. Samples were frozen for 24 hours then freeze dried for 48 hours to obtain the encapsulated butterfly pea and sappan wood dye products (16).

2.4.2. Tumeric extract microencapsulation

To formulation of coating material (formula A: 85% maltodextrin and 15% carrageenan; formula C: 75% CMC and 25% starch) were used. The coating materials (10g) was dissolved in 100 ml of distilled water and homogenized at 3000 rpm for 5 minutes. After homogeneous condition was reached, 5g of turmeric extracts were added gently and homogenization was continued for 5 minutes. Samples were frozen for 24 hours then freeze dried for 48 hours to obtain the encapsulated turmeric dye products (16).

2.5. Color Design

Each of the encapsulated natural dye products (0.5g) were mixed with 50 mL water to obtain three basic colors (cyan, magenta, yellow). Two of three basic colors were mixed slowly (drop by drop) to obtain secondary colors. The encapsulated butterfly pea (cyan) and turmeric (yellow) extracts were mixed to obtain green color. The encapsulated sappan wood (magenta) and turmeric (yellow) extracts were mixed to obtain orange color. The encapsulated butterfly pea (cyan) and sappan wood (magenta) extracts were mixed to obtain purple color. The amount of each extract used to obtain the secondary color were noted.

2.6. Determination Parameters

2.6.1. Moisture Contents

Moisture contents of encapsulated natural dye products (0.5g each) were analyzed using the oven method at 105°C. The samples were first dried for three hours, cooled in desiccator for 15 minutes and then weighed. The samples were dried again for 30 minutes, cooled for 15 minutes and weighed. This process was repeated until a constant weight was reached with a maximum successive weighing difference of 0.2 mg.

2.6.2. Antioxidant Activity

An antioxidant test was carried out by the free radical scavenging activity method. A solution of 2,2-diphenyl-1-picrylhydrazyl (DPPH) was prepared with a concentration of 50 M or 0.197 mg of DPPH dissolved to 100 mL of methanol. A total of 2 ml of the DPPH solution was mixed with 2 mL of encapsulated natural dye dissolved in 200, 250, 300, 350, and 400 ppm. After that, the mixture was incubated in a dark place for 15 minutes at room temperature, and the absorbance was measured at a wavelength of 515 nm. The measurements were carried out in triplicate. The free radical scavenging activity was determined using equation (1).

$$\% \text{ Inhibition} = ((A_b - A_a) / A_b) \times 100 \quad (1)$$

Description:

A_a = Sample absorbance (sample + DPPH)

A_b = Control absorbance (methanol + DPPH).

2.6.3. Total Dissolved Solids

The encapsulated natural dye was weighed (2.25g) and dissolved in water to a total volume of 100 mL. Total dissolved solids (ppm) were measured using a TDS Meter.

2.6.4. Intensity and Storage stability of color

The stability of color of natural dye during storage was determined by dissolving one gram in 100 mL of water. The solution was divided into two parts, one part was stored at room temperature in open container, and the other part was stored in close container. The storage stability and intensity test were carried out by measuring the color intensity of the dye product consecutively for seven days. The results obtained are expressed as L*, a*, and b*, which represent lightness (L*), redness (+a*), greenish (-a*), yellowish (+b*), and bluish (-b*). L*a*b* value obtained were to CMY color number using software color converter available in the colorimeter instrument (22).

3. Results and Discussions

3.1. Moisture Content

The effect of water content is very significant on the shelf life of products. Therefore, it is very important to reduce the moisture content of the encapsulated product. Natural dyes produced by the same drying method have different water content. Figure 1 showed that moisture contents of butterfly pea and sappan wood which were encapsulated with mixture of maltodextrin (85%) and carrageenan (15%) (formulation A) and maltodextrin (90%) and carrageenan (10%) (formulation B) were not significantly different. However, the moisture content of turmeric which was encapsulated with maltodextrin (85%) and carrageenan (15%), and CMC (75%) and starch (25%) was significantly different. The use of CMC (75%) and starch (25%) (formulation C) as encapsulating materials for turmeric extract resulted in higher moisture. Carrageenan and CMC form gel which under certain conditions can entrap water (23,24), which can cause higher moisture content. The moisture contents of all encapsulated samples of natural dye were <12% that meets SNI 01-3709-1995 regarding powdered materials.

