



Extraction Characteristic and Microencapsulation of Anthocyanin as Natural Food Colouring From Roselle Calyces by Ultrasound-Assisted Extraction

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Abstract

Anthocyanins are widely used as a food additive, and further study in production process development is required in order to obtain an efficient and superior process. This article presents the anthocyanin extraction by ultrasound-assisted extraction and the characterization of solid form anthocyanin extract. In addition, a simple kinetic analysis for the extraction process is investigated. Extraction was conducted by ultrasound-assisted extraction with a solute-solvent ratio of 1:4 and 1:8 at a temperature of 30°C, 40°C and 60°C. Anthocyanin content was analyzed by UV-VIS spectrophotometer. Drying process was performed by a freeze dryer with the addition of maltodextrin and followed by characterization of powder comprising moisture content, solubility and colour intensity. The result shows that the extraction temperature has an effect on anthocyanins extracted. Temperatures rise increased the diffusion coefficient and triggered the driving force of solids into the solvent. This result had a correlation with the second-order kinetic model where the rate of extraction increases along with temperature rise. Characterization of anthocyanin extracts in solid form showed that the addition of maltodextrin provided better results than the product without maltodextrin. The anthocyanin powder added with maltodextrin fulfils the Indonesian standards for food colouring powders, having a low moisture content (5.6%) and high solubility (91.4%). Moreover, colour intensity analysis of anthocyanin powder showed that the powder with maltodextrin has a tendency of a lighter colour with low value of L*, a* and b*.

INTRODUCTION

Food processing is intended to comply with the market demand for food products such as food colour. As a consequence, food colouring substance plays a major role in food production in accordance with the consumer's demand. The food colouring is classified into four groups comprising natural food colouring, identical natural colouring, synthetic food colouring, and caramels (Downham & Collins 2000). The use of natural food colouring has increased by 5-10%, whereas the growth of synthesis food colouring was only 3 to 5%. Natural food colouring has been implemented to replace

synthetic food colouring due to public concern on the safety issue; the healthier and safer food colouring (Durge et al., 2013).

Anthocyanin is a type of natural dye that can be applied as food colouring. Anthocyanins and its derivatives have been listed as natural food colouring according to Indonesian Agency for Drug and Food Inspectors. Moreover, based on Codex Alimentarius Commission list in the European Union, anthocyanins are included as one of the natural food colourings having a code of E163. Anthocyanins also show health benefits, such as modulating cardiovascular disorders, preventing tumour growth, and preventing DNA destruction

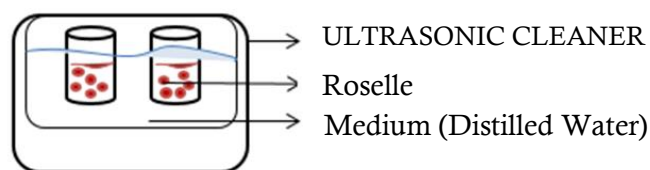


Figure 1. Ultrasonic extraction equipment set

(Lin et al., 2016). Anthocyanins are water-soluble flavonoids found in plants, fruits, and vegetables and provide in red, purple, and blue colour (Mane et al., 2015). Anthocyanins is obtained by extraction from blackberry, cherry (Oancea et al., 2013), Jabuticaba skin (*Myrciaria cauliflora*) (Rodrigues et al., 2015), wine (*Vitis vinifera*) (Ghafoor et al., 2009), *Aronia melanocarpa* (blackberry) (D'Alessandro et al., 2014) and red Roselle calyces.

Some modern methods such as Microwave-Assisted Extraction (MAE), pulsed-electric-field, and Ultrasonic-Assisted Extraction (UAE) have been implemented for obtaining anthocyanins (Mane et al., 2015). The UAE is one of the superior extraction methods since it enhanced solvent penetration into the material and as a result, this increase the mass transfer. The use of ultrasonic reduced the solvents requirement, increase extraction rate, and speed up the extraction process as well as save costs (Wang et al., 2016; Xu et al., 2015). The UAE has been applied for the extraction of anthocyanins from sugar beet molasses (Chen et al., 2015), *Aronia melanocarpa* (D'Alessandro et al., 2014) and *Delonix regia* flowers (Adjé et al., 2010).

