

Investigation on the Composition and Extraction Mechanism of the Soluble Species from Oily Sludge by Solvent Extraction

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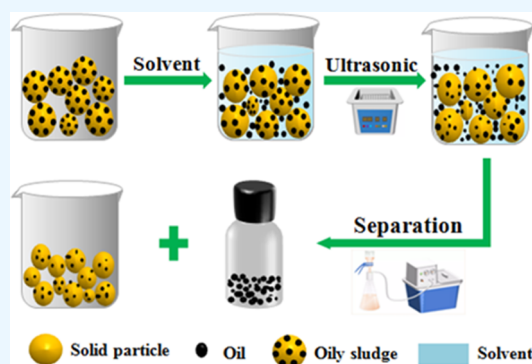


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Supporting Information

ABSTRACT: Oily sludge (OS) was extracted with petroleum ether (PE), methanol, carbon disulfide (CDS), acetone, and isometric CDS/acetone mixture (IMCDSAM), respectively, to obtain soluble species (E_1 – E_5) and extraction residues (R_1 – R_5). The soluble species were analyzed by gas chromatography/mass spectrometry (GC/MS), and the extraction residues were characterized by Fourier transform infrared spectrometry (FTIR) and thermogravimetric analysis (TGA). Results showed that the extract yield of the soluble species from OS using CDS and IMCDSAM as the solvent was 61.0 and 67.3%, respectively. GC/MS results exhibited that the compounds detected in E_1 – E_5 are mainly hydrocarbons and oxygen-containing compounds. E_1 – E_5 are rich in alkanes, alkenes, ketones, alcohols, and other oxygen-containing compounds. Double-bond equivalence (DBE) and carbon numbers (CNs) of the compounds detected in E_1 , E_2 , and E_4 are distributed in 0–4 (DBE) and 10–20 (CNs), respectively, while the DBE and CNs of the detected compounds in E_3 and E_5 are concentrated in 0–6 and 15–35, respectively. Thermogravimetry-differential thermogravimetry (TG-DTG) profiles presented that pyrolysis of OS occurred mainly in the temperature range of 150–750 °C, while pyrolysis of R_1 – R_5 took place in the range of 350–750 °C. In the temperature range of 150–550 °C, the weight losses of OS and each extraction residue differ significantly, with OS having a much higher weight loss than the extraction residues. Meanwhile, the possible mechanism of oily sludge extraction was considered. Results revealed that selecting a low-polar or nonpolar solvent capable of selectively destroying hydrogen bonds and/or aromatic interactions is critical for improving the extract yield of OS.



1. INTRODUCTION

OS is a complex mixture produced during the exploitation, transportation, storage, and refining of petroleum, generally composed of water, petroleum hydrocarbons, and solid particles. OS contains a large amount of toxic and harmful substances, such as benzenes, phenols, anthracenes, and pyrenes; it has been listed as a hazardous waste in various countries.^{1,2} Massive accumulations of OS will be extremely hazardous to the environment and human health.^{3,4} However, there are plenty of aliphatic, aromatic, and asphaltene organic components in OS, of which aliphatic and aromatic components account for 75–85%.⁵ And these organic components are essential raw materials for fuel and fine chemical production. Therefore, using appropriate technology to recover organic components from OS is of great significance for sludge reduction, harmless treatment, and resource utilization.

As a hazardous waste in the production process of the oil industry, OS has the characteristics of a large output and high oil content (especially heavy oil), which is a major problem that disturbs the treatment of solid waste sludge. At present, the treatment methods of OS mainly include biodegradation,^{5–7} mechanical separation,^{8,9} thermal decomposition,^{10,11} and solvent extraction.^{12,13} Among them, the method of

solvent extraction is faster, efficient, and applicable, which is widely considered to be the most promising technology for achieving resource treatment of oily sludge.¹⁴

