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Optimisation of *Swietenia macrophylla* Seed Oil Extraction using Ultrasound-assisted Method

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Swietenia macrophylla is a large deciduous tree that mainly grows in open rain forests. Its seeds reportedly have many medical efficacies, such as antidiabetic properties and antioxidative effects. This study used ultrasound-assisted extraction (UAE) and Central Composite Design (CCD) Response Surface Methodology (RSM) to optimise the oil yield extracted from *S. macrophylla* seed. The properties of the extracted *S. macrophylla* seed oil, namely antioxidant activity and bioactive compounds, were also examined. For the CCD RSM, the seed oil yield was set as the response while the parameters for optimization were solid-to-solvent (SS) ratio (1:3, 1:4, and 1:5), amplitude (70 %, 80 %, and 90 %), and extraction time (5 min, 10 min, and 15 min). The optimum oil yield extracted via UAE was then subjected to several analyses. First, the oil's DPPH (2,2-diphenyl-1-picrylhydrazyl) radical scavenging activity was analysed to determine antioxidant activity. Then, Gas chromatography–Mass spectrometry (GC-MS) analysis was used to evaluate the oil's bioactive compounds (methyl ester fatty acids). The UAE method extracted the optimum yield (27.68 ± 0.3 wt%) of *S. macrophylla* seed oil under the following conditions: 1:4.5 S/S ratio, 90 % amplitude, and 14.4 min extraction time. The analysis of antioxidant activity revealed a DPPH radical scavenging activity of 87.96 ± 0.04 %. The GC-MS analysis revealed several fatty acids in the oil, including arachidonic acids, stearic acid, palmitic acid, oleic acid, and linoleic acid. This study shows that UAE is a promising method for extracting *S. macrophylla* seed oil and that *S. macrophylla* is a potential oral hypoglycaemic agent that could be used for pharmaceutical applications.

1. Introduction

Swietenia macrophylla has been used by many world communities because every part of the plant has various ethnomedical uses. The seeds of this plant have especially been reported to have many significant medical properties. It is also a common folk medicine with antimicrobial, anti-inflammatory, antioxidant effects, as well as antidiabetic and anticancer properties. *Swietenia macrophylla* is commonly used in Malaysia to treat hypertension and diabetes by direct chewing and swallowing (Moghadamtousi et al., 2013).

Although a few synthetic oral hypoglycaemic agents, such as Tolbutamide and Chlorpropamide, have been introduced to treat diabetes, most of these agents cause detrimental side effects, such as gastrointestinal disturbance and lactic acidosis.*S. macrophylla* is reported to be a safer oral hypoglycaemic agent for treating diabetes (Dewanjee et al., 2009). The method for extracting plant oil is crucial for synthesising the oil extract. The conventional extraction method—Soxhlet extraction—usually requires a longer extraction time, a relatively high consumption of solvent, and has insufficient reproducibility (Dawidowicz et al., 2008). Ultrasound-assisted extraction (UAE) is an extraction method that could be applied in oil extraction industries to improve oil yield and reduce processing time, and can benefit the edible oil industry. Previous studies have used UAE to extract chemical compounds from flaxseed oil (Gutte et al., 2015), *Stevia rebaudiana* (Alupului et al., 2009), grape seeds (Da Porto et al., 2013), rice bran oil (Krishnan et al., 2015), and *S. macrophylla* (Gumaling et al.,

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2018). In their study, Gumaling et al. (2018) used n-hexane to extract oil from *S. macrophylla*. However, ethanol can be used to replace n-hexane because it is cheaper, more bio-renewable, and has less handling risks compared to n-hexane. It also has negligible toxicity. In this study, the SS ratio, amplitude, and extraction time of the UAE method were varied via RSM to optimise the oil yield extracted from *S. macrophylla*. Then, the bioactive compounds and antioxidant activity of the *S. macrophylla* seed oil were analysed.

2. Methodology

2.1 Plant sample and chemical

S. macrophylla seeds were bought from a local market in Bangi, Selangor, Malaysia. The seeds came with a peel and were peeled off prior to drying. The seeds were rinsed to remove foreign particles and dirt. The cleaned seeds were then dried in an oven (Memmert, Germany) set to 100 °C for 24 h. Following that, a blender (Panasonic blender, Japan) was used to grind the dried seeds. The ground seeds were then sieved to approximately 0.75 mm particle size.

