Food Colorant from Natural Sources: Suji Leaves (*Pleomale angustivoria*) and Dragon Fruit Peel (*Hylocereus sp.*)

Yeni Rahmawati¹, Intan Febrianti², Detya Firly Priadi³, Orchidea Rachmaniah⁴

Received June 25, 2023; Revised August 15, 2023; Accepted September 28, 2023 DOI: <u>10.12962/j2964710X.v4i1.17299</u>

Abstract— Synthetic colorants have become more prevalent than natural colorants due to their ease of acquisition, affordability, stable and consistent colors, and availability in various options. However, long-term excessive use of synthetic colorants can potentially lead to carcinogenic or mutagenic effects. As a result, research on natural colorants remains promising. Suji leaves (*Pleomale angustivoria*), and dragon fruit (*Hylocereus sp.*) peel are potential sources of natural dyes. Suji leaves contain chlorophyll, which is commonly used as a natural colorant in food products, while dragon fruit peel contains betacyanin, an agricultural waste that is yet to be widely utilized. Chlorophyll imparts a green color, while betacyanin contributes a red color. These natural colorants offer safer and healthier alternatives to synthetic ones. Therefore, this study extracts a suji leaf (Pleomale angustivoria) and dragon fruit (Hylocereus sp.) are extracted, obtaining their natural colorants by simple extraction method. The research will be conducted in two stages: extracting active natural colorant compounds, namely chlorophyll and betacyanin, followed by solvent evaporation. The optimum maceration time for dragon fruit peel is 24 hours, while the optimum maceration time for suji leaves is known to be 3 x 24 hours. The maceration of suji leaves and dragon fruit peel was carried out at room temperature. The containers were airtight and protected from light. The green color (chlorophyll) of suji leaves and the red color (betacyanin) of dragon fruit peel are sensitive to temperature, air, and light. Experiment results show that betacyanin extracts with organic acid

stabilizers, citric acid, and ascorbic acid have the same λ_{max} of 325 nm. The betacyanin extract without organic acid has $\lambda_{max} = 525$ nm. Adding ascorbic acid (75 mL) up to pH 4.5 is more suitable for increasing the betacyanin yield. 96%. In contrast, using ascorbic acid with ethanol as solvent gave a higher yield value of 21.42% than citric acid. Ethanol, 96% v/v, is more suitable as a solvent for chlorophyll than acetone,85 v/v. The yield of chlorophyll from suji leaves was 22.1% (total chlorophyll 2.36 mg/L) with 96% ethanol solvent. While when using Acetone 85% as a solvent, a lower yield ca. 17% (1.90 mg/L).



Keywords-Dragon fruit peel, Extraction, Rotary evaporator, Suji leaf, Natural dyes

I. INTRODUCTION

Suji leaves have several characteristics that are better than other chlorophyll sources. Suji leaves do not contain a gelling component like grass jelly extract. The presence of gelling hydrocolloids will block the release of chlorophyll from the leaves during the extraction process. Moreover, suji leaves do not have toxic components such as cyanide in cassava leaves; therefore, suji leaf chlorophyll extract is safe to use as a food additive (BTP) [6].

Dragon fruit skin contains betacyanin, almost the same as the fruit flesh. Hence, the waste from dragon fruit

¹ Departement of Chemical Engineering, Institut Teknoogi Sepuluh Nopember, Sukolilo, Surabaya, 60111, Indonesia. E-mail: rifqah_18des@chem-eng.is.ac.id.

² Chemical Engineering, Institut Teknoogi Sepuluh Nopember, Sukolilo, Surabaya, 60111, Indonesia. E-mail: intanfebrianti60@gmail.com.

³ Chemical Engineering, Institut Teknoogi Sepuluh Nopember, Sukolilo, Surabaya, 60111, Indonesia. E-mail: detvapriadi@gmail.com.

⁴ Food Engineering Programme, Chemical Engineering, Institut Teknoogi Sepuluh Nopember, Sukolilo, Surabaya, 60111, Indonesia. Email: orchidea@chem-eng.its.ac.id.

(dragon fruit skin) can be used as a source of betacyanin dye [4].

Based on those problems, the innovation of dyes from natural materials that are safe and easy to apply as well as synthetic is needed. At the same time, liquid dyes are less efficient in storage and transportation and have a shorter shelf life than concentrated filtrate preparation. So to overcome this, suji leaf extract and dragon fruit skin are processed as filtrate to obtain pure filtrate without any solvent content using a rotary evaporator.

II. METHOD

A. Tools and Material

Various simple glasses were used in the study, a desiccator, pycnometer filter paper, cotton cloth, rotary vacuum evaporator, blender, and sieve. UV-VIS spectrophotometer, rotary vacuum evaporator, and high-performance liquid chromatography (HPLC).

