Recent advances in applications of power ultrasound for petroleum industry

Xiaoming Luo, Haiyang Gong, Ziling He, Peng Zhang, Limin He

Shandong Key Laboratory of Oil & Gas Storage and Transportation Safety, China University of Petroleum (East China), Qingdao 266580, China
Surface Engineering Pilot Test Center, China National Petroleum Corporation, Daqing 163453, China

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ABSTRACT

Power ultrasound, as an emerging green technology has received increasing attention of the petroleum industry. The physical and chemical effects of the periodic oscillation and implosion of acoustic cavitation bubbles can be employed to perform a variety of functions. Herein, the mechanisms and effects of acoustic cavitation are presented. In addition, the applications of power ultrasound in the petroleum industry are discussed in detail, including enhanced oil recovery, oil sand extraction, demulsification, viscosity reduction, oily wastewater treatment and oily sludge treatment. From the perspective of industrial background, key issue and resolution mechanism, current applications and future development of power ultrasound are discussed. In addition, the effects of acoustic parameters on treatment efficiency, such as frequency, acoustic intensity and treatment time are analyzed. Finally, the challenges and outlook for industrial application of power ultrasound are discussed.

1. Introduction

As the cornerstone of industrial society, fossil energy still plays a major role in the world energy consumption [1]. It accounts for 65% of total production capacity, and this trend will likely continue in the next 100 years [2]. However, the petroleum industry is presented with various challenges due to the rapid exploitation of fossil energy, among which are the following: economic exploitation of remaining crude oil, reclamation of unconventional energy sources, dehydration and viscosity reduction of crude oil, and harmless treatment of oily waste.

In the middle and late stages of production, a considerable amount of crude oil remains in the reservoirs and cannot be driven out by naturally or by water flooding [3]. This oil has not only high viscosity but also high absorbability to rock, showing poor fluidity. In order to achieve high oil field, developing green economic enhanced oil recovery (EOR) technologies is a practical and challenging topic. In addition to accelerating the exploitation of conventional energy sources, many countries have gradually turned their attention to the utilization of unconventional energy sources to meet the growing energy consumption. Among them, oil sand has been attracting great interests of the petroleum industry due to its large reserves, efficient distribution and easy mining [4]. However, the rapid exploitation of oil sand brings many problems to subsequent extraction, including overuse of surfactants and water pollution. High energy consumption in crude oil transportation and distillation is also a significant drawback. In order to minimize energy waste and security risks, appropriate technologies are required to dehydrate crude oil and reduce its viscosity [5,6]. In addition, a great amount of waste containing high concentration of petroleum hydrocarbons (PHCs) is produced through oil production, storage, transportation and refining. Highly toxic waste not only causes irreparable pollution to the environment but also poses threat to human health, and is not legally allowed to be discharged without treatment. Increasing use of fossil fuels leads to the increase of oily waste products, thus imposing a burden on treatment facilities [7].

Ultrasound shows great promise for both upstream and downstream of the petroleum industry due to its cleanliness, low cost and high efficiency [8]. Especially, power ultrasound (20–100 kHz) not only has favorable penetration in oil/water media [9], but can also generate and transmit high energy specific density (10–1000 W·cm⁻²) [10]. When the cavitation bubble implodes, it induces high temperature of about 5000 K and high pressure of about 2000 atmospheres, accompanied with shock waves and micro-jets. Such cavitation effects can break long chains of oil molecules, thus improving crude oil quality. In addition, the ultrasonic technique meets the requirements of sustainable development since it operates under moderate conditions and produces no volatile organic chemicals (VOCs). However, complicated physical and chemical characteristics of acoustic cavitation hinder our understanding of the mechanism of power ultrasound for various applications, which is required in order to optimize the operating parameters and increase efficiency. Nonetheless, few reviews comprehensively discuss the mechanism and applications of acoustic cavitation for petroleum industry.

* Corresponding author.
E-mail address: upclxm@163.com (X. Luo).

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This review presents the physical and chemical effects of acoustic cavitation from the perspective of bubble dynamics. In addition, the applications of power ultrasound for petroleum industry are discussed in detail including enhanced oil recovery, oil sand extraction, demulsification, viscosity reduction, oily wastewater treatment and oily sludge treatment. Finally, we discuss the outlook for the industrialization of power ultrasound.

2. Acoustic cavitation characteristics

2.1. Acoustic cavitation

2.1.1. Acoustic cavitation and cavitation threshold

Ultrasound is a mechanical wave with pressure oscillation which can travel through gaseous, liquid and solid media [10]. Especially, power ultrasound (20–100 kHz) can transmit high specific energy density to the medium and cause intense acoustic cavitation, which then results in high temperatures and pressures, as well as mechanical effects (shock waves, micro-jets and shear force). These effects are attributed to the evolution of cavitation bubbles. Under ultrasonic irradiation, the medium undergoes a cycle of pressure expansion and compression. When negative pressure overcomes the cohesive force between fluid particulates, cavitation bubbles appear as nuclei in the medium. Cavitation bubbles form in three ways: (i) single bubble, (ii) bubble chains and (iii) small bubbles that spread from existing bubbles. In order to cause acoustic cavitation, the acoustic intensity must reach a certain value, known as the cavitation threshold. The increasing frequency can lead to a higher acoustic cavitation threshold, indicating that acoustic cavitation tends to occur at low frequency. The cavitation threshold is highly dependent on medium properties, such as viscosity, interfacial tension and vapor pressure [11]. In addition, the cavitation threshold is sensitive to dissolved gas in the medium. Microbubbles in fluid facilitate the nucleation of cavitation bubbles, which significantly lowers cavitation threshold.