2.2 Ultrasound-assisted extraction

The method of Da Porto et al. (2013) was used to extract the seed oil, with slight modifications. The extraction was assisted by 750 W and 20 kHz ultrasonic processors (SONICS model VCX 750) equipped with a titanium alloy tip probe (13 mm diameter). The *S. macrophylla* seeds (50 g) and (200 mL) ethanol were mixed in a 250 mL beaker. The solvent was fixed at 200 mL of ethanol, while the seed was changed based on the ratio of solid to solvent. A water bath was prepared to which the beaker and its contents were immersed and a temperature of 60 ± 5 °C was maintained. The probe was submerged about 4 cm under the surface of the mixture. Then, Whatman No. 1 paper was used to filter the mixture under vacuum. A rotary vacuum evaporator was used at 55 °C for approximately 20 min to remove part of the solvent. The oil was separated from the remaining solvent by centrifuging the mixture at 5000 rpm for 15 min. The extracted oil was pipetted out from the mixture into a vial. Samples were stored in the refrigerator at 4 °C for further analysis.

2.3 Determination of oil yield

Eq(1) was used to determine the yield of the extracted *S. macrophylla* seed oil:

$$\text{Yield (wt\%)} = \frac{\text{weight of oil extract produced (g)}}{\text{weight of ground sample (g)}} \times 100 \quad (1)$$

2.4 Design of experiment and statistical analysis

Design Expert software (version 7.1.5, Stat-Ease Inc., Minneapolis, US) was used to set the design-of-experiment for this study. In the software, six-centre-point Central Composite Design (CCD) was applied. Extraction time (A), solid-to-solvent ratio (B), and amplitude (C) were the three parameters (independent variables) varied in the Response Surface Methodology (RSM), while the response (dependent variable) was the *S. macrophylla* seed oil yield. The parameters—extraction time, and amplitude—were set based on Gumaling et al. (2018) and Wong et al. (2019).A preliminary study was conducted prior to the optimisation process to select the solid-to-solvent ratio, as shown in Table 1.

Table 1: Experimental range and independent parameters used in RSM

Parameters	Coded Factor	Factor Level				
		-∝	-1	0	+1	+∝
Extraction time (min)	A	1.59	5	10	15	18.41
Solid to solvent ratio (g/mL)	B	2.32	3	4	5	5.68
Amplitude (%)	C	63.18	70	80	90	96.82

Eq(2) was used to fit the parameter values into a polynomial equation:

$$Y = \alpha_0 + \alpha_1 A + \alpha_2 B + \alpha_3 C + \alpha_{11} A^2 + \alpha_{22} B^2 + \alpha_{33} C^2 + \alpha_{12} AB + \alpha_{13} AC \quad (2)$$

Where Y is the total oil yield; α_i and α_{ij} values represent constant regression coefficients; A: Extraction time, B: Solid to Solvent Ratio and C: Amplitude.

2.5 Evaluation of antioxidant activity via DPPH radical scavenging assay

Antioxidant activity was determined per the method outlined in Ruttarattanamongkol and Petrasch (2016) and Abu Bakar (2019), with slight modifications. First, 1100 µL DPPH solution was added to 200 µL of *S.*

macrophylla solution (5 mg/mL) and mixed. To prepare the DPPH solution, 1.97 mg of DPPH reagent was diluted in 50 mL of 95 % ethanol. All measures were done in triplicate so the mixture was poured into several vials. The next step was to incubate the reaction mixture in room temperature in the dark for 1 h. Next, a spectrophotometer (Jenway model 7300 Instrument Inc., UK) was used to record the absorbance at 517 nm against ethanol as a blank. Eq(3) was used to determine the percentage of inhibition or the scavenging activity of the free radicals:

$$\text{DPPH Scavenging Activity}(\%) = \frac{(A_0 - A_1)}{A_0} \times 100 \quad (3)$$

The samples were analysed in triplicate. A_0 is the absorbance of the control and A_1 is the absorbance of the sample.