85% Acetone and 96% ethanol (Merck, Darmstadt, Germany) were used as organic solvents. While citric acid and ascorbic acid used as stabilizers in betacyanin dyes were purchased from a cake supplier in the Mulyorejo area, Surabaya, East Java. Suji leaves that are approximately two months old were used in this study. On the other hand, dragon fruit peel came from dragon fruit sold in various traditional markets in the Sedati area, Sidoarjo, East Java.

B. Research Procedures

1) Suji Leaves Natural Dye Extraction

The production of natural colors from suji leaves is (1) chlorophyll extract. Extraction was done by cutting 80 g of suji leaves into small pieces, crushing them with a blender, and then macerating them with 85% acetone and 96% ethanol for 3x24 hours at room temperature. Each extract is filtered through a cloth to separate the pulp from the filtrate. The suji leaf extract is then concentrated in a rotary evaporator. The concentrated portion containing chlorophyll was then analyzed for total chlorophyll using a UV-VIS spectrophotometer. The yield of the concentrated extract, the density of the concentrated extract, and the Thin Layer Chromatography TLC test were then analyzed.

2) Extraction of Dragon Fruit Skin Natural Colourant

While the manufacture of dyes from dragon fruit peels includes betacyanin extraction carried out using 500 g dragon fruit skin, cut into small pieces, and then crushed with a blender. Sample 1 was then macerated in 96% ethanol solvent for 24 hours at room temperature, and then 1% ascorbic acid was added in 75 ml volumes until the pH of the solution reached 4.5. Sample 2 was macerated in 96% ethanol solvent for 24 hours at room temperature, and then 1% citric acid was added to 55 ml until the pH of the solution was 4.5. Each extract was filtered through a filter cloth to separate the pulp from the filtrate. The dragon fruit peel filtrate containing ethanol solvent was then concentrated in a rotary evaporator to remove the solvent, resulting in a concentrated extract (betacyanin filtrate).

The betacyanin filtrate was then analyzed for absorbance using a UV-VIS spectrophotometer, yield analysis, density analysis, and HPLC test compared with the betacyanin powder color standard.

The preparation of natural colors by other methods was carried out as a comparative method; namely: 100 g of suji leaves and 500 g of dragon fruit peel were washed and drained. They were then cut to \pm 0.5 cm and dried in the sun. Once the fresh material was dried, it was pulverized in a blender to produce a dry natural coloring powder. The same test analysis was also carried out on the two dry natural dye products (suji leaf & dragon fruit peel), namely: product yield, solubility analysis, total chlorophyll analysis for suji leaf dye, absorbance analysis for dragon fruit peel dye and moisture content analysis.

Both extraction methods were carried out at room temperature (± 30 °C) for 24 hours for the amplification method. The solvent evaporation conditions were 60 °C rotary evaporators at 40 rpm.

C. Analysis Procedure

1) Analysis of Moisture Content by Oven Method [1]

The porcelain cup was dried in an oven for 15 minutes and then cooled in a desiccator for 10 minutes. 3 g of sample was added to the cup and dried in an oven (105 oC, 6 hours). The cup was then removed from the oven, cooled in a desiccator for 15 minutes (to room temperature), and weighed. This step was repeated until the weight of the dried sample was constant. The moisture content and total solids were calculated as follows:

Water content (%bb) =
$$\frac{a - (b - c)}{a} \times 100\%$$
(1)

Percent bb is the moisture content per wet material (%), a is the weight of the sample before drying (g), b is the weight of the sample + cup after drying (g), and c is the weight of the dried empty cup (g).

2) Solubility Test In Ethanol or Water [2]

A 0.2 g sample was weighed and dissolved to 10 mL with 96% ethanol or distilled water. Heat the sample to 76 oC while stirring. Filter the sample with filter paper that has been weighed before. Next, the filter paper was dried in the oven until the weight of the filter paper was constant. Solubility was calculated based on the following formula (2):

Solubility (%w) =
$$\frac{S - (K_2 - K_1)}{S} \times 100\%$$
 (2)

S represents the wet weight of the sample (g), K_1 represents the dry filter paper (g), and K_2 represents the weight of the filter paper after filtering.

3) Chlorophyll Qualitative Analysis [16]

A qualitative test for chlorophyll-A and chlorophyll-B in the sample was performed by thin-layer chromatography (TLC) [16]. A total of 0.1 g concentrated extract was dissolved in 2 mL of Acetone, and the dissolved sample was photographed on a silica plate measuring (3.5 cm x 9.0 cm). The plate was developed in a closed chamber using a mobile phase of n-hexane: acetone = 70/30 (v/v) up to the specified limit. The resulting color spots were compared with the results in [16].

