Box-behnken design for the optimization using solvent-free microwave gravity extraction of garlic oil from *Allium sativum* L.

Y Variyana, R S C Muchammad and M Mahfud*
Department of Chemical Engineering, Institut Teknologi Sepuluh Nopember, Sukolilo, Surabaya 60111, Indonesia

*E-mail: mahfud@chem-eng.its.ac.id

**Abstract.** Garlic (*Allium sativum* L.) has compounds that could be used as anti-microbial, anti-atherosclerosis, anti-oxidant, anti-viral, anti-cholesterol, anti-diabetic, anti-hypertension, and anti-cancer. In this paper, oils were extracted from garlic using Solvent-Free Microwave Extraction (SFME) method without solvent which is as an alternative technique to produce oil and it has several advantages in terms of product quality and quantity. This research used technique of gravity for extraction process and produce high yield of garlic oil. The highest results were obtained from this study when at 100 g, 450 W, 10 min, and 0.5 cm. Response surface methodology was designed to evaluate the effects of mass of raw material (g), microwave power (W), extraction time (min) and material size (cm) for optimization of analytical methods. Regression models have designed and predicted experimental values well. Gas chromatography-mass spectrometry (GC-MS) technology was used to analyze the chemical compositions of garlic oil. The results showed that sulphide compounds, like diallyl disulfide, trimethylene trisulfide, ester and alcohol which is found in garlic oil.

1. **Introduction**

Garlic (*Allium sativum* L.) is quite easily found in tropical countries, in Asia such as Indonesia. This is because garlic has become the main ingredient in Asian cuisine and is easy to grow in tropical countries. Choosing of good garlic is either consumed raw or cooked form. As the main ingredient of cooking, garlic has a role to give a savory and slightly spicy flavor into cuisine. Garlic has also been functioning as an aroma enhancer in cooking. Besides that, garlic is a natural flavor enhancer that is free of chemicals. Therefore, most Indonesian dishes use garlic as the main ingredient. There is interest in garlic (*Allium sativum*) and not only garlic extract for applications as food ingredients, but also as pharmaceutical sector. Garlic is used as antihypertensive [1,2], antithrombotic agents [2], antimicrobial activities [2,3], antibacterial agents [4], fibrinolytic [5], immunomodulatory [6], cancer prevention [7], and etc. Moreover, Garlic oil is used in pharmaceutical applications, one of which is reported to have anticoagulation activity of plasma to inhibit diabetes mellitus because of biological activities [8]. One of the chemical components in garlic is sulfur. Sulfur as macro elements was contained in garlic (*Allium sativum*).

The extraction of solid-liquid is the process of taking bioactive compounds from natural plants. Several techniques of conventional separation are currently used for the extraction of oils from natural plants, including maceration, soxhlet, reflux, and stirring methods. However, these methods have several disadvantages that have long extraction time, low yield, and high cost to operate the extraction.
process. Recently, several methods of microwave applications were used to extract oil from garlic. It has been reported by comparison of simultaneous distillation solvent extraction (SDE), microwave-assisted hydro-distillation extraction (MWHD) and ultrasound-assisted extraction (UAE) to isolate of the compounds from garlic [9]. For faster production and lower temperature, supercritical carbon dioxide extraction was done for extracting the garlic oil [10]. In addition, The solvent extraction (SE) and supercritical fluid extraction (SFE) of product were applied for sensitive heat compounds of garlic volatile oil [11]. However, the extraction process using supercritical method requires operating complex conditions, high pressure and expensive of capital cost. Nevertheless, in supercritical fluid extraction (SFE), the method has also several disadvantages for the use of its industry because of small extraction capacity and high equipment investment [11,12]. The research was reported by to analyze the effect factors using microwave-assisted extraction and conventional extraction method for the extraction process from garlic [13]. Extraction technology using microwave energy is likely to produce effective heating in the process and to obtain good quality and quantity of oil products from the extraction process. Microwave heating is very effective when used in moist materials. Because of contains of water who can well absorb energy from microwaves, in separation on microwave called dielectric constant [14]. Garlic has a water content about 65% [15]. The extraction performance of garlic using microwave has the potential to reduce extraction time, produce higher yields and better extract quality.

