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Advance in Ultrasound-Assisted Extraction of Edible Oils: A Review

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ABSTRACT

Ultrasonic waves cause an increase in oil extraction efficiency through their mechanical effects, cavitation, and the energy generated by this phenomenon. Temperature, time, solvent to sample ratio, solvent type, and ultrasonic power are the most effective factors in the ultrasound-assisted extraction of oils. Temperature increase reduces the surface tension and vapor pressure of the solvent, leading to a rise in the solvent diffusion into the cell, and thus, the extraction efficiency increases. However, as the temperature approaches the solvent boiling point, its vapor pressure rises. Due to the smaller pressure difference between the interior and exterior of the cavitation bubbles, they collapse less intensively and cause the extraction efficiency to decrease. The increase in ultrasonic power has a similar effect on extraction efficiency. An elevation of the extraction time improves this parameter in the early minutes of extraction, by destroying the cell walls and making the oil diffuse out of the cell-matrix. An increase in the solvent to sample ratio up to an optimal level elevates the concentration gradient of the solvent towards the sample matrix by reducing the solution viscosity and brings about an improvement in the extraction efficiency. The application of the solvents with high vapor pressures results in a decrease in the cavitation energy because of the pressure difference between the cell interior and exterior. Therefore, such solvents do not have a positive effect on the extraction efficiency. Each of the aforementioned factors has its optimum level to enhance the ultrasound-assisted extraction efficiency.

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Introduction

Edible oils are water-insoluble compounds with plant or animal origins [1]. Such oils and fats have drawn scientists` and investors` attention from ancient times for both their health-promoting effects and commercial uses [2] .Various techniques are applied for oil extraction, among which the solvent and mechanical (cold or hot press) extraction methods are the conventional ones. Since the extraction method considerably influences the oil quality, the selection of the most appropriate method is of great importance [3]. The disadvantages of the conventional extraction techniques such as oil degradation caused by thermal stresses and high pressures, which leads to higher solvent consumption and longer extraction time [4], in addition to the environmental issues due to the consumption of organic solvents and the presence of their residues in the final product, have had researchers pay attention to the application of novel non-thermal techniques for oil extraction. Ultrasound-assisted extraction (UAE), microwave-assisted extraction (MAE), and subcritical and supercritical fluid extraction (SFE) are some of such methods (figure 1). These methods are employed as an assistant in the extraction process accompanied by safe organic solvents. Moreover, they have enormous potential for the reduction or removal of toxic chemical solvents and the enhancement of the extraction efficiency through improving the cell membrane permeability and the extract quality. Such methods are also known as cold extraction as they do not use high temperatures during the process [5].

UAE is one of the most significant methods for the extraction of raw materials from natural products, and valuable substances from herbal sources. In this method, the solvent diffuses better into the plants and disrupts their cell wall; thus, the extraction efficiency is improved [6,7]. Ultrasound is known as an efficient extraction method whose optimization is easier than that of other techniques, due to its few affecting parameters (matrix moisture content, solvent

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type, and time). Furthermore, it has some advantages:
Cheaper extraction, mass transfer improvement, better solvent diffusion, less dependence on the solvent (lower solvent consumption), extraction at low temperatures, faster extraction and shorter extraction time, more purity of the extract, higher and more reproducible extraction efficiency, lower fossil energy consumption, expansion of the number of the solvents used, selective extraction, application of small-sized equipment, having safety and health benefits and being environmentally friendly [6–10].