4) Quantitative Analysis of Chlorophyll-A and Chlorophyll-B [26]

The quantitative analysis of chlorophyll A and B was carried out spectrophotometrically by weighing exactly 1,0 mg of the concentrated extract sample and dissolving it in 10 ml of distilled water until thoroughly mixed. The resultant sample is ready to measure the absorbance value by UV-VIS spectrophotometry at 645 nm and 663 nm. The absorbance value at each wavelength was used to calculate the chlorophyll A (mg/L) and chlorophyll B (mg/L) contents using the following equations (3) and (4):

Chlorophyll-A (mg/L) =
$$12,72A_{663} - 2,59A_{645}$$
 (3)
Chlorophyll-B (mg/L) = $22,9A_{645} - 4,67A_{663}$ (4)

5) Quantitative Analysis of Total Chlorophyll [1]

At 1000 rpm for 10 minutes, then collect the supernatant substance. Take 1 mL of the supernatant and dilute it to 10 mL with distilled water. The diluted solution was then measured for absorbance at a wavelength of 663 μ m and 645 μ m using a UV-Vis spectrophotometer. The concentration was calculated using the equation [26]:

$$C (mg/L) = 20,31x A_{645} + 8,05 A_{663}$$
(5)

C represents total chlorophyll concentration, A645 nm represents the absorbance value of chlorophyll extract at 645 nm wavelength, and A663 nm represents the absorbance value of chlorophyll extract at 663 nm wavelength.

As for the dry extract of suji leaves, 1 mg of powder was dissolved and diluted to 10 mL in a volumetric flask. The resulting solution was centrifuged (1000 rpm, 10 min). The resulting supernatant was measured for chlorophyll concentration at 663 μ m and 645 μ m wavelengths with a UV-Vis spectrophotometer. The chlorophyll concentration was calculated using equation (5).

6) Spectrophotometric Analysis of Betasianin [4]

Before measuring the absorbance of betacyanin, the maximum wavelength of betacyanin was first determined for the concentrated extract obtained. One mg of the concentrated extract obtained was diluted to 5 mL in a volumetric flask with distilled water. Absorbance measurements were then performed at different wavelengths (300-700 nm) with 50 nm as an interval. The absorbance measured at each wavelength was recorded and plotted using Absorbance vs. Wavelength. The wavelength with the highest absorbance was read as the maximum for betacyanin.

The concentrated extract of dragon fruit peel obtained was centrifuged (1000 rpm, 10 min), and 1 mL of the

supernatant was taken and diluted in a volumetric flask to 10 mL with distilled water. Absorbance measurements were performed at the maximum wavelength obtained in the previous step. As for the dried powder of the dragon fruit peel, 1 mg was dissolved and diluted in a volumetric flask with distilled water to make up to 10 ml; 1 ml was taken, and the absorbance was measured at the appropriate maximum wavelength.

7) Prediction of Total Betasianin Content [14]

The total betacyanin content can be predicted using spectrophotometry [14]. The concentrated extract sample was weighed exactly 1 mg and dissolved in 10 mL of distilled water. The absorbance was then measured at the appropriate maximum wavelength for each sample. The samples of betacyanin with ascorbic acid have an absorbance value of 350 nm, while the samples of betacyanin with the addition of citric acid have an absorbance value of 325 nm. The measured absorbance was then used to calculate the total betacyanin in the extract (6).

Betacyanin total =
$$\frac{A x (MW) x V x (DF) x 100}{E x L x W}$$
(6)

A denotes absorbance at λ_{max} , L denotes 1 cm, DF denotes dilution factor, V denotes the volume of extract (mL), E denotes the weight of dragon fruit peel base (g), and MW denotes 550 gr/mol.

8) Calculation of Yield

The yield calculation for the concentrated extract obtained is according to equation (7) below:

yield (%) =
$$\frac{filtrate volume (mL)}{weight of raw material} \times 100\%$$
 (7)

As for the dry dye powder, the calculation of the yield is done according to equation (8):

yield (%) =
$$\frac{dry \ weight \ of \ obtained \ material}{weight \ of \ raw \ material} \ge 100\%$$
 (8)

III. RESULTS AND DISCUSSION

A. Extracting Green Color From Suji Leaves (Pleomale angustivora)

Before measuring the absorbance of betacyanin, the wavelength of betacyanin was first determined for the concentrated extract obtained. The maceration extraction method was used to extract the green dye in suji leaves (for 72 hours). The maceration was carried out under airtight conditions, minimizing the presence of light at room temperature using a magnetic stirrer. This step was done to reduce the green coloring being degraded. Oxygen (O₂) in the extraction process triggers chlorophyll degradation and oxidative stress [9].

