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REVIEW ARTICLE

Status and prospect of oil recovery from oily sludge: A review

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Abstract Oily sludge is a kind of solid emulsified waste produced by the petroleum industry. It is generally composed of water, crude oil, and solid particulate matter. Because it contains large amounts of cycloalkanes, benzene series, polycyclic aromatic hydrocarbons, and other toxic and harmful substances, it poses a substantial threat to human health and the surrounding environment; therefore, it must be treated to reduce its toxicity. However, a large component of oily sludge is crude oil, which has great recycling value. Therefore, various crude oil recovery technologies, such as solvent extraction, pyrolysis, centrifugation, ultrasonic treatment, electronal treatment, flotation, supercritical treatment, and combined processes, have been developed for the treatment of oily sludge. The main purpose of this review is to discuss the development of these recycling technologies and to summarize and compare their advantages, disadvantages, and mechanisms of action. On this basis, the future development direction of recycling technology is prospected.

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Contents

1. Introduction

Oily sludge is mainly generated during the production, refining, storage, and transportation of petroleum (Deng et al., 2016; Liang et al., 2017), and includes mud from the drilling process, waste oil in the well, emulsified solids created during the crude oil refining process, and sediment in the storage tank (Fig. 1) (Deng et al., 2015; Vivana et al., 2015; Wang et al., 2017). In China, the annual sludge production is close to 5 million tons (Gong et al., 2018; Wang et al., 2019). With the continuous growth of the oil industry, the development of unconventional oil and gas fields has continued to increase (Santos et al., 2014), s has the production of oily sludge (Fig. 2) (BP, 2019). Oily sludge is generally a complex emulsifying mixture composed of water, heteroatoms (N, O, S), heavy metals (Ca, V, Fe, Ni), crude oil, solid particles, and various surfactants (Wang et al., 2018a; Castaneda et al., 2014). The chemical compositions and properties of oily sludge vary

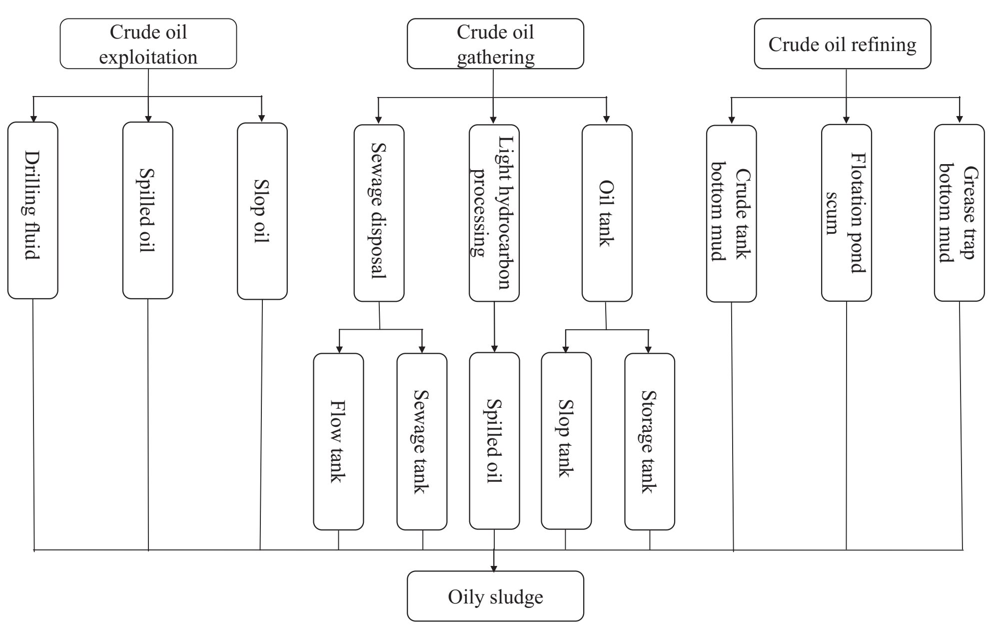

Fig. 1 A schematic diagram of the oily sludge source.

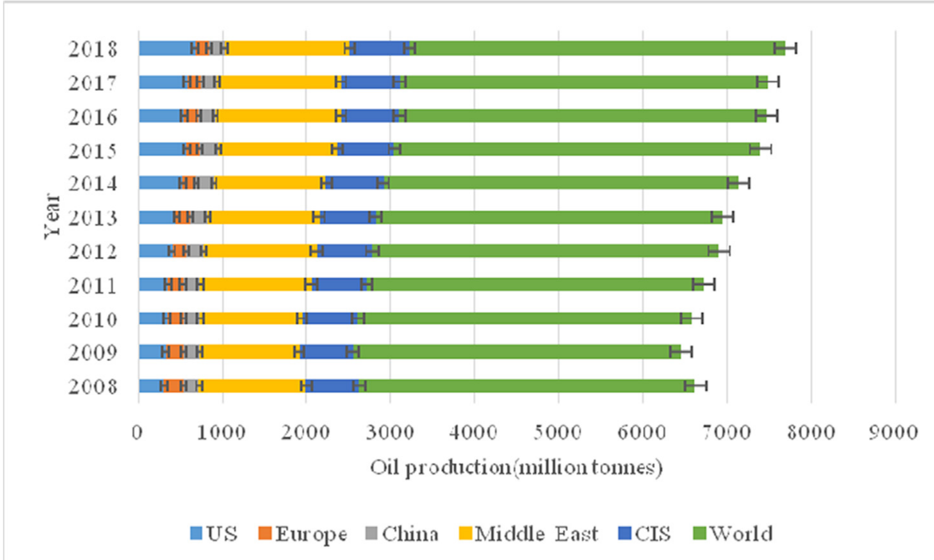

Fig. 2 Worldwide regional oil production in recent years (BP, 2019).

greatly due to numerous factors such as the oilfield type, soil composition, and storage conditions; thus, the physicochemical properties of oily sludge from different sources are not the same (Wang et al., 2019). The components of crude oil mainly include aliphatic hydrocarbons, aromatic hydrocarbons, asphaltenes, resins, etc., among which benzene, xylene, cycloalkanes, polycyclic aromatic hydrocarbons, and other volatile and refractory organic compounds are relatively common (Hu et al., 2013; Rudyk, 2018; Huang et al., 2014a); this is also one of the reasons why oily sludge is highly viscous, toxic, and acidic, and why its common form is highly stable oil-in-water (W/O) or water-in-oil (O/W) emulsions (Langevin and Argiller, 2015; Filho et al., 2012). Therefore, oily sludge poses a serious threat to the surrounding ecological environment and human health (Rudyk, 2018), and China (No.HW08, National Catalogue of Hazardous Wastes, 2007, Ministry of Environmental Protection of the People's Republic of China) and many other countries have identified the oily sludge as hazardous waste solids (Qu et al., 2019; Yang et al., 2019).

However, with the continuous increase in the consumption of petroleum products in recent years (Fig. 3) (BP, 2019), the harmlessness, resource recovery and reduction treatment of oily sludge have become one of the major problems in the petroleum industry. In the past decade, researchers have innovatively developed many disposal and treatment technologies to adapt to stricter environmental regulations and the increasing public concern about the environmental damage caused by traditional oily sludge disposal methods (Hu et al., 2013). Traditional oily sludge disposal and treatment methods, such as landfilling, incineration, solidification, and biodegradation, all have drawbacks, such as low efficiency, high cost, resource wastefulness, and a high risk of environmental pollution (Hu

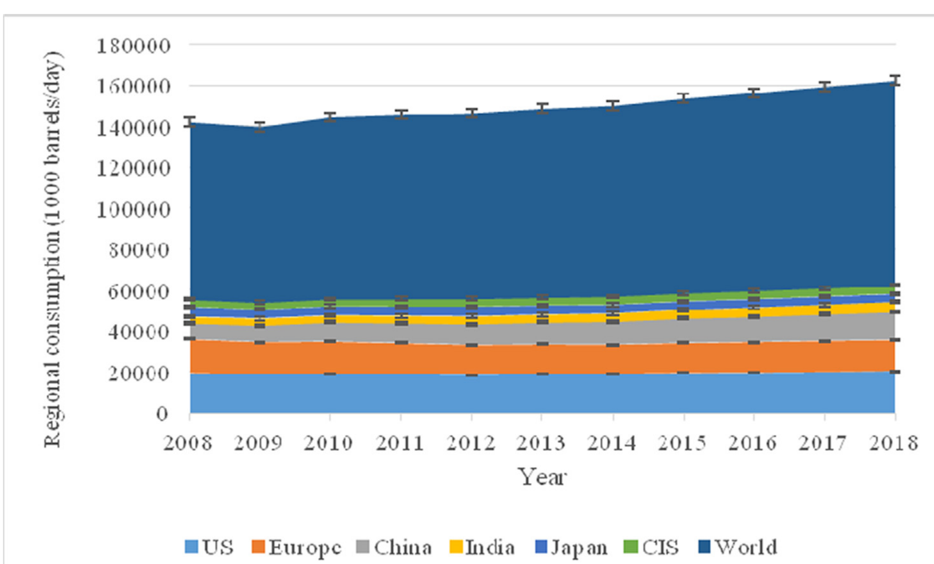

Fig. 3 Worldwide daily regional consumption of oil in recent years (BP, 2019).

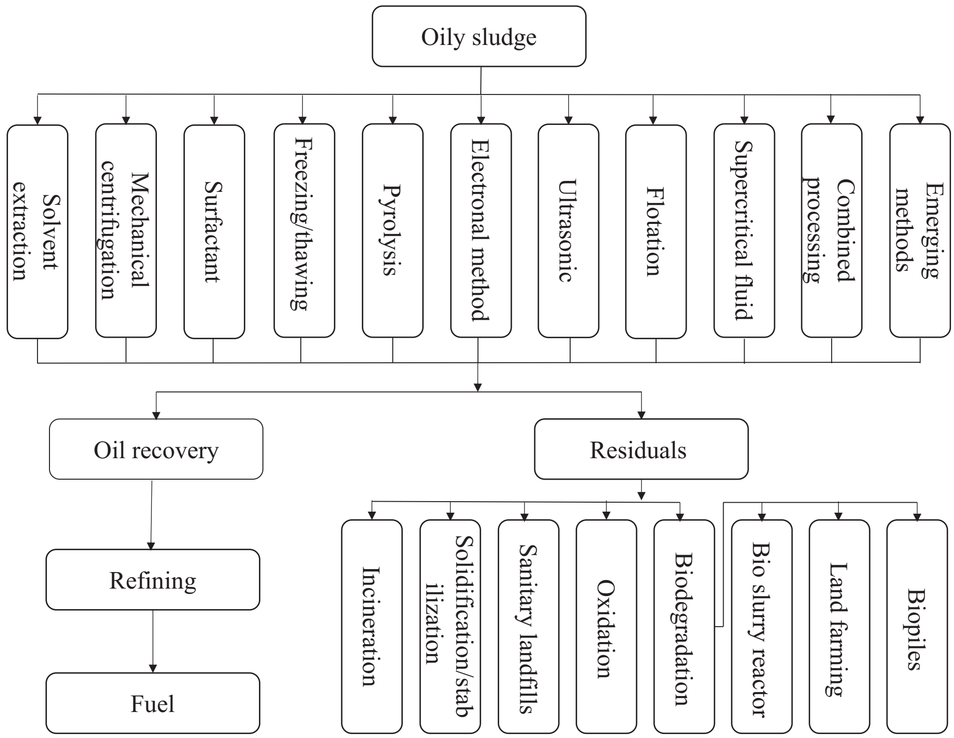

Fig. 4 A schematic diagram of the oily sludge disposal.

et al., 2020). For example, during landfill disposal, organic pollutants may spread through leachate or landfill gas to the surrounding environment and endanger both human and animal health (Luo et al., 2019; Gautam et al., 2019). Incineration disposal requires a large amount of fuel to aid in combustion, and will produce harmful refractory gas and ash (Deng et al., 2016; Zhou et al., 2009). For the use of curing methods to dispose of oily sludge, substantial amounts of cement and other curing agents are consumed, and the cost is high (Karamalidis and Voudrias, 2007). Voudrias Biodegradation treatment is both inefficient and time-consuming (Jasmine and Mukherji, 2015).

