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*Review Article*

# Research Progress and Prospects on the Treating and Disposal for Waste Oil-Based Drilling Cuttings from Shale Gas Wells

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Oil-based drilling cuttings (OBDC) from shale gas wells is a kind of typical hazardous waste produced during the exploration and development of shale gas reservoirs, which will endanger the natural ecosystem and human health. This article summarized the OBDC treating and disposal methods commonly used all over the world. The treating methods were divided into restriction, separation, and degradation, while the disposal methods were divided into landfill, land farming, and resource recovery. The technical principles, technological process, advantages, and limitations of existing OBDC treating and disposal methods were overviewed in detail, and the development prospects of OBDC and resource recovery were illustrated as well. Finally, we proposed to maximize the resource recovery of OBDC by multitechniques processing, tracking the processed products to improve the reliability of product quality. In addition, the data system for original materials used for different methods should be established to service industrial development.

## 1. Introduction and Motivation

China has enormous shale gas resources, which has considerable commercial development prospects. In recent years, with the rapid development of shale gas exploration and production industry in China, a large amount of hazardous waste has been produced, known as oil-based drill cutting (OBDC) [1]. OBDC is a kind of organic mixtures, which is composed of crushed formation cuttings, drilling waste fluid, mud, and sand in drilling operation. Different from the exploitation of traditional natural gas reservoirs, shale gas exploitation requires more drilling ports and long horizontal wells to form large-scale production. According to previous works, the average production of waste OBDC in a shale gas well was about $200\,m^3$ [2, 3]. Evidently, OBDC belongs to hazardous waste that contains some pollutants, e.g., petroleum hydrocarbons and heavy metals, which has the dual attributes of waste and resource. In the view of waste, if the waste OBDC is piled nakedly, the volatile organic compounds will pollute the air in the vicinity of the well site and endanger the human health. Additionally, bare stacking will cause soil compaction around the stacking site. Then the toxic pollutants would enter the surface and ground water via the action of rainwater erosion, and finally enter the food chain, which seriously endanger ecological security and human health [4–6]. In the view of resource, OBDC is a kind of recyclable resources with considerable oil content. By scientific and efficient methods, the recovery and utilization of petroleum hydrocarbons and rational disposal of harmful substances would be realized.

Currently, the main treating methods for waste OBDC primarily are stabilization/solidification, incineration, thermal desorption, etc. Although these methods can meet the environmental requirements to a certain extent, they still cannot achieve high-efficient recovery of hydrocarbons and complete degradation of harmful substances. Due to the limitations of existing treating methods and the increasingly stringent environmental requirements, further upgrades, the key public relations are urgent to carry out in areas such as waste resource recycling and harmless disposal. In addition, with the reformation of China's energy structure and the implementation of carbon peak and carbon neutrality strategy, it is necessary to promote the exploration and development of deep shale gas resources in the next decade, which will produce much more waste OBDC. Therefore, in this work, the treating and disposal methods of waste OBDC are systematically overviewed through different classifications, including the technical principles and research progress. At last, the future research priorities and industrial development trends are also discussed.

## 2. Physicochemical Properties of OBDC

OBDC is a multiphase system containing oil (15%-20%), water (5%-10%), and solid impurities (-70%) [7–9]. The main organic matters in oil component are hydrocarbons, nonhydrocarbons, resins, and asphaltenes. Hydrocarbons include alkanes and aromatics, and alkanes account for a content of 73%, while the nonhydrocarbons mainly include fatty acid methyl ester (FAME), stearic acid salt, and so on. Asphaltene and colloid are mainly heterocyclic compounds containing heteroatoms such as S/N [10]. Due to the variety of pollutants, e.g., anthracene, pyrene, polychlorinated biphenyls (PCBs) in oil [11], it can be known the higher the oil content of OBDC, the greater the toxicity is. The particle surface OBDC is attached to the base oil as the main component of the oil-based mud, so its appearance is black, as shown in Figure 1 [12]. The leaching liquid produced by rain-washing of OBDC has the characteristics of high COD value, high suspended matter content, and deep color [13], which will contaminate the groundwater, surface water, and soil around the drilling site. The previous studies show that OBDC mainly contains eight kinds of heavy metals: Mn, Cd, Cu, Hg, Ni, Pb, Zn, and Cr [14], which is mainly derived from formation materials, drilling additives, and corrosion of drill bits under high temperature and pressure. All those heavy metals in OBDC present varying degrees of pollution except for Cr.

Above all, organic matters, heavy metals, and alkaline salts are the main pollutants in OBDC. If they are not effectively treated and disposed, the pollutants will gradually migrate to the environment, leading to secondary pollution. Therefore, how to achieve the harmless treatment and resource utilization of OBDC are the key and difficulty issues for environmental protection in the exploration and development of shale gas reservoirs in China.

## 3. Treating Methods for OBDC

According to the conversion mode of pollutants in the treating process, OBDC treating methods can be divided into restriction, separation, and degradation.

Figure 1: Appearance of OBDC from shale gas wells [12].

*3.1. Restriction Methods.* In the treatment of waste OBDC, restriction refers to encapsulating the pollutants in a confined space through a series of physical or chemical methods, to achieve the purpose of limiting contaminant migration and transformation [15]. Thereinto, stabilization/solidification is the most common used restriction methods. Stabilization refers to the conversion of contaminants into inert matters with low toxicity and solubility, and then reducing their environmental risks in the subsequent disposal [16]. Solidification refers to mixing the curing agent with OBDC, and the contaminants would be adhered, wrapped, and solidified. Finally, a kind of stable solid with certain structural strength is formed [17]. The solidified object can be the original OBDC or cuttings residue after treatment [15]. Common stabilization/solidification methods include fly ash-based solidification [18, 19], cement-based solidification [20], and plastic solidification. The flow chart is shown in Figure 2.

