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Extraction of Peanut Oil Using Thermosonication: Modeling and Multiobjective Optimization of Process Parameters Using Box–Behnken Design

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Abstract: The extraction of peanut oil was investigated using the combination of ultrasound and heat application, which is known as a novel technology called thermosonication. The study was set up using the Box–Behnken design and the models based on quadratic equations were established. The effects of extraction time (4-12 min), extraction temperature (40-60°C), solvent–to–solid ratio (SSR) (3:1-9:1)(v/w) and ultrasound power (60-100%) on the extraction yield and the oleic acid concentration of extracted oils were investigated. Results showed that the extraction yield was primarily affected by the extraction temperature and SSR. The average maximum yield of 39.93% was achieved when variables were set to 12 min of time, 50℃ of temperature, 9:1(v/w) of SSR and 80% of ultrasound power. Thermosonication did not significantly affect the fatty acid composition. Since it was targeted to determine an optimum point where the maximum extraction yield and oleic acid concentration were obtained, a multiobjective optimization was performed. The optimum thermosonication conditions were determined as 4 min of time, 60℃ of temperature, 9:1(v/w) of SSR and 100% of power with a maximum extraction yield of 39.86%. Also, the oleic acid concentration was determined as 63.51% in this optimum condition.

Key words: thermosonication, Box–Behnken, peanut oil, oleic acid, multiobjective optimization

1 Introduction

Peanut oil is considered as an important vegetable oil extracted from *Arachis hypogaea* L., or commonly known as peanut or groundnut. According to the latest available production data of FAO[1], more than 5 million tonnes of groundnut oil was produced worldwide in 2014, and two countries –China and India– are leading this production with 1.8 and 1.25 million tons, respectively. The importance of peanut oil rises from its higher content of unsaturated fatty acids (UFA), especially oleic acid (*C18:1*) followed by linoleic acid (*C18:2*). Some of the reported oleic and linoleic acid contents of peanut oil are 69% and 43%[2], 67.4% and 35.4%[3], 58% and 21%[4], respectively. The rest of the fatty acid composition is mainly constituted by such saturated fatty acids (SFA) as palmitic (*C16:0*), stearic (*C18:0*), arachidic (*C22:0*) and behenic acids (*C24:0*) in varying concentrations; however, it has been reported that none of these SFAs except palmitic acid is found higher than 10% in peanut oil[3]. The characteristic distribution of the SFA and UFA of peanut oil has been reported to result in better oxidative stability[5]. Besides, the enhanced quality attributes due to low free fatty acid (FFA) value and the pleasant flavor of peanut oil makes it a preferable food oil[4]. From all these aspects, this study focused on extracting peanut oil without causing any significant changes in its fatty acid composition using a novel extraction technique such as thermosonication.

Ultrasound technology has recently gained increasing interest in utilization for such purposes as cleaning, homogenization, degassing, emulsification and extraction. The types of equipment for ultrasonication are either ultrasonic baths or immersible probes both of which can operate at different sonication parameters such as frequency, ultrasonic power, time and temperature. The extraction processes using ultrasound depend on the formation of small bubbles by ultrasound waves; therefore resulting in cavitation[6]. These rapidly growing and shrinking bubbles cause an increase in the cell diffusivity of the material[7], and this leads to more solvent penetrating into the material resulting in a better extraction yield of the targeted component.

Thermosonication is a combined treatment of both ultrasound and thermal processing. The use of ultrasound in

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food processing has been investigated for recent years, and many studies have been published in the literature so far. However, heat application at mild or higher temperatures in addition to ultrasound processing –as called thermosonication– is a novel and relatively intact field; thus, it needs further investigation. Thermosonication is generally used in the inactivation of microorganisms[8–11] or enzymes[12–14] in mostly risky food products such as fruit juices and milk rather than the extraction of valuable or bio–active compounds[15] from the food material.

On the other hand, ultrasound–assisted extraction (UAE) method has been intensively studied for the extraction of oil from various oil sources. Many studies involving modeling and/or optimization of the extraction process have been published in the literature. The use of ultrasound for the extraction of oil has a wide range from conventional oil sources such as olive[16, 17], rapeseed[18], soybean[19] or almond[20] to lesser–known sources such as avocado[21], pepper seed[22], papaya seed[23], perilla[24], radish seed[25], and some specific plants[26] or fungi[27].

Despite recent studies in the field, it has been noticed that there is only few information about the extraction of peanut oil using ultrasound technology in the literature. Therefore, this study aimed to investigate the effects of different thermosonication conditions on the extraction yield and the oleic acid concentration of peanut oil. Besides, the thermosonic extraction was modeled using Box–Behnken design of the response surface methodology (RSM) and the process was optimized in order to find the optimum extraction conditions where maximum yield and oleic acid concentration could be reached.

2 Materials and Methods

2.1 Materials

Unroasted and unsalted peanuts were purchased from a local market in Çankırı province of Turkey in mid-2019. Prior to analyzes, the samples were kept in a dark, dry and cool place in order to prevent undesired oxidation of lipids. The peanuts were separated from all possible residues of leaves, hard shells and miscellaneous parts of the plant before analyzes. Analytical grade n-hexane used for oil extraction was purchased from Merck (Merck KGaA, Darmstadt, Germany). All other chemicals used for preparing the methyl esters of fatty acids were purchased from Sigma-Aldrich (Sigma Aldrich, St. Louis, USA).