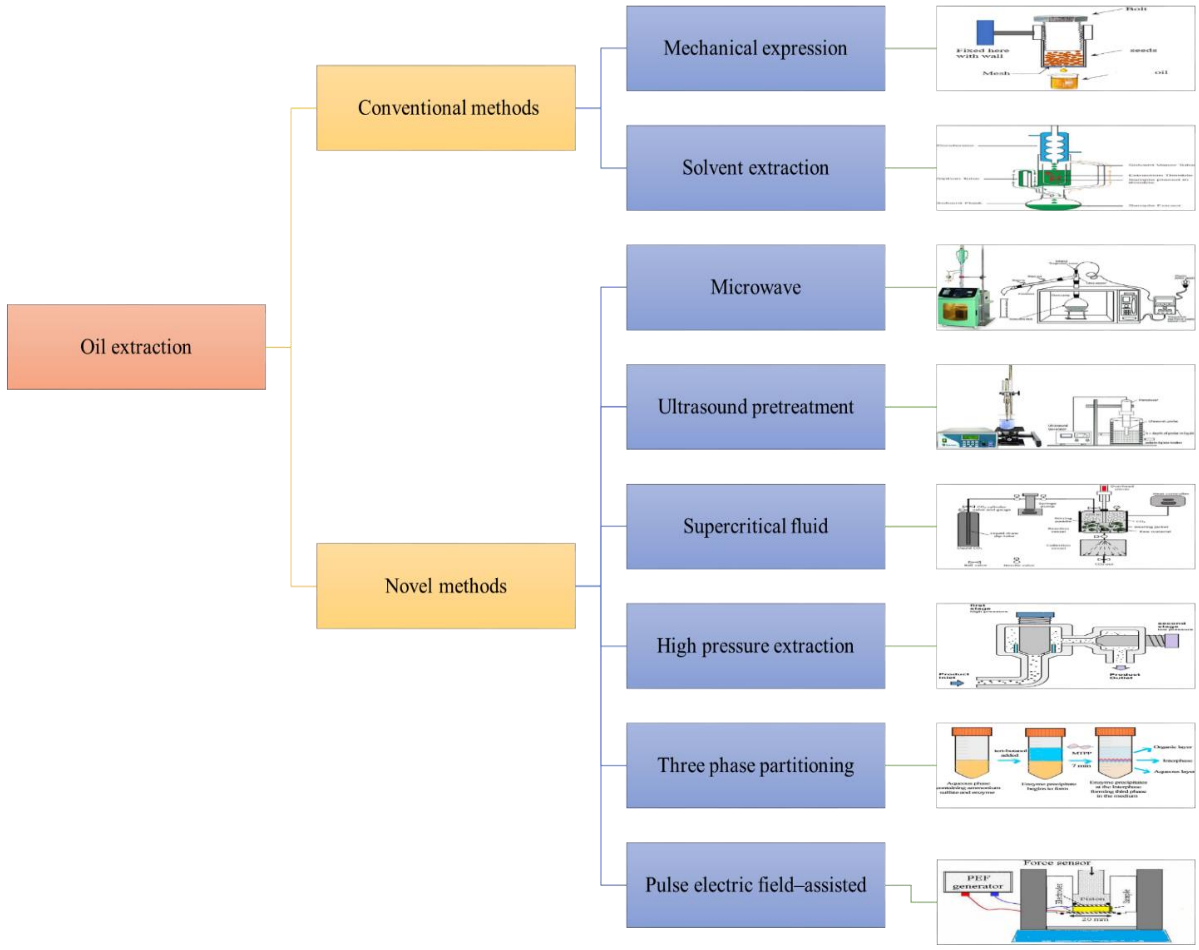

Figure1. Ideal scheme of various oil extraction methods

In addition to the benefits mentioned, as reported in several studies, the qualitative properties of the UAE extracted oils are comparable with those extracted by conventional methods. A study showed not only did UAE significantly reduce the extraction time of the essential oil of orange peel but also the organoleptic properties of this oil were similar to those extracted using the conventional method [11]. Some other studies that examined the UAE of ginseng saponins from ginseng roots, [12] the essential oils of peppermint, lavender, garlic, and citrus declared that the use of low-temperature and short-time extraction could maintain and improve the quality of heat-unstable compounds (such as saponins) because low temperatures reduce heat degradation in the product [13–16]. In various studies, no significant differences were observed in the fatty acid profiles of the canola, soybean, sunflower, and cottonseed oils extracted by both UAE and hexane[17]. The quality characteristics of the pepper seed oil extracted by UAE and conventional solvent extraction were compared. The ultrasonic bath method was carried out with the conditions of 50 °C, 50 minutes, and 200W, and the conventional solvent extraction was conducted with the circumstances of 50 ml of hexane, 50°C, and 6 hours. The oil extracted by UAE had more transparency, lower refractive index, lower free fatty acid content, lower peroxide value, and lower iodine value compared with the solvent-extracted one, but the fatty acids of both oils were similar [18]. Similarly, in another study, the physicochemical properties of the avocado oil extracted by the UAE and the conventional solvent extraction were compared. The results indicated that the UAE-extracted oil had a higher iodine value than the conventionally-extracted, while its melting point, free fatty acid content, color compounds were lower. The high temperature used in the solvent extraction method inactivates the bioactive compounds. Chemical residues, harmful to human health, may also be present in the oils extracted with organic solvents [19].

