

Research Article

Ultrasonic Extraction of Oil from *Caesalpinia spinosa* (Tara) Seeds

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Oil extracted from the seeds of *Caesalpinia spinosa* (common name: tara) can be used in a number of applications. In the present study, tara seed oil was obtained by ultrasonic extraction. The effects of different solvents, particle sizes in the ground seed samples, extraction times, ultrasonication powers, extraction temperatures, and liquid–solid ratios on the yield of tara seed oil were investigated. The yield from the ultrasonic extraction was compared with that from traditional Soxhlet extraction. The results showed that ultrasonic extraction could be completed in a shorter time with reduced solvent consumption. The yield of tara seed oil increased with increasing ultrasonication power and extraction temperature. Gas chromatography was used to analyze the fatty acid compositions of the oils extracted by the two methods. The fatty acid compositions of the oils from both extraction methods were similar, which indicates that ultrasonic extraction is a viable alternative means of extraction. It is a rapid, efficient, and simple method for production of lipids from tara seeds.

1. Introduction

A number of new methods for extracting oils from seeds have been investigated in recent years, including mechanical compression [1], ultrasonic extraction [2], microwave extraction [3], and supercritical fluid extraction [4]. Compared with traditional Soxhlet extraction, ultrasonic extraction provides higher selectivity, is less time-consuming, has lower energy consumption and reduced emissions [5, 6], and produces higher-quality oil [7]. It is also environmentally friendly because most of the extraction solvent can be recovered, and the equipment is inexpensive.

Caesalpinia spinosa (common name: tara) is an evergreen thorny shrub native to South America. It is valuable economically as it is used as a raw material in various applications [8, 9]. In China, tara plants are widely distributed in Yunnan province, and tara is used in a number of applications with high economic value [10]. Tara is rich in tannin [11], which can be used as a diluent in oil drilling, in the production of gallic acid and pyrogallol, as an antibiotic synergist and food additive, and in medicinal applications [9, 12–14]. Tara seeds have a thick endosperm layer [10, 15] and good thermal and

physicochemical stability [16, 17]. They are rich in a host of nutrients, including glucomannan, and have a high half milk protein content. The oil extracted from tara seeds contains numerous fatty acids and a high content of unsaturated fatty acids [13]. To date, no studies have investigated how the extraction process affects the quality or composition of tara seed oil. An understanding of the effect of the extraction on the oil is required to maximize its application potential.

In the present study, ultrasonic extraction of tara seed oil was optimized [18–20], and the effects of different extraction conditions on the yield of tara seed oil were determined [21–24]. Ultrasonic extraction was also compared with traditional Soxhlet extraction [25–28]. Gas chromatography was used to determine the fatty acid composition of the tara seed oil. The results provide a theoretical basis for optimizing tara oil extraction processes.

2. Results and Discussion

2.1. Effect of the Solvent on Ultrasonic Extraction of Tara Seed Oil. The organic solvent used for an extraction directly

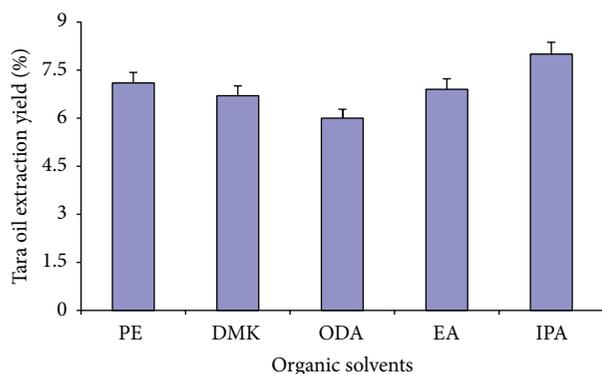


FIGURE 1: Tara seed oil yields with different organic solvents. Samples of dry tara powder (10 g, 60–80 mesh) were extracted with different organic solvents. The ultrasonic extraction was performed for 45 min at 250 W and 30 °C.

TABLE 1: Physical characteristics of tara seed oils extracted using different solvents.

Extraction solvents	Tara oil physical characteristics
Petroleum ether	Lustrous and transparent, clear, having no impurity, having nonstick centrifuge tube wall
Isopropyl alcohol	Dark brown, not clear, having impurities, accompanied by a substance similar to wax precipitation
n-Hexane	Oil clarification, relatively thick
Acetone	Oil was dark brown, thick
Ethyl acetate	Clarification, color and luster being deep, thick

affects the final oil yield. To select the most appropriate solvent for ultrasonic extraction of tara seed oil, petroleum ether, isopropyl alcohol, n-hexane, acetone, and ethyl acetate were all used for tara seed extraction while holding the other extraction conditions constant. The results are shown in Table 1. The solvent used affected both the tara seed oil yield and its physical characteristics (color and clarity).