2.2 Thermosonic extraction of oil

Peanuts were grounded using a Sinbo SCM2934 coffee grinder (Sinbo, Turkey) and 10 ± 0.05 g of the grounded sample was transferred to a glass beaker. 30, 60 and 90 mL of n-hexane was introduced to the beaker to obtain the solvent-to-solid ratios (SSR) of 3:1, 6:1 and 9:1 (v/w), respectively. The samples were then placed in the center position of an Elmasonic P60-H ultrasonic bath (Elma Schmidbauer GmbH, Germany) with a maximum power outlet of 580 W. The applied ultrasonic power was 60, 80 and 100% of the maximum power. The operation frequency of the ultrasound bath was 37 kHz. The duration of the extraction process ranged from 4 to 12 min, and the applied extraction temperatures were 40, 50 and 60℃. Overheating due to ultrasonic vibration was prevented by placing plastic ice packs in the ultrasonic bath. The schematic representation of the thermosonication process is given in Fig. 1. After the extraction process, the solvent was removed from the solution using a rotary evaporator (Hei–VAP Precision ML/G1, Heidolph Instruments GmbH & CO.KG, Germany) for 5 min. In the next step, the samples were kept in an oven at 40℃ overnight in order to remove the possible traces of solvent.

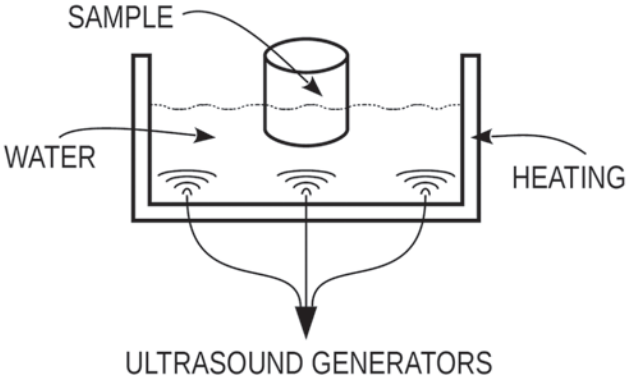


Fig. 1 The schematic representation of thermosonication process used in the study.

After the removal of the solvent, the yield of the thermosonic extraction was calculated as expressed in Eq. 1

$$Yield\,(\%) = \left(\frac{m_o}{m_i}\right) \times 100 \qquad \text{Eq. (1)}$$

where (m_o) is the mass of extracted oil and (m_i) is the mass of grounded peanut.

2.3 Determining the fatty acid composition of oils by gas chromatography (GC)

Fatty acids were converted into their methyl esters according to the official ISO method no: 12966-2:2011[28] prior to GC analysis. The esters were then injected into a gas chromatography unit (GC 2010 Plus, Shimadzu, Japan) equipped with a TR–CN100 capillary column (60 m × 0.25 mm inner diameter × 0.20 μm film thickness) (Teknokroma, Spain) and a flame ionization detector (FID). The injection port, oven and detector temperatures were set to 230℃, 190℃ and 240℃, respectively. The column flow was 0.15 mL/min with a split ratio of 1:150. Helium was used as the carrier gas, and the duration of the analysis was 60 min. The identification of the peaks was performed by making a comparison using the data obtained from the injection of

fatty acids methyl esters mixture (Supelco® 37 Component FAME Mix, Sigma-Aldrich, St. Louis, USA). The quantification was performed by the built–in integrator of the GC software.

2.4 Design of experiment (DOE)

The Box–Behnken model was used as the design of the study. The effects of the extraction time (x_1), extraction temperature (x_2), SSR (x_3) and ultrasound power (x_4) were investigated on the extraction yield (y_1) and the oleic acid concentration (y_2) of the extracted peanut oils. Each factor had three levels as follows:

- x_1: 4, 8 and 12 (min)
- x_2: 40, 50 and 60 (℃)
- x_3: 3:1, 6:1, 9:1 (v/w)
- x_4: 60, 80 and 100 (% of maximum power)

Because the boiling point of *n*-hexane is about 68.7℃, the maximum extraction temperature was chosen as 60℃ to avoid undesired boiling and consequent changes in SSR. All experiments were conducted in two replicates and a total of 54 runs were performed with six measurements for the center points. The data were fitted to full quadratic models as expressed in Eq. 2

$$y_i = c_0 + c_1 x_1 + c_2 x_2 + c_3 x_3 + c_4 x_4 + c_{11} x_1^2 + c_{22} x_2^2 + c_{33} x_3^2 + c_{44} x_4^2 + c_{12} x_1 x_2 + c_{13} x_1 x_3 + c_{14} x_1 x_4 + c_{23} x_2 x_3 + c_{24} x_2 x_4 + c_{34} x_3 x_4 \quad \text{Eq. (2)}$$

where y_i is the response to be predicted from the model (y_1 or y_2), c_0 is the constant, c_1, c_2, c_3 and c_4 are linear coefficients, c_{11}, c_{22}, c_{33} and c_{44} are the quadratic coefficients and c_{12}, c_{13}, c_{14}, c_{23}, c_{24} and c_{34} are the interaction coefficients.

2.5 Data analysis and optimization

The data obtained from experiments were analyzed using Minitab statistical software (Version 16, Minitab Inc., USA). The confidence level for analysis of variance (ANOVA) was chosen as 95%. For a better presentation of the simultaneous effects of coupled variables on the extraction yield and oleic acid concentration, experimental data were plotted on 3D and contour graphs using Statistica v10 (Statsoft Inc., Tulsa, OK).