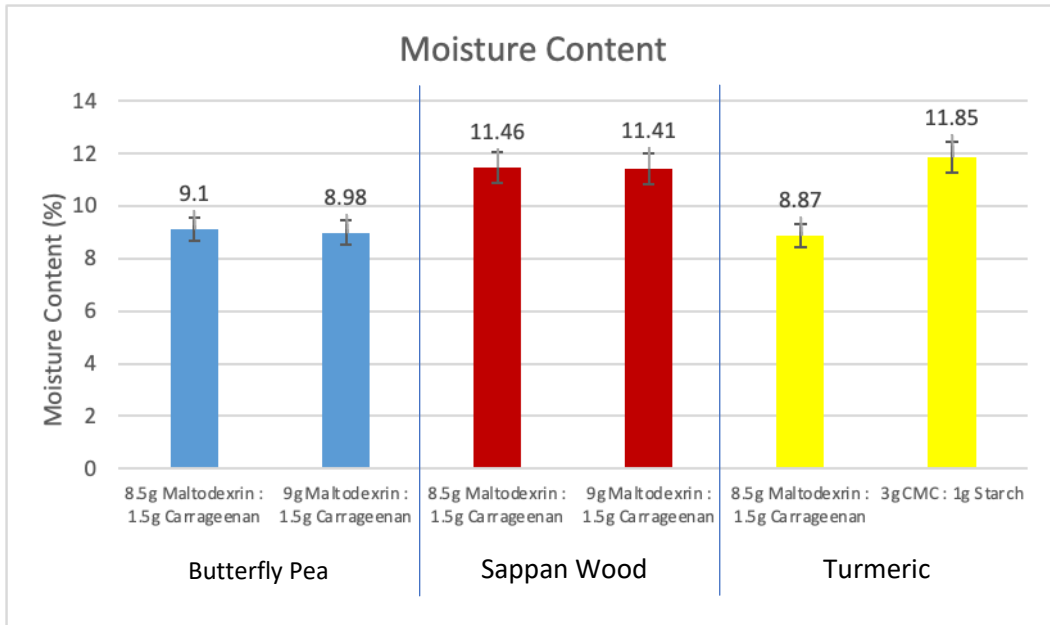


Figure 1. Effect of coating on butterfly pea, sappan wood, and turmeric extracts on the moisture content.

3.2. Total Dissolved Solids

Total dissolved solids (TDS) must be known in order to determine how much solids are dissolved when the encapsulated extract was dissolved in water. In the total dissolved solids analysis, in addition to inorganic and organic substances, microgranular substances will also be detected. Figure 2 shows that the average TDS in the encapsulated butterfly pea sample was higher than those in the sappan wood and turmeric. In addition, the comparison of maltodextrin:carrageenan treatment was not significant in the encapsulated of butterfly pea and sappan wood extracts, but significant in the encapsulated turmeric extracts. This shows that the encapsulation of the sappan wood extract forms a microencapsulation which is less soluble with water.

