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Optimization of Ultrasound-Assisted Oil Extraction from Peony Seeds Based on Operational Research

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Research Article

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ABSTRACT

Operational research methodology was employed to optimize the ultrasonic-assisted extraction of oil from peony seeds. Five variables, namely the particle size (mesh), solvent: raw material ratio (ml/g), ultrasonic power (W), extraction time (min), and extraction temperature (°C) were investigated. Regression equations were developed, and nonlinear programming model was derived with an operation research theory. The ANOVA analysis indicated that all the quantities determined had significant quadratic and linear effects on oil yield (Y), contents of A-Linolenic Acid (ALA), linolein acid (LA) and oleic acid (OA), and energy consumption (Q). Optimum conditions for the extraction of oil were found to be: particle size of 62 mesh, solvent: raw material ratio of 14 ml/g, ultrasonic power of 323 W, extraction time of 45 min, and extraction temperature of 44°C. The optimal predicted Y, ALA, LA, OA, and Q for peony seed oil of 28.21%, 122.11 mg/g, 65.22 mg/g, 62.53 mg/g, and 2,394.99 KJ, respectively. While the experimental values were in agreement with the predicted values

INTRODUCTION

Tree peony (*Paeonia* section *Moutan* DC.) is a perennial deciduous shrub with excellent ornamental and medicinal values. It is indigenous to China and possesses a history of more than 2,000 years [1]. Tree peony seeds, which are a by-product of its flower, have long been neglected, yet recently have been used as a resource for woody oil [2]. Oilseed peony refers to the tree peony that produces considerable seeds used as an edible oil resource [3].

The unsaturated fatty acids (UFAs) content of edible oil is an important guideline in evaluating its nutritional value [4]. Tree peony seed oil is notable for its high UFAs content (>90%) [3,5]. Higher α-linolenic acid content (ALA, ± 43.18%) compared to other oleaginous seed oils, e.g., peanut oil: 0.4%, olive oil: 0.7%, rap oil: 8.4%, soybean oil: 6.7%, and tea oil: 1% has been reported in tree peony seed oil [3]. It is generally known that n-3 FAs are essential dietary nutrients that cannot be independently synthesized by the human body and has been linked to the prevention of various diseases such as cancer, cardiovascular, inflammatory, and autoimmune diseases [6-8]. Two other important UFAs in peony seed oil are linoleic acid (LA) and oleic acid (OA), which account for about 26% and 21% of its total oil content, respectively. Both of them play important biological roles in promoting human health by enhancing immune responses or reducing the risk for cardiovascular diseases [9,10].

To date, several oil extraction techniques in peony seed have been reported, which include pressing, microwave-assisted or ultrasound-assisted extraction, and supercritical CO_2 extraction [5,11]. Ultrasound-assisted extraction is an interesting process to obtain high valuable compounds. The main benefits will be a more effective extraction, thus saving energy, and also the use of moderate temperatures, which is beneficial for heat-sensitive compounds. In the last few years, ultrasound technology has been considered as a valuable tool in food engineering processes, and this field of research has become a very active one [11-14]. Previous studies in this area have mainly focused on the effect of factors on oil yield (Y). However, our understanding of how ultrasonication affects composition content is limited.

The UFAs in peony seed oil are prone to oxidative rancidity, and the degree of oxidation is affected by its molecular structure, temperature and moisture content [15,16]. The reduction in ALA, LA, and OA in walnut oil varied with different drying methods [17,18]. Zhou studied the influence of different heat treatments on fatty acid composition and content in betel nut and found that

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UFAs contents decreased and saturated fatty acid contents increased as temperature and heating time increased. Moreover, energy consumption is a very important factor that should be considered in reducing environmental impact and in maximizing the profitability of operations [19,20].

Although it is well known that the Y increases with an increase in solvent: raw material ratio, ultrasonic power, extraction time and temperature, it is still necessary to further determine the relationship between these parameters and UFAs content in peony seed oil, as well as energy consumption (Q) for unit mass (g), to obtain an optimal manufacturing process. In an attempt to increase ALA content and reduce energy consumption during oil extraction, the manufacturing parameters, particle size, solvent: raw material ratio, ultrasonic power, extraction time and temperature were investigated as functions of Y, ALA, LA, OA, and Q. Using nonlinear regression and the operation research theory, we developed models to estimate the optimal properties of peony seed oil and energy consumption based on specific practical production constraints.

MATERIALS AND METHODS

The *P. ostii* fruits were collected from Shuangquan town of Changqing District (36.35°N, 116.72°E; altitude: 111 m), Jinan, Shandong Province, China, in August 2015. The seeds were dried immediately after being removed from fruits in a shady, well-aired place for one month, then packed in paper bags and stored in a dark and dry place at room temperature. Moisture content, determined by drying at 105°C to constant mass, was around 6%.

The seeds were ground by using an electrical mill equipped with a fast rotating knife (1,500 rpm; 1 min). The ground seeds were sieved through a set of standard screens and classified into five particle size classes (40-80 mesh). Other extraction factors to be tested were solvent: raw material ratios ranging from 5 to 15, ultrasonic power ranging from 100 to 400 W, extraction time ranging from 20 to 60 min, and extraction temperature ranging from 20 to 60°C **(Table 1)**. The time for reaching a designated temperature in an ultrasonic cleaner and rotary evaporation was examined. The Q of extraction was calculated based on time and power consumption.

Table 1. Ultrasound-assisted extraction process for peony seeds.