Based on the amount of the energy produced, ultrasonic power, ultrasonic intensity or the ultrasonic energy density, applications of ultrasound are divided into two groups, namely high power (low frequency: 16-100 kHz and high intensity: 10-1000 w/cm^2) and low power (high frequency: 100 kHz-1 MHz and low intensity: <1 w/cm^2). The high power ultrasound is used to change the physicochemical properties of foods, and to accelerate and improve sample preparation, including the preparation of a liquid sample to assist the liquid-liquid extraction, homogenization, and emulsification. The low power ultrasound is exploited for the analysis of the physicochemical properties (acidity, sugar content, etc.) of food products [6].

Ultrasonic waves are the mechanical waves which are propagated in elastic media. The wave frequency is the reason behind the difference between sound and ultrasound as the sonic wave frequencies are within the human hearing range (16 Hz to 20 kHz) and the ultrasonic wave frequencies are higher than the human hearing range and lower than microwave (20 kHz to 10 MHz). The physical effects of ultrasound are produced in the frequency range of 20-100 kHz, while its chemical effects are produced within the range of 200-500 kHz [6,9]. Considering the widespread applications of ultrasound in oil extraction, no comprehensive investigation has ever been conducted on the influential factors of UAE such as solvent type, extraction time and temperature, and ultrasonic power. Consequently, this review article intends to examine the effective factors that have been performed for the extraction of edible oils by ultrasound.

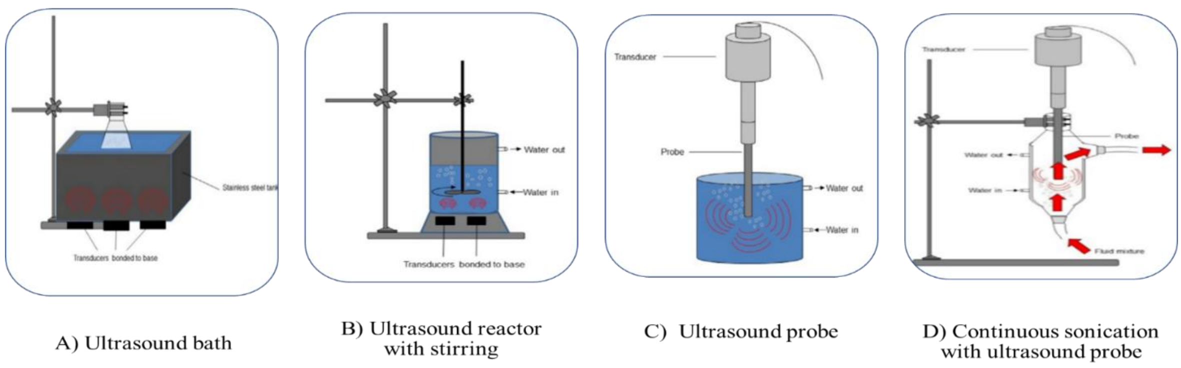

Figure2. Commonly used ultrasonic systems
A) Ultrasound bath, B) Ultrasound bath equipped with a stirrer, C) Ultrasound probe, and D) Ultrasound probe for continuous sonication [9].

UAE systems

Ultrasonic bath and probe are the commonly used systems used for UAE. The bath has a stainless steel wall. Its frequency is almost 40 kHz and its temperature can be controlled. The more modern types of ultrasonic baths have been designed, which are equipped with a stirrer and water can be circulated inside them and their frequency is approximately 25 kHz [9]. The probe system is stronger because of generating more intensive waves. Its frequency is almost equal to 20 kHz and its temperature is not usually controlled [20,21]. Given that the probe is mostly made of nickel, the metal ions may migrate from the probe to the oil and oxidize it. A newer type of this system has been designed which has a glass chamber in which the extraction process can be continuously controlled [figure 2) [9].