Production of anthocyanin extract is usually found as a liquid extract, which in this form the product is easily degradation. Several studies conducted research on the characteristics of conventional extraction of anthocyanin from Roselle calyces (Zaidel et al., 2014; Selim et al., 2008; Kouakou et al., 2015). In addition, kinetics analysis of ultrasonic extraction have been investigated, for example, ultrasonic polysaccharide extraction of *C. Sinesis* (Cheung & Wu, 2013), polyphenols from *Picea abies* (Lazar et al., 2016), oleanolic acid and ursolates from *Hedyotis diffusa* (Wei and Which, 2014). This research is focused on the production of solid anthocyanin extract from Roselle calyces. Specifically, this research studied the effect of the solvent and solute ratios as well as the effect of temperature on the results of liquid anthocyanin extract. Further, the liquid extract was

encapsulated, and solid anthocyanin extract was characterised. In addition, specific reaction kinetic study of UAE for anthocyanins extraction is investigated. Based on the result of the kinetic model, the total anthocyanin in the extract at a specific time can be predicted. The reaction kinetics of UAE can be predicted based on the 2nd order reaction kinetic model (Wei & Yang, 2014; Jokić et al., 2010; Lazar et al., 2016; Tao et al., 2014).

RESEARCH METHOD

Types of Equipment and Materials

In this study, dried red Roselle from Roselle garden in Blitar area, East Java is utilised. Distilled water was used as a solvent, and supporting materials were HCl, Na-Acetate, and Maltodextrin. An Ultrasonic Cleaner (Krisbow, Indonesia) is applied as a ultrasonic-assisted extractor, and Freeze Dryer (Power L 1500) is the drying equipment. UV-VIS Spectrophotometer (UV Mini 1240 Shimadzu) and Chromameter (Color Reader CR-400 / 410Minolta Chroma) were used for total anthocyanin and colour intensity analysis, respectively.

Research Methods

Raw Material Preparation

The preparation process consisted of size reduction and screening in order to enlarge the extraction contact between the material and the solvent. Dried Roselle size reduction was performed by a blender (Vienta Vlb-460) for 30 seconds. Then, the product was sieved to obtain dry Roselle having a size less than 5mm.

Anthocyanin Extraction

The extraction process was carried out by sonication using ultrasonic wave. Prior to the sonication process, the dried Roselle and solvent were mixed at the solute-solvent ratio of 1: 4 and 1: 8. The extraction was conducted at temperatures of 30°C, 40°C and 60°C with a frequency of 40 kHz

$$[(A_{510} - A_{700})_{pH\ 1,0} - (A_{510} - A_{700})_{pH\ 4,5}] \cdot MW \cdot \frac{1000}{\epsilon \cdot l} \quad (1)$$

for 60 min (Melecchi et al., 2006). Roselle extract was then filtered using filter paper and vacuum pump.

Analysis of Anthocyanin Levels

The anthocyanin content was analysed based on the total anthocyanin content defined as cyanidin-3-glucoside. The analysis was performed by a UV-VIS spectrophotometry (UV Mini 1240 Shimadzu) with different pH method (Giusti & Wrolstad, 2001) at pH 1.0 and 4.5. KCl buffer solution having pH of 1.0 and Na-Acetate buffer solution with pH of 4.5 were used to dissolve the Roselle extract. Each solution was examined on absorbance at visible maximum ($\lambda_{vis-max}$) and 700 nm wavelength. The visible-maximum wavelength for cyanidin-3-glucoside was 510 nm (Jurd & Asen, 1966). The total anthocyanin content was determined according to equation (1).

Where A is the absorbance at each wavelength and pH, MW is the molecular weight of cyanidin-3-glucoside (449.2 g/mol), ϵ is the cyanidine-3-glucoside absorbency molar coefficient (26.900 L/mol/cm), and l is the length of the cuvette (cm).

Study of Kinetics Model

The kinetic model of anthocyanin extraction from Roselle calyces is determined by the equation of the 2nd order reaction rate based on the Pelleg model. This model had been previously used to evaluate the kinetics model of oleanolic and ursolic acid (Wei & Yang, 2014), polyphenols (Lazar et al., 2016) and phenolic (Tao et al., 2014). The 2nd order kinetics model is illustrated by equation (2).

$$\frac{dC_t}{dt} = k(C_s - C_t)^2 \quad (2)$$

With C_t is the concentration of extracted anthocyanin (mg/L) at t specific time (minutes), C_s is the anthocyanin concentration in the liquid extract in saturation (mg/L), and k is the coefficient of the second order extraction rate (L/g.min). The kinetic parameter was determined by integrating equation (2) at the boundary conditions $C_t = 0$ to C_t , and t = 0 to t, hence the equation (3) is obtained.

