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Influence of ultrasonic waves on the removal of different oil components from oily sludge

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Ultrasonic technology is a promising tool for washing oily sludge to recover oil. In this study, the influence of ultrasonic conditions on the removal of different oil components from oily sludge was investigated to optimize ultrasonic washing technology. Among the three frequencies (25, 50 and 100 kHz) applied, ultrasonic washing at 25 kHz exhibited the best performance in terms of oil extraction rate and efficiency. An ultrasonic intensity of 0.33 W/cm² was necessary to overcome the energy threshold for oil washing. Application of a standing wave or dual frequencies did not improve the oil removal performance perceptibly. The optimum conditions for the removal of oil from oily sludge were an ultrasonic frequency of 25 kHz, intensity of 0.33 W/cm² and sludge/water ratio of 1/2 (in volume). Thin-layer chromatographic flame ionization detection showed that ultrasonication could overcome high energy thresholds, resulting in an increase in the removal of asphaltenes and resins, with the removal of asphaltenes particularly affected by frequency. This study could provide valuable information for the application of ultrasonic technology in oily sludge treatment.

Keywords: oily sludge; ultrasound; oil removal; oil family compositions; ultrasonic washing

1. Introduction

A large amount of oily sludge, which contains 30–45% of oil and diverse chemical compounds with a wide toxicity range to biota, is generated during petroleum production and processing activities.[1–3] Because of its hazardous nature, the disposal of oily sludge is a big challenge for oil production.[4–6]

Many technologies, including centrifugation, thermal desorption, biodegradation, solvent extraction, hydrothermal processing and ultrasonic cleaning, have been tried for the treatment of oily sludge.[7] Among them, ultrasonic cleaning, which has been applied widely to precision or critical cleaning, has shown strong promise.[8,9] Ultrasonic waves generate and evenly distribute cavitation implosions in a liquid medium. The released energy can reach and penetrate crevices, blind holes and areas that are normally inaccessible through other cleaning methods.[10,11] The removal of contaminants is consistent and uniform, regardless of the complexity and geometry of the substrates, making it suitable for the treatment of oily sludge characterized with a complicated interface.[11] Zhang et al. found that oil removal could be as high as 64.1% under an ultrasonic frequency of 20 kHz and work power of 60 W, and could be further increased to 73.2% by combining ultrasonication with surfactants.[12] However, recent studies have focused on the removal effect of oily sludge by ultrasonic cavitation, with little reported on the ultrasonic washing process.

Oil in oily sludge usually has complex family compositions, which can be divided into saturates, aromatics, resins and asphaltenes.[13,14] Different oil components have different wettability to solid particles. It is therefore easy to speculate that different oil components may exhibit different surface stripping efficiencies by ultrasonic washing, leading to differences in removal efficiencies for different components. Because oil family compositions are very different in different oil fields, it is important to understand the impacts of ultrasonic conditions on different oil components in oily sludge.

In this study, we optimized ultrasonic conditions based on the effects of ultrasonic frequency, intensity and sludge/water ratio on the removal of oil and oil components from oily sludge generated during oil production. The removal efficiencies of different oil components were evaluated using thin-layer chromatography (TLC). At the same time, the application effects of a standing wave and dual frequencies on oil removal efficiency were also investigated. The results derived from this study could provide valuable information for establishing a sound oily sludge treatment technology.

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2. Materials and methods

2.1. Samples and reagents

Oily sludge samples were taken from an oilfield in north China and stored at 4°C before use. The major components of the samples were oil (54.5%, by mass), water (25%) and sand (20.5%).

Carbon tetrachloride, anhydrous sodium sulphate, magnesium sulphate, magnesium silicate and all other reagents were purchased from the Beijing Chemical Company, Beijing. All chemicals were of reagent grade and used without further purification.