Group	Particle size (mesh)	Solvent: raw material ratio (ml: g)	Ultrasonic power (W)	Extraction time (min)	Extraction temperature (°C)
1	40, 50, 60, 70, 80	10	300	30	30
2	60	5, 7.5, 10, 12.5, 15	300	30	30
3	60	10	100, 250, 300, 350, 400	30	30
4	60	10	300	20, 30, 40, 50, 60	30
5	60	10	300	30	20, 30, 40, 50, 60

In the first experiment, to investigate the influence of particle size on Y, ALA, LA, OA, and Q, a solvent: raw material ratio of 10 was preset, and the ultrasonic power, time, and temperature were fixed at 300 W, 30 min, and 30°C, respectively. In the second experiment, to investigate the influence of the solvent: raw material ratio on Y, ALA, LA, OA, and Q, the particle size used was 60 mesh, and the ultrasonic power, time, temperature was 300 W, 30 min, and 30°C, respectively. In the third experiment, to investigate the influence of ultrasonic power on Y, ALA, LA, OA, and Q, the particle size was preset to 60 mesh, and the solvent: raw material ratio, extraction time, and temperature were fixed at 10, 30 min, and 30°C, respectively. In the fourth experiment, to investigate the influence of extraction time on Y, ALA, LA, OA, and Q, the particle size and solvent: raw material ratio were preset to 60 mesh and 10, respectively, the ultrasonic power and temperature were fixed at 300 W and 30°C, respectively. In the fifth experiment, to determine the influence of extraction temperature on Y, ALA, LA, OA, and Q, the particle size and solvent: raw material ratio were preset to 60 mesh and 10, the ultrasonic power and time were fixed at 300 W and 30 min, respectively.

Chemicals and Standards

Five fatty acid methyl ester (FAME) standards, namely, methyl heptadecanoate (C17:0), methyl α-linolenic acid (C18:3, 12c, 15c, ALA), methyl linoleate (C18:2, 12c, LA), and methyl oleate (C18:1, OA), and Supelco37-component FAME Mix (C4-C24 unsaturates) were used in the present study. The FAMEs were named using the formula C*x*:y*n*c, where "*x*" is the number of carbon atoms, "*y*" is the number of double bonds, and "n" is the position (relative to carboxyl end) of the double bonds. Standard stock solutions were prepared in *n*-hexane at an appropriate concentration and then diluted to the desired range of concentrations. Among these, methyl heptadecanoate was used as the internal standard (IS). All standards and stock solutions were kept in the dark at 4˚C, while ALA was kept at -20°C.

Methanol and *n*-hexane were of chromatographic grade and purchased from Alltech Scientific (Beijing, China). Other analytical grade chemicals containing concentrated sulfuric acid were ordered from Tianjin Kermel Chemical Reagent Co., Ltd. (Tianjin, China). High performance liquid chromatography (HPLC)-grade water was obtained from a Milli-Q System (Millipore, Billerica, MA, USA).

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Lipid Extraction

Oil in tree peony seeds were extracted according to procedures described in quince seeds and flax seeds, with minor modifications [21,22]. The peony seed powder and the predetermined volume of *n*-hexane were placed in three 100-mL Erlenmeyer flasks, comprising three replicate samples for each test. Ultrasound- assisted extraction was conducted in an ultrasonic cleaner. After extraction, the samples were transferred to the centrifuge tube, and centrifuged at 3,000 rpm and 4°C for 15 min in a HITACHI RX series centrifuge. After centrifugation, the supernatant was collected, and the *n*-hexane was removed from the samples by evaporation under vacuum at 40°C on a rotary evaporator. At the end of the extraction cycle, the samples were dried with a nitrogen blowing instrument and weighed.

The Y of peony seed was calculated by using the following formula:

$Y=(M_1 - M_2)/M \times 100\%$;

Where M is the weight of peony seed powder (g); M_1 is the total weight of the receiving bottle and sample (oil) (g); and M_2 is the weight of the receiving bottle (g).

Q for unit mass (g) comprised three parts, namely, ultrasonic working energy consumption, heating energy consumption, and rotary evaporation energy consumption.

Fatty Acid Methylation

The sulfuric acid-methanol method with higher methyl esterification efficiency was selected and used in the present study because acetyl chloride is highly irritating and toxic [23]. The concentrated seed lipids earlier described were re-dissolved in 1.0 mL of a methanol solution containing 5% concentrated sulfuric acid in a tightly capped vial with N_2-headspace. The vial was vortexed for 1 min and placed in a 90°C water bath for 1 h to induce derivatization. Afterwards, the samples were removed and allowed to cool down to room temperature. Then, 1.0 mL of deionized water was added to terminate the derivatization reaction. The FAMEs were subsequently extracted with 1.5 mL of *n*-hexane, centrifuged at 3,000 rpm at 4°C for 15 min, the supernatant was collected, and the samples were dried with a nitrogen blowing instrument. The concentrated solution was re-dissolved in 1.5 mL *n*-hexane volume, transferred to 2 mL vials in aliquots of 1.0 mL, and covered with a plastic cap with a polytetrafluoroethylene septum. In addition, 40 µL of methyl heptadecanoate (4.0 mg/mL in *n*-hexane) was employed as the internal standard.

GC-MS Instrument

FAMEs analysis was conducted by using a gas chromatograph-mass spectrometer (GC6890/MS5973, Agilent) equipped with a 7683 autosampler tray module and a 7683 autoinjector module (Agilent). The column was an (5%-diphenyl) dimethyl polysiloxane packed capillary column (HP-5; 30 m × 0.25 mm i.d., 0.25-µm film thickness; Agilent). Operating conditions were as follows: ultrahigh-purity nitrogen was used as the carrier gas at a flow rate of 1.0 mL/min, and analyses were performed in a constant flow mode, and a split liner with glass wool was installed in the injector.

The temperature of the transfer line, ion source, and quadrupole were 280, 230 and 150 ˚C, respectively. The injector temperature was set at 250°C for split injection at a split ratio of 20:1. The injection volume was 1 µL. The initial oven temperature was maintained at 100°C for 2 min and then increased by 15°C/min to 230°C and kept isothermal for 5 min. The ionization potential of the mass-selective detector was 70 eV and the scan range was 30-450 amu. Analyses were performed in triplicate. Identification of compounds were achieved by a mass spectra database search (NIST05 Library) and co-eluted with corresponding standards.