Similarly, the presence of light is also a cause of chlorophyll degradation [9]. Adding stirring in maceration extraction will increase the collision between particles to turbulence during extraction. As a result, the thickness of the layer between the surface of the solid and the liquid is thinned, and the mass transfer surface area is increased.



When both 85% acetone and 96% ethanol were used as solvents, there was a color change in the concentrated chlorophyll extract (Table 1). In general, the concentrated chlorophyll extract produced is bluish-green [6]. However, the concentrated chlorophyll extract was brownish-green after evaporation in the 85% acetone and 96% ethanol solvents. Therefore, it is assumed that chlorophyll is degraded during drying. High temperatures during evaporation may induce chlorophyll degradation. In general, the enzymatic degradation of chlorophyll is enhanced by high temperatures. The formation of chlorophyll derivatives upon heating can be classified into two groups based on the presence or absence of Mg^{2+} atoms in the tetrapyrrole center.

Chlorophyll derivatives that contain Mg^{2+} are green. Chlorophyll derivatives that do not have Mg^{2+} are olivebrown or brownish-green. Two H⁺ atoms can easily replace the Mg^{2+} atom in chlorophyll. This results in the formation of pheophytin, which is olive brown. The appearance of pheophytin in chlorophyll A is more common than the formation of pheophytin in chlorophyll B. The more excellent stability of pheophytin-B is due to the electron-withdrawing effect of the C-3 formyl group contained in chlorophyll-B (Figure 1) [21].

In addition to thermal effects, chlorophyll degradation in suji leaf crude extract is also influenced by pH. In general, chlorophyll is heat stable at alkaline pH (pH = 9.0). However, it is unstable at acidic pH (pH = 3.0). If the pH is lowered, acid can be released. The acid condition has a very detrimental effect as it can accelerate the rate of chlorophyll degradation. Therefore, pH measurements were carried out on the crude chlorophyll extract and the

J. Fund. Appl. Chem. Eng., **2023**, *4* (1), 1-8 eISSN 2964-710X

concentrated chlorophyll extract (Table 1) because the concentrated chlorophyll extract produced had a visual appearance that did not correspond to the literature [6]. The decrease in pH in the extract and both extracts, which are relatively acidic in pH (Table 1), indicates the degradation of chlorophyll [8]. Chlorophyll degradation occurs by converting chlorophyll to pheophytin (Figure 1). The degradation leads to the release of organic acids, causing the pH of the extract obtained to decrease.



Figure 1. Reaction of chlorophyll degradation due to heating [21].

The process of chlorophyll degradation itself begins with the decay of green plants. In this process, Mg²⁺ ions are released from the chlorophyll structure to form ferritin, followed by the release of phytol groups. Chlorophyll A, initially green-blue, is converted to ferritin A, which becomes grey. Meanwhile, chlorophyll that was originally yellow-green becomes pheophytin-B, which is brown. Acidic conditions also influence the conversion of chlorophyll to pheophytin. During processing, organic acids are released from the suji leaves, causing the pH to drop [21]. Efforts to control the requirements during the extraction process are made by adding alkaline compounds to maintain the chlorophyll structure during the extraction process. Alkaline materials added to the solution as stabilizers include NaHCO3, NaOH, Na2HPO4, and MgCO₃ [5]. Therefore, considering the extracts' color observation and pH measurement, both extracts produced in this study experienced browning [13].

The obtained yields of chlorophyll A and B in the extracts are in Table 2. Ethanol, 96% v/v, gave the highest yield, 22.06% (v/b), compared to 85% acetone (Table 2). Ethanol is more suitable for the extraction of chlorophyll due to is a polar-protic solvent. Solvents that can provide

Journal of Fundamentals and Applications of Chemical Engineering

OH- ions, making it easier to interact with polar functional groups on chlorophyll [11]. Meanwhile, Acetone is a polar aprotic solvent that cannot provide OH- ions, so the solubility of chlorophyll in Acetone will be relatively lower than in ethanol. In addition, ethanol is more water soluble than Acetone. Therefore, the solubility of chlorophyll from suji leaf chloroplast in ethanol will be easier.