Cai et al. [18] took the OBDC from shale gas well as the solidification object, the inorganic gel materials (cement, fly ash, and active reinforcement materials) and small quantity of interfacial modifier was used. The comprehensive influence of various components' contents on the solidification efficiency and solidification mechanism of solidifying reagents on OBDC was investigated. Their results show that the unconfined compressive strength of the OBDC solidified body was significantly increased. The hydrated calcium silicate gel (C-S-H) and ettringite (AFt) generated in the solidification process filled the pores of the solidified body, as a result, the porosity and density of solidified body was decreased (as shown in Figure 3). In addition, Cai et al. [19] used the residual drilling cuttings produced in pyrolysis process as the solidification object, self-made solidification additives CS combined with fly ash and active reinforcing materials was used to solidify. In their work, the compressive strength, water stability, and frost resistance of the solidified body increased remarkably. And when the content of CS between 6% and 15%, the leaching toxicity of the solidified

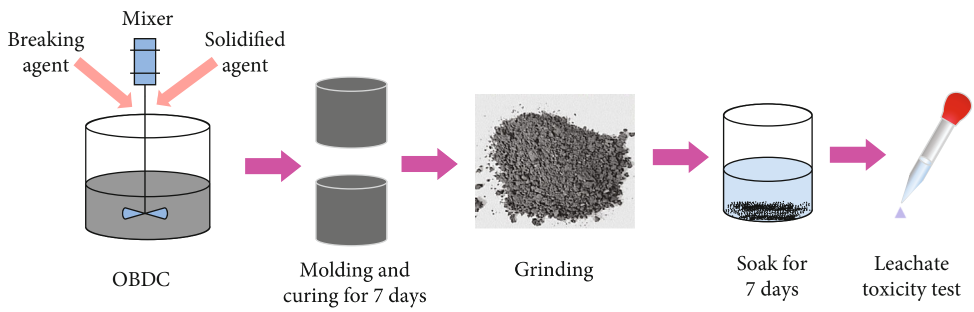
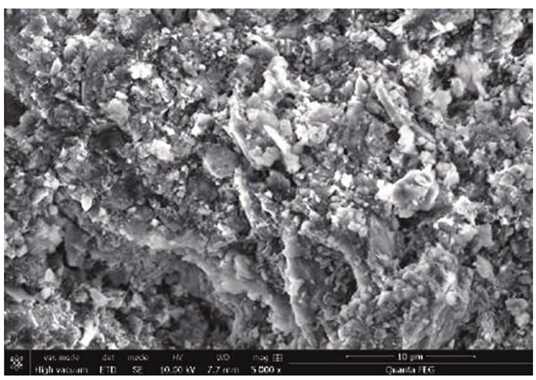
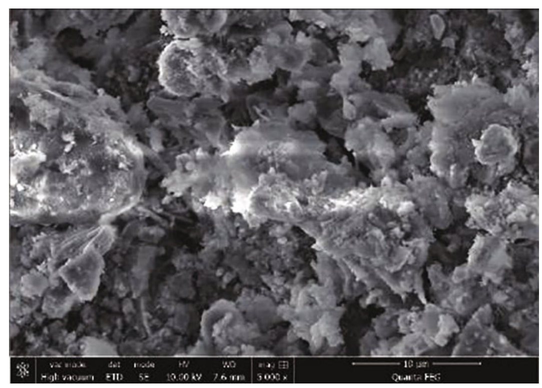



Figure 2: Flow chart of stabilization/solidification method.



(a) Original OBDC



(b) Solidified body with CS

Figure 3: Microstructure of original OBDC and OBDC solidified body (25°C and 28 d) [18].

Table 1: Analysis results of leaching liquid of OBDC solidified system with different curing agent contents (20 ∘ C, 7 d) [19].

| Curing agent | pH | COD/$(mg \cdot L^{-1})$ | Suspended mater/$(mg \cdot L^{-1})$ | Hydrocarbon/$(mg \cdot L^{-1})$ | Chrominance |
|---|---|---|---|---|---|
| 6%CS | 7.90 | 79.8 | 52 | 1.2 | 32 |
| 10%CS | 8.30 | 62.5 | 46 | 0.8 | 16 |
| 15%CS | 8.85 | 55.7 | 41 | 0.5 | 8 |
| NES | 6 ~ 9 | 100 | 70 | 5.0 | 50 |

body could meet the requirements of national environmental standards (NES), as shown in Table 1.

Meanwhile, Zhang et al. [20] used modified magnesium oxychloride cement to solidify drilling waste. The results show that the compressive strength of solidified body could reach 38 MPa, and the leaching toxicity could meet the requirements of the *Hazardous Waste Identification Standard (GB5085.3-2007)* and the *Integrated Wastewater Discharge Standard (GB8978-1996)*. In the work of Xie et al. [13], ferric trichloride ($FeCl_3$) was firstly used to break the gel of the waste drilling mud, and then the composite solidification agent was used to solidify it (fly ash 45 g, lime 6 g, loess 45 g, and cement 4 g per 100 mL drilling waste). The influencing factors of the curing effects were studied as well. The results show that the compressive strength of the solidified body was correlated with the slurry density, and the alkaline condition at 45°C in favor of forming more stable solidified body.

As we can see, stabilization/solidification is an economical and efficient treating method for organic waste. However, when the oil content of OBDC is high, it is necessary to conduct pretreating before solidification, e.g., pyrolysis and desorption, to reduce environmental risks [21, 22]. On the other hand, the solidification will increase the volume and weight of the waste, which will require a large area of land resources. As time goes by, the solidified body may produce weathering, which leads to leakage risks.

### 3.2. Separation Methods

*3.2.1. Supercritical Fluid Extraction.* Supercritical fluid extraction is a new physicochemical treating method, which mainly make use of the high solubility and mass-transfer diffusion capacity of supercritical fluid to extract the target object from the mixtures. When the temperature and pressure of the fluid exceed the critical values, it will reach the

