

Optimization of Supercritical Carbon Dioxide Extraction of *Quercus infectoria* Oil

Liza Md Salleh^{a,b,*}, Stashia ELeaness Rosland Abel^b, Gholamreza Zahedi^c, Russly Abd Rahman^d, Hasmida Mohd Nasir^a, Syed Anuar Syed Faua'ad^{a,b}

^aCentre of Lipids Engineering & Applied Research (CLEAR), Ibnu Sina Institute for Scientific and Industrial Research, Universiti Teknologi Malaysia, 81310 UTM Johor Bahru, Johor, Malaysia

^bDepartment of Bioprocess Engineering, Faculty of Chemical Engineering, Universiti Teknologi Malaysia, 81310 UTM Johor Bahru, Johor, Malaysia

^cProcess System Engineering Centre (PROSPECT), Universiti Teknologi Malaysia, 81310 UTM Johor Bahru, Johor, Malaysia

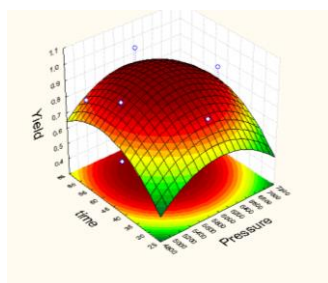
^dDepartment of Food and Process Engineering, Faculty of Engineering, Universiti Putra Malaysia, 43000 Serdang, Selangor, Malaysia

*Corresponding author: i.liza@cheme.utm.my

Article history

Received : 2 March 2015
Received in revised form :
24 April 2015
Accepted : 10 May 2015

Graphical abstract



Abstract

This current study focuses on the modelling and optimization of supercritical fluid extraction of *Quercus infectoria* galls oil. In this case, response surface methodology (RSM) and artificial neural network (ANN) were applied for the modelling and prediction of extraction yield of galls oil. A 17-run Box-Behnken Design (BBD) was employed to statistically optimize the process parameters of SC-CO₂ extraction of *Quercus infectoria* galls at a condition as follows: pressure (5000, 6000, 7000 Psi), temperature (40, 50, 60°C) and extraction time (30, 45, 60 min). The maximum yield of the extracted oil is 1.12 % and the optimum conditions are at an extraction pressure of 5574 Psi; extraction temperature of 75°C and extraction time of 54 min. Under the optimal conditions, the experimental results agree with the predicted values obtained through analysis of variance (ANOVA). This indicates a successful response surface methodology and highly satisfactory goodness of fit of the model used. The analysis of experimental design for process optimization results demonstrates that temperature and extraction time are the main parameters that influence the oil extraction of *Quercus infectoria*.

Keywords: *Quercus infectoria*(Manjakani); supercritical CO₂ extraction; optimization; response surface methodology (RSM); artificial neural network

© 2015 Penerbit UTM Press. All rights reserved.

1.0 INTRODUCTION

Quercus infectoria or also known as Manjakani (oak galls) is produced through the penetration by an insect, *Cynips Gallae* at the cortical parts of the tree. Manjakani in Malaysia is a small tree native of Greece and Asia Minor with four to six feet in height. The stems are crooked, shrubby looking with smooth and bright-green leaves borne on short petioles of 1 to 1.5 inches long. In the last century, a number of Asian people namely Chinese, Indians and Malays have used Manjakani in the ingredients of traditional medicine. According to previous researches, the best Manjakani can be found in Aleppo, Syria which is known as 'Mecca Manjakani' or 'Aleppo Manjakani'.

The usefulness of Manjakani is not limited to only in treating vaginal discharge, but also effective in curing sore throat, ulcer and skin problems. Manjakani contains elements of *astringent* which can help to remove bacteria that cause vaginal discharge and also in curing external tissue damage after childbirth process. Galls are mainly imported from Syria and Turkey, though some costly grades are brought in smaller quantity from other countries namely China and Japan. Galls are purely and effectively

astringent, scarcely stimulant. They may be used as an injection in bad leucorrhoea; in which cases they arrest putrefactive tendencies, and may be combined with suitable stimulants. The galls have been used to treat dysentery, internal hemorrhages, gonorrhoea, impetigo, tonsillitis, and menorrhagia [1]. Apparently, they also used as an antidote, febrifuge, ophthalmic, haemostatic agent, ointments and suppositories [2, 3]. Besides that, manjakani also can act as an anti-microbes and anti-inflammatory [4]. In addition, Manjakani could be used in all cases where astringents are indicated such as in chronic dysentery, diarrhoea, passive haemorrhages, and in cases of poisoning by strychnine and other vegetable alkaloids [5].

During the past several years, the increasing demand on high quality specification has required wide research on the improvement of control and optimization procedures for a multi scale process systems. Optimization is a process of determining the optimum values which provide maximum and minimum results of desired specifications. In 1991, Box and Wilson introduced a technique on optimization analysis, which is known as Response Surface Methodology (RSM). By using the design of experiment (DOE) regression techniques, the approximation

of the functional relationship between input variables of the coefficient and a real structural response variable of an approximate polynomial model (first, second and third order) can be estimated. In addition, RSM has been applied to determine the optimum conditions of factors that could achieve a maximum yield of production.