Ultrasound mechanism

Cavitation is the principal mechanism of UAE, during which micro-bubbles are formed in the liquid, grow rapidly to a critical size, and finally collapse. This releases a large amount of energy which is exerted on its surroundings in the form of shear stress. Also, the collapse brings about the creation of intense radial vibrations through which mass transfer is enhanced. It should be noted that different studies have revealed that the bubbles collapse asymmetrically around solid particles so that an extremely rapid flow is directed from the liquid toward the particle surfaces. The collision of these micro-jets with the surfaces causes the breakdown and destruction of cell walls. Solvent extraction includes the immersion of the plant tissue in the solvent (for solvent absorption), the tissue swelling, and finally, the transport of the solutes from the tissue into the solvent through diffusion or osmosis. Ultrasonic waves facilitate and accelerate the tissue swelling and the solute transport by creating pores in the cell walls (Figure 3 and 4) [6,22].

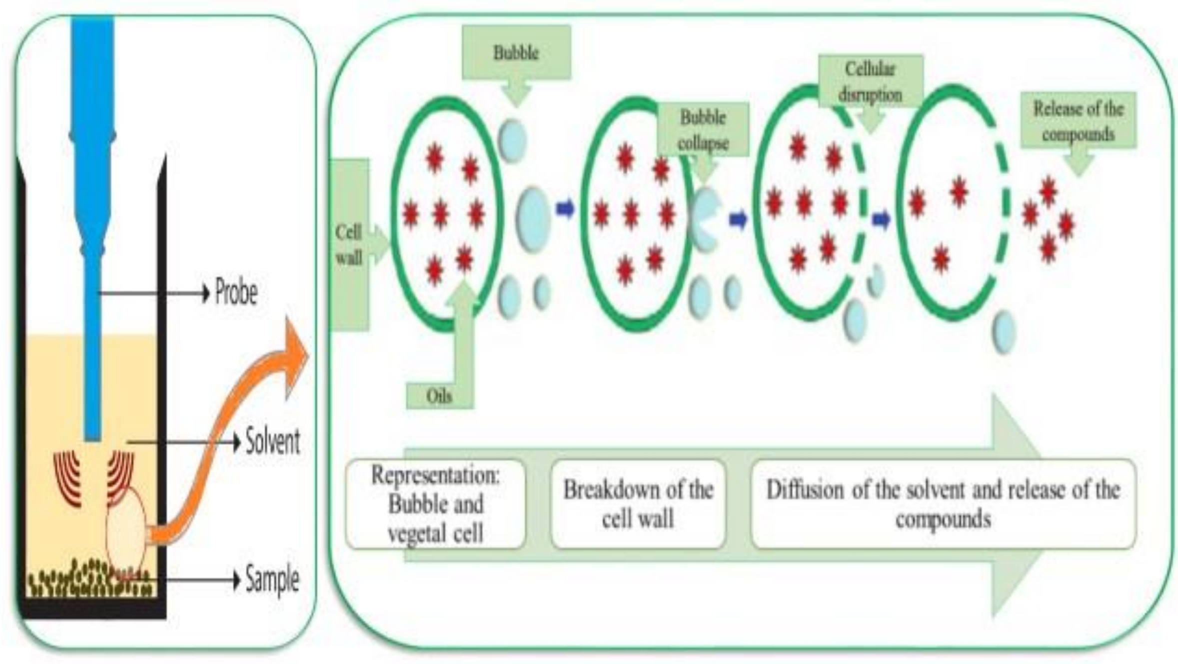

Figure 3. The principle of acoustic cavitation. Modified from Soria and Villamiel