The tara seed oil obtained using petroleum ether had the best physical characteristics among the extracted oils. In addition, the yield of oil obtained with petroleum ether was higher than with the other solvents (Figure 1). Consequently, petroleum ether was selected as the optimal extraction solvent for subsequent experiments.

2.2. Effect of Tara Seed Powder Particle Size on Ultrasonic Extraction of Tara Seed Oil. To investigate the correlation between the tara seed oil extraction yield and the particle size in the tara seed powder, samples with six particle sizes were compared (Figure 2(a)). Grinding of the tara seeds after drying breaks the internal structure of the cell wall, and this effectively improves solvent access to the cell contents. Consequently, the extraction yield improved as the particle size reduced. In the present study, the 60–80-mesh particle size was selected for the optimized tara seed oil extraction, because this particle size provided a good extraction yield

while not requiring too much grinding of the seeds for sample preparation.

2.3. Effect of the Liquid–Solid Ratio. During extraction, the liquid–solid ratio of a reaction mixture can affect the degree of interaction between the solid and the solvent, which in turn can affect the extraction yield. The best yield is achieved when the solution reaches saturation concentration. In the present study, with liquid–solid ratios of <14 : 1, the tara seed oil extraction yields were lower than at higher liquid–solid ratios. When the liquid–solid ratio was increased to 14 : 1, the extraction yield reached 6.7%. With a further increase in the liquid–solid ratio to 16 : 1, the extraction yield of tara seed oil increased only marginally (Figure 2(b)). Higher liquid–solid ratios result in a larger concentration gradient, which is beneficial for transfer of the tara seed oil from the powder to the solvent. Therefore, a liquid–solid ratio of 14 : 1 was selected for the optimized ultrasonic extraction of tara seed oil.

2.4. Effect of the Ultrasonication Power on Ultrasonic Extraction of Tara Seed Oil. Four levels of ultrasonication power were investigated (Figure 3(a)), and the yield of tara seed oil increased with increasing ultrasonication power. With the ultrasonication power at <250 W, the extraction yield was lower than when the ultrasonication power was at 250 W. This increase in yield could be attributed to more rapid collapse of bubbles in the solution by ultrasound waves at a higher ultrasonication power compared with at a lower ultrasonication power [29]. Collapse of bubbles in the solution will promote solvent penetration into the cell tissue and accelerate release of cell contents into the extraction solution to improve the extraction yield. From these results, an ultrasonication power of 250 W was selected for the optimized method.

2.5. Effect of Extraction Temperature on Ultrasonic Extraction of Tara Seed Oil. Changing the temperature used for ultrasonic extraction affects the interaction between the solid and liquid. In the present study, the extraction yield increased with increasing of the extraction temperature (Figure 3(b)). The highest extraction yield was obtained with an extraction temperature of 60 °C, which was selected as the optimum temperature for subsequent experiments. Higher temperatures would enhance the speed of bubble collapse in the solvent, and this would promote solvent penetration into the cell tissue and accelerate release of the cell contents into the extraction solution.

2.6. Effect of Extraction Time on Ultrasonic Extraction of Tara Seed Oil. Extraction times between 15 and 90 min were investigated (Figure 3(c)), and the extraction yield increased as the extraction time increased. This could be because a longer time gave the ultrasound wave more time to disrupt the cell walls and release the cell contents. When the extraction time was <60 min, the extraction yields were lower than that with an extraction time of 60 min. With an extraction time of 90 min, the extraction yield increased modestly compared with that for 60 min. This could be because the extraction solution became saturated and large increases in the yield were not