2.6 Evaluation of bioactive compounds via GC-MS

The bioactive compounds (methyl ester fatty acids (FAMEs)) in the extracted oil were determined using GC-MS, as described by Kandhro et al. (2008). A HP-5MS capillary column (5 % phenyl methylsilyloxane), with dimensions of 30 m × 0.25 mm internal diameter × 0.25 µm film thickness (Agilent 1909Is-433, Agilent Technologies, Palo Alto, CA, USA), was used to separate the methyl ester fatty acids. The temperature was initially maintained for 2 min at 150 °C, after which it was increased to 230 °C.

3. Results and discussions

3.1 Optimisation of ultrasound-assisted extraction conditions and verification of the model

Table 2 shows the actual response (oil yield) and the predicted response, over 20 runs, with an experimental design consisting of different combinations. The data was fitted to a quadratic equation model to determine the conditions for the optimum oil yield based on varying the three independent variables (extraction time (A), solid-to-solvent ratio (B), and amplitude (C)) and observing the effect of this variation on the response (Y) (oil yield), as shown in Eq(4):

$$Y = 24.91 + 3.09\,A + 0.44\,B + 1.60\,C - 0.16\,AB - 0.36\,AC + 0.68\,BC - 1.51\,A^2 - 1.10\,B^2 + 0.21\,C^2 \quad (4)$$

The results of the Analysis of Variance (ANOVA) are presented in Table 3. The coefficient of determination (R^2) and the adjusted coefficient of determination (Adj. R^2) for the quadratic regression model was 0.9270 and 0.8614. Both are high R^2-values, indicating that the model could be used to explain and predict the oil yield extracted using UAE. The R^2-value can be expressed as the proportion of variance in a set of data explained by a statistical model. When the R^2-value approaches 1, the model can be said to be well fitted to the actual data (Sin et al., 2006). The model is significant if the P-value is less than 0.05. The 'Lack of Fit' was not significant ($P > 0.05$), so the reliability of the model in predicting the response is confirmed. The interaction term with the largest effect on the oil yield was BC.

Figure 1 shows the 3D contour plots showing the correlation between the independent variables and the oil yield of the *S. macrophylla* seeds. The link between extraction time and solid-to-solvent ratio at a constant 80 % amplitude is shown in Figure 1 (a), where increased extraction time and increased solid-to-solvent ratio also increased the *S. macrophylla* seed oil yield. A prolonged time can ensure that the oil can be extracted completely from the seed into the solvent via ultrasonic waves. Increasing the solid-to-solvent ratio can improve the concentration gradient, which, in turn, enhances the amount of oil that can be recovered from the solvent. Since the main mass transfer limitation in this range is within the solid matrix, this effect is attenuated with a higher solvent-to-solid ratio (Mohammadpour et al., 2019). The relationship between extraction time and amplitude with oil yield at a constant solid-to-solvent ratio of 4 g/mL is shown in Figure 1 (b). The result shows that the oil yield increased with increased time and amplitude. This result is due to the increased cavitation effect as the amplitude increases. The high amplitude of the ultrasonic waves and the longer sonication time could damage more cell walls so more oil is released into the solvent (Krishnan et al., 2015). Figure 1 (c) shows the effect of solid-to-solvent ratio and amplitude on the oil yield at a constant extraction time of 10 min. The result shows that the oil yield increased with increased solid-to-solvent ratio and amplitude. Cracks will form in the cell wall due to cavitation. Then, bubbles start to form due to the changes in temperature and pressure. The bubbles subsequently collapse onto the solid material surface and releases high pressure and temperature, generating microjets directed towards the solid surface. The permeability of the plant tissues will increase, and facilitate the entry of the solvent into the inner part of the material, as well as help wash out the extract. In the UAE method, the plant cell wall can be disrupted even faster if a larger ultrasonic wave amplitude is applied. This wave travels through the liquid medium and helps the solvent penetrate into the solvent wall more effectively to release the intracellular product into the solvent more rapidly. The greater the volume of solvent, the faster the diffusion rate and the higher the mass transfer between the seeds and the solvent. An overly high volume of solvent could nevertheless decrease the dispersion of ultrasound energy.

Also, when the concentration of solvent is increased beyond a certain point, the mass transfer will still be limited to the interior of the solid, so the driving force will no longer increase (Gutte et al., 2015).