2.1.2. Bubble dynamics

The physical and chemical effects of acoustic cavitation are determined by bubble dynamics. As shown in Fig. 1, the evolution of a cavitation bubble in the oscillating pressure field has two stages: expansion and collapse. When the bubble reaches unstable size, it implodes and causes a series of physical and chemical phenomena. The radial dynamics of the bubble is described by the Keller-Miksis equation [12]:

\[
(1 - \dot{M})R \ddot{R} + \frac{3}{2} \left(1 - \frac{M}{3}\right) \dot{R}^2 = \frac{1 + M}{\rho_0} + \frac{R}{\rho_0 c} \int \left[ P_B - P_0 - P_0 \right] \, dt
\]

where \( M \) is the Mach number that is equivalent to \( R/c \); \( R \) is the bubble radius; \( c \) is the sound velocity; \( \rho_0 \) is the liquid density; \( P_0 \) is the pressure in the bubble; \( P_0 \) is the static hydraulic pressure; \( P_B \) is the acoustic pressure amplitude.

During negative pressure phase, the nucleated bubble expands and reaches maximum radius \( R_{\text{max}} \) at the end of the expansion. The temperature inside the expanding bubble remains constant in the absence of chemical reactions. The pressure inside the bubble decreases and reaches a minimum at the end of the expansion. The temperature \( T \) and pressure \( P_B \) inside the bubble during expansion are given by [13]:

\[
T = T_{00} = \text{cte} \quad (2)
\]

\[
P_B = P_v + \left( P_0 + \frac{2\sigma}{R_0} - P_v \right) \left( \frac{R_0}{R} \right)^3 \quad (3)
\]

where \( T \) is the temperature in the bubble; \( T_{00} \) is the fluid temperature; \( P_v \) is the vapor pressure of the fluid; \( \sigma \) is the surface tension of the fluid; \( R_0 \) is the ambient radius of the bubble.

During the positive pressure phase, cavitation bubbles rapidly collapse accompanied with a series of physical and chemical phenomena. This process occurs in an adiabatic state since the extremely short collapse time inhibits the heat exchange inside and outside the bubble. Temperature and pressure inside the bubble increase as the size of the bubble decreases [14]. When the bubble radius reaches minimum radius \( R_{\text{min}} \) temperature of nearly 5000 K and pressure of nearly 2000 atmospheres can be reached inside the compressed bubble. Such high temperature causes a complex series of elementary reactions to occur, thus producing various oxidants. In conclusion, high temperature, high pressure and oxidation cause sonochemistry reactions. The temperature \( T \) and pressure \( P_B \) inside the bubble during collapse are given by [13]:

\[
T = T_{00} \left( \frac{R_{\text{max}}}{R} \right)^{3(\gamma-1)}
\]

\[
P_B = \left( P_v + \left( P_0 + \frac{2\sigma}{R_0} - P_v \right) \left( \frac{R_0}{R_{\text{max}}} \right)^3 \right) \left( \frac{R_{\text{max}}}{R} \right)^{\gamma}
\]

where \( R_{\text{max}} \) is the maximum radius of the bubble; \( \gamma \) is the adiabatic factor.

The bubble radius \( R_0 \) at 0 instantaneous acoustic pressure is the ambient radius, also known as the equilibrium radius. The equilibrium radius \( R_0 \) reflects the average radius of bubbles at a given frequency. During collapse, the radius of the cavitation bubble decreases from the maximum to the minimum. The compression ratio \( R_{\text{max}}/R_{\text{min}} \) is identified as a key parameter affecting the cavitation intensity [12]. Both maximum temperature and pressure reached inside the bubble at the end of the collapse increase with increasing compression ratio. Moreover, the maximum temperature increases linearly, while the maximum pressure increases exponentially with the compression ratio. The compression ratio is also used to distinguish active from inactive bubbles [15]. Both decreasing the frequency and increasing the acoustic intensity can effectively increase the compression ratio, enhancing acoustic cavitation [12].