Oily sludge contains large amounts of oil emulsions, water, heavy metal ions, and other recyclable resources composed of hydrocarbons and refractory petroleum hydrocarbons, which are a renewable energy with high potential value (Hu et al., 2020). In addition, with the continuous updating of environmental friendliness, sustainability, and other eco-environmental development modes, recycling has become the optimal environmental solution for oily sludge disposal and treatment. The resource utilization of oily sludge can not only effectively reduce the disposal volume and pollution degree of hazardous waste solids, but can also reduce the use of non-renewable resources. According to a relevant report by the American Petroleum Institute (API), the main environmental factor that must be considered for oily sludge treatment is the maximum recovery and utilization of crude oil (Hu et al., 2013). Therefore, in recent years, increasingly more attention has been paid to the research on the technology for the recycling and harmless treatment of oily sludge; in particular, demulsification and other oil-water and solid-liquid separation processes can effectively separate the crude oil, water, and solid particles in oily sludge, which creates favorable conditions for the recovery of crude oil (Liang et al., 2017). In recent years, physical recovery and treatment technologies, such as solvent extraction (Zubaidy and Abouelnasr, 2010), and chemical treatment technologies, such as pyrolysis (Ma et al., 2014), have been applied in engineering, as have combined physical and chemical processing technologies. These technologies can not only be used to effectively recover crude oil resources from oily sludge, but also effectively reduce its harm to the human body and the surrounding ecological environment. However, the energy consumption, mechanism, recovery efficiency, and environmental hazards of various oily sludge recovery technologies are different, and there remain many shortcomings in their strict compliance with ecological environmental standards and economic practicality (Hu et al., 2013). Therefore, this paper introduces some popular technologies for the recovery of crude oil from oily sludge (Fig. 4), and discusses and summarizes their use conditions, mechanisms of action, advantages, and disadvantages, based on which the application prospects of these technologies are predicted.

2. Recovery technology of crude oil from oily sludge

Existing solvent extraction, mechanical centrifugation, surfactant, freezing/thawing, pyrolysis, electronal, ultrasonic, flotation, supercritical, combined processing and some emerging technologies as many as a dozen crude oil recovery methods. The following part summarizes and compares the development process, application conditions, advantages and disadvantages, and mechanism of action of various methods.

2.1. Solvent extraction

Solvent extraction is a technology that completely mixes the oily sludge with the extraction solvent in a certain proportion,

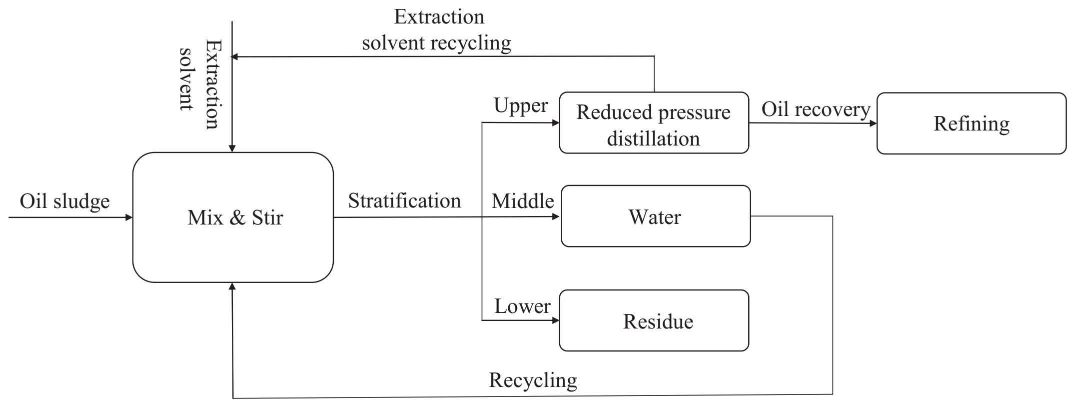

Fig. 5 Flowchart of recovery of crude oil by solvent extraction.

removes solid particles, water and other impurities in the mixture, distills the mixture, and separates and recovers the crude oil in the sludge from the extractant (Fig. 5). This method is based on the similar phase solution principle of solid-liquid two-phase extraction, that is, the solubility of different crude oil components in oily sludge is different in different extractants (Hu et al., 2017a). The method costs more than 300 US$/m^3, depending on the extraction agent, the extraction process, and the properties of the sludge (Hu et al., 2013).

In general, the amount of extractant used in solvent extraction methods is particularly important. For this reason, researchers (Zubaidy and Abouelnasr, 2010) have respectively used methyl-ethyl ketone and liquefied petroleum gas condensate as extractants, and have focused on the amount of the extraction agent. It has been found that, with a ratio of extractant to oily sludge of 4:1, the recovery rate of crude oil can reach up to 39%; however, the recovered oil was found to have a high sulfur content and some carbon residue impurities. Aiming at the problem of the existence of solid residue after extraction, Liang et al. (2014) used four different extraction solvents to investigate the influences of the solid concentration of oily sludge on oil recovery and the distribution coefficient. Their experiments revealed that the extraction process is not in an ideal state; instead, there is an interaction force between solid particles that varies with the change of the solid concentration, which ultimately results in the variation of oil recovery and the distribution coefficient. The results showed that the distribution coefficient of oily sludge can accurately describe the effect of the solid concentration on the solvent extraction process, which is essentially the adsorption and desorption behavior of crude oil between the solid and liquid phases. To investigate the desorption mechanism of oily sludge in detail, Zhao et al., 2018b used kinetic and thermodynamic equations to model and analyze the desorption isotherm of oil slime in the solvent extraction process, and found that the crude oil components existed in the form of a colloidal dispersion system. The desorption process is mainly controlled by two mechanisms, namely solvent effects, which can promote the rapid desorption of aromatic hydrocarbons, saturated hydrocarbons, and resin compounds, and residual effects caused by hydrogen bonding, electrostaticity, and π-π conjugation, which are also important for the desorption of asphaltenes and oily sludge. These effects are closely related to the pore structure of oily sludge and the channel effect of hydrogen bonds; the mesoporous structure will limit the desorption of oil, and the desorption efficiency and speed are mainly affected by the components of asphaltene and resin. Under the best extraction conditions, the recovery rate of crude oil has been found to reach 87.9% (Zhao et al., 2019). Identifying these mechanisms can aid in the improvement of the extraction process to improve the quality of the recovered oil, especially the recovery proportion of light components. In addition, comparative research has been conducted on different types of extractants. Nezhdbahadori et al. (2018) used the response surface method to compare the effects of different polar extractants on the recovery efficiency of crude oil. The experimental results showed that, at a temperature of 20 °C, a treatment time of 19 min, and a mixing ratio of 6.4/4.2, the optimal recovery efficiency of methyl-ethyl ketone extract was 30.41%, while that of toluene extract was 37.24%. It was found that the physical properties of the oily sludge itself, such as the petroleum hydrocarbon content and polarity, the metal ions, and the solid particles, had great influences on the extraction recovery efficiency. An extraction agent with the same polarity was found to be more efficient than an extraction agent with a different polarity. These results laid a solid foundation for the utilization of oily sludge.

On the whole, solvent extraction is one of the simple and efficient techniques. This technology can separate the oily sludge into crude oil and solid residue in a short time and effectively reduce the volume of oily sludge. However, in the design of the extraction tower for the treatment of a large amount of oily sludge, the solvent evaporation steam recovery unit needs to be added to prevent the secondary pollution of volatile petroleum hydrocarbon gas to the atmosphere. In addition, compared with other technologies such as pyrolysis and mechanical centrifugation, this treatment method also requires the use of a large number of organic extraction agents. So how to reduce the recovery cost and avoid the secondary pollution caused by organic solvents to the environment may be the focus of future research.

2.2. Mechanical centrifugation

Mechanical centrifugation mainly uses centrifugal force generated by high-speed rotation of centrifugal equipment to separate the components with different densities such as crude oil, water and solid impurities in the oily sludge (Fig. 6). According to the principle of centrifugal settlement, centrifugal force compels particles in oily sludge to move to the bot-

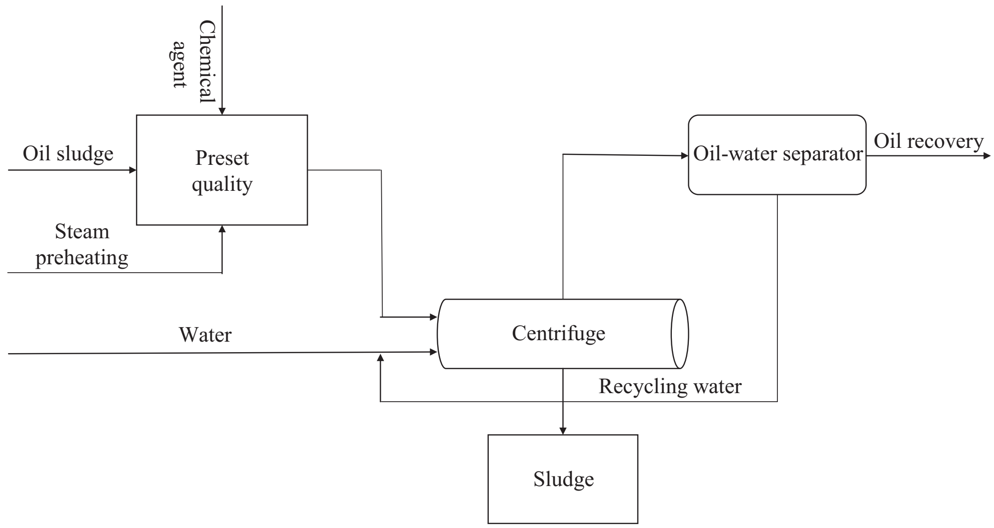

Fig. 6 Flowchart of recovery of crude oil by mechanical centrifugation.

tom of the centrifugation device in the radial direction during centrifugal operation. After centrifugation, the oily sludge will be stratified, the upper oil-water emulsion liquid can be recovered, and the lower solid particle impurities require further disposal (Pinheiro and Holanda, 2013). The centrifugal settlement distance of solid particles is strongly related to the viscosity of oily sludge, the particle size, and the centrifugation speed. Moreover, the larger the settlement distance, the easier the separation. Different centrifugation conditions are set depending on the characteristics of the sludge, so the application cost of this method ranges between 100 and 300 US$/m^3 (Drelich et al., 2010; Da Silva et al., 2012).