Qualitative and Quantitative Analysis of Fatty Acids

FAs were initially identified through a mass spectra database search (NIST05 Library). Three major unsaturated fatty acids were analyzed by reference to corresponding authentic standards. Other minor components were confirmed by comparing retention time and MS spectra with the 37-component FAME Mix (C4-C24 unsaturates). A standard curve with an internal standard was used as the quantitative approach to construct three calibration plots of the analyte/internal standard peak-area ratio versus the standard concentration, as determined by the least squares method. The three FAMEs in each sample were quantified in absolute terms by linear regression of their corresponding standard. The FAMEs were expressed as milligrams per gram dry weight of the sample. All samples were analyzed in triplicate.

Statistical Analysis

The mean values of each sample were obtained from three replicates and used for further analysis. Variance analysis was conducted using SPSS 18.0. To figure out the relationships among the five parameters (particle size, solvent: raw material ratio, ultrasonic power, extraction time and temperature) and Y, ALA, LA, OA, and Q, multiple variable nonlinear and linear regressions were used. Once the regression equation was built from experimental data, it could be directly applied to predict the properties of the peony seed oil by mill personnel.

Operations research is one of the popular managerial decision tools used by industry and complex organizational systems. Optimization is a branch of operation research, which uses mathematical techniques such as linear and nonlinear programming

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to derive values for system variables that will optimize performance. Nonlinear programming was used in the present study, because the regression equations obtained were nonlinear (Equations (1)-(5)). In order to solve the nonlinear and constrained programming problem, the penalty function approach was used. This method attempts to approximate a constrained optimization problem with an unconstrained one and then applies standard search techniques to obtain the optimal solution. The approximation is accomplished using penalty methods by adding a term to the objective function that prescribes a high cost for violation of the constraints.

RESULTS AND DISCUSSION

Effect of Particle Size

Particle size had a significant effect on Y. In theory, Y increases with a decrease in particle size because of shorter diffusion paths [24,25]. However, as shown in **Figure 1**, the highest Y was obtained from the intermediate class of the peony seed powder, which is consistent with the findings in previous studies [5,26-29]. The reasons for this may be as follows: smaller particles are more prone to compaction, which in turn increases mass transfer resistance and causes non-uniformity in the extraction; smaller particles possess liquid surface tension that prevents solvent immersion and hinders oil extraction; and oil could be lost during peony seed grinding when particle size is too small.

Three UFAs were higher at 60 mesh, while relatively low at a particle size of 80 mesh **(Figure 1)**. UFAs of smaller particle size are prone to oxidation due to its larger surface area. Because solvent: raw material ratio, ultrasonic power, extraction time and temperature were all the same, the total energy consumption was equal. A lower Q of 1,646.41 KJ was obtained at a particle size of 60 mesh because Y was higher at the particle size of 60 mesh **(Figure 1)**.

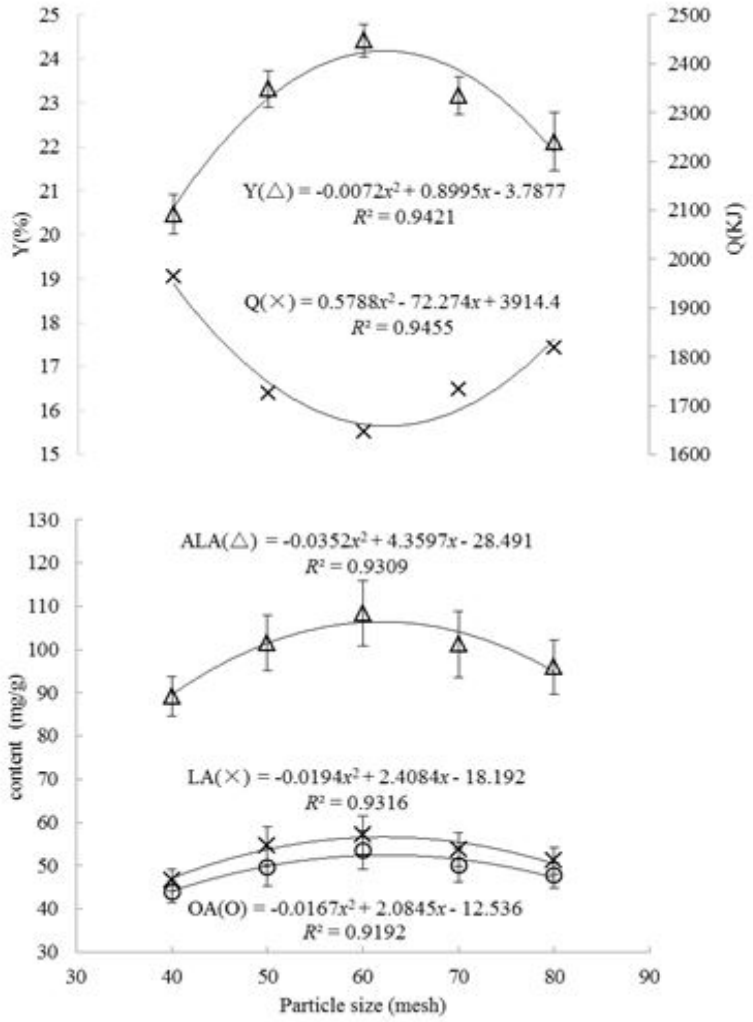

Figure 1. Oil properties (Y, ALA, LA, and OA) and energy consumption (Q) as functions of particle size.