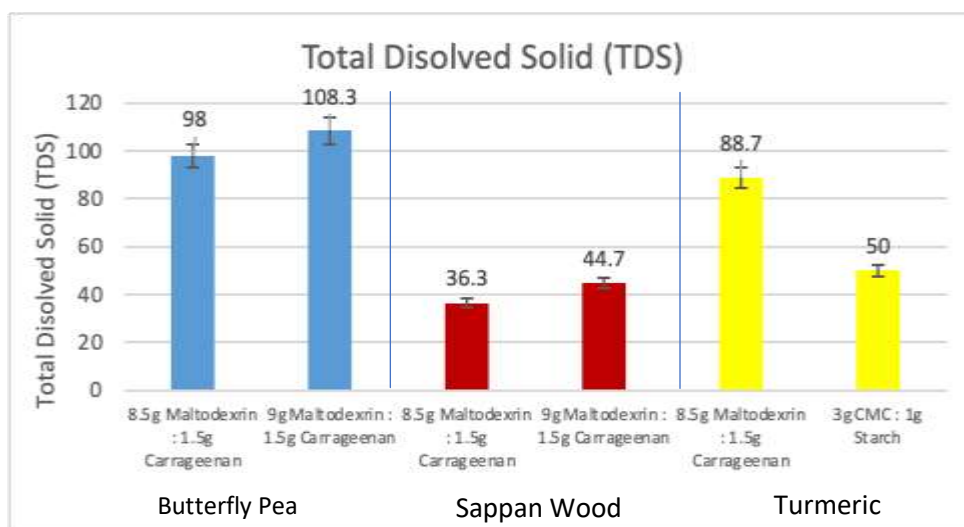


Figure 2. Effect of coating on butterfly pea, sappan wood, and turmeric extracts on total dissolved solids value.

3.3. Antioxidant Activity (IC_{50})

One of the advantages of natural dyes compared to synthetic dyes is that they have antioxidant properties that are beneficial to health. The antioxidant activity was determined using the free radical scavenging activity (IC_{50}) method. Figure 3 shows that antioxidant activity (IC_{50}) of butterfly pea which was encapsulated with a mixture of maltodextrin (85%) and carrageenan (15%) (formulation A), maltodextrin (90%) and carrageenan (10%) (formulation B), and turmeric which was encapsulated with maltodextrin (85%) and carrageenan (15%) (formulation A) and CMC (75%) and starch (25%) (formulation C) were not significantly different. However, antioxidant activity of sappan wood which was encapsulated with a mixture of maltodextrin (85%) and carrageenan (15%) (formulation A), and maltodextrin (90%) and carrageenan (10%) (formulation B), was significantly different. Substances with lower IC_{50} value have higher antioxidant activity (15). This indicates that maltodextrin (90%) and carrageenan (10%) coating treatment can coat the particles better than other formulas which result in higher antioxidant activity.

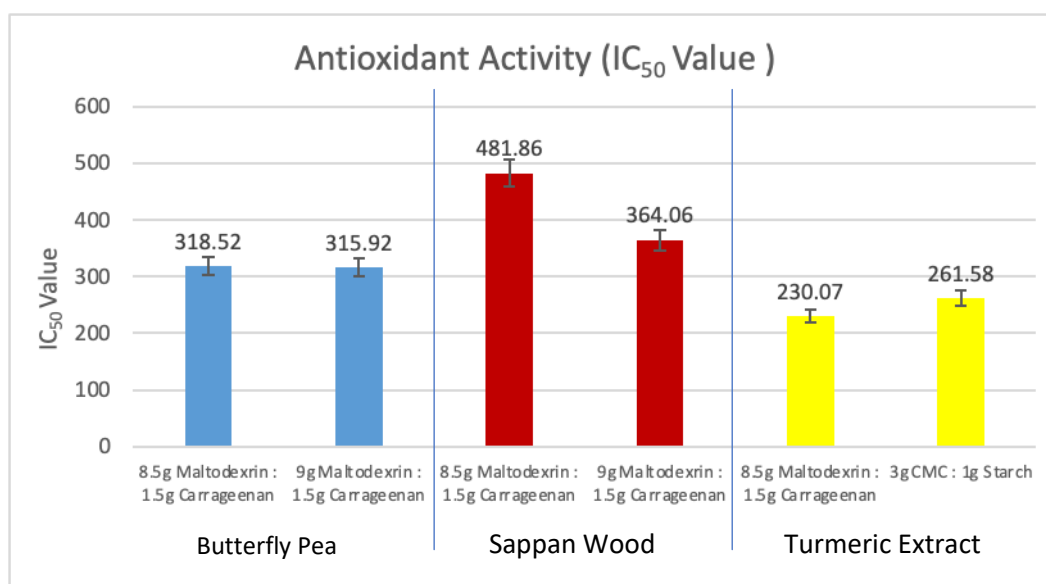


Figure 3. Effect of coating ratio on butterfly pea, sappan, and turmeric extracts on the IC_{50} value