To explore the centrifugal settlement and separation process of solid particles in oily sludge, Huang et al. (2014b) proposed a model for the prediction of the behavior of solid particles in the centrifugal process based on the particle size distribution and oil viscosity, and evaluated two different types of oily sludge samples with a Sorvall ST 40 Centrifuge device at room temperature. In the experiment, solid particles were divided into three groups according to different particle sizes and viscosities. The experimental results were found to be consistent with the model predictions, and the water-oil emulsion recovery rates in each group were found to be 86%, 81%, and 75%, respectively. Large particles were found to be effectively removed by centrifugation, while small particles were retained in the upper water-oil emulsion. For medium-sized particles with a certain viscosity, preheating or preconditioning is required to reduce the viscosity and improve the removal rate of solid particles. The prediction model can effectively determine the optimum centrifugation time and pretreatment method. However, due to the high viscosity of oily sludge, pre-conditioning is required to reduce energy consumption and improve the efficiency of mechanical centrifugal treatment. For example, the addition of a demulsifier, surfactant, or other organic agents, or the conduction of preheating, ultrasonic irradiation, or other pretreatment methods, can further reduce the viscosity of the oily sludge and improve the convenience of subsequent centrifugal operation. For this reason, Shao et al. (2014) evaluated various pre-conditioning methods, and found that mechanical centrifugation after preconditioning and tempering is faster, is easy to operate, has a higher oil removal rate than biological treatment, and can be used to recover crude oil.

In a word, the recovery effect of this method is affected by many factors, such as centrifugal power, speed, time, temperature, type and concentration of preconditioned additives (Philemon and Benoit, 2013). Benoit Besides, compared with other methods such as surfactant method and solvent extraction method, the technology still has the problems of high cost, high noise and secondary pollution of organic agents. So it is necessary to further research and develop new green and efficient pre-conditioning agents, also improve the treatment process, as well as design and optimize more efficient centrifugal equipment.

2.3. Surfactant

The surfactant treatment is based on the principle of chemical cleaning, which uses the hydrophilic group of the surfactant and the water phase solution of the oil mixture to improve the solubility of petroleum hydrocarbon (Fig. 7). During oil recovery, hydrophobic groups will gather at the oil-water interface (Zhao et al., 2015), thereby reducing the viscosity and surface tension of the oily sludge, enhancing the migration ability of petroleum hydrocarbons between the oil and water phases, and ultimately achieving the purposes of oil-water demulsification, solid-liquid separation, and the recovery of crude oil (Seo et al., 2018). In fact, surfactants have been extensively used in the pretreatment stages of other recovery technologies, such as mechanical centrifugation (Shao et al., 2014), electric methods (Elektorowicz et al., 2005), and flotation methods (Da Silva et al., 2019), to increase the recovery efficiency of crude oil. However, as compared with these technologies, which are characterized by high costs and complex equipment, the surfactant method is simple to implement and does not require large mechanical equipment (Hu et al., 2013). Depending on the type and amount of surfactant used, the technology costs about 300 US$/m^3 (Lima et al., 2011). The methods can be categorized as either chemical or biological techniques according to the types of surfactants used.

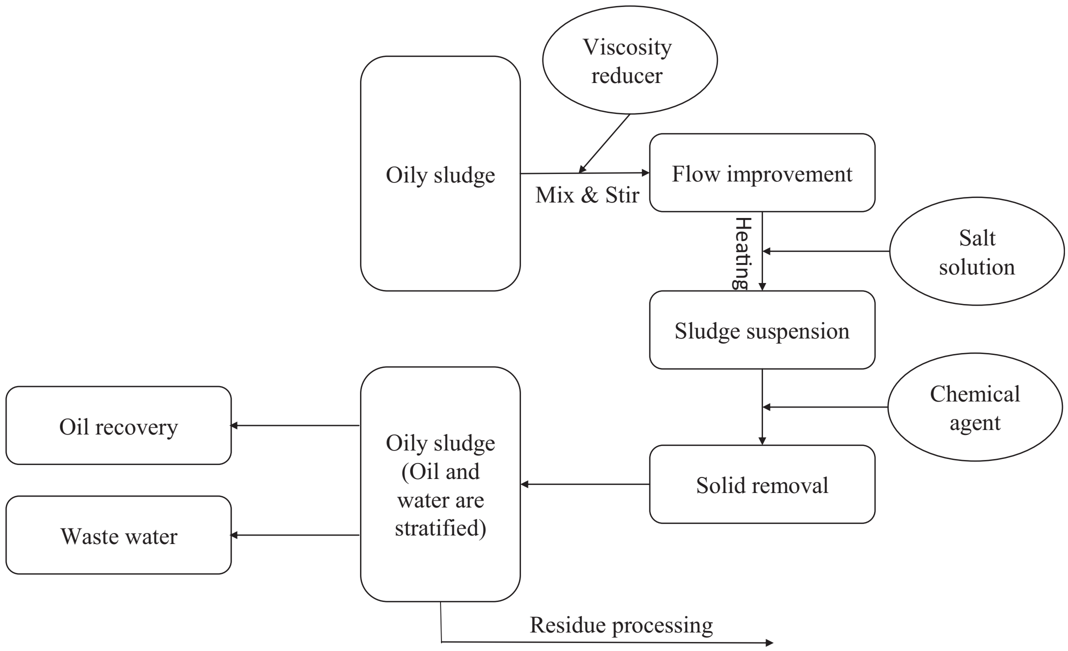

Fig. 7 Flowchart of recovery of crude oil by surfactant method.

2.3.1. Chemical surfactant

The chemical surfactant method uses various chemical surfactants to treat the oily sludge for the recovery of crude oil. Of course, for oily sludge with different properties, the chemical surfactants used are different. For example, Azim et al. (2011) mixed nonylphenol ethoxide, inorganic acid, and isopropyl alcohol in proportion to make a demulsification system for the treatment and recovery of petroleum sludge. The results showed that the sludge could be decomposed into different components; the oil phase was found to contain many recoverable petroleum hydrocarbons, but water and solid impurities also existed in the oil phase, which required further centrifugation to purify the recovered oil. To address the problem of residual solid particles after the use of this method, Liang et al. (2017) used four different surfactants to investigate the influences of the behavior of solid particles of oily sludge on the oil-water distribution coefficient and oil removal efficiency. It was found that the deoiling process of oily sludge via chemical agents was controlled by the solid concentration, and a set of SCA models was established to accurately describe this effect. The results revealed that the solid concentration and the experimental temperature can effectively improve the deoiling efficiency, and the SCA model can be used to analyze the deoiling behavior of oily sludge via the calculation of the effect of the solid concentration, which is helpful for the extraction and recovery of high-quality crude oil from the oil phase. Via comparison, it is evident that, regardless of whether the demulsification system is composed of a single surfactant or a mixture of various chemical agents, it will be affected by the solid concentration, and the strength of the effect of the solid concentration is not related to the surfactant, but changes with the variation of temperature. In addition to the effect of the solid concentration, the recovery efficiency of the chemical surfactant method is affected by many other factors. To maximize the recovery of crude oil, Ramirez and Collins (2018) investigated the characteristics of several commonly used surfactants (namely their activity, micelle concentration, and size), the applied dosage, and the proportion of oily sludge. The results indicated that the surfactant plays a decisive role in this method, as it can effectively reduce the interfacial tension of the W/O emulsion in the sludge, accelerate the rupture of the emulsion, and promote the stratification of oil and water. Moreover, the ratio of the surfactant to the oily sludge is very important for the improvement of the recovery rate of crude oil. The results showed that the use of Triton X-100 surfactant achieved the highest oil recovery efficiency at a lower concentration and dose. These findings are beneficial to the improvement of the cost performance of the chemical surfactant method and the reduction of recovery costs.

2.3.2. Biosurfactant

Because chemical surfactants are mostly mixed with organic solvents, they may cause secondary pollution to the environment and are difficult to biodegrade. Therefore, in recent years, researchers have turned to the study of environmentally friendly surfactants with good surface activity, low biological toxicity, good demulsification performance, and strong selectivity; among them, rhamnose tallow is the most widely used. Yan et al. (2012) used rhamnose tallow to recover crude oil from refinery sludge, which was found to directly recover 91.5% of crude oil from sludge under the optimal conditions. A pilot plant was also used to further verify the advantages of the surfactant, including its high dehydration property, strong operability, and low toxicity. In addition, Liu et al. (2018) investigated the solid-liquid ratio, operating temperature, stirring power, dosage of active agent, chemical cleaning time, and other influencing factors when using rhamnose fat to recover the oil sludge from the bottom of the tank. Under the best operating conditions, the recovery rate of crude oil was found to reach 83.17%, and the moisture content of oil could be less than 0.42%. However, regardless of the use of chemical or biological surfactants, the key step of oil recovery is effective