2.1.3. Mechanical effects of acoustic cavitation: shock waves and micro jets

As shown in Fig. 2, periodic oscillation and implosion of the cavitation bubble cause mechanical effects, such as shock waves and micro
In the case of oscillating sound pressure, the expansion and collapse of the bubble are driven by the moving bubble wall. The direction of the bubble wall velocity instantly changes from inward to outward at the start of the expansion. This rapid transition causes the bubble wall to launch continuous shock waves into the surrounding fluid [16]. The shock waves produced by the cavitation bubble are shown in Fig. 2(A) [17]. When the cavitation bubble reaches an unstable state, it implodes and generates micro jets. The cavitation jet can penetrate the bubble wall, considered as characteristic sharp extrusion. The micro jets excited by the imploding bubble and the velocity field are shown in Fig. 2(B) [18]. When shock waves and micro jets act on the phase interface, the shear forces cause stripping and disintegration.

2.1.4. Sonochemistry effects: oxygen free radicals

During the bubble collapse, high temperature causes the water and oxygen molecules inside the bubble to decompose. A variety of oxides are formed by the reaction system such as -OH, H₂O₂, H, O [19]. Among them, hydroxyl radicals can react with organics indiscriminately due to their high oxidation capacity (oxidation potential up to 2.80 eV) [20]. Within a single cycle, the total amount of free radicals increases with collapsing bubble, and is kept constant as the bubble expands [13]. After the bubble implodes, free radicals diffuse into surrounding medium and oxidize the organic compounds. The main chemical reactions that produce oxides in the cavitation bubble are presented as following:

\[
\begin{align*}
H_2O &\rightarrow \cdot H + \cdot OH \\
O_2 &\rightarrow 2\cdot O \\
\cdot OH &\rightarrow H_2O_2
\end{align*}
\]

(6) (7) (8)

2.2. Effects of acoustic parameters and medium properties on acoustic cavitation

2.2.1. Acoustic parameters

Frequency, acoustic intensity, treatment time and field type are the main parameters determining acoustic cavitation.

(1) The increase of frequency increases the cavitation threshold, hindering bubble nucleation [21]. In addition, the number of bubbles increases, while the average size of the bubbles decreases [19].

(2) Increasing acoustic intensity results in higher acoustic pressure amplitude, causing stronger bubble oscillation [22].

(3) The fluid temperature increases with treatment time, reducing the cavitation threshold [23].

(4) Acoustic cavitation is more pronounced in the standing wave field than traveling wave field [11].

2.2.2. Medium properties

The properties of the medium that determine acoustic cavitation include viscosity, interfacial tension, saturated vapor pressure, dissolved gas and solid porosity.

(1) High viscosity promotes negative pressure in the expansion phase, thus increasing the cavitation threshold [11].

(2) The increase of interfacial tension decreases the nucleation velocity of the bubble, but increases the intensity of bubble implosion [24].

(3) The increase of saturated vapor pressure promotes acoustic cavitation since the amount of vapor inside the bubble is increased [11].

(4) Bubble nucleation is facilitated by gas dissolved in the medium, resulting in reduced cavitation threshold [25].

(5) Gas trapped in gaps and holes of a solid can act as a cavitation source [25].

3. Enhanced oil recovery

As oil production abates, the development of technologies for enhanced oil recovery is a worldwide challenge of increasing importance [26]. In chronological order, oil production goes through three stages: primary, secondary and tertiary oil recovery. Primary oil recovery refers to the recovery of oil based solely on various natural energies, such as rock expansion and natural gas expansion. Under secondary oil recovery, the reservoir pressure is maintained by water flooding or gas injection. Tertiary oil recovery, also known as enhanced oil recovery, is based on reducing viscosity and adsorbability of crude oil by various physical and chemical methods, thereby increasing its fluidity. After each stage, oil production gradually decreases until the well is no longer commercially viable.

Current EOR technologies are based primarily on chemical reagents, electromagnetic waves, hydraulic fracturing and direct current heating. However, adding surfactants and acids has the disadvantages of high input cost and reservoir contamination. The electromagnetic wave method is limited by the penetration depth in the conductive medium [9]. Hydraulic fracturing is effective in increasing oil production, however the environmental contamination caused by the infiltration of fracking fluid into groundwater is a potential hazard. As a new alternative technology, high power low frequency ultrasonic EOR has the advantages of low cost and straightforward operation, while eliminating the risk of contamination.
The mechanisms of ultrasonic EOR can be classified as: oil viscosity reduction, wax crystallization inhibition and oil reservoir permeability enhancement [27]. The effects of power ultrasound on viscosity are presented in Section 6. When wax nucleates, grows and is deposited in the horizontal portion of the well, it blocks oil flow, reducing oil production [28]. Ultrasonic cavitation is effective in inhibiting the growth of wax crystals. The mechanical vibrations caused by ultrasonic cavitation break long chain alkanes into short chain alkanes, i.e., C–C bond cleavage [3]. The increase of light hydrocarbon proportion improves the oil/water separation test performed in Siberian wells reached an overall success rate of 85%. Currently, coaxial special cables and multi-core special cables are widely used for ultrasonic EOR [38]. With the increase of oil well depths and underground temperatures, special cross-linkable PE with excellent heat resistance is a promising alternative to conventional PE as cable insulation material. Moreover, smaller optical cables with less attenuation are increasingly replacing copper lines.