Since the study aimed to extract the oil from peanuts as much as possible while remaining the oleic acid concentration at the maximum, a multiobjective optimization was performed to determine an optimum point where both responses could reach to desired maximum levels. The built–in response optimizer of Minitab was used and the starting points of variables were chosen as their corresponding minima, i.e. 4 min, 40℃, 3:1 v/w and 60% of the power. Since both functions were equally desired, the importance values were chosen equally as 1. After the optimum point was determined, a further thermosonication experiment was performed in order to verify the optimization procedure.

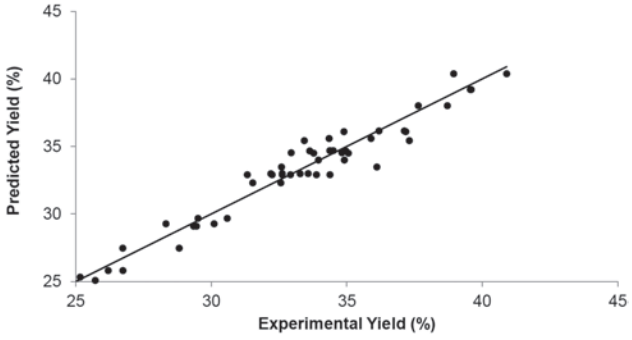


Fig. 2 The relationship between the experimental and predicted extraction yields.

3 Results and Discussion

3.1 Assessment of the experimental data

The experimental design with both coded and uncoded variables, the experimental and the predicted responses of thermosonically extracted peanut oils using Box-Behnken design are presented in Table 1. The average extraction yields ranged from 24.23% to 39.93%, while no significant differences were observed in the oleic acid concentration which had an average of 61.4%. It could be considered that the extraction yield data were in agreement with other studies in literature where the extraction of peanut oil was investigated using the conventional Soxhlet method. The reported oil contents of peanut kernels belonging to different cultivars ranged from 31.52% to 44.09%[29], and from 32.7% to 45.4%[30]. The minimum and the maximum yields were observed in the thermosonication conditions of 8 min of extraction time, 40℃ of extraction temperature, 3:1 (v/w) of SSR, 80% of power and 12 min-50℃-9:1 (v/w)-80%, respectively. The experimental data were then used to construct full quadratic models that would fit the design best. The models with uncoded units for both y_1 and y_2 are given in Eqs. 3-4. The correlation between the experimental and predicted extraction yields is given in Fig. 2.

$$y_1 = -9.37296 - 0.00246X1 + 1.19439X2 - 0.57334X3 + 0.06003X4 + 0.06799X1^2 - 0.0068X2^2 - 0.18717X3^2 + 0.00161X4^2 - 0.03691X1X2 + 0.20257X1X3 - 0.00436X1X4 + 0.05737X2X3 - 0.00488X2X4 + 0.00088X3X4 \quad \text{Eq. (3)}$$

$$y_2 = 96.811 - 0.14X1 - 0.9651X2 + 0.1242X3 - 0.2732X4 + 0.0336X1^2 + 0.006X2^2 + 0.0385X3^2 + 0.001X4^2 + 0.0069X1X2 - 0.0434X1X3 - 0.007X1X4 - 0.0008X2X3 + 0.0038X2X4 - 0.0044X3X4 \quad \text{Eq. (4)}$$

The fatty acid compositions of the extracted oils are

Table 1 Experimental and predicted responses under different thermosonication conditions.

Coded Variables				Uncoded Variables				Extraction Yield (%)		Oleic Acid (%)	
x_1 (min)	x_2 (°C)	x_3 (v/w)	x_4 (%)	x_1 (min)	x_2 (°C)	x_3 (v/w)	x_4 (%)	*Experimental*	*Predicted*	*Experimental*	*Predicted*
0	0	0	0	8	50	6:1	80	32.90 ± 1.11	32.90	60.55 ± 0.26	60.55
0	+	+	0	8	60	9:1	80	39.58 ± 0.02	39.20	60.19 ± 0.96	60.92
0	0	−	+	8	50	3:1	100	27.77 ± 1.47	27.46	61.91 ± 0.06	62.06
0	0	+	−	8	50	9:1	60	36.66 ± 0.66	36.14	61.35 ± 1.44	61.11
+	0	0	+	12	50	6:1	100	35.12 ± 1.09	35.58	60.58 ± 0.10	60.62
0	−	0	−	8	40	6:1	60	29.22 ± 1.25	29.27	62.67 ± 0.69	62.42
0	−	−	0	8	40	3:1	80	24.23 ± 0.62	25.30	62.92 ± 0.70	62.04
0	−	+	0	8	40	9:1	80	32.05 ± 0.74	32.30	62.13 ± 0.05	61.14
+	0	+	0	12	50	9:1	80	39.93 ± 1.38	40.38	60.84 ± 0.21	60.11
0	+	0	−	8	60	6:1	60	34.44 ± 0.08	34.69	60.85 ± 1.07	60.73
0	+	−	0	8	60	3:1	80	24.88 ± 0.40	25.32	61.08 ± 0.48	61.92
0	0	−	−	8	50	3:1	60	26.47 ± 0.39	25.81	61.80 ± 0.81	61.52
−	−	0	0	4	40	6:1	80	30.05 ± 0.76	29.67	61.76 ± 0.14	62.39
+	−	0	0	12	40	6:1	80	34.35 ± 2.49	33.47	60.05 ± 0.21	61.18
−	0	0	−	4	50	6:1	60	32.74 ± 0.76	32.97	61.46 ± 1.19	61.27
+	0	0	−	12	50	6:1	60	33.89 ± 1.32	34.52	60.66 ± 0.21	61.74
0	+	0	+	8	60	6:1	100	34.42 ± 0.91	34.50	61.76 ± 0.55	62.25
−	0	+	0	4	50	9:1	80	34.30 ± 0.93	34.66	60.78 ± 0.44	61.82
+	0	−	0	12	50	3:1	80	25.31 ± 0.59	25.08	62.90 ± 0.12	62.10
−	0	0	+	4	50	6:1	100	35.37 ± 2.74	35.43	63.64 ± 1.14	62.41
+	+	0	0	12	60	6:1	80	34.43 ± 0.68	33.99	62.27 ± 0.22	61.55
0	0	+	+	8	50	9:1	100	38.18 ± 0.76	38.01	60.39 ± 0.01	60.58
−	+	0	0	4	60	6:1	80	36.04 ± 1.62	36.08	62.88 ± 0.06	61.66
0	−	0	+	8	40	6:1	100	33.10 ± 0.68	32.98	60.56 ± 0.12	60.92
−	0	−	0	4	50	3:1	80	29.40 ± 0.07	29.08	60.75 ± 0.33	61.72