2.2. Treatment procedure

The experiments were carried out in a thermo-stated stainless vessel \((L \times W \times H = 0.3 \text{ m} \times 0.2 \text{ m} \times 0.18 \text{ m})\), Figure 1) equipped with an ultrasonic generator applicable at different frequencies (25, 50, 100, 25/50, 25/100 and 50/100 kHz) and output powers (0–300 W). A transducer was mounted under the bottom of the vessel. Water and oily sludge were filled into a one-litre glass vessel used as the reactor at a proper ratio under 30–60 rpm mixing and 20°C. Ultrasonication was applied under different frequencies (including dual frequencies) and output powers. A standing wave was generated by adjusting the distance between the vibration plate and the liquid level to the multiple of half a wavelength \((\lambda/2)\). For the ultrasonic wave at 25 kHz, velocity in the oily sludge/water mixture (weight ratio, 1/2) was 1449 m/s (measured by the Institute of Acoustics, Chinese Academy of Sciences), and its corresponding wave amplitude was 0.058 m. A control treatment experiment was performed under the same conditions except that ultrasonication was not applied.

2.3. Analytical methods

Oil content in oily sludge was determined using an infrared oil content analyser (JDS-106U, Jiling Beiguang Analyzer Co., China) following the extraction of oil by carbon tetrachloride according to the Standard Method (GB/T16488–1996).[15] The major components of oil (saturates, aromatics, resins and asphaltenes) were analysed by a thin-layer chromatograph equipped with a flame ionization detector (TLC-FID) (MK6s, Iatroscan, Japan).[16,17] Ultrasonic pressure was measured using a CS-3 hydrophone (sensitivity, 2.2 μV/Pa; Institute of Acoustics, Chinese Academy of Sciences). The ultrasonic intensity \(I \text{ (W/m}^2\text{)}\) was calculated using the following equation [18]:

\[
I = \frac{P^2}{\rho v},
\]

where \(P \text{ (Pa)}\) is the ultrasonic pressure, \(\rho \text{ (kg/m}^3\text{)}\) is the density of the sludge–water mixture and \(v \text{ (m/s)}\) is the ultrasound spreading velocity in the sludge–water mixture.

3. Results and discussion

3.1. Effects of ultrasonic wave frequency on removal of oil and oil components

The frequency, intensity and form of ultrasonic waves exert a great influence on the intensity and abundance of cavitations during ultrasonication in a given medium.[10,11] Figure 2 shows the effect of ultrasonic frequency on oil removal. Compared with the control treatment without ultrasonication, ultrasonication treatment enhanced the removal of oil from oily sludge. Among the three frequencies tested, ultrasonic washing at the lowest frequency (25 kHz) exhibited the best performance in terms of oil extraction rate and efficiency. Under this condition, approximately 49.5% of oil was recovered from oily sludge within 30 min. Similarly, Xu et al. also found that oil removal at 28 kHz was higher than that at 40 kHz.[7] At low frequency (20–30 kHz), fewer cavitations of large size and greater energy were generated, while at higher frequency, much denser cavitations with moderate or lower energy were formed.[19] The lower cavitation threshold and higher cavitation intensity at low frequency might be favourable for ultrasonic washing of oily sludge.[7]

![Figure 1. Ultrasonic system.](image)

![Figure 2. Effect of frequency on removal of oil. Sludge/water ratio by weight, 1:2; \(T = 20^\circ\text{C}\); output power, 300 W.](image)
Swamy et al. reported that the application of dual frequencies (20 and 40 kHz) could increase copper yield in the leaching of oxide ores. [19] However, such an enhancing effect was not observed for the removal of oil in our study. The combination of 25/50 or 25/100 kHz at the same output power (300 W) exhibited decreased oil removal (39.3% and 38.5%, respectively) in comparison with the treatment at 25 kHz (data not shown). At the same output power, the use of dual frequencies resulted in the reduction in cavitation intensity at the lower frequency, which might be the main reason for the decrease in oil removal. At the same time, the effect of a standing wave (or stationary wave) on oil removal was also investigated. A standing wave was produced by emitting two waves with the same frequency, wavelength and amplitude in opposite directions. However, no perceptible difference was observed between the standing wave treatment and the non-standing wave treatment (data not shown).

