International Journal of Applied Pharmaceutics

ISSN- 0975-7058 Vol 11, Issue 2, 2019

Original Article

SUPERCRITICAL CARBON DIOXIDE EXTRACTION OF MANGO GINGER (*CURCUMA MANGGA* ROXB.): PROCESS OPTIMIZATION USING TAGUCHI METHOD

RINALDI SALEA, ERWAN Y. DARUSSALAM, STEVANUS HIENDRAWAN, BAMBANG VERIANSYAH, RAYMOND R. TJANDRAWINATA*

Dexa Laboratories of Biomolecular Sciences (DLBS), Industri Selatan V Block PP No. 7, Jababeka Industrial Estate II, Cikarang 17550, West Java, Indonesia
Email: raymond@dexa-medica.com

Received: 31 Nov 2018, Revised and Accepted: 30 Jan 2019

ABSTRACT

Objective: Extraction of *Curcuma mangga* (*C. mangga*) using supercritical carbon dioxide (SC-CO_2) was investigated to provide information about the optimum extraction condition.

Methods: A Taguchi method with L_9 orthogonal array design was used to determine the optimum extraction conditions. Effects of extraction pressure, temperature, CO_2 flow rate and dynamic extraction time on *C. mangga* oil yield were investigated at levels ranging between 150-350 bar, 40-60 °C, 10-20 g/min and 120-240 min, respectively.

Results: The highest *C. mangga* oil yield (5.223%) from SC-CO_2 extraction was obtained at a pressure of 350 bar, temperature of 60 °C, CO_2 flow rate of 20 g/min and dynamic extraction time of 240 min. The experimental *C. mangga* oil yield at optimum condition was in a good agreement with the values predicted by computational process using Taguchi method. Based on S/N ratio calculation, the most influencing parameters in maximizing *C. mangga* oil yield is extraction temperature, followed by extraction pressure, dynamic extraction time and CO_2 flow rate.

Conclusion: In this study**,** Taguchi method was successfully applied to optimize SC-CO_2 extraction of *C. mangga.* Taguchi method was able to simplify the experimental procedure of SC-CO_2 extraction.

Keywords: Taguchi, Supercritical carbon dioxide extraction, *Curcuma mangga,* optimization

DOI: http://dx.doi.org/10.22159/ijap.2019v11i2.31079

INTRODUCTION

Curcuma genus (*Zingiberaceae* family) consists of more than 80 species and some are widely used as spices and medicinal preparations [1]. It is native to the warm and humid environments of South and Southeast Asia. *Curcuma mangga*, which also known as the mango ginger, is one of the significant species in the *Zingiberaceae* family cultivated for food and medicinal purposes. The rhizomes of *C. mangga* are similar to ginger but possess a raw mango taste mainly due to the presence of car-3-ene and cis-ocimene [2, 3]. Medicinally, *C. mangga* rhizomes have been known for its uses to treat stomachache, fever and chest pain. It is also used in postpartum care, specifically to aid the healing of wounds [1, 4]. Literature has shown diverse therapeutic effects of *C. mangga* extract, such as antibacterial, antifungal, insecticidal, aphrodisiac, antipyretic, anti-inflammatory, anti-mycobacterial, anti-hyper-cholesterolemic and antioxidant properties [1, 3].

Pharmacologically-active compounds in plants are usually present in low concentrations. In recent years, our laboratories have developed and investigated various extraction techniques and biological assays on natural products [5-8]. The most widely-used method is extraction based on organic solvent [9]. Organic solvent extraction methods have been known to possess several drawbacks such as the production of hazardous wastes and few adjustable parameters to control the selectivity of extraction process. Therefore, the development of more reliable, simpler, less chemical-intensive and less expensive technique is necessary for commercial-scale production. In recent years, supercritical carbon dioxide (SC-CO_2) extraction has widely been utilized for the extraction of oil and bioactive compounds from plant materials [8,10]. SC-CO_2 extraction offers several advantages due to its non-toxic nature and unique physicochemical properties which include low viscosity, fast diffusion and tunable physical properties with temperature and pressure. The low critical temperature and pressure of carbon dioxide (T_c=31 °C and P_c=7.38 MPa) offer the possibility to operate extraction process at low temperature (below 80 °C) and moderate pressure (100–450 bar), which may be an ideal condition for the extraction of thermolabile compound [11, 12]. Despite the high potential of SC-CO_2 for the extraction of oil and bioactive compounds, various parameters of SC-CO_2 extraction (i.e. temperature, pressure, CO_2 flow rate and time of extraction) have become the major obstacle in finding the optimum extraction condition for each plant material [13, 14].

