Vietnam Journal of Science and Technology ***56*** *(4A) (2018) 229-237*

OPTIMIZATION OF GINGER OLEORESIN EXTRACTION FROM FRESH GINGER BY USING MICROWAVE-ASSISTED ENERGY

Nguyen Trung Dung*, Ta Hong Duc, Nguyen Dang Binh Thanh

Hanoi University of Science and Technology, No. 1 Dai Co Viet road, Ha Noi

*Email: *dung.nguyentrung@hust.edu.vn*

Received: 24 September 2018; Accepted for publication: 7 October 2018

ABSTRACT

Nowadays, the active compounds from natural products are interested in not only research fields but also the industrial areas. Steam distillation is widely employed for the isolation of bioactive substances from natural sources. However, the process is usually time-consuming, high energy consumption and unwanted chemicals can be entrained with the main products. Microwave-assisted extraction is now recognized as an efficient extraction technique that dramatically reduces extraction time, increases yields and higher quality of the extracts can be obtained. The aim of this study is the evaluation of the major factors affected by operating parameters such as concentration of solvent, radiation time,ratio of microwave power to material, volume of solventon yield of products by microwave-assisted extraction. In addition, comparison between traditional extraction methods and microwave-assisted extraction is also carried out. The results show that the application of microwave energy for the extraction of active substances from fresh ginger have faster heating, reduce thermal gradients and extraction time.

Keywords: ginger oleoresin, microwave - assisted extraction, modeling, kinetics, response surface methodology.

1. INTRODUCTION

The active compounds from natural plants can be extracted using traditional methods such as steam distillation andSoxhlet extraction, and non-traditional methods such as microwave-assisted extraction (MAE), ultrasound - assisted extraction (UAE), and supercritical fluid extraction (SFE). However, traditional methods have some disadvantages, for example, long timeoperation, high energy consumption, and decomposition of active components. Therefore, modern extractive methodswere paid attentionin recent years. One of the potential methods is microwave - assisted extraction (MAE) [1]. This technology was first demonstrated in 1992 by Pare [2]. The main advantage of microwave – assisted extraction is fast, uses low quantities of solvent, and is adaptable not only at laboratorybut also atindustrial scales. The parameters whichinfluence the extraction efficiency are namely solvent and volume, extraction time, microwave power, plant matrixcharacteristics and temperature. Figure 1 shows the typical systems of microwave-assisted extraction, which is often classified byopened and closed vessel extraction. Closed vesselextraction is performed under controlled pressure and temperature while openedvessel extraction is only irradiatedby microwave energy. Principal elements of a microwave device include four major components: microwave generator, wave guide, applicator where the sample is placed, and circulator [3].

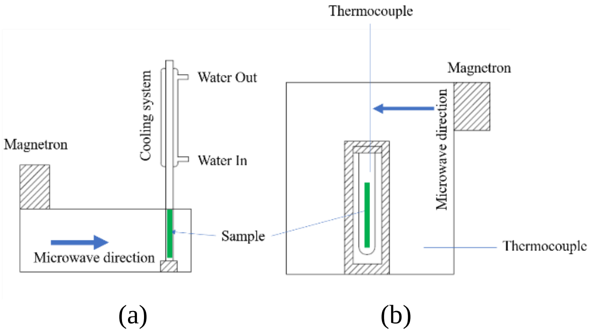

Figure 1. Opened (a) and closed (b) vessel microwave - assisted extraction.

One of the main advantages of MAE process is short extraction time. The reason is here that the mass and heat transfer are progressed in the same direction while in conventional extraction these two processes are developed inreverse directions as shown in Figure 2 [1,2]. Moreover, in MAE, the microwave energy is delivered directly to materials through molecular interactions with the electromagnetic field via conversions of electromagnetic energy into thermal energy.

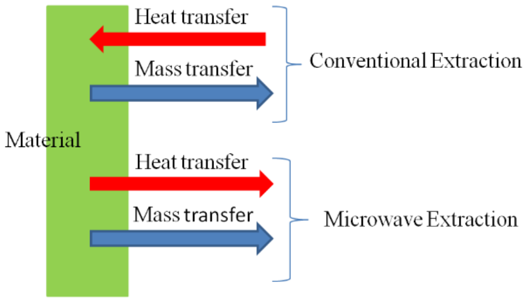

Figure 2. Heat and mass transfer mechanisms in conventional and microwave extraction.

Ginger has been used for a medicine, cooking, and condiment. Ginger essential oils and oleoresins are main products from fresh ginger. Both can beused in food and pharmaceutical industries. The main compounds of the ginger oleoresin are series of gingerols and shogaols. Figure3 shows the structure of major active compounds of ginger oleoresin.

