Optimization of Agitation and Kinetic Studies on Proanthocyanidin Compound Extraction from Red Sorghum Grains in Agitated Vessel

D Y Susanti 1,2, W B Sediawan 1, M Fahrurrozi 1, M Hidayat 1

Abstract. Proanthocyanidin extraction for antioxidant material from intact red sorghum grains in agitated vessel had been investigated. The optimization and the appropriate kinetic evaluation were useful to conduct the engineering design of the developed process. Concentrations of the proanthocyanidin compounds in the aqueous extract were affected by the agitation speed and extraction time. The objectives of this research were to determine the optimum values of agitation speed and time for getting maximum extraction performance and to develop the appropriate kinetic model for illustrating the extraction phenomena. This research also evaluated the characteristics of the obtained extract. The optimum conditions of extraction were determined using Response Surface Methodology. The concentration of proanthocyanidin compound in the extract was predicted to be maximum at 405.05 rotations per minute (rpm) of agitation speed and 133.03 minutes of extraction time. The values of $R^2$ of the regression equations were 99.45%. In the kinetic modeling, the mechanistic model turns to be much more accurate than the second order kinetic. The extract from the optimum process had 0.837 mg/ml of proanthocyanidin concentration and was proven to be effective in scavenging 64.02% of free radicals in the 100 $\mu$M DPPH in 30 minutes.

Keyword: optimization; agitation; kinetic; proanthocyanidin; extraction; red sorghum

1. INTRODUCTION

Recently, natural medicines containing phenolic compounds including phenolic acid, flavonoids and polyphenols are widely explored to be used for maintaining health of 80% citizen in developing countries. It is related to their ability to prevent oxidative damage of materials in plants, vegetables, fruits and to their role as natural antioxidants for humans. [1,2].

The need of the beneficial compounds for healthy supplements and food antioxidants is increasing rapidly. Therefore, designing effective extraction systems of natural antioxidant has been an interesting object to be developed [3,1] in order to substitute the unsafe synthetic antioxidant.

Red sorghum grain is known as a potential source of natural antioxidants because it is easily cultivated and contains the high phenolic compounds in its pericarp which has powerful antioxidant activity. The phenolic compounds have different structures and physical properties, including proanthocyanidin (condensed tannin) and polymeric phenolic[4,5]. Red sorghum pericarp has 11.9-15.5 mg of proanthocyanidin/g of grain[6]. Proanthocyanidin is proven as an antioxidant agent [7], which can be applied in food and medicine.

In order to design antioxidant production systems, numbers of extraction techniques are useful to be developed and optimized to get the suitable and efficient method to extract antioxidant compound from their natural source [8,1]. The method should be simple and safe for the proposed extracts and the environment or known as green technology. Commonly, proanthocyanidin extraction from red sorghum
was conducted from pericarp debris and whole grain by organic solvent such as water, ethanol, etc. In this paper, proanthocyanidin was extracted from intact red sorghum pericarp in agitated vessel using water as solvent. The whole grain extraction was expected to avoid gelatinization of sorghum flour mixed with pericarp debris that could hamper the extraction process from the solid matrix. Furthermore, it could also eliminate dehulling process prior to the extraction. So, it could be the simplest method to be applied in wide range of industries before the red sorghum grain was further processed into flour. In this method, water had been a good choice of solvent as they had the affinity to the solute better than the solid matrix and other components [9] and more effective than nonpolar solvent [10]. It has the same polarity with the compound, non-toxic and cheap for reducing extraction cost [11]. Using water as the basic of solvent, proanthocyanidin were removed from the inside of pericarp to solvent, and the grain were processed to produce sorghum flour.

Several studies had focused on the development of accelerated water extraction (AWE) as a solid-liquid extraction technique that was developed to substitute organic solvents in antioxidant extraction for medicinal compounds or food additives [3]. As polar solvent, the dielectric constant of water is high because of the hydrogen bond in its structure [12]. Water extraction become an interesting method which is widely applicable in bioactive extraction because it is natural, non-toxic, low cost green solvent and has been recognized as a safe solvent by food and nutraceutical industries [13,2]. This method is an alternative of green technology in collecting functional compounds located inside of materials without a harmful impact on the environment and biomaterial by using water as non-toxic solvents. To create the faster process than conventional techniques, the ability of water in extracting compounds the embedded materials, this method can be supported by agitation, temperature treatment, extraction cycles optimization [3,10].