Taguchi method is one of the simple and systematic approaches for designing an experiment with less number of experimental runs. It could be employed to optimize the performance characteristics of process parameters, which is proved to be a powerful tool that differs from the traditional full factorial method. This approach could economically satisfy the needs of problem-solving and design optimization. Thus, it is possible to reduce the time and cost for the experimental investigations [15-17]. Taguchi method has been employed in some applications related to SC-CO_2 extractions. Ansari *et al.* (2012) employed Taguchi method to optimize supercritical extraction of oil from spearmint (*Menthaspicata* L.) leaves [18]. Previously, we compared Taguchi and full factorial method to optimize various parameters on the supercritical extraction of black cumin (*Nigella sativa*) seeds. It was shown that Taguchi method was able to simplify the experimental procedure and maintain experimental cost at a minimum level without affecting the quality of the result. On the other hand, the oil yield difference between full factorial and Taguchi method is insignificant (±0.1%) [19]. In previous study, we also utilized Taguchi method to optimize SC-CO_2 extraction oil from Javanese turmeric (*Curcuma xanthorrhiza* Roxb.) and emprit ginger (*Zingiber officinale var.* Amarum) rhizome [8, 11]. The objective of this present study was to investigate the optimum condition for *C. mangga* oil extraction through SC-CO_2 extraction and to investigate the effect of extraction pressure, temperature, CO_2 flow rate and dynamic extraction time on *C. mangga* oil yield using Taguchi method.

MATERIALS AND METHODS

Materials

Dried rhizomes of *C. mangga* were obtained from CV Cipta Pratama (Magelang, Central Java, Indonesia). The dried rhizomes were comminuted in a milling machine (Quadro Comil, Canada) with 040 G screen and square impeller at 3500 rpm. The moisture content of the sample before extraction was 12.5% wet basis (w. b). Food-grade liquid CO_2 (purity of 99.99%) was supplied in a cylinder tube by PT Inter Gas Mandiri (Cikarang, Indonesia).

Supercritical carbon dioxide (SC-CO_2) extraction

SC-CO_2 extraction was carried out using a customized supercritical fluid extractor with CO_2 cycle system (KIST-Korea) described in previous study [19]. Extractor vessel with 1 L capacity was loaded with 50 g of grounded *C. mangga*. Food grade liquid CO_2 was delivered to the extraction vessel using a high-pressure pump (Thar, USA). Extraction pressure was varied from 150 to 350 bar and temperature from 40 to 60 °C. The pressure in extraction vessel was controlled by a back pressure regulator (Tescom, USA). Heat exchangers (Lab. Companion, USA) were provided in the system to maintain the temperature in extractor and separator vessel. The CO_2 flow rate was varied from 10 to 20 g/min. Static time was 60 min for all experiments and dynamic time was varied from 120 to 240 min. After experiments had been finished, the extract was separated from SC-CO_2 by pressure reduction and collected from the separator.

Taguchi method

Taguchi method utilizes orthogonal array design (OAD) to study a large number of parameters with a small number of experiments. It significantly reduces the number of experimental configurations to be studied without affecting the quality of results and maintains the experimental cost at a minimum level [20, 21]. Taguchi method employs a special set of OAD in each number of experimental conditions to systematically look for favorable operating conditions [22]. A Taguchi method with L_9 OAD was used to investigate the optimum condition in SC-CO_2 extraction since it is the most suitable design for the investigated conditions, i.e. four factors with three levels (values). The factors and levels are listed in table 1, whereas the structure of Taguchi method with L_9 OAD is shown in table 2. S/N ratio calculation is an evaluation of output performance which measures the level of performance and effect of noise parameters on performance. A target value of 'larger is better' was used since the purpose of this study was to obtain the highest *C. mangga* oil yield. In this study, the main effects of the three parameters were studied excluding the interactions between variables.

S/N ratio was calculated using the following Eq. (1):

$$\frac{S}{N} = -10log\left(\frac{1}{n}\sum_{i=1}^{n}\frac{1}{y_i^2}\right) \quad (1)$$

Where y_i represents the experimentally observed value of the i^{th} experiment, and n is the number of trial at the same level. Statistical analysis was performed using MINITAB v.15 (Minitab Inc., USA) statistical software package.