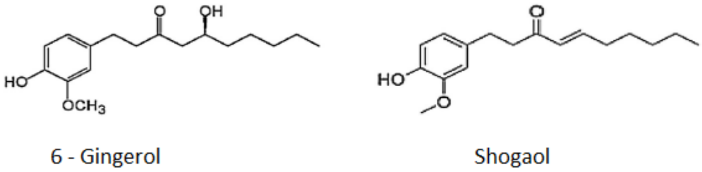

Figure 3. Major active compounds in ginger oleoresin.

Rahul *etal.* extracted essential oil from ginger using water as a solvent on a modified conventional microwave oven. In this paper, the results showed that the solvent and size of raw material affected the extraction process [4]. Kubra *et al.* studied the effects of extraction temperature, operation time and solvent at constantmicrowave power on the performance of process [5]. In their work, aqueous ethanol (50 % volume) and microwave irradiation time of 60s were reported to be the optimal operating conditionproducing the highest extracted yield. This paper also commented that MAE could produce higher recoveries compared to conventional extraction processesalong with the improvement in the quantity and quality of extracts in very short operating time (3 to 5 min). Liu *et al.* optimized microwave assisted extraction of 6-gingerol from zinger [6]. The optimum extraction conditions were microwave power of 528 W at the liquid/solid ratio of 26 ml/g, extraction time of 31 s using 78 vol.% ethanol-water solution as a solvent. Alfaro *et al.* studied influences of solvent, matrix dielectric properties, and applied power on the extraction of ginger [7]. In their work, microwave assisted liquid phase extraction of ginger had been optimized using open vessel apparatus. They stated that the highest extraction ginger yield was obtained with ethanol solvent and water as modifier at power levelof 150W during 120s. Spiro *et al.* studied kinetics of ginger rhizome extraction with different solvent such as dichloromethane, ethanol, 2–propanol and acetone – water mixture [8]. They reported that the yield of extraction decreased in the sequence acetone, acetone and water, dichloromethane, ethanol, 2-propanol at 30 ^{o}C. Tian *et al.* studied the influence of microwave irradiation treatment on the extraction of ginger [9]. Results obtained from their work indicated that the most satisfying results can be achieved by pretreatment of microwave irradiation under the output power of 480 W during 6 min.

The main objectives of the present study are to produce ginger oleoresin from Vietnam fresh ginger using microwave - assisted extraction. Four parameters that effect on the yield of ginger oleoresin including radiation time, microwave power to raw material ratio, volume of solvent and concentration of solvent are examined. The response surface methodology is also applied to find optimization ginger oleoresin extraction by MAE.

2.MATERIALS AND METHODS

2.1. Materials

Fresh ginger (*Zingiberofficinale*Rosc)is collected from the local farm at Tam Dao district (~ 7 month age) and is used as raw material in this study. The samples are cleaning, peeling and cutting into pieces (1 – 2 mm). The microwave assisted extraction is performed undertaken with ethanol and water mixture as a solvent. The filtration process is used to remove the solid phase. The solution gets from the filtration is then evaporated in a rotary concentrator. Figure4 shows the diagram of produce ginger oleoresin extraction.

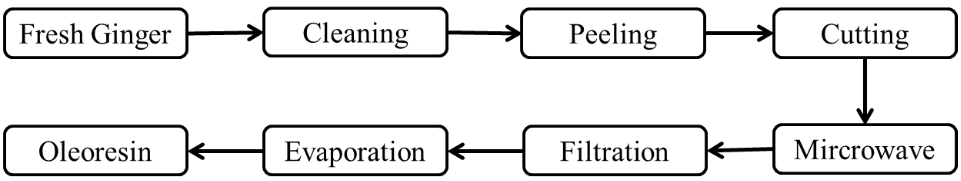

Figure 4. Flow diagram of ginger oleoresin extraction.

2.2. Method of microwave-assisted extraction

Microwave-assisted extraction experiments are performed on a commercial microwave oven [Model: KOR-4A17, microwave input power: 900 W, microwave energy output: 500W]. The extraction processiscarried out in a closed vessel microwave assisted extraction system at 500 W.