TABLE 2.				
CHLOROPHYLL ANALYSIS RESULTS OF CONCENTRATED SUJI LEAF				
EXTRACTS WITH DIFFERENT SOLVENTS.				
Solvent	Chlorophyll Concentration (mg/mL)			Yield (v/b)
~~~~	А	В	Total	
Ethanol 96 % v/v	1.4725	0.8861	2.3586	9.40
Acetone 85 % v/v	1.0914	0.8065	1.8979	8.46

Thin-layer chromatography (TLC) was used to determine the presence of chlorophyll-A and chlorophyll-B in suji leaf extracts (section 2.3.3) (Figure 2). Both chlorophyll-A and chlorophyll-B dissolved well in the two solvents used, 96% ethanol and 85% acetone.



Figure 2. Results of TLC analysis using 85% acetone (a) and 96% ethanol (b) solvents on a concentrated extract of suji leaves.

## B. Extraction of red color from dragon fruit peel (Hylocereus sp.)

Maceration extraction was also carried out on dragon fruit peel. However, the extraction time was only 24 hours. This time was sufficient to extract maximum betacyanin from the dragon fruit peel, as indicated by the pale color of the dragon fruit peel from the 24-hour maceration (Table 3).

Betacyanin has good solubility in ethanol solvent, as its polarity is similar to that of ethanol. Therefore, ethanol was chosen as the solvent [12]. A magnetic stirrer (350 rpm) performed maceration at room temperature. Maceration was carried out under airtight conditions, and the light was avoided to prevent the degradation of betacyanin in dragon fruit peel extract [2]. Oxygen also plays a role in the photocatalytic degradation of betacyanin pigments. The presence of oxygen (O2) in the extraction process affects the stability of betacyanin, which is sensitive to oxidation [18]. The stability of betacyanin is also affected by light (UV or visible light) and temperature. Light excites betacyanin chromophore electrons to higher energy levels ( $\pi^*$ ), resulting in lower activation energy and higher molecular reactivity [23] [24] [10]. Therefore, the effect of pH, temperature, and light on the red dye (betacyanin) was also observed (Table 4).

TABLE 3.			
THE PHYSICAL APPEARANCE OF DRAGON FRUIT RIND BEFORE AND AFTER MACERATION.			
Time	Description		
Before Maceration	The peel of the dragon fruit, which has been cut into small pieces and blended in a blender, has a visually intense red color.		
After 24 hours of maceration	After maceration for 1x24 hours, the dragon fruit began to fade, visually turning whitish pink.		
	TABLE 4.		

RESEARCH INTO THE EXTRACTION OF DRAGON FRUIT PEEL			
Condition (Stabilizer-	the pH of the crude extract		pH Concentrated extract
Solvent)	before	after	
Ethanol 96 % +	57	4.46	3 77
Citric Acid	5.7	4.40	5.22
Ethanol 96 % +	57	1 16	2.40
Citric Acid*	5.7	4.40	5.40
Ethanol 96 % +	57	4 57	2 20
Ascorbic Acid	3.7	4.37	5.50

* The resulting extract undergoes a color change

The crude extract from the maceration was added to stabilizing agents in organic acids, citric acid, or ascorbic acid. 55 ml of citric acid or 75 ml of ascorbic acid are added until a pH of  $\pm 4.5$  is obtained. Betacyanin is stable at acidic to neutral pH but is most stable at pH = 4.5. Therefore, pH = 4.5 is assumed to be the optimum pH for maintaining betacyanin stability [4]. Adding ascorbic acid and citric acid also removes oxygen from the environment. However, after the evaporation process, the concentrated betacyanin extract experienced a decrease in pH (Table 4). The decrease in pH of the extract is due to the volume of the extract being reduced by evaporating the solvent so that the extract becomes concentrated, and the pH of the extract becomes lower [20].

As with the Suji leaf extract results (Table 1), decreasing the pH of the betacyanin concentrated extract was also followed by a change in color, primarily when citric acid was used as a stabilizer (Table 5). Degradation of betacyanin may also occur by adding organic acid stabilizers, citric acid, or ascorbic acid. Acidic environmental conditions in concentrated extracts of betacyanin (Table 4) can lead to the recondensation of betalamic acid with amine groups from residual additions, isomerization, and dehydration of the C15 group [21] [3].