### 4. Oil sand extraction

According to published data [4], conventional oil reserves are about 1.62 × 10¹¹ m³, while heavy oil and oil sands reserves are about 8.9 × 10¹³ m³. Oil sands, also called bitumen sand usually consists of 3–20% bitumen, 80–87% sand and clay, and 3–6% water [39]. In the petroleum industry, both oil sand and oil sludge are non-homogeneous mixtures of oil, water and soil particles. In comparison with oil sludge, oil sand has lower water content and larger solid particle size. According to current data, 85% of the world’s proven oil sand resources are concentrated in Canada. Canada’s oil sand has high density, high viscosity, high acid value and high bitumen content. In order to recover tar sand, adequate process should be used to separate bitumen from oil sand prior to bitumen dilution and modification. In Canada, water washing is the most widely used in industry for oil sand extraction [40]. However, high water consumption and water contamination caused by this method are significant drawbacks.

Power ultrasound is considered to be a promising alternative to conventional technology for oil sand extraction. The oscillating pressure wave penetrates the bitumen and acts on the oil-solid interface, which is inapplicable to other methods. Acoustic cavitation disintegrates the bitumen, which is attributed to a combination of mechanical vibrations, chemical reactions and enhancing the mass transfer [41]. As shown in Fig. 5, micro jets caused by the cavitation bubble erode the bitumen surface, resulting in micro holes [42]. The holes expand, reach solid surface, and eventually the bitumen layer is entirely stripped. Through sonochemical reactions, the bitumen is converted into small organic molecules, such as resins and oils. According to [43], the average density of bitumen increased from 8° to 15° API and the ratio of H/C also increased after ultrasonic treatment, indicating that light hydrocarbons were being formed. In order to cause acoustical cavitation, ultrasonic disintegration of oil sand is performed in water, i.e., water-based ultrasonic extraction. In [44], adding alkali further improved ultrasonic efficiency of oil sand extraction. Liu [45] studied the effects of pH on the bitumen-silica interaction using atomic force microscopy (AFM). It was found that rising pH resulted in decreased force of adhesion, and higher repulsion barrier. During sonication, alkalis react with carboxylic acids in the bitumen, creating in situ surfactants [41]. These anionic surfactants not only reduce the bitumen/water interfacial tension but also increase the zeta potential of both bitumen droplets and silica particles, facilitating the separation of the bitumen from solid surface [46]. Three dynamical regions of ultrasonic oil sand extraction in dependence on sonication time are shown in Fig. 6. In region A, the oil component in bitumen is released by ultrasound. The recovered light oil and non-polar oil increase extraction efficiency. In region B, the separation of asphaltenes from solid particles dominates the extraction process. In situ surfactants generated by the neutralization reaction destroy the exposed asphaltenic micelles and form Hartley micelles. In region C, asphaltene micelles are cracked by acousti-
Cavitation. This effect demonstrates the advantage of using power ultrasound with respect to other methods. Under ultrasonic treatment, the autocatalytic reaction can be sustained by maintaining pH, without consuming additional surfactants.

Ultrasound in the range of 20–40 kHz is usually used to study the effects of various factors (acoustic power, treatment time, alkali concentration and temperature) on bitumen recovery efficiency [47,48]. The published results on ultrasonic oil sand extraction are listed in Table 1. According to current literature, Sadeghi et al. did the first systematic study on oil sand extraction using ultrasound in the 1980s and 1990s [49,50]. Mason et al. proposed two mechanisms of power ultrasound for soil cleaning (abrasion of soil particles and leaching of entrenched particles) [51] and a leaching model [52]. The equipment for ultrasonic oil sand extraction is developing towards large scale and high power in order to meet industrial demands. Various ultrasonic devices have been developed, such as the flow vibrating tray, the reactor equipped with multiple high power vibrators and the tube equipped with external surface transducers [51,53]. An industrial ultrasonic device capable of producing sound waves of audible frequency and up to 6 mm of amplitude has been designed in Canada. In [54], equipment for ultrasonic oil sand extraction developed by Russian Academy of Sciences is reported. It operates in a frequency range of 25–40 kHz and a power range of 1–7 kW. The pilot-scale testing for extracting contaminated soil was carried out, which verified the results obtained in the laboratory.

5. Crude oil demulsification

The process of oil production consists of water injection and surfactant addition into the oil reservoir, and results in stable water-in-oil (W/O) emulsions. Before crude oil can be transported and refined, dehydration is required since emulsified water increases the oil viscosity and leads to excessive friction loss [58]. In addition, the ions in emulsified water (such as Na+, Mg2+, Ca2+, Cl−) not only corrode refinery equipment but also deactivate the catalysts [59]. The factors affecting the emulsion stability are water content, oil viscosity, droplet size distribution, oil–water interface property, and others [60]. In order to separate dispersed water droplets from oil, the oil–water interface must be broken for further coalescence of droplets. The surfactants used for oil recovery reduce the oil–water interfacial tension, thus stabilizing the interface film [61]. In addition, the asphaltenes in crude oil are surface-active materials and dominate the stability of the W/O emulsion. The heterocyclic molecules of asphaltenes possess hydrophilic functional groups and adsorb on the oil–water interface [62]. This rigid interfacial film is elastic and difficult to drain out, which impedes the coalescence of droplets as they approach.