Solvent-free microwave extraction (SFME) has been applied to the extraction of a number of fresh natural plants with high potential for larger extraction applications [16-18]. In terms of quality and quantity of extract, SFME seems to be more economical and competitive than conventional methods [19]. SFME is a combination of microwave heating and distillation occurring at atmospheric pressure. Then, microwave gravity technique e is a green technique for extracting essential, volatile and aromatic oils. According to Vian et. al (2008) that a physical phenomenon in gravity technique, known as hydro-diffusion, allows extracts (water and essential oils), to spread beyond plant material, to descend by earth gravity out of the microwave reactor and fall through hollow Pyrex disks [20]. Gravity is not only economical and efficient technology, but also main advantages are green technology and environmental friendly because it works without using any water being added under the influence of microwave and earth gravity at atmospheric pressure [20]. In this paper, combination of solvent-free microwave gravity extraction has been potential technique.

In this study, the box-behnken design (BBD) was applied by response surface methodology (RSM) to investigate and analysis the effect of extraction parameters for garlic oil results using the solvent-free microwave extraction in gravity. Furthermore, the chemical composition of the product is analyzed by gas chromatography-mass spectrometry technology (GC-MS).

2. Materials and Methods
2.1 Materials
Garlic (fresh condition) was purchased from a local market in Surabaya, Indonesia and n-hexane was used of analytical grade from Merck (Darmstadt, Germany). Then, garlic is peeled and chopped manually using a hand with several variables and does not appear to be damaged the external part is selected.

2.2 Methods
2.2.1 Solvent-free microwave gravity extraction. Microwave with gravity technique of extraction process did not require any solvent. The microwave was used for experiment which is Electrolux of the EMM2308X model with the following specifications: Maximum power of 800 W, 220V voltage, 1250W power, Magnetron frequency of 2450 MHz (2.45 GHz). Reactor as to contain material inside microwave connected to Condensor below it and in the bottom separating funnel as container. For SFME method, 1000 ml sized reactor (round bottom flask) containing mass of raw material (50;100;150 g) was placed in the microwave. Microwave power were set at 300;450;600 W and extraction time was 5;10;15 min. After extraction process, the garlic oil was separated by n-hexane to
collect the pure extract. Then, it was evaporated over to remove excess of n-hexane at atmospheric condition. Garlic oil was weighed and stored in vial at 4°C for further analysis. The extraction yield of garlic oil was calculated according to the equation as follows:

\[
\text{Extraction yield (\%) } = \frac{\text{Mass of garlic volatile oils}}{\text{Mass of fresh garlic (1-moisture content(\%))}} \times 100
\]  

(1)

2.2.2 Experimental design and statistical analysis of extraction yield from Allium sativum L. Table 1 shows of independent variable responses and code levels derived after preliminary runs. Four-factors from Box-Behnken Design (BBD) was applied by Li (2010) to optimize the extraction conditions in order to obtain a high extract from fresh Allium sativum L [11]. The independent variables were set of mass of raw material (g), microwave power (W), extraction time (min) and material size (cm).

| Factors                  | Unit | Low level | High level |
|--------------------------|------|-----------|------------|
| Microwave power          | W    | 300       | 600        |
| Mass of raw material     | g    | 50        | 150        |
| Extraction time          | min  | 5         | 15         |
| Material size            | cm   | 0.5       | 1.5        |

A total of 29 experiments were designed in table 2 where each experiment of extraction yield was responded, Y (yield, %). This is the same as the design carried out by Ye & Lai (2012) [21]. Regression analysis was performed for the experimental data and was fitted into the second-order model, as follows equation below:

\[
Y = \beta_0 + \sum_{i=1}^{3} \beta_i X_i + \sum_{i=1}^{3} \beta_{ii} X_i^2 + \sum_{i=1}^{3} \sum_{j=i+1}^{3} \beta_{ij} X_i X_j
\]

(2)

where \( \beta_0 \), \( \beta_i \), \( \beta_{ii} \) and \( \beta_{ij} \) are regression coefficients in the intercept, linear, quadratic, and interaction terms, respectively; \( X_i \) and \( X_j \) are the independent variables. The Design-Expert 11 software package (trial version; State-Ease Inc., Minneapolis, USA) was used to get the coefficients of the polynomial quadratic model.