Solvent extraction of oily sludge is mainly focused on extraction conditions, including solvent type, extraction temperature and time, solid/liquid ratio, extraction times, and auxiliary methods. Ávila-Chávez et al. used supercritical ethane to extract hydrocarbons from petroleum sludge. It was found that the extract yield was higher, the quality of the recovered oil was good.¹⁵ Naggar et al. recovered oil from oily sludge using different solvents and investigated the effect of extraction time, temperature, and solvent volume on the extract yield.¹⁶ Results showed that the oil yield with naphtha and kerosene as the solvent reached 83.99%, and the solvents used could be easily separated from the recovered oil. Nezhdbahadori et al. used the polar solvent methyl ethyl

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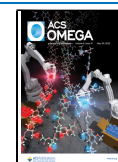


Table 1. Proximate and Ultimate Analyses of OS (wt %)

proximate analysis				ultimate analysis				
WC	SC	VHC	C	H	O ^a	N	S	H/C
3.50	74.36	25.64	76.74	11.33	8.63	1.03	2.27	1.77

^aBy difference.

ketone (MEK) and the nonpolar solvent toluene to recover hydrocarbons from oily sludge.¹⁷ Results showed that the hydrocarbon yields with MEK and toluene were 30.41 and 37.24%, respectively. It was found that the major components of OS were nonpolar, and the oil yield with the nonpolar solvent MEK was higher than that with toluene. Al-Doury compared the extraction performance of OS with five solvents, light naphtha, heavy naphtha, kerosene, gasoline, and MEK, to extract and recover oil.¹⁸ It was found that MEK presented better extraction performance with crude oil yield of 95%. Simultaneously, it was discovered that the extract yield was increased with increasing solvent volume and temperature, and the effect of solvent volume was more significant than that of temperature. Meng et al. used petroleum ether (60–90 °C) to recover oil from oily sludge by ultrasonic-assisted extraction.¹⁹ By a single-factor experiment, the extraction conditions were determined: an ultrasonic temperature of 55 °C, an ultrasonic intensity of 320 W, and a liquid–solid mass ratio of 7.5. Under the above conditions, the extract yield of oil was 94%. Han et al. recovered oil from the mixture of oily sludge and oil sand using toluene and ethanol.²⁰ Results showed that the best extraction conditions were as follows: the oil sand to oily sludge ratio of 0.5, the solvent to mixture ratio of 2, the extraction temperature of 60 °C, and the extraction time of 20 min. And the total recovery yield of oil was 81.8%.

These studies indicate that solvent extraction has high potential for separating valuable hydrocarbons from waste oily sludge. However, most of these studies reported the effect of various solvents on oil recovery, while the relationship between the solvent used and the components of the recovered oil was rarely discussed. Studies have shown that the type of solvent has a significant effect on the extraction efficiency of OS, and the composition of hydrocarbons recovered by different solvents is remarkable difference.^{4,21} The selection of extraction solvent is closely related to the composition of sludge and its physical and chemical properties. Therefore, it is necessary to reveal the relationship between the solvent and the composition of the recycled oil.

In this study, OS was subjected to extract with PE, CDS, methanol, acetone, and IMCDSAM. Molecular composition of the soluble species extracted from OS was detected by GC/MS, while the extraction residue was characterized by FTIR and TGA to understand the change of physicochemical properties of oil sludge.

2. EXPERIMENTAL SECTION

2.1. Materials. OS was collected from the Karamay Oilfield, Xinjiang Uyghur Autonomous Region, China. Before the extraction experiment, OS was pulverized and passed through a 70-mesh sieve and dried in a vacuum oven at 80 °C for 12 h. PE, methanol, CDS, and acetone were purchased commercially and purified by rotary evaporator distillation prior to use. The proximate and ultimate analyses of OS are listed in Table 1.

2.2. Solvent Extraction. 30 g of OS and 300 mL of PE were added to a 1000 mL beaker, and the extraction process was carried out at the ultrasonic condition (45 kHz) for 2 h at room temperature. Then, the mixture was filtered into a filtrate and a filter cake, and the filter cake was repeatedly extracted with 300 mL of PE every time until the filtrate was almost colorless. All of the filtrates were collected and concentrated using a rotary evaporator to obtain extract E₁, and the final residual filter cake was dried in a vacuum oven at 80 °C for 8 h and labeled as residue R₁. Using the same procedure, oily sludge was extracted with methanol, CDS, acetone, and isometric CDS/acetone mixed solvent to obtain extract E₂ and residue R₂, extract E₃ and residue R₃, extract E₄ and residue R₄, extract E₅ and residue R₅, respectively. The extract yield of OS was calculated according to the formula: $m_E/m_{OS,daf}$ where m_E and $m_{OS,daf}$ denote the mass of E (E₁–E₅) and OS (dry and ash-free basis), respectively.