Experimental values are represented as mean ± standard deviation of replicates.

summarized in Table 2. Seven major fatty acids including palmitic (*C16:0*), stearic (*C18:0*), oleic (*C18:1*), linoleic (*C18:2*), arachidic (*C20:0*), eicosenoic (*C20:1*) and behenic (*C22:0*) acids were identified using the GC analysis. Oleic acid was the most abundant fatty acid ranging from 60.05% to 63.64%, followed by linoleic acid (approximately 21-22%) and palmitic acid (approximately 9-9.5%) in the first 3 rankings. The oleic acid range of this study was found higher than the reported data from the conventional extraction studies in literature. The reported oleic acid concentrations of peanut kernels ranged from 43.13% to 55.07%[29], and from 48.4% to 57.3%[30]. Although this difference might be due to the physicochemical characteristics of the material, the analyzed oleic acid concentrations in this study were good proofs of the efficiency of ultrasound technique. Additionally, it was observed that the use of different thermosonication conditions did not significantly affect the overall fatty acid composition of the extracted oils, and this effect was also reported by other researchers[18−20, 25, 26].

3.2 Visualization of the data

The interaction effects of process variables on the extraction yield and oleic acid concentration are visualized using contour and 3D surface plots in Fig. 3 and Fig. 4, respectively. According to Fig. 3a, the interaction effect of time and temperature had a significant effect on the yield. The extraction yield was greatly increased at elevated temperatures when the extraction time was kept constant. No significant effect was observed for the extraction time at constant SSR as seen in Fig. 3b. From Fig. 3d, the simultaneous increase in temperature and power greatly affected the extraction yield, as this finding was similar to the research of Samaram *et al.*[23]. According to Figs. 3e and 3f, there were no considerable changes in the extraction yield when low SSR values were applied with either increasing

Table 2 Fatty acid compositions (%) of peanut oils extracted under different thermosonication conditions.