Table 2: Experimental design and responses for actual and predicted values

Run	Extraction Time (min) (A)	Solid to solvent ratio (g/mL) (B)	Amplitude (%) (C)	Oil yield (%)	
				Actual	Predicted
1	5.00	3.00	90.0	19.72	20.09
2	10.00	5.68	80.0	22.55	22.53
3	10.00	4.00	80.0	24.59	24.91
4	5.00	5.00	90.0	21.44	22.64
5	15.00	3.00	90.0	24.32	25.87
6	10.00	4.00	80.0	25.78	24.91
7	5.00	5.00	70.0	18.64	17.13
8	10.00	4.00	96.8	30.15	28.18
9	15.00	3.00	70.0	25.66	24.74
10	10.00	4.00	80.0	24.73	24.91
11	10.00	2.32	80.0	21.42	21.05
12	15.00	5.00	70.0	24.04	23.95
13	10.00	4.00	80.0	25.82	24.91
14	5.00	3.00	70.0	17.66	17.40
15	15.00	5.00	90.0	27.38	27.79
16	10.00	4.00	63.2	21.22	22.80
17	18.41	4.00	80.0	26.25	25.82
18	1.59	4.00	80.0	15.39	15.78
19	10.00	4.00	80.0	25.10	24.91
20	10.00	4.00	80.0	23.36	24.91

Table 3: ANOVA for the quadratic model

Source	Sum of Square	df	Mean Square	F Value	P Value
Model	222.16	9	24.58	14.12	0.0001
A (Extraction Time)	130.43	1	130.43	74.60	0.0001
B (Solid to solvent ratio)	2.67	1	2.67	1.53	0.2446
C (Amplitude)	35.05	1	35.05	20.05	0.0012
AB	0.20	1	0.20	0.11	
AC	1.02	1	1.02	0.58	
BC	3.67	1	3.67	2.10	
A^2	33.06	1	33.06	18.91	
B^2	17.52	1	17.52	10.02	
C^2	0.61	1	0.61	0.35	
Residual	17.48	10	1.75		
Lack of Fit	13.32	5	2.66	3.21	0.1135
Pure Error	4.16	5	0.83		
Cor Total	239.64	19			
R-Squared			0.9270		
Adj R-Squared			0.8614		
Pred R-Squared			0.5400		

By applying optimal conditions, a 28.17 w/w% oil yield was predicted. In the experimental procedure with conditions of 90 % amplitude, 1:4.5 g/mL solid-to-solvent ratio, and 14.4 min extraction time, the actual yield was 27.68 ± 0.3 w/w%. A 1.77 % (less than 5 %) percentage error was observed between the predicted and actual yield, so the optimal conditions are reasonable and adequate. This result also indicates the validity and acceptability of the response model for optimisation purposes.

Previous studies have reported various methods for extracting *S. macrophylla* seed oil using, including advanced extraction methods (Microwave-Assisted Extraction (MAE) (Abu Bakar, 2019) and Supercritical Carbon Dioxide Extraction (SC-CO_2) (Abu Bakar et al., 2020), as well as conventional methods (Soxhlet Extraction (SE) (Suliman et al., 2013)). In previous studies, UAE with hexane as a solvent produced the highest oil yield (46.7 ± 0.1 %) (Gumaling et al., 2018) followed by MAE (43.69 ± 0.092 %) (Abu Bakar, 2019), SE (42.7 %) (Suliman et al., 2013), and SC-CO_2 (28.88 ± 0.419 %) (Abu Bakar et al., 2020).