2.3 Gas Chromatography–Mass Spectrometry (GC-MS)
GC-MS analysis was analyzed by Agilent 6980N GC system (Agilent, USA). The detector was used an Agilent 5973 inert MSD. The performed and analyzed were separated using an Agilent J&W Scientific column (0.25 mm x 30 m x 0.25 um), which was inserted directly into the ion source of the MS system. The oven temperature was set as follows: 50°C for 5 min, then programmed to 280°C at 10°C/min, where it was maintained for 15 min. Flow rate provided of 1 ml/min (constant). The split ratio was 1/2. The detector temperature was set at 250°C. The Mass Spectrometry (MS) of quad and sources were set in 150°C and 230°C. The mass range was scanned in 20–600 amu for the full-scan acquisition mode and solvent delay was 0 min. Each compound was identified by comparison of its retention time and mass spectrum in NIST library.

3. Result and Discussion
3.1 Model Summary Statistics for Fitting a Second-Order Model
Design-Expert tests to fit a series of models from response surface to the data are linear (first-order), 2FI (two factor interaction), quadratic (second-order), and cubic. The significance of p-value are less than 0.05 of quadratic model vs 2FI (see table 3). Therefore, model summary in table 4 showed that standard of deviation less than 0.05 is only quadratic model. The accuracy of lack of fit is quadratic and cubic models. In this optimization, the quadratic model is preferred. Moreover, adjusted R² and
predicted $R^2$ should close to 1. In addition, the difference between predicted $R^2$ and adjusted $R^2$ is less than 0.2. Therefore, summary statistics suggest from the response to select the quadratic model for this extraction process, which is model recommended by Design-Expert.

Table 2. Box-behnken design (BBD) matrix and response for the extraction yield.

