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To cite this article: Megawati *et al* 2020 *IOP Conf. Ser.: Earth Environ. Sci.* **572** 012041

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Kinetics of soursop leaves antioxidant extraction using microwave-assisted extraction

Megawati^{1*}, D S Fardhyanti¹, D Widjanarko², Hanifah¹, G M Bungsu¹, and M H F Rizky¹

¹ Chemical Engineering Department, Faculty of Engineering, Build. E1, 2nd Floor, Sekaran, Gunungpati, Universitas Negeri Semarang, Indonesia

² Mechanical Engineering Department, Faculty of Engineering, Universitas Negeri Semarang, Indonesia

*Email: megawati@mail.unnes.ac.id

Abstract. The purpose of this research is to study the kinetics of soursop leaves antioxidant extraction using microwave-assisted extraction. The kinetics was approached by a homogeneous mechanism using the first and second-order rate law. The extraction was conducted with ethanol as the solvent (1:13 v/v) using a modified microwave oven operated at various powers (300, 450, and 600 W) for 2-10 minutes. The soursop leaves extraction with ethanol using microwave-assisted extraction obtained a maximum yield of 33.98% (with total phenol of 24.10 mgGAE/g extract) at 600 W and 8 minutes. The kinetics of soursop leaves antioxidant extraction can be quantitatively described by the homogeneous rate law, where the second-order model is better than the first one. The extraction capacity and constant rate increase as the microwave power does. At 300, 450, and 600 W, respectively, the extraction capacities were about 500, 588, and 667 mg/l and the constant rates were 8.5×10^{-4} , 9×10^{-4} , and 1.18×10^{-3} l/mg/min.

1. Introduction

Soursop trees (*Annona muricata* L.) are the easiest plant to grow in a warm and humid tropical climate, containing antioxidants and acetogenins [1]. In general, their leaves also contain bioactive compounds, tannins, phenols, phytosterols, flavonoids, saponins, and alkaloids [2]. Flavonoids are phenolic compounds that can function as antioxidants since they have a molecular structure that can transfer electrons to free radical molecules [3]. Specifically, a compound is said to be a strong antioxidant when the IC₅₀ value is less than 50 ppm, strong if it is in the range 50-100 ppm, moderate 100-150 ppm, and weak 151-200 ppm [4]. Besides, soursop leaves antioxidant is powerful because the IC₅₀ value is 1.36 mg/ml (ppm) [5]. In addition, soursop leaves antioxidant has been used as a bio-diesel B20 additive to prevent its degradation because of air oxidation [6].

An isolation of plant antioxidants can be done in several ways such as conventional and non-conventional extraction techniques. The conventional techniques include Maceration and Soxhlation, and non-conventional include ultrasound-assisted, microwave-assisted, and subcritical water extraction technique [7, 8]. The conventional extraction techniques have several disadvantages; they need a large amount of solvents and long process, while the yields obtained are few [9]. The microwave-assisted technique is often called microwave-assisted extraction (MAE). MAE is an extraction method which



utilizes microwave as a heating medium. This technology is suitable for extracting thermolabile antioxidants because it has better temperature control than conventional heating methods. In addition, MAE also has several advantages. Accordingly, it needs a short extracting process and many yields can be obtained [10]. This research used MAE to extract soursop leaf antioxidant efficiently and economically. The data obtained were used to study the extraction kinetics.

Extraction kinetics is important to facilitate the design of unit operation. On the other hand, the mass transfer of an extractive solute from the inner particle of the solid to the bulk of the liquid occurs in two stages. First, the diffusion of solute in the inner particle and second the convective mass transfer at the solid-liquid interphase. In the case of tiny particles, the diffusion of solute in the inner particles was assumed very fast, which can be negligible. Hence, the rate-limiting step is only the mass transfer from the particle surface to the bulk of the solution [11].

2. Materials and methods

2.1. Antioxidant extraction of soursop leaf using microwave-assisted extraction

The materials used in this study were: soursop leaves (from Sapta Marga, Semarang); 70% ethanol; gallic acid (Sigma Aldrich G7384-100G); folin ciocalteau reagent (Merck 109001); Na₂CO₃ (Merck 106392), methanol (Merck 106009), NaOH (Merck 106462), and distilled water. Before being used, the soursop leaves were dried in an oven (Mammert) at 90 °C for 2 hours, then blended using a blender (Philips) until they became powder. The powder size is maintained by sieving it using a 500 µm screening (Endecotts).