There are several approaches applied in the industry to effectively determine the optimum conditions of the process variables, including the pressure, temperature, extraction time, and size of the samples. Those strategies are classified as experimental design of statistical optimization. Apparently, the common strategies used in setting up robust extraction methods are using experimental designs. In the experimental designs, sufficient information on the interaction of the factorial design strategy is very significant where; numerous factors are varied during the designing process. For example, in the extraction of bioactive flavonoid compound from *Strobilanthes crispus* leaves [6], the experimental RSM engaged full factorial composite design involving three factors, namely; temperature, pressure and extraction time. In addition, three levels were applied in order to obtain a second order polynomial model for the lycopene maximization.

Zahedi and Azarpour applied artificial neural network (ANN) technology to simulate the supercritical fluid extraction of *Passiflora* seed oil [7]. They compared the use of ANN and RSM to optimize the extraction process. The results proved the estimation of oil yield using the network is better than the RSM model for three independent variables; pressure, temperature and extraction time. These depend on the equation of both models, ANN applied long and complicated model, whereas RSM only controls the process by quadratic polynomial which will neglect some of the terms regarding supercritical fluid extraction process.

Recently, RSM and ANN have been compared to investigate data fittings and estimation abilities, such as in curdlan production from *Paenibacillus polymyza* [8], optimizing *Bauhinia monandra* seed oil [9] and shea butter oil biodiesel production [10]. ANN was reported to represent the data effectively than RSM.

To date, no research has been reported on optimization of process parameters for supercritical carbon dioxide extraction of *Quercus infectoria*. Therefore, in this work, the response surface methodology (RSM) was performed using multiple regression techniques to find the optimum condition which can produce maximum yield extraction. In addition, a Box-Behnken statistical experimental design was employed to evaluate the effects of various parameters such as pressure, temperature and extraction time on the oil yield from *Quercus infectoria*.

2.0 MATERIALS AND METHODS

2.1 Materials

Quercus infectoria galls were obtained from local market (Kota Tinggi, Johor, Malaysia). The galls were dried in an oven at 45°C for 24 hours. The dried galls were then ground into powder with a grinder (Panasonic) and sieved into 200 µm of particle sizes. After that, the powder was sealed in a plastic container and stored at room temperature.

2.2 Chemicals

The solvent used for extraction was carbon dioxide liquid with a purity of 99.99% and was purchased from Malaysian Oxygen (MOX), Penang.

2.3 Supercritical Carbon Dioxide Extraction of Galls Oil

The laboratory assembles of supercritical CO₂ extraction apparatus is shown in Figure 1, consisting of a carbon dioxide cylinder, a chiller (BL-730, Yih Der Taiwan), a CO₂ syringe pump (ISCO, Model 100DX), a modifier syringe pump (ISCO, Model 100DX), an extraction chamber (ISCO, SFX 220), an extraction cartridge, controller (ISCO, SFX 200) and a restrictive temperature controller (ISCO).

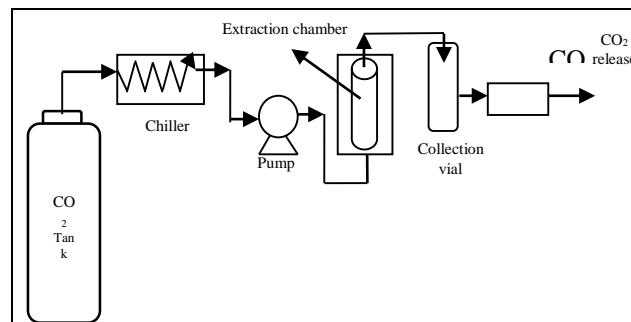


Figure 1 Diagram of supercritical carbon dioxide (SC-CO₂) extraction apparatus

For extraction, approximately two grams of ground plant material were loaded into the extraction chamber. The small amount of ground sample used in this experiment is to prevent any physical surge of the sample out of the extractor and to avoid prolonged extraction. After an initial air purge, liquefied carbon dioxide from the cylinder was passed through a chiller and pumped into the extractor chamber by using a high pressure pump. During the extraction process, the operating temperature, pressure and extraction time were set to the desired value, at the control panel of the extraction unit. When the desired pressure and temperature were reached, the extraction started by opening the valve between the pump and the sample cartridge to allow the CO₂ to flow through the sample. After each extraction, the extracted oil was collected in the glass vial and the yield of the oil was then measured gravimetrically. The extraction yield is expressed as the percent ratio of the mass of extracting oil to the mass of *Quercus infectoria* galls loaded into the extraction vessel, as follows:

$$\text{Oil yield (\%)} = \frac{\text{Mass of extracted oil}}{\text{Mass of dried material}} \times 100 \quad (1)$$

At this stage, the optimization process of SC-CO₂ extraction will be carried out by Box-Behnken Design (BBD) using Statistica 6.0 Software in order to determine the optimum value of (temperature, pressure, and extraction time).