Table 1: Factors and levels used in experimental design

Level	Factors			
	Pressure (bar)	Temperature (°C)	CO_2 flow rate (g/min)	Dynamic time (min)
	A	B	C	D
1	150	40	10	120
2	250	50	15	180
3	350	60	20	240

Table 2: Standard for L_9 orthogonal arrays

Run	Independent variables			
	A	B	C	D
1	1	1	1	1
2	1	2	2	2
3	1	3	3	3
4	2	1	2	3
5	2	2	3	1
6	2	3	1	2
7	3	1	3	2
8	3	2	1	3
9	3	3	2	1

RESULTS AND DISCUSSION

Effect of SC-CO_2 extraction conditions on *C. mangga* oil yield

Effects of four operating conditions of SC-CO_2 extraction, namely pressure, temperature, CO_2 flow rate and dynamic extraction time on the extraction of *C. mangga* oil were investigated using Taguchi method. The experimental responses in terms of oil yield are summarized in table 3. Results showed that the obtained *C. mangga* oil yield was varied from 0.524-4.889%.

A total of 9 experiments were performed in order to obtain the results of *C. mangga* oil yield from SC-CO_2 extraction. Since a higher oil yield was the desirable purpose, the quality characteristic was set as 'larger is better'. The optimization of *C. mangga* oil yield from SC-CO_2 extraction was considered from S/N ratio in each level of parameters. Fig. 1 depicted the S/N ratio of each individual factor. The higher number of S/N ratio represents a higher oil yield.

Table 3: Results of *C. mangga* oil using SC-CO_2 extraction

Run	Pressure (bar)	Temperature (°C)	CO_2 flow rate (g/min)	Dynamic time (min)	Yield (%)
1	150	40	10	120	0.524
2	150	50	15	180	2.766
3	150	60	20	240	4.222
4	250	40	10	120	4.280
5	250	50	15	180	3.274
6	250	60	20	240	4.889
7	350	40	10	120	4.128
8	350	50	15	180	4.419
9	350	60	20	240	4.292

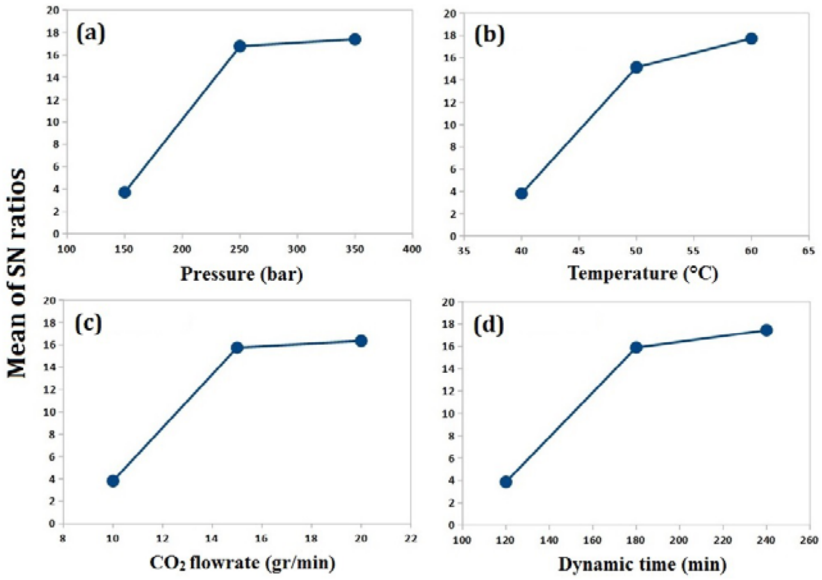

Fig. 1: Effects of pressure (a), temperature (b), CO_2 flow rate (c), and dynamic time (d) on *C. mangga* oil yield

Fig. 1 shows the main effect plot of *C. mangga* oil yield to different extraction factors and levels. Main effect plot was performed based on the average response for each combination of control factor levels. It can be seen in fig. 1a that increased pressure from 150 to 250 bar resulted in a significant increment of *C. mangga* oil yield, while further increasing pressure from 250 to 350 bar resulted in a smaller oil yield increment. Generally, using a higher pressure at isothermal conditions resulted in an increased solvent density and subsequently solvent power and solubility of the compounds. An increase in CO_2 density accelerated the mass transfer between analytes and solvent during the extraction process and thus it improved the extraction yield. As the density increased, the distance between molecules was decreased and the interaction between compounds and CO_2 was increased, which led to a greater solubility of compounds in CO_2 [14].

The selection of operating conditions depends on the specific compound or compound family to be extracted. Molecular weight and polarity of the compound have to be taken into account case by case. The extraction temperature for thermolabile compound has to be fixed between 35 and 60 °C, e.g. in the vicinity of the critical point and as low as possible to avoid degradation [8]. At a constant pressure, an increase in temperature reduced the density of SC-CO_2, hence reducing the solvent power of SC-CO_2, but it increased the vapor pressure of the compounds and thus it became easier to extract [14]. These two competing factors changed differently and it depends on extraction conditions. Above the crossover pressure, the effect of temperature on density became less than that of solute vapor pressure. Therefore, the yield tends to increase with temperature. Below the crossover pressure, the effect of temperature on density became more than that of solute vapor pressure [23]. From fig. 1b, it can be seen that as temperature increased from 40 to 60 °C, the oil yield was also increased. This can be explained that the vapor pressure of the oil was increased and its solubility and extraction was improved at higher temperature [24].