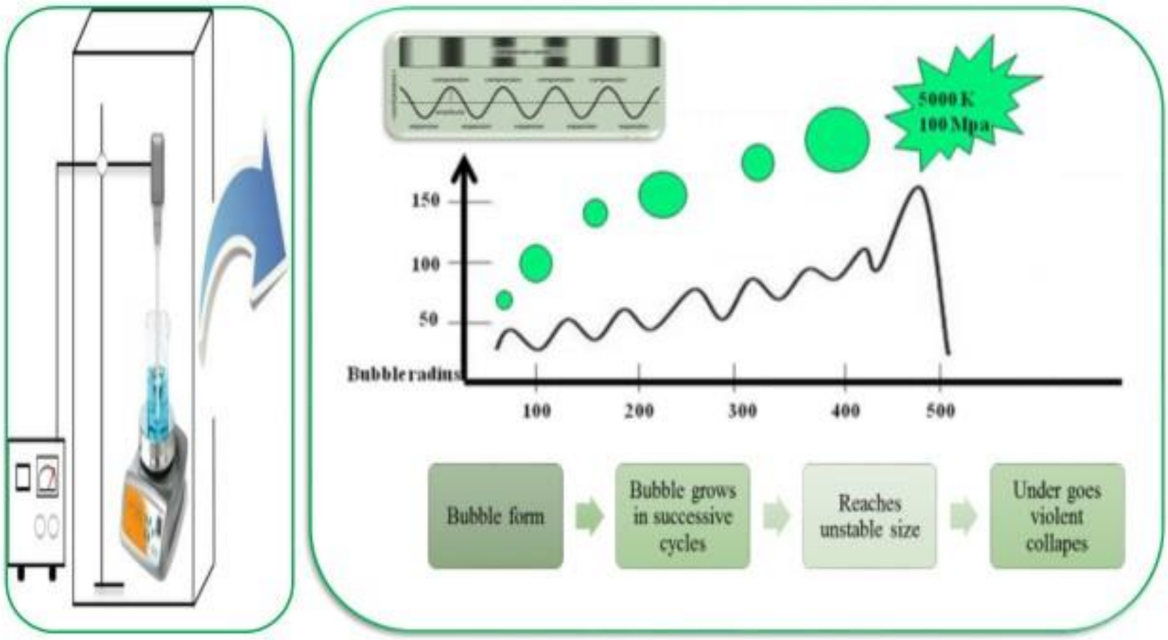

Figure 4. Graphical representation of cavitation-bubble collapsing and releasing plant material in three steps.

The sequential vibration created in the fluid by the ultrasonic waves cause shear stress and reduces viscosity in the fluid. At the same time, such waves influence the interface between the fluid and the cell wall, which leads to the rupture and breakdown of the wall. This results in an increase in temperature and thus, a decrease in viscosity. With a reduction in the fluid viscosity, it further absorbs the energy generated by the mechanical effects of the waves. The absorbed energy makes the cavitation bubbles grow and creates a balance between the gas inside the bubbles and the liquid outside. The bubbles gradually grow to their critical size by absorbing the ultrasonic energy; collapse during compression and release a large amount of energy which is exerted on the surrounding particles in the form of shear stress. The bubble collapse is asymmetrical in the vicinity of the solid particles and creates a very fast flow from the liquid to the particle surfaces. When the cavitation bubbles collapse on the particle surfaces, the high heat and pressure released, directly produce micro-jets and shock waves on the solid surfaces. The collision of these micro-jets with the surfaces causes the disruption and breakdown of cells and consequently, mass transfer is improved [22–24]. In general, ultrasonic waves produce large amounts of energy and pressure through the mechanical effects of cavitation bubbles collapse, thus leading to the thinning of the cell membranes. As a result, the penetration of the solvent into the cell and the extraction of the target samples from the sample matrix to the solvent are enhanced. Additionally, the bubbles collapse causes turbulence in the microscopic scale, and strong collisions between particles and their stimulation, thus facilitating and accelerating mass transfer. Accordingly, the UAE enhances the extraction efficiency and decreases solvent consumption [9,20,21]. The equipment used in the UAE is simpler than that of MAE and SFE, thereby lowering the operational costs. In contrast to MAE and SFE, a variety of organic solvents could be used in UAE [25].

Effective factors in the UAE

Sample nature, its components, temperature, time, solvent to sample ratio, solvent type, solvent viscosity, solvent vapor pressure, and ultrasonic power are the main factors influencing the UAE of oils [9,26].