2.3 Experimental Design

Response Surface Methodology (RSM) was applied to optimize the process parameters of supercritical carbon dioxide extraction oil yield. In this study, three independent variables were used; pressure (X_1), temperature (X_2) and extraction (X_3). The independent variables were transformed into a range between -1 and 1 for the appraisals of factors. The variables were coded according to the following equation:

$$Z_j = (X_i - X_0) / \Delta_j, \quad i=1, 2, 3 \quad (2)$$

Where Z_{ij} is the coded value of the independent variable; X_{i0} is the real value at the centre point; and Δ_{ij} is the step change in the variable X_i . The coded and uncoded levels of the independent variables used in the RSM design are listed in Table 1 below. Meanwhile, the dependent variable is the total oil yield of *Quercus infectoria* galls.

Table 1 Uncoded and coded levels of independent variables used in RSM design

Symbol	Independent variables	Coded levels		
		-1	0	1
Factor levels		-1	0	1
X_1	Pressure (Psi)	5000	6000	7000
X_2	Temperature (°C)	40	50	60
X_3	Time (Min)	30	45	60

A second-order polynomial regression model was used to express Y as a function of the independent variables as follows:

$$Y = \beta_0 + \sum \beta_i X_i + \sum \beta_{ii} X_i^2 + \sum \sum \beta_{ij} X_i X_j \quad (3)$$

Where Y represents the response variables, β_0 is a constant, β_i , β_{ii} and β_{ij} are the linear, quadratic and interactive coefficients, respectively. X_i and X_j are the levels of the independent variables. The analysis includes the Fisher's test (overall model significance), its associated probability P (F), correlation coefficient (R), and determination coefficient (R^2) which measures the goodness of fit of the regression model. Models were then used to plot 3D response surface and contour plots. The contour plots for three variables were superimposed to define the optimum combinations of the independent variables. The analysis of variance (ANOVA) was carried out to determine the significant differences between the independent variables at the confidence limit of 95% [11].

2.4 Artificial Neural Network

Artificial neural networks are the results of the scientists' interest in how the human brain can make decisions based on noisy data [12]. Because of their ability to find nonlinear and complex relationships, ANN application in various branches of science and also in industrial managements has been increasing [13]. Strictly speaking, ANNs do not belong to any specific branch of science due to its applicability in almost all science disciplines like image processing [14], document analysis [15], engineering tasks [16], financial modelling [17] and biomedical [18]. Neural networks generally can be divided into three major components as listed below [19]:

- The computational units
- The network architecture
- The learning algorithm to train the network

The computational units, which are known as "neuron", are fundamental units of the structure, which in fact are functions that change input signals to output signals.

Signals from other units enter each unit via weighted connections. Each input signal is multiplied by its associate weight and then the summation plus a special weight, which is called "bias" and its input signal is always 1, makes net input for

the function. Based on the function definition, this input will be changed to neuron outputs (Equation 4). Figure 2 illustrates a schematic of a neuron.

$$\text{Output} = f[\sum_{j=1}^n (w_{ij}x_j) + b] \quad (4)$$

Where x_j is a signal from unit j that enters to unit i , w_{ij} is its weight and b is the bias of this unit.

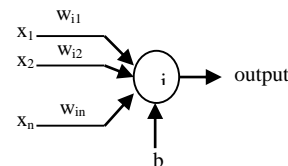


Figure 2 Schematic of a neuron

The network structure is made of layers of neurons which are connected together. First layer is called input layer that contains number of neurons equal the number of input data. The number of last layer neurons is the same as number of output data. This layer is called output layer. Intermediate layer(s) is/are known as hidden layer(s). The number of hidden layer(s) neurons may vary and are depended on the designer decision.

There is a lot of styles for connecting neurons in the network. However, they are divided into two main categories: feed forward and feedback structures. In the feed forward networks, information signals always propagate towards the forward direction, but in the feedback networks, the final outputs are again fed back at the input [16]. Figure 3 illustrates these two types of structures schematically.

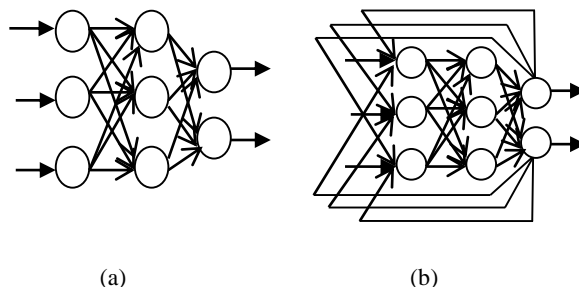


Figure 3 Schematic of (a) feed forward and (b) feedback structure for 3 inputs/2 outputs network with 3 neurons in hidden layer

After determining the type, number and structure of neurons of a network, it is necessary to find out the weights of connection between the neurons. These parameters should be selected so that the network can produce outputs closed to target data. In this case; in the first step, all parameters will be started in random, and then the outputs of the network will be compared to the target values. If the error is not small enough, the parameters will be changed with a mathematical algorithm. This procedure will be repeated many times until the desired error is obtained and the network will be robust enough to produce accurate outputs.