Within this paper, the increasing of water capacity in extraction phenolic and proanthocyanidin compounds in the vessel had been conducted by agitation and determination the optimum of extraction time in order to create the best mixing and sufficient mass transfer. Agitation can accelerate the spread of the functional compound to all parts of the solvent and trigger the mass transfer of the compound from the inside to the thin layer of solvent covering to the grain. Therefore, the extraction rate will be faster than maceration method, is estimated to be affected by the rotational speed and extraction time. So, the correlation between extraction rate on the agitation speed is useful as an interesting object to describe extraction regime [14,15].

The rotational speed of agitation and extraction time affect the recovery of extraction process, so optimization of those variables is useful to design the extraction process systems. This optimization is related to the energy consumption and the intactness of the grain. Furthermore, to complete the feasibility study of the developed extraction from intact red sorghum pericarp, the optimization of some parameters and kinetic studies of process are useful to explore. The kinetic analysis is useful in selecting, designing, describing, predicting the operational process and scale up its feasibility [15]. The quantitative analysis is very important to improve the accuracy in determining the most efficient and effective extraction conditions in designing extraction system. The kinetics were described by different application of mathematical techniques[1]. As described in several papers which evaluate the performance of process based on the optimization and kinetics studies [9,16], the most suitable kinetic and optimization result are useful to be applied in industrial scale process design. The preliminary of this method feasibility has been focused on determining the best rotational speed and extraction time and the mass transfer process kinetics. The appropriate kinetic of the mass transfer process from the inside pericarp of red sorghum to the solvent is expected to describe the mechanism and process performance. The aim of this paper are to determine the optimum values of agitation speed and extraction time to reached the maximum value proanthocyanidin concentration as antioxidant compound from intact sorghum; to determine the appropriate kinetic model to quantitatively describe the extraction process and to evaluate the characteristic of obtained extract.
2. MATERIALS AND METHOD

2.1. Material
The red sorghum grains (*Sorghum bicolor* L. Moench) from local market Wonogiri, Indonesia were cleaned and dried before extraction. Aquadest, Folin-Ciocalteu reagent, gallic acid, sodium bicarbonate, 1-butanol, and HCl were obtained from Merck Co. (Darmstadt, Germany). Fe NH$_4$(SO$_4$)$_2$ x 12 H$_2$O was obtained from J.T.Baker Avantor.Inc (Center Valley, Pennsylvania). Procyanidin B1 was obtained from Chengdu Biopurify Phytochemicals Ltd. (Sichuan, China). All other reagents were at analytical grade.

2.2. Determination sorghum grain characteristic
The characteristic of the red sorghum grain were measured, consist of the thickness of pericarp, the radius and the initial tannin concentration in pericarp.

2.3. Extraction procedure
Extraction system was conducted in miniplant of agitated vessel in Figure 1 at 60 °C of the solvent temperature in three variation rotational speed, involved 300, 400, and 500 rpm. Thirty five grams of red sorghum grain was extracted in 350 ml of aquades in each rpm during 150 minutes. The datas of aqueous extract was taken within 30 minutes of interval by the appropriate procedure, consist of proanthocyanidin concentration (PC), colour intensity, electric conductivity and radical scavenging activity of extract.

![Figure 1. The proanthocyanidin extraction system from intact red sorghum](image)

2.4. Determination of proanthocyanidins concentration (PC) in extract
Proantocyanidin was determined by acid butanol assay [17,18].

2.5. Optimization using Response Surface Methodology (RSM)
The optimum conditions of extraction were determined using minitab of RSM in two level factorial design based on the experimental proantocyanidin concentration (PC) in the extract solution, derived from the extraction using distilled water as solvent at 60°C of temperature; 10 of sorghum-solvent ratio.

2.6. Determination appropriate kinetic model
For generalization of the process, quantitative analysis using second order kinetics and the proposed mechanistic model were performed. The most appropriate model is determined by their fitness to the experimental data.

2.6.1. Second order kinetic model
The change of PC in extract is evaluated using second order extraction in equation 1 [3,19,11].

$$\frac{dC_A}{dt} = k_A \cdot (C_{Ae} - C_A)^2$$  \hspace{1cm} (1)

where $k_A$ ($cm^3_{solvent} \cdot g^{-1} \cdot minute^{-1}$) = the constant value of extraction rate based on second order kinetic; $C_{Ae}$ ($g/cm^3$) = the equilibrium concentration of proanthocyanidin in extract; and $C_A$ ($g/cm^3$)= the concentration of proanthocyanidin in solvent at t time of extraction.
By integrating second order kinetic (equation 1) under the boundary conditions \( C_{At} = 0 \) at \( t = 0 \) and \( C_{At} \) at \( t = t \), equation 2 was obtained. The value of \( k_A \) were determined by the slope and the intercept of the linear equation 3.