2.3. Characterization. **2.3.1. Proximate Analysis.** Proximate analysis was carried out according to refs 4, 22, 23.

2.3.1.1. Water Content (WC). 1.00 g of oily sludge was put in an oven (with ventilation) at 105 °C for 2 h, and the weight loss of the sample was attributed to water content. WC was calculated as follows

$$WC = \frac{\text{reduced mass, g}}{\text{mass of tested sample, g}} \times 100\% \quad (1)$$

2.3.1.2. Volatile Hydrocarbon Content (VHC). Dried oily sludge (105 °C) with known mass was put into an electrical muffle furnace at 800 °C for 2 h, and the residue was weighed

$$VHC = \frac{\text{mass of tested sample, g} - \text{mass of residue, g}}{\text{mass of tested sample, g}} \times 100\% \quad (2)$$

2.3.1.3. Solid Content (SC). After measuring volatile hydrocarbon content, the solid content can be calculated by difference as follows

$$SC = 100\% - VHC \quad (3)$$

2.3.2. Ultimate Analysis. Ultimate analysis was performed on an Eurovector EA3000 Elementar (Italy).

2.3.3. Analysis of E₁–E₅ with GC/MS. E₁–E₅ were analyzed by GCMS-6890B/5977A equipped with an HP-5MS capillary column (30 m length, 0.25 mm inner diameter, 0.25 μm film thickness) at a constant helium flow of 1.0 mL/min and a quadrupole analyzer with the *m/z* range from 30 to 550 amu. The oven temperature was held at 40 °C for 3 min and increased from 40 to 150 °C with a rate of 5 °C/min and then from 150 to 280 °C with 10 °C/min, followed by holding at 280 °C for 5 min. The compounds detected were identified by comparing mass spectra with NIST11 library data.

2.3.4. FTIR Analysis. FTIR was carried out using a PerkinElmer spectrum 100. Each sample was uniformly mixed with dried KBr powder with a ratio of 1:100 and the mixture was pressed in the form of pellets. The FTIR spectrum

of each sample was recorded from 400 to 4000 cm^{-1} at a resolution of 4 cm^{-1} and scanned 32 times.

2.3.5. TG-DTG Analysis. TG analysis of each sample was tested by an SDTQ600 analyzer. The sample was heated from room temperature to 1000 $^{\circ}\text{C}$ at a rate of 10 $^{\circ}\text{C}/\text{min}$, and the purge gas was nitrogen (99.999%) at a flow rate of 100 mL/min.

3. RESULTS AND DISCUSSION

3.1. Extract Yield. As displayed in Figure 1, the yields of E_2 and E_4 are 34.89 and 45.60%, respectively, less than those of E_1

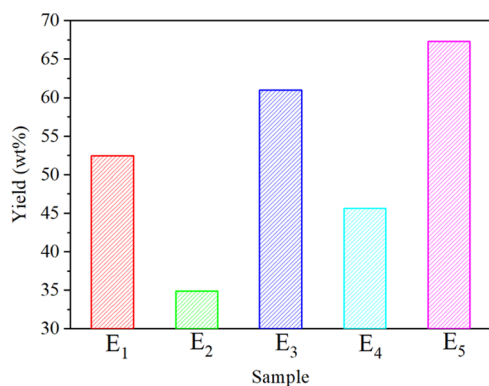


Figure 1. Extract yield of E_1 – E_5 from OS.

and E_3 . The higher yield of E_5 is attributed to the mixing effect of IMCDSAM (mixed solvents); therefore, E_5 is likely to contain most of the organic components in E_3 and E_4 . The different yields of E_1 – E_4 are attributed to the properties of the four solvents, PE, methanol, CDS, and acetone. Methanol is a strong polar compound; the polarity of acetone is weaker than methanol; and petroleum ether is a less polar compound, while CDS is a nonpolar compound. However, the yield of the extract from OS by methanol is lower, which is higher by CDS, indicating that the yield of the extract is related to the polarity of the solvent. According to the principle of similar compatibility, it can be speculated that organic components in OS are mainly nonpolar compounds.¹⁷