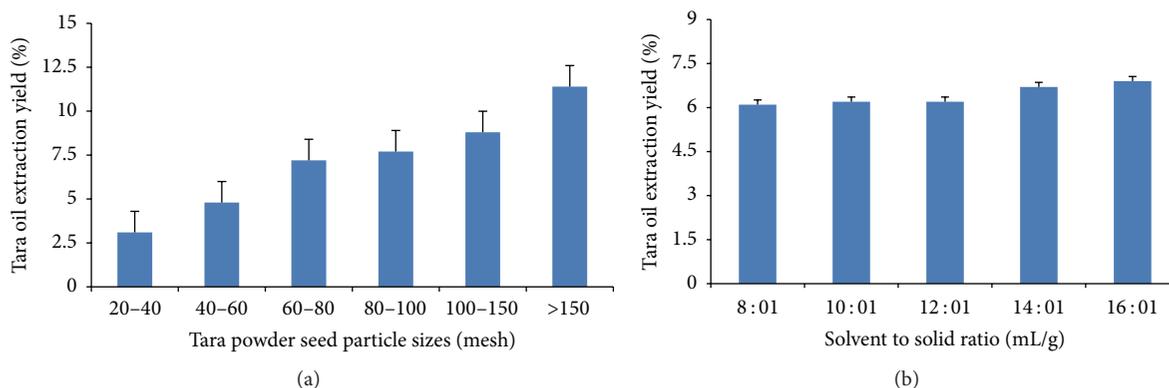


FIGURE 2: (a) Tara seed oil yields with different particle sizes. Samples of dried tara powder (10 g) ground to different sizes (20–40, 40–60, 60–80, 80–100, and >150 mesh) were extracted with petroleum ether. Ultrasonic extraction was performed for 45 min at 250 W and 30°C. (b) Tara seed oil yields with different liquid–solid ratios. Dry tara powder (10 g, 60–80 mesh) was extracted with different liquid–solid ratios (8 : 01, 10 : 01, 12 : 01, 14 : 01, and 16 : 01). Ultrasonic extraction was performed at 250 W and 30°C.