3.0 RESULTS AND DISCUSSION

3.1 Experimental Data

A 17 run Box-Behnken Design with three factors, three levels and five replicates at the centre point was used to fit a second-order response surface in order to optimize the extraction conditions. The replications of the centre point were carried out in order to measure the process stability and inherent variability of the response. The design parameters of BBD are given in Table 2,

along with the experimental and predicted values of the response. In this study, three parameters, namely; pressure, temperature and extraction time were used as the input process variables in the supercritical carbon dioxide (SC-CO₂) extraction. Based on the result, it is shown that the highest yield of oil is 1.00 % and was obtained after 60 min extraction at 60°C and 6000 Psi. Overall observation of the results shows that the oil yield generally increases with the linear effects of temperature, extraction time and pressure of the SC-CO₂ extraction process.

Table 2 Experimental and predicted data for the oil yields obtained from Box-Behnken experimental design

Run	P (Psi)	T (°C)	t (Min)	Yield (%) (Observed)	Yield (%) (Predicted)	Standard Deviation
1	5000	40	45	0.56	0.63	0.05
2	7000	40	45	0.69	0.65	0.03
3	5000	60	45	0.93	0.89	0.03
4	7000	60	45	0.89	0.90	0.01
5	5000	50	30	0.61	0.62	0.01
6	7000	50	30	0.64	0.63	0.01
7	5000	50	60	0.80	0.76	0.03
8	7000	50	60	0.74	0.77	0.02
9	6000	40	30	0.63	0.60	0.02
10	6000	60	30	0.84	0.86	0.01
11	6000	40	60	0.73	0.74	0.01
12	6000	60	60	1.00	1.00	0.00
13	6000	50	45	0.93	0.93	0.00
14	6000	50	45	0.93	0.93	0.00
15	6000	50	45	0.93	0.93	0.00
16	6000	50	45	0.94	0.93	0.01
17	6000	50	45	0.93	0.93	0.00

3.2 Model Fitting and Statistical Analysis

3.2.1 Regression Model

The multiple regression coefficients, obtained by employing a least squares technique to predict a second-order polynomial model for the yield, are summarized in Table 3. The significance of each coefficient was checked using the *t*-test and *P*-value (Table 3). The *P*-value was used to check the significance of each coefficient and also to indicate the interaction strength between each independent variable. For the oil yield, examination of these coefficients with the *t*-test indicated that linear terms (pressure, temperature and extraction time), quadratic terms (pressure and extraction time) are highly significant ($p < 0.01$). Meanwhile, there is significant interaction between the pressure and extraction time, also the quadratic terms of temperature ($0.01 < p < 0.05$) for the oil yield. In addition, there is no significant interaction between temperature and extraction time ($p > 0.05$) within the experimental range. Therefore, these results suggest that the linear, quadratic and (or) interaction effects of the independent variables may become the main factors affecting the extraction of oil yield. The developed regression model for the relationship between oil yield (*Y*) and the coded values of independent variables of pressure (*X*₁), temperature (*X*₂) and extraction time (*X*₃) and their interaction is shown in the following equation:

$$Y = -7.9205 + 0.0019X_1 + 0.0651X_2 + 0.0490X_3 - 0.0003X_2^2 - 0.0005X_3^2 + 0.0001X_2X_3 \quad (5)$$

To fit the response function and experimental data, the linearity and quadratic effect of the independent variables, and also the interactions on the response variables were evaluated by analysis of variance (ANOVA). The fitness and adequacy of the model was judged by the coefficient of determination (*R*²), which was defined as the ratio of the explained variation to the total variation, used as a measure of the degree of fitness. The closer the *R*² value to the unity, the better the empirical model fits the actual data. The ANOVA results of the model are shown in Table 4 indicating a good model performance with an *R*² value of 0.991 and adjusted *R*² of 0.979 among linear, quadratic, cross-product and total model. This suggests that the predicted second order polynomial models define the real behaviour of the system well. From the model, the optimum condition for oil yield is at 5574 Psi, 75 °C and 54 Min with the maximum yield at 1.12%.

Table 3 Regression coefficients of the second order polynomial model for *Quercus Infectoria* oil yield extract

Model parameters	Regression Coefficients	Standard Error	t	Significance level (P value)
Intercept				
X ₀	-7.9205	0.5847	-13.546	0.000
Linear				
X ₁ , Pressure	0.0019	0.0001	13.936	0.000
X ₂ , Temperature	0.0651	0.0123	5.304	0.001
X ₃ , Extraction time	0.0490	0.0068	7.250	0.000
Quadratic				
X ₁ ²	-0.0000	0.0000	-13.221	0.000
X ₂ ²	-0.0003	0.0001	-3.070	0.019
X ₃ ²	-0.0005	0.0000	-10.003	0.000
Interaction				
X ₁ X ₂	-0.0000	0.0000	-4.103	0.004
X ₁ X ₃	-0.0000	0.0000	-2.172	0.041
X ₂ X ₃	0.0001	0.0001	1.448	0.178

Table 4 Analysis of Variance (ANOVA) of the response surface model of *Quercus infectoria* oil yield