The linearization of equation (3) is used to calculate the required kinetic parameters, according to equation (4).

$$C_t = \frac{C_s^2 \cdot k \cdot t}{1 + C_s \cdot k \cdot t} \quad (3)$$

$$\frac{t}{C_t} = \frac{1}{k \cdot C_s^2} + \frac{t}{C_s} = \frac{1}{h} + \frac{t}{C_s} \quad (4)$$

Where h is the initial extraction rate (g/L/min) at the time of t (min), and C_t is 0. The value of the extraction rate coefficient is determined by plotting the t/C_t data versus t from the experiment. According to equation (4), the k and C_s values are calculated based on the intercept and slope, respectively.

Freeze Drying

Liquid roselle extract was then powdered based on matrix microencapsulation with the freeze-drying method. Maltodextrin as an encapsulating agent in the amount of 10% of the liquid weight is added for maintaining the solid form of the drying product. The extracts mixed with the encapsulating agent were then frozen at -35°C for 24 hours (King et al., 2001). Furthermore, the extract was dried in the freeze dryer at temperature of -70 ° C and vacuum pressure for \pm 48 hours (Pérez-Gregorio et al., 2011). The result of the drying was milled by mortar to form powder extract (solid extract).

Characterization of Solid Products

The moisture content of solid extract as well as its solubility, and colour intensity was analysed. The moisture content and solubility analysis were performed by standard oven method from AOAC (Association of Official Analytical Chemists). The colour intensity was analysed based on the values of * L, * a, and * b with the Chromameter (Color Reader CR-400 /410 Minolta Chroma), using Hunter's Lab Colorimetric System colour system. Before being used the chromameter was calibrated using standard white colour calibration plate. Twenty-five grams of dried anthocyanin powder was placed in a clear container and closed, and then L * (Lightness), a * (Redness) and b * (Yellowness) were measured. The L * value

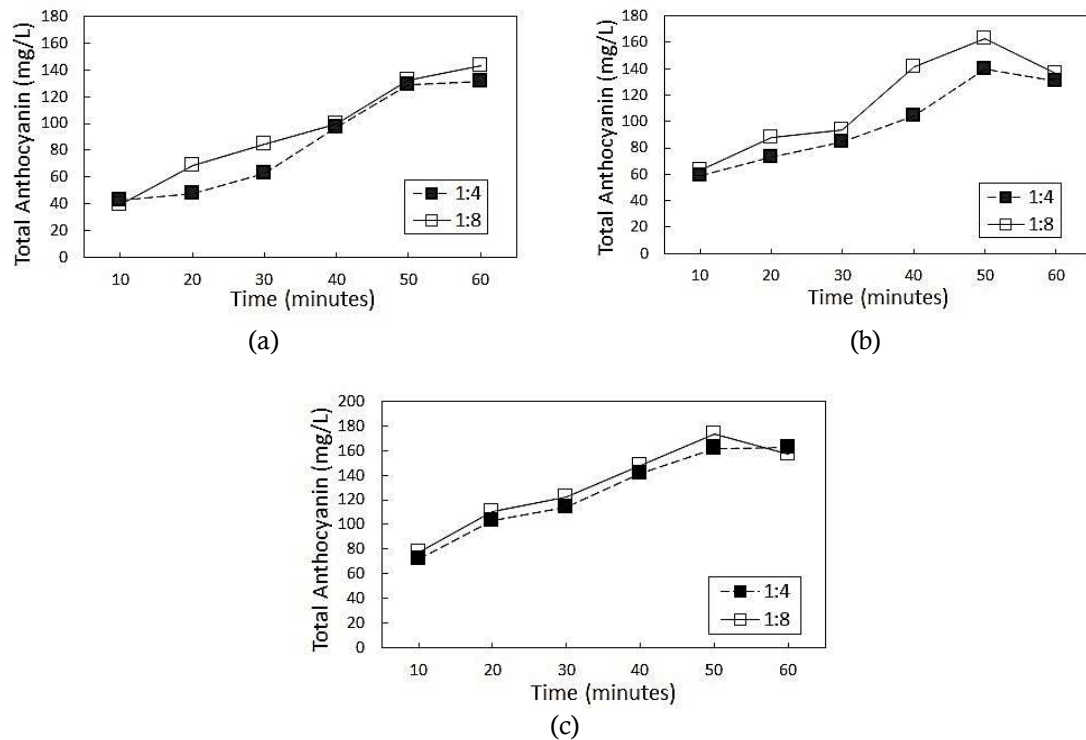


Figure 2. Anthocyanin level at various time, temperature, and solvent ratio (a) Temperature of 30°C (b) Temperature of 40°C (c) Temperature of 60°C.