The soursop leaves extraction was done using MAE method. First, about 20 g of soursop leaves powder was inserted into a glass extractor (2 L) and 260 ml of 70% ethanol was added. The antioxidant phenolic concentrations were analysed in every 2 minutes. The extractions were conducted at various powers of 300, 450, and 600 W. The extraction time was 10 minutes. After the extraction was complete, the mixture was vacuum filtered to separate the filtrate and residue. The solvent in the filtrate was recovered by distillation at 78 °C until the extract volume was 20 ml. The extract was purified by evaporating the solvent by heating it in an oven at a temperature of 100 °C to obtain a pure extract. All of these treatments were done in triplets to ensure the accuracy of the data. The pure antioxidant extract that obtained was stored in a dark coloured glass bottle and placed in a refrigerator (10 °C).

The phenolic concentration analysis was conducted by adding 0.5 ml samples with 0.5 ml folin ciocalteau reagent (v/v) and 4 ml Na₂CO₃ 7.5% (w/v). The solution was left for 60 minutes. Then, the solution was placed into spectrophotometry to measure the absorbance at a wavelength of 765 nm. The antioxidant phenolic concentration was calculated based on the absorbance produced using a calibration curve. The calibration curve was obtained using gallic acid solutions with concentrations of 50, 100, 150, 200, 250, and 300 ppm. The calibration equation for phenolic concentration based on its absorbance is written as equation (1), with A = absorbance and C_t = concentration (ppm).

$$A = 0.0094C_t - 0.0433 \quad (1)$$

2.2. Kinetic of soursop leaf extraction with ethanol as solvent using microwave assisted extraction

The mass transfer of solute in the solid-liquid extraction can be expressed by the first and second-order rate laws, as described in equation (2) and (3), with C_t = antioxidant concentration during the extraction (mg/l), C_s = extraction capacity (mg/l), t = time (min), and k_e = extraction constant rate (l/mg/min) as well as the initial condition of $C_{t(t=0)} = 0$ and $C_{t(t=t)} = C_t$ [12].

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

$$-\frac{dC_t}{dt} = k_e (C_s - C_t)^2 \quad (3)$$

Equation (2) and (3) can be solved by an integral method. The results, equation (4) and (5) were, then linearly regressed to obtain the kinetic parameters, k_e and C_s [13].

$$\ln\left(\frac{C_s}{C_s - C_t}\right) = k_e t \quad (4)$$

$$\frac{t}{C_t} = \frac{1}{k_e C_s^2} + \frac{t}{C_s} \quad (5)$$

3. Results and Discussion

The absorbance measurement results on time and power variations are presented in table 1. The absorbance is substituted into equation (1) to obtain phenolic concentration (C_t). The first-order model was solved using a linear regression from equation (4). A plot graph between $\ln(C_s/(C_s - C_t))$ with t was made. The plot was then linearized and the intercept was set at (0,0) to get the slope and coefficient determinant (R^2) (as presented in figure 1). Trial-and-error attempts were made to obtain the C_s which gives the highest R^2 value. After obtaining the C_s value, then the slope value can be used as the extraction constant rate value (k_e). Figure 1 shows that the R^2 values in each power variation are 0.96; 0.81; and 0.33 for 300, 450, and 600 W, respectively. Thus, the soursop leaves extraction kinetics is less approachable using the first-order model, especially at high power (600 W).

Table 1. The absorbance and phenolic concentration of soursop leaves extraction with 70% ethanol using MAE (operating condition: raw material weight of 20 g, raw material size of 500 μm , 70% ethanol volume as solvent of 260 ml).