Demulsifiers and electrostatic methods are widely used for crude oil demulsification [59]. Demulsifier molecules can replace the emulsifier molecules on oil–water interface, which destabilizes the interfacial film [63]. However, emulsifiers cause environmental pollution and their overuse leads to re-stabilization of the emulsion [59]. Under electrostatic treatment, the water droplets are polarized and coalesce into larger droplets, which facilitates their separation from crude oil. However, in emulsions with high water content, the electric field is prone to break down due to water droplets that form between electrodes [59]. Power ultrasound has been used for numerous studies on demulsification at the lab scale due to its low cost, cleanliness and high adaptability [65,66]. The research on crude oil demulsification using ultrasound is listed in Table 2.

When the distance between the emitting and reflecting surfaces is an integer multiple of half a wavelength, sound waves overlap and

![Fig. 5. Cavitation bubble disintegrates the bitumen in oil sand.](image)

![Fig. 6. Dynamical model of ultrasonic oil sand extraction.](image)

Table 1

| Oil sand origin | Irradiation mode | Frequency | Oil sand quality | Oil content | Effect | Reference |
|-----------------|------------------|-----------|-----------------|-------------|--------|-----------|
| Canada          | Continuous, single frequency | 40 kHz | 200 g         | 14.5 wt%    | Disintegration | [43]     |
| Canada          | Continuous, single frequency | 22 kHz | 15 g          | 13.5–14.5 wt% | Disintegration | [41]     |
| Canada          | Continuous, single frequency | 28 and 200 kHz | 2.97 g   | 12.3 wt%    | Disintegration | [55]     |
| China           | Continuous, dual frequency | 28 and 68 kHz | 200 g | 9.8 wt%     | Disintegration | [4]      |
| China           | Pulsed, single frequency | 28 kHz | –             | 12.53 wt%   | Disintegration, interface effect | [56]     |
| Indonesia       | Continuous, single frequency | 40 kHz | 50 g          | 25.2 wt%    | Disintegration, interface effect | [57]     |
generate standing waves. Ultrasonic standing waves (USWs) can cause the water droplets in oil to aggregate into bandings, which increases the probability of droplet collision and coalescence. Due to the difference in compressibility and density between oil and water, water droplets in oil are driven to nodal or antinode planes by USWs [72]. The aggregated droplets are equally spaced in the direction of acoustic propagation, forming lateral droplet bandings. The secondary Bjerknes force comes into action once the droplets approach to each other, facilitating droplet coalescence [73]. Pangu et al. proposed a mathematical model of dynamic behavior of binary droplets [74], and developed a global model of the droplet coalescence [75]. Luo et al. studied the droplet suspension and banding characteristics in USWs [76, 77]. Under ultrasonic treatment, the dispersed droplets vibrate with the continuous phase. The droplets of different particle sizes vibrate at different velocities, increasing the probability of their collision and coalescence, thus facilitating oil–water separation [78]. Acoustic cavitation results in secondary emulsification, which reduces the demulsification efficiency [69]. However, Antes et al. [71] showed that mechanical effects of acoustic cavitation can break the barrier between droplets and facilitate their coalescence. It was found that the optimum frequency for crude oil demulsification is 45 kHz; however, the dehydration effect vanishes at frequencies above 130 kHz. Power ultrasound delivers high specific energy density to the droplets and causes them to vibrate [79]. The mechanical vibration causes the surfactants adsorbed on the oil–water interface to re-disperse, reducing the film strength. Xie et al. [80] studied the effects of ultrasound on the oil–water interface using an interfacial shear viscometer. After ultrasonic treatment, the interfacial shear strength decreased since the asphaltenes on the interface were redissolved in the oil phase. Moderate acoustic cavitation can break the interfacial film of the droplet, which facilitates crude oil demulsification. The coalescence evolution of droplets near the cavitation bubbles is shown in Fig. 7 [81]. However, excessive cavitation breaks droplets into smaller ones, causing secondary emulsification. The acoustic parameters affecting demulsification efficiency primarily include frequency, acoustic intensity and irradiation time. It is generally accepted that lower frequency leads to higher demulsification efficiency of crude oil [30]. This is attributed to the decrease of frequency that enhances cavitation intensity. Under ultrasonic irradiation at 20 kHz, the coalescence behavior of droplets [82] and increase of the droplet size [80] in W/O emulsions were reported. According to [67], the demulsification efficiency of crude oil reaches maximum for a certain acoustic intensity. Excessive acoustic intensity not only reduces the demulsification efficiency, but also causes tiny oil droplets to separate in water [83]. At high acoustic intensity, cavitation bubbles frequently implode and produce micro jets that inject and disperse oil into the water phase. The demulsification efficiency increases and then saturates with treatment time [70]. The emulsion properties determining the demulsification efficiency are primarily oil viscosity, interfacial tension and water content. High viscosity inhibits not only acoustic cavitation but also droplet vibration. It is well-known that low interfacial tension hinders droplet coalescence. Compared with gravitational sedimentation, ultrasound has significant advantages for treating emulsions with low interfacial tension [83]. The water content has complex effects on ultrasonic demulsification. The increase of droplets can increase the probability of droplet collision and coalescence. However, increased droplet concentration causes excessive dissipation of acoustic energy and ultrasound load [67]. In addition, the increase of water content increases the saturated vapor pressure of emulsions, reducing the cavitation threshold [10]. The design of the ultrasonic device is also a key point for improving demulsification efficiency. In order to maintain the cavity resonant frequency, two independent ultrasonic transducers were used to treat emulsions with varying temperatures [84]. In addition, the energy utilization of the device can be more efficient by optimizing the model, which considers both medium properties and device parameters [85].