3.4. Color Intensity

The color intensity parameter was measured to see the effect of the coating material on the color intensity of natural dyes. The benefits of coating materials in the encapsulation process of bioactive compounds include increasing color stability. However, encapsulation can reduce the color intensity of natural dyes. Figure 4 showed that color intensity of butterfly pea, which was encapsulated with a mixture of maltodextrin (85%) and carrageenan (15%) (formulation A), maltodextrin (90%) and carrageenan (10%) (formulation B), and turmeric which was encapsulated with maltodextrin (85%) and carrageenan (15%) (formulation A) and CMC (75%) and starch (25%) (formulation C) were not significantly different. However, the sappan wood, which was encapsulated with a mixture of maltodextrin (85%) and carrageenan (15%) (formulation A), and maltodextrin (90%) and

carrageenan (10%) (formulation B), was significantly different. This indicate that maltodextrin (90%) and carrageenan (10%) could coat the dye particles well. This is in accordance with the findings by Ribeiro and Veloso which indicate that microencapsulation can maintain the color from degradation and external influence if the particles are coated well (25).

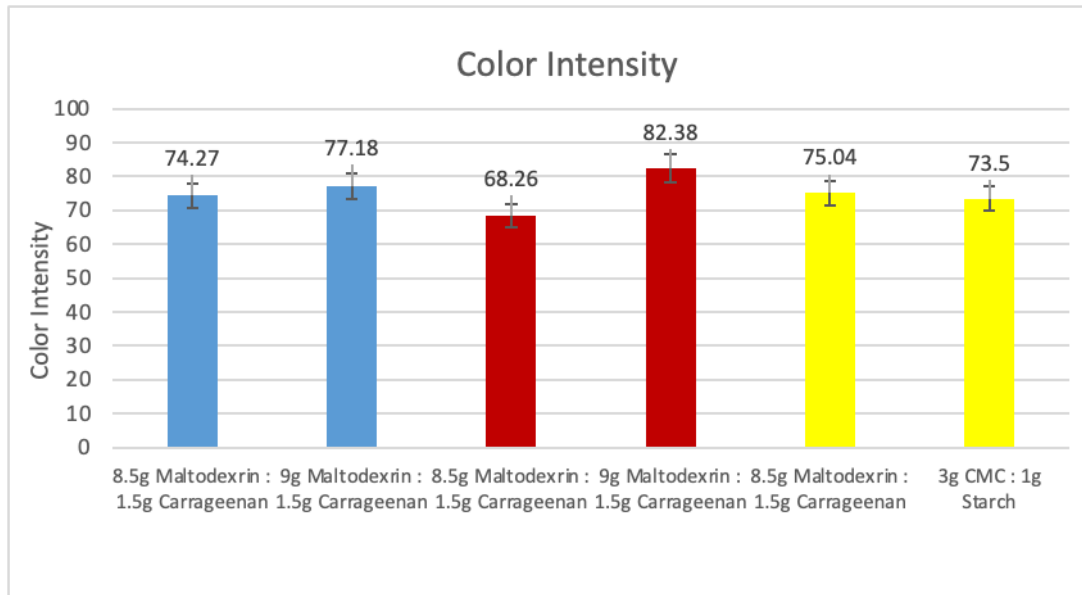
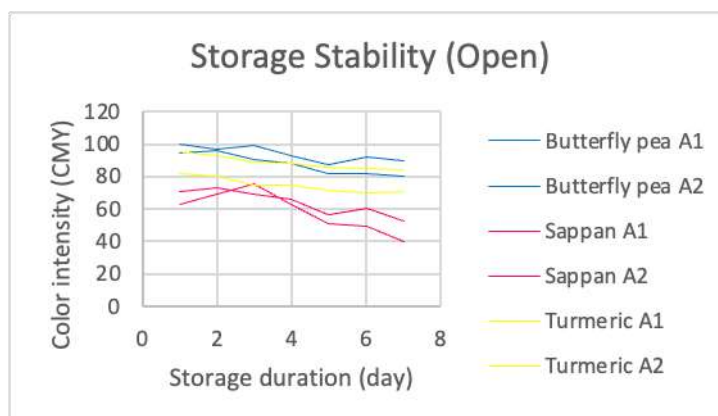
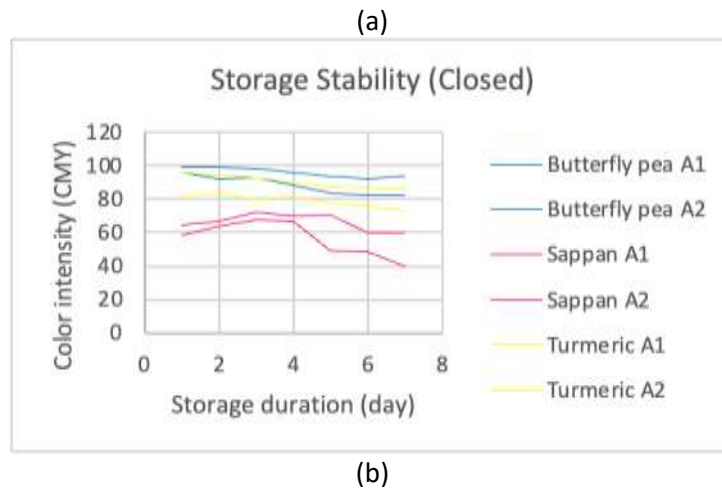