\[
C_{At} = \frac{C_{Ae}^2 \cdot k_A \cdot t}{1 + C_{Ae} \cdot k_A \cdot t} \\
\frac{t}{C_{At}} = \frac{1}{k_A \cdot C_{Ae}^2} + \frac{t}{C_{Ae}}
\] (2) (3)

2.6.2. Mechanistic model

The proposed mechanistic model was developed based on the assumption that the intraparticle diffusion was to be very fast, so the process was controlled by the mass transfer rate of the solute from the surface of the solid to the bulk of the liquid solvent. So the proanthocyanidin concentration inside the grain was homogenous. The material balance of solute in the liquid then produces equation 4:

\[
\frac{dVC_A}{dt} = \frac{dm_x \cdot X}{dt} = N_b \cdot S \cdot k_c \cdot (C_A^* - C_A)
\] (4)

in which the equilibrium proanthocyanidin concentration in the liquid \( (C_A^*) \) is approximated by:

\[
C_A^* = H \cdot X.
\] (5)

The overall solute material balance produces equation 6.

\[
X = X_0 + \frac{V}{m_s} \cdot C_{Ae}. \quad (6)
\]

The combination of equation 4, 5 and 6 resultes in equation 7.

\[
\frac{dC_A}{dt} = \frac{N_b \cdot S \cdot k_c}{V} \left[ H \left( X_0 \cdot \frac{V}{m_s} \cdot C_{Ae} \right) \right] - \frac{N_b \cdot S \cdot k_c}{V} \left( \frac{H \cdot V}{m_s} + 1 \right) \cdot C_A
\] (7)

In equation 4,5,6 and 7, \( C_A \) (g/cm\(^3\))= the concentration of proanthocyanidin in solvent; \( t \) (minute) = time; \( N_b \) (grains)= the number of red sorghum grains immersed in solvent; \( k_c \) (cm.minute\(^{-1}\)) = the coefficient of mass transfer; \( V \) (ml) = solvent volume; \( m_s \) (g) = mass of sample sorghum; \( C_Ae \) (g/cm\(^3\)) = initial concentration of proanthocyanidin in solvent; \( H \) (gram pericarp/cm\(^3\) solvent) = distribution coefficient. \( S \) (cm\(^2\))= the large of pericarp surface; \( X \) (g/g solid sample)= the proanthocyanidin content in solid matrix; \( X_0 \) (g/g solid sample)= the initial proanthocyanidin content in solid matrix

The mass of the solid matrix was assumed to be constant during the extraction. The geometric form of the grains was assumed to be spherical and their radius are uniform. The proposed model can be solved numerically to produce extract concentration \( (C_A) \) at various time. By curve fitting to the experimental data, the value of \( k_c \) and \( H \) can be evaluated. The value of \( k_c \) and \( H \) can also be evaluated by the slope and intercept of the linear form of the analytical solution of equation 7.

2.7. Determination physicochemical characteristic aqueous extract

The color intensity was measured using chromameter CR400/410, while the electric conductivity was measured using the electric conductivity meter. Furthermore, the antioxidant activity was analyzed by the radical scavenging activity (RSA) method. The absorbance profile of the 0.1 ml sample in 3.9 ml 100 \( \mu \)M the radical 2,2-diphenyl-1-picrylhydrazyl (DPPH) in methanol solution was detected using spectrotomter UV-VIS (Genesys 10S) on wave length 515 nm [20,21].

3. RESULT AND DISCUSSION

3.1. Concentration of the proanthocyanidin compound in the extract during extraction

Preliminary measurement showed that the red sorghum grain had 0.01 cm of pericarp thickness; 0.19 cm of radius, and 0.1263 g of proanthocyanidin B1 equivalent/cm\(^3\) of initial proanthocyanidin concentration in pericarp. The embeded compound was released from the inside to outside of grain pericarp and transferred to the solvent. The profile of the proanthocyanidin concentration (PC) in the solvent at various time and at various agitation speed was shown in Figure 2.
Figure 2. The proanthocyanidin concentration (PC) in solvent during extraction

As presented in Figure 2, the proanthocyanidin concentration value increased faster at the beginning of extraction and tended to be slower at the longer time of extraction. The fast process had been triggered by the high solute concentration difference of the surface of pericarp and of the bulk of liquid. However, as the time progresses, the difference became smaller, hence the rate of extraction would be slower.