3.2. Group Compositions of the Soluble Species in E_1 – E_5 . As exhibited in Tables S1–S5, 66, 68, 56, 59, and 53 compounds can be identified by GC/MS in E_1 – E_5 , respectively. These GC/MS-detectable organic compounds can be classified into 10 groups, including alkanes, alkenes, arenes, ketones, alkanols, ethers, carboxylic acids (CAs), aldehydes, other oxygen-containing organic compounds (OOCOCs), and sulfur-containing organic compounds (SCOCs). And the relative content distribution of group components in E_1 – E_5 is shown in Figure 2. Alkanes and alkenes are the most abundant groups in E_1 – E_5 , with the total content of more than 40%, indicating that the organic compounds in OS are probably alkanes and alkenes. In addition, there are a few amounts of arenes detected in E_4 and E_5 . And there are many oxygen-containing organic compounds in the extracts from OS. Among them, ketones, alkanols, and ethers are the main oxygen-containing organic compounds, and only small amounts of CAs and aldehydes are detected in E_1 – E_2 and E_3 – E_4 . For OOCOCs, containing two or more oxygen-containing functional groups, the relative content is \sim 10%, indicating that there are indeed some complex oxygen-containing components in the oily sludge. The detected

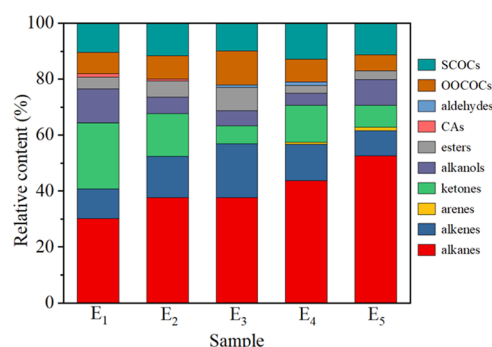


Figure 2. Distribution of group components in E_1 – E_5 .

SCOCs exist in two forms: hexathiane and cyclic octa atomic sulfur, accounting for around 11%.

3.2.1. Distribution of CH and O_x Class Species in E_1 – E_5 . In order to analyze GC/MS-detectable compounds in E_1 – E_5 , the compounds are divided into CH and O_x class species (x is the number of oxygen atoms per compound). CH class refers to the compounds containing only C and H atoms, while the O_x class contains C, H, and O atoms. The distribution of the CH and O_x class species in E_1 – E_5 is displayed in Figure 3. CH

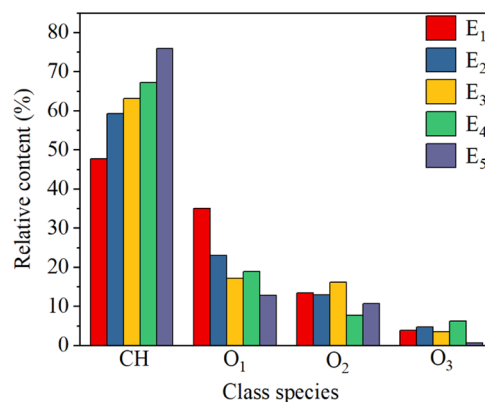


Figure 3. Distribution of CH and O_x class species in E_1 – E_5 .

species are most abundant in E_1 – E_5 , especially in E_5 , with a relative content of 75.84%. For O_1 species, E_1 accounts for around 35%, which is significantly higher than the other samples. This is due to the fact that O_1 species are mainly alkanols, alkenols, and ketones, which can be easily dissolved in PE. The relative content of O_2 and O_3 species in E_1 – E_5 did not change obviously, and O_2 species are mainly ethers, while O_3 species are compounds with three oxygen-containing functional groups, such as ester group, hydroxyl group, or carbonyl group. This indicates that E_1 – E_5 are rich in hydrocarbons and oxygen-containing compounds with low oxygen atoms.