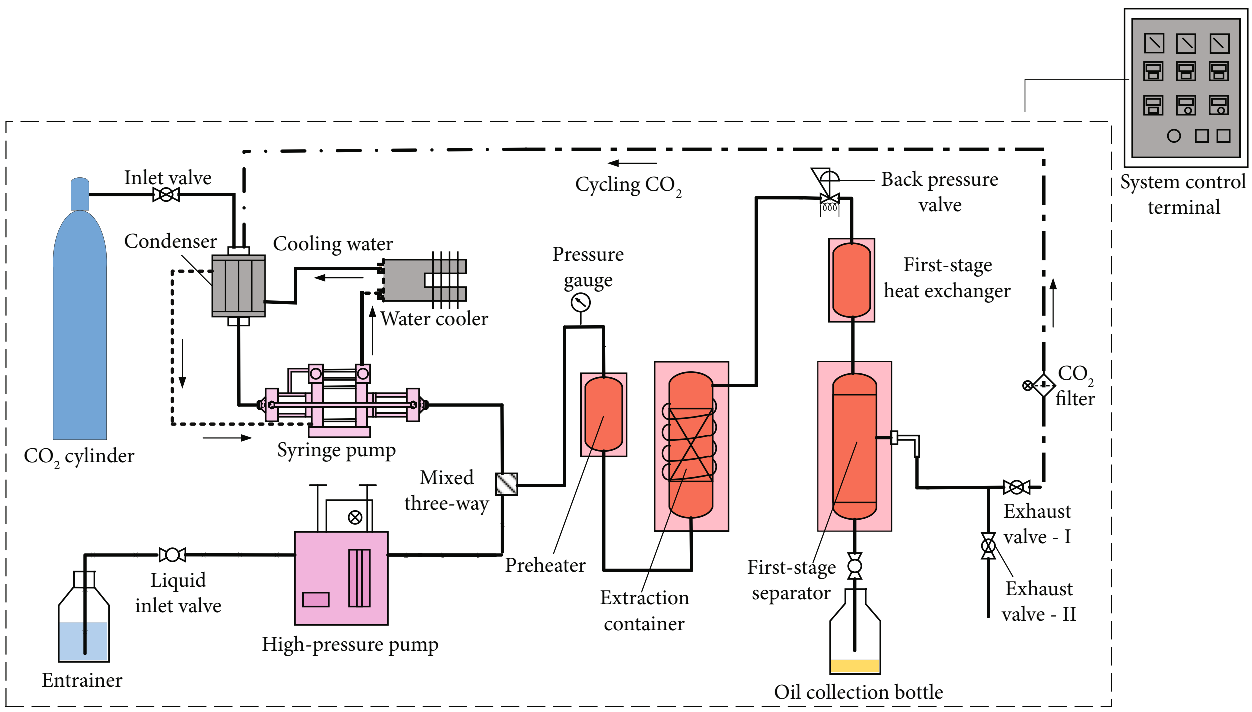
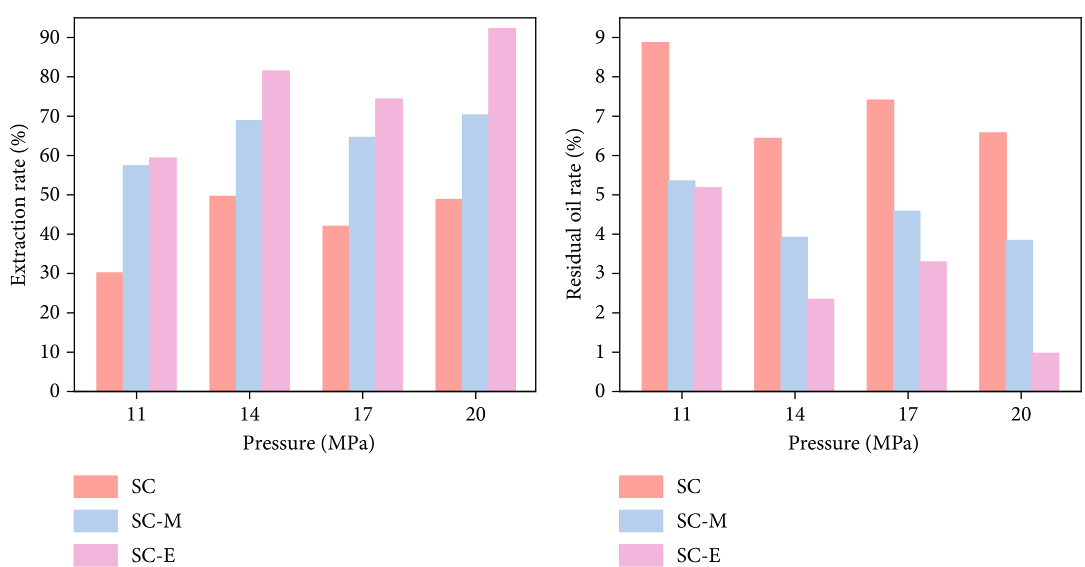



FIGURE 4: Supercritical $CO_2$ extraction technology device diagram.




(a) Extraction pressure vs. extraction rate

(b) Extraction pressure vs. residual oil rate

FIGURE 5: Influence of extraction pressure on supercritical $CO_2$ extraction efficiency [35].

supercritical state (a special state between gas and liquid) with liquid-like high density and solubility and gas-like high diffusion, which could quickly penetrate into matrix of OBDC to achieve high extraction efficiency in extraction process [23, 24]. In the extraction work of waste OBDC, $CO_2$ is usually considered as the ideal extraction solvent. The critical temperature and pressure of $CO_2$ is 31.1°C and 7.38 MPa, respectively [25–27]. Besides, $CO_2$ is a kind of nonpolar molecule with relatively stable chemical properties, so it can easily dissolve mineral oil, diesel, and other organic hydrocarbons according to the principle of *like dissolves like*. And, it hardly reacts with the target product, which shows high-efficiency extraction [28]. Compared with organic solvents, supercritical $CO_2$, as a reaction medium, can eliminate gas-liquid interface resistance and greatly improve the reaction efficiency. By adjusting the temperature and pressure, the physical properties of supercritical $CO_2$, e.g., viscosity, density, and polarity can be changed. It will increase the solubility of target product, and then improving the selectivity of supercritical $CO_2$ extraction.

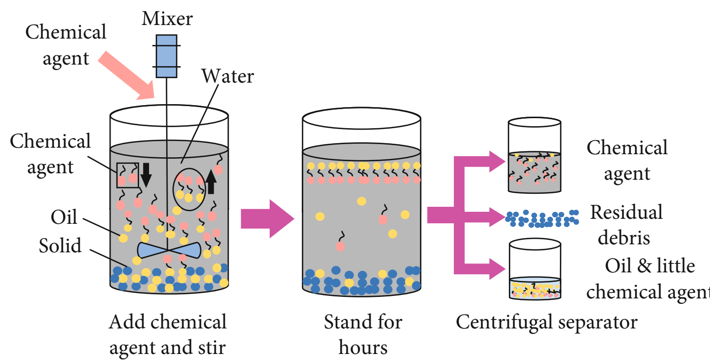



FIGURE 6: Flow chart of chemical cleaning.

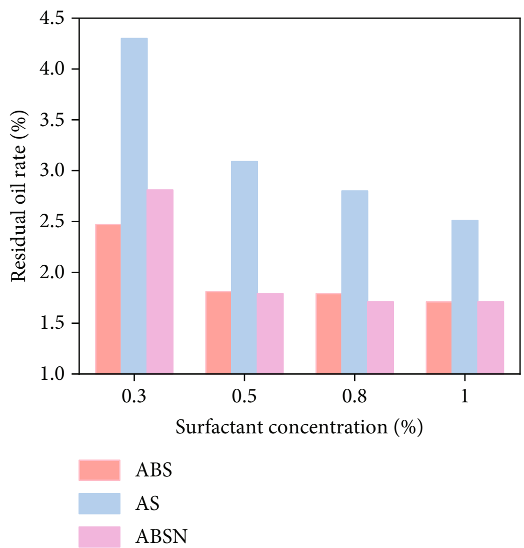



FIGURE 7: Oil content of drilling cuttings after cleaning with various surfactants solution [37].