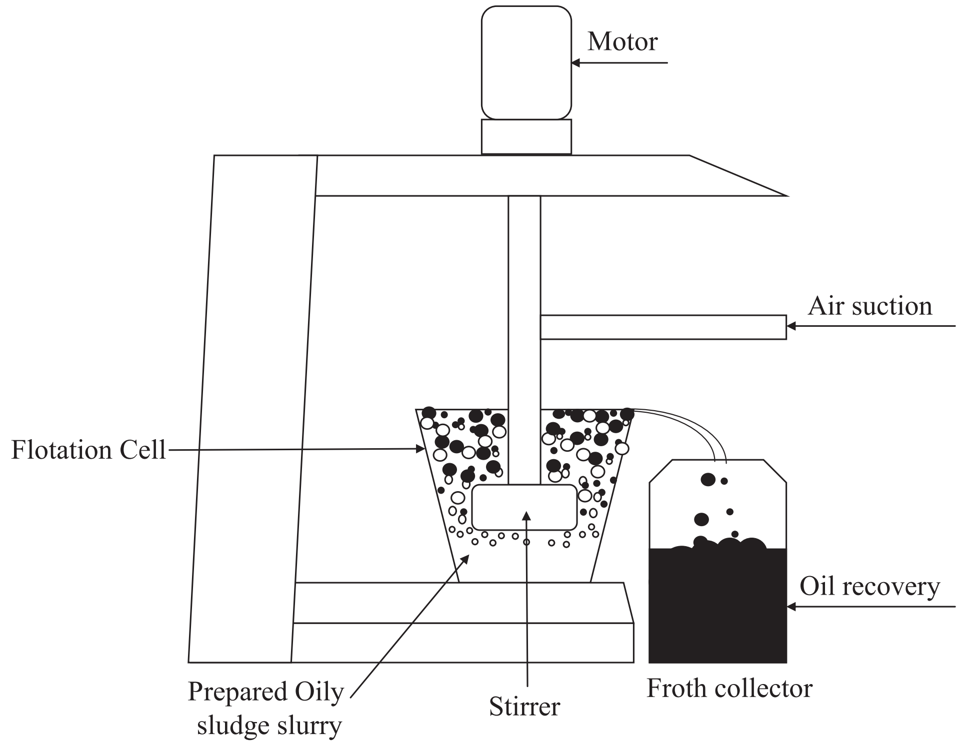

Fig. 8 Schematic layout of recovery of crude oil by flotation method.

demulsification. Therefore, Sahebnazar et al. (2018) introduced zero-valent iron nanoparticles to purify rhamnoides. By characterizing the critical micelle concentration and surface tension, the purity of rhamnose fat was increased to 83.33%. As compared with the unpurified rhamnose lipid surfactant, the results showed that the viscosity of the oily sludge could be reduced by 27.2%, which could effectively improve the demulsification degree of the oily sludge and increase the recovery rate of crude oil. However, to make better use of waste resources to reduce costs, Chirwa et al. (2017) reported a bacterial culture isolated from a waste oil field that could produce biological surfactants and be used as an inoculant in the recovery of oily sludge. After 10 days of operation in a fed-batch piston flow reactor (FB-PFR) system, the crude oil recovery reached 99.7%. After the purification of the crude biosurfactants produced by the culture, it was found via instrumental analysis that the molecular structure of the biosurfactants was highly compatible with lipid peptides. The results showed that this method can not only be used to recover crude oil from oily sludge, but can also degrade the aromatic organic impurities in the oil to produce cleaner oil products.

Overall, the surfactant method can be used to recover crude oil from oily sludge. As compared with mechanical centrifugation, solvent extraction, and other methods, the process is simple and convenient, does not require large and complex machinery and equipment, and is characterized by a large treatment capacity and high efficiency. Although this method has been used on a large scale, there are many factors that may affect its outcomes, including the type of surfactant, cost, effectiveness, toxicity, degradability, and recyclability, among others. In particular, biosurfactants, which have gained popularity in recent years, generally have a high cost of preparation, which may limit their large-scale application. Therefore, in the future, energy can be saved and costs can be reduced by recycling waste and increasing the production rate.

2.4. Flotation

Another method of oil recovery from oily sludge is the flotation method. The process principle of flotation is similar to that of the air flotation pool in sewage treatment (Li et al., 2016). In the air flotation device, the oily sludge, water, and surfactant are first mixed in proportion, and a liquid slurry is created by the demulsification of the surfactant. Then, air is injected to generate bubbles in the slurry. When the bubbles move to the surface of the slurry due to buoyancy, they will collide with the oil droplets, thus continuously thinning the water-phase film between the oil-phase film and bubble film until it finally breaks. The oil-phase droplets will then rapidly spread and attach to the surface of the bubble film. Because the density of the oil phase is less than that of the water phase, the bubbles attached to the oil-phase droplets will quickly float to the surface of the mixed slurry (Radzuan et al., 2016; Ramaswamy et al., 2007). After a period of time, the oil droplets floating on the surface of the slurry can be scraped off, collected, and further purified, ultimately achieving the purpose of crude oil recovery (Fig. 8).

In this method, surfactants act as foaming and trapping agents to separate the oil phase and solid phase from the slurry mixture to the surface foam. Some researchers have used 5 g sodium dodecylbenzenesulfonate as a foaming agent, and, after 12 min of air flotation operation, the recovery rate of the crude oil was found to reach 55%. When the amount of the foaming agent was increased to 20 g, the recovery rate was found to increase by another 12%. It was found that the air flotation efficiency increased with the amount of the foaming agent, but was not strongly related to the original oil content in the oily sludge (Ramaswamy et al., 2007). In fact, the key to the efficiency of this method lies in increasing the solubility of the oily sludge, thus reducing the difficulty of separating each phase of the slurry mixture and further optimizing the dehydration performance of the sludge. Therefore, before using the flotation method, Guo et al. (2011) acidified oily sludge, which then settled after it was allowed to stand for 120 min. When the pH value was decreased to 4, 77% of the water in the oily sludge was removed. The reason for this is that the acid destroyed the structure of the flocculation skeleton in the slurry, resulting in the release of a large amount of water inside. The solubility of the oily sludge was then increased, thereby facilitating the subsequent operation of crude oil recovery via air flotation. In addition to pre-acidification, Da Silva et al., 2019 also reported a microemulsion system for the solubilization of the oily sludge in the flotation device. The system comprises saponified coconut oil as a surfactant, n-butanol as an auxiliary agent, kerosene as an oil phase, and 2% sodium chloride solution as a water phase mixed in a certain proportion. When the ratio of the microemulsion to oily sludge was 4:1, after mixing at 60 °C for 1 h, more than 90% of the sludge was dissolved. This is of great benefit to the subsequent flotation separation operation, and can effectively improve the recovery efficiency of crude oil.

As mentioned previously, the most important factor of air flotation is the effective solubilization of oily sludge, and this process requires the strong cooperation of surfactants. Therefore, as compared with the surfactant method, the air flotation method does not have advantages in terms of its cost and operation convenience due to the need for complex air flotation treatment equipment. In addition, factors such as the flotation temperature, time, properties of the oily sludge, pH value, surfactant, and solvent addition can affect the flotation efficiency. Among these factors, the viscosity of oily sludge is greatly influential, so the operations of viscosity reduction and solubilization are crucial. However, at present, the recovery of crude oil from oily sludge via air flotation has mostly been conducted in laboratory studies. Furthermore, when the viscosity of oily sludge is too high, large amounts of water and surfactant are required for solubilization. This will produce an excess of wastewater, increase the cost of subsequent treatment and environmental risks, and hinder the large-scale application of this method.

2.5. Freezing/thawing

Another method of oil recovery from oily sludge is the freezing/thawing method. Due to the various compositions of petroleum hydrocarbons in oily sludge in different regions and the diverse freezing points of various hydrocarbons, the water-phase and oil-phase separation process of the freezing/thawing method can be generally divided into two situations. The first situation is that when the freezing point of the hydrocarbons is lower than that of water; in this case, the water phase in the oily sludge will first be cooled and frozen, and its volume will expand. At this point, the equilibrium of the two phases in the emulsion mixture will be gradually broken, and the oil phase will slowly begin to coagulate with the continued decrease of the temperature. In the subsequent thawing process, the dual effects of oil-phase gravity and surface tension will gradually stratify the oil-water emulsion with the water phase, resulting in the instability of the oil-water emulsion and ultimately achieving the separation and recovery of crude oil (Ghosh and Rousseau, 2009). The second situation is that when the freezing point of the hydrocarbons is higher than that of water; in this case, the oil phase will start to freeze earlier than the water phase. In addition, because the oil phase is less dense, ice formed by the freezing of the oil phase will cover the water phase. With the continued decrease of the temperature, the water phase will also begin to freeze, causing its volume to increase and the bursting of the reservoir. During the thawing process, the oil and water phases will be gradually stratified and separated due to the effect of gravity, after which the oil phase can be recycled (Lin et al., 2008).

In 1999, Jean et al. (1999) became the first to verify the feasibility of the freezing/thawing method for the separation of crude oil from oily sludge. Gas-mass chromatographic analysis and identification revealed that the method can be used to separate more than 50% of the oil content. Subsequently, He and Chen (2002) found via comparison that, during the lubricating pretreatment of oily sludge, more than 90% of the water could be removed with an efficiency significantly better than those of centrifugation, surfactant use, and ultrasonic and other methods, and was conducive to the recovery of oil products in a refinery. However, the rate of the freezing process of this method is too fast, which is not conducive to oil separation. Therefore, Chen and He (2003) further optimized the rate of the freezing process and found that the milder the freezing/thawing process, the better the dehydration effect, and the better the oil recovery. However, the COD content in the removal of wastewater was found to be relatively high, generally around 10,000–15,000 mg/L. In response to this problem, Feng et al. (2017) used the freezing/thawing method to recover oil from a waste cutting oil-water emulsion; in addition to further verifying the separation mechanism of the method, they also optimized the experimental conditions. It was found that the cyclic change of temperature during the freezing-thawing process could lead to the change of the oil and water phase, resulting in demulsification and the instability of the oil-water emulsion mixture with high stability, high viscosity, and multi-solid particles, and ultimately the separation of the two phases. Under the best operating conditions, the recovery of waste oil was found to reach 3700 mg/L, and nearly 80% of the COD content in the wastewater was removed.

In general, the demulsification of oil/water mixtures and the recovery of oil products are affected by a number of factors in the freezing/thawing process. For example, the temperature, time and rate of freezing and thawing, the content of water phase, oil phase, oxygen, impurities and so on. In addition, in the process of freezing and thawing, the temperature cannot rise or fall too fast, and it requires a lot of time and energy, in comparison with surfactants, mechanical centrifugation, flotation, etc., which makes the method more restrictive, more costly and less economical. So, this method has a certain application prospect in the case of high cold regions or natural freezing in winter.