indicates how bright the object is with a maximum value of 100 = white and 0 = black. The a^* value shows a red-green color tendency where $+a^*$ = red, $-a^*$ = green). The value b^* represents yellow to blue ($+b^*$ = yellow, $-b^*$ = blue) (Chung et al., 2016).

RESULTS AND DISCUSSION

Antosianin Extraction Using Ultrasonic Method

Figure 2 shows the effect of temperature and solvent ratio on the total anthocyanin content. Solute to solvent ratio of 1:8 produces an extract with higher anthocyanin levels than the 1: 4 ratio, for the temperature of 30°C, and 40°C. In contrast, at temperature of 60°C, 60 minutes extraction showed opposite result. The level of anthocyanin extract at ratio of 1:4 is higher than the extract with the solute to solvent ratio of 1:8. The greater the solute-solvent ratio increases the concentration gradient or driving force during mass transfer in solids and have an effect on an increase in the diffusion of the solid component to the solvent. This cause the extraction rate depends on the particle concentration gradient. In addition, extraction was influenced by how fast the component was dissolved and equilibrium in the liquid was achieved (Cacace and Mazza, 2003). The extraction of anthocyanin and phenolic components of black currants also shows a similar phenomenon in which

the amount of extracted anthocyanin increased as the solute-solvent ratio increases (Cacacea & Mazza, 2003). It is also reported for the ultrasonic-assisted extraction of anthocyanins from blueberries, cherry (Wang et al., 2016), and *Jatropha integerrima* (Xu et al., 2015).

Extractions performed on variations in temperature and solvent ratios show that the anthocyanin content increases with increasing the time of extraction. This suggests that the extraction of anthocyanin components in the sample by the solvent increases at the time. Specific period is required for the solute to dissolve into the solvent. The longer the contact time between the solvent and the tissue, the better the equilibrium of the particle transferred to the solvent (Dutta et al., 2007). The use of ultrasound waves for anthocyanin extraction from Roselle calyces can also help to improve process efficiency. Vibration on extraction by ultrasonic method provides intensive process agitation. This agitation increases the osmosis between the material and the solvent and as a result, enhance the efficiency of the extraction process. An ultrasonic method with a frequency of 36 kHz can destroy leaf cells, thus accelerating the process of mass transfer of bioactive compounds from cells to solvents (Dean, 1998). The frequency resulted in cavitation in the water medium, and the ultrasonic cavitation generated a breaking force that breaking

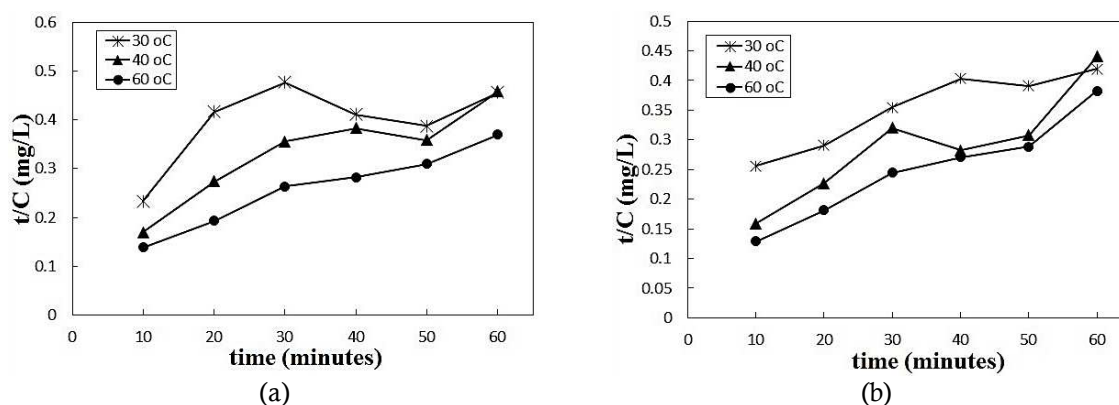


Figure 3. Anthocyanin extraction kinetics of 2nd order at various temperature (a) solute-solvent ratio of 1: 4 (b) solute-solvent ratio of 1: 8.