3.2.2. Analysis of DBE and CN for GC/MS-Detectable CH and O_x Class Species in E_1 – E_5 . Figure 4 gives plots of DBE versus CNs for the GC/MS-detectable CH and O_x class species in E_1 – E_5 . As shown in Figure 4, the DBE values and CNs of the GC/MS-detectable compounds in E_1 are primarily concentrated in the ranges 0–4 and 10–20, respectively. Compared with the distribution of DBE and CNs in E_1 , the distribution of CNs in E_2 is more concentrated (14–20), and the compounds with high DBE values are more abundant in E_2 . The DBE value of the compounds in E_3 is widely distributed in 10–30, while CNs are mainly distributed in 0–3,

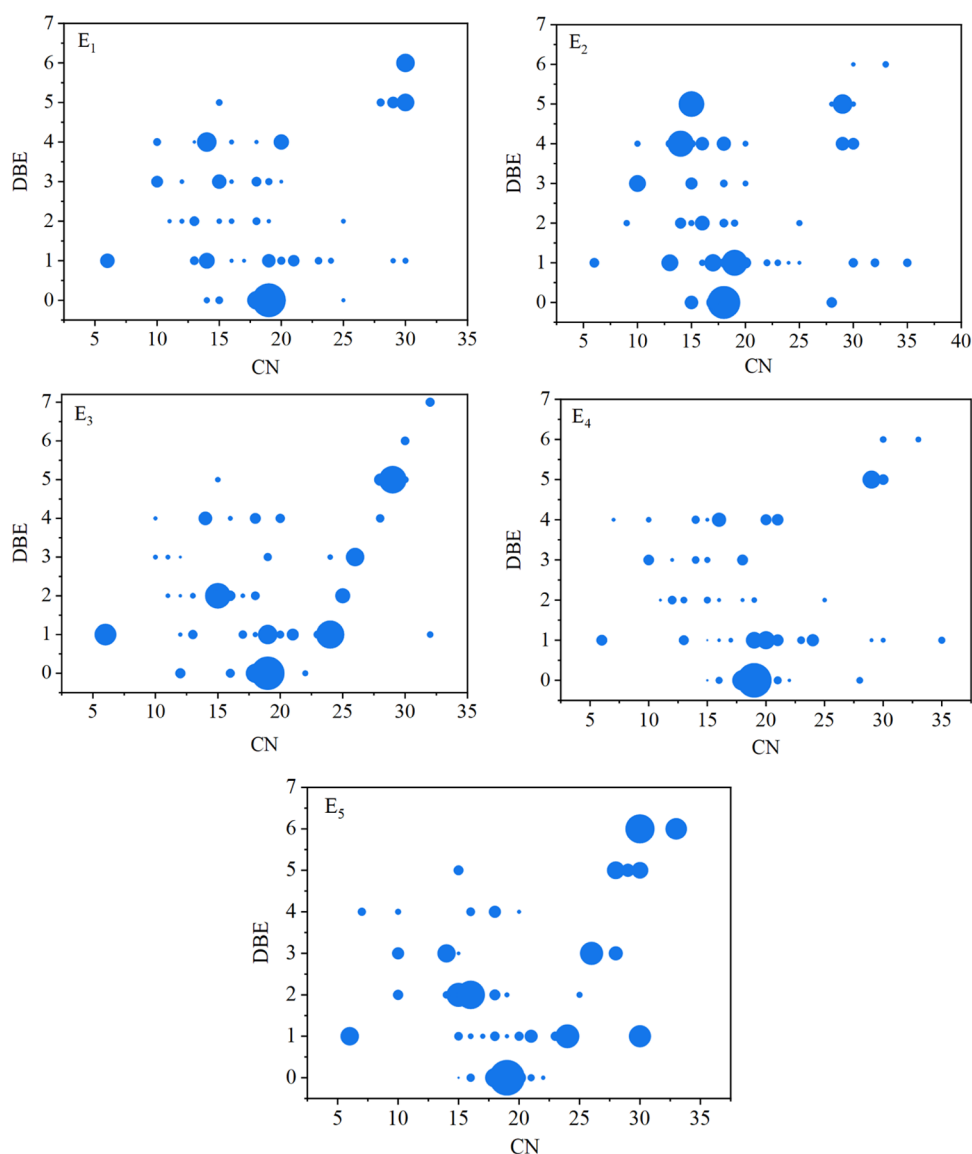


Figure 4. Plots of DBE versus CN for the GC/MS-detectable compounds in E_1 – E_5 .