| Run | Microwave power (W) | Mass of raw material (g) | Extraction time (min) | Material size (cm) | Observed | Predicted | Residual |
|-----|---------------------|--------------------------|-----------------------|-------------------|----------|-----------|---------|
| 1   | 150                 | 300                      | 10                    | 1                 | 0.1521   | 0.1506    | 0.0015  |
| 2   | 100                 | 450                      | 15                    | 0.5               | 0.2237   | 0.2316    | -0.0079 |
| 3   | 100                 | 300                      | 10                    | 0.5               | 0.1624   | 0.1669    | -0.0045 |
| 4   | 100                 | 450                      | 10                    | 1                 | 0.2432   | 0.2380    | 0.0052  |
| 5   | 150                 | 450                      | 5                     | 1                 | 0.1642   | 0.1588    | 0.0054  |
| 6   | 100                 | 600                      | 10                    | 0.5               | 0.2197   | 0.2095    | 0.0102  |
| 7   | 150                 | 450                      | 10                    | 0.5               | 0.2112   | 0.2052    | 0.0060  |
| 8   | 50                  | 450                      | 15                    | 1                 | 0.2015   | 0.1981    | 0.0034  |
| 9   | 100                 | 450                      | 10                    | 1                 | 0.2346   | 0.2380    | -0.0034 |
| 10  | 50                  | 450                      | 10                    | 1.5               | 0.1201   | 0.1362    | -0.0161 |
| 11  | 100                 | 300                      | 5                     | 1                 | 0.1021   | 0.1114    | -0.0093 |
| 12  | 150                 | 600                      | 10                    | 1                 | 0.1678   | 0.1804    | -0.0126 |
| 13  | 100                 | 600                      | 5                     | 1                 | 0.1292   | 0.1390    | -0.0098 |
| 14  | 100                 | 450                      | 10                    | 1                 | 0.2400   | 0.2380    | 0.0020  |
| 15  | 100                 | 450                      | 10                    | 1.5               | 0.2219   | 0.2380    | -0.0161 |
| 16  | 100                 | 450                      | 5                     | 1.5               | 0.1325   | 0.1232    | 0.0093  |
| 17  | 100                 | 450                      | 10                    | 1                 | 0.2501   | 0.2380    | 0.0121  |
| 18  | 50                  | 600                      | 10                    | 1                 | 0.1586   | 0.1588    | -0.0002 |
| 19  | 100                 | 450                      | 5                     | 0.5               | 0.1879   | 0.1870    | 0.0009  |
| 20  | 100                 | 300                      | 15                    | 1                 | 0.1699   | 0.1702    | -0.0003 |
| 21  | 100                 | 450                      | 15                    | 1.5               | 0.1986   | 0.1982    | 0.0004  |
| 22  | 50                  | 300                      | 10                    | 1                 | 0.1454   | 0.1314    | 0.0140  |
| 23  | 50                  | 450                      | 10                    | 0.5               | 0.2093   | 0.2139    | -0.0046 |
| 24  | 100                 | 600                      | 10                    | 1.5               | 0.1602   | 0.1469    | 0.0133  |
| 25  | 100                 | 300                      | 10                    | 1.5               | 0.1309   | 0.1323    | -0.0014 |
| 26  | 150                 | 450                      | 10                    | 1.5               | 0.1801   | 0.1857    | -0.0056 |
| 27  | 50                  | 450                      | 5                     | 1                 | 0.1379   | 0.1344    | 0.0035  |
| 28  | 100                 | 600                      | 15                    | 1                 | 0.1989   | 0.1998    | -0.0009 |
| 29  | 150                 | 450                      | 15                    | 1                 | 0.2198   | 0.2145    | 0.0053  |

The fitting parameter showed that actual data could be evaluated to match the statistical result. The model coefficients are represented by constant terms, linear coefficients for independent variables, interaction coefficients, coefficients of quadratic. Then, the determination coefficient ($R^2$), the adjusted coefficient of determination (Adj-$R^2$) and adequate precision are used to evaluate the adequacy of the model that is focus on the model maximizing of the adjusted $R^2$ and the predicted $R^2$; this model is adequate when the value is $p < 0.05$; lack of fit ($p > 0.05$); and $R^2 > 0.9$ [22].
Table 3. Model fitting for the extraction process.

| Source            | Sum of squares | df   | Mean Square | F-value | p-value |
|-------------------|----------------|------|-------------|---------|---------|
| Mean vs Total     | 0.9591         | 1    | 0.9591      |         |         |
| Linear vs Mean    | 0.0215         | 4    | 0.0054      | 4.95    | 0.0047  |
| 2FI vs Linear     | 0.0013         | 6    | 0.0002      | 0.1559  | 0.9853  |
| **Quadratic vs 2FI** | **0.0229**   | **4** | **0.0057**  | **42.88** | **< 0.0001** |
| Cubic vs Quadratic| 0.0011         | 8    | 0.0001      | 0.9763  | 0.5268  |
| Residual          | 0.0008         | 6    | 0.0001      |         |         |
| Total             | 1.01           | 29   | 0.0347      |         |         |

Table 4. Model summary of statistics for the extraction yield using response surface.

| Source   | Std. Dev. | R²   | Adjusted R² | Predicted R² | Lack of Fit p-value |
|----------|-----------|------|-------------|--------------|---------------------|
| Linear   | 0.0330    | 0.4521 | 0.3608      | 0.3096       | 0.0147              |
| 2FI      | 0.0371    | 0.4792 | 0.1898      | 0.0353       | 0.0085              |
| **Quadratic** | **0.0116** | **0.9607** | **0.9214** | **0.8132** | **0.4414** | **Suggested** |
| Cubic    | 0.0116    | 0.9829 | 0.9203      | -0.1171     | 0.3043              |