The TLC-FID method can analyse oil family compositions. [16,17] We found that the oil extracted from the oily sludge sample contained 47% of saturates, 24.5% of aromatics, 19% of resins and 9.5% of asphaltenes. Figure 3 shows the effects of ultrasonic frequency on the removal of different oil components. Although the use of ultrasonication resulted in an increase in removal for all four components, the effects on the removal of resins and asphaltenes were significant. Without ultrasonication, the removal of resins and asphaltenes were only 5.0% and 1.3%, respectively, showing that it was difficult to strip these two components from solid particles. By employing ultrasonication, their removals increased to 27.9–31.9% and 21.0–31.9%, respectively, under different frequencies. Furthermore, lower frequencies favoured the removal of oil components, particularly for resins and asphaltenes. The removals at 100, 50 and 25 kHz were 21.0%, 27.0% and 27.9%, respectively, for resins, and 13.9%, 16.2% and 42.3%, respectively, for asphaltenes. The removal of aromatics also increased slightly at 25 kHz, but the effect on saturates was negligible.

3.2. Effects of ultrasonic wave intensity on removal of oil and oil components

Although oil removal efficiency can be significantly affected by ultrasonic intensity, most previous studies have only provided output powers of the experimental equipment. [18,19] which make comparison and plant design difficult. In the present study, output power was transformed into ultrasonic intensity. With the increase in output power from 100 to 300 W (corresponding to power density from 0.17 to 0.5 W/cm²), ultrasonic intensity increased from 0.13 to 0.33 W/cm². As shown in Figure 4, with the increase in ultrasonic intensity from 0.13 to 0.33 W/cm², the 30 min oil removal increased from 13.8% to 49.5%. It should be noted that an increase in ultrasonic intensity from 0.26 to 0.33 W/cm² led to a significant improvement in oil removal, showing that there might be a cavitation threshold for oily sludge washing. In other words, the ultrasonic waves must generate enough pressure amplitude for oil components to overcome the adhesion force of sludge. [15]

Figure 5 shows the effect of ultrasonic intensity on the removal of different oil components. Removal of asphaltenes began when the ultrasonic intensity was increased to 0.19 W/cm², and the removal of resins began when the intensity was 0.33 W/cm². For the enhancement of saturates removal, an ultrasonic intensity of 0.19 W/cm² was sufficient. Therefore, a higher ultrasonic intensity might be required for the treatment of oily sludge containing abundant asphaltenes and resins.

3.3. Effect of sludge/water ratio on oily sludge washing

Other important factors on oily sludge washing are the colligative properties of liquid media, including viscosity, surface tension and density. [19–21] The effect of the

![Figure 3](image1.png)

Figure 3. Effect of ultrasonic frequency on the removal of different oil components. Sludge/water ratio by weight, 1:2; $T = 20\degree C$; ultrasonic output power, 300 W; ultrasonic time, 120 min.

![Figure 4](image2.png)

Figure 4. Effect of ultrasonic intensity on removal of oil. Sludge/water ratio by weight, 1:2; ultrasonic frequency, 25 kHz; $T = 20\degree C$. 
sludge/water ratio on oil removal is shown in Figure 6. Oil removal increased from 12.3% to 46.8% when the sludge/water ratio was decreased from 1:0.5 to 1:2. However, a further decrease in the sludge/water ratio to 1:4 did not improve the oil removal performance markedly. It has been reported that the cavitation process, including the formation, growth and collapse of bubbles due to the propagation of a mechanical wave through liquid, is important for stripping oil from oily sludge. When a sound wave or any mechanical wave propagates through liquid, it does so as a series of rarefactions and compressions.[22] Thus, an increase in the ratio of water to oil could lead to a decrease in the viscosity of the mixture and an enhancement in cavitation. The decrease in viscosity by dilution improved its mobility and ultrasonic intensity. Considering that a larger reactor would be needed for a lower sludge/water ratio, we determined that the optimum ratio was 1:2.

4. Conclusions

The present study demonstrated that the optimum conditions for oily sludge treatment were a frequency at 25 kHz, ultrasonic intensity of 0.33 W/cm² and soil/water ratio of 1/2. The use of a standing wave or dual frequencies was not effective in improving oil removal efficiency. The TLC-FID analysis indicated that asphaltenes and resins had a higher energy threshold. Because ultrasonication alone did not achieve satisfactory oil removal, the combination of ultrasound and some other measure, including the use of surfactants, may be required.

Disclosure statement

No potential conflict of interest was reported by the authors.

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