and the DBE values of the compounds with CNs in the range of 20–30 tended to increase with CNs. The distribution of DBE and CNs in E_4 is similar to that in E_1 . The above results indicate that CDS may be easy to extract high-molecular-weight compounds with high unsaturation from oil sludge. For E_5 , the DBE value and CN distribution are obviously different from E_3 and E_4 . The CN value of the compounds in E_5 is varied from 14 to 33, and the DBE values of the compounds with CNs of 14–25 are concentrated in 0–3, while the DBE values of the compounds with CNs of 25–33 are concentrated in 5–6. The relative content of the compounds with DBE = 5–6 and CNs = 25–33 in E_5 is higher than those in E_3 , indicating that IMCDSAM presents a strong ability to extract highly unsaturated and high-molecular-weight compounds.

3.3. Analysis of Extraction Residue by FTIR. As shown in Figure 5, the characteristic peaks of aliphatic moieties around 2850 and 2920 cm^{-1} in R_2 and R_4 are slightly stronger than those of other residue samples, while the characteristic peaks of aliphatic moieties almost disappeared in R_1 , R_3 , and R_5 , indicating that R_2 and R_4 are rich in aliphatic moieties. This result is related to the low extract yields of E_2 and E_4 . The

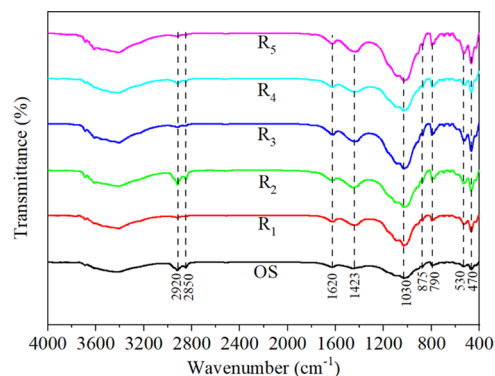


Figure 5. FTIR spectra of OS and R_1 – R_5 .

characteristic peaks around 1620 and 1423 cm^{-1} are ascribed to the aromatic ring $\text{C}=\text{C}$ and the aliphatic chains $-\text{CH}_3$ or $-\text{CH}_2$, respectively.²⁴ The intensity of the peaks in R_1 – R_5 is significantly stronger than that of OS, implying that R_1 – R_5 may be rich in macromolecular aromatic compounds. This may be due to the enrichment of insoluble organic compounds in

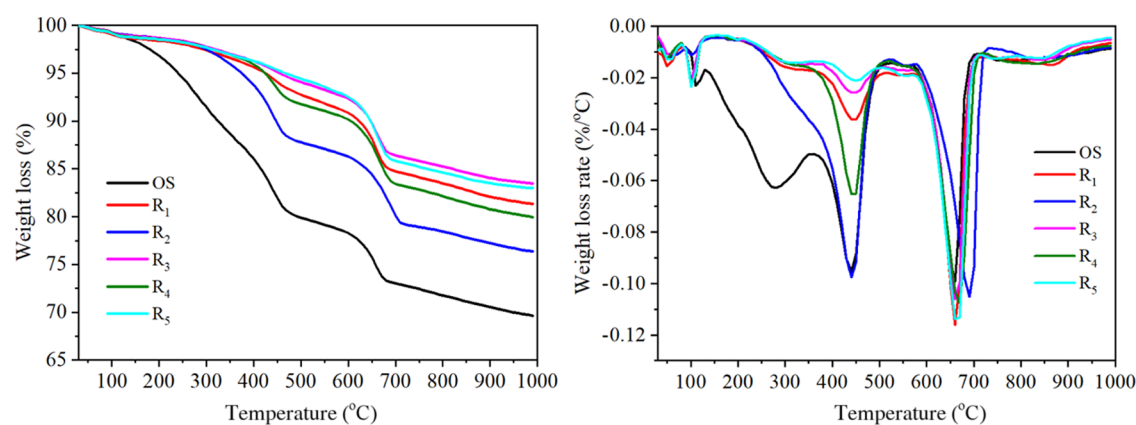


Figure 6. TG and DTG curves of OS and R_1 – R_5 .