From Figure 2, it can be observed that the maximum value of PC during extraction were 0.698 mg/ml for 300 rpm; 0.837 mg/ml for 400 rpm; and 0.712 mg/ml for 500 rpm, reached at 120 minutes of time extraction. The decreases of PC at the long time of extraction may be due to the random variation when the extraction reached at the equilibrium condition or the sign of the degradation of proanthocyanidin compound. Figure 2 showed also the highest extract concentration was obtained at 400 rpm of agitation speed. At the low speed, the dispersion of particle was not well dispersed in the liquid, so the mass transfer becomes slow. But the very high agitation speed made the particle motion completely follow the fluid velocity, hence the velocity of the particle relative to the fluid velocity became very small, resulting the slow interface mass transfer.

3.2. Optimization using RSM

The more accurate optimum value of agitation speed and extraction time are further investigated by Response Surface Method (RSM), which is simple and more comprehensive [4,10]. The limit range for optimizing agitation speed and time to get the maximum PC using RSM were shown in Table 1. The optimization data in Table 2 was conducted using two level factorial design by minitab RSM.

The effect of each parameter and their interaction also have been analyzed by RSM [22,23,24] using pareto chart of standardized effects shown in Figure 3. Their individual effect, their synergism or antagonism effect between each factor can be detected by the large proportion. Figure 3 shows that the agitation value has more prominent effect to PC value in extract better than extraction time. Furthermore, the graph of counter and surface plot was shown in Figure 4.

The optimization by RSM in Figure 5 resulted that the optimum condition has been 405.05 rpm and 133.03 minutes for proanthocyanadin extraction, as effective condition to accelerate extraction and get the highest point of concentration. The increasing value of parameters became ineffective to be applied.

**Table 1.** Experimental range and level of independent variable

| Independent variables | unit | Range and level |
|-----------------------|------|-----------------|
| Agitation speed (X₁)  | rpm  | 300 400 500     |
| Extraction time (X₂)  | minute | 90 120 150     |
Table 2. Experimental data for optimization using RSM

| Experiment | Independent variables | Dependent variables (R = Response) |
|------------|-----------------------|------------------------------------|
|            | No                    | X₁ = Agitation speed (rpm)          | X₂ = time (minute) | PC (mg/ml) |
| 1          | 1                     | 300                                 | 90                | 0.495     |
| 2          | 2                     | 300                                 | 120               | 0.698     |
| 3          | 3                     | 300                                 | 150               | 0.689     |
| 4          | 4                     | 400                                 | 90                | 0.644     |
| 5          | 5                     | 400                                 | 120               | 0.837     |
| 6          | 6                     | 400                                 | 150               | 0.808     |
| 7          | 7                     | 500                                 | 90                | 0.565     |
| 8          | 8                     | 500                                 | 120               | 0.712     |
| 9          | 9                     | 500                                 | 150               | 0.709     |

The predicted PC can be determined using equation 8 with 99.45 % of the R² value.

\[
PC = -3.221 + 0.010127X_1 + 0.03035X_2 - 0.000012X_1X_1 - 0.000004X_1X_2 (8)
\]

Figure 3. Pareto chart of standardized effect of rpm and time to PC

Figure 4. Surface plot of PC value

Figure 5. The optimum condition (rpm & time) and the maximum PC in extract

3.3. The kinetic discussion

In general, each model has been developed by dissimilar degree of complexities [16]. In this paper, the second order kinetic model and the mechanistic model approach are applied and evaluated in the proanthocyanidin extraction from intact sorghum pericarp.
3.3.1. Second order kinetic

The second order kinetic model is an empirical approach which had been reported as the appropriate model for solid-liquid extraction of water soluble compound [19,3]. The model described the process in two simultaneous processes, which are the removal of solutes from the outer surface of particle in constant velocity and the diffusion of solute material from inner to outer part of particle [11].

In general, the proanthocyanidin concentration in solvent (Cₚ) increases faster at the beginning and then slower, leading to an equilibrium concentration (Cₑ). Table 3 showed the parameter values of second order kinetic model in extraction process from the fitting of PC data on equation 4.

Table 3. The constant values of second order kinetic model

| Rpm  | kₑ (cm³ solvent. gPA⁻¹. minutes⁻¹) | Cₑ (gram cm⁻³) | Cₑ (mg ml⁻¹) | SSE  |
|------|-----------------------------------|----------------|--------------|------|
| 300  | 23.561                            | 8.63.10⁻⁴      | 0.863        | 3.192.10⁻⁴  |
| 400  | 33.622                            | 9.47.10⁻⁴      | 0.947        | 3.88.10⁻⁴   |
| 500  | 36.716                            | 8.29.10⁻⁴      | 0.829        | 2.75.10⁻⁴   |

Table 2 illustrated the influence of agitation in improving extraction rate (kₑ) by increasing the dispersion of the solute materials in the interface. The performance of second order kinetic on predicting the PC value during the process was shown in Figure 6. From Figure 6, it can be seen that the second order model was only visually moderately accurate.