Source	Sum of Squares	Degree of freedom	Mean sum of squares	F-value	F _{0.05}
Regression	0.31875	9	0.0354	88.50	> 2.54
Residual	0.00301	7	0.0004		
Total	0.32175	16			
R ²	0.991				
Adjusted R ²	0.979				

3.3 Response Surface Analysis

The 3-dimensional plots of the response surfaces are presented in Figure 4-6. Each surface plot represents an infinite number of combinations of two test factors while the third factor is fixed at a constant level. Figure 4 shows the effect of temperature, pressure and their reciprocal interaction on extraction yield, when extraction time is fixed at 45 min. By referring to Figure 4, there is a linear increase in the oil yield with an increase in extraction temperature from 40 to 60°C. This is most likely due to the increase of mass transfer speed. Meanwhile, a greater increase in extraction yield resulted when the pressure is increased in the range from 5000 to 6000 Psi, and then the yield started to go down after 6000 Psi.

Figure 5 is the response surface showing the effect of pressure and extraction time on the oil yield at a constant temperature of 50°C. Pressure has a positive linear effect on the oil yield at low pressure levels. This is most likely due to the improvement of oil solubility resulted from the increase in carbon dioxide density with the rise of pressure. However, once the pressure reaches a high level (about 6000 Psi), the oil yield slightly decreased. This is probably due to the increase of repulsive solute-solvent interactions resulting from highly

compressed CO₂ at high pressure levels. As presented in Figure 5, an increase in oil yield is observed by increasing the extraction time in an early stage of extraction (about 30 to 45 min). However, further increase in the extraction time up to 60 min resulted in decreasing oil yield extraction. For very long extraction time, however, the negative quadratic effect also became significant. This was reflected in the plateau of the oil yield for extraction time over 60 min [20].

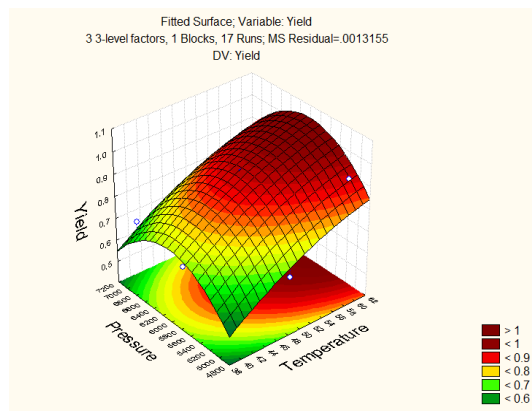
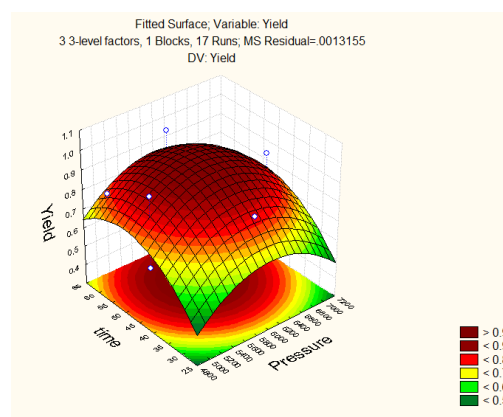
**Figure 4** 3D response surface plots showing effects of pressure and temperature on the oil yield and their interaction. The extraction time is constant at 45 min.**Figure 5** 3D response surface plots showing effects of pressure and extraction time on the oil yield and their interaction. The temperature is constant at 50°C.

Figure 6 illustrates the effect of temperature and extraction time on the oil yield extraction at a constant pressure of 6000 Psi. How extraction temperature affect the oil yield is described in Figure 5. As shown in Figure 6, temperature has a positive linear impression on the oil extraction yield, while further increase in the temperature resulted in little change in the yield of oil extraction. Meanwhile, for extraction time, it shows different results compared to the extraction temperature. The oil yield increases rapidly with the extraction time and reaches the maximum value, followed by a decline with further increase. As described in Figure 5, the negative effect on oil yield extraction was reflected in the plateau of the oil yield. In general, from the surface plots (Figure 4-6), the optimum extraction process parameters for the oil yield extraction are; pressure of 5574 Psi, temperature of 75 °C and extraction time of 54 min. Under these conditions, the oil extraction yield is 1.12 %.

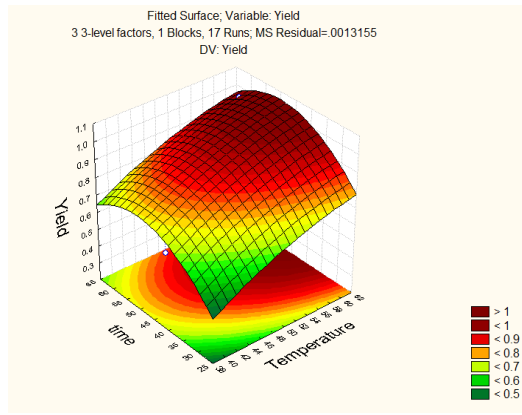


Figure 6 3D response surface plots showing effects of temperature and extraction time on the oil yield and their interaction. The pressure is constant at 6000 Psi.