ANOVA in table 5 using quadratic model is generally significant. The factors are mass of raw material (A), microwave power (B), extraction time (C), and material size (D) show important values for all factors with responses. Moreover, the quadratic model could evaluate and analyze of the interaction of each factors. The interaction between the mass of raw material and microwave power (AB), mass of raw material and extraction time (AC), microwave power and extraction time (BC), microwave power and material size (BD), and extraction time and material size (CD) were insignificant (p >0.05) which means that the factors was less influenced for the extraction process from *Allium sativum*. However, the mass of raw material and material size (AD) interaction showed only significant results (p <0.05). Generally, the response of the all interaction of various factors and independent variables could be optimized using quadratic model.

The analysis of variance in ANOVA (table 5) for the quadratic model gives the result of p <0.05; lack of fit is p >0.05 that the model is logic and accurate relative to the pure error [23]; the model of F-value of 30.28 implies the model is significant; the results were indicated the goodness of fit from correlation of factors in regression analysis [24,25]. Thus, all factors were influenced for optimizing to produce yield. On the other hand, the model was indicated that significant result has been affected to analyze the optimal conditions. From these experimental data and predictive model could be applied and accepted for obtaining the highest yield.

The quadratic model (see table 6) resulted R² of 0.9607 (R² value is closer to 1), showing good correlation. The predicted R² of 0.8132 is in reasonable agreement with the adjusted R² of 0.9214. It is adeq precision >4 which indicates an adequate signal and model could be used to navigate the design. The standard deviation of the model is 0.0116. A small value of the standard deviation shows a good correlation that has a close value between the experimental data and predicted model for the response [26]. Moreover, high values of adequate precision (15.2273), which means the model could be used for optimization [27].
Table 5. Analysis of variance (ANOVA) for response surface from quadratic model to identify significant factors.

| Source of variations | Sum of Squares | df | Mean Square | F-value | p-value |
|----------------------|----------------|----|-------------|---------|---------|
| Model                | 0.0457         | 14 | 0.0033      | 24.44   | < 0.0001 significant |
| A - mass of raw material | 0.0012     | 1  | 0.0012      | 9.35    | 0.0085 |
| B - microwave power  | 0.0025         | 1  | 0.0025      | 18.37   | 0.0008 |
| C - extraction time  | 0.0107         | 1  | 0.0107      | 80.22   | < 0.0001 |
| D - material size    | 0.0071         | 1  | 0.0071      | 53.12   | < 0.0001 |
| AB                   | 1.562E-06      | 1  | 1.562E-06   | 0.0117  | 0.9154 |
| AC                   | 0.0000         | 1  | 0.0000      | 0.1198  | 0.7344 |
| AD                   | 0.0008         | 1  | 0.0008      | 6.32    | 0.0248 |
| BC                   | 9.025E-07      | 1  | 9.025E-07   | 0.0068  | 0.9357 |
| BD                   | 0.0002         | 1  | 0.0002      | 1.47    | 0.2458 |
| CD                   | 0.0002         | 1  | 0.0002      | 1.72    | 0.2110 |
| A²                   | 0.0061         | 1  | 0.0061      | 45.60   | < 0.0001 |
| B²                   | 0.0175         | 1  | 0.0175      | 131.28  | < 0.0001 |
| C²                   | 0.0062         | 1  | 0.0062      | 46.28   | < 0.0001 |
| D²                   | 0.0032         | 1  | 0.0032      | 23.65   | 0.0003 |
| Residual             | 0.0019         | 14 | 0.0001      |         |         |
| Lack of Fit          | 0.0014         | 10 | 0.0001      | 1.27    | 0.4414 |

Table 6. Fit statistics from ANOVA for the extraction process of garlic oil.