Time (minute)	300 W		450 W		600 W	
	Absorbance	C_t (mg/l)	Absorbance	C_t (mg/l)	Absorbance	C_t (mg/l)
2	2.31	250	2.82	305	3.97	427
4	2.81	303	3.32	358	4.37	469
6	3.46	373	3.71	399	5.60	600
8	3.65	393	4.12	443	5.62	603
10	3.89	418	4.73	507	5.49	589

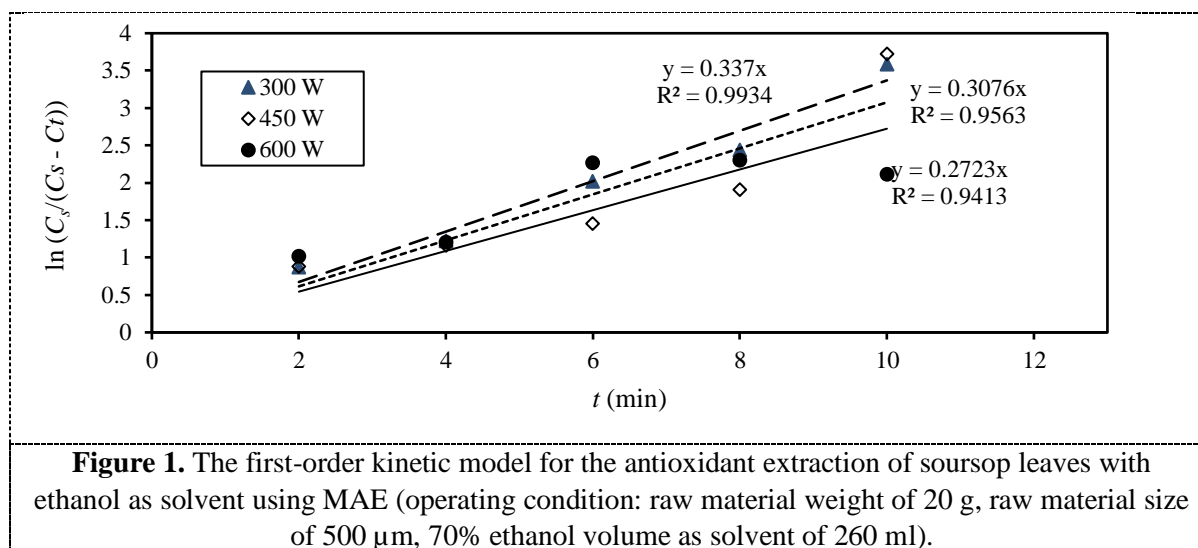


Figure 1. The first-order kinetic model for the antioxidant extraction of soursop leaves with ethanol as solvent using MAE (operating condition: raw material weight of 20 g, raw material size of 500 μm , 70% ethanol volume as solvent of 260 ml).

The second-order model was calculated using equation (5). Similar to the first-order model, the second-order model was also solved using a linear regression, as depicted in figure 2. Figure 2 shows that the extraction kinetics of soursop leaves can be approached by the second-order model well, even better than the first-order one. The accordance between the data and the result of calculations is presented in figure 3. Because the size of the soursop leaf powder is too small, the solute diffusion from inside to solid's surface can be ignored. This mass transfer can be expressed by the homogeneous model,

especially the second-order rate law. The result of this study is in accordance with previous works. They extracted *Jatropha* seeds with hexane and petrochemical ether solvents. This condition also occurs in the flavonoid extraction of *Terminalia bellerica* using MAE [12, 14].