the cell wall mechanically and increased the transfer of material (Liu et al., 2010). The longer the extraction process, the more material transfer also occurs so that the anthocyanin levels increase. However, anthocyanin levels decreased after extraction time of 50 minutes. In this condition, it is possible to have an equilibrium, and hence the extract solubility decreased (Dutta, 2007).

Moreover, Figure 2 shows the effect of extraction temperature on anthocyanin levels, after ultrasonic assisted extraction for 60 min. Higher temperature extraction provides more extracted anthocyanin. This is due to the correlation between temperature, solubility, diffusivity coefficient, viscosity and extraction time. The solute characteristics depend on the nature of the solute (entropy fusion and melting point) and mixed properties such as the activity coefficient. It was confirmed that low melting point and high temperature increased the solubility (Cacace & Mazza, 2003). On the other hand, temperature rise decreased the viscosity and causing the increase in diffusion coefficient (Olawale, 2013). As the diffusion coefficient increased, a driving force of the solid mass to the solvent was present, and the increase of the gradient coefficient results in the higher concentration.

However, excessively high temperatures in ultrasonic-assisted extraction may cause anthocyanin degradation and lead to a decrease in anthocyanin levels in the extract (Wang et al., 2016). As well as anthocyanin levels obtained from temperature extraction 60 °C, in which in the 60th-minute extraction with a 1: 8 ratio decreased to have a smaller number than the ratio of 1: 4. Since the beginning of extraction, the amount of anthocyanin extracted at a 1: 8 ratio was more significant than the ratio of 1: 4, so at a ratio of 1: 8 more

anthocyanins may be degraded. This allows for a more significant decrease in anthocyanin levels. Research conducted by Chumsri et al. (2008) showed optimum anthocyanin levels in dry red Roselle using water solvents at a ratio of 1: 8, temperature 50°C with 60 minutes duration had bright red colour and high anthocyanin levels. Extremely high temperatures and too much time length of process produce an anthocyanin extract that has a lighter red colour and low anthocyanin levels. Anthocyanin can be degraded due to changes in acidity and thermal conditions, where anthocyanins will undergo co-pigmentation and structural changes (Gradinaru et al., 2003; Idham et al., 2010; Patras et al., 2010).

Kinetic of Anthocyanin Extraction

Kinetic of anthocyanin extraction from Roselle calyces is determined by plotting of t/C values versus time as shown in Figure 3.

According to Figure 3, the value of kinetic parameters, i.e. the rate of extraction constant (k), the concentration in the saturation state (C_s), and the initial rate of extraction (h) can be calculated. The determination of kinetic parameters is obtained by the mathematical equation of regression as defined in Table 1.

The solute-solvent ratio of 1: 4 or 1: 8 indicates that the extraction rate increase as the temperature of extraction raises. This confirms that the extraction of anthocyanin from Roselle calyces was related to the operating temperature. Higher reaction rate indicates that the reaction is faster at higher extraction temperatures. Similar results were also found in ultrasonic-assisted extraction for polyphenols from *Picea abies*, ursolic acid extraction from *Ocimum sanctum*, and anthocyanin extraction from blackberries. It as also reported that the rate of

Table 1. Kinetic parameters for 2nd order kinetic models

Ratio of solute-solvent	T (K)	B=1/C _s	A=1/h	C _s =1/b (mg/l)	h=1/A	k=h/C _s ²
1:4	303	0.0026	0.3032	384.6154	3.29	2.23 x10 ⁻⁵
	313	0.0049	0.1605	204.0816	6.23	14.96 x10 ⁻⁵
	333	0.0044	0.1068	227.2727	9.36	18.13 x10 ⁻⁵
1:8	303	0.0033	0.2358	303.0303	4.24	4.62 x10 ⁻⁵
	313	0.0046	0.1284	217.3913	7.78	16.5 x10 ⁻⁵
	333	0.0046	0.0882	217.3913	1.34	23.9 x10 ⁻⁵