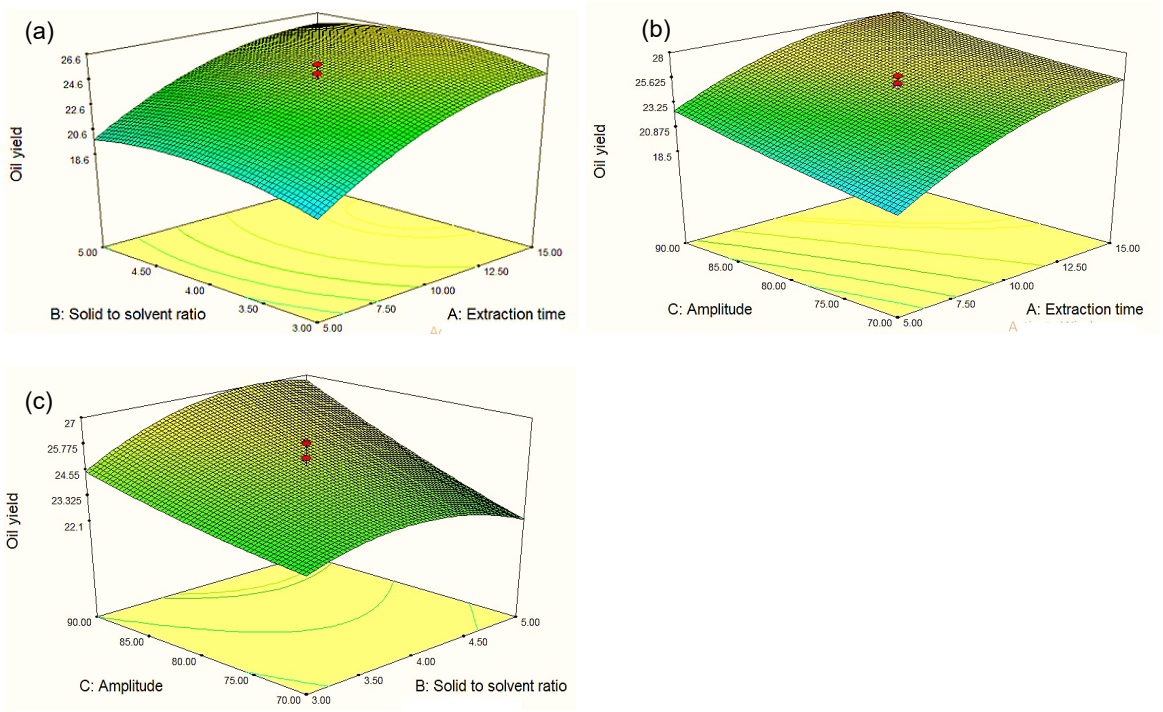

Figure 1: 3D contour plot showing correlation between (a) extraction time (min) and solid to solvent ratio (g/mL) (b) extraction time (min) and amplitude (%) (c) solid to solvent ratio (g/mL) and amplitude (%) to the oil yield of S. macrophylla seeds

3.2 Antioxidant activity by DPPH assay

The *S. macrophylla* seed oil extracted using UAE showed a 87.96 ± 0.04 % DPPH scavenging activity, which is a higher radical scavenging activity compared to MAE (64.80 ± 0.008 %), SC-CO_2 (45.9 ± 0.005 %), and SE (34.68 ± 0.003 %) (Abu Bakar et al., 2020). The UAE method produced oil with a higher DPPH radical scavenging activity because it involved no heat and required a lesser reaction time, so the oil yield retained more antioxidant activity.

3.3 Bioactive compounds by GC-MS

From the GC-MS results, 22 compounds were identified, of which the major fatty acids observed were palmitic acid (16:0) and stearic acid (18:0), and monounsaturated fatty acids, which are saturated fatty acids; and oleic acid (18:1 n-9c), and linoleic acid (18:2 n-6c), arachidic acid (20:4), and docosatrienoic acid (22:4), which are polyunsaturated fatty acids.

4. Conclusions

This study successfully demonstrated the potential of ultrasound-assisted extraction (UAE) to optimise the extraction of *S. macrophylla* seed oil. Using UAE, the highest *S. macrophylla* seed oil yield (27.68 ± 0.3 %) was obtained at 90 % amplitude, 14.4 min extraction time, and 1:4.5 g/mL solid-to-solvent ratio. The RSM model produced an R^2-value of 0.9270, considered satisfactory and indicating a good correlation between the predicted data and the actual data. The antioxidant activity of the *S. macrophylla* seed oil extracted using UAE was 87.96 ± 0.04 %, as determined from the DPPH radical scavenging activity (%). Compared to other extraction methods, UAE managed to extract oil with more types of fatty acids, such as linoleic acid, stearic acid, palmitic acid, oleic acid, linolenic acid, and arachidonic acids. UAE has a lot of advantages compared to the conventional extraction method. For example, the former requires lesser time and a lower volume of solvent than the latter, so it also leads to lower energy consumption and lower solvent cost. The oil extraction industry commonly applies UAE to extract bioactive compounds because the method has advantageous speed and is more economical than conventional extraction methods. It also involves low-cost technology. The oil extraction system can be simplified by replacing the pressing step with ultrasound methods.

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