The degradation of betacyanin can occur in acidic or high-temperature environments; it leads to isomerization, decarboxylation, or cleavage, which results in a gradual reduction of the purple-red color [3].

TABLE 5.			
COLOR CHANGES OCCUR IN EXTRACTS OF DRAGON FRUIT PEEL (HYLOCEREUS SP).			
Condition (Stabilizer- Solvent)	Crude Extract	Concentrated Extract	
Ethanol 96 % + Citric Acid	Intense red color	Brownish red color	
Ethanol 96 % + Citric Acid*	Orange color	Brownish red color	
Ethanol 96 % + Ascorbic Acid	Intense red color	Brownish red color	

^{*} The resulting extract undergoes a color change

Exposure of crude dragon fruit rind extract to light will also cause betacyanin to degrade. Betacyanin degradation can also be triggered by light. The presence of light can trigger the photochemical degradation of betacyanin [7]. Betacyanin is unstable when exposed to sunlight [17]. There may also be a pause/waiting period before the following process occurs. These things may have triggered betacyanin oxidation events.

The discoloration of betacyanin in the concentrated extract is thought to be caused by betacyanin degrading during evaporation. Betacyanin is sensitive to high temperatures [24]. Therefore, temperatures <65 °C are recommended when extracting and purifying betacyanin [23]. Contact of betacyanin with light is also thought to occur during the evaporation process. Before measuring total betacyanin with the spectrophotometer, the maximum wavelength of betacyanin was first determined ( $\lambda_{max}$ ) as described in subtopic 2.3.5. The maximum wavelengths for the betacyanin extracts obtained are shown in Figure 3.

The betacyanin ethanol + ascorbic acid extract has  $\lambda_{max} = 350$  nm, while the concentrated betacyanin ethanol + citric acid extract, whether discolored or not, both have  $\lambda_{max} = 325$  nm.

Betacyanin has a characteristic wavelength at 534-552 nm ( $\lambda$ max) [4]. The existence of a shift  $\lambda_{max}$  (Figure 3) may be due to the different pH of the concentrated extract (pH <3.5). Betacyanin extracts with a pH range = 3.5 < pH < 7 give a decrease in wavelength [25].



Figure 3. Spectrogram of dragon fruit peel extract. Legenda: ◊: Concentrated extract + Ascorbic acid; ★: concentrated extract + citric acid (without discoloration); and △: concentrated extract + citric acid (with discoloration).

The density, the yield of the concentrated extract, and the prediction of the total betacyanin content [14] in the extract obtained are shown in Table 6. It can be seen that yield is directly proportional to the concentration of acid stabilizer added to the extract [25]. The predicted total betacyanin content was based on the method of Priatni (2015); the 96% ethanol + citric acid sample had a higher total betacyanin content, 4279.78  $\mu$ g/100 g, than when ascorbic acid was used as a stabilizer.

Citric acid has a higher acidity (pKa = 3.15) than ascorbic acid (pKa = 4.1). Strong acids give higher yields of betacyanin extracts than lower acids (ascorbic acid) [15]. The addition of organic acids is used to obtain more stable extracts.

TABLE 6. ORTAINED EXTRACT OF DRAGON EPUIT PEEL ( $Hylocepeus$ sp)			
Condition (Solvent-Stabilizer	Density (g/mL)	Extract Yield (%)	Total Betasianin (μg/100 g peel)***
Ethanol 96% v/v + Citric Acid	0.41	9.29	4279.78
Ethanol 96% v/v + Citric Acid [*]	0.41	6.90	N.D**
Ethanol 96% v/v + Ascorbic Acid	0.179	21.42	1229.04
* The resulting extract undergoes a color change			

**N. D = not determined (tidak dihitung).

** calculated using the equation used by Priatni (2015).

## C. Simplified Production of Colour Powder (household scale)

Suji leaves, and dragon fruit peel was sun-dried to produce the dry color powder. The dragon fruit peel was cut into  $\pm 0.5$  cm pieces and dried in the sun (3 x 24 hours); then, the dried dragon fruit peel was ground with a blender and homogenized using a 40-mesh sieve. The dried powder of dragon fruit peel was prepared as a red dye (Figure 4A), while the powder of suji leaves (Figure 4B) was prepared with a shorter drying time of 1x24 hours. In addition, the moisture content, water solubility, and yield of both dried color powders were calculated (Table 7).