the oily sludge after extraction. The absorption peaks around 875 and 790 cm^{-1} are attributed to the deformation vibration of C–H outside the aromatic ring, which further proves that there are more aromatic compounds in R_1 – R_5 . And the characteristic peaks appeared at 1030, 530, and 470 cm^{-1} belong to minerals.²⁵

3.4. Analysis of Extraction Residue by TG-DTG. As is presented in Figure 6, the weight loss trend of R_1 – R_5 is similar to OS, while the weight loss of R_1 – R_5 is significantly lower than that of OS. According to the TG-DTG curves, the pyrolysis process of OS and R_1 – R_5 can be divided into three stages. The first stage ranges from ambient temperature to 150 $^{\circ}\text{C}$, which is related to the loss of moisture.²⁶ The second stage occurs in the range of 150–550 $^{\circ}\text{C}$, and the weight loss of OS is about 20%. In this stage, two obvious peaks of the weight loss rate can be observed at 240 and 440 $^{\circ}\text{C}$ in OS, respectively, whereas there is only one peak at 440 $^{\circ}\text{C}$ for R_1 – R_5 . The pyrolysis of OS in the 150–350 $^{\circ}\text{C}$ range is attributed to the release of volatiles and light oil components with relatively low boiling points, whereas the pyrolysis in the 350–550 $^{\circ}\text{C}$ range is caused by the decomposition of heavy oil components with higher boiling points.^{27,28} Since most of the light oil components with low boiling points are easily soluble in organic solvents, the peak of weight loss rate at about 240 $^{\circ}\text{C}$ disappears in R_1 – R_5 . In the third stage, the weight loss mainly took place between 550 and 750 $^{\circ}\text{C}$, which is mainly caused by the decomposition and condensation of heavy macromolecular organic compounds, and the decomposition of inorganic matter might also be carried out in this stage.²⁹ It is noteworthy that the weight loss of R_3 is higher than that of R_5 before 650 $^{\circ}\text{C}$, and after that temperature, the weight loss of R_5 is higher than that of R_3 , which might be related to less macromolecular heavy organic compounds in R_3 . This suggests that CDS may be more effective in the extraction of macromolecular heavy organic compounds from oil sludge.

3.5. Mechanism of OS Extraction. Petroleum mixture in OS can be separated into four fractions, including saturates, aromatics, resins, and asphaltenes. Saturate fraction is rich in nonpolar hydrocarbons with a linear or branched chain and aliphatic cycloalkanes, which are soluble in most organic solvents.²⁴ Aromatic fraction is composed of aromatic hydrocarbons with various condensation degrees, alkyl substitution, and heteroatom contents.³⁰ The composition of resin fraction is similar to that of aromatic fraction, but the condensation degree and heteroatom content of resin fraction are higher than those of aromatic fraction. Asphaltene fraction

is composed of macromolecular organic compounds with polycondensed aromatic rings and side aliphatic chains, which cannot be dissolved in general organic solvents.²⁴

Zhao et al. considered that the existing form of oil in OS is a relatively stable colloidal dispersion system rather than a homogeneous solution.³¹ The dispersion phase in the colloidal system is the micelle structure, in which asphaltene is the colloidal nucleus, and resin is the solvation shell around asphaltene, while the dispersion medium in the colloidal dispersion system is mainly composed of saturates and aromatics. Some strong interactions including π – π interactions and hydrogen bonds exist between resins and asphaltenes. Lou et al. found that heteroatoms (S, N, and O) in resins and asphaltenes can form hydrogen bonds with hydroxyl functional groups in the solid surface, such as O–H...O, O–H...N, and O–H...S, resulting in strong interaction between macromolecular micelles and solid surface.³² It is concluded that the composition of the colloidal dispersion system and the interaction between the colloidal system and the solid particle are the main factors affecting the extract yield.³³