Effect of the Solvent: Raw Material Ratio

Increasing solvent: raw material ratio significantly ($p<0.05$) increased Y and three UFAs. A marked increase was shown in Y (4.73%) when the solvent: raw material ratio ranged from 5 to 12.5, whereas Y showed non-significant increase (0.11%) at higher

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solvent: raw material ratio ranging from 12.5 to 15 **(Figure 2)**. The variation trend in Y with the solvent: raw material ratio was similar to previous reports by others [24,30,31]. Theoretically, for a fixed amount of raw material, a higher quantity of solvent is utilized, the driving force for the mass transfer of oil is stronger, and the extraction rate is faster. However, if the solution is very dilute, an extra solvent increase would not lead to a sufficient increase in the concentration difference, and the increase in extraction yield would be limited.

ALA, LA, and OA increased from 88.70, 46.40, and 44.10 mg/g to 108.40, 57.09, and 53.60 mg/g, respectively, when the solvent: raw material ratio increased from 5 to 10, but these indexes weakened as the ratio further increased **(Figure 2)**. No significant differences in UFAs among solvent: raw material ratios of 10, 12.5 and 15 were observed. A similar trend was reported in watermelon seeds oil [29].

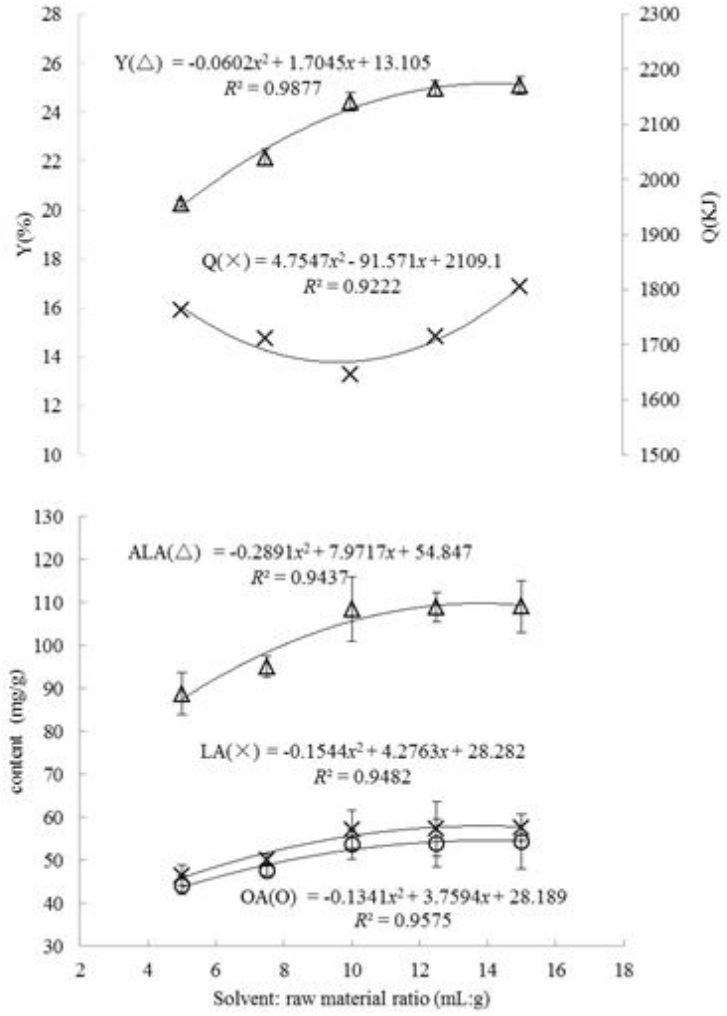

Figure 2. Oil properties (Y, ALA, LA, and OA) and energy consumption (Q) as functions of solvent: raw material ratio.

With an increase in the solvent: raw material ratio, Q initially decreased and then increased. Q reached the bottom (1,646.41 KJ), when the solvent: raw material ratio was 10. Ultrasonic working energy consumption and heating energy consumption were equal when ultrasonic power, extraction time, and temperature were same. When a higher amount of solvent was used, a higher amount of rotary evaporation energy was required. The increase in Y was greater than that in Q when the solvent: raw material ratio increased from 5 to 10; therefore, Q decreased at higher solvent: raw material ratios; however, the increase in Y was less than that of Q when the solvent: raw material ratio ranged from 10 to 15, and Q increased at higher solvent: raw material ratios.

Effect of Ultrasonic Power

Ultrasonic power could significantly affect Y, UFAs, and Q. Y increased by 4.99% when ultrasonic power was increased from 100 to 300 W, while it slightly decreased at 350 W and 400 W **(Figure 3)**. The result indicated that the increase in Y was not positively correlated with ultrasonic power at a specific reaction time, which was in agreement with the reports by [32,33]. Ultrasonic treatment of samples can induce acoustic cavitation and rupture of plant cells, which in turn promotes the release of internal components of plant cells into the solvent and mass transfers between the solid matrix and solvent. However, excessively high power could also increase the occurrence of bubbles in the solvent during cavitation, which in turn might reduce the efficiency of the ultrasound energy that is transmitted into the medium. Furthermore, an extremely high power may lead to the decomposition of oil components [34].

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UFAs and Y showed a similar profile, whereas Y, ALA, LA, and OA reached its maximum levels when ultrasonic power was 322.22 W, 316.13 W, 318.2 W, and 319.8 W (calculated from the regression equations), respectively. These findings may be related to the structure of UFAs. ALA was trienic acid, LA was dienoic acid; both of these were more easily oxidized than OA at higher ultrasonic power [35]. Ultrasonic power had a positive effect on Q **(Figure 3)**. Under the same solvent: raw material ratio, extraction time, and extraction temperature, higher ultrasonic power consumed more ultrasonic working energy.