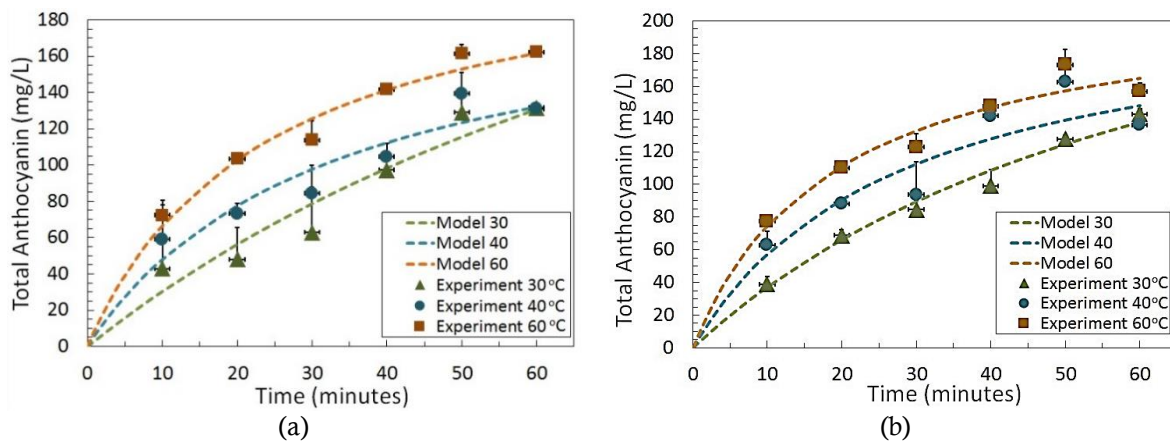
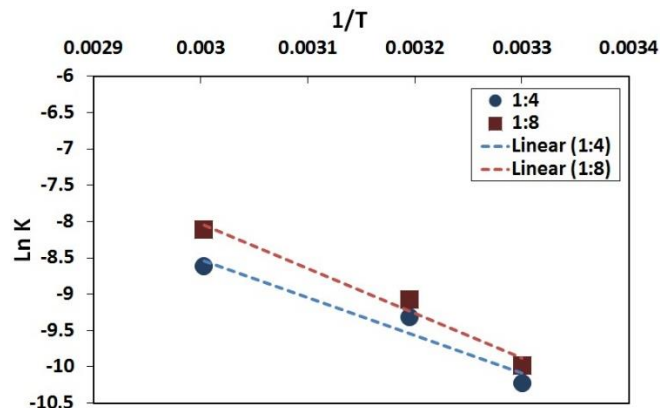


Figure 4. Total ratio of anthocyanin from Roselle calyces extraction of experimental data and calculation result, (a) solute-solvent ratio of 1:4 (b) solute-solvent ratio of 1:8.

Figure 5. The correlation of temperature and 2nd order reaction rate constant

reaction had a tendency to increase with the increasing of extraction temperature (Lazar et al., 2016; Vetal et al., 2012; Oancea et al., 2013).

Figure 4 shows that the results of calculations with the second order kinetics model give a similar trend to the experimental data at temperatures of 30 ° C, 40 ° C and 50 ° C. Calculation of error analysis between experimental results and model calculations shows that the average deviation that occurs only ranges between 4 to 14%.

The value of the activation energy can be calculated by the linearization of the Arrhenius equation (equation 5).

$$\ln k = \ln k_0 + \left(\frac{-E_a}{R} \right) \frac{1}{T} \quad (5)$$

Figure 5 shows the correlation of $\ln k$ (kinetics rate) to $1/T$ (temperature) at the solute-solvent ratio of 1:4 and 1:8.

The value of activation energy on anthocyanin ultrasonic-assisted extraction from roselle calyces at solute-solvent ratio of 1: 4 and 1:8 are 42.45 kJ.mol⁻¹ and 52.48 kJ.mol⁻¹, respectively. The activation energy is the minimal energy required for the anthocyanin molecule to be leached out of the roselle. The value of positive activation energy indicates that the reaction is running in an endothermic state. The value of activation energy

Table 2. Moisture content and Solubility of Red Roselle Solid Extract Products

	With Maltodextrin	Without Maltodextrin
Moisture content	7.2%	5.6%
Solubility	95.2%	91.4%

Table 3. Color Intensity of Solid Product

Type of extract	Color intensity		
	L*	a*	b*
Solid extract with maltodextrin	37.186	53.671	1.418
Solid extract without maltodextrin	13.654	71.539	6.092

with positive notation is also shown in the calculation of kinetics model of ultrasonic-assisted polyphenol extraction on *Picea abies* (Lazar et al., 2016), anthocyanin extraction of grape juice (Anna et al., 2012), and anthocyanin extraction from berries Cacace & Mazza, 2003).