3.4 ANN Structure

The LM training algorithm was employed for modelling of the oil extraction yield of the galls. There is no any general and precise method to achieve the optimum number of hidden layers of the neurons and it is obtained by trial and error. The optimum number of hidden layer neurons was determined to be 50 for this network. Table 5 describes the parameters of the used network.

Table 5 Parameters of the used network

Training Algorithm	Levenberg-Marquardt
Network	Feedforward
Hidden Layer Transfer Function	tansig
Output Layer Transfer Function	Purelin
Number of Hidden Layer Neurons	24
Number of Output Layer Neurons	1
Performance Function	MSE
Divide Function	Dividrand
Best Performance	6E-06
mu	1e-07
Gradient	2.73e-10

Some statistical methods were used for comparison. The criterion for the comparison in this work is root-mean-squared-error (RMSE) between the net output and the training data. MSE is defined as:

$$RMSE = \sqrt{\frac{\sum(X_{exp} - X_{sim})^2}{n}} \tag{6}$$

Where X_{exp} is the target value, X_{sim} is the output value, and n is the number of experimental data. The RMSE value for the defined system was calculated giving 0.00217 which reveals a very good precision of the proposed network.

Figure 7 gives information on the oil yield extraction by comparing the experimental data against the ANN model predicted values for training data by assuming the oil yield obtained is equal of 100 g basis. Figure 8 implies the experimental data versus the simulated ones derived by ANN model for testing data, which have not been applied for the training data, for the extraction yield. The figures show that the data obtained from the

model are in a very good agreement with the laboratory results. Whereas, the predictions that match measured values should fall on the diagonal line. Almost all data fall close to this line, which confirms the accuracy of the ANN model.

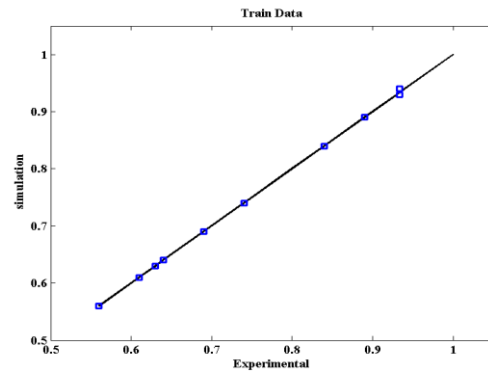


Figure 7 Scatter plot of experimental data (the oil extraction yield) vs. predicted values by ANN model for train data

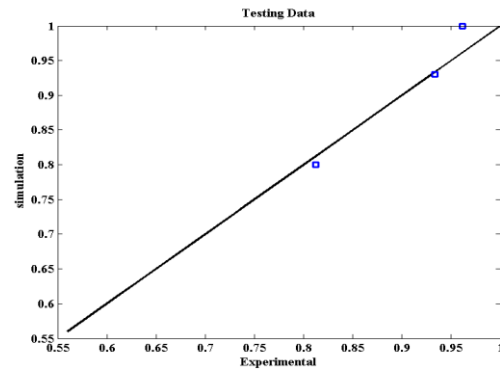


Figure 8 Scatter plot of experimental data (the oil extraction yield) vs. predicted values by ANN model for testing data

Figure 9 and 10 indicate the influence of temperature and pressure on the oil extraction yield at definite extraction time. At low temperature, the oil yield improves with the growth of temperature, most probably due to the boosted mass transfer rate. Whereas, at higher temperature the oil extraction yield declines with the rise of temperature, most possibly due to the lowered density of carbon dioxide. Pressure expresses a positive linear effect on the oil extraction yield at low pressure levels. This is most possibly owing to the progress of oil solubility originated from the increased carbon dioxide density with the increase of pressure. At higher pressures, because of the increased unpleasant solute–solvent interactions originated from the noticeably pressurized carbon dioxide, the negative effect of pressure on the oil extraction yield also becomes significant.

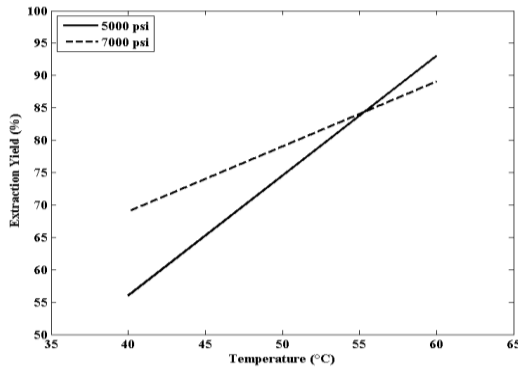


Figure 9 Effect of temperature on the yield of extraction at constant extraction time (30 min)

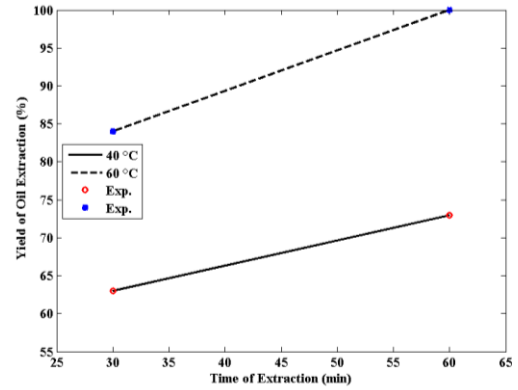


Figure 12 The influence of Temperature rise on the yield of oil extraction at constant pressure, 6000 psi