### 6. Crude oil viscosity reduction

The pressure energy consumed during crude oil transportation causes significant losses for the petroleum industry. Particularly, extraction and transportation of heavy oil remain a challenge due to its high viscosity (greater than 1000 cP). The heavy oil has a large proportion of asphaltenes that are highly dense, polar, and insoluble in straight chain alkanes. In the colloidal dispersion system of crude oil, the asphaltenes are in the form of micellar nuclei, which are stabilized by the adsorbed resins [86]. The stability of asphaltene micelles is also affected by the ratio of asphaltenes and resins, aromatic degree, and other parameters [87]. The increase in the size of asphaltene micelles results in a higher viscosity of crude oil [88]. In general, the asphaltenes tend to precipitate from crude oil and agglomerate. When the asphaltene particles in oil reach a certain size, the interactions between asphaltene particles come into action and dominate the rheological characteristics of the system [89]. In [90], it was found that the heavy oil viscosity increases exponentially with asphaltene content.

### Table 2

| Crude oil details       | Irradiation mode            | Frequency   | Oil viscosity          | Water content | Effect                  | Reference  |
|-------------------------|-----------------------------|-------------|------------------------|---------------|-------------------------|------------|
| Gachsanran crude oil    | Continuous, single frequency| 20 kHz      | 19.1 mm²/s (25 °C)     | 10-25 v.%     | Standing waves           | [67]       |
| Lu-Ning crude oil       | Pulsed, single frequency    | 10-80 kHz   | 1565.2 mm²/s (20 °C)   | 56 v.%        | Standing waves           | [68]       |
| Iranian crude oil       | Continuous, single frequency| 28 kHz      | 16.82 mm²/s (20 °C)    | 7 v.%         | Standing waves           | [69]       |
| Lu-Ning crude oil       | Continuous, single frequency| 10 and 20 kHz| 108 mm²/s (30 °C)       | 5 v.%         | Standing waves           | [6]        |
| SAGD heavy oil          | Continuous, single frequency| 10-30 kHz   | –                      | 30-90 v.%     | Mechanical vibrations    | [70]       |
| Brazilian heavy crude oil| Continuous, single frequency| 35 kHz      | 133.4 mm²/s (45 °C)    | 12-50 v.%     | Mechanical vibrations    | [71]       |

![Fig. 7. Coalescence evolution of droplets near the cavitation bubbles.](image-url)
small molecules and free radicals. The combination of these two effects results in a significant reduction of oil viscosity. In Phase 3, small molecules and free radicals reinte-grate to produce large oil molecules, increasing the oil viscosity. Najafi et al. [88] investigated the viscosity reduction of two oil samples using ultrasound, and found the optimal treatment time increases with the augmentation of asphaltene content. Acoustic cavitation occurs when the negative pressure of ultrasound overcomes the cohesive force in the fluid. The approximate relationship between the acoustic cavitation threshold and the viscosity is presented in Eq. (9) [5]. For heavy or ultra-heavy oil, sufficient acoustic intensity is required to produce cavitation bubbles to break oil molecules. After sonication, the decomposed molecules are dispersed in colloidal systems of crude oil. The asphaltene micelles can absorb small molecules, which increases the size of the micelle as well as the viscosity of crude oil [96]. However, the viscosity cannot be fully recovered since the total amount of asphaltenes in crude oil is reduced.

\[
P_C = 0.08(\log \eta + 5)
\]

where \(P_C\) is the acoustic cavitation threshold (MPa); \(\eta\) is the kinetic viscosity (Pa·s).

7. Oily wastewater treatment

In the later stages of oil production, numerous tertiary oil recovery methods have been developed, such as alkaline flooding, polymer flooding and alkaline-surfactant-polymer (ASP) flooding. Among them, the ASP system is a promising approach for improving oil production, in use since 1980s [100]. However, the use of ASP produces a large amount of oily wastewater that is more difficult to manage in comparison with general wastewater. The high concentration of polymers increases the viscosity of water (by a factor of 4–6), reducing the flotation velocity of oil droplets. In addition, polymers and surfactants adsorb on the oil–water interface and thus impede the droplet coalescence. In China, the maximum oil content of oily wastewater allowed to be discharged is only 10 mg/L because of its high pollution risk and toxicity [101]. Various techniques are developed to treat oily wastewater in the petroleum industry, such as flotation, membrane filtration, electrochemical methods and chemical oxidation.