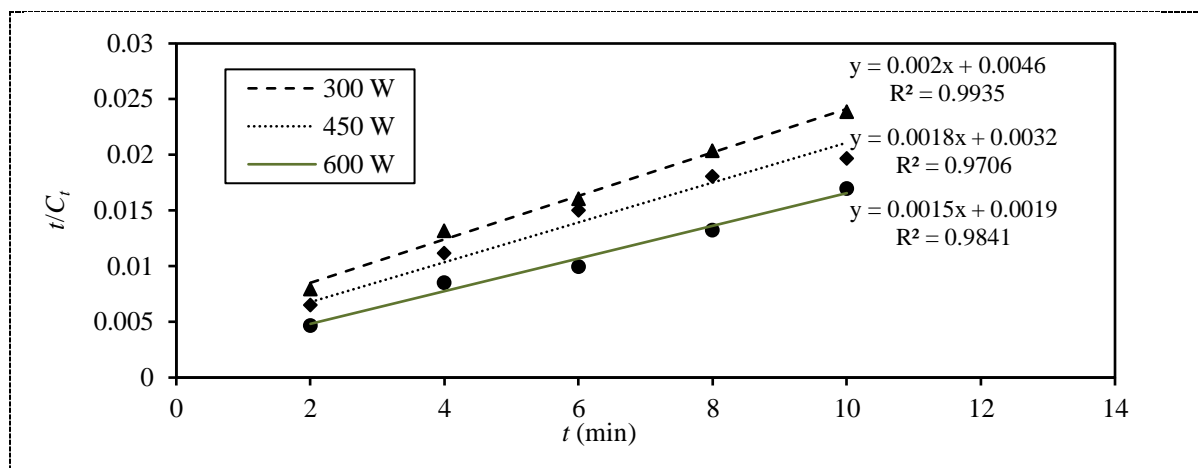


Figure 2. The second-order rate law for the antioxidant extraction of soursop leaves with ethanol as solvent using MAE (operating condition: raw material weight of 20 g, raw material size of 500 μm , 70% ethanol volume as solvent of 260 ml).

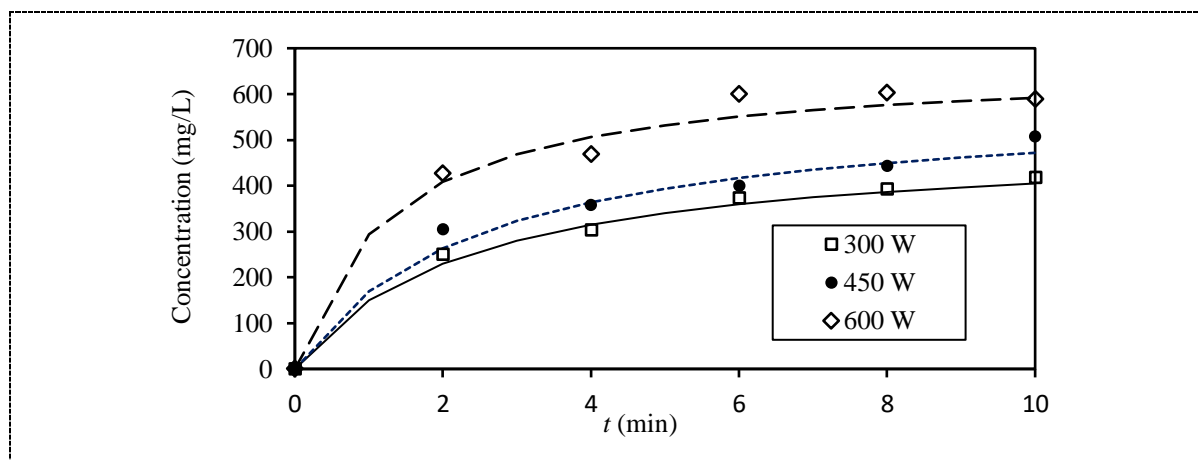


Figure 3. The second-order rate law for the antioxidant extraction of soursop leaves with ethanol as solvent using MAE (operating condition: raw material weight of 20 g, raw material size of 500 μm , 70% ethanol volume as solvent of 260 ml).

Figure 4 shows the detail information regarding the similarity between the phenolic concentration data and the calculation results (close to the graph's diagonal line). It appears that the second-order model can quantitatively describe the soursop leaves antioxidant extraction with ethanol using MAE properly.

The values of the extraction rate (k_e) and extraction capacity (C_s) constants obtained from the study are presented in table 2. Based on the results, the extraction speed and extraction capacity constants increase in proportion to the increase in power. This occurs because the extraction temperature will increase when higher powers are used. This is in line with research conducted on flavonoid extraction on *Terminalia bellerica* using MAE. The increase in extraction temperature is proportional to the increase in kinetics constant, as well as the extraction capacity [14]. It is important to remember that at

high powers or temperatures, phenolic compounds can be damaged. In addition, the extraction speed constant also increases with the addition of the ratio of materials to solvents and particle size. The constant extraction speed can be doubled from 6.53×10^{-4} to 1.28×10^{-3} l/mg/min at a material-solvent ratio of 1:10 g/ml. Also, the smaller the particle size of the material from 1.2 to 0.175 mm, the extraction speed constant increases fivefold [15]. The extraction speeds will be faster in smaller-sized materials because the surface area covered by the solvent is greater, so the amount of materials dissolved will increase [16].