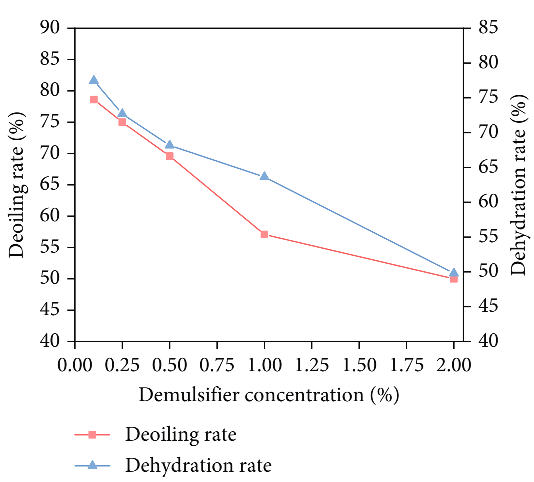



FIGURE 8: Influence of demulsifier concentration on dehydration and deoiling [38].

Figure 4 shows the technological flow for supercritical $CO_2$ extraction, and the process of supercritical $CO_2$ extraction including extraction and separation. (1) In the extraction container, organic hydrocarbons in OBDC are dissolved and carried by supercritical $CO_2$ above the critical pressure and temperature; (2) adjusting the temperature or pressure lower than its critical point, the supercritical $CO_2$ will change to a gas state. Its solubility decreased sharply, causing separation of the supercritical $CO_2$ and extracted objects in first stage separator [23, 29]. Due to the $CO_2$, properties remain unchanged, the $CO_2$ not only can be recycled but also can be easily removed without leaving a solvent residue.

Liang [29] carried out an experimental investigation on the extraction of oily sludge by supercritical $CO_2$. The results showed that the oil phase recovery rate of oily sludge was 33.15% after 40 h at the pressure of 20 MPa and the temperature of 55°C. The extracted components were saturated components and some aromatic components, but the heavier components such as gum and asphalt were not detected. Ma et al. [30] used supercritical $CO_2$ to extract hydrocarbons from oil-based mud and studied the extraction efficiency of different extraction parameters. The results demonstrated that the extraction efficiency of the target substance could reach 98% under the conditions of 60 min, 20 MPa, and 35°C. They show that the extraction process did not alter the hydrocarbon properties of the organic hydrocarbons in the waste oil-based mud, and the retrieved hydrocarbons could be recycled after processing, which can achieve the green recycling of petroleum resources.

Moreover, previous studies shown when using supercritical $CO_2$ extraction to treat OBDC, adding a specific amount of entrainer (coextraction agent) could significantly improve the extraction rate of target products [31–33]. Wei et al. [34] found that the coextraction agent greatly improve the extraction efficiency of supercritical $CO_2$. When 0.5% coextraction agent was added, the oil content in off waste residue was less than 0.3%, which meets the requirements by *China's National Standard of Sewage Sludge Applied to Agricultural Soils (GB 4284-1984)*. Peng et al. [35] used two kinds of entrainer (ethanol and methanol) to assist the extraction of OBDC, and compared the effects of the above two entrainers on the extraction rate of supercritical $CO_2$. It was shown that better extraction performance can be obtained by using

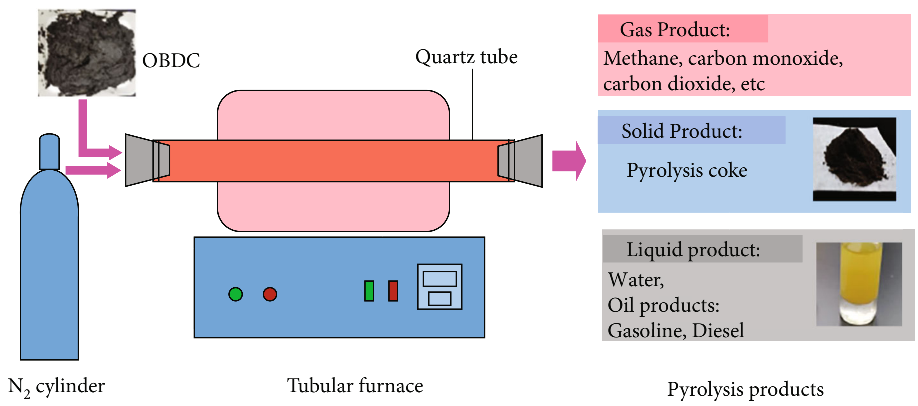



Figure 9: Flow chart of pyrolysis treating.

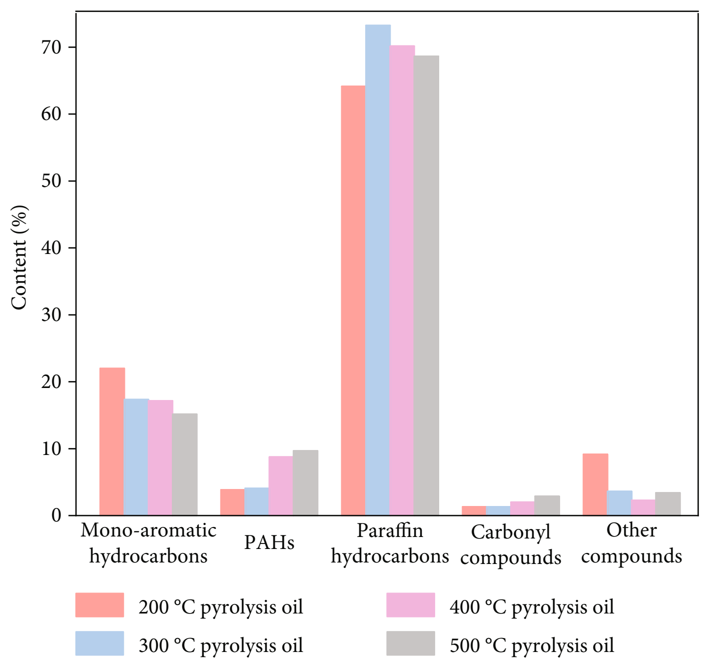



Figure 10: Compositions of pyrolytic oil at different temperatures [39].

ethanol as entrainer. Under the extraction pressure of 20 MPa, the highest extraction rate can reach 92.2% and the lowest residual oil ratio is 0.9% (as illustrated in Figure 5).

It can be seen that the key to this treatment is the tunability of supercritical $CO_2$. The solubility of a compound in supercritical $CO_2$ can be tuned precisely by changing the temperature and pressure, which allows the whole process to be simple. At low temperatures, the target oil phase is hardly thermally degraded and its chemical properties will not be altered during the extraction process. Moreover, the extraction and separation efficiency of this method is relatively high, and there is no solvent in the residues.