Solvent type

The solvent type influences the UAE yield [17]. The choice of solvent depends on the viscosity, surface tension, and vapor pressure of the solvent, which affect the cavitation and its threshold. Increasing the viscosity and /or surface tension, increases give to the cavitation threshold and resistance of the sample against the movement of the device. Therefore, the intensity of the vibrations must increase to create cavities. Solvent vapor pressure depends on the temperature of the liquid and low vapor pressure of the solvent causes the cavitation bubbles to collapse more sharply [27]. The solubility of a solute in a solvent remarkably depends on the nature and strength of the solute-solute, solvent-solute, and solvent-solvent interactions. A general principle says: “like dissolves like”. Therefore, non-polar solvents extract non-polar substances and similarly, polar solvents extract polar compounds. Based on their polarity, solvents are divided into the three groups of polar, semi-polar, and Non-polar. Considering the non-polarity of oils, non-polar solvents have the highest efficiency in oil extraction. The higher the polarity of a solvent, the lower the oil extraction efficiency. N-hexane and petroleum ether are included in the non-polar solvents which have the highest efficiency in oil extraction. In such solvents, only the weak Van der Waals forces exist between the molecules which are why they are volatile and have very low boiling points. Similarly, only the Van der Waals forces exist between the non-polar solute molecules. Therefore, all the molecules of the solution are affected by this type of force, which makes the solution formation possible [28]. In a study carried out on the UAE of pomegranate seed oil, the effects of different solvents were investigated on the oil extraction efficiency. The highest efficiency belonged to petroleum ether followed by hexane, ethyl acetate, diethyl ether, acetone, and isopropanol [29]. In another work that examined the UAE of safflower, n-hexane had the most dramatic effect on increasing the extraction efficiency, followed by ethyl acetate, petroleum ether, and ethanol [24]. In the UAE of Tobacco oil, hexane was more effective than petroleum ether [30]. In the UAE of rapeseed oil using hexane, isopropanol, and ethanol, the highest extraction efficiencies were associated with hexane and isopropanol respectively, whereas ethanol was not successful in extracting this oil. N-hexane is less polar than isopropanol; however, ethanol is a semi-polar solvent and not suitable for oil extraction[31]. The application of mixed solvents elevates UAE efficiency. In the studies conducted on the UAE of rapeseed and canola oils using n-hexane and the mixture of n-hexane (non-polar) and isopropanol (polar), the results showed that the mixture of the solvents enhanced the extraction efficiency of the oils, because polar lipophilic compounds (e.g., phospholipids and phosphoproteins) had more tendency to mixed solvents, because of the reactions among phenolic compounds and hydroxyl radicals during cavitation [17,32]. In the UAE of sunflower oil with hexane and the mixture of ethanol and hexane, the extraction efficiency of hexane was higher than that of the mixture. At the same time, the oil extracted by the solvents mixture contained a high content of wax compounds that were extracted by ethanol. It was reported in this paper that ethanol was no appropriate for oil extraction [7].

Solvent to sample ratio

As the solvent to sample ratio rises, oil extraction efficiency is increased, too. At high ratios, the decrease in the solution viscosity results in an increase in the solvent diffusion into the sample matrix, and the solvent concentration gradient increases toward the matrix and mass transfer increases. Consequently, the extraction efficiency is improved [17,26,33] with an elevation of the solvent concentration, due to more driving force and high osmotic pressure, the concentration difference between the interior of the sample and the surrounding medium increases, and based on the diffusion law, the oil migrates from the sample matrix (high concentration) to the solvent (low concentration) and the extraction efficiency is enhanced [17]. An increase in the solvent to sample ratio raises the concentration gradient, changes the driving force, and gives rise to the extraction efficiency. A rise in the ratio up to an optimal level alters the driving force and increases the extraction efficiency [29]. The optimum solvent to sample ratio has been reported to be equal to 4 to 1 in the UAE of grapeseed oil using petroleum ether [34], 10 to 1 in UAE of tomato seed oil using hexane [35], 7 to 1 in the UAE of perilla oil using hexane [36], 12 to 1 in the UAE of chia oil using ethyl acetate [37], and 12 to 1 in the UAE of canola oil using hexane [17].

Table1. Effects of the solvent type and the physicochemical properties of the seed oils whose oil has been extracted using UAE.