| Fit statistics                  | Response |       |
|---------------------------------|----------|-------|
| Standard deviation              | 0.0116   |       |
| Mean                            | 0.1819   |       |
| Coefficient of variance (CV) %  | 6.36     | 0.9607|
| R²                              |          | 0.9214|
| Adjusted R²                     | 0.8132   |       |
| Predicted R²                    |          |       |
| Adeg Precision                  | 15.2273  |       |

3.2 Analysis single factor

The effect of the mass of raw material on extraction yield is shown in figure 1. This result is generated from the single effect shown for the optimization process using response surface. As expected, on the influence in mass of raw material, there is an increase for extraction yield at mass of 50 g to 100 g. However, there was decreased in yield at 150 g. The results increase with the increase in the ratio of solvents to raw materials under certain conditions which are more suitable for producing the higher yield [11]. This increase results from a decrease in the concentration of the extract solution, which leads to an increase in the solvent to dissolve the oil in extract. Thus, from this study, the effective in mass of raw material of Allium sativum is 100 g.

Figure 2 showed that is microwave heating, due to the effects of volumetric heating, a faster increase in temperature could be obtained, depending on microwave power and the dielectric loss factor of the irradiated material. The increase in yield from microwave power 300 W to 450 W. Then, in microwave power 600 W there was a significant decrease in yield. This strong absorption gives an increase in temperature in the sample which causes the cell to rupture in situ. However, the strength
should not be too high if there is no loss of secondary metabolites [28]. Therefore, the recommended microwave power for the extraction process from *Allium sativum* of 450 W.

![Figure 1](image1.png) **Figure 1.** Effect of single parameter is mass of raw material.

![Figure 2](image2.png) **Figure 2.** Effect of single parameter is microwave power.

![Figure 3](image3.png) **Figure 3.** Effect of single parameter is extraction time.

![Figure 4](image4.png) **Figure 4.** Effect of single parameter is material size.

As shown in figure 3 that the extraction time has a significant impact on the yield of garlic oil. The extraction yield of essential oil increased with the increase of extraction time in the range of 5-10 min after that slightly decreased in the condition of 15 min. Generally, longer extraction time would increase for the extraction yield. It might declined after reaching 10 min are occured the decomposition of material and material might degradation of extract. The extract will give decreasing yield with the increase of extraction time. It will easily decompose when exposed to high temperatures for long periods of time [29]. Therefore, the optimal extraction time was chosen at 10 min for gravity extraction technique.

Particle size was also an important factor for the extraction process from natural materials. The results from figure 4 indicated that effect of material size was analyzed. The result showed that the
yield of garlic oil increased as the material size decreases. This is explained by the fact that when the mass of *Allium sativum* is peeled, the cells containing oil were broken, making infiltration of water as internal solvent. Similarly, as the material is peeled into smaller, materials containing the oil are broken in larger quantities, making water diffusing into the oil from *Allium sativum* more quickly [23]. Moreover, particle size was small that caused for high surface area, high diffusivity and permeability into material [30]. This rapidly pushes the extract of oil out under the influence of microwave energy, leading to higher performance. The extraction time of 60 min is recommended based on the experimental results.

3.3 Analysis of Contour Plots and Interaction

From figure 5 showed that contour plot of mass of raw material and microwave power (AB). Interaction both of two show that maximum extraction yield of garlic oil (0.2423%) was obtained under the experimental conditions of extraction in 100 g and 450 W. Clearly, an increase in mass of raw material and microwave power would be resulted the maximum yield. The extraction process happened from research to get the highest garlic oil yield at 450 W. However, the obtained garlic oil content ceases to rise, and then, starts diminishing after this one will cause decrease for the extraction yield. Then, the response of mass of raw material to get the highest yield at 100 g in this research. It also had a proportional value for contributing the garlic oil. Indeed, from contour plots, the process might be slightly as sensitive as between mass of raw material and microwave power.