*3.2.2. Chemical Cleaning.* Chemical cleaning refers to the application of detergents, e.g., demulsifiers and surfactants, to reduce oil-water interfacial tension, change interfacial viscosity, and enhance oil emulsifying properties so that oil substances on the surface of OBDC are easy to be stripped or emulsified [8]. To achieve a better oil-water separation efficiency, this method is often combined with mechanical separation, such as centrifugal separation and pressure filtration separation (as shown in Figure 6).

Ma et al. [36] developed a demulsifier, and applied the demulsification-flocculation-separation process to treat waste OBDC; the oil recovery efficiency could reach 98.6%. Zhao et al. [37] chose surfactants to remove the oil on the surface of oily cuttings, which are produced during the deep water drilling in West Africa. Three surfactants, sodium alkyl sulfonate (AS), sodium dodecyl sulfonate triethanolamine (ABSN), and sodium alkyl benzene sulfonate (ABS) were chosen, and their oil removal effects were compared. The results revealed that the ABSN and ABS were suitable as the cleaning agents that could be seen in Figure 7. Via this cleaning method, the oil content of clean drilling cuttings was as low as 2%, which can meet the requirement of

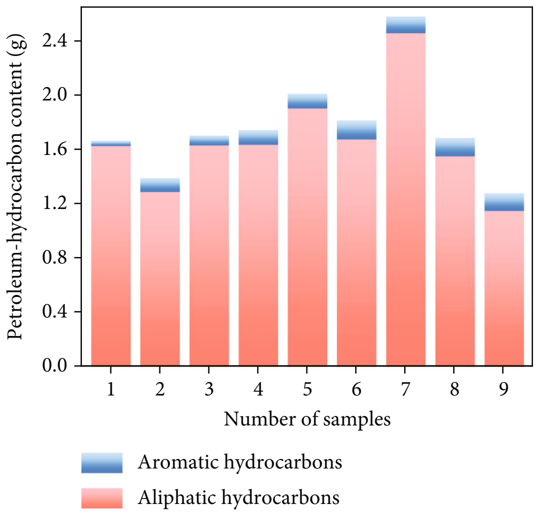



Figure 11: The yield and distribution of aliphatic and aromatic hydrocarbons [12].

oceanic disposal. Meanwhile, Yucheng et al. [38] used thermo-chemical demulsification and centrifugal separation method to treat waste oil-based drilling fluid and selected BASF L62 as a demulsifier. The results shown that, under the conditions of demulsifier dosage of 300 mg/L, rotate speed of 8000 r/min, demulsification temperature of 80°C, demulsification time of 3 h, centrifugal time of 25 min, and demulsifier concentration of 0.25%, the dehydration rate and oil removal rate are 75% and 72.73%, respectively, as shown in Figure 8.

This method has relatively mature treating system and stronger effectiveness without hi-tech equipment, so it can achieve better treating efficiency. However, the commonly used cleaning agents are toxic, and a large amount of wastewater will be generated in treating process, causing secondary contamination.

*3.2.3. Pyrolysis.* Pyrolysis is a physical separation process, which directly or indirectly heating the material to the temperature above the boiling point of oil and water under adiabatic conditions to separate the oil and water from the OBDC (as shown in Figure 9). This method is designed to just change the phase of organic oil in the OBDC but would not damage the structural properties of organics [39], which can be divided into three stages: water evaporation, light-hydrocarbons cracking, and heavy-hydrocarbons cracking [40].

Xia et al. [39] conducted the pyrolysis experiments of OBDC from shale gas wells and investigated the effects of final pyrolysis temperature and heating rate on the pyrolysis efficiency under nitrogen atmosphere. They found that the final pyrolysis temperature was a key factor for the pyrolysis efficiency of OBDC. Under their optimum pyrolysis conditions, the oil content of pyrolysis ash residue was only 0.039%, and the pyrolysis oil obtained at different temperatures show a relatively high content of alkanes (as shown in Figure 10). To further reduce the pyrolysis energy consumption and improve the yield of target substances, Xu [12] used fly ash as a catalyst to optimize the pyrolysis process of OBDC. They found that fly ash can increase the weight loss rate of OBDC by 14.5%, and decrease the oil content of pyrolytic slag to below 0.1%, which meets the *Agricultural Sludge Pollutant Control Standard (GB 4284-2018)*. The liquid phase products of pyrolysis contained a large number of aliphatic hydrocarbons and a small number of aromatic hydrocarbons, as displayed in Figure 11.

In the process of pyrolysis, OBDC is in an inert and medium-low temperature environment, which can effectively prevent the generation of secondary pollutants, stabilize heavy metals, and improve the quality of pyrolytic oil [40]. And, the application scope of solid residues and oil products generated in pyrolysis is wide [41]. However, the particles and gaseous pollutants produced in pyrolysis process will cause air pollution, and the exhaust gas needs to be treated again, which increases the processing procedures and costs. In other words, the problems of high-energy consumption and initial investment in pyrolysis treating have not been solved [42].

*3.2.4. Microwave Heating.* Microwave is a kind of electromagnetic waves with a wavelength between 0.1 mm and 1 m [43]. The principle of microwave heating is similar to the traditional heating treatment, where the oil and water are evaporated by heating, and then condensed and separated to achieve the purpose of harmless treatment. However, compared with traditional heating method, microwave heating is a heating method at the molecular level, which each atom can be equably heated with the faster speed and high efficiency. Microwave energy is transferred to the material through the interaction of electromagnetic fields and molecules, while traditional heating transfers heat energy to the surface of the material through conduction, convection, and radiation [44, 45]. Different ways of conventional heating and microwave heating are shown in Figure 12.

Hou et al. [46] found that in the microwave heating treatment of OBDC, a certain amount of graphite can increase the heating rate of OBDC. As we can see from Figure 13, when the mass fraction of graphite was 6%, the highest temperature of OBDC could reach 565°C, which was about 40°C higher than that without graphite. After microwave heating, the petroleum hydrocarbon content in the drill cuttings residue was decreased to 0.23%, where the microwave processing power was 1200 W, the temperature was 250°C, and the time was 30 min. Chen [47] found that without adding other substances, the highest temperature of OBDC heated by microwave only reached 300-400°C, and the oil content of the treated residue was 2.53%. While adding certain absorbing materials (such as activated carbon, graphite worm, and SiC powder) in the microwave heating process can significantly improve the microwave treating efficiency. This is because the absorbing materials cannot only convert microwave energy into heat energy but also increase the porosity of OBDC. The results also show that the addition of absorbing materials can shorten the processing time, and the oil content of the final treated cuttings is only 0.82%, which can be discharged with drilling.