Independent Variables				Fatty Acids (%)						
Time (min)	Temperature (℃)	SSR (v/w)	Power (%)	Palmitic *(C16:0)*	Stearic *(C18:0)*	Oleic *(C18:1)*	Linoleic *(C18:2)*	Arachidic *(C20:0)*	Eicosenoic *(C20:1)*	Behenic *(C22:0)*
8	40	6:1	60	9.18 ± 0.01	2.88 ± 0.41	62.67 ± 0.69	21.11 ± 0.06	1.17 ± 0.27	0.76 ± 0.12	2.23 ± 0.05
8	40	3:1	80	9.22 ± 0.04	2.92 ± 0.28	62.92 ± 0.70	21.09 ± 0.08	1.15 ± 0.34	0.68 ± 0.08	2.01 ± 0.12
8	40	9:1	80	9.35 ± 0.07	3.13 ± 0.10	62.13 ± 0.05	21.20 ± 0.07	1.27 ± 0.09	0.65 ± 0.05	2.27 ± 0.23
4	40	6:1	80	9.27 ± 0.09	3.31 ± 0.08	61.76 ± 0.14	21.02 ± 0.09	1.45 ± 0.01	0.83 ± 0.02	2.36 ± 0.02
12	40	6:1	80	9.56 ± 0.02	3.26 ± 0.11	60.05 ± 0.21	22.30 ± 0.15	1.46 ± 0.08	0.91 ± 0.10	2.46 ± 0.08
8	40	6:1	100	9.49 ± 0.03	3.03 ± 0.01	60.56 ± 0.12	22.57 ± 0.07	1.36 ± 0.04	0.80 ± 0.04	2.20 ± 0.01
8	50	3:1	100	9.49 ± 0.22	2.71 ± 0.37	61.91 ± 0.06	22.04 ± 0.47	1.08 ± 0.35	0.69 ± 0.10	2.09 ± 0.06
8	50	9:1	60	9.52 ± 0.04	2.83 ± 0.68	61.35 ± 1.44	22.24 ± 0.34	1.12 ± 0.42	0.80 ± 0.25	2.14 ± 0.39
12	50	6:1	100	9.48 ± 0.04	3.19 ± 0.08	60.58 ± 0.10	22.21 ± 0.08	1.39 ± 0.07	0.86 ± 0.01	2.30 ± 0.08
12	50	9:1	80	9.41 ± 0.06	3.21 ± 0.29	60.84 ± 0.21	21.82 ± 0.73	1.44 ± 0.13	0.83 ± 0.14	2.44 ± 0.01
8	50	3:1	60	9.44 ± 0.06	2.98 ± 0.40	61.80 ± 0.81	21.58 ± 0.25	1.22 ± 0.29	0.76 ± 0.21	2.21 ± 0.22
4	50	6:1	60	9.48 ± 0.11	3.00 ± 0.58	61.46 ± 1.19	21.79 ± 0.07	1.21 ± 0.51	0.76 ± 0.22	2.30 ± 0.06
12	50	6:1	60	9.44 ± 0.02	3.40 ± 0.12	60.66 ± 0.21	21.75 ± 0.08	1.52 ± 0.05	0.83 ± 0.07	2.41 ± 0.04
4	50	9:1	80	9.39 ± 0.08	3.42 ± 0.08	60.78 ± 0.44	21.47 ± 0.15	1.52 ± 0.05	0.91 ± 0.05	2.52 ± 0.13
12	50	3:1	80	9.18 ± 0.05	3.04 ± 0.08	62.90 ± 0.12	20.56 ± 0.03	1.29 ± 0.01	0.77 ± 0.04	2.25 ± 0.19
4	50	6:1	100	9.20 ± 0.17	2.69 ± 0.58	63.64 ± 1.14	20.51 ± 0.05	1.12 ± 0.41	0.70 ± 0.16	2.14 ± 0.22
8	50	9:1	100	9.54 ± 0.02	3.12 ± 0.01	60.39 ± 0.01	22.85 ± 0.08	1.28 ± 0.10	0.71 ± 0.02	2.11 ± 0.01
4	50	3:1	80	9.48 ± 0.07	2.87 ± 0.16	60.75 ± 0.33	22.78 ± 0.04	1.15 ± 0.16	0.73 ± 0.06	2.24 ± 0.01
8	50	6:1	80	9.59 ± 0.02	3.01 ± 0.09	60.55 ± 0.26	22.57 ± 0.03	1.23 ± 0.07	0.77 ± 0.07	2.27 ± 0.11
8	60	9:1	80	9.48 ± 0.09	3.12 ± 0.41	60.19 ± 0.96	22.55 ± 0.10	1.40 ± 0.22	0.88 ± 0.13	2.38 ± 0.21
8	60	6:1	60	9.42 ± 0.01	3.07 ± 0.61	60.85 ± 1.07	22.13 ± 0.32	1.31 ± 0.36	0.82 ± 0.18	2.40 ± 0.24
8	60	3:1	80	9.50 ± 0.04	3.03 ± 0.30	61.08 ± 0.48	22.25 ± 0.17	1.29 ± 0.23	0.74 ± 0.12	2.12 ± 0.04
8	60	6:1	100	9.15 ± 0.07	3.35 ± 0.23	61.76 ± 0.55	20.87 ± 0.17	1.49 ± 0.13	0.92 ± 0.12	2.45 ± 0.17
12	60	6:1	80	9.11 ± 0.03	3.05 ± 0.12	62.27 ± 0.22	21.03 ± 0.01	1.33 ± 0.10	0.78 ± 0.01	2.43 ± 0.02
4	60	6:1	80	9.12 ± 0.06	2.84 ± 0.13	62.88 ± 0.06	21.05 ± 0.01	1.13 ± 0.01	0.70 ± 0.03	2.28 ± 0.09

Values are represented as mean ± standard deviation of replicates.

temperature or increasing ultrasound power.

As stated in Section 3.1, the concentrations of oleic acid did not significantly change during the extractions; however, it can be concluded from Fig. 4 that the extraction temperature and the SSR were the most effective variables on the oleic acid concentrations. According to Figs. 4b, 4e and 4f, lower SSR values revealed higher oleic acid concentrations when the other variable was kept constant. Even there were no significant decreases in the oleic acid, this phenomenon might be due to the transition of other fatty acids to the extract at high solvent concentrations; thus, resulting in relatively lower oleic acid concentrations among other fatty acids.

The experiments revealed that the single effects of the extraction temperature and SSR were more significant than those of extraction time and ultrasound power. The overall trend of the single assessment of the variables showed that extracting at higher temperatures resulted in more oil yield, which is in agreement with previous studies[16, 20, 24, 25]. Samaram *et al.*[23] stated that obtaining higher oil yields at elevated temperatures was because of the easier transfer of oil from the cells which were softened by heat. However, Mohammadpour *et al.*[26] reported the decrease in the extraction yield at increasing temperatures. According to the authors, there were two reasons for this: a) as the vapor pressure increased with heat, less pressure difference occurred between the bubbles and their surroundings, and b) the surface tension was lowered. The yield was also increased when SSR was increased because increasing the concentration gradient helped to transfer more oil droplets to the solvent phase. This is also in agreement with previous studies[18, 20, 23–26]. The experimental data and the interaction plots reveal that the extraction time was not as effective as SSR and temperature. In a relevant study, researchers stated that most of the oil was extracted at the beginning of the ultrasound process; therefore, lower extraction times would be beneficial since the extraction rate decreases in time[25].