Based on the optimized factors, figure 6 show the independent and interaction of parameters with the yield of the garlic oil could be evaluated. Generally, the extraction yield increases linearly with condition of factors. Then, the effects of mass and extraction time (AC) gives response around maximum conditions. The interaction both of two affects for contributing to produce on the yield. Surface plots proved that mass of raw material is more sensitive to produce the extraction yield than extraction time. Generally, the longer of the extraction time and mass would be obtained the higher on the yield. As similar result was conducted by Wang Y (2008) which states that longer of extraction time, the higher yield of extraction. Nevertheless, the yield is decreasing with the length of time when it has passed its optimum time [31]. It shows the maximum extraction yield near center of extraction number and mass of raw material.

Regarding yield, figure 8 shows the response for interaction of microwave power and extraction time (BC). Contours showing of microwave power and extraction time contributes to the yield. Visually, the longer extraction time the higher yield was. The position for the optimum state is around 450 W and extraction time is 10 min. The optimization resulted that the optimum point is in range of it. Actually, the extraction time can be extended, however, the components in extraction will be sensitive because of the addition of microwave power and extraction time. Therefore, the decrease of extraction yield with along extraction time or high extraction temperature could be explained by thermal degradation of garlic oil.

In the last interaction, the microwave power and material size (BD) were investigated. Contour from figure 9 shows the influence of microwave power which is more dominant than the material size. If increased microwave power, it will result in a decrease of extraction yield.

The experimental data and predicted model from figure 10 have intersect at that be considered as good correlation. This confirms that using a quadratic model could be used as an optimization method to determine the optimal conditions for the extraction process from *Allium sativum* L. The results of the graph show clearly the point between experimental data and predictive model are good and almost precision. Besides that, it corresponds to predicts and experimental data are scattered in linear lines at close range, indicating the experiment results are accurately predicted.
Figure 5. Response surface plots showing the interaction effect of mass of raw material and microwave power (AB).

Figure 6. Response surface plots showing the interaction effect of mass of raw material and extraction time (AC).

Figure 7. Response surface plots showing the interaction effect of mass of raw material and material size (AD).

Figure 8. Response surface plots showing the interaction effect of microwave power and extraction time (BC).

Figure 9. Response surface plots showing the interaction effect of microwave power and material size (BD).

The result in figure 11 showing a plot of the normal probability of residual students, each group of additional terms was assessed sequentially using the partial that F-test discussed before. The
experimental model is considered quite appropriate because the residue calculated follows a random pattern. Points are represented as squares according to the actual running experiments. The colors indicate an increase in the order of actual results. In addition, it shows that data points are match to modeling. The correspond to predictive and actual data are spread over a 45-degree line with a short distance, indicating that actual results are accurately predicted from factor values [23]. Thus, based on the data analysis of the extraction yield from experiments, the final quadratic model is showed as follows:

\[
Y_{\text{ield}}(\%) = 0.23796 + 0.0102A + 0.0143B + 0.02432D + 0.000625AB + 0.014525AD + 0.000475BC - 0.007BD + 0.007575CD - 0.03064A^2 - 0.05199B^2 - 0.03087C^2 + 0.02207D^2
\]  

Figure 10. Model graph for comparison between experimental data and predicted model.

3.4 Confirmation experiments

Design-Expert uses direct search methods to maximize the desirability function. The response for each noise factor after this optimization is done using a desirability solution. The study reported by Raissi & Farsani (2009), the desirability function approach of target value when the desirability value is 1 [33]. The desirability function from this optimization is 1. Solution 1 has the highest overall desirability. Furthermore, in the study from *Allium sativum* was to evaluate the effect of parameters using solvent-free microwave extraction method and microwave with gravity technique. Based on the solution model of optimization, the optimum conditions calculated of garlic oil when mass of raw material (103.395 g), microwave power (464.281 W), extraction time (12.213 min) and material size (0.676 cm). While, the results of the experimental data to obtain the highest yield conditions at 100 g, 450 W, 10 min, and 0.5 cm is 0.2432%, which is very close to the predicted value of 0.2510%. This indicates that the model is accurate to predict the expected optimal condition. On the other hand, the error rates between the experimental and predicted model which are less than 5%. Therefore, the extraction process from *Allium sativum* for any parameters of mass of raw material, microwave power, extraction time, and material size could be accurately predicted by the regression models designed by response surface methodology.