Microwave heating can heat the inner and outer parts of the material synchronously with its strong penetrability,

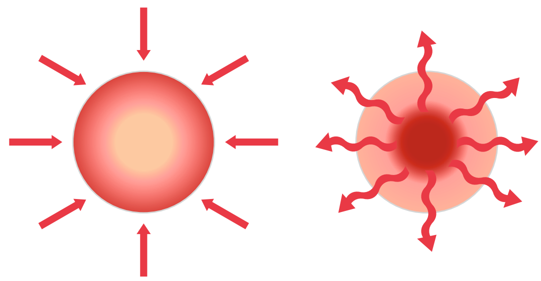



Figure 12: Conventional heating versus microwave heating [43].

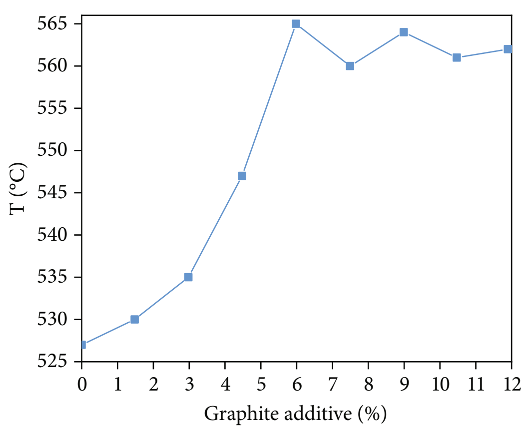



Figure 13: Influence of graphite content on heating characteristics of cuttings [46].

without the influences of its thermal conductivity. In addition, the power consumption of the microwave heating method is 55-80% of the traditional heating [48]. From the economic and environmental point of view, the microwave heating technology has certain application prospects in the treatment of waste OBDC.

### 3.3. Degradation Methods

*3.3.1. Microbiological Degradation.* Biodegradation is defined as a process that using selected microorganisms to biologically degrade petroleum hydrocarbons in OBDC into nontoxic residues. The commonly used microorganisms for degrading organic hydrocarbons are bacteria, fungi, and algae [49]. The biodegradation method can eliminate oil pollutants without secondary pollution, and composting, land cultivation, and bioenhancement are the more commonly used biodegradation methods.

Babaei et al. [50] studied the degradation extent of total petroleum hydrocarbons (TPHs) and polyaromatic hydrocarbons (PAHs) during composting in the dark and at room temperature. The results indicated that the highest removal rate of TPHs and PAHs was obtained when OBDCs were composted with 15% bagasse, and the PAHs and TPH were removed up to 24.8% and 67.5%, respectively, after 70 days of incubation. Alavi et al. [51] investigated the degradation effects of oil-based mud by indigenous microbial consortium. They found that pretreatment could significantly improve the efficiency by using microbiological to degrade oil-based mud directly. The oil content of the residue decreased from 13.54% to 1.14% after degradation. Haixia et al. and He et al. [52, 53] studied the ability of OBT, ZL, and PKUA/B strains for treating OBDC. It can be seen from Figure 14 that the treating efficiency of the oil removal bacteria OBT was the best, the oil content in the drill cuttings was decreased to 3.5%, and the oil removal rate was more than 74%. However, the effects of microbiological treating for degrading OBDC solely were weak, so a combined method with normal temperature cleaning and microbial degradation was used widely. The experimental results indicated that the oil phase recovery rate was more than 85%, and the total petroleum hydrocarbon content (TPH) of waste residues was less than 2% after the cleaning. In addition, some scholars used biobased surface cleaning agents instead of chemical cleaning agents to remove hydrocarbons in drilling cuttings before microbiological treatment, and the content of hydrocarbons in biologically treated drilling cuttings decreased to less than 1% [54, 55]. Compared with single biodegradation or chemical cleaning, the combination of chemical cleaning and biodegradation is more efficient for treating OBDC.

*3.3.2. Supercritical Water Oxidation.* When the ambient temperature and pressure of water exceed 37°C and 22.1 MPa, respectively, it will enter the supercritical state. And the changes in properties of supercritical water are as follows: the number of hydrogen bonds decreases, the density and viscosity decrease, the surface tension gradually disappears, and the polarity becomes weaker. Supercritical water is an ideal reaction medium, and its physical properties mainly include low viscosity, high solubility, and high permeability [8, 56]. Supercritical water oxidation (SCWO) method is considered as an advanced oxidation technique, which can effectively degrade refractory organic compounds, stabilize heavy metals, and neutralize alkaline salts in OBDC [57]. Supercritical water is used as the reaction medium and excess oxygen or air as the oxidant to oxidize organic matter into carbon dioxide and water (as shown in Figure 15). Moreover, the SCWO reaction is carried out in a closed reactor, where the oxidation reaction is thorough and rapid without secondary pollution [58, 59].

Yao et al. [59] used a batch-type SCWO reactor to treat OBDC and investigated the influence mechanism of oxygen coefficient, temperature, and reaction time on the degradation efficiency of petroleum hydrocarbons and the stability of heavy metals. The results illustrated that nearly 100% of naphthalenes and more than 97% of alkanes could be removed (as shown in Figure 16). The heavy metals, such as Ba, Cr, and Pb, in solid-phase products were almost completely stabilized, and only a small amount of Zn and Cu were detected in liquid products. Chen et al. [60] investigated the SCWO for OBDC in a batch reactor under the conditions of various oxygen coefficients, temperatures, and reaction times. The results indicated that the organic matter in the OBDC was distributed in the gas phase, liquid phase, and solid phase in preheating experiments. The proportion of organic matter oxidized was less than 9.8%, but

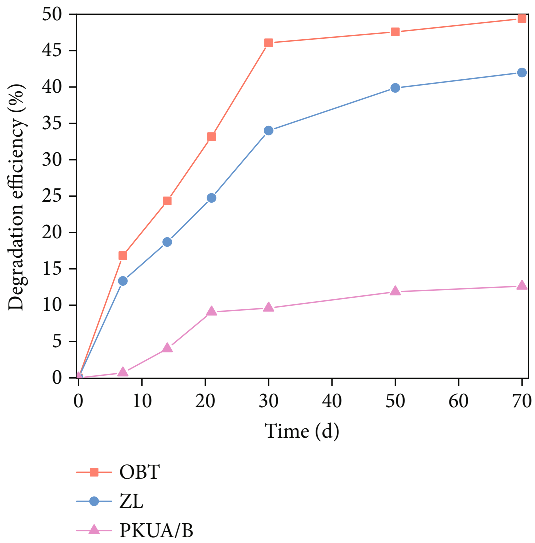



FIGURE 14: Effects of different bacteria strains on oil degradation [52].

the carbon compounds will eventually transform into $CO_2$. According to the Figure 17, The SCWO reaction had higher efficiency when the temperature was higher than 475°C, and the removal rate of total organic carbon (TOC) reached 89.2% at 500°C. The analysis of the reaction pathways suggested that the SCWO reaction was dominated by homogeneous reactions and supplemented by heterogeneous reactions that exist in the reactor. The homogeneous reaction was a supercritical oxidation reaction governed by a free radical mechanism, while the heterogeneous reaction was mainly a mass transfer process.