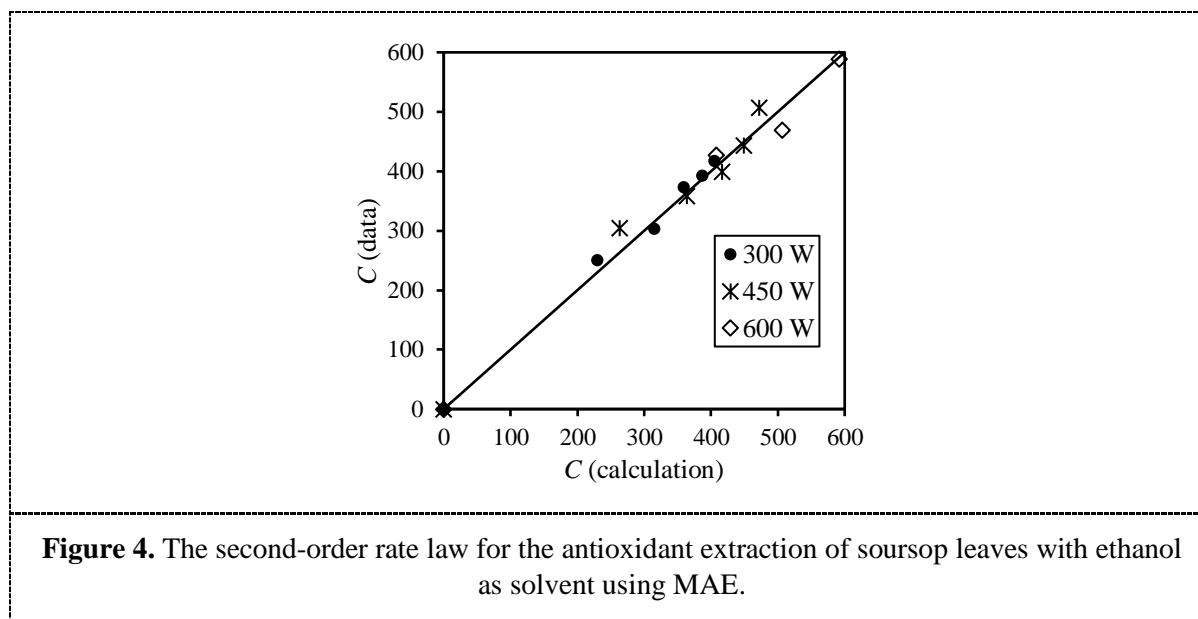


Figure 4. The second-order rate law for the antioxidant extraction of soursop leaves with ethanol as solvent using MAE.

Table 2. The extraction constant rate (k_e) and extraction capacity (C_s) of second-order rate law of soursop leaf extraction using microwave-assisted extraction (operating condition: raw material weight of 20 g, raw material size of 500 μm , 70% ethanol volume as solvent of 260 ml).

Power (W)	Extraction capacity (C_s) (mg/l)	Extraction constant rate (k_e) (l/mg/min)
300	500	0.00085
450	588	0.00090
600	667	0.00118

4. Conclusion

Kinetics of soursop leaves antioxidant extraction with ethanol using microwave-assisted extraction can be quantitatively described by the homogeneous rate law. The second-order model approach is better than the first one. The extraction capacity and constant rate increase as the microwave power does. At 300, 450, and 600 W, respectively, the extraction capacities were about 500, 588, and 667 mg/l and the constant rates were 8.5×10^{-4} , 9×10^{-4} , and 1.18×10^{-3} l/mg/min.

Acknowledgments

We would like to show our gratitude to Ministry of Research, Technology, and Higher Education of the Republic of Indonesia for financial support through research grant 2019 (192/SP2H/LT/DRPM/2019).

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