3.5 GC-MS analysis

The results of GC-MS analysis showed the composition of garlic oil using the method solvent-free microwave gravity extraction given in Table 5. In addition, The typical compositions from mass spectra of the resulted garlic oil by GC-MS is almost similar compounds with others research [9,11], [34–36].
Based on the analysis, the components contained in volatile oils could be classified into several compounds, namely monoterpenes, oxygenated monoterpenes, sesquiterpenes, oxygenated sesquiterpenes, other compounds, and other oxygenated compounds. Furthermore, the components of oxygenated compound have more influence on the aroma of volatile oils compared to monoterpene compounds. In this study, based on GC-MS test, the amount of oxygenated compound found in garlic oil is 15.35%. The method applied to extraction process for garlic has the advantage of having the highest levels of oxygenated compound so that the aroma of the oil produced is better. Some things that affect oxygenated compound levels, namely, a reduction in thermal and hydrolytic effects on this combination method.

Table 5. Chemical composition derived from garlic oil by GC-MS.

| Compound                                          | Retention time (min) | % Area  |
|---------------------------------------------------|----------------------|---------|
| 2,6,6-trimethyl-                                  | 2.542                | 0.645   |
| Octamethylocyclotetrasiloxane                     | 2.806                | 2.609   |
| 6,6-dimethyl-2-methylene-                          | 2.912                | 0.417   |
| Diallyl disulphide                                 | 3.970                | 1.735   |
| 6-Methyl-ergoline-8-carboxylic acid methyl ester  | 4.594                | 0.982   |
| Trimethylene trisulfide                            | 5.044                | 1.398   |
| 4-Fluoro-o-xylene                                  | 5.149                | 1.156   |
| Naphthalene                                        | 5.409                | 0.955   |
| Isobutylamine                                      | 6.572                | 1.358   |
| 1,1-cyclobutanedimethanamine, N,N’-dimethyl-       | 6.688                | 1.735   |
| 1,2,4,6-Tetrathiepane                             | 8.032                | 1.197   |
| Benzenecethanamine, N,N-bis(trimethylsily)-beta.,3,4-tris[(trimethylsily)oxy]- (R)-2-Dimethylaminopyrimidine | 8.238 | 0.551 |
| Caryophyllene                                      | 8.693                | 1.008   |
| 1,4-dimethyl-7-(prop-1-en-2-yl)-1,2,3,4,5,6,7,8-octahydroazulene | 8.814 | 1.412 |
| aromadendrene                                      | 7.476                | 7.476   |
| Guaiol                                             | 6.790                | 6.790   |
| Eicosane                                           | 10.459               | 1.560   |
| 2-Furanmethanol                                    | 13.442               | 49.133  |

4. Conclusions
The extraction process of garlic oil using solvent-free microwave gravity extraction could be established. The effects of extraction factors are mass of raw material, microwave power, extraction time, and material size affecting the extraction process to produce the garlic oil. From the results of the optimization, it was found that the interaction of mass of raw material and material size (AD) is only showed significant. The quadratic model was designed to obtain a goodness fitting between experimental data and predicted model. Based on experiment, the highest yield is 100 g, 450 W, 10 min and 0.5 cm. Besides that, the optimum condition predicted in 103.395 g, 464.281 W, 12.213 min and 0.676 cm to produce the highest yield that was suggested by response surface methodology. In addition, GC-MS result showed that garlic oil is contain in organosulfur compound using gravity technique. Therefore, from these studies providing an improved new technique of extraction process and optimizing of parameters involved by RSM that are the important factors for the industrial sectors.
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