According to published literature, few studies focus on oily wastewater treatment using solely ultrasound. It is common to combine ultrasound with other techniques in order to enhance oil removal or compensate for their drawbacks. Table 4 lists published research on oily wastewater treatment with ultrasound. Ultrasound-assisted advanced oxidation methods (AOPs) can be used to degrade the organics in water. The effect of advanced oxidation is attributed to the production of OH radicals, which react with organic molecules at the rate of 10^9 to 10^10 mol·L^-1·s^-1 [102]. During the final stage of collapse, acoustic cavitation bubbles can also produce OH radicals and release them into the aqueous phase when imploding. According to [103], the ultrasonic degradation of water-soluble polymers is effective in the frequency range of 200–600 kHz. Combined ultrasound-Fenton application shows better oxidation effect on oily wastewater than using one method alone [104]. The reaction process enhancement by ultrasound pre-oxidation was also reported in [105]. The combined application of ultrasound and chemical or electric flocculation causes organics in water to

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**Table 3**

Published research on crude oil viscosity reduction with ultrasound.

| Crude oil details           | Frequency | Oil viscosity         | Asphaltene content | Effect                        | Reference |
|-----------------------------|-----------|-----------------------|---------------------|-------------------------------|-----------|
| Samara crude oil            | 20 kHz    | 1014 mPa·s (20 °C)    | 6.1 wt%             | Breaking chemical bonds, heating | [95]      |
| Iran heavy crude oil         | 20 kHz    | 796.28 mPa·s (22 °C)  | 11.34 wt%           | Breaking chemical bonds, heating | [93]      |
| Daqing heavy oil            | 28 kHz    | 2830 mPa·s (30 °C)    | 8.81 wt%            | Breaking chemical bonds        | [96]      |
| Ultra-heavy residual oil    | 20–24 kHz | 659.86 Pa·s (30 °C)   | –                   | Breaking chemical bonds, heating | [5]       |
| Paraffin oil                | 25 and 68 kHz | 31.73 mPa·s (25 °C) | –                   | Heating                       | [97]      |
| Saudi light vacuum residuum | 40 kHz    | 4665.15 mPa·s (100 °C) | 17.14 wt%           | Breaking chemical bonds, heating | [98]      |
agglomerate and separate. The oil droplets in oilfield wastewater have high negative Zeta potential and strong electrostatic repulsion between each other, resulting in a stable dispersion system. The cationic flocculants play the roles of adsorption bridging and charge neutralization on oil droplets; however, they have low affinity to hydrophobic oily colloids [106]. Ultrasound can induce the polymerization of flocculants with hydrophobic groups, thus enhancing the removal efficiency of oil from wastewater. According to [107], the ultrasonically improved electroflocculation technology is effective in reducing the oil content of wastewater in industrial tests. In addition, ultrasound can be used to deal with membrane fouling for improving the filtration efficiency. The deposition and adsorption of particles on the membrane reduce the membrane flux and even block membrane pores [108]. The shock waves and micro jets produced by ultrasonic cavitation cause particles to initiate and detach from the membrane [109]. In addition, the interaction of the membrane and contaminant can be disrupted by imploding cavitation bubbles, improving membrane flux [110].

It is feasible to aggregate oil droplets in water by establishing an ultrasonic standing wave field. In [112], radioactive particles suspended in wastewater were removed with an efficiency of up to 97.68% using USWs. As shown in Fig. 9, the oil droplets in water aggregate and flocculate into oil clusters in USWs of 2 MHz. Before ultrasonic treatment, the droplet size was concentrated in the range of 2 to 6 μm. After ultrasonic treatment, the size of oil clusters varies from 0.11 to 0.18 mm, while the rising rate of oil clusters is in the range of 0.45 to 1.1 mm/s. In general, the ultrasonic separation of micron-sized particles is performed around 1 MHz [113]. The applications of high frequency ultrasound for separating particles from biomass are reviewed in [114]. The advantages of using high frequency USWs for particulate separation are twofold. The increase of frequency can increase the primary acoustic force acting on particulates, which improves their migration velocity. In addition, the higher frequency results in a shorter interval between bandings, thus reducing the migration distance of particulates.