Effect of Extraction Time

Y increased by 3.83% when extraction time ranging from 20 to 40 min, then increased by 0.39% when the time from 40 to 50 min, and decreased by 0.26% from 50 to 60 min. The results showed clearly that Y increased rapidly in the first 40 min, and then reached a steady state, finally, slightly decreased when extraction time was prolonged **(Figure 4)**. Such a trend was similar for all three UFAs **(Figure 4)**. Ultrasonic facilitated the release of oil inside the plant cells to the exterior solvent and gave a large yield at the early stage of extraction. The rinsing step may have released most of the oil from the broken cells at the first 40 min. Moreover, the concentration difference was large at the beginning of the extraction, which resulted in high osmotic pressure and diffusion driven. As the diffusion front moved towards the interior of the tissue, the diffusion area decreased, the diffusion distance increased, and the diffusion rate decrease accordingly. Therefore, no distinct increase in Y was observed in subsequent phases. As extraction time was prolonged, the chemical decomposition of the oil and oxidation of UFAs may have occurred [32]. Similar explanations can be applied to the changes in the three UFAs. **(Figure 4)** demonstrates a linear increase in the response of Q over time. A positive correlation between extraction time and energy consumption was observed.

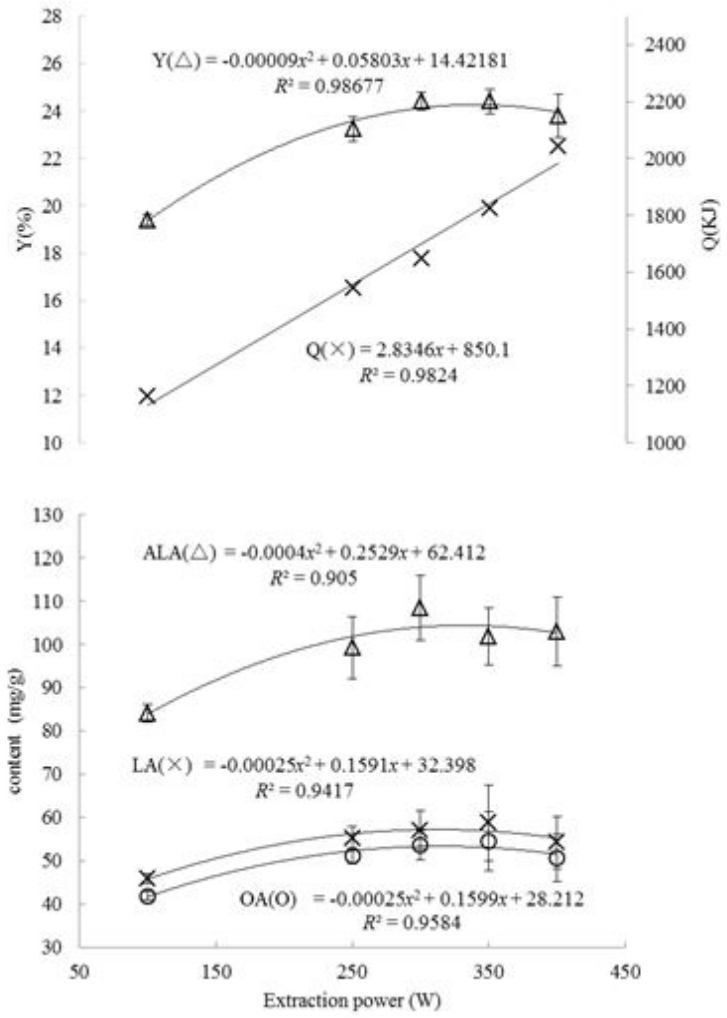

Figure 3. Oil properties (Y, ALA, LA, and OA) and energy consumption (Q) as functions of extraction power.

Effect of Extraction Temperature

With an increase in temperature, Y initially increased and then subsequently decreased **(Figure 5)**. The three UFAs also showed similar trends **(Figure 5)**. At a higher temperature, a larger Y was obtained. Previous studies have shown that the extraction rate of most compounds increase with temperature. Two reasons may explain this phenomenon: mild heating might soften the

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plant tissue, and weaken the cell wall integrity the increase of the oil diffusion coefficient and the enhanced solubility of the oil in the extracting solvent at higher temperatures caused the increase of the oil mass going out from the seeds into the solution [27,36]. However, the solvent often evaporates at higher temperatures, which in turn decreases the contact area of the solvent and the material, as well as reduces the diffusion rate. Moreover, earlier studies have shown that as the temperature approaches the boiling point of the solution, sonication becomes ineffective due to a decrease in surface tension and an increase in vapor pressure with microbubbles, which in turn causes the damping of ultrasonic waves [24,37]. Furthermore, the decomposition of oil and the oxidation of UFAs may occur at higher temperatures [15]. These factors may have been responsible for the observed decrease in Y and UFAs. Extraction temperature had a significant effect on Q, which increased by 492.39 KJ when the temperature was increased from 20°C to 60°C. The result indicated that higher extraction temperatures required more heating energy.