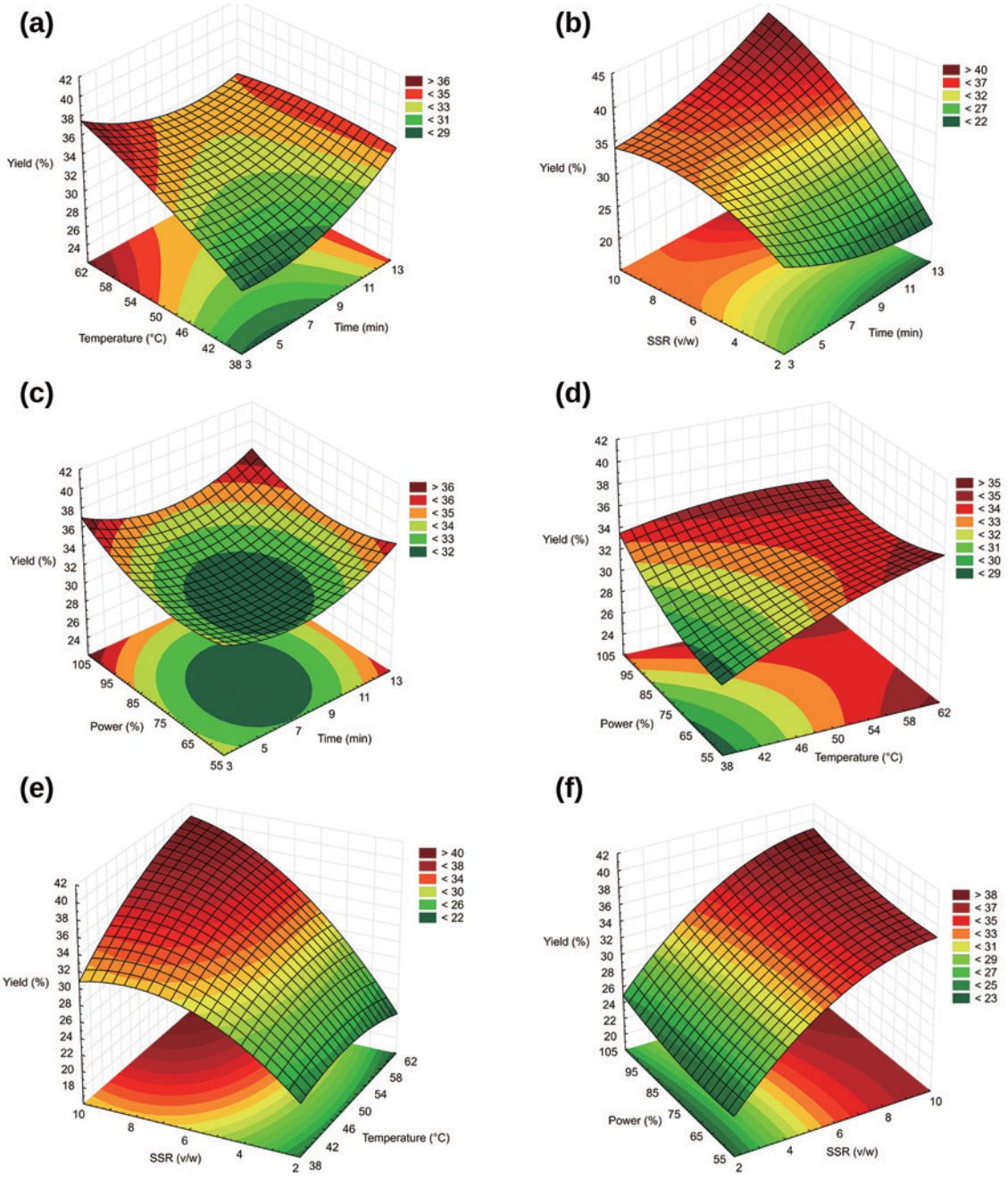


Fig. 3 3D surface and contour plots of extraction yields.

3.3 Statistical assessment of the models

The ANOVA outputs from the statistical analyzes for y_1 and y_2 are presented in Tables 3 and 4, respectively. According to Table 3, a high F value for the regression of the extraction yield (54.84) and very low p value ($p<0.05$) indicated that the regression model was statistically significant for the representation of the experimental data. The adjusted determination coefficient (Adj R^2) was 0.9343, which meant that the experimental and predicted responses were matching well. The ANOVA revealed that only time × power ($p>0.05$) and SSR × power ($p>0.05$) interactions were statistically insignificant among other interactions. Even the single extraction time and SSR effects had high p values ($p>0.05$) individually, ${x_1}^2$ ($p=0.002$) and ${x_3}^2$ ($p=0.000$) were statistically significant. The p value for lack-of-fit was 0.688 ($p>0.05$); therefore, it was concluded that the size of the design with 54 runs was adequate to successfully investigate how the thermosonication conditions affected the extraction yield.