Figure 4. Effect of coating ratio on butterfly pea, sappan, and turmeric extracts on the color intensity

3.5. Storage Stability

The color stability of the encapsulated powdered dye product was tested by assessing the color intensity for seven days. Visually, all samples did not show significant change during storage. The color test results also showed the CMY value was relatively stable, except for the sappan wood encapsulated with maltodextrin (85%) and carrageenan (15%). In this case the value of magenta number significantly decrease and indicated that the maltodextrin 85% and carrageenan 15% treatment cannot coat the particles well. The imperfection of the coating process causes the compounds in natural dyes to be oxidized, resulting in a decrease in color intensity. Lei et al. (2018) suggested that oxidation by oxygen can cause loss of biological function of plant compounds (26). This can be seen in Figure 5.





(b)
Figure 5. Effect of coating on the extracts of butterfly pea, sappan, and turmeric on storage stability: (a) The storage










3.6. Color Design

The encapsulation with maltodextrin (90%) and carrageenan (10%) was the best of encapsulation formula for butterfly pea and sappan wood extract. However, the encapsulation with maltodextrin (85%) and carrageenan (15%) was the best encapsulation formula for turmeric extract. The encapsulated butterfly pea (cyan) and turmeric (yellow) extracts were mixed to obtain green color. The encapsulated sappan wood (magenta) and turmeric (yellow) extracts were mixed to obtain orange color. The encapsulated butterfly pea (cyan) and sappan wood (magenta) extracts were mixed to obtain purple color. The amount of each extract used to obtain the secondary color were shown in Table 1 and the result is shown in Figure 6.



(a) (b) (c)
Figure 6. Color Designs of the encapsulated natural primer dye product: (a) Combination of the cyan butterfly pea and yellow turmeric dye into green color; (b) Combination of the cyan butterfly pea and magenta sappan dye into orange color; (c) Combination of the magenta sappan dye and yellow turmeric dye into orange color.

Table 1. Color Design Ratio and the resulting secondary color.

No.	Primary Color (CMY)	Ratio	Combination Result
1		1	
		4	
2		1	
		8	
3		2	
		1	

4. Conclusions

The encapsulation with maltodextrin (90%) and carrageenan (10%) was the best encapsulation formula for butterfly pea and sappan wood extract. However, the encapsulation with maltodextrin (85%) and carrageenan (15%) was the best of encapsulation formula for turmeric extract. The color test results also showed the CMY value was relatively stable, except for the sappan wood encapsulated with maltodextrin (85%) and carrageenan (15%). In addition, from the color mixing, the green color was obtained from mixing butterfly pea and turmeric dyes in 1:4 ratio, purple from mixing butterfly pea and sappan dyes in 1:8 ratio, and orange from mixing turmeric and sappan dyes in 1:2 ratio.

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Author Contributions

Musdalifa and Februadi Bastian conceived and designed the experiments; Musdalifa Muthia Chairany, and Nur Haliza performed the experiments; Musdalifa and Nur Haliza analyzed the data; Muthia Chairany, Nur Haliza, and Februadi Bastian contributed reagents/materials/analysis tools; Musdalifa and Muthia Chairany wrote the paper.

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