![Graph A](image)

(A) 

![Graph B](image)

(B) 

3.4. Mechanistic model

The mechanistic model quantitatively describes the extraction process comprehensively. Since the experimental data show that the agitation speed influences the overall mass transfer rate, the main assumption taken is that the overall mass transfer was controlled by the solute mass transfer from the surface of the solid to the bulk of solvent. Later, this assumption will be tested by the experimental data.

The solution of mechanistic model by equation 5 was conducted on the real condition in extraction of 1252 red sorghum grain which has 0.01 cm of pericarp thickness; 0.19 cm of grain radius and 0.1263 g/cm³ of initial proanthocyanidin content in pericarp in 350 cm³ of water. The value of kₑ and H was evaluated by curve fitting to the experimental data. The results of the parameter value were shown in Table 4 and the comparison of the calculated and experimental data were shown in Figure 7.

Table 4. The value of parameters in mechanistic model

| Rpm  | H (gram pericarp.cm⁻³ solvent) | kₑ (cm⁻³. minutes⁻¹) | SSE      |
|------|-------------------------------|----------------------|----------|
| 300  | 8.89.10⁻³                     | 4.511.10⁻¹           | 5.992.10⁻⁹ |
| 400  | 8.32.10⁻³                     | 7.562.10⁻¹           | 6.140.10⁻⁹ |
| 500  | 7.274.10⁻³                    | 7.312.10⁻²           | 2.627.10⁻⁹ |
Visual observation concludes that the mechanistic model are superior than the second order model. More detailed comparison between the two models will be discussed later.

The $k_c$ value at 400 rpm was observed to be higher than the ones at 300 rpm and 500 rpm. The $k_c$ value in 400 rpm is higher than 300 rpm because the increasing agitation speed decreases the stagnant layer in interfacial area by the mixing of bubbles [25], hence improves the diffusion [15]. So, the rate of diffusion is faster than the one of chemical reaction [15]. Agitation also decreased the resistance of interfacial mass transfer by decreasing the thickness of stagnant layer and increasing friction between the two phases.

But, the $k_c$ value at 500 rpm was less than the ones at 400 rpm. As discussed in section 3.1, the decreasing of mass transfer coefficient ($k_c$) was caused by the decrease of the relative particle velocity to fluid because the particle movement completely follows the movement of the fluid, hence the slip velocity decreased, so as the interface mass transfer rate.

Based on those discussions, it can be concluded that the highest value of mass transfer coefficient ($k_c$) had been attained at around 400 rpm, inline with the result of RSM. It is the limit that the agitation speed can increase the friction between two phases and decrease the thickness of the stagnant film. After 400 rpm, the particle motion will follow the fluid motion, so the increasing of speed is not effectively increasing the mass transfer rate.

In the point of view of accuracy, the mechanistic model was visually superior to the second order model in predicting the proanthocyanidin concentration in the extract, as shown in Figure 8.

3.5. The colour intensity, electric conductivity and antioxidant activity of the extract

Several research evaluated the correlation between the concentration of bioactive compound such as phenolic, anthocyanin, carotenoid etc. with the color of extract using colorimeter [26]. In this paper, the proanthocyanidin concentration in the extract has also be visually observed by the increasing of b value as yellowish indicator shown at Figure 9A, and so as the electric conductivity at Figure 9B.

The antioxidant activity of the extract from the optimum condition was evaluated. The activity was was expressed as radical scavenging activity (RSA) to radical 2,2-diphenyl-1-picrylhydrazyl (DPPH) as, shown in Figure 10C. Within 30 minutes of incubation, the RSA value of the extract was stable in scavenging 64.02% of 100µM.DPPH in methanol.

**Figure 7A.** The predicted and the experimental PC from mechanistic model

**Figure 7B.** The correlation of the predicted & experimental PC from mechanistic model
Figure 8. The comparison between second order kinetic and mechanistic model in this case.

Figure 9. The color value (A), electric conductivity value (B) and antioxidant activity (C) of extract

4. Conclusion

The optimum conditions for proanthocyanidin extraction from intact red sorghum grain at the temperature of 60°C and sorghum-solvent mass ratio of 10 obtained by RSM was agitation speed of 405.05 rpm and extraction time of 133.03 minutes. The proposed mechanistic model turns to be much more accurate for predicting proanthocyanidin concentration in the extract than the second order kinetic. The antioxidant activity of extract from the optimum process was supported by its RSA value which reached 64.02% in scavenging free radicals in 100 μM DPPH in 30 minutes.

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