**Figure 4.** The dried color powder was obtained from the peel of dragon fruit (a) and the leaves of suji (b) by a simple drying process.



**Figure 5.** Spectrogram of betacyanin from treated dry powder ( $\blacklozenge$ ) and betacyanin standard ( $\blacklozenge$ ).

Measurements of absorbance values at different wavelengths were also carried out to obtain the maximum wavelength for the betacyanin extract from the simple processed dry powder (Figure 5) and the previous measurement of the betacyanin concentrated extract (Figure 3). Betacyanin from the processed dry powder and standard betacyanin have the same  $\lambda_{max} = 525$  nm, so both dragon fruit peel powders have the same betacyanin content as the standard betacyanin compound.

TABLE 7.
ANALYTICAL RESULTS OF DRIED COLOR POWDER BY SIMPLE DRYING
METHOD

Dry colored powder	Water content (b/b)	Solubility (%)	Yield (%)	Color concentration (mg/L)
Dragon fruit skin	6,20	98,85	6,60	$\mathbf{N.N}^{*}$
Suji leaves	30,87	97,28	23,00	15,29**

J. Fund. Appl. Chem. Eng., **2023**, *4* (1), 1-8 eISSN 2964-710X

*betacyanin total; **chlorophyll total.

## IV. CONCLUSION

The optimum maceration time for dragon fruit peel is 24 hours, while the optimum maceration time for suji leaves is known to be 3 x 24 hours. The maceration of suji leaves and dragon fruit peel was carried out at room temperature. The containers were airtight and protected from light. The green color (chlorophyll) of suji leaves and the red color (betacyanin) of dragon fruit peel are sensitive to temperature, air, and light.

Both betacyanin extracts with organic acid stabilizer (citric acid or ascorbic acid) have the same  $\lambda_{max}$  of 325 nm. The betacyanin extract without organic acid has  $\lambda_{max} = 525$  nm. Adding ascorbic acid (75 mL) up to pH 4.5 is more suitable for increasing the betacyanin yield. 96% ethanol + ascorbic acid gave a higher yield value of 21.42% compared to citric acid.

96% ethanol is a better solvent for chlorophyll than 85% acetone. The yield of chlorophyll from suji leaves was 22.1% (total chlorophyll 2.36 mg/L) with 96% ethanol solvent. While Acetone 85% gave a 5% lower yield (1.90 mg/L).

### V. ACKNOWLEDGMENT

The authors would like to thank the Institut Teknologi Sepuluh Nopember (ITS), through Direktorat Riset dan Pengabdian Masyarakat (DRPM), for funding this research under Contract No. 1577/PKS/ITS/2021. We would also like to thank Kintan Rizky Salsabila for assisting with the research.

#### VI. REFERENCES

- [1] N. Aryanti, A. Nafiunisa dan F. M. Willis, "Ekstraksi dan Karakterisasi Klorofil dari Daun Suji (Pleomele Angustifolia) sebagai Pewarna Pangan Alami," Jurnal Aplikasi Teknologi Pangan Jurusan Teknik Kimia, Fakultas Teknik, Universitas Diponegoro, vol. 5, no. 4, 2016.
- [2] E. L. Attoe, dan J. H. von Elbe, "Photochemical degradation of Betanine and Selected Anthocyanins," Journal of Food Science, vol. 46, pp. 1934-1937, 1981.
- [3] H. Azeredo, "Betalains: properties, sources, applications, and stability—a review," Int J Food Sci Technol, vol. 44, pp. 2365–2376, 2009.
- [4] E. Bestari, R. Hastuti, K. Khabibi, "Ekstraksi dan Uji Kestabilan Zat Warna Betasianin dari Kulit Buah Naga (Hylocereus polyrhizus) serta Aplikasinya sebagai Pewarna Alami Pangan," Jurnal Kimia Sains dan Aplikasi, vol. 13, no. 2, 2010.
- [5] L. G. Guerrero, "Pigment changes during the processing of green table olive specialties treated with alkali and without fermentation," Food Research International, Sevilla, 2014.
- [6] D. Indrasti, N. Andarwulan, E. H. Purnomo, N. Wulandari, "Klorofil Daun Suji: Potensi dan Tantangan Pengembangan Pewarna Hijau Alami,"

Jurnal Ilmu Pertanian Indonesia, vol. 24, no. 2, pp. 109-116, 2019.

- [7] A. D. Khuluq, S. B. Widjanarko, E. S. Murtini, "Ekstraksi Dan Stabilitas Betasianin Daun Darah (Alternanthera Alternanthera Dentata) (Kajian Perbandingan Pelarut Air: Etanol Dan Suhu Ekstraksi)," Jurnal Teknologi Pertanian, vol. 8, no. 3, 2007.