Fig. 1c shows the effect of CO_2 flow rate on oil yield. *C. mangga* oil yield was increased at CO_2 flow rate of 10-15 g/min, while further increasing CO_2 flow rate from 15 to 20 g/min resulted in a smaller oil yield increment. Mass transfer was increased with an increased CO_2 flow rate since there was a higher amount of SC-CO_2 passed through the extractor and it decreased the film thickness around the particles throughout the bed, which lowered external mass transfer resistance. However, increased CO_2 flow rate resulted in a reduced residence time of CO_2. In this case, the extraction process deviated from the equilibrium and SC-CO_2 left the extractor unsaturated [25-27].

For all experiments, the static time was fixed at 60 min. During the static time, the system in extractor is allowed to reach an equilibrium that involved an interaction between the supercritical fluid and plant matrix. For a system in which the extraction rate is determined by the transfer of a solute from particle surface into the fluid, it can be assumed that the equilibrium between supercritical fluid and the matrix is established in a suitable period of static extraction time and will be released during dynamic extraction time. Fig. 1d shows that *C. mangga* oil yield was increased along with increased extraction time by up to 180 min and it did not vary with further increase in extraction time. Similar phenomena were also observed in our previous study on *C. xanthorrhiza* Roxb extraction [8].

Optimum condition

In this study, since the target value was 'larger is better', the optimum condition for each factor was defined as the level that gives the highest point of means of each plot for *C. mangga* oil yield. Based on the results of Taguchi method, the highest *C. mangga* oil yield from SC-CO_2 extraction was obtained at a pressure of 350 bar, temperature of 60 °C, CO_2 flow rate of 20 g/min and dynamic extraction time of 240 min. S/N ratio calculation results are shown in table 4. Greater difference value (Δ) of the average S/N ratio indicated that the control factors have a greater substantial effect on S/N ratio. Based on S/N ratio calculation, the most influencing parameters in this process is extraction temperature, followed by extraction pressure, dynamic extraction time and CO_2 flow rate.

Table 4: S/N value calculated for each parameter and level

Level	Pressure	Temperature	CO2 flow rate	Dynamic time
1	3.712	3.796	3.819	3.865
2	16.761	15.146	15.748	15.902
3	17.389	17.718	16.360	17.450
Δ (max–min)	13.677	13.922	12.541	13.585
Rank	2	1	4	3

Experimental validation

Experimental validation is the final step in the modeling process to investigate the accuracy and robustness of the established model. The final analysis involved a comparison between the predicted values of the established model and experimentally validated values. The predicted value at optimized condition is 5.997%, while the actual value at this condition is 5.223%. Percentage error found in this study was 12.9%. It was found that the average percentage error was below 15%, confirming and concluding that the methodology used in establishing the model in this scientific research was systematic. *C. mangga* oil yield obtained in this study was higher than previous studies using SC-CO_2 extraction. In the previous study, Krishna *et al.* used SC-CO_2 extraction to extract *C. mangga* oil. They varied the extraction pressure from 100 to 350 bar, extraction temperature from 40 to 60 °C and extraction time from 5 to 15 h. Results of previous study showed that the best *C. mangga* extraction yield was 2.83% at 350 bar and 60 °C for 15 h [28].

CONCLUSION

Optimization of SC-CO_2 extraction on the yield of *C. mangga* oil was successfully carried out using Taguchi method. The optimum condition to obtain the highest *C. mangga* oil yield (5.223%) was achieved at a pressure of 350 bar, temperature of 60 °C, CO_2 flow rate of 20 g/min and dynamic extraction time of 240 min. The experimental oil yield at optimum condition was in accordance with the predicted computational results (5.997%). S/N ratio calculation identified extraction temperature as the most influencing parameters in maximizing *C. mangga* oil yield followed by extraction pressure, dynamic extraction time and CO_2 flow rate.

ACKNOWLEDGEMENT

The authors acknowledge Dexa Laboratories of Biomolecular Sciences (DLBS)–PT. Dexa Medica for financial support. The authors would like to thank Isabela Anjani for critical review on this manuscript.

AUTHORS CONTRIBUTIONS

All the author have contributed equally

CONFLICT OF INTERESTS

The authors declared no conflicts of interest with respect to the authorship and/or publication

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