The efficiency of closedvessel MAE strongly relies on the selection of the operating conditions. The factors that may affect the performance of closed vessel MAE are solvent, solvent to feed ratio, extraction time, microwave power to material ratio, temperature, and sample characteristic. It is important to understand the influences and interactions of these factors on the closedvessel MAE processes. In the extraction process, there are some solvents can be used for ginger extraction such as ethanol, methanol, water, hexane and acetone [10]. Oleoresin of ginger is obtained from raw material by extraction with a non-aqueous solvent. The most widely used co-solvent is ethanol and water as modifier with concentration ranging from 10 % to 100 % volume [5]. The physical properties of water and ethanol solvents as shown in Table 1 indicate that these two liquids are strongly activated by microwave irradiation. In general, the high extraction yield is obtained with the increase of microwave power. However, extraction yield becomes decline due to degradation of thermal sensitive compounds if microwave power is higher. On the other hand, microwave power applied to the extraction depends on the mass of the plant. In this study, the power level is set at 500 W with 50 $\pm$ 0.1 g of fresh material. Depending on materials, the extraction time of MAE usually varies from few minutes up to an hour due tothe risk of thermal degradation. The short radiation durationof 1, 2, 3, 4, and 5 min. are investigated in the present work since the extraction is carried out in a closed vessel.

Table 1. Physical properties of water and ethanol as the solvent in MAE.

Solvent	**Dielectric constant[(a)] ε'**	**Dissipator factor tan δ (x 10^{-4})**	**Boiling point[(b)] t (^{o}C)**	**Viscosity[(c)] (cP)**
Ethanol	24.3	2,500	78	0.69
Water	78.3	1,570	100	0.89

(a) Determined at 20 ^{o}C
(b) Determined at 101.4 kPa
(c) Determined at 25 ^{o}C

The yield of ginger oleoresin Y (%) is determined by mass of extracted active compound × 100/mass of sample.

In this study, response surface methodology (RSM) is used for optimizing the yield of ginger oleoresin extraction. According to the Box-Behnken design, 29 experimental runs are prepared for combinations of independent variables including concentration of solvent, ratio of microwave power to raw material, extraction time, and volume of solvent.

3. RESULTS AND DISCUSSION

3.1. Preliminary analysis

3.1.1. Effect of solvent concentration

In this section, four different solutions with ethanol concentrations of 10 %, 50 %, 70 % and 100 % volume are used, respectively. The ratio of power level and material is 500/50 W/g, the volume of solvent is 50 ml, and microwave exposing time remains at 2 min. The results of ginger oleoresin with various concentration of solventare given in Table 2.

Table 2. Effect of ethanol concentration on ginger oleoresin yield.

Run	**Solvent (% volume)**	**Power lever/ material (W/g)**	**Volume of solvent (ml)**	**Time (min)**	**Oleoresin (%)**
1	Ethanol 100 %	500/50	50	2	1.8
2	Ethanol 70 %	500/50	50	2	2.1
3	**Ethanol 50 %**	**500/50**	**50**	**2**	**3.2**
4	Ethanol 10 %	500/50	50	2	1.9

It can be seen from Table 2 that ethanol concentration is the hopeful at 50 % to obtain 3.2 % ginger oleoresin. The reason can be explained that the dielectric constant of aqueous ethanol at this concentration is highleading to the increasein absorption ability of microwave energy. At a higher or lower concentration the oleoresin yield tends to be reduced (see Table 2). This phenomenon may be explained that, at higher concentration, ginger oleoresin yield is decreased due to the solubility of oleoresin. On the other hand, at a lower concentration, the interaction between solvent and matrix of material is not sufficient for the extraction [8]. The results obtained from this study are suitable with the work of Kubra [5].

3.1.2. Effect of radiation time

Another important parameter concerned in the paper is radiation time. The experiments performed under various extraction times. Based on the results given in Table 2, concentration of ethanol is 50 % volume and power level and material ratio are 500/50 W/g, the volume of solvent is 50 ml, and extraction time changes in the range 1 min to 5 min. Table 3 shows the effect of the radiation time on yield of ginger oleoresin.

Table 3. Effect of extraction time on ginger oleoresin yield.

Run	**Solvent (% volume)**	**Power level/material (W/g)**	**Volume of solvent (ml)**	**Time (min)**	**Oleoresin (%)**
1	Ethanol 50 %	500/50	50	1	0.8
2	Ethanol 50 %	500/50	50	2	3.8
3	Ethanol 50 %	500/50	50	3	6.0
4	**Ethanol 50 %**	**500/50**	**50**	**4**	**6.6**
5	Ethanol 50 %	500/50	50	5	5.7

The results showed that the highest yield of ginger oleoresin of 6.6 % can be achieved at 4 min of extraction time. At shorter radiation time, mass and heat transfer between materials and solvent is very low and not enough of time to extract active compounds from raw material. Therefore, ginger oleoresin yield is smaller. At longer radiation time, operation temperature and pressure in extraction vessel are too high. Thus, ginger oleoresin may be decreased because of decomposition into the solvent.