Characterization of Solid Extract

Moisture content and Solubility of Solid Extracts

The table shows that red Roselle solid extract using maltodextrin and without maltodextrin has a moisture content of 7.2%, and 5.6%, respectively. Based on the Indonesian standard SNI 01-0222-1995, the permitted maximum moisture content on powder food colouring is in the range of 4.5% - 5%. In this research, higher moisture content than the standard is resulted due to the freeze-drying process. One of the freeze-drying product characteristics is its hygroscopic character which is able to absorb water molecules from environment either through absorption or adsorption (Mosquera et al., 2012). In addition, the principle of removal of water molecules in freeze-drying process is sublimation in which the water in solid form was converted to gas at vacuum pressure (Oberoi & Sogi, 2015).

Encapsulant agent (maltodextrin) is a material used to coat core substances (anthocyanin). The use of maltodextrin aims to stabilize and reduce the hygroscopic characteristic of the food colouring (Mosquera et al., 2010). The addition of maltodextrin also has a function to avoid clumping of the extract and reduce surface stickiness during the drying process, resulting in an excellent free-flowing product (Goula and Adamopoulos, 2008). However, according to Canuto et al. (2014), the addition of maltodextrin causes the moisture content of the dried extract increases. This is due to maltodextrin blocks water in the extract to come out at the time of drying occurs.

The solubility rate of red Roselle dye using maltodextrin and without maltodextrin were 95.2% and 91.4%, respectively. The value is relatively high and presents an excellent point as a food additive. The solubility of freeze-drying material is strongly affected by its filler (maltodextrin). Maltodextrin may affect the physical properties of the encapsulated material because the maltodextrin is highly soluble in water. This makes it very popular as an additive in the food industry (Mosquera et al., 2010). The addition of maltodextrin increases the solubility of the product to water and relieves digestion in the body.

Color Intensity of Solid Extract

Maltodextrins have been widely used as additives to the drying process of natural materials such as fruit juice, sumac extract, and borojo extract (Gabas et al., 2007; Caliskan & Dirim, 2016; Mosquera et al., 2010; Mosquera et al. 2012). Table 2 shows the brightness level with the L* notation on the solid extract by using maltodextrin was higher than the solid extract without maltodextrin with the value of 37,186 and 13,654, respectively. This was because anthocyanin has been encapsulated in maltodextrin which had white color with intensity value of $L^* = 98,18 \pm 0,15$; $a^* = -0.185 \pm 0.05$ and $b^* = 2.91 \pm 0.15$ (Caliskan and Dirim, 2016). As a result, the use of maltodextrin in the extract at the time of drying could reduce the colour intensity. The encapsulation process protected the core material by coating the exterior, thereby preventing exposure to adverse environmental conditions (Gabas et al., 2007; Saikia et al., 2015).

The intensity indicating the anthocyanin colour is the a^* notation with the green-red chromatic colour. The value of a^* in the extract with maltodextrin is lower than that of the extract without maltodextrin at the value of 53,671 and 71,539, respectively. It was also reported by Caliskan & Dirim (2016), the use of maltodextrin

encapsulated the anthocyanin in white-colour maltodextrin. Wang et al. (2011) confirmed that the higher levels of maltodextrin as encapsulating agent lead to a decrease in chroma (a^*) value.

CONCLUSION

Anthocyanin extraction from Roselle calyces using ultrasound-assisted extractor results in optimum yield at a solute-solvent ratio of 1: 8 and a temperature of 60°C. Along with the extraction time, the total anthocyanin shows an increase of up to 50 minutes then the total anthocyanin decreases afterwards. The anthocyanin extraction presents conformity with the second order kinetics model. Characterization of anthocyanin extract in solid form confirms that the addition of maltodextrin provides a better result than drying product without maltodextrin. The powder extract has moisture content of 5.6%, 91.4% solubility and bright colour due to the addition of maltodextrin.

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