In contrast to the extraction yield, the low F and high p

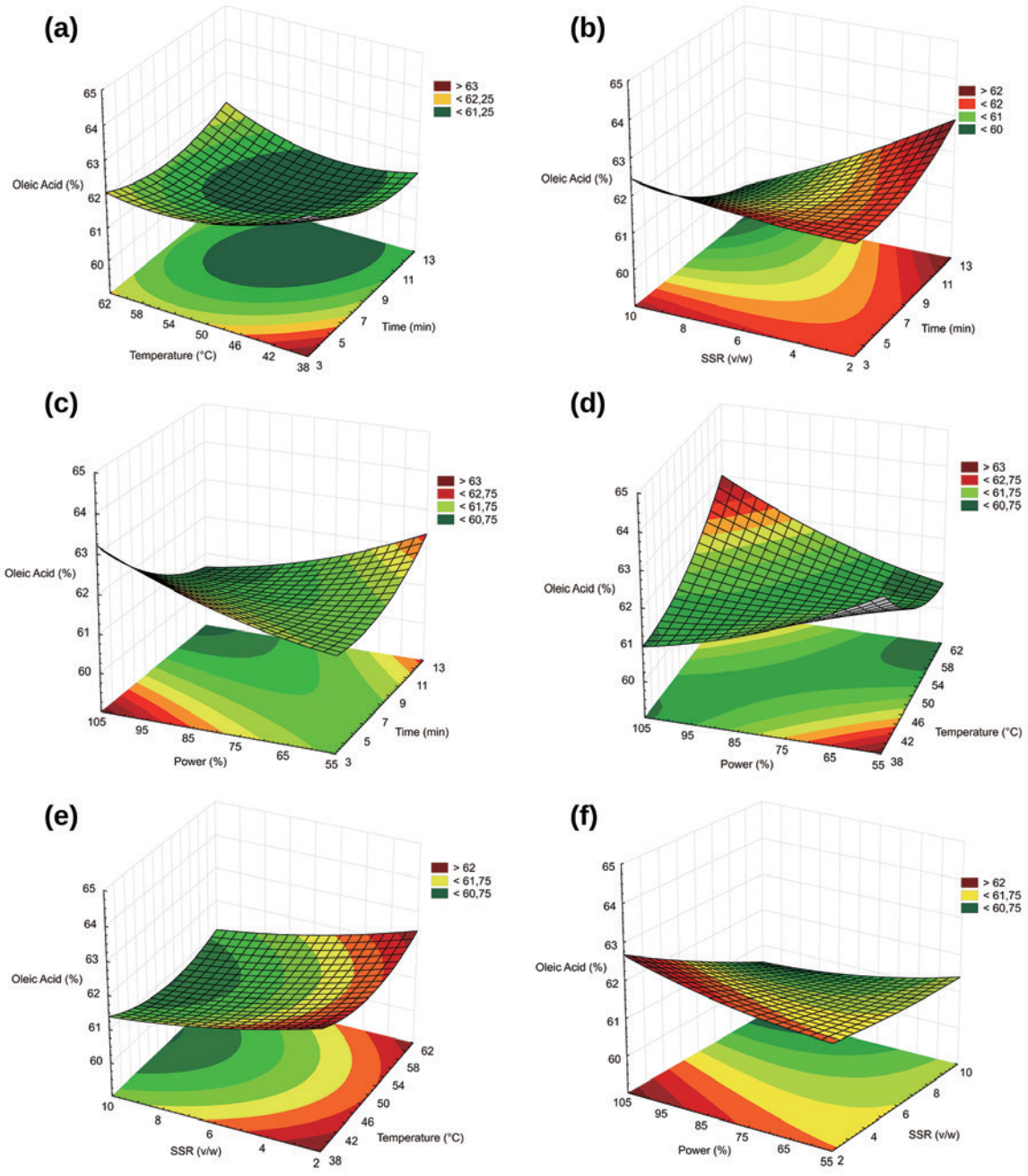


Fig. 4 3D surface and contour plots of oleic acid concentrations.

values for the regression of the oleic acid concentrations given in Table 4 represent that the model was statistically insignificant and the oleic acid concentrations were not significantly affected by the process variables. As aforementioned in Section 3.1, there were only minor changes in the fatty acid compositions and this was also reported by other researchers. Therefore, the adjusted determination coefficient (Adj R^2) was 0.1849, meaning an independent relationship between the variables and the response.

3.4 Optimization and verification of process variables

Since the peanut oil is a good source of oleic acid, the models were subjected to a multiobjective optimization in order to determine the optimum conditions where most of the oil could be extracted from the peanut with the highest available oleic acid concentration. By the aid of the experimental data, the desired ranges for y_1 and y_2 were set to 30-40% and 60-65%, respectively. The built-in response optimizer of the statistical software achieved to determine an optimum point at the following conditions:

Table 3 ANOVA outputs for the extraction yield.