Effect on oil quality	Solvent type	Frequency (kHz)	Optimal sound process temperature (° C)	Optimal audio process time (minutes)	Type of ultrasound device	Type of oil	Study
Increasing the number of polyphenols and antioxidant activity of oil compared to extraction without applying the ultrasound process	Petroleum ether	40	25 to 35	60	Ultrasonic bath	Grapeseed	[34]
No change in fatty acid profile compared to extraction without the ultrasound process	Hexane	38	60	60	Ultrasonic bath	Tomato seeds	[35]
No change in fatty acid profile compared to extraction without the ultrasound process	Hexane & Isopropanol	20	-	0 - 180	Probe system	Soybeans	[32]
No change in fatty acid profile compared to extraction without the ultrasound process	Ethyl acetate	40	50	40	Ultrasonic bath	chia seed	[37]
-	Hexane	50	41	17	Probe system	Seed of Perilla	[36]
No change in fatty acid profile compared to extraction without the ultrasound process	Hexane Petroleum ether	40	25	60	Ultrasonic bath	Tobacco leaves	[30]
No change in fatty acid profile compared to extraction without the ultrasound process	Petroleum ether	-	40	30	Probe system	Pomegranate seed	[29]
No change in fatty acid profile compared to extraction without the ultrasound process	Hexane	24	30	30	Probe system	Tea leaf	[38]
-	Hexane	-	30	30	Probe system	safflower seed	[24]
No change in fatty acid profile compared to extraction without the ultrasound process	Hexane	20	30	30	Probe system	Flaxseed	[10]
The lower acid number and a lighter color than extraction without applying the ultrasound process	Hexane	20	-	70	Ultrasonic bath	Rice bran	[39]

Effect on oil quality	Solvent type	Frequency (kHz)	Optimal sound process temperature (° C)	Optimal audio process time (minutes)	Type of ultrasound device	Type of oil	Study
Increase in the amount of peroxide compared to extraction without applying the ultrasound process	Hexane	40	62/5	38/5	Ultrasonic bath	Papaya	[40]
-	Hexane	40	51/7	43/9	Ultrasonic bath	Apricot kernels	[20]
No change in peroxide number, conjugate number, and anizidine number and increase in the amount of tocopherol and tocotrinol compared to extraction without applying the ultrasound process	Hexane	30	50	77/5	Probe system	Kolkhoung (Pistacia khinjuk) kernels	[33]

- No result has been reported in the manuscript

Extraction time

As the extraction time increases, the cell wall is disrupted under the influence of the cavitation energy, so the contact area between the disrupted cell walls and the solvent increases and the oil diffuses a lot more into the solvent. At a constant solvent-to-sample ratio, the elevation of the UAE time from 40 to 72 min can promote the extraction yield of oil from 23.46 to 26.71%. With further increasing the extraction time, the oil concentration reaches a balance between the sample matrix and the solvent, and hence, the oil diffusion into the solvent decreases, and the extraction efficiency is lowered [17,24]. The other reasons for the reduction in the extraction efficiency at long extraction times could be the diffusion of some impurities such as insoluble compounds into the extract and the decrease in the solvent diffusivity into the sample matrix. Additionally, according to the diffusion law, due to the reduction in the oil concentration in the matrix, the oil will be absorbed again in the destructed tissues of the sample, and the extraction efficiency is decreased [29]. As well as the free radicals, peroxide value, and the decomposition of phenolic compounds might be reinforced with temperature increase, in some oils [17]. An increase in the UAE time of flaxseed oil up to 15 min did not affect the extraction efficiency. However, a gradual increase was observed in the efficiency from 15 to 30 min. At longer durations, no significant rise was seen in the oil extraction efficiency [10]. In the UAE of pomegranate seed oil, as the extraction time was elevated to 30 min, the extraction efficiency increased, too [29]. In the UAE of tea seed oil, the extraction efficiency increased with an elevation of the time up to 30 min. Nevertheless, it subsequently decreased to 120 min [38]. The extraction duration longer than 70 min caused a reduction in the UAE efficiency of rice bran oil [39].