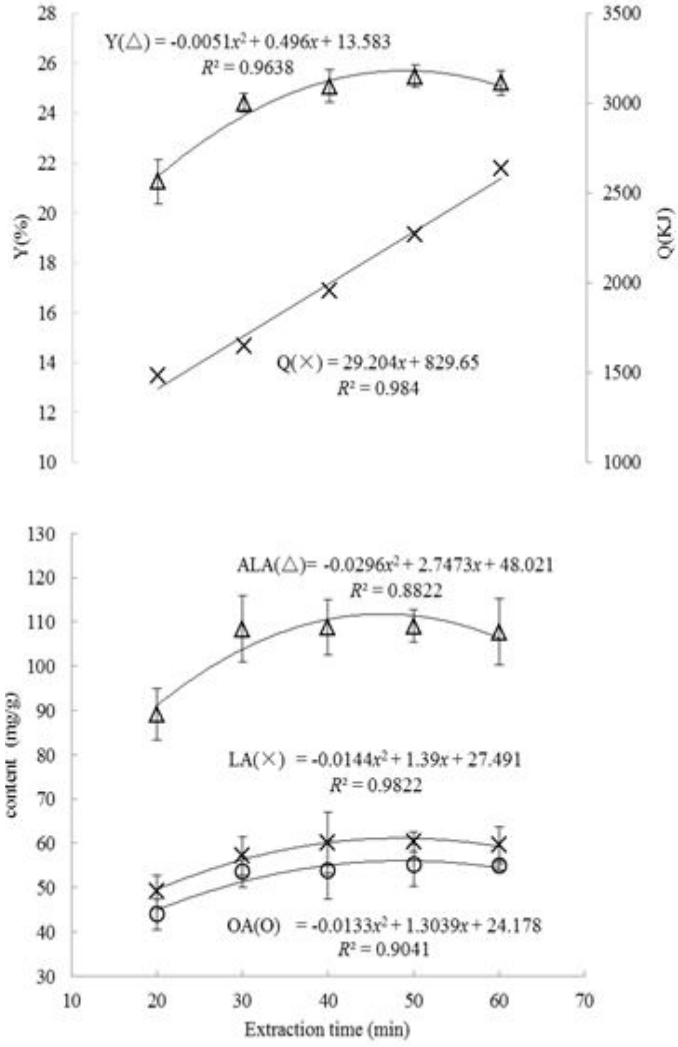

Figure 4. Oil properties (Y, ALA, LA, and OA) and energy consumption (Q) as functions of extraction time.

Nonlinear Regression

Based on the experimental data, five regression equations (particle size, solvent: raw material ratio, ultrasonic power, extraction time, and extraction temperature as functions of Y, ALA, LA, OA, and Q) were obtained as follows:

$Y=-42.54 + 9.01(x_1/10) - 0.72(x_1/10)^2 + 1.72x_2 - 0.06(x_2)^2 + 5.84(x_3/100) - 0.86(x_3/100)^2 + 5.29(x_4/10) - 0.56(x_4/10)^2 + 3.42\ (x_5/10) - 0.37(x_5/10)^2$,

R^2= 0.9738..... (1)

$ALA=-208.94 + 39.18(x_1/10) - 3.15(x_1/10)^2 + 7.39x_2 - 0.26(x_2)^2 + 27.93(x_3/100) - 4.33(x_3/100)^2 + 29.03(x_4/10) - 3.20(x_4/10)^2 + 20.98(x_5/10) - 2.40(x_5/10)^2$,

R^2= 0.9131..... (2)

$LA=-120.38 + 23.32(x_1/10) - 1.87(x_1/10)^2 + 4.95x_2 - 0.19(x_2)^2 + 13.66(x_3/100) - 2.11(x_3/100)^2 + 13.96(x_4/10) - 1.44(x_4/10)^2 + 11.55(x_5/10) - 1.33(x_5/10)^2$,

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R^2= 0.9495..... (3)

OA=-115.64 + 21.29(x_1/10) - 1.71(x_1/10)^2 + 3.87x_2 - 0.14(x_2)^2 + 13.89(x_3/100) - 2.13(x_3/100)^2 + 14.56(x_4/10) - 1.56(x_4/10)^2 + 12.35(x_5/10) - 1.33(x_5/10)^2,

R^2=0.9350..... (4)

Q=3364.25 - 713.07(x_1/10) + 57.07(x_1/10)^2 - 95.77x_2 + 4.96(x_2)^2 + 94.05(x_3/100) + 38.68(x_3/100)^2 + 38.62(x_4/10) + 32.07(x_4/10)^2 - 98.81(x_5/10) + 27.42(x_5/10)^2,

R^2=0.9953..... (5)

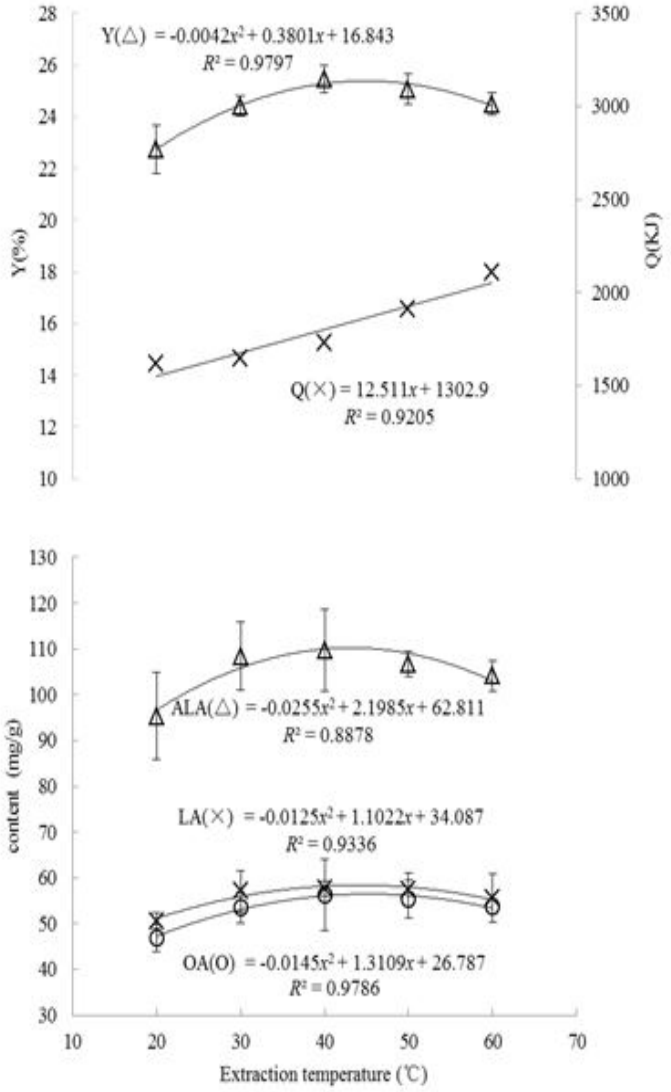

Figure 5. Oil properties (Y, ALA, LA, and OA) and energy consumption (Q) as functions of extraction temperature.