2.6. Pyrolysis

In the pyrolysis method, oily sludge is separated via the pyrolysis and gasification of the organic components in the sludge at

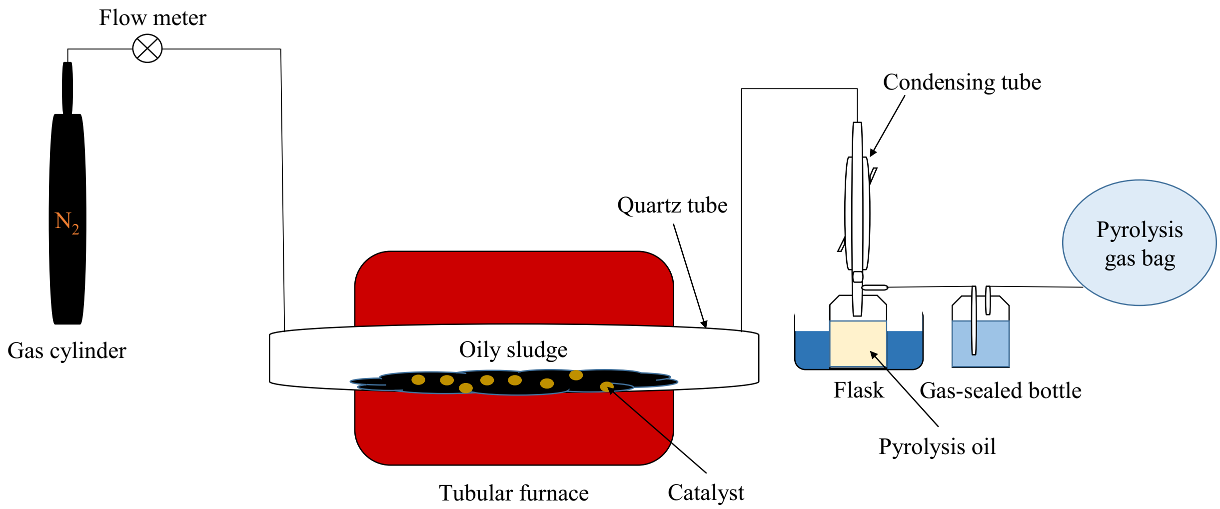

Fig. 9 Schematic layout of recovery of crude oil by pyrolysis method.

high temperatures with indirect heat transfer under anaerobic conditions. The pyrolysis gas is condensed into oil and recovered, while the remainder of the solid impurities are carbonized (Fig. 9). During this process, macromolecular hydrocarbons are pyrolytically converted into petroleum hydrocarbons with low molecular weight (Egazaryant et al., 2015). The mechanism of pyrolysis can be divided into four stages according to the temperature ranges. The first stage is the evaporation of water, which occurs in the temperature range of 300–380 K. The second stage is the vaporization of light organic components, which occurs in the temperature range of 380–590 K. The third stage is the cracking decomposition of medium and heavy organic components and carbonates, which occurs in the temperature range of 590–1100 K. The fourth and final stage is the reduction and decomposition of coke and other inorganic materials, which occurs in the temperature range of 1100–1200 K (Miao et al., 2019; Cheng et al., 2018). Pyrolysis methods can be roughly categorized into three types, namely ordinary pyrolysis, catalytic pyrolysis, and microwave pyrolysis. Depending on the pyrolysis process, the type of catalyst, and the characteristics of the sludge, the technology can cost anywhere from 300 to 500 US$/$m^3$.

2.6.1. Ordinary pyrolysis

The ordinary pyrolysis method mainly adopts the electric furnace heating way, and without additives, directly pyrolysis the oily sludge to recover the oil. For example, Schmidt and Kaminsky (2001) conducted an experiment on the pyrolysis recovery of oily sludge in a fluidized bed reactor. When the raw material characteristics and pyrolysis conditions were appropriate, up to 84% of the oil was recovered. It was found that the higher the temperature, the easier it was to decompose the sludge into light organic matter with a low boiling point. However, when the temperature was increased to a certain value, the oil recovery rate no longer increased. For this reason, Liu et al. (2009) studied the characteristic changes of the common pyrolysis process of oily sludge via thermogravimetric and elemental analysis. They found that the faster the pyrolysis heating rate, the higher the proportions of carbon and sulfur in the solid residue of the oily sludge; however, this was found to be accompanied by the decrease of the hydrogen content. The results revealed that the recovery rate of petroleum hydrocarbons changed significantly under the influence of the heating rate, and nearly 80% of the total organic carbon in the oily sludge was converted into petroleum hydrocarbons. Subsequently, Ma et al. (2014) conducted a rapid pyrolysis experiment of oily sludge in a rotary kiln reactor. The results showed that, when the pyrolysis temperature was 550 °C and the mixing ratio was 1:2, the recovery rate of the pyrolysis oil could reach 87.9%, and its components were found to be mostly linear alkanes with a low molecular weight. By comparing the infrared spectra of the extracted oil, it was found that the pyrolysis process is more conducive to the formation of long-chain n-alkanes and 1-olefin.

2.6.2. Catalytic pyrolysis

In recent years, with the vigorous development of unconventional oil and gas field resources, the viscosity of oily sludge has become increasingly higher, and the components have become increasingly more complex. Therefore, the quality of oil recovered by the ordinary pyrolysis method has been unable to meet the requirements of reuse. To improve the quality and selectivity of recovered oil and further reduce the use cost, researchers have developed a variety of catalytic pyrolysis technologies based on common pyrolysis. For example, Shen et al. (2016) introduced the catalytic pyrolyzation-reforming process into a method of the recovery of fuel oil from heavy oily sludge. In this process, a small amount of oily sludge was first burned into ash residue, which was then mixed with several catalysts and added to a reactor. The experimental results revealed that the oily recovery rate reached 35.5%. In addition, the addition of an ash catalyst not only reduced the boiling point range of the recovered oil, but also improved the quality of the oil. However, to obtain recovered oil products with better quality, Cheng et al. (2017) proposed an oily sludge pyrolysis process that combined steam injection and the addition of sludge ash. The steam injection was found to effectively inhibit the secondary cracking reaction and prevent the oil from further cracking into gas. The light oil/heavy oil ratio was also found to decrease with the increase of the steam flow; therefore, the steam injection improved the stability of the recovered oil and the proportions of the heavy and medium fractions. However, when the injected steam flow rate was increased to a certain extent, the output of coke was found to slowly increase, i.e., excessive steam injection led to a decrease in oil production. The addition of sludge ash can convert the heavy fraction into the light fraction and reduce the carbon content. The synergistic effect of steam injection and the addition of sludge ash was found to improve the oil recovery and quality. In the subsequent comparative experiments of various oily sludges, the characteristics of sludge at different stages of pyrolysis were studied, and the effects of the addition of sludge ash were thoroughly analyzed. It was found that the activity of ash was mainly manifested in the second and fourth stages of pyrolysis process, which provides a reference for the optimization of pyrolysis processes in the future (Cheng et al., 2018). Some researchers have tried to use CO_2 as a gas reaction medium when conducting oily sludge pyrolysis experiments. The results have indicated that CO_2 can accelerate the pyrolysis and dehydrogenation of hydrocarbons in oily sludge and increase the production of light components such as H_2, CH_4, and C_2H_6. The formation of CO can effectively inhibit the production of benzene series and reduce the aromaticity of recovered crude oil products (Kim et al., 2019). Moreover, catalysts (e.g., activated alumina, natural zeolite, ZSM-5, HZSM-5, KOH, dolomite, etc.) commonly used in the refining industry have also been introduced to the pyrolysis process. For example, Lin et al. (2017a) added a KOH catalyst during a pyrolysis experiment of oily sludge to improve the quality of the recovered oil and the recovery rate of straight-chain hydrocarbons. Then, a Zn/HZSM-5 zeolite catalyst was used to selectively recover aromatics. The results showed that the total aromatics yield increased from 48.7% to 92.2% with the increase of the residence time from 1.0 s to 7.6 s (Lin et al., 2017b). Moreover, Shen et al. (2016) analyzed and compared the characteristics of five different catalysts, namely oil field sludge ash, oil tank sludge ash, activated alumina, natural zeolite, and ZSM-5 molecular sieve, as well as their catalytic effects on the pyrolysis of oily sludge. The experimental data revealed that the catalytic effect of sludge ash was the best and its oil recovery rate was the highest, which is primarily because the ash contains sulfur and iron. The oil tank sludge ash and ZSM-5 molecular sieve catalysts were found to have great influences on the boiling point range of the recovered oil, which is helpful for the improvement of the quality of the oil. Researchers have also explored innovations in the selection and optimization of catalytic pyrolysis treatment equipment to improve the oil recovery efficiency. For example, Gao et al. (2018a) designed a new type of fixed-bed reactor that uses a ceramic membrane to conduct the pyrolysis of oily sludge. The ceramic membrane installed in the pyrolysis reactor has a filtering role in the entrainment of particles in the adsorbed volatiles. For the use of this reactor, the pyrolysis temperature of 500 °C was found to be the best operating condition, and the highest recovery rate reached 52.95%. Lin et al. (2018a) developed a special U-shaped reactor for the continuous catalytic pyrolysis of oily sludge using dolomite. Due to the promoting effect of the dolomite catalyst on the dehydrogenation and polymerization of petroleum hydrocarbons, the highest oil recovery rate was found to reach 76.6%, and the main components were determined to be saturated light oils, such as aromatic hydrocarbons.

However, with the stricter requirements of ecological protection and the continuous development of green economy, researchers have begun to study the use of biomass and other organic waste as catalysts, and have conducted co-pyrolysis reactions with oily sludge to improve the utilization of waste solids and oil recovery. For example, Hu et al. (2017b) and Lin et al. (2018b) respectively studied the co-pyrolysis reaction of wood chips, rice husks, and oily sludge, and the results revealed a synergistic effect between biomass and oily sludge during co-pyrolysis. The contents of saturated and aromatic hydrocarbons in the recovered oil were found to be increased by 55% and 86%, respectively, the contents of resin and asphaltene were found to be reduced by 31% and 68%, respectively, and the relative content of oxygen-containing compounds was found to be significantly reduced by 46–93%. Via further analysis, it was found that this synergistic mechanism originated from the catalysis of alkali metals and ash in the biomass, which promoted the secondary reaction of recovered oil liquid products, thus producing more H_2, CO, CO_2, and C_1-C_2 hydrocarbons. In addition, the synergistic effect was also found to inhibit the release of hydrogen sulfide, promote the total distribution of sulfur in oil and gas products, improve the quality of oil products, and reduce the risk of environmental pollution caused by recycling.

2.6.3. Microwave pyrolysis

In addition to the ordinary and catalytic pyrolysis methods, microwave pyrolysis technology has also been developed in recent years. This technique uses electromagnetic waves to heat particles of oily sludge and promotes the movement and collision of particles. It can not only rapidly increase the temperature of oil-water mixtures and accelerate demulsification processes, such as the separation of oil-water molecules (Martínez-Palou et al., 2013; Abdurahman et al., 2017), but can also decompose the large molecules of petroleum hydrocarbons into small molecules. Moreover, the transfer of acid and solid particles from the oil phase to the water phase is promoted, thereby improving the quality of the recovered oil (Silva et al., 2014). For example, Lin et al. (2017c) used microwave pyrolysis technology to treat oily sludge from oil refinery wastewater. In the experiment, the rapid microwave heating organically combined water evaporation with carbonation, thereby accelerating the conversion of sludge biomass, increasing the yield of gaseous products, reducing the residue of solid impurities, and effectively enriching trace metals. When the pyrolysis temperature was 600 °C, the yield of recovered oil reached 33%. Hou et al. (2018) further demonstrated the advantages of microwave heating by comparing the effects of both microwave and ordinary electric furnace heating on oil-based drilling cuttings. The results demonstrated that microwave heating can promote the pyrolysis of petroleum hydrocarbons better than can electric heating, and is beneficial to the recovery of oil products.