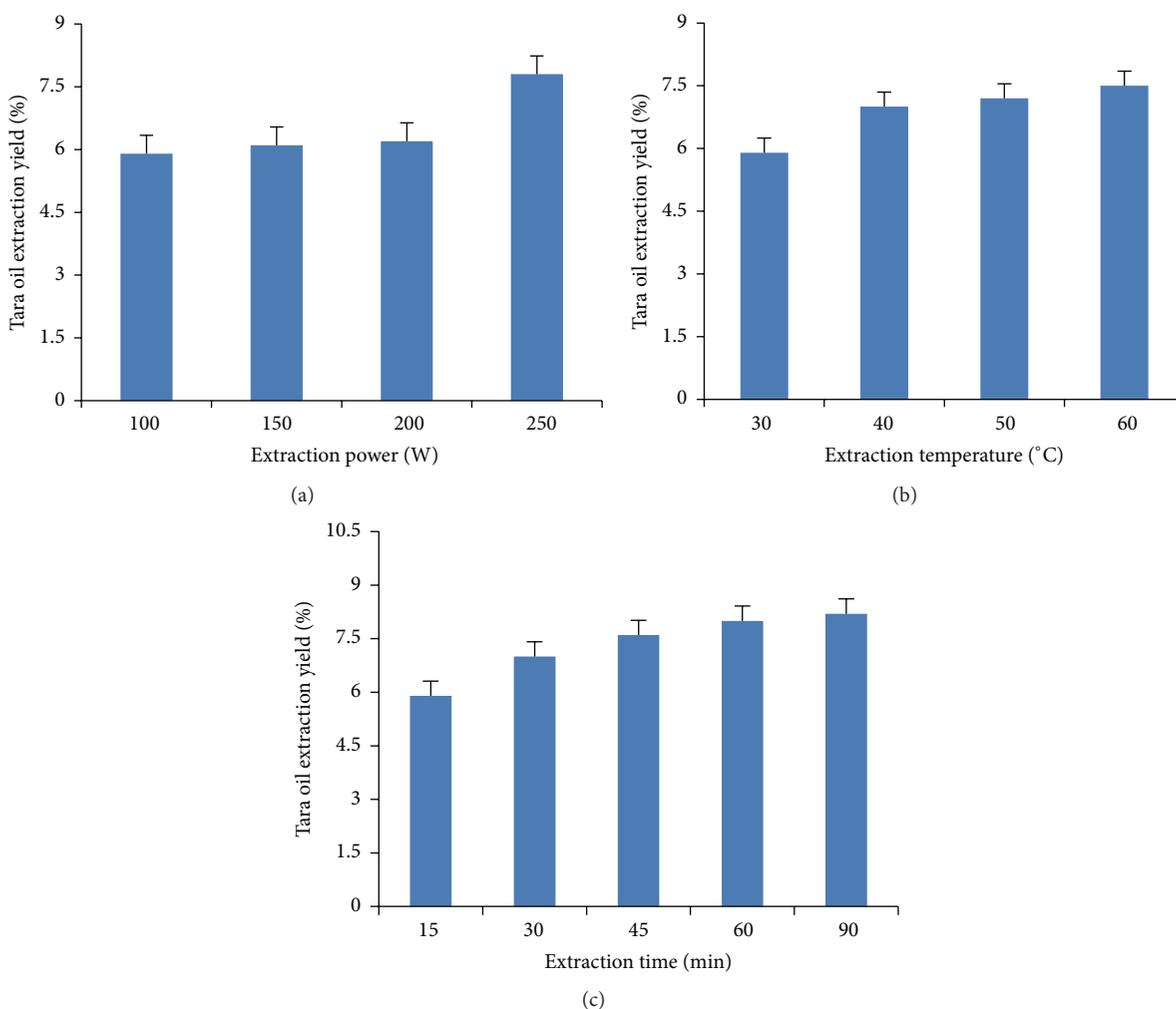


FIGURE 3: (a) Tara seed oil yields with different extraction times. Dry tara seed powder (10 g, 60–80 mesh) was extracted for 15, 30, 45, 60, or 90 min. Ultrasonic extraction was performed at 250 W and 30°C. (b) Tara seed oil yields with different ultrasonication powers. Dry tara powder (10 g, 60–80 mesh) was extracted using four different ultrasonication power settings (100, 150, 200, and 250 W). Ultrasonic extraction was performed for 45 min at 30°C. (c) Tara seed oil yields with different ultrasonication temperatures. Dry tara powder (10 g, 60–80 mesh) was extracted ultrasonically at 30°C, 40°C, 50°C, or 60°C. Ultrasonic extraction was performed at 250 W for 45 min.

TABLE 2: Percentage yield of tara seed oil.

Extraction method	Tara oil extraction yield (%)
Ultrasonic assisted extraction	10.53
Soxhlet extraction	9.40

TABLE 3: Fatty acid compositions (% of total fatty acids) of tara seed oils extracted by different methods.

Fatty acid	Ultrasonic assisted extraction (%)	Soxhlet extraction (%)
C16:1	9.03	11.69
C16:2	0.41	0.42
C18:1	3.01	3.93
C18:2	10.96	14.28
C18:3	69.94	62.32
C18:4	0.9	1.14
C20:1	0.47	0.52
C22:1	1.96	1.96

possible after saturation. Therefore, 60 min was selected as the optimum extraction time for subsequent experiments.

2.7. Comparison of Yields of Tara Seed Oil from Ultrasonic and Soxhlet Extraction. In Sections 2.1–2.6 the ultrasonic extraction was optimized as follows: extraction solvent (petroleum ether), particle size (60–80 mesh), extraction time (60 min), ultrasonic power (250 W), extraction temperature (60°C), and liquid–solid ratio (14 : 1).

The optimized ultrasonic extraction was compared with a traditional Soxhlet extraction performed as follows: extraction solvent (petroleum ether), particle size (60–80 mesh), extraction time (240 min), extraction temperature (90°C), and liquid–solid ratio (14 : 1).

A comparably higher yield of tara seed oil was obtained using the ultrasonic extraction (Table 2). This extraction method also used less organic solvent and was faster (60 min compared with 240 min).

2.8. Effect of Ultrasonic Extraction on the Fatty Acid Composition of Tara Seed Oil. Table 3 shows that the fatty acid composition of tara seed oil from ultrasonic extraction was similar to that from Soxhlet extraction.

3. Experimental Section

3.1. Chemicals and Materials. Tara seeds (wonderful variety) were collected from Yunnan province, China. The seeds were separated from the shell, cleaned, and dried at 60°C for 48 h. The dried seeds were ground to powder and then sieved (20–40, 40–60, 60–80, 80–100, 100–150, and >150 mesh). The dry powders were stored in desiccators at 4°C. Petroleum ether was purchased from Tianjin Chemical Reagents Co. (Tianjin, China). All other solvents and chemicals used in this experiment were of analytical grade and were purchased from Beijing Chemical Reagents Co. (Beijing, China).

3.2. Apparatus. The ultrasonic extraction experiments were carried out in a rectangular (23.5 cm × 13.3 cm × 10.2 cm) bath (KQ-250DB, Kunshan Ultrasonic Co. Ltd., Kunshan, China) with 50 kHz transducers annealed to the bottom. The power rating of the ultrasonic bath was 250 W, and power could be adjusted from 0 to 100%. The temperature was controlled by replacing the water in the bath. To examine the effect of ultrasonic power on the extraction yield, four power levels were compared: 100% (250 W), 80% (200 W), 60% (150 W), and 40% (100 W). A Soxhlet extraction apparatus was used to extract the samples by a traditional method for comparison purposes. A 1/10,000 analytical balance was used for weighing samples.