- [8] N. Koca, "Effect of pH on Chlorophyll degradation and color loss in blanched green peas," Food Engineering Departement Ankara University, Ankara, pp. 609-615, 2007.
- [9] E. Lee, H. Ahn, E. Choe, "Effects of Light and Lipids on Chlorophyll Degradation," Department of Food and Nutrition Inha University, Incheon, 2014.
- [10] X. Liu, Y. Gao, H. Xu, Q. Wang, B. Yang, "Impact of high-pressure carbon dioxide combined with thermal treatment on the degradation of red beet (Beta vulgaris L.) pigments," J Agric Food Chem vol. 56, pp. 6480– 6487, 2008.
- [11] T. Marnoto, "Ekstraksi Tannin Sebagai Bahan Pewarna Alamu dari Tanaman Putrimalu (Mimosa Pudica) menggunakan Pelarut Organik," Reaktor, vol. 14, no. 1, pp. 39-45, 2012.
- [12] N. Naderi, H. M. Ghazali, A. S. M. Hussin, M. Amid and A. M. M. Yazid, "Characterization and quantification of dragon fruit (Hylocereus polyrhizus) betacyanin pigments extracted by two procedures," Pertanika J. Trop. Agric. Sci., vol. 35, no. 1, pp. 33-40, 2012.
- [13] S. Prasetyo, H. Sunjaya, Y. Yohanes. "Pengaruh rasio massa daun suji/pelarut, temperature dan jenis pelarut pada ekstraksi klorofil daun suji secara batch dengan pengontakan disperse," Lembaga Penelitian dan Pengabdian kepada Masyarakat Universitas Katolik Prahayangan, 2012.
- [14] S. Priatni, A. Pradita, "Stability Study of Betacyanin Extract From Red Dragon Fruit (Hylocereus polyrhizus) Peels," Indonesian Institute of Sciences, 2015.
- [15] S. Priyadi, P. Darmaji, U. Santoso, P. Hastuti, "Profil Plumbum (Pb) dan Cadmium (Cd) sebagai Kontaminan Dampak Penggunaan Agrokimia serta Remediasi Biji Kedelai Menggunakan Swelling Agent pada Khelasi dengan Asam Sitrat," Program Studi Agroteknologi, Fakultas Pertanian, Universitas Tunas Pembangunan, Surakarta, 2013.
- [16] H. T. Quach, R. L. Steeper, W. G. Griffin, "An Improved Method for the Extraction and Thin-Layer Chromatography of Chlorophyll A and B from Spinach," Journal of Chemical Education, Science Departement, Bunker Hill Company. Boston, vol. 81, no. 3, 2004.
- [17] P. Rengku, A. Ridhay, P. Prismawiryanti, "Ekstraksi Dan Uji Stabilitas Betasianin Dalam Ekstrak Buah Kaktus (Opuntia Elatior Mill.)," Jurusan Kimia Fakultas MIPA, Universitas Tadulako, 2017.

- [18] R. Reynoso, F. A. Garcia, D. Morales, E. G. de Mejia, "Stability of betalain pigments from Cactacea fruit," J Agric Food Chem, vol. 45, pp. 2884–2889, 1997.
- [19] M. Saadah, N. Nurdiana, D. Wahyudiati, "Uji Kadar Zat Warna (B-Karoten) Pada Cabe Merah (Capsicum Annum. Linn) Sebagai Pewarna Alami," Jurusan Tadris IPA Biologi FITK IAIN Mataram, 2016.
- [20] E. Saati, R. R. D. Theovilla, S. B. Widjanarko, A. Aulanni'am, "Optimalisasi Fungsi Pigmen Bunga Mawar Sortiran sebagai Zat Pewarna Alami dan Bioaktif Pada Produk Industri," Jurnal Teknik Industri, vol. 12, no. 2, pp. 96-104, 2010.
- [21] S. Schwartz, et al., "Fennema's Food Chemistry 4th Edition," CRC Press. Boca Raton. United States of America, 2008.
- [22] S. Sudarmadji, B. Haryono, Suhardi, "Prosedur Analisa untuk Bahan Makanan dan Pertanian," Malang: Penerbit Liberty. 1989.
- [23] Y. M. Wong, L. F. Siow, "Effects of heat, pH, antioxidant, agitation, and light on betacyanin stability using red-fleshed dragon fruit (Hylocereus polyrhizus) juice and concentrate as model," Journal of Food Science and Technology, School of Science, Monash University Malaysia, 2015.
- [24] K. K. Woo, F. H. Ngou, Ngo L. S., Soong W.K., and P. Tang. "Stability of Betalain Pigment from Red Dragon Fruit (Hylocereus polyrhizus)," American Journal of Food Technology, vol. 6, pp. 140-148, 2011.
- [25] L. Yusmita, A. Kasim, dan H. Nurdin, "Pengaruh suhu ekstraksi dan konsentrasi asam sitrat terhadap pigmen betasianin daun kremah merah (Alternanthera dentata) dan aplikasinya pada pangan," Pro Food Jurnal Ilmu dan Teknologi Pangan, vol. 3, no. 1, pp. 178-185, 2017.
- [26] Z. Jinheng, H. Chao, L. Zhiheng, "Absorption spectrum estimating rice chlorophyll concentration: Preliminary investigations," Institute of Ecoenvironment and Agriculture Information and College of Environment and Safety Engineering University of Science and Technology Guizhou University. Guizhou, 2009.