SCWO method has fast processing speed and high efficiency. However, the critical pressure and temperature of water are more higher than that of $CO_2$, which needs more energy and has some hidden dangers. In addition, it needs more oxygen, high equipment operation cost, and it cannot eliminate the danger of heavy metals are still not be improved [61].

## 4. Disposal Methods for OBDC

Treatment refers to the weakening or removal of contaminants in OBDC, while the disposal is to find the ultimate destination for OBDC. OBDC is solid organic waste with more than 70% inorganic ash. The treating methods can only reduce the organic matter and moisture in OBDC, but cannot remove the ash, so final disposal is required. The original OBDC is often reinjected or buried locally, which exists great environmental risks [62]. China's environmental regulations state that the untreated OBDC is not allowed to be discarded or directly landfilled, and can generally be used as the final disposal methods of residue after other treating techniques.

The disposal methods of OBDC produced by offshore drilling platforms and land drilling platforms are different. Taking land-drilling platforms as an example, the commonly used OBDC disposal can be divided into land farming, landfill, and resource utilization.

*4.1. Land Farming.* The land farming mainly used the microbial flora in the soil to degrade the hydrocarbon organic matter in the OBDC [63]. Compared to other disposal methods, land farming holds many merits such as simple operation, low cost, low energy consumption, and no need to deliberately cultivate the microbial flora. However, the original chemical additives in OBDC are difficult to degrade by microorganisms, and will migrate to soil and groundwater, resulting in secondary pollution. In addition, macromolecular substances (such as asphalt and wax) in OBDC need a long time to degrade [64]. This method requires strict site selection and control of leachate during degradation to reduce the risk of secondary pollution. Given the shortage of land resources in China, it is currently not a recommended disposal method [65].

*4.2. Landfill.* The landfill method refers to burying the treated OBDC into underground; it is the most commonly used onshore method for land drilling platforms. In the past decade, the platform OBDC was landfilled without treating, but in recent years, it has gradually transformed into the final disposal method of residue after pretreatment of OBDC. The landfill requires oil content below 3%, and the burial depth should be below 1.5 m of the surface and above 1.5 m of groundwater [66]. Although landfill disposal is a simple and cheap method, with the increasingly stringent environmental protection in China, it has been explicitly prohibited.

*4.3. Resources Utilization.* The resource utilization of OBDC refers to using treated OBDC to prepare cement, concrete, and bricks, which can recover petroleum resources and reduce the volume and toxicity of OBDC to achieve safe disposal. It is the most promising disposal method at present. IOGP (International Association of Oil and Gas Producers) pointed out that one of the principles for the sustainable reuse of hazardous waste is to ensure it meets the prescribed environmental guideline before expected using, aiming at protecting the current environment and avoiding future risks. This requires a series of performance tests on drilling cuttings after dehydration and deoiling to ensure the resource utilization of OBDC while meeting environmental protection requirements [67].

As an efficient method to realize the resource utilization of solid waste, cement kiln collaborative disposal has become increasingly widespread abroad in recent years. Solid hazardous waste was been put into the high-temperature cement kiln together with cement raw material to achieve the harmless disposal of hazardous waste while producing cement clinker [68]. It is a special incineration technique whose basic principle is to decompose all organic matter into small molecules at high temperature (1350-1650°C), and finally into $CO_2$ and $H_2O$ [69], while heavy metals are fixed in the clinker matrix, making it difficult to leach.

Cement kiln co-processing disposal is a zero-emission technique, which uses inorganic minerals and organic

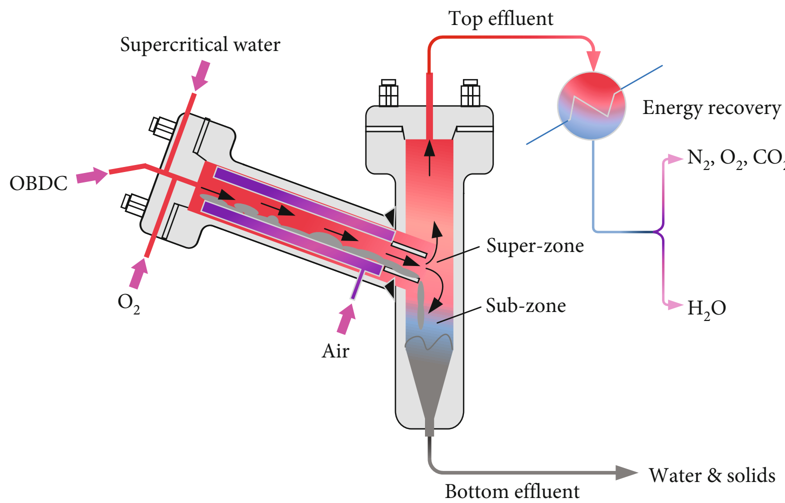



Figure 15: Flow chart of supercritical water oxidation [57].

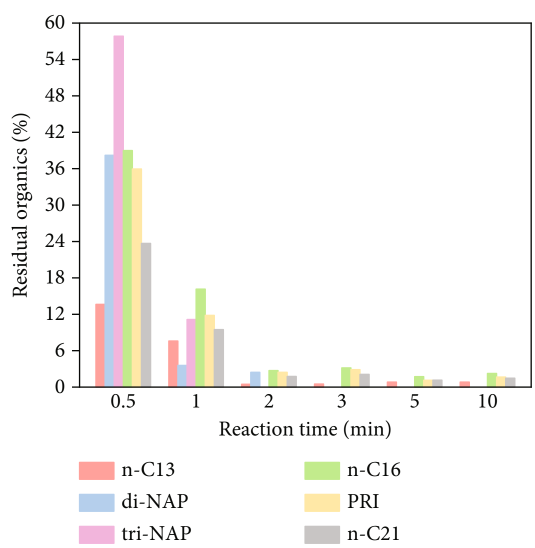



Figure 16: Effects of reaction time on residual rate of organic matter ($T = 475 \circ$ C and OC = 2.5) [59].

substances in hazardous waste as raw materials and fuels for cement production, which shows good ecological and economic benefits [70]. Lawrence Cement Plant of Canada used combustible hazardous waste as an alternative fuel in cement production in the 1970s [71]. A large number of tests had proven that it is feasible to produce cement by using combustible hazardous waste instead of traditional fuel in a rotary kiln, and it has not negative effects on cement quality and environment [72]. Wen et al. [73] used cement kiln to dispose of OBDC, the main inorganic units of the samples are $SiO_2$, $Al_2O_3$, BaO, $Fe_2O_3$, etc. Its mineral compositions are similar to cement raw materials, which are suitable for Cement kiln co-processing disposal. The combustion calorific value of OBDC is high, which can be used as a substitute for fuel in the cement production process. The synergistic treatment could reduce the consumption of coal/natural gas in the cement industry, which achieves the resource utilization of waste. However, the content of $SO_3$ and $Cl^-$ in OBDC is relatively high. When using cement kilns to co-process OBDC, after a reasonable determination of the mixing ratio of OBDC and raw materials, the effects of this factor on cement clinker can be avoided.