In summary, in the ordinary, catalytic, and microwave pyrolysis processes, the efficiency of oil recovery is affected by the pyrolysis temperature, catalyst, reactor, and other treatment conditions. The advantage of pyrolysis methods is not only that the recovered oil is liquid, which is convenient for storage and transportation, but also that the quality of the oil is similar to that of the low-quality distillate oil of refineries, and can therefore be directly used in diesel engines. Moreover,

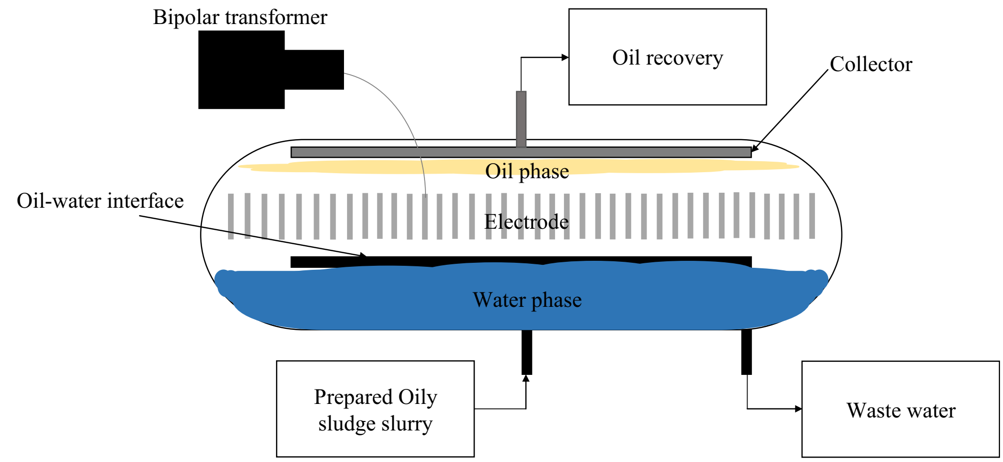

Fig. 10 Schematic layout of recovery of crude oil by electronal method.

pyrolysis produces much less harmful waste gas than incineration. Currently, pyrolysis has large-scale engineering applications. As compared with other recovery technologies, such as solvent extraction, mechanical centrifugation, surfactant use, air flotation, freezing/thawing, etc., pyrolysis technology is more efficient, faster, has a larger processing capacity, and is more widely used. However, it should also be noted that pyrolysis is worthy of further study to improve the yield and quality of the recovered oil and reduce the contents of harmful substances, such as polycyclic aromatic hydrocarbons. Moreover, in terms of equipment simplification and the reduction of costs, energy, and consumption, especially the reduction of the moisture content of oily sludge and the pyrolysis temperature, the future market prospects are very broad.

2.7. Electronal method

The electronal method uses the current generated by the electrodes to act on the oil colloidal mixture, which forms an electroosmotic pressure in the mixture that causes ions or charged particles to migrate to the corresponding electrode (Fig. 10). When using this method, the electric field generated by the current will break the original oil emulsion in the aggregation state, forcing the solid particles to slowly migrate to the anode under the action of electrophoresis, while the liquid phases, namely oil and water, migrate to the cathode (Ali and Alqam, 2000; Elektorowicz and Habibi, 2005). Then, in the anode region, due to the presence of the electroosmotic pressure, the solid particles that have been separated will collide and coagulate, gradually forming a precipitate. In the cathode region, due to the effects of molecular and electrostatic forces, the water and oil droplets in the oil-water emulsion mixture separate and coalesce separately. Finally, the two phases of water and oil are gradually formed to achieve the goal of oil recovery (Mhatre et al., 2015; Ghazanfari et al., 2012; Pamukcu et al., 2016).

When this method is used, the effects of positive and negative electrodes should not be underestimated; therefore, Elektorowicz et al. (2006) studied the effects of different potential gradients on oily sludge. The results showed that a lower potential (0.5 V/cm) could produce a higher demulsification rate, and that the solid phase transition would become denser and more stable. Through pH changes, resistance evolution, and hydrocarbon polarity analysis, it was confirmed that the smaller the potential gradient, the longer the exposure time, the better the phase separation effect, and the better the recovery operation. The continuous control of the electrical process and a prolonged processing time is the key to maintaining efficient phase separation, but operation control remains difficult for long-term, stable, and low-potential output. For this reason, Jahromi et al. (2018) developed a new controller based on seepage theory and installed it in the electrical equipment system. The treatment of oily sludge was found to prolong the exposure time of the electric field and effectively reduce the energy consumption, and experiments demonstrated that the controller can enhance the demulsification effect of oily sludge and improve the quality of separated fractions. To further understand the surface characteristics of sludge solids and the electric-induced reaction mechanisms of different electric fields, Kariminezhad and Elektorowicz (2018) comprehensively compared the physical and chemical characteristics and thermal behavior changes of solid particles during the electronal treatment of oily sludge with four different types of currents. It was found that the applied electric field improved the migration ability of molecules and oil recovery. The reaction of electrolytic products could cause the surface properties of solid particles to change, but, under the combined action of the oil-phase electroosmotic flow caused by viscous coupling (Pamukcu et al., 2016) and the electrophoretic movement of the solid to the anode, the oil-phase accelerated separation. Based on these findings, various additives were introduced into the electronal treatment of oily sludge by Taleghani et al. (2019) to improve the recovery rate of crude oil. Their experiment proved that, when the constant potential/voltage gradient was 1 V/cm, an appropriate addition of $FeCl_3$ solution enhanced the electrocoagulation effect. The turbidity of the water phase was significantly reduced, and the recovery rate of light oil was increased to 52%. These research results can therefore be applied to the upstream and downstream oil industry.

However, the use of electronal method to recover crude oil from oily sludge is affected by pH, potential, resistance, additives, voltage gradient, and other conditions. As compared with mechanical centrifugation, pyrolysis, solvent extraction, and other recovery technologies, this method is characterized by high efficiency and low energy consumption. In particular,

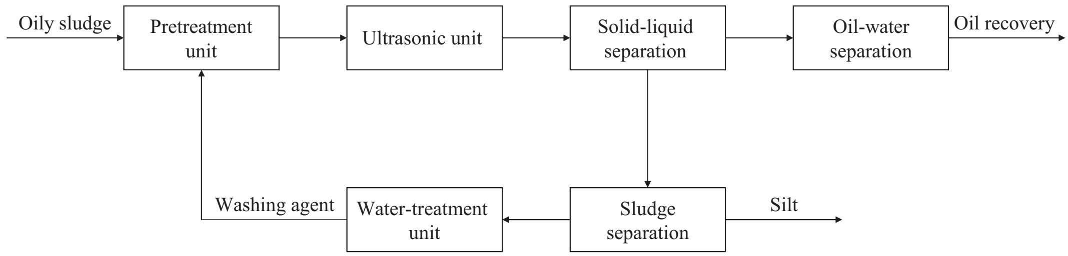

Fig. 11 Flowchart of recovery of crude oil by ultrasonic method.

the sludge collection tank can be directly transformed into an electric battery for recovery and treatment by the electronal method, which can greatly reduce the cost of equipment manufacturing and use. In addition, the energy consumption cost of the electronal method modified by the new controller can be as low as 2.22 Canadian dollars per ton of sludge (Jahromi and Elektorowicz, 2018). However, most research on this method has been conducted in laboratory environments. Moreover, the recovery efficiency is relatively low for oily sludge with high viscosity and solid particle impurities. Therefore, it remains necessary to further expand the scale of pilot tests or even field applications, and also to improve and optimize the electric processing equipment, to verify the advantages of this process.

2.8. Ultrasonic

In the ultrasonic method, ultrasonic waves are used to change the physical properties and state of oily sludge. The cavitation effect and mechanical vibration caused by the acoustic radiation will strip the crude oil droplets originally attached to the surface of the solid particles, thereby reducing the amount of crude oil on the surface of the solid particles. These processes mainly rely on cavitation, especially at the solid-liquid boundary (Hamdaoui and Naffrechoux, 2007; Xu et al., 2009). Under the continuous irradiation of ultrasonic waves, the viscosity of oil-water emulsions decreases continuously. The small droplets in the emulsion mixture accelerate the movement and collide and coalescence, ultimately achieving the purpose of separating the aqueous phase and the oil phase to recover the crude oil (Hamida and Babadagli, 2008; Xie et al., 2015) (Fig. 11). In addition, ultrasonic waves are penetrative to a certain extent, and oil droplets or impurities can be removed from the pores of the sludge solids, thereby facilitate subsequent recovery operations.

However, the use of the ultrasonic method is influenced by a number of factors, including the sound frequency, intensity, treatment time, temperature, sludge properties, additives, and salt or other impurities. For example, Abramov et al. (2009) investigated the influences of these factors when using the ultrasonic method to recover crude oil and asphalt from oil sands and tar sands. The results showed that the physicochemical properties of the oil sands, the properties of the solid particles, the concentration of additives, and the temperature play decisive roles in the recovery of crude oil. However, Wang et al. (2018b) used the ultrasonic method to conduct dehydration experiments on heavy oil in a high-temperature static state, and the effects of the temperature, ultrasonic frequency, intensity, treatment time, and precipitation time on dehydration were also verified. It was found that the increase of the temperature is beneficial to the ultrasonic dehydration of heavy oil, but the ultrasonic frequency is inversely proportional to the dehydration rate. When the ultrasonic intensity was increased, the dehydration rate of heavy oil was found to increase; however, with the continued increase of the intensity of the sound wave to the threshold, the rate of dehydration stopped increasing and gradually began to decrease. A too-long ultrasonic irradiation time is not conducive to the dehydration of heavy oil, and the rate of ultrasonic dewatering was found to decrease with the increase of the settling time. To solve this problem, the "two-stage ultrasonic irradiation process with equal irradiation time" (Check, 2014) was developed. In this process, ultrasonic waves with a power of 75 W were used for primary irradiation, while ultrasonic waves with a power of 50 W were used for secondary irradiation. The duration of both stages was 45 s. Experimental results demonstrated that the process can effectively reduce the settling time, and the dehydration rate of heavy oil was found to reach 96%; additionally, the salt content was reduced to 2.56 PTB, which improved the maneuverability of the further recovery of crude oil. To determine the strength threshold of the ultrasonic method, some researchers optimized the ultrasonic power to 0.24 W/cm^2 in a pilot experiment of the use of this method to treat oily sludge. These ultrasonic waves broke through the deoiling threshold at a frequency of 25 kHz and increased the crude oil recovery from 46% to 60.7%. Moreover, the combination of a surfactant, such as sodium sulfonate, and ultrasonic waves was found to further increase the recovery rate of crude oil to 82–90% (Gao et al., 2018b).