As shown in Figure 7, colloidal clusters are composed of saturates, aromatics, resins, and asphaltenes. Carboxyl and other heteroatom groups in colloidal clusters can be easy to form hydrogen bonds with the hydroxyls on the surface of solid particles, resulting in strong interaction between colloidal clusters and solid particles. The addition of solvent molecules

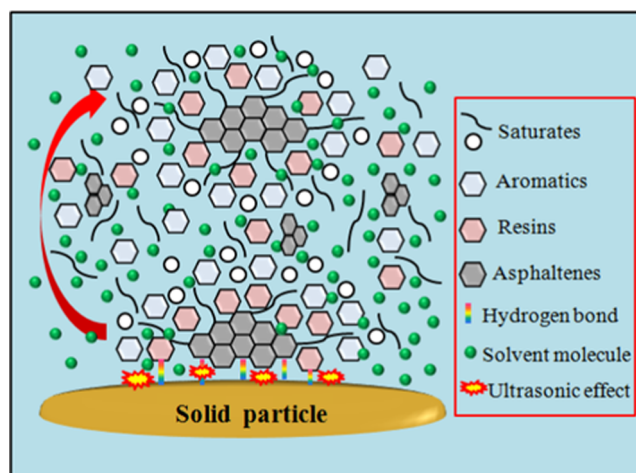


Figure 7. Schematic diagram of the removal of colloidal clusters from solid particles.

can form strong interaction with the colloidal clusters, resulting in the destruction of the hydrogen bonds between the colloidal clusters and the solid surface, thereby releasing the colloidal clusters from the solid surface into the solvent. Of course, it is more effective to destroy the hydrogen bonds between colloidal clusters and solid particles in the ultrasonic field. The colloidal clusters separated from the solid surface are surrounded by solvent molecules and interact with the solvent molecules. The stronger the interaction between the solvent molecules and one of the components in the colloidal cluster, the easier it is to separate the component from the colloidal cluster.

3.6. Solvent Selection. PE, methanol, CDS, acetone, and IMCDSAM are often used as solvents to extract coal. It is found that these solvents can selectively destroy some intermolecular interactions in coal, such as alkyl entanglement effect, weak hydrogen bonds, aromatic π - π interaction, strong hydrogen bonds, hydrogen bond/ π - π interaction effect, and so on.³⁴⁻³⁶ The extract yield of E_4 is much higher than that of E_2 , indicating that acetone can destroy hydrogen bonds more effectively, and it is beneficial for separating colloidal clusters from solid particles. The extract yield by CDS is higher for single solvent extraction, implying that the aromatic π - π interaction was destroyed during the extraction process. While the aromatic π - π interaction mainly exists in aromatics, resins, and asphaltenes, which further indicates that the three components in colloidal clusters can be easily extracted by CDS. Therefore, the selection of less polar (e.g., PE, acetone) or nonpolar (e.g., CDS) solvents that can selectively destroy hydrogen bonds and aromatic π - π interaction is critical for improving the extract yield of OS.

4. CONCLUSIONS

OS was extracted by PE, methanol, CDS, acetone, and IMCDSAM. Most of the organic matter in OS can be extracted by weakly polar and nonpolar solvents, with the nonpolar solvent CDS being more effective. Soluble species from oily sludge consist mainly of hydrocarbons and oxygen-containing compounds with low oxygen atoms, including alkanes, alkenes, ketones, alcohols, ethers, and so on. The CNs of these compounds are distributed in the range 0-35, while the DBE of the compounds is concentrated in the range 0-6. The organic matter in OS exists as relatively stable colloidal clusters, consisting mainly of saturates, aromatics, resins, and asphaltenes, which interact with each other mainly through π - π interactions and hydrogen bonds. Heteroatoms in the colloidal clusters interact by forming hydrogen bonds with hydroxyl groups on the surface of solid particles. Therefore, the selection of less polar or nonpolar solvents that can selectively destroy hydrogen bonds and aromatic π - π interaction is critical for improving the extract yield of OS.

■ ASSOCIATED CONTENT

SI Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acsomega.2c08188>.

Compounds detected in E_1 , E_2 , E_3 , E_4 , and E_5 with GC/MS (PDF)

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Notes

The authors declare no competing financial interest.

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