Extraction temperature

Temperature is one of the effective factors in a solvent [27]. As temperature increases, the number of cavitation bubbles raises leading to an increase in the contact area between the solvent and the cell walls and a decrease in the solvent surface tension and viscosity. The less viscous solvent is abler to diffuse into the sample matrix; therefore, the extraction efficiency is improved with an enhancement in mass transfer. Even a constant solvent-to-sample ratio, elevation of temperature from 40 to 50 °C promoted the extraction yield of oil from 18 to 20% [17,29]. Temperature increase up to an optimal level is suitable for improving the extraction efficiency. As the process temperature approaches the solvent boiling point, the solvent vapor pressure is elevated and more bubbles are formed. Nonetheless, owing to the lower pressure difference between the interior and exterior of the formed bubbles, the power of the bubbles collapse, and the generated energy decrease, and the extraction efficiency is reduced at higher temperatures. The other reason for such reduction is the excessive decrease in the solvent surface tension which harms the extraction efficiency [10]. It should be noted that temperature rise, at a constant time, likely increases the peroxide value [17]. Also, the choice of extraction temperature is important to prevent the destruction of heat-sensitive compounds. Although, the temperature rise leads to a higher extraction yield the majority of studies have considered low temperatures to be suitable for UAE [27]. In the UAE of apricot kernel oil, the process temperature ranged from 30 to 60°C and the highest efficiency was obtained at 44°C [20]. The UAE temperature of papaya oil increased from 25 to 50°C, and the optimum temperature was found to be 38.5°C [40]. In the UAE of perilla oil, the temperature was elevated from 20 to 60°C and the optimum temperature was reported to be 40°C above which the extraction efficiency was lowered [36]. The optimum UAE temperature of Kolkhoung (Pistacia khinjuk) kernel oil was equal to 50°C [33] and in the UAE of canola, the temperature rise of 40 to 50°C caused the peroxide value to increase from 1.5 to 1.8 (meqg $O2$ kg^{-1}) [17].

Ultrasonic power

During sonication, cavitation bubbles are subject to vibration, growth, and collapse. These physical and mechanical effects cause an increase in the contact area and oil extraction [41]. Ultrasonic power impacts the occurrence of cavitation. An increase in this feature promotes efficiency. As this feature is increased, the process temperature is increased, too. At a constant process time, the solvent viscosity and surface tension decrease, while its vapor pressure rises. The cavitation threshold is lowered at high temperatures, which is advantageous to the formation of bubbles. Consequently, the extraction efficiency is improved. Considering the rise in the solvent vapor pressure and the processing time as well as the negative effects of these parameters at high temperatures, an increase in the ultrasonic power up to an optimum level positively influences the extraction efficiency, and at higher levels, as the cavitation energy is reduced, the extraction efficiency is lowered, too [24,27]. At a constant UAE time of 15 min, the extraction efficiency of perilla oil firstly increased when the ultrasonic power was raised from 300 to 400 w, whereas it decreased subsequently as the power rose to 500 w [36]. In the UAE of pomegranate seed oil, the rise in the ultrasonic power from 80 to 160 w caused an increase in the extraction efficiency at a constant duration of 30 min. However, the power of 200 w slightly lowered efficiency [29]. The optimum ultrasonic power was reported to be 400 w at 60 min to enhance the grapeseed oil UAE efficiency [34]. The ultrasonic power of 300 w was effective in the UAE of safflower oil and increased its efficiency, while higher powers reduced the extraction efficiency [24]. Ultrasonic frequency affects the size of cavitation bubbles and their resistance to mass transfer. Generation and intensity of cavitation in a liquid may decrease at high ultrasonic frequencies because it is difficult for the acoustic cavitation and greater intensity required to produce cavitation bubbles [27]. Also, with a rise in time, temperature, and ultrasonic frequency, the cavitation intensity will be higher, which may accelerate the oxidation of some of the extracted oil such as peanut oil which is extracted at ultrasonic frequencies up to 40 kHz[41].

Conclusion

Ultrasound is one of the novel non-thermal methods of extracting edible oils through which the efficiency of oil extraction from plant tissues is raised. To optimize this extraction method; The parameters of time and solvent - to - sample ratio should be such that the largest amount of oil and the least amount of impurities would be extracted. If the contact time of the solvent with the plant tissues containing the oil exceeds its optimum, impurities and insoluble compounds also enter the solvent and the oil is re-absorbed into the tissue according to the law of diffusion and the difference in the concentration of inside and outside the plant tissue, as a result, the extraction efficiency is reduced. The solvent - to - sample ratio should be to the extent that increases the gradient of the solvent concentration towards the sample matrix to elevate the oil extraction efficiency. The ultrasonic power should be such that it does not cause a great increase in temperature and decrease in the extraction efficiency because increasing the temperature reduces the surface tension and vapor pressure of the solvent as long as it is not close to the boiling point of the solvent. The solvent vapor pressure is raised when the temperature rises so high that it approaches the boiling point of the solvent and the pressure difference inside and outside the cavitation bubbles is reduced. Thus, less energy is produced following the lower intensity of the bubble burst, and consequently, solvent diffusivity and oil extraction are reduced.

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Conflict of interests

Authors have not conflicted of interest.

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