To sum up, compared with the recovery technologies such as freezing/thawing method, electronal method and surfactant method, ultrasonic method has the advantages of short treatment time, good viscosity reduction effect and no secondary pollution. However, although this method has entered the pilot scale application stage, we still need to clearly see that, large ultrasonic emission equipment, high cost of use and maintenance, will be the biggest obstacle to the technology's popularity in the future.

2.9. Supercritical fluid

The method refers to the use of supercritical fluid to treat oily sludge in order to recover crude oil and other useful resources. According to the different treatment methods, it can be divided into supercritical oxidation and supercritical extraction. The common supercritical fluids are water, ethane, ethylene and carbon dioxide.

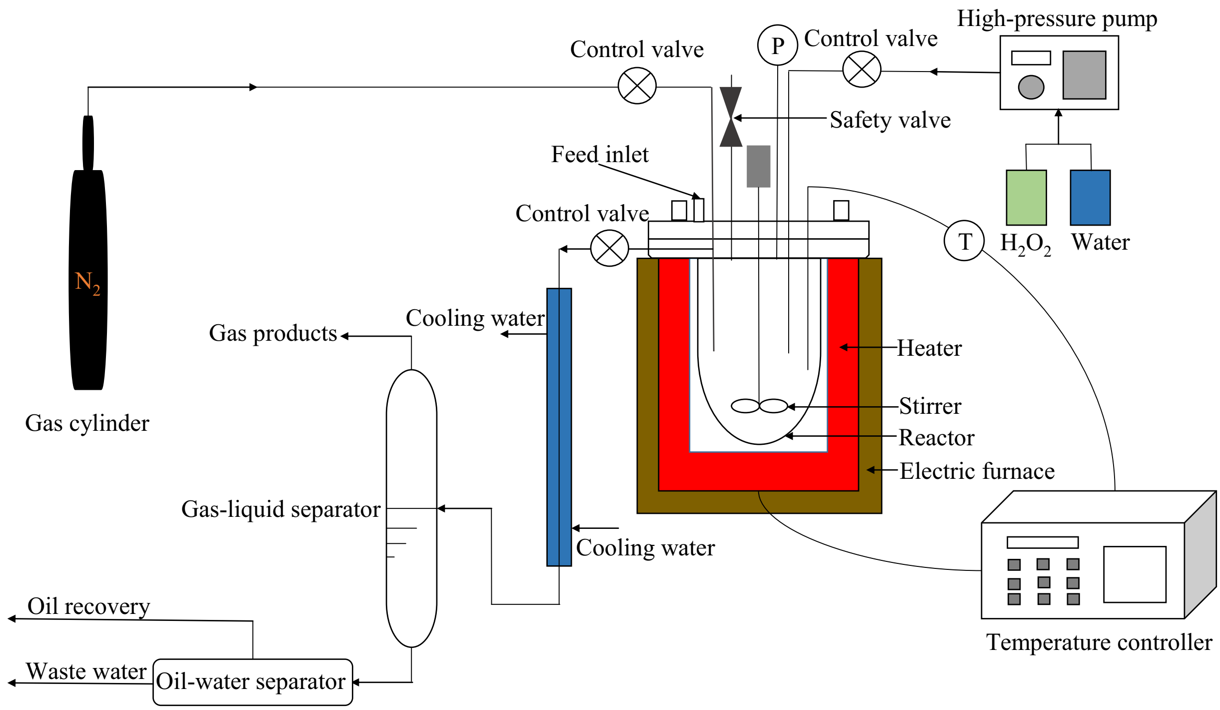

Fig. 12 Schematic layout of recovery of crude oil by supercritical oxidation method.

2.9.1. Supercritical oxidation

Supercritical oxidation refers to the use of supercritical fluid oxidation to treat oily sludge to reduce both the viscosity of the oil phase and the content of heavy component from the sludge recovered after separation (Fig. 12). Supercritical water is often used as a medium for oxidation reactions, and is characterized by an ultra-low viscosity, no surface tension, a high diffusion rate, and high solubility for organic compounds (Savage, 1999). It can transform and upgrade the crude oil emulsifiers in the oily sludge into useful oil products, and each phase of the oily sludge mixture becomes a homogeneous single phase. This can overcome the limitations of material transfer between phase interfaces to reduce the contents of recombination fractions, surfactants, and impurities (Cui et al., 2009). To further clarify the mechanism of the supercritical water oxidation method, Cui et al. (2011) treated oily sludge by a parallel-chain reaction of supercritical water oxidation. The oxidation mechanism was found to be roughly divided into three steps. The first step is the decomposition of the conjugated double bond of the macromolecular organic matter into the intermediates of aliphatic ketones, aldehyde, and carboxylic acid. The second step is the decomposition of these intermediates into small organic acids. The final step is the decomposition of these organic acids into water and carbon dioxide. According to experimental data, four lumped kinetic models were summarized to describe the reaction process of the oxidation of oily sludge by supercritical water. The predictions made by the model were found to be in good agreement with the actual results, which is helpful for the improvement of the performance of the batch reactor. Based on the same technology, a pilot experiment of supercritical oxidation for oil recovery was conducted, and it was found that the use of supercritical water as a solvent and dispersant could oxidize the recombination residue in the oily sludge into light component fuels, such as oil and natural gas mixtures, by various free radical reactions. The recovery rate of oil was reported to reach 18.6 wt%, and a higher temperature resulted in a deeper reaction degree, and thus a higher recovery rate of crude oil (Radfarmia et al., 2015). However, in recent years, the exploitation of unconventional oil resources with high viscosity, density, and acidity has increased (He et al., 2015), thus increasing the proportions of heavy metals and heavy components in the sludge and increasing the difficulty of crude oil recovery. For this reason, Khan et al. (2019) developed a new method of using supercritical water to treat oily sludge and recover crude oil. At a temperature of 400 °C, a pressure of 30 MPa, and a 16.7 wt% content of emulsion, the recovery rate of oil products was found to reach 78.6 wt% after reacting for 60 min. After the upgrading and detoxification of the oily sludge, the contents of heavy metals and heteroatoms were significantly reduced, including a 79% reduction of vanadium and a 23% reduction of sulfur.

2.9.2. Supercritical extraction

In addition to supercritical oxidation, supercritical fluid extraction is also used to recover crude oil from oily sludge. The method is based on the advantage of supercritical fluid regarding its two-phase, gas-liquid properties. When approaching the critical point of the fluid, the solubility of each component will change with the change of the temperature and pressure. In this way, the solubility of each component and the selectivity of the extraction solvent can be easily regulated, thus facilitating the separation and recovery of useful resources in the oily sludge (Oliveira et al., 2011). Supercritical ethane and carbon dioxide are characterized by non-toxicity, no residue, non-inflammability, easy separation, and high solubility; they can dissolve and extract the non-polar components of crude oil, and they are therefore often used as reaction media in this method (Khanpour et al., 2014). For example, non-polar supercritical ethane fluid has been used to extract the oily sludge at the bottom of the tank. Under the optimal conditions, the petroleum hydrocarbon recovery rate was found to reach up to 58.5 wt%. After analysis and characterization, it was found that the recovered hydrocarbons were significantly better than the original oily sludge, and could be further refined and purified for fuel use (Ávila-Chávez et al., 2007). Recently, a method of crude oil recovery from waste oil-based mud via supercritical CO_2 extraction was reported. Under the conditions of an operating temperature of 35 °C, a pressure of 20 MPa, and a mass ratio of fluid to oil-based mud of 3, the recovery rate of crude oil after 60 min of extraction reached 98.6%. The longer the extraction time, the higher the efficiency; however, when the extraction pressure and density of the supercritical fluid increased simultaneously, the extraction efficiency first increased and then gradually decreased. Moreover, light petroleum components recovered from oil slurries, such as n-alkanes, can be reused in drilling operations to reduce drilling costs (Ma et al., 2019).

In summation, both supercritical oxidation and supercritical extraction are affected by the physical properties of the oily sludge, the type and amount of supercritical fluid, the type of reactor, the reaction temperature, the pressure, and other factors. Among these factors, temperature and pressure are more important when maintaining a supercritical state. Compared with traditional recovery technologies such as pyrolysis, surfactant use, mechanical centrifugation, and solvent extraction, supercritical oxidation and extraction are characterized by high efficiency, environmental friendliness, recyclability, and high cost performance, and they have thus attracted increasingly more attention. However, supercritical fluid places high requirements on the reactor and may cause the corrosion of the reactor after long-term use. In addition, because the saturated vapor pressure may conflict with the change of the critical fluid density, the extraction efficiency will increase and then decrease with the continued increase of the extraction temperature, which may become a difficult point for future researchers to overcome.

2.10. Combined processing

As various methods for the recovery of crude oil from oily sludge have become increasingly more mature, the advantages and disadvantages of various technologies have been gradually discovered and verified. For example, the recovery rate of crude oil is not ideal when high viscous oily sludge is treated by freezing/thawing method (Jean et al., 1999); however, the pyrolysis method (Schmidt and Kaminsky, 2001) has higher efficiency and cost in the treatment of highly water-cut oily sludge. Therefore, researchers have considered complementary advantages and creatively merged various methods to develop a series of combined processing methods. For instance, to overcome the lack of the driving force of highly viscous oily sludge in the freezing/thawing process, Zhang et al. (2012) used a combination of the ultrasonic method and freezing/thawing method to recover crude oil from refinery oily sludge. The results showed that the energy generated by the ultrasonic wave method promoted the rapid separation of oil droplets and solid particles, while the freezing/thawing method promoted the separation of oil droplets and water in the system. As a result, the combined process achieved a much higher crude oil recovery rate than did a single sludge treatment method. Similarly, Hu et al. (2015) introduced a combination of solvent extraction and freezing/thawing to improve the recovery efficiency of crude oil and solvents, as well as the quality of crude oil, based on its high water content. In addition to the combination of various recovery technologies, some scholars have combined the technologies for the recovery of crude oil and waste treatment to reduce the contents of heavy components of sludge and further improve the recovery efficiency of crude oil. For example, a combined ultrasonic and Fenton reaction process has been used to treat oil spill sludge, and it was found that the ultrasonic method plays a role in dissolution, while Fenton reactions can oxidize large molecules of petroleum hydrocarbons into light oil products for recycling and reuse (Sivagami et al., 2019). Moreover, Guo et al., 2014 combined Fenton reactions and biomass technology to dehydrate oily sludge; biomass was added to absorb iron ions from the sludge and some organic matter, which was found to be beneficial to the subsequent recovery of waste oil. Due to the increasingly complex compositions and higher viscosities and emulsification degrees of oily sludge recently produced by some unconventional oil and gas fields, the combination of only two technologies cannot meet the requirements of recovery treatment. Therefore, Su et al. (2019) combined the ultrasonic, microwave, and chemical treatment processes for the treatment of highly emulsified oil mud in a gas field. It was found that the combined physical-chemical method exhibited a synergistic effect, and greatly improved the dehydration rate of oil mud and created conditions for the recovery of crude oil.