3.3. Tara Seed Ultrasonic Extraction. Fresh tara seeds were dried in an oven at 45°C until their weights were constant, and they were then ground into powders (20–40, 40–60, 60–80, 80–100, 100–150, and 150 mesh). The tara seed powders were dried in an oven at 60°C. The general method for extraction involved weighing tara seed powder (10 g) into a 250 mL flask. Then extraction solvent was added and the flask was placed in the ultrasonic bath. The following extraction conditions were investigated for optimization: extraction solvent, particle size in the tara seed powder, extraction time, ultrasonication power, extraction temperature, and liquid–solid ratio. After the extraction, the solution was reduced on a rotary evaporator (temperature 45°C, 0.07–0.08 MPa, and approximately 20 min), and the solvent was recovered. The residue was dried to a constant weight in a vacuum drier to obtain tara seed oil. The tara seed oil was weighed and the extraction yield was calculated.

3.4. Soxhlet Extraction. Tara seed powder (10 g, 60–80 mesh) was weighed into a 200 mL Soxhlet extractor. Petroleum ether was used as the extraction solvent at a 14 : 1 liquid–solid ratio. After extraction, the solution was filtered through a Buchner funnel and the filtrate was collected in a round-bottom flask. The filtrate was reduced under vacuum (RE-52AA rotary evaporator, Shanghai Ya Rong Biochemical Instrument Co. Ltd., Shanghai, China) to obtain a residue that contained tara seed oil. This residue was dried in a vacuum drier to remove any residual petroleum ether.

3.5. Yield Calculation. Each extraction was repeated three times and the results were averaged. The tara seed oil percentage yield was calculated as follows:

$$\text{Yield (\%)} = \frac{W_2 - W_1}{W_2} \times 100\%, \quad (1)$$

where W_2 is the initial weight of tara powder (g) used in the Soxhlet extraction and W_1 is the weight of the dried residual tara powder after the Soxhlet extraction (g).

3.6. Fatty Acid Composition of the Tara Seed Oil. The fatty acid compositions of the tara seed oils obtained by ultrasonic extraction and Soxhlet extraction were determined using gas chromatography after derivatization to fatty acid

methyl esters. Methyl ester derivatization of the oil was performed by saponification in 0.5 mol/L KOH solution with 40 mL of methanol added to each 4 g sample of oil. After derivatization the samples were analyzed using a gas chromatograph (6890N, Agilent Technologies, Santa Clara, CA, USA) equipped with a flame ionization detector. A capillary column HP-INNOWax (30 m × 0.25 mm × 0.25 μm; Agilent Technologies) was used for separation of the methyl esters. The column temperature was initially 150°C (held for 1 min) and then increased to 250°C at 5°C/min (held for 1 min). The carrier gas was helium (purity: 99.99%) at a flow rate of 40 mL/min. The injection volume was 1.0 μL, and the samples were injected in split mode with a split ratio of 10:1. The injector temperature was 250°C, the transfer line temperature was 280°C, and the ion source temperature was 250°C.

4. Conclusions

In the present study, ultrasonic extraction of tara seed oil was optimized. The yield of tara oil increased with increasing particle size in the tara seed powder, ultrasonication time, ultrasonication power, extraction temperature, and liquid–solid ratio. The optimized ultrasonic extraction method used petroleum ether as the extraction solvent, tara seed powder with a particle size of 60–80 mesh, an extraction time of 60 min, ultrasonication power of 250 W, extraction temperature of 60°C, and liquid–solid ratio of 14:1. The extraction yields achieved with the optimized ultrasonic extraction were similar to those from traditional Soxhlet extraction. Gas chromatography analysis showed that the fatty acid composition of the tara seed oil obtained by ultrasonic extraction was similar to that of oil obtained by Soxhlet extraction. Ultrasonic extraction is a useful and environmentally friendly extraction method that could be applied to the production of other plant oils and active substances.

Competing Interests

The authors declare no conflict of interests.

Authors' Contributions

Zhan-jun Li and Lei Yang contributed equally to performing the research, analyzing the data, and writing the manuscript. Feng-jian Yang and Yuan-Gang Zu approved the final manuscript.

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