8. Oily sludge treatment

Oil sludge is hazardous waste discharged during the process of crude oil extraction, transportation, storage and refining. According to [115], nearly 3 × 10^6 tons of oil sludge is produced per annum in China, the total world production is equal to 160 million metric tonnes per year. In general, oily sludge is composed of 15–50% PHCs, 30%–85% water and 5%–46% solid [7]. The carcinogenic substances (aromatic and polycyclic aromatic hydrocarbons) and heavy metals (Ni, V, Fe) in oily sludge not only harm the human health but also contaminate the environment [116]. The oil phase in oily sludge is in the form of a stable colloidal dispersion, which consists of saturates, aromatics, resins and asphaltenes (SARA). These stable organics are resistant to microorganisms and cannot be degraded completely in natural stacking [117]. The strong interactions between oil molecules and solid particles also impede oil removal from oily sludge. Based on the kinetic model fitting for the desorption behavior of oil components, it was found that the adsorption between oil molecules and solid particles is physical adsorption [118]. The desorption coefficient of asphaltenes is the lowest among four components, indicating that the asphaltenes dominate the oil adsorption [86]. In addition, the asphaltenes are the most complex polar molecules in oil, possessing heteroatoms S, N and O. The stable adsorption of oil on solid particles can be attributed to the formation of hydrogen bonds between heteroatoms and hydroxyl groups on the surfaces of solid particles [119].

Developing an efficient and low risk treatment technology for oily sludge has attracted widespread attention. Incineration is a simple and efficient method, however, it has a drawback of releasing VOCs [120]. Centrifugation [7] and pyrolysis [121] provide a more thorough treatment of oily sludge, however, the high energy consumption is a significant drawback. Water-based ultrasound can remove oil from oily sludge, which produces no harmful gases and requires low energy input. Due to its deep penetration, ultrasound can reach blind holes and gaps, providing deep cleaning. Published research on oily wastewater treatment with ultrasound is listed in Table 5.

Pulsed pressure waves of ultrasound force the water droplets and solid particles in sludge to vibrate and thus destabilize oily sludge. Water droplets collide with each other and coalesce, facilitating oil–water separation [128]. Ultrasound can penetrate into each phase of oily sludge and disintegrate sludge entirely [129]. After ultrasonic treatment, the average particle size of sludge is found to be reduced [130]. When oily sludge is broken into smaller pieces, the effective area between cavitation bubbles and sludge increases, enhancing deep oil removal from solid surfaces. The effects of cavitation bubbles on oil removal from oily sludge include not only mechanical disintegration but also oxidative degradation [131]. The oxidation process can be neglected since its reaction constant takes a small percentage of the total reaction constant. Therefore, the mechanical effects of ultrasonic cavitation are dominant causes for oil removal from oily sludge. As shown in Fig. 10, the shock waves and micro jets produced by cavitation bubbles break the hydrogen bonds between oil and solid particles, resulting in oil desorption [132]. Both resins and asphaltenes have polar heterocyclic compounds which exhibit strong adsorption on solid surfaces [133]. According to [127], ultrasound is effective in desorbing resins and asphaltenes, thus improving removal efficiency of all oil components. The combined application of ultrasound and other techniques (Fenton oxidation, thermochemical cleaning, freezing, etc.) is viable for more thorough oil removal. A variety of physical and

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Table 4

| Oil sample details | Frequency | Oil content | Method | Reference |
|--------------------|-----------|-------------|--------|-----------|
| Olive oil          | 20 kHz    | –           | Combined with Electro-Fenton method | [104] |
| Oil products       | 20 kHz    | 223.5 mg/l  | Combined with electrocoagulation    | [106] |
| Paraffin oil       | 40 kHz    | 1000 mg/l   | Combined with membrane filtration | [111] |
| Canola oil         | 2 MHz     | 5 v.%       | Standing waves                       | [66] |

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![Fig. 9. Oil droplets in water aggregate and flocculate into oil clusters in USWs.](image-url)
chemical processes can be enhanced due to accelerated heat and mass transfer by ultrasonic cavitation. It was reported that the ultrasound-assisted thermochemical treatment resulted in higher oil removal efficiency than using one method alone [123].

9. Conclusion and outlook

In recent years, low oil recovery efficiency, high energy consumption and high pollution have been key issues impeding the sustainable development of petroleum industry. As an emerging green technology, power ultrasound possesses great prospects in both upstream and downstream of the petroleum industry. In certain oil fields, power ultrasound is presently being used, successfully increasing oil production.

Low frequency and high power are likely to remain the trend of ultrasonic industrial development in the future. The decrease of frequency not only reduces the acoustic cavitation threshold but also increases the compression ratio of cavitation bubbles. The lower the frequency, the lower the attenuation coefficient of acoustic energy and the longer the penetration distance. In addition, high power is required to produce strong acoustic cavitation. At present, the transducers are mainly made of piezoelectric ceramics which are not resistant to high temperature and contain heavy metals (Pb, Ba, etc.). Lithium niobate shows promise as an alternative material for the preparation of ultrasonic viscosity reduction technology should focus on heavy and ultra-heavy oil with high asphaltene content, since higher initial crude oil viscosity results in superior relative effectiveness of the ultrasonic viscosity reduction process. In addition, the combined application of ultrasound and other techniques is promising for achieving enhanced oil removal from oily waste.

CRediT authorship contribution statement

Xiaoming Luo: Conceptualization, Methodology, Software, Writing - review & editing. Haiyang Gong: Data curation, Writing - original draft. Ziling He: Supervision. Peng Zhang: Visualization, Investigation. Limin He: Software, Validation.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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