3.2.Statistical analysis

Table 4. Experimental design for Box – Behnken method.

Run	Code Variables				Response Y (%)
	Factor A (W/g)	Factor B (min)	Factor C (%)	Factor D (ml)	
1	1.00	-1.00	0.00	0.00	3.2
2	-1.00	0.00	1.00	0.00	1.4
3	0.00	0.00	1.00	-1.00	1.1
4	0.00	0.00	1.00	1.00	1.3
5	0.00	1.00	0.00	-1.00	2.5
6	1.00	0.00	0.00	1.00	9.2
7	0.00	-1.00	1.00	0.00	1.3
8	-1.00	1.00	0.00	0.00	6.4
9	-1.00	-1.00	0.00	0.00	4.1
10	0.00	0.00	-1.00	1.00	3.5
11	0.00	1.00	-1.00	0.00	4.1
12	-1.00	0.00	-1.00	0.00	2.6
13	0.00	-1.00	0.00	-1.00	6.2
14	0.00	1.00	1.00	0.00	1.6
15	0.00	-1.00	0.00	1.00	4.2
16	0.00	0.00	0.00	0.00	8.5
17	1.00	1.00	0.00	0.00	10.2
18	0.00	-1.00	-1.00	0.00	3.5
19	1.00	0.00	0.00	-1.00	6.9
20	-1.00	0.00	0.00	-1.00	4.6
21	0.00	0.00	0.00	0.00	8.2
22	0.00	0.00	-1.00	-1.00	1.5
23	-1.00	0.00	0.00	1.00	7.5
24	0.00	1.00	0.00	1.00	9.3
25	0.00	0.00	0.00	0.00	8.4
26	1.00	0.00	-1.00	0.00	4.1
27	1.00	0.00	1.00	0.00	1.8

In the section, RMS is utilized to the modeling of microwave-assisted extraction process [11]. Using the above preliminary analysis to determine optimal conditions, four-factor Box – Behnken method is employed to design experimental matrix as shown in Table 4 with 27 experimental runs. Four independent variables are considered in which ratio of microwave power to raw material (Factor A: 5 W/g, 10 W/g, and 15 W/g), extraction time (Factor B: 1 min, 5.5 min, and 10 min), concentration of ethanol (Factor C: 10 %, 50 %, and 90 %), and volume of solvent (Factor D: 10 ml, 50 ml, and 90 ml). The experimental data shows that value of ginger oleoresin varied from 1.1 % to 10.2 %. The experimental results are validated by using response surface analysis (F- value, p – value, LOF value, and R - squared) and to estimate the influencing factors. The model F - value is 12.58 that implies the model is significant. Values of

Prob> F (p – value) is less than 0.05 indicate model terms are significant. R – squared is 0.9263 and LOF value is 0.087. Based on the result, the quadratic model is suitability for predicting the optimum response value. The equation is an empirical relationship between ginger oleoresin yield and code variables is given Equation (1) as follows.

$$Y (\%) = 8.36 + 0.73A + 0.97B – 0.9C +1.02D + 1.18AB - 0.27AC - 0.15AD – 0.075BC + 2.20BD – 0.45CD – 0.68A^2 – 1.36B^2 – 4.96C^2 – 1.21D^2 \quad (1)$$

Based on the ANOVA analysis, B, C, D, BD, B^2, C^2, D^2 are significant model terms (p – value < 0.05). Therefore, we can attempt to simplify model by removing items that are insignificant. The equation (2) represents the mathematical model for maximum ginger oleoresin yield.

$$Y (\%) = 8.36 + 0.97B – 0.9C +1.02D + 2.2BD – 1.36B^2 – 4.96C^2 – 1.21D^2 \quad (2)$$

The interactions of independent variables are described using three – dimensional response surface plots as shown in Figure 5.

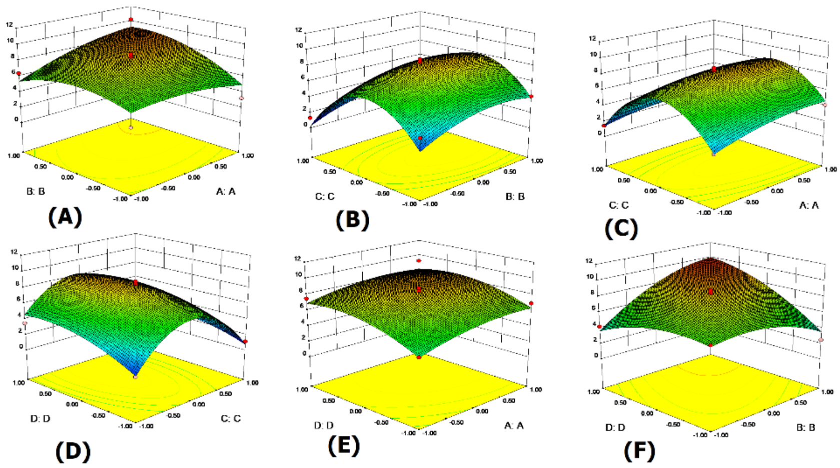

Figure 5. Response surface plots showing the effects of variables.