Overall, as compared with single treatment technologies, combined treatment methods can integrate the advantages of various recovery technologies, thereby effectively improving the recovery efficiency of crude oil. However, most of the existing research on combined treatments has focused on the separation and recovery of crude oil from oily sludge. Although the resource recovery and reduction of sludge can be realized, the research on how to deal with the residual sludge, and how to reduce the harm to the environment, is insufficient. In addition, after combining the advantages of two or more technologies, combined methods may also have the disadvantages of increased equipment manufacturing and maintenance costs, tedious operation, and long processes. Therefore, in the future, combined methods should focus on the improvement of process design, the optimization of equipment manufacturing, and the recovery and treatment of other harmful substances in the sludge, such as heavy metal elements and heteroatomic compounds.

2.11. Emerging methods

The technology for the recovery of crude oil from oily sludge has gradually matured, and the determination of how to recover crude oil efficiently, quickly, without pollution, and at a low cost has become a focus of technological research and development. A variety of new methods and technologies have successively emerged, some of which are based on the improvement and optimization of the original sludge treatment technology. For example, inspired by the wet-air oxidation (WAO) process (Jing et al., 2012), Zhao et al. (2018b) proposed that a secondary WAO process could be used to treat

Table 1 Summary and comparison of crude oil recovery technologies.

Technology	Scale	Efficiency (%)	Cost (US$ /m^3)	Duration (day)	By-products	Advantages	Disadvantages	References
Solvent extraction	Field application	30–88	> 300	1–2	VOCs, Unrecoverable extraction slurry	Simple, Fast , Efficient	The cost of organic solvents is high, Secondary pollution	(Zhao et al., 2018a, 2019; Zubaidy and Abouelnasr, 2010; Hu et al., 2017a; Liang et al., 2014; Nezhdbahadori et al., 2018)
Mechanical centrifugation	Field application	40–70	100–300	1–2	Unrecoverable sewage and mud	Convenient, Fast, Efficient, High yield	Equipment maintenance cost is high, High energy consumption and noise, Pre-conditioning treatment is required	(Pinheiro and Holanda, 2013; Drelich et al., 2010; Huang et al., 2014b; Shao et al., 2014; Philemon and Benoit, 2013
Surfactant	Pilot application	70–90	> 300	2–10	Unrecoverable sewage and mud	Simple, Efficient, Large handling capacity	Cost is high, Chemical surfactants are toxic and cause secondary contamination, The treatment of heavy metals is limited	(Liang et al., 2017; Lima et al., 2011; Yan et al., 2012; Zhao et al., 2015; Seo et al., 2018; Azim et al., 2011; Ramirez and Collins, 2018; Liu et al., 2018; Sahebnazar et al., 2018; Chirwa et al., 2017)
Freezing/ thawing	Laboratory application	50–70	/	2–5	Unrecoverable sewage and mud	Convenient, Long lasting, Suitable for high-cold area	High energy consumption, High cost, Low efficiency	(Ghosh and Rousseau, 2009; Lin et al., 2008; Jean et al., 1999; He and Chen, 2002; Chen and He, 2003; Feng et al., 2017)
Pyrolysis	Field application	50–90	300–500	1–2	VOCs, Coke	Efficient, Quick, High quality oil recovery, Large handling capacity	Equipment maintenance cost is high, High energy consumption, Not suitable for high water sludge	(Lin et al., 2017a, 2017b, 2018a, 2018b, 2017c; Ma et al., 2014; Schmidt and Kaminsky, 2001; Liu et al., 2009; Shen et al., 2016; Cheng et al., 2017; Gao et al., 2018a; Hu et al., 2017b; Kim et al., 2019; Hou et al., 2018)
Electronal method	Laboratory application	50–70	/	2–3	Sewage, Mud	Low energy consumption, Efficient and fast	Small processing capacity, Equipment manufacturing complex	(Elektorowicz et al., 2006; Jahromi et al., 2018; Kariminezhad and Elektorowicz, 2018; Taleghani et al., 2019)
Ultrasonic	Pilot application	45–70	/	1–2	Sewage, Mud	Efficient, Fast, Environmentally friendly	Small handling capacity, High equipment manufacturing and maintenance costs	(Abramov et al., 2009; Wang et al., 2018b; Check et al., 2014; Gao et al., 2018b)

Table 1 (*continued*)

Technology	Scale	Efficiency (%)	Cost (US$ /m^3)	Duration (day)	By-products	Advantages	Disadvantages	References
Flotation	Laboratory application	55–70	/	1–2	Waste water	Convenient and low energy consumption	High water consumption, Low efficiency, Not suitable for high viscosity sludge	(Ramaswamy et al., 2007; Guo et al., 2011; Da Silva et al., 2019)
Supercritical fluid	Pilot application	70–90	/	1–2	Waste water	Efficient and quick	High energy consumption, Water consumption	(Cui et al., 2011; Radfarmia et al., 2015; He et al., 2015; Khan et al., 2019; Ávila-Chávez et al., 2007; Ma et al., 2019)
Combined processing	Laboratory application	/	/	/	/	Fast, Efficient and energy saving	The equipment is complicated and the operation process is long	(Zhang et al., 2012; Hu et al., 2015; Sivagami et al., 2019; Guo et al., 2014; Su et al., 2019)

oily sludge; they used 80 ml of hydrogen peroxide as an oxidant and 4 g of sodium carbonate as a demulsifier, and conducted two-stage WAO at a temperature of 240 °C for a reaction time of 90 min. The separation degree of the oily sludge and water was significantly improved, and a volume reduction of about 85.4% was achieved. Moreover, the removal rate of crude oil achieved 93.1%, and the separated waste oil could be recovered and reused. It was found that the effect of the demulsifier was the greatest, the effects of the reaction time and temperature were the second-greatest, and the effect of the oxidizer was the least. Similarly, Duan et al. (2019) developed a multistage chemical treatment method for the recovery of crude oil from oily sludge based on the surfactant method and the solid separation process. Three different surfactants, namely limonene, sodium sulfate solution, and octadecyl dimethylammonium bromide + citric acid, were used in different stages of sludge treatment. The separation difficulty of the traditional surfactant method lies in the residual solids at the interface, which have strong interaction with the resin and asphaltenes due to electrostatic attraction or chelation; thus, it is difficult to separate them via the demulsification process of the surfactant. The proposed technology innovatively introduces organic polyacids to break up the strong electrostatic interaction, thus facilitating the separation of these residual solids. At a temperature of 80 °C and a water/sludge ratio of 4, the solid particle content in the recovered crude oil was found to be less than 1% after 40 min of stirring and separation at a rate of 500 rpm. If the recovered oil is mixed with the new crude oil emulsion in proportion, it is more beneficial to the removal of the excess water in the emulsion. The advantage of this method lies in the direct solid-liquid separation of oily sludge into the oil-water phase and recovery of crude oil, and there is no need for subsequent mechanical centrifugation or other operations.

With the continuous innovation and progression of science and technology, some new recycling technologies are currently emerging. Recently, for example, Wu et al. (2019) reported a new technology for the water extraction of carbon dioxide, in which water and carbon dioxide are mixed with oily sludge at a certain proportion. The dissolution and expansion of carbon dioxide force the crude oil to separate from the solid particles of the sludge, and the presence of the water-phase interface prevents the separated crude oil from readhering to the surface of the solid particles. The results demonstrated that the recovery rate of crude oil reached up to 80% under the optimal conditions. Compared with the ultrasonic method, solvent extraction method, and supercritical fluid extraction method, this new method not only has the advantage of a high crude oil recovery rate, but also has low requirements for processing equipment due to its low reaction temperature and pressure and less environmental pollution. In addition, Xia et al. (2019) designed a new method for the recovery of oil from sludge via a steam jet; a clean vertical steam jet is used to inject layered oily sludge, and the light oily sludge in the upper layer can be quickly and efficiently separated from the heavy oily sludge in the bottom layer. With the continuous heating of the high-temperature steam, the viscosity of the oily sludge will gradually decrease, and the high pressure generated by the steam jet will promote the separation of oil and water. Moreover, the steam can disperse the metal impurities in the oil droplets into the water to achieve the recovery and purification of crude oil. The use of this method was found to recover nearly 90% of crude oil and remove about 80% of metal impurities under certain conditions.

To sum up, various emerging methods for the recovery and treatment of oily sludge crude oil have advantages of rapidity, high efficiency and energy saving. In particular, the efforts and results of the researchers have been fruitful in improving the efficiency and reducing the cost of recycling. However, we should also see that most of these emerging methods are still in the stage of laboratory research, which is far from the pilot test or even large-scale engineering application. Therefore, the amplified verification experiment of new methods will be the focus of researchers' next work.

3. Conclusion and outlook

In recent years, with the continuous development of the oil industry, the development of unconventional oil and gas fields has continued to increase, which has resulted in the increased production of highly viscous, toxic, and acidic oily sludge. Because oily sludge contains many heavy metal elements, heteroatomic compounds, polycyclic aromatic hydrocarbons, and other toxic and harmful pollutants, its harm to both human and animal health and the surrounding ecological environment should not be overlooked. Consequently, the development mode of waste recycling has also attracted increasingly more attention. Therefore, a variety of methods have been developed for resource recovery and the reduction and harmless treatment of oily sludge, among which technologies such as pyrolysis have been applied in large-scale engineering. Based on the comparison presented in Table 1, it is evident that many methods, including solvent extraction, ultrasonic and other physical methods, catalytic pyrolysis, supercritical oxidation and other chemical treatment methods, various combined processing methods, and some new technologies, have emerged in recent years. The most significant problem of the dehydration and recovery of crude oil is the determination of how to separate the emulsified mixture of oil and water in the oily sludge to achieve solid-liquid separation and oil-water separation. Therefore, demulsification technology is crucial for the improvement of the recovery efficiency of crude oil. In addition, the compositions of oily sludge produced by different oil fields or refineries are varied. Therefore, the concepts of environmental protection, energy conservation, and consumption reduction should be simultaneously considered, and full play should be given to the interdisciplinary advantages of physics, chemistry, and biology. Different crude oil recovery and treatment processes are optimized or combined for different types of oily sludge, and continuous improvement and innovation have been made in technical optimization, demulsification and quality improvement, economy and efficiency improvement, and equipment research and development.

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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