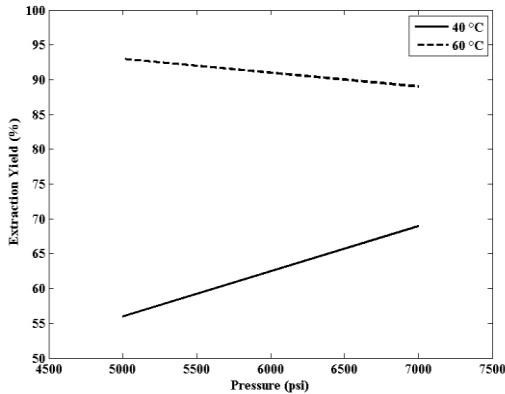


Figure 10 Effect of pressure on the yield of extraction at constant extraction time (45 min)

Figure 11 presents the experimental versus simulated data derived by ANN model. The figure shows that the data obtained from the model are in very good agreement with the laboratory results. Meanwhile, Figure 12 displays the effect of temperature on the oil yield extraction over time. It reflects rising oil yield extraction with increasing temperature at constant pressure during the time of extraction revealing the decline of oil yield extraction rate due to density reduction of carbon dioxide.

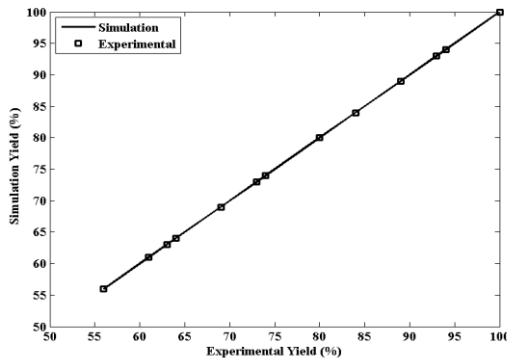


Figure 11 Comparison between the results of simulation with experimental data

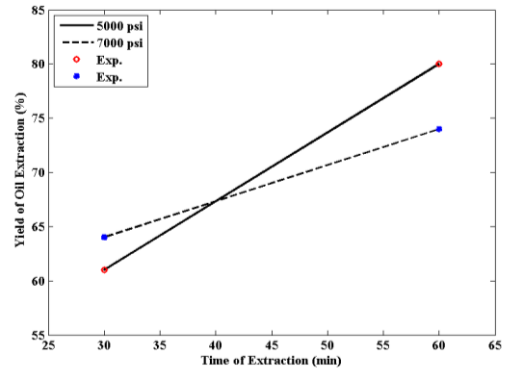


Figure 13 The influence of pressure rise on the yield of oil extraction at constant temperature, 50 °C

Figure 13 presents the influence of pressure on the oil yield at a constant temperature along the time of extraction, which implies a positive effect of pressure on the yield at lower pressure values. As can be seen the rate of oil yield extraction is reduced by intensifying the pressure and the yield massively declines after a definite period of time owing to the high interaction between solvent and solute. The last two graphs provide us with more historical data, which are costly and time consuming if experimental are needed. Table 6 represented the summary for comparison of relative error between ANN prediction and RSM.

Table 6 Comparison of the ANN prediction with RSM for training data

Temperature (°C)	Pressure (Psi)	Extraction time (Min)	Oil yield (exp.)	Predicted value (ANN)	Predicted value (RSM)	Relative error (%) (ANN)	Relative error (%) (RSM)
40	5000	45	0.56	0.56	0.63	0.000000	-12.50000
40	7000	45	0.69	0.69	0.65	0.000000	5.79710
60	7000	45	0.89	0.89	0.90	0.000000	-1.12360
50	7000	30	0.64	0.64	0.63	0.000000	1.56250
50	5000	60	0.80	0.80	0.76	0.000000	5.00000
50	7000	60	0.74	0.74	0.77	0.000000	-4.05405
40	6000	30	0.63	0.63	0.60	0.000000	4.76190
60	6000	30	0.84	0.84	0.86	0.000000	-2.38095
50	6000	45	0.93	0.932	0.93	-0.21505	0.00000
50	6000	45	0.93	0.932	0.93	-0.21505	0.00000
50	6000	45	0.93	0.932	0.93	-0.21505	0.00000

4.0 CONCLUSION

Response surface methodology was successfully applied for optimization of the *Quercus infectoria* oil using supercritical carbon dioxide (SC-CO₂) extraction. It is found that the independent parameters of pressure, temperature and extraction time significantly influence the oil yield extraction. The graphical optimization method was employed in order to find the best extraction condition and it predicted that the optimum conditions to be at pressure of 5574 Psi; temperature of 75 °C and extraction time of 54 min. Under this condition, about 1.12 % of oil was extracted from the *Quercus infectoria* galls. In addition, the comparison between ANN and RSM models has been implemented by RMSE and relative error methods. For instance, RMSE value for ANN model has been enumerated to be 0.00217 which reveals a very good precision of the proposed network. From the results it can be concluded that the ANN model by using MLP neural network architectures is the best forestimation of the values of the targets in comparison with RSM model by using STATISTICA SOFTWARE because in RSM model, quadratic polynomial can be used as a maximum effect to approximate the targets (where the other terms are neglected), whereas in ANN model very long and complicated algorithms are used for the same aim. These algorithms can give the fit approximation without neglecting any term.