where x_1 is the particle size (mesh); x_2 is the solvent: raw material ratio; x_3 is the ultrasonic power (W); x_4 is the extraction time (min); x_5 is the extraction temperature (°C); and R is the correlation coefficient of each regression. To avoid the overfitting of the data, the forward selection method was used during the regression process. The order of a testing polynomial was started from one. A good fit to the data was achieved when the order of 2 was used for the polynomial regression. Using these five equations, Y, ALA, LA, OA, and Q of peony seed oil extraction can be predicted based on the production conditions, such as particle size, solvent: raw material ratio, ultrasonic power, extraction time and temperature at a significance level of 0.05. The relative error between experimental and estimated values derived from the regression equations is presented in **Table 2**. For Case One and Case Two, the relative error of these five indexes fell into the range of -3.05 to 1.39% and was within ± 5%, suggesting that the results were reliable and acceptable for industrial applications.

Operations Research

Because the oil content of peony seeds was 35% (as determined by using a Soxhlet apparatus), Y should be <35%, and ALA, LA, and OA would be <350 mg/g, respectively. In cases where maximum Y is desired, the maximum ALA, LA, OA of 350 mg/g and the minimum Q of 0 KJ were required, and non-linear programming was performed as follows:

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Table 2. Relative error between experimental value and estimated value derived from the regression equations.

Oil properties and energy consumption	Case one			Case two		
	Estimated value	Experimental value	Relative error (%)	Estimated value	Experimental value	Relative error (%)
Y	24.34	24.41	-0.29	25.44	25.09	1.39
ALA	105.09	108.40	-3.05	109.54	109.05	0.45
LA	56.31	57.09	-1.37	57.31	57.57	-0.45
OA	52.46	53.60	-2.13	54.31	54.34	-0.05
Q	1,663.76	1,646.41	1.05	1,804.91	1,805.19	-0.01

Case one: particle size of 60 mesh, solvent: raw material ratio of 10, ultrasonic power of 300 W, extraction time of 30 min, and extraction temperature of 30°C; Case two: particle size of 60 mesh, solvent: raw material of 15, ultrasonic power of 300 W, extraction time of 30 min, and extraction temperature of 30°C.

Max $F(x)$=-42.54 + 9.01(x_1/10) - 0.72(x_1/10)^2 + 1.72x_2 - 0.06(x_2)^2 + 5.84(x_3/100) - 0.86(x_3/100)^2 + 5.29(x_4/10) - 0.56(x_4/10)^2 + 3.42 (x_5/10) - 0.37(x_5/10)^2, which is subject to the following:

- 208.94 + 39.18(x_1/10) - 3.15(x_1/10)^2 + 7.39x_2 - 0.26(x_2)^2 + 27.93(x_3/100) - 4.33(x_3/100)^2 + 29.03(x_4/10) - 3.20(x_4/10)^2 + 20.98 (x_5/10) - 2.40(x_5/10)^2 < 350;

- 120.38 + 23.32(x_1/10) - 1.87(x_1/10)^2 + 4.95x_2 - 0.19(x_2)^2 + 13.66(x_3/100) - 2.11(x_3/100)^2 + 13.96(x_4/10) - 1.44(x_4/10)^2 + 11.55(x_5/10) - 1.33(x_5/10)^2 < 350;

- 115.64 + 21.29(x_1/10) - 1.71(x_1/10)^2 + 3.87x_2 - 0.14(x_2)^2 + 13.89(x_3/100) - 2.13(x_3/100)^2 + 14.56(x_4/10) - 1.56(x_4/10)^2 + 12.35(x_5/10) - 1.33(x_5/10)^2 < 350; and

3,364.25 - 713.07(x_1/10) + 57.07(x_1/10)^2 - 95.77x_2 + 4.96(x_2)^2 + 94.05(x_3/100) + 38.68(x_3/100)^2 + 38.62(x_4/10) + 32.07(x_4/10)^2 - 98.81 (x_5/10) + 27.42(x_5/10)^2 > 0

The experimental condition constraints are as follows:

40 < x_1 < 80, 0 < x_2 < 15, 250 < x_3 < 400, 20 < x_4 < 60, 20 < x_5 < 60

The optimal solution for the problem is (x_1, x_2, x_3, x_4, x_5) = (62.57, 14.33, 339.53, 47.23, 46.21), with a maximum $F(x)$=28.28%. The results indicated that a maximum Y of 28.28% for peony seed oil extraction can be obtained with a particle size of 62.57 mesh, solvent: raw material ratio of 14.33, ultrasonic power of 339.53 W, extraction time of 47.23 min, and extraction temperature of 46.21°C. The conditions result in ALA, LA, OA, and Q values of 121.73 mg/g, 65.08 mg/g, 62.58 mg/g, and 2,575.28 KJ, respectively.