		Yield			
Source	DF	Adj SS	Adj MS	F	P
Regression	14	923.084	65.9345	54.84	0.000
Linear	4	15.639	3.9098	3.25	0.021
Time	1	0.000	0.0000	0.00	0.997
Temperature	1	11.489	11.4891	9.56	0.004
SSR	1	0.413	0.4128	0.34	0.561
Power	1	0.142	0.1418	0.12	0.733
Square	4	77.290	19.3225	16.07	0.000
*Time*Time*	1	12.622	12.6224	10.50	0.002
*Temperature*Temperature*	1	4.929	4.9290	4.10	0.050
*SSR*SSR*	1	30.267	30.2674	25.18	0.000
*Power*Power*	1	4.420	4.4195	3.68	0.063
Interaction	6	97.015	16.1692	13.45	0.000
*Time*Temperature*	1	17.437	17.4368	14.50	0.000
*Time*SSR*	1	47.274	47.2739	39.32	0.000
*Time*Power*	1	0.972	0.9721	0.81	0.374
*Temperature*SSR*	1	23.694	23.6936	19.71	0.000
*Temperature*Power*	1	7.616	7.6165	6.34	0.016
*SSR*Power*	1	0.022	0.0222	0.02	0.893
Residual Error	39	46.889	1.2023		
Lack-of-Fit	10	9.458	0.9458	0.73	0.688
Pure Error	29	37.431	1.2907		
Total	53				
R^2			0.9517		
Adj. R^2			0.9343		

- x_1: 4 min
- x_2: 60℃
- x_3: 9:1 v/w
- x_4: 100%

The optimized responses at this point were calculated as 39.86% and 63.51% for y_1 and y_2, respectively. The desirability functions for y_1 and y_2 were 0.986 and 0.702, respectively and the composite desirability was 0.832. In order to verify these data, an additional extraction was performed at the optimum condition. The verified responses and the fatty acid composition of the extracted oil at the optimum condition are presented in Table 5. The experimental yield was found higher than the predicted optimum data –which was desired for extracting the maximum amount of oil from the peanut–, and the oleic acid concentration was achieved to remain above 62%. From the verification data, it can be concluded that the optimization process was applicable and beneficial when the functions were desired to reach their maximum simultaneously.

4 Conclusion

The effects of thermosonication parameters including extraction time, extraction temperature, solid–to–solvent ratio (SSR) and ultrasound power on the extraction yield and the oleic acid concentration of the thermosonically extracted peanut oil were investigated. The average maximum extraction yield was 39.93% when the thermosonication parameters were set to 12 min-50℃-9:1(v/w)-80%, respectively. The extraction yields of thermosonication were comparable and mostly in agreement with the extraction yields of such conventional extraction methods as Soxhlet method. The highest oleic acid concentration of the extracts was 63.64% with thermosonication parameters of 4 min-50℃-6:1(v/w)-100%. The results revealed that the overall fatty acid compositions did not significantly change during the extractions with an average oleic acid concentration of 61.4%. It was also observed that the single effects of the extraction temperature and SSR were more effective on the extraction yield than the remaining

Table 4 ANOVA outputs for oleic acid concentration.

Oleic Acid					
Source	DF	Adj SS	Adj MS	F	P
Regression	14	24.1328	1.72377	1.86	0.064
Linear	4	9.0816	2.27039	2.45	0.062
Time	1	0.0438	0.04376	0.05	0.829
Temperature	1	7.5011	7.50109	8.09	0.007
SSR	1	0.0194	0.01936	0.02	0.886
Power	1	2.9359	2.93587	3.17	0.083
Square	4	5.5047	1.37618	1.48	0.226
*Time*Time*	1	3.0864	3.08645	3.33	0.076
*Temperature*Temperature*	1	3.9016	3.90158	4.21	0.047
*SSR*SSR*	1	1.2819	1.28190	1.38	0.247
*Power*Power*	1	1.8797	1.87973	2.03	0.162
Interaction	6	10.4384	1.73973	1.88	0.110
*Time*Temperature*	1	0.6050	0.60500	0.65	0.424
*Time*SSR*	1	2.1736	2.17361	2.34	0.134
*Time*Power*	1	2.5425	2.54251	2.74	0.106
*Temperature*SSR*	1	0.0050	0.00500	0.01	0.942
*Temperature*Power*	1	4.5451	4.54511	4.90	0.033
*SSR*Power*	1	0.5671	0.56711	0.61	0.439
Residual Error	39	36.1690	0.92741		
Lack-of-Fit	10	26.2666	2.62666	7.69	0.000
Pure Error	29	9.9024	0.34146		
Total	53				
R^2			0.4002		
Adj. R^2			0.1849		

Table 5 Optimized responses from the extraction at the optimum conditions.

Optimized variables						
x_1: 4 min		x_2: 60℃		x_3: 9:1 v/w		x_4: 100%
Optimum responses from verification run (%)						
y_1: 48.68 ± 0.31						
y_2: 62.61 ± 0.64						
Fatty acids (%)						
Palmitic *(C16:0)*	Stearic *(C18:0)*	Oleic *(C18:1)*	Linoleic *(C18:2)*	Arachidic *(C20:0)*	Eicosenoic *(C20:1)*	Behenic *(C22:0)*
9.58 ± 0.03	2.62 ± 0.01	62.61 ± 0.64	21.86 ± 0.14	0.65 ± 0.02	0.62 ± 0.04	2.06 ± 0.73

Values are represented as mean ± standard deviation of replicates.

parameters. Mathematical models were built using Box-Behnken design of the response surface methodology, and an optimum condition was determined where both extraction yield and oleic acid concentration were maximized. The optimum conditions were 4 min-60℃-9:1(v/w)-100% for x_1, x_2, x_3 and x_4, respectively. The extraction yield and oleic acid concentration at the optimum condition were calculated as 39.86% and 63.51%, respectively. These values were verified with an additional extraction run which resulted in close agreement with the predicted data from the optimization. It is concluded that thermosonication can be utilized as a novel and alternative way to

extract oil from peanuts with high extraction yields while remaining the fatty acid composition almost same.

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Conflict of Interest

The author declares no conflict of interest.

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