Acknowledgement

This work was financially supported by Research Grant of Malaysia Ministry of Higher Education (MOHE).

References

- Warrier, P.K., Nambiar, V.P.K., Ramankutty, C. 1995. *Indian Medicinal Plants*. 4th Edition. Orient Longman, Madras, India.
- Perumal, S.R., Gopalakrishnakone, P., Sarimuthi, M., Ignacimuthu, S. 2006. Wound Healing Potential of *Tragia Involucrate* Extract in Rats. *Fitothérapie*. 77: 300–302.
- Kudi, A.A., Ngbede, J.E. 2006. In vitro Antibacterial Activity of Aqueous Garlic (*Allium sativum* Linn.) Extract on Isolates from Surface Wounds. *Journal of Food Agriculture and Environment*. 4: 15–16.
- Ikram, M., Nowshad, F. 1977. Constituents of *Quercus infectoria*. *Planta Med.* 31:286–287.
- Dar, M. S., Ikram, M., Fakouhi T. 1976. Pharmacology of *Quercus infectoria*. *J Pharm Sci.* 65: 1791–1794.
- Liza, M. S., Abdul Rahman, R., Mandana, B., Jinap, S., Rahmat, A., Zaidul, I.S.M., Hamid, A. 2010. Supercritical Carbon Dioxide Extraction Of Bioactive Flavonoid from *Strobilanthes crispus* (Pecah Kaca). *Food and Bioproducts Processing Journal*. 88: 319–326.
- Zahedi, G., Azarpour, A. 2011. Optimization of Supercritical Carbon Dioxide Extraction of *Passiflora* Seed Oil. *Journal of Supercritical Fluids*. 58: 40–48.
- Rafiqh, S.M., Yazdi, A.V., Vossoughi, M., Safekordi, A.A., Ardjmand, M. 2014. Optimization of Culture Medium And Modeling Of Curdlan Production from *Paenibacillus polymyxa* by RSM and ANN. *International Journal of Biological Macromolecules*. 70: 463–473.
- Akintunde, A.M., Ajala, S.O., Betiku, E. 2015. Optimization of *Bauhinia monandra* Seed Oil Extraction Via Artificial Neural Network And Response Surface Methodology: A Potential Biofuel Candidate. *Industrial Crops and Products*. 67: 387–394.
- Betiku, E., Okunolawo, S.S., Ajala, S.O., Odelele, O. S. 2015. Performance Evaluation of Artificial Neural Network Coupled with Generic Algorithm and Response Surface Methodology in Modeling and Optimization of Biodiesel Production Process Parameters from Shea Tree (*Vitellaria paradoxa*) Nut Butter. *Renewable Energy*. 76: 408–417.
- Montgomery, D.C. 2001. *Design and Analysis of Experiments*. 5th Edition. John Wiley and Sons Inc.
- Minns, A. W., Hall, M. J. 1996. Artificial Neural Networks As Rainfall-Runoff Models. *Hydrolysis Science Journal*. 41: 399–417.
- Aggarwal, K. K., Singh, Y., Chandra, P., Puri, M. 2005. Sensitivity Analysis of Fuzzy and Neural Network Models. *ACM SIGSOFT Software Engineering Notes*. 30: 1–4.
- Durantoni, M. 1996. Image Processing By Neural Networks. *IEEE Micro*. 16: 12–19.
- Marinai, S., Gori, M., Soda, G. 2005. Artificial Neural Networks For Document Analysis And Recognition. *IEEE Trans. on Pattern Analysis and Machine Intelligence*. 27: 23–35.
- Jin, X., Cheu, R. L., Srinivasan, D. 2002. Development and Adaptation of Constructive Probabilistic Neural Network in Freeway Incident Detection. *Transportation Research Part C*. 10: 121–147.
- Abu-Mostafa, Y. S. 2001. Financial Model Calibration Using Consistency Hints. *IEEE Transaction on Neural Networks*. 12: 791–808.
- Nazeran, H., Behbehani, K. 2000. Neural Networks in Processing and Analysis of Biomedical Signals. In: *Nonlinear Biomedical Signal Processing-Volume1: Fuzzy Logic, Neural Networks And New Algorithms*. Akay, M. (Ed.). New York: IEEE Press.
- Kamaruzzaman, J., Begg, R. K., Sarker, R. A. 2006. Artificial Neural Networks In Finance And Manufacturing. *IDEA Group Publishing*. 2–27.
- Liu, S., Yang, F., Zhang, C., Ji, H., Hong, P., Deng, C. 2009. Optimization of Process Parameters for Supercritical Carbon Dioxide Extraction of *Passiflora* Seed Oil by Response Surface Methodology. *The Journal of Supercritical Fluids*. 48: 9–14.