When a maximum ALA is desired, a maximum Y, LA, and OA of 35%, 350 mg/g, and 350 mg/g, and a minimum Q of 0 KJ are required, and nonlinear programming can be performed as follows:

Max $F(x)$=- 208.94 + 39.18(x_1/10) - 3.15(x_1/10)^2 + 7.39x_2 - 0.26(x_2)^2 + 27.93(x_3/100) - 4.33(x_3/100)^2 + 29.03(x_4/10) - 3.20(x_4/10)^2 + 20.98 (x_5/10) - 2.40(x_5/10)^2, which is subject to the following:

- 42.54 + 9.01(x_1/10) - 0.72(x_1/10)^2 + 1.72x_2 - 0.06(x_2)^2 + 5.84(x_3/100) - 0.86(x_3/100)^2 + 5.29(x_4/10) - 0.56(x_4/10)^2 + 3.42 (x_5/10) - 0.37(x_5/10)^2 < 35%;

-120.38 + 23.32(x_1/10) - 1.87(x_1/10)^2 + 4.95x_2 - 0.19(x_2)^2 + 13.66(x_3/100) - 2.11(x_3/100)^2 + 13.96(x_4/10) - 1.44(x_4/10)^2 + 11.55(x_5/10) - 1.33(x_5/10)^2 < 350;

- 115.64 + 21.29(x_1/10) - 1.71(x_1/10)^2 + 3.87x_2 - 0.14(x_2)^2 + 13.89(x_3/100) - 2.13(x_3/100)^2 + 14.56(x_4/10) - 1.56(x_4/10)^2 + 12.35(x_5/10) - 1.33(x_5/10)^2 < 350;

3,364.25 - 713.07(x_1/10) +57.07(x_1/10)^2 - 95.77x_2+4.96(x_2)^2 + 94.05(x_3/100) + 38.68(x_3/100)^2 + 38.62(x_4/10) + 32.07(x_4/10)^2 - 98.81 (x_5/10) + 27.42(x_5/10)^2 > 0.

The optimal solution is (x_1, x_2, x_3, x_4, x_5)=(62.19, 14.21, 322.52, 45.36, 43.71), with a maximum $F(x)$=122.13 mg/g. It is suggested that the maximum ALA for peony seed oil extraction can be obtained at a particle size of 62.19 mesh, a solvent: raw material ratio of 14.21, ultrasonic power of 322.52 W, extraction time of 45.36 min, and extraction temperature of 43.71°C. Also, Y, LA, OA, and Q for extraction were 28.21%, 65.17 mg/g, 62.51 mg/g, and 2,410.25 KJ, respectively.

To increase LA, the optimal solution is (x_1, x_2, x_3, x_4, x_5)=(62.35, 13.03, 323.70, 48.47, 43.42), with a maximum $F(x)$=65.58 mg/g. The results indicated that at a particle size of 62.35 mesh, solvent: raw material ratio of 13.03, ultrasonic power of 323.70 W, extraction time of 48.47 min, and extraction temperature of 43.42°C, the maximum LA was 65.58 mg/g, with a maximum Y, ALA, and OA of 28.12%, 121.45 mg/g, and 62.40 mg/g, respectively, and a minimum Q of 2,469.33 KJ for peony seed oil extraction.

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To attain a maximum OA, the optimal solution is (x_1, x_2, x_3, x_4, x_5) = (62.25, 13.82, 326.06, 46.67, 46.43), with a maximum F(x)=62.66 mg/g. Thus, at a particle size of 62.25 mesh, solvent: raw material ratio of 13.82, ultrasonic power of 326.06 W, extraction time of 46.67 min, and extraction temperature of 46.43°C, a maximum OA of 62.66 mg/g, with a maximum Y, ALA, LA of 28.25%, 121.85 mg/g, and 65.29 mg/g, respectively, and a minimum Q of 2,489.55 KJ for extraction can be obtained.

In addition, to achieve a minimum Q process, the optimal solution is (x_1, x_2, x_3, x_4, x_5)=(62.47, 9.65, 250, 30, 20), with a minimum F(x)=1,467.99 KJ. Thus, at a particle size of 62.47 mesh, solvent: raw material ratio of 9.65, ultrasonic power of 250 W, extraction time of 30 min, and extraction temperature of 20°C, a minimum Q of 1,467.99 KJ can be obtained, as well as a maximum Y, ALA, LA, and OA of 22.05%, 93.41 mg/g, 50.06 mg/g, and 45.37 mg/g, respectively.

The nonlinear programming approach can help mill personnel in designing production conditions that include particle size, solvent: raw material ratio, ultrasonic power, extraction time, and extraction temperature to produce desired oil with optimal oil properties and energy consumption.

Peony seed oil has been a resource of interest because of its higher ALA [38,39]. Higher Y, LA and OA, as well as a lower Q can be achieved using the process that produces a maximum ALA. In the present study, based on comprehensive analysis of nutrition value, market value, production cost, simple operation, an optimal process was acquired with five parameters rounded: a particle size of 62 mesh, solvent: raw material ratio of 14, ultrasonic power of 323 W, extraction time of 45 min, and extraction temperature of 44°C. Under optimized conditions, a Y of 28.21%, ALA of 122.11 mg/g, LA of 65.22 mg/g, OA of 62.53 mg/g, and Q of 2,394.99 KJ were obtained.

CONCLUSION

The present study revealed that, although energy consumption of ultrasound-assisted extraction in peony seed oil was increased, the oil yield, the content of α-linolenic acid, linolein acid and oleic acid were increased as the ultrasonic power, extraction time and temperature increased. Five nonlinear regression equations (Equations. (1)-(5)) were developed. Given the conditions for seed oil extraction (particle size, solvent: raw material ratio, ultrasonic power, extraction time, and extraction temperature), the oil properties and energy consumption can be estimated by mill personnel. Using the nonlinear programming model, the optimal Y, ALA, LA, OA, and Q could be obtained under certain practical constraints in cases where a maximum ALA is desired and a maximum Y, LA, and OA of 35%, 350 mg/g, and 350 mg/g, respectively, and a minimum Q of 0 KJ are required. The optimal solution was the maximum ALA of 122.11 mg/g, which can be obtained when a particle size of 62 mesh, solvent: raw material ratio of 14, ultrasonic power of 323 W, extraction time of 45 min, and extraction temperature of 44°C were used, which makes Y, LA, OA, and Q for peony seed oil equal to 28.21%, 65.22 mg/g, 62.53 mg/g, and 2,394.99 KJ, respectively. The results of the present study provide a promising seed oil extraction technology for the peony industry.

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