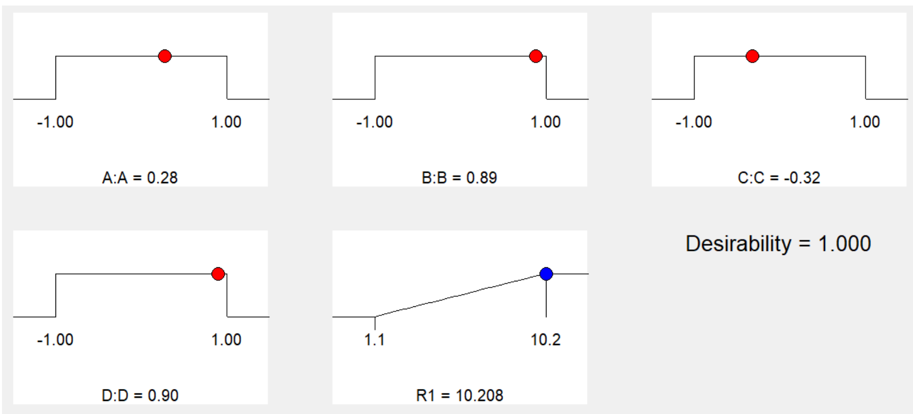

Figure 6. Optimization of the process by Ramp function.

For example, when the concentration of solvent (factor C) from 90 % to 10 %, the yield of ginger oleoresin first increases, and then declines as shown in Figure 5B– 5C – 5D. The ratio of microwave power to material (factor A), the extraction time (factor B), and the volume of solvent (factor D)havea similar effect on the yield of ginger oleoresin but have a less effect on the yield of ginger oleoresin than concentration of solvent (see in Figure 5A to 5F).

In order to verify the model's accuracy, the maximum ginger oleoresin yield is tested using recommended optimum condition (Factor A = 0.28; Factor B = 0.89; Factor C = -0.32; and Factor D = 0.90; Y (%) = 10.208) (see in Figure 6). The yield of ginger oleoresin is 9.7 % obtained from the experiment. Thus, the predicted value from equation model and experimental data are in good agreement (Relative error is 5.23 %). The small difference between actual and predicted values reflects themodel's accuracy.

3.3.Comparison with traditional extractions

In order to compare to traditional extraction methods, maceration and Soxhlet extraction methods are studied. Maceration extraction is performed using 50g of fresh ginger in 50 ml of ethanol 50 vol.% solvent for 45 hours. After filtration process, ginger oleoresin is achieved by removing solvent in rotary concentrator. Yield of ginger oleoresin is 7.5 % and smaller than that using microwave assisted extraction (10.2 %). Moreover, the maceration time of 45 hours is much longer compared to microwave extraction time of 4 min. In the case of Soxhlet extraction, similar fresh ginger is extracted bythis method for 8 hours using ethanol 50 vol.% solution at the temperature of 80^{o}C. The results show that the yield of ginger oleoresin is 12.5 %. Clearly, that ginger oleoresin collected from the Soxhlet extraction is the highest compared to other methods. However, the maximum yield of ginger oleoresin obtained in this study is 10.2 % with a very shortextraction time. Thus, it is clear that the energy consumption in MAE is the lowest compared to the other two methods, while the recovery of the ginger oleoresin is good.

4. CONCLUSIONS

Closed vessel MAE system is used to extract ginger oleoresin of Vietnamesefresh ginger. Response surface methodology is used to optimization and predict the yield of ginger oleoresin by microwave-assisted extraction method. Four – factor Box Behnken is designed with 27 experimental runs. It is conclusion that the concentration of solvent, the ratio of microwave power to raw material, the extraction time, and the volume of solvent are important parameters to effect on the yield of ginger oleoresin in microwave assisted extraction. The optimal condition of extraction process should be chosen:ratio of microwave power to raw material 12.8 W/g, extraction time from 8 min, concentration of solvent from 40 % and volume of solvent from 80 ml.

Acknowledgements. This work is funded by Hanoi University of Science and Technology under the project T2017-PC-010.

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