In the cement kiln co-processing disposal, the gas temperature in the cement rotary kiln is as high as 1650∘C and the residence time is long, so the organic matter in the hazardous waste is completely decomposed [74, 75]. And then, cement rotary kiln has large space and strong treating capacity, which can maintain a stable and continuous burning atmosphere and ensure the efficiency of hazardous waste disposal [76, 77]. Under the negative pressure condition, smoke and dust will not overflow, which fundamentally prevents recontamination in the

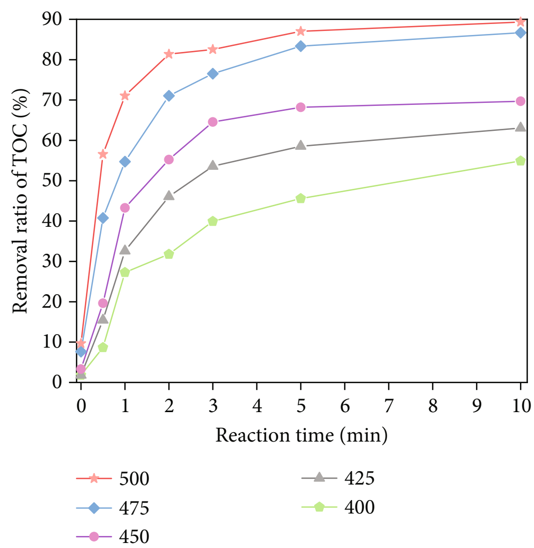



Figure 17: The total organic carbon removal efficiency under various times and temperatures at OC = 2.5 [60].

Table 2: Overview of the current treating and disposal methods commonly used.

| No. | Category | Name | Advantages | Limitations |
|---|---|---|---|---|
| 1 | Treating methods | Stabilization/ solidification | Low cost and simple process flow | Waste of oil and land resources, high environmental risk, unsuitable for treating OBDC with high oil content |
| 2 | | Supercritical $CO_2$ extraction | $CO_2$ is cheap and recyclable, the process is safe and reliable, the treatment is fast and efficient,weak damage to the useful components | High requirements for equipment, hard to apply on-site |
| 3 | | Chemical cleaning | Simple process equipment and low energy consumption | Chemical cleaning agents are expensive and specific, secondary contamination |
| 4 | | Microwave heating | High efficiency and speed, waste minimization | High complexity of microwave heating equipment, hard to achieve industrial application |
| 5 | | Pyrolysis | High oil recovery and waste minimization without treating agents | High energy consumption, extra treatment for waste gas and water |
| 6 | | Microbiological degradation | Low investment, no secondary contamination, complete degradation of organic pollutants | Long treating cycle, low efficiency, hard to cultivate microorganisms suitably for degrading petroleum hydrocarbons |
| 7 | | Supercritical water oxidation | High efficiency, no secondary contamination | High energy consumption and treating costs, more restrictive reaction conditions |
| 8 | Disposal methods | Land farming | Simple and low cost | Waste of oil and land resources, high environmental risk |
| 9 | | Landfill | Simple process, easy to construct, large quantity, low cost | Waste of oil resources, risk of soil and underground water contamination |
| 10 | | Resources utilization | High-value utilization of resources | Not mature, hard to popularize widely |

process. Moreover, cement kiln co-processing disposal can use hazardous waste with high ash and organic components as raw materials and part of heat sources for cement production to achieve resource utilization of effective components in hazardous waste [78].

## 5. Characteristics of Treating and Disposal Methods for OBDC

As mentioned, based on the conversion mode of pollutants, the OBDC treating methods are divided into restriction, separation, and degradation, and the disposal methods include land farming, landfill, and resources utilization. The advantages and limitations of treating and disposal methods for OBDC are displayed in Table 2, comparatively.

## 6. Remarks and Perspectives

Due to the complex components and increasingly stringent environmental requirements, the efficient treating and harmless disposal of OBDC have become a key issue in industrial development. The maximized resource utilization is the ultimate goal of OBDC treating and disposal. The treating objective is the original OBDC, and the disposal is the product after treatment. The disposal determines the treatment, and the treated products must meet the local environmental requirements. Resource utilization of the treated products is the ultimate goal of OBDC disposal. The significance of the selection of treating and disposal technical route for OBDC is to choose the disposal methods suitable for local conditions and the processing techniques that can meet the disposal requirements of its products. Actually, those works are carried out around the contradiction between the utilization of beneficial substances and the reduction of harmful substances. The development and progress of relevant techniques all over the world are also aimed at weakening this contradiction.

The harmless treatment, high-efficiency oil recovery technique, and resource utilization of drilling cuttings can achieve ideal results under certain conditions, but the various treating and disposal methods commonly used around the world also have specific limitations. Based on the principles of harmless treatment and high-value utilization, the following suggestions are put forward for reference:

(1) The raw materials database should be established to collect basic information of OBDC in different regions (composition, source, output, and processing technology used). State and local supervision departments should implement real-time tracking, while the disposal enterprises need to provide real-time feedback. To ensure the treated drilling cuttings residues meet the local environmental requirements, attention should be paid to oil content and heavy metals

(2) The existing OBDC processing technical parameters should be continuously optimized, and new techniques need to be developed. At present, more studies could focus on supercritical fluid extraction and pyrolysis. Pyrolysis is the most efficient method for treating OBDC; some new catalysts can be developed to further reduce energy consumption. Meanwhile, supercritical $CO_2$ extraction has considerable application prospects, and its technical parameters could be further improved through pilot plant tests

(3) For different types of raw materials, if a single process is not ideal, on the premise of minimizing potential environmental hazards and achieving the resource utilization of oil, a combination of multiple processes could be considered

## Conflicts of Interest

The